Habilitation theses

NORMs: Embedding Radioactivity in the Built Environment

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2026.

Content

Introduction

The challenges we face today require us to change our approach to achieving economic growth in a sustainable way. The concept of sustainable development was defined in the 1987 Brundtland Report (UN), the essence of which is that we should meet our needs in such a way that future generations will also be able to do so [1]. Therefore, we must strive to minimise the environmental impact of various industries and reduce the amount of waste generated during processes. Our twofold goal is to focus on developing existing industrial activities and creating a circular economic model while also examining the possibilities for reusing waste that has already been generated. We must view this waste not as waste, but as industrial by-products. This step is important in several respects: it can reduce the amount of material going to landfill and also reduce the need for raw material extraction.

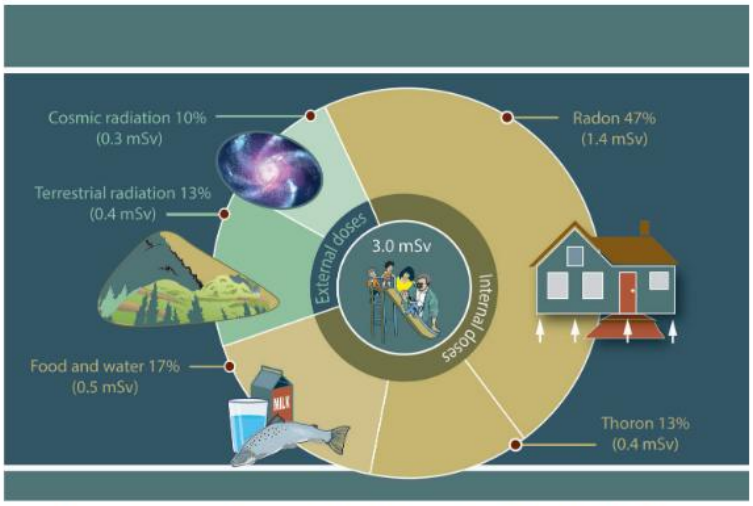
From an environmental, technological, or resource perspective, the need for reuse is not in question. However, in many cases, it poses a risk due to the hazardous content or physical and chemical properties of the material in question. In certain industrial activities, such as mining, oil and gas extraction, energy production and some chemical processes, naturally occurring radioactive materials (NORM) can accumulate in waste [2-7]. These radioactive materials present a potential threat to both the environment and human health, particularly if they are not handled or stored properly. Their presence can increase radiation exposure levels for humans. Due to the presence of NORM materials in waste, classification is necessary prior to use, and the legal regulatory framework for classification must be in place to ensure stronger radiation protection. In addition, radioecological monitoring of NORM materials in landfills may be necessary.

For over a decade, the Department of Radiochemistry and Radioecology has been monitoring NORM deposits, classifying NORM materials and investigating ways to reuse them. My thesis summarises our work and presents new scientific results from our research, using a few examples to illustrate the work of our research group.

Theoretical background

The two main types of radiation exposure affecting humans are natural and artificial. Most of the latter originates from medical applications, while a smaller proportion comes from the nuclear fuel cycle, various industrial processes, and nuclear weapons testing [8]. Around 80%

of radiation exposure is due to naturally occurring radiation. This includes primordial radionuclides, which are radiation-emitting elements with long half-lives that were present when the Earth formed, as well as cosmic radiation and cosmogenic nuclides induced by nuclear reactions. A number of environmental factors influences the level of environmental background radiation. Figure 1 shows the distribution of natural radiation exposure [9].



Source: Adapted from UNEP, *Radiation: Effects and Sources*. Copyright 2016 by UNEP.

Figure 1 Distribution of natural radiation exposure

In terms of radiation exposure, the most significant terrestrial radiation sources are U-238, U-235, Th-232 and their decay series, as well as the K-40 isotope. Radioisotopes of natural origin are present in all elements of the environment as a result of natural cycles: in soil, air, water, and living organisms. As such, their presence can cause both external and internal radiation exposure. External radiation exposure occurs when the radiation source is located outside the human body, while internal exposure occurs when the source enters the body. This entry can occur via inhalation, ingestion or absorption through the skin [10]. The radiation exposure pathways affecting humans are shown in Figure 2.

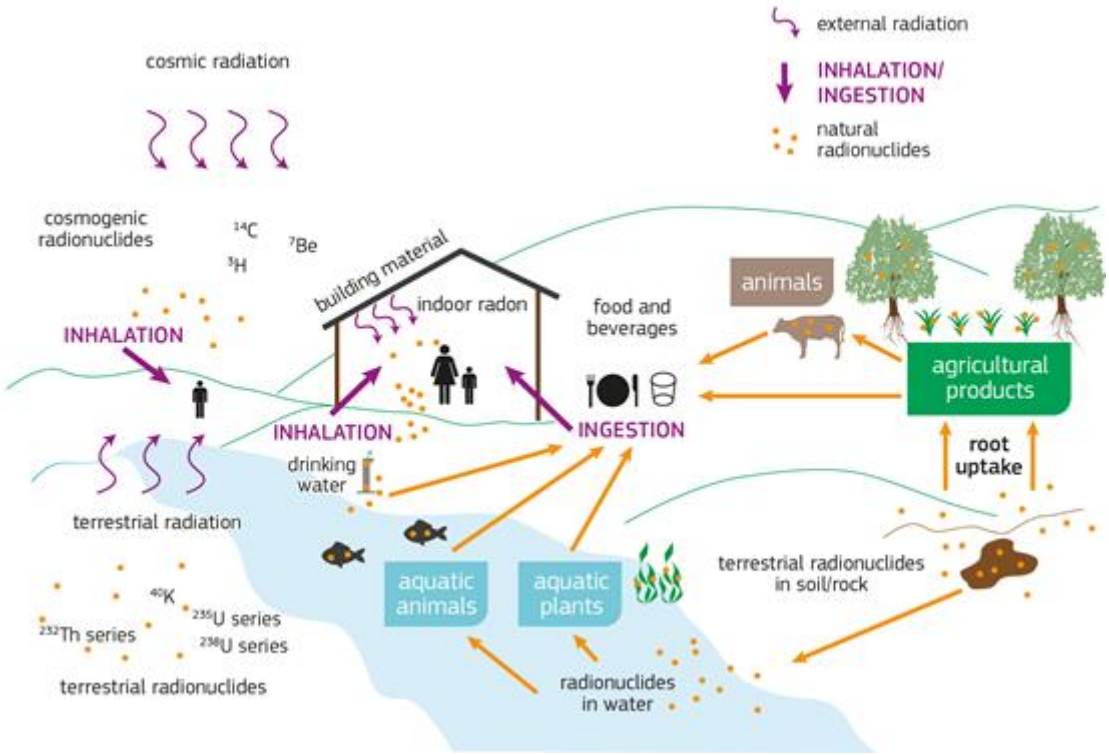


Figure 2 The radiation exposure pathways affecting humans [11]

We can be exposed to external radiation both outdoors and indoors: this is caused by the presence of mainly gamma-emitting isotopes in the Earth's crust. Although the shielding effect of buildings can be taken into account in the latter case, building materials can also be considered a source of radiation due to their naturally occurring radionuclide content [2, 8].

Regarding internal radiation exposure, various isotopes of radon (primarily Rn-222 and Rn-220) have a significant role to play, but the K-40 isotope, which occurs in almost every element of the food chain, can also be a contributory factor. It is estimated that the Rn-222 isotope and its daughter elements cause around 60% of the internal radiation exposure to humans. Radon gas, released from rocks and soil, accumulates in enclosed spaces (e.g. buildings, caves) when it reaches the surface, where it is inhaled and undergoes further alpha decay in the lungs. The short-lived alpha-emitting daughter products (e.g., Po-218, Po-214) adhere to the walls of the alveoli, which increases the long-term risk of lung cancer [12, 13]. Another risk factor is the elements in the uranium and thorium decay chains, which can increase radiation exposure when they enter drinking water or the food chain. The amount of these isotopes, and thus the magnitude of the radiation exposure they cause, depends on a number of factors (e.g. the chemical properties of the medium, temperature parameters), and the chemical properties of the isotope in question are also decisive [14, 15].

It should be noted that certain environmental elements may become enriched, a phenomenon that can be attributed to both natural environmental conditions and anthropogenic effects [14, 16-17]. This accumulation can result in an increase in external and/or internal radiation exposure to humans, thereby posing a health risk. Radioecology is responsible for identifying areas, applications and uses that may pose a risk. It is also responsible for developing and implementing monitoring procedures as necessary, performing dose estimates and managing the existing radiation situation.

NORMs

According to the guidelines of the International Atomic Energy Agency (IAEA), NORM (Naturally Occurring Radioactive Material) refers to materials that do not contain significant amounts of radionuclides other than those occurring naturally [18]. In this interpretation, the precise definition of "significant quantities of naturally occurring radionuclides" is a regulatory issue. NORM materials are defined as those containing naturally occurring radionuclides of crustal origin that result from natural or artificial enrichment processes. These materials have the potential to cause a significant increase in radiation exposure [19].

In order to protect the population and any workers who may be affected, it is essential to map NORM materials and areas and carry out the necessary risk assessments. Numerous research groups worldwide are investigating this issue, and a set of criteria for the use of NORM materials is being developed in the legal frameworks of an increasing number of countries. Hungarian legislation differentiates between building materials made from natural materials (alunite-containing clay slate, granite rocks, porphyry, tuff, trass and lava) and materials containing residues from industries that process naturally occurring radioactive materials (fly ash, phosphorous gypsum, slag containing phosphorus compounds, tin slag, copper slag, red mud and steel production residues) [2, 19].

It is an established fact that primordial radionuclides are present in all environmental elements in various forms. They can be found either as the main constituent of certain rocks or as trace elements. The presence of these materials poses a radiological risk, depending on the quantity, geochemical processes, and use. During specific industrial procedures, physical-chemical separation can lead to the accumulation of these radionuclides in the end product or certain by-products. When handling them, it is essential to consider radiation protection considerations. This requires the clear identification of the areas and industries concerned, and the prioritisation of radiation protection considerations. The industries in which this saturation

may occur are listed in Annex 6 to Regulation 2/2022 (IV.29.) OAH Decree 6. Annex, may include the following: rare earth mining, processing, production of thorium compounds and thorium-containing products, niobium and tantalum ore processing, oil and gas production, geothermal energy production and use, TiO_2 pigment production, phosphorus production, zirconium and zirconium industry, phosphate ore processing, phosphate fertilizer production, cement production, coal mining, coal-fired power plants, phosphoric acid production, iron production, tin, lead and copper metallurgy, groundwater filtration facilities, ore mining, ore processing, alumina production and processing [19]. Table 1 lists some of the NORM industries that are important from a radiation hazard perspective according to the IAEA [20, 21].

Table 1 NORM industries' radiation hazard perspective

Industry	Typical radionuclides	Avarage activity concentration [Bq/g]
Mining rare earth metals	Ra-228	10
	Th-232	
Production and use of thorium and its compounds	Th-232	20
Production of niobium and ferro-niobium	Th-232	100
	Po-210	100-500
	Pb-210	5
	Ra-226	
	U-238	300
Uranium mining	U-238	10
Oil and gas production	Pb-210	1000
	Ra-228	15000
	Ra-226	
Production of titanium dioxide pigments	U-238	<2
	Th-232	
	Ra-226	1-1600
	Ra-228	
Phosphate industry	Th-232	<3
Zirconium and zirconium oxide industry	U-238	2-4
	Po-210	200-600
Manufacture of tin, copper, aluminum, zinc, lead, iron, and steel	Po-210	1-200
Coal combustion	Po-210	> 100
	Th-232	

External radiation exposure from NORM materials is primarily caused by the presence of the isotopes U-238 (T: $4.47*10^9$ years) and Th-232 (T: $1.4*10^{10}$ years) and their daughter elements. Due to its long half-life, the isotope U-235 (T: $7.04*10^8$ years) is also present in the Earth's crust, but its abundance is much lower than that of the isotope U-238 (natural occurrence

approx. 0.72%). In the case of secular equilibrium, all members of the decay series have significantly lower activity concentrations than the elements of the U-238 decay series, which has a natural occurrence of approx. 99.27%. The average abundance of the U-238 isotope is 33 Bq/kg, that of the Th-232 isotope is 45 Bq/kg, and that of the K-40 isotope is 412 Bq/kg. It should be noted here that these values may vary significantly due to natural enrichment or human activity [2, 14].

NORM materials commonly contain the following radionuclides: U-238; Ra-226, Rn-22, Pb-210, and Po-210 from its decay series; Th-232 and Ra-228 from its decay series; and K-40.

The presence of radon is also decisive in the case of internal radiation exposure from NORM materials. Radon is a noble gas that is colourless and odourless. It is found in all decay series of the Earth's crust and is produced during the alpha decay of radium. Due to the structure of the element, it is not able to form chemical bonds with other chemical elements. It has excellent dissolution properties in water and organic solvents, depending on the temperature. There are three naturally occurring isotopes: radon (Rn-222), thoron (Rn-220) and actinium (Rn-219) [15, 22]. Following its formation, it can be expelled from solid particles and reach the surface. During the decay of its parent element, the radium isotope, radon can escape into the pore space (emanation), from where it can escape into the environment through diffusion and conversion processes (exhalation) [23-24]. The extent of escape is influenced by the initial radium activity concentration of the material, the particle size, the material's structure, and the diffusion path length. Figure 3 shows the process of radon emanation and exhalation.

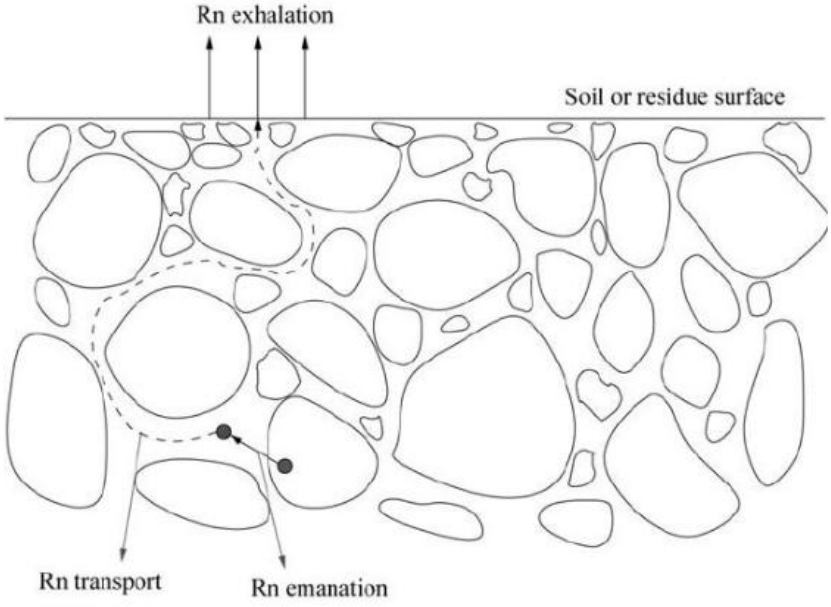


Figure 3 Radon emanation and exhalation

Handling of NORMs

NORM materials generated during industrial processes have traditionally been identified as waste and most often deposited in landfills. When disposing of them, the primary consideration was not radiochemical parameters, but rather the appearance and chemical parameters of the waste.

The presence of NORM materials in landfill sites represents a potential hazard to both the environment and the surrounding population. Inadequate design and maintenance can result in the mobilisation of radionuclides, potentially contaminating surface and groundwater, as well as the surrounding soil and vegetation. This may lead to an increase in external and internal radiation exposure. This increase can be influenced by a number of environmental factors, including weather conditions. The storage of NORM materials and the minimisation of risks arising from inadequate storage is an extremely complex task that requires cooperation between various fields (regulation, environmental protection, radiation protection, etc.). It is regrettable that this area still requires intervention in many respects. If inappropriate methods are used, a disaster such as the red sludge disaster that occurred in Ajka, Hungary, on October 4, 2010, could occur [25]. The incident occurred at the northwestern corner of the Ajka Alumina Plant's sludge reservoir No. 10 (Figure 4), leading to flooding in the surrounding areas.

Figure 4 Red mud disaster in Ajka: mud storage reservoir burst

In accordance with the principles of sustainable development, the efficient use of available resources and raw materials is our primary task, so industrial by-products can be identified as potential secondary raw materials. However, it is important to note that, as with the storage of industrial by-products containing NORM materials, their use can only be considered acceptable if the environmental impact and radiological risk are negligible. In other

words, the radiation safety of the manufacturer, the user and the population must be ensured. The construction industry has the greatest demand for these industrial by-products as an additives.

Building materials fulfil a dual role in radiation protection: Their physical properties make it possible to shield against cosmic and terrestrial background radiation, thereby minimising the external radiation exposure inside buildings. However, due to their composition and natural radionuclide content, the building materials used can contribute to the radiation exposure of those inside. Consequently, the radiological characteristics of NORM materials used as additives directly influence the annual effective dose to the population. The increase in gamma dose is primarily caused by the Ra-226, Th-232, and K-40 isotopes originating from building materials, while the increase in internal radiation exposure is contributed to by radon escaping into the body. The indoor radon activity concentration is dependent on the type of material and the amount used during construction. A number of factors, including the composition, structure, porosity, air pressure, humidity, and temperature of the building material, also influences the magnitude of the additional dose from radon. In view of all this, it is mandatory for NORM materials intended for use in the construction industry to undergo radiological testing. The objective of this testing is to ensure that the radiation exposure of the population resulting from their installation complies with the relevant radiation protection requirements [22, 26-27].

Dose assessment

When ionising radiation interacts with living organisms, the resulting changes at the cellular level can lead to systemic processes. The occurrence of these changes depends on the size of the dose range, and various physical, chemical, and biological factors can influence their appearance by modifying the effects of radiation. Depending on the size of the dose range, stochastic or deterministic dose-response relationships can be described during the interaction. The former is used in the case of small dose ranges and estimates the probability of harmful effects occurring as the dose increases, while in the latter case, we must expect harmful effects to occur above a so-called threshold dose, where the severity of the effects increases as the dose increases.

In NORM situations, the primary concern is the increased background radiation, which can lead to elevated radiation exposure for the population and the environment. Estimates can be calculated using dosimetric methods. Whenever direct dose measurement is not possible, the

expected radiation exposure of the population is determined based on measurement results obtained during radioecological monitoring studies. These results are then taken into account alongside the given circumstances, environmental parameters, living conditions (such as food consumption) and typical residence times, among other relevant factors.

Estimates of external radiation dose exposure can generally be calculated based on the measured dose value. It should be noted that the dose rate values measured in the air need to be adjusted due to the human body's own shielding effect. At the same time, the amount of time spent outdoors or in buildings must also be taken into account. It should be remembered that the building material itself can also be a source of radiation. With regard to gamma radiation, the effective dose can be estimated using the following relationship, with a semi-infinite approximation and at a height of one meter above the ground surface:

$$E_f = K_f \cdot \tau \cdot F \cdot \phi$$

where: K_f is the surface dose coefficient [(Sv/s)/(Bq/m^2)], Φ is the surface contamination (Bq/m^2), τ is the duration of exposure and F is the ratio of time spent outdoors to time spent indoors, as well as a reduction factor that takes into account the shielding effect of buildings, with a value of 0,1-0,8 [8].

As radionuclides in environmental elements can come into direct or indirect contact with the human body through incorporation into the food chain, it is also necessary to estimate the radiation exposure resulting from inhalation and ingestion. This can be calculated from the concentration of the radionuclide activity, taking into account the route of exposure. In the event of inhalation, the following relationship can be utilised:

$$E_h = K_h \cdot V \cdot c \cdot F \cdot \tau$$

where E_h is the effective dose, K_h is the inhalation dose coefficient [Sv/Bq], V is the respiratory rate [m^3/nap], c is the activity concentration of the given isotope [Bq/m^3], F is the reduction factor taking into account the ratio of time spent outdoors and indoors, as well as the shielding effect of buildings, with a value of 0.1-0.8, and τ is the duration of stay [days].

In case of ingestion, the extra dose can be calculated using the following formula:

$$E_l = K_l \cdot G \cdot c \cdot \tau$$

where E_l is the effective dose, G is the amount of food consumed [kg/nap], K_l is the ingestion dose conversion factor [Sv/Bq], c is the concentration of the given isotope in the food [Bq/kg] and τ is the consumption period of the given food [days] [28].

When estimating radiation exposure, the location-specific parameters of the given environment and local consumption habits must be taken into account. The results obtained must also be aggregated according to the exposure pathways for each isotope.

Materials and methods

Measured NORMs

One of the primary focus of our research group is radioecological mapping of NORM deposits, alongside investigating and applying biomonitoring possibilities and examining potential uses for NORM materials. In this theses, we examine oil drilling (drilling mud), coal processing (coal slag, fly ash), and waste generated during alumina production (red mud) as NORM materials.

Oil is an invaluable source of energy. The production of oil often results in the generation of various by-products, including sludge, slag, water, shale, and ash. Elevated concentrations of uranium and thorium decay series may occur, which is why these wastes are categorised as NORM materials, although their radionuclide content can vary significantly (0-1000 Bq/kg) depending on the phase of the technology they originate from and their geographical location. Due to the expected risks, their radiological classification, disposal, possible use, and continuous monitoring may be necessary [29-31].

The by-products and waste generated during coal processing are classified as NORM material due to their radionuclide content. The concentration of uranium and radium in slag and ash is higher than in the original coal [14]. These by-products are used widely, primarily in the construction industry. In addition to their widespread use, their significant radioactivity must be taken into account, as confirmed by several surveys [32-35].

Red mud is a by-product of alumina production. Bauxite is extracted using an alkaline solution of concentrated sodium hydroxide, and the red mud produced during this process is highly alkaline. The quantity produced is significant, roughly equivalent to the amount of aluminum oxide produced using this technology [36]. In Hungary, 4 million tons of red mud are generated annually during alumina production. Due to its chemical properties, it is classified

as Category II hazardous waste [37], and is deposited in sludge storage facilities. This product is suitable for use in small quantities in the production of pigments and paints, in catalytic processes, and as an additive in the construction industry [38].

Measurement methods

In our research, we focused on three main areas: identifying NORM materials/areas, radioecological monitoring of these areas, and, where relevant, classifying NORM materials and examining possible areas of reuse. This is a complex process, but it provides us with a clearer picture of the sources of potential additional radiation exposure in our immediate environment. In order to estimate the radioecological risk posed by NORM materials, it is necessary to know not only their composition, but also the release characteristics of the individual radioactive isotopes. With knowledge of the mobility data of radioisotopes and the irradiation pathways, the increase in radiation exposure to humans can be estimated. The main steps of the process are outlined in Figure 5.

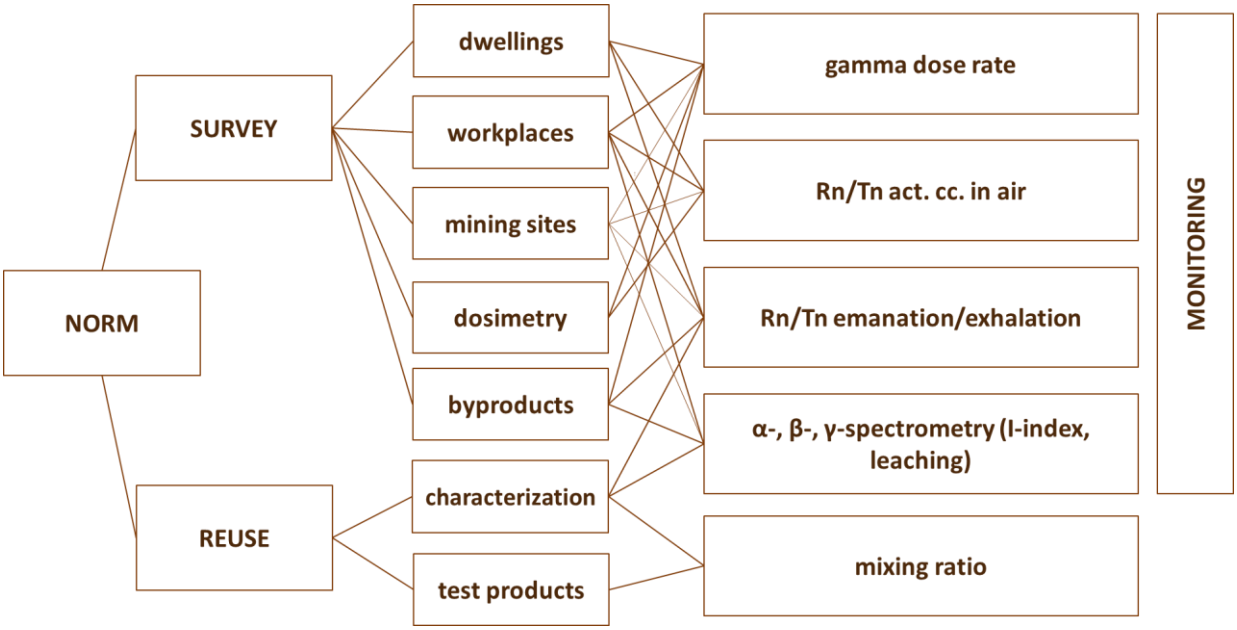


Figure 5 RRIT protocol for testing NORM materials

As a first step in identifying NORM materials, we performed on-site gamma dose rate measurements, laboratory gamma spectrometry measurements, and field and laboratory radon emanation/exhalation measurements. Following a thorough examination of the results obtained, we explored the potential of utilising the specified industrial by-product as a raw material in the construction industry, considering various mixing ratios. To this end, we calculated the activity

concentration index (I-index) value for gamma radiation emitted by building materials specified in the legislation using the following equation: [19]:

$$I = C_{Ra\text{-}226}/300\ Bq/kg + C_{Th\text{-}232}/200\ Bq/kg + C_{K\text{-}40}/3000\ Bq/kg$$

where $C_{Ra\text{-}226}$, $C_{Th\text{-}232}$ és $C_{K\text{-}40}$ activity concentration of relevant radionuclides in the building material, express in Bq/kg.

If the resulting I-index value does not exceed 1, the material in question can be used as a raw material in construction. In this case, it is important to examine the mixing ratio of NORM materials added as additives, because the index refers to the construction material and not to the constituent additives. In the case of leaching tests, we performed isotope-specific analysis, for which we carried out alpha, beta, and gamma spectrometry measurements.

In addition to the classification step, it is important to monitor the area using both active and passive methods. Bioindication and biomonitoring studies, which assess the state of the environment and its changes, are becoming increasingly important in today's global context. This is due to the fact that plants are in constant dynamic interaction with the soil, air and water. This interaction has the potential to enhance the suitability of certain plant species for monitoring the quality of the environment. Specifically, these species could be utilised for the assessment of the geosphere, atmosphere, and hydrosphere near the surface, as well as for the tracking of specific processes. In the context of biomonitoring studies, we utilise bioindicator organisms, defined as living beings or communities that exhibit a response, whether morphological, histological, cellular, metabolic or compositional, to the presence and/or alteration of specific elements or chemicals. Lower-order species and simpler vegetation are generally used as bioindicators. It is expected that they will respond selectively to the component under investigation. They should not damage or pollute the area under investigation, and in the case of bioindication, they should respond differently and selectively. In the case of bioaccumulation, they have a selective and high bioconversion factor [39]. n our case, we conduct tests using accumulation monitor species with the aim of determining the quantity of pollutants present in the environment. Accumulation species are distinguished by the accumulation of substances within the organism without significant damage. These compounds are not subject to decomposition or utilisation by the organism's metabolism. Consequently, the absorption and storage time of these substances in the accumulating organism is greater than their excretion time [40-41].

Moss, for example, can be used as a bioindicator. It plays a very important role in certain plant communities, and its significant biomass mass is suitable even under unfavorable climatic conditions, making many species suitable for biomonitoring studies, primarily for the detection of heavy metal pollution in the atmosphere [42-44]. Fungi and tobacco plants can also be used as accumulation bioindicators, as polonium accumulation can be observed in both cases [45-47]. Biomonitoring studies enable long-term radioecological testing of an area: mosses and fungi are passive species that occur naturally in the study area, while tobacco plants were planted by our research team as a bioindicator species in the given areas. In both cases, the analysis is performed after sample collection and laboratory testing.

This theses focuses on the classification of NORM materials/deposits and their usability in construction, so I will briefly present these below.

Sampling

The oil and gas production recultivated drilling sludge depository examined is located near Zalatárnok (Figure 6). It consists of three storage facilities, which we examined with and without cover (1.5 m thick soil layer).

Figure 6 Drilling sludge depository near Zalatárnok

The coal ash reservoir in Ajka is located within the city (Figure 7) and rises approximately 20 metres above its surroundings. The landscaping has been completed, but due to the specific nature of the recultivation process used, the thickness of the cover layer is uneven and the Ra-226 content varies. Erosion dust from the storage facility can be carried by the wind to nearby agricultural and residential areas.

Figure 7 Coal ash depository in Ajka

The red mud reservoir is located between Ajka and Kolontár (Figure 8). The reservoirs are strategically located west of Ajka, on a gently sloping area near the Torna stream valley. Red mud is deposited in two ways: dry or wet. In Ajka, wet deposition was used, whereby the sludge was placed in the reservoir without dewatering. Due to the risks associated with wet storage, following the aforementioned dam breach on 28 February 2011, they transitioned to dry technology, which involves the reduction of the moisture content of the red mud through vacuum filtration (or high-pressure technology) [48]. This solution does carry some inherent risks. For example, there is a possibility of dust dispersion in certain weather conditions. For instance, when wind conditions are favourable, red mud dust containing NaOH can visibly cover surrounding areas.

Figure 8 Red mud depository near Ajka

Sample preparation

The collected samples were prepared for laboratory measurements. For gamma spectrometry measurements, the collected plant and soil samples must be dried in a drying oven

(to constant weight, 90 °C), pulverized, and homogenized. The prepared sample is then placed in a foil-lined Marinelli vessel (Figure 9) of known mass and sealed for 30 days to allow secular equilibrium to be established between the parent and daughter elements [49]. In the case of water samples, after any necessary pre-concentration (evaporation), they are also stored in sealed sample containers for 30 days.

Figure 9 Marinelli-beaker

For radon exhalation measurements, the process is similar to gamma spectrometry sample preparation: the prepared sample is placed in an accumulation chamber, the accumulation chamber is then flushed with N_2 gas (Figure 10), and the sample holder is sealed airtight. The sealed sample is then stored for between three and four days, after which the radon concentration of the sample can be determined.

Figure 10 Accumulation chamber

Alpha spectrometry measurements require a sophisticated, multi-stage sample preparation process due to the short length of alpha radiation. The first step is the chemical processing, pre-concentration, and dissolution of the sample, followed by isotope-selective

separation and source preparation. The measurement is typically carried out in a vacuum using a semiconductor (PIPS) detector alpha spectrometer. A tracer isotope is used to monitor the process. A tracer is an isotope that is chemically identical to the isotope being studied, but does not occur in the sample. Its use allows the efficiency of the entire process to be determined, as it is added in a known quantity to the sample to be measured at the beginning of the process [50].

The sample can be transferred into the solution by complete or partial extraction, according to various protocols. The main differences between these protocols are the amount of sample to be measured, the type and amount of chemicals used, and the contact time. Their selection depends on the purpose of the measurement, which may be the analysis of a single fraction (e.g., water-soluble, plant-available fraction) or the total content, and may involve one or more steps. Currently, there is no universally accepted method for detecting radionuclides within the EU. As a starting point, researchers generally choose methods suitable for determining heavy metals.

Our research group performs preparation in accordance with the Hungarian standard MSZ-21470-50:2006, a method used in environmental practice to determine the presence of toxic elements, heavy metals, and Cr(VI) in soils. The method recommends several parallel, single-step extractions: it includes two complete extraction steps, which involve aqua regia and nitric acid extraction (in the case of soil samples, other chemicals may also be required due to the SiO2 content, e.g. HF). The standard also deals with the assessment of the likelihood of release into the environment, measuring the possibility of entry into the water cycle using distilled water preparation with rain, while the Lakanen-Erviö solution method examines uptake through the root system of plants. It is also possible to carry out traditional chemical or microwave detection using a specific protocol [51].

The Tessier sequential extraction method [52] was also originally developed for heavy metal speciation studies. The method examines the binding forms and environmental mobility of elements in five consecutive steps, focusing on the ion-exchangeable fraction, the carbonate-bound fraction, the iron- and manganese-oxide-bound fraction, the oxidizable fraction, and the residue, in that order.

Following the preparation of the samples, the next step is to prepare an alpha source suitable for measurement. The source preparation options depend on the alpha-emitting isotope to be measured: evaporation, vacuum evaporation, micro-(co)precipitation separation,

electrolysis (electrodeposition) [50]. The purpose of source preparation is to deposit a nearly mononuclear layer onto a surface in order to minimise self-absorption in the sample. The most widely employed technique is electrodeposition, where alpha-emitting isotopes function as cathodes in an aqueous environment. During the process, a charge transfer occurs between the metal (electrode) and the electrolyte solution. The electrolyte is an organic or inorganic solution containing the metal that forms the alpha source. The metal used as the electrode can be platinum, copper, aluminium, etc [53].

We use a spontaneous deposition method to determine polonium. This process involves the separation of the polonium on high-nickel stainless steel discs in an 80 °C water bath for a period of three hours, with continuous stirring [54]. This can be seen in Figure 11.

Figure 11 Po-210 source preparation

In the process of determining uranium and thorium, it is essential to first perform a radiochemical separation step using extraction chromatography with UTEVA resin [55]. This preparatory step is integral to ensure the efficient extraction and purification of the desired elements. The alpha source was prepared for both uranium and thorium using an electrodeposition method, employing a Canberra Electro α system, with separation performed on a high-nickel-content acid-resistant steel disc.

Gamma-spectometry

Structurally, the atomic nucleus can be described by its discrete energy levels. Following a transformation (whether radioactive decay, an artificial or natural nuclear reaction),

the atomic nucleus may remain in an excited state. The process of excitation involves the emission of electromagnetic radiation with an energy corresponding to the transition between the levels of the atomic nucleus. This electromagnetic radiation is gamma radiation, and the energy of gamma photons is specific to each isotope due to the defined energies of the levels. This provides a good opportunity to identify the radionuclide and determine its activity. The process of detection is based on the interaction between gamma radiation and the detector material. This interaction can occur in three main mechanisms, depending on the energy of the gamma radiation: the photoelectric effect, Compton scattering and pair production. The interaction results in electron emission, and the electrons can create ion pairs and light photons during further ionisation and excitation processes. The photons and charged particles produced are then collected and converted into processable voltage or current pulses at the detector output. The amplitude of these pulses is proportional to the energy of the absorbed gamma photons.

Gas ionization, scintillation, and semiconductor detectors can be used to detect gamma radiation, while scintillation and semiconductor detectors can be used for spectrometric purposes. Due to its excellent energy resolution, HPGe semiconductor detector spectrometry is commonly used in laboratory tests for the analysis of environmental samples. The disadvantage of this is that it requires continuous cooling. For field measurements and samples containing few isotopes, simpler scintillation detectors (mainly NaI(Tl)) are used, which have poorer energy resolution but do not require cooling [8, 50, 56].

During qualitative determination, the device's energy is first calibrated using radioactive isotopes with known energy spectra, after which the spectrum of the unknown sample is recorded using the required measurement time. Following a calibration process, the type of radionuclide present in the given sample can be determined using a nuclide table. The amplification must be selected so that the signal generated by the gamma photon with the highest energy to be measured still falls within the measurement range. Both relative and absolute methods are utilised for the quantitative determination of the radioactivity of a sample. The relative method can be employed if the sample to be determined has the appropriate composition (i.e., identical isotopes are present) and if a standard with known activity is available. It is imperative that the geometry and density of the samples are also identical. Using the standard sample, whose activity is known, the peak area corresponding to unit activity can be determined. The corresponding peak of the sample with unknown activity can then be selected, and the peak area can be calculated by comparing the two. For the absolute method, a

sample with known activity consisting of any isotope is required, as well as a standard. This is used to determine the energy dependence of the detector's efficiency.

Figure 12 shows the gamma-measurement setup used at the Department of Radiochemistry and Radiology.

Figure 12 Semiconductor HPGe detector at RRIT

An ORTEC GMX40-76 high-purity germanium semiconductor detector was utilised for gamma-spectrometry. The detector demonstrated a relative efficiency of 40%. The spectra were recorded using an ORTEC DSPEC LF 8196 MCA multichannel analyser and then evaluated using Aptec MCA software. The IAEA 326 soil sample was used as the reference material. The activity concentration of Ra-226 was calculated from the 295 keV gamma line of Pb-214 and the 609 keV gamma line of Bi-214, while the activity concentration of Ra-228 was determined from the 911 keV line of Ac-228 and the 2614 keV line of Tl-208.

In-situ gamma-dose rate measurements

The measurement of radiation originating from the earth's crust can be carried out using in situ gamma spectrometry. During the measurement process, an unshielded gamma detector is utilised, which is positioned at a height of 1 m above the ground surface. During the measurements, we used a NaI(Tl) scintillation detector (Automess 6150AD-b – Figure 13), which has a detection limit of 1.3 nSv/h.

Figure 13 Automess 6150AD-b

Radonemanation, exhalation

During measurement, radon can be determined directly or through its daughter products. The direct method involves the detection of the alpha particles produced during radon decay, along with those originating from its daughter products (Po-218, Po-214). The indirect method involves the detection of alpha, beta, and gamma radiation from radon decay products using a suitable detector. When measuring the radon concentration in a given air space, it is important to note that the instantaneous measured value is only indicative, as a number of environmental parameters (e.g. pressure) influences the radon concentration. Active and passive detectors can be used to determine radon concentration. The former are used for shorter-term measurements (e.g. a few days), while the latter are used for longer-term measurements (e.g. a year), allowing us to observe long-term (seasonal) changes.

The accumulation method involves collecting exhaled radon in a closed space and determining the amount exhaled based on the elapsed time, decay, and radon concentration. The setup used at the department is shown in Figure 14.

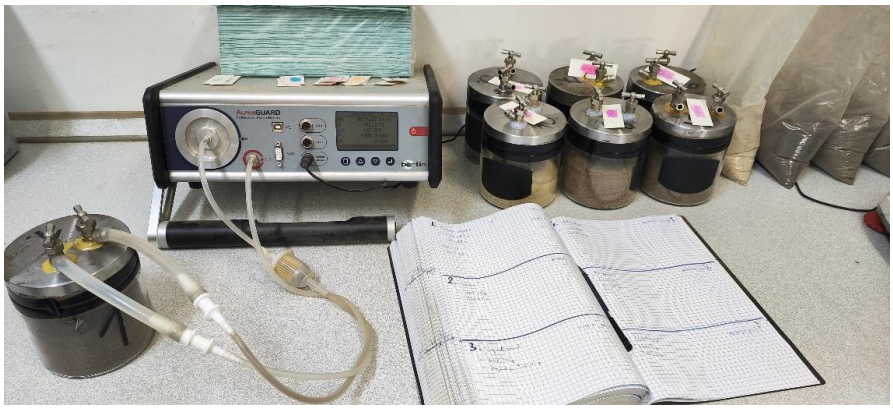


Figure 14 AlphaGUARD 2000 radonexhalation measurement system

Alfa-spectrometry

An alpha particle is a helium nucleus with a mass number of 4 and a double charge. Structurally, it consists of two protons and two neutrons. Alpha radiation is emitted by heavy atomic nuclei. The energy of an alpha particle emitted from an atomic nucleus is a specific, discrete value. During alpha decay, the energy released is between 3 and 9 MeV, 98-99% of which is carried by the alpha particle. The energy of the alpha particle is indicative of the emitted isotope; therefore, spectroscopic methods can be used to make both qualitative and quantitative determinations for a given radiation source [57, 58].

As previously mentioned, a critical part of alpha spectrometry measurement technology is the production of a highly accurate, infinitely thin alpha source. Due to self-absorption, a proportion of the energy of the alpha-emitting particles is already absorbed in the sample. This results in significant variations in the alpha spectrum of the sample, such as deterioration of resolution or elongation of the low-energy parts of the spectrum energy peaks.

During our measurements, an Ortec Alpha Duo semiconductor PIPS detector dual-chamber spectrometer was used with an energy resolution of <20 keV. The detector is shown in Figure 15.

Figure 15 Ortec Alpha Duo with semiconductor PIPS detector

Results

The results are presented as outlined above. First, I will discuss the monitoring tests of the NORM reservoirs examined, then I will present the results of the related dissolution tests and the associated radiation exposure estimates, and finally I will present the usability of the examined NORM materials in the construction industry.

Radioecological investigation of NORM depositories

Oil sludge depository near Zalatárnok

We took measurements in the covered and uncovered parts of the Zalatárnok reservoir. During the field measurements, the environmental dose equivalent (H*(10)) was examined at a height of 1 m using a 6150AD-b Automess dose rate meter, the radon activity concentration in soil gas using an AlphaGUARD PQ2000 radon monitor, and the gas permeability of the soil using a RADON-JOK permeability meter. Following the appropriate steps of sample preparation, the activity concentration of the main gamma-emitting radionuclides in the samples was determined using a semiconductor HPGe detector gamma spectrometry method. The specific radon exhalation and radon emanation factor of the samples was measured using an accumulation method with the AlphaGUARD radon monitor.

Based on the measurements, it can be concluded that the dose equivalent rate ranged from 67 to 85 nSv/h in the uncovered state, increasing to between 83 and 89 nSv/h after the application of the cover layer. The dose equivalent rate measured on the covered landfills was approximately 86 to 90 nSv/h. The calculated gamma dose rate values were similar to the measured data, but generally resulted in slightly higher values. Following the application of the cover layer, a slight increase in both the measured and calculated gamma dose values was observed. This can be explained by the radionuclide content of the cover layer. The spatial dispersion of the dose rate in the vicinity of the measurement points remained below 10%, indicating the formation of a homogeneous radiation field.

During the outdoor radon concentration test, we measured values between 9 and 16 Bq/m³ at the uncovered landfill, with an average concentration of 11 Bq/m³. Following the application of the cover layer, the concentrations remained within a comparable range, averaging approximately 13 Bq/m^3. For the landfills already covered, we obtained average values of around 16 Bq/m^3. The measured concentrations in both cases were close to the global

average, and the landfills do not cause significant radon exposure above background levels. Due to the open location, wind and atmospheric turbulence promote the rapid mixing of radon with the ambient air, thus preventing the accumulation of locally released radon.

Based on in situ radon exhalation measurements, the exhalation rate on uncovered landfills was 9 mBq/m²s. Following the application of the cover layer, this rate increased to 14 mBq/m²s. Exhalation on covered landfills was around 13 mBq/m², which in all cases remained below the global average given in the literature. Meteorological effects and the radionuclide content of the cover layer can explain the slight increase in exhalation observed during the application of the cover layer. The extent of exhalation is determined by the amount of radon generated in the soil and its ability to reach the surface. This is also influenced by porosity, permeability, and moisture content.

The examination of radionuclide activity concentrations showed that the activities of the isotopes U-238, Ra-226, Th-232, and K-40 were within the range of the global average or the average values in Hungary. The calculated radium equivalent activity remained well below the limit value for NORM materials in all cases. An analysis of the isotope ratios indicated a slight imbalance in the U-238 decay series, as the Ra-226 concentration slightly exceeded the equilibrium value. This can be explained by the enrichment of Ra-226 during the use of drilling mud. The activity concentrations of the cover layer samples were slightly higher than the Hungarian soil averages, which is consistent with the use of locally mined clay.

It can be concluded that the examined landfills do not cause significant gamma dose exposure or radon concentration exceeding natural background radiation. The application of a cover layer resulted in only minor alterations to the radiation parameters, with the measured values remaining close to the average values reported in international literature in all cases. The physical and radiological properties of the soil, together with meteorological factors, determine the development of radon exhalation and soil gas radon concentration.

Coal ash depository in Ajka

In the case of the Ajka coal ash repository, we conducted field and laboratory measurements similar to those performed for the drilling mud storage facility: During the field measurements, the environmental dose equivalent (H*(10)) was examined at a height of 1 m using a 6150AD-b Automess dose rate meter, the radon activity concentration in soil gas using an AlphaGUARD PQ2000 radon monitor, and the gas permeability of the soil using a RADON-

JOK permeability meter. Following the sample preparation procedure, the activity concentration of the main gamma-emitting radionuclides in the samples was determined in the laboratory using a semiconductor HPGe detector gamma spectrometry method. The specific radon exhalation and radon emanation factor of the samples was measured using an accumulation method with an AlphaGUARD radon monitor.

The objective of the study was to evaluate the radiological risk posed by the recultivated coal-fired power plant fly ash storage facility in Ajka, with a focus on gamma dose rate, radon exhalation, and Rn-222 concentration in the air. The recultivation of the Ajka storage facility was primarily aimed at reducing dust emissions, without setting radiological limits. The average gamma dose rate measured in the recultivated, covered areas was 277 nSv/h, the exhalation value was 486 mBq/m²s, while the radon concentration measured at a height of 150 cm was 36 Bq/m³. The results of the measurements showed that at various points in the storage facility – especially along the embankment and in sloping areas – the radiological parameters exceeded the environmental background values. In the inner line and the sloping zone, the exhalation flux reached 1100 mBq/m^{2}s, the Rn-222 concentration measured in the air was 250 Bq/m^3, while the gamma dose rate was around 380 nSv/h. As the affected, uncovered areas are easily accessible to both humans and animals, there is a potential increase in the health risk from radiation exposure.

Red mud depository near Ajka

In the case of the red mud reservoir, in line with the field and laboratory measurements presented earlier, we also conducted biomonitoring tests. The decision to extend the monitoring tests was driven by the red mud disaster and its consequences, as previously outlined. Our research focused on two key areas: firstly, assessing the contamination of the area based on its distances from the reservoir, and secondly, investigating the detectability of potential radionuclide contamination caused by dust pollution resulting from dry deposition technology. For the latter tests, tobacco plants were planted around the reservoir and our research was supplemented by collecting moss and mushroom samples. For the present studies, the collected samples were analysed using HPGe detector gamma spectrometry and semiconductor (PIPS) detector alpha spectrometry methods after sample preparation.

During the measurements, we determined the activity concentrations of the Ra-226, Th-232, and K-40 isotopes. Based on the results, the activity concentration of Ra-226 in the contaminated areas ranged from 139.5 to 168 Bq/kg, while a greater variation was observed in

the control areas (36.7 to 216.2 Bq/kg), which was mainly caused by an outlier measurement point. According to previous analyses, this outlier value is unrelated to red mud contamination. Rather, it is attributable to local environmental pollution from ceramic industry activities, which have been shown to increase NORM levels. The activity concentration of Th-232 was found to be comparable in both contaminated and uncontaminated areas, ranging from 19.6 to 104.9 Bq/kg. This indicates that the distribution of thorium in the studied area was not significantly impacted by the red mud spill. In the case of the K-40 isotope, there was also no detectable difference between the two types of areas, with values ranging from 306.9 to 479.6 Bq/kg, which falls within the typical range of natural soil radioactivity. Overall, the comparison of the radiological parameters of the soil samples showed that red mud contamination did not cause a significant additional burden in terms of the radionuclides examined. Following a thorough evaluation of the NORM activity data for the soil, it is concluded that the radiological risk in the areas examined can be considered low. Found deviations can be explained by local anthropogenic sources or natural variability.

As previous studies have demonstrated, tobacco plants have the potential to act as bioindicators in contaminated areas. The studies concentrated initially on the absorption process through the root system. Following a notable increase in dusting during the transition to dry processing in the case of the red mud reservoir, we examined whether tobacco plants could serve as bioindicators for polonium uptake through the leaf surface. The polonium activity concentrations in most sampling periods were consistent with the international average values reported in the literature. The Po-210 activity concentration measured in tobacco leaves increased in contaminated and recultivated areas after the red mud spill. The increase in Po-210 activity concentrations measured in tobacco leaves can be linked to the change in red mud storage technology in 2011 and the dry, windy weather conditions experienced in 2012. Our measurements confirm the suitability of tobacco as a biomonitor for monitoring atmospheric deposition and airborne dust. In other words, the leaf surface of the plant is suitable for risk assessment of NORM isotopes spread by atmospheric transport outside the soil. This is also supported by the lower transfer factors observed during the rainy growing season of 2014.

During the measurements, the atmospheric Rn-222 activity concentration near the soil surface (0.6 m) ranged from 9.32 to 92.06 Bq/m³ in 2012 and from 12.49 to 133.41 Bq/m³ in 2014. The increased values are probably due to changes in technological parameters and the formation of airborne dust caused by dry weather conditions. The findings indicate a moderate negative correlation between the Po-210 content of tobacco leaves and the environmental Rn-

222 level (2012: r = –0.43; 2014: r = –0.39). This suggests that the dominant process in the transport mechanism of NORM isotopes in the vicinity of red mud storage facilities is not gas-phase diffusion but solid-phase atmospheric deposition.

Tobacco plants have the potential to serve as biomonitoring indicators, facilitating the tracking of annual and distance-dependent changes in radionuclide contamination bound to atmospheric dust. Measurements of transfer factor indicate that, in conditions of dry and warm weather, airborne dust may have a significant impact on the uptake of Po-210, in addition to radionuclide spread from groundwater. However, it should be noted that measurements can be influenced by a number of variables, including the filtering and shielding effect of local forest belts. Therefore, it is of the utmost importance to select a representative sampling site.

The usability of NORMs in construction

As previously discussed, one potential area of application for NORM materials is their use as base or additive materials in the construction industry. We examined their applicability in the case of wood ash, fly ash, slag, red mud, and coal ash. The main gamma-emitting components of the samples were determined after sample preparation using a semiconductor HpGe detector gamma spectrometry method, while radon emanation and exhalation were determined using the accumulation method described earlier with the help of an AlphaGUARD radon monitor. The I-index value required for certification, as specified by the OAH regulation, was determined using the correlation described above. For the purpose of certification, we also ascertained the radium equivalent value of the materials that were tested and estimated the radiation exposure.

The natural radionuclide content of the raw materials examined was analysed using a semiconductor detector gamma spectrometry method, which is suitable for determining the activity concentration of Ra-226, Th-232, and K-40 with high accuracy. The measured activity concentration ranges were 9 ± 0.6 and 494 ± 25 Bq/kg for Ra-226, 1 ± 0.1 and 119 ± 8 Bq/kg for Th-232, and 24 ± 1 and 730 ± 28 Bq/kg for K-40. The results show that although the radionuclide content of some samples exceeded the global average soil values given in Radiation Protection 112 (50 Bq/kg for Ra-226 and Th-232, 500 Bq/kg for K-40), the majority of the samples tested were within the concentration ranges reported in the international literature.

Comparison with identical samples from different countries confirmed that the measured activity values do not differ significantly from published data for materials of similar geological and industrial origin. This finding lends strength to the interpretability of the radiological characteristics of the materials studied in an international context.

As part of the radioecological risk assessment process, we carried out measurements to determine the leaching of radionuclides from red mud, and therefore the mobility of the individual radionuclides themselves. During these tests, we performed leaching experiments in accordance with the mentioned protocols to determine the mobility of the different isotopes and their subsequent entry into the food chain. Following sample and source preparation, the samples were analysed using alpha spectrometry with a semiconductor (PIPS) detector. While the Tessier method and the MSZ-21470-50 standard are suitable for characterising red mud and similar materials, they do not provide sufficient information for radiological risk assessment on their own as no single test method can fully describe environmental risk. Using complex methods to estimate the fractions that can be absorbed by individual plants and the pH dependence of mobilisation can improve the accuracy of dose estimation.

Several radiation protection indicators were used to quantify the radiological risk, including radon exhalation and emanation rates, the absorbed gamma dose rate in indoor air, the annual effective dose of exposure and the I-index. The annual effective dose ranged from 0.1 to 3.6 mSv/year, with most samples remaining below the acceptable radiation protection limit. The lowest dose rate was associated with the cement sample, while the highest value was associated with the mine tailings sample due to radionuclide enrichment of geological origin.

Based on the I-index assessment, two samples – mine tailings and red mud – exceeded the recommended threshold of 1 mSv/year, indicating that the use of these materials in construction is only recommended in limited quantities or under appropriate technological control.

Correlation analysis showed a statistically significant positive correlation between radium concentration and radon mass and surface exhalation rates, which physically confirms that radon emissions are fundamentally dependent on radium content.

The results of the indoor dose exposure study showed that increasing the proportion of red mud, fly ash and mine tailings in the mixture leads to an increase in the external gamma dose rate and the radon-induced indoor dose exposure. Therefore, it is necessary to determine optimised mixing ratios for safe construction applications. When interpreting dose calculations,

the physical properties of building materials must be considered, such as density, porosity and diffusion coefficient. Radon emanation efficiency must also be considered, as well as the ventilation conditions of the living space and the position of people inside the room. These parameters significantly influence actual radiation exposure.

In conclusion, the potential building materials examined can be used safely in residential buildings from a radiological point of view, provided they are used at the correct concentrations and under technological control. However, it is essential to comply with regulatory limits and continuously monitor material properties affecting dose exposure.

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