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Natural Radioactivity of Solid Fuels and Ash-And-Slag Materials in the Context of their Environmental Impact

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Abstract. The indicators of natural radioactivity of solid fuels and ash-and-slag materials are examined in the context of assessing their environmental impact. The distribution of naturally occurring radionuclides in fuels and combustion products is analyzed based on gamma-spectrometric and neutron activation studies. It is shown that fuel combustion leads to a significant concentration of natural radionuclides in ash-and-slag materials, resulting in an increase in their specific activity compared to the parent fuel. Radionuclide concentration factors and effective specific activity values are calculated. An assessment of the potential radioecological burden is performed. Statistically significant correlations between rare-earth elements and radioactive nuclides are identified. The obtained results demonstrate the feasibility of using natural radioactivity indicators as an informative tool for environmental monitoring of coal-based power generation and for assessing anthropogenic impacts on the environment.

Keywords: ash-and-slag materials, solid fuels, naturally occurring radioactive elements, rare-earth elements, radioecological monitoring

Introduction

Coal-based power generation plays a key role in the energy balance of the Republic of Kazakhstan. A substantial share of electricity production is provided by the combustion of coal from the Ekibastuz and Karaganda basins, which results in a persistent anthropogenic impact on the environment. One of the most significant environmental factors associated with the operation of coal-fired thermal power plants is the generation and accumulation of ash-and-slag wastes containing a wide spectrum of chemical and radioactive components.

According to data from the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [1], the average concentrations of naturally occurring radionuclides in coal are as follows: U-238–16–110Bq/kg; Ra-226–17–60Bq/kg; Th-232 – 11–64 Bq/kg; K-40 – 40–850 Bq/kg. The concentrations of radioactive elements vary over a wide range depending on coal genesis, quality, and the compositional characteristics of both organic and mineral constituents. The issue of natural radioactivity of Kazakhstan's coals and the distribution of individual radionuclides remains insufficiently studied. In general, Kazakhstan's coals are characterized by low levels of radioactivity, with concentrations of naturally occurring radionuclides (U-238, Th-232, Ra-226, K-40) close to Clarke values [2,3].

According to the International Atomic Energy Agency (IAEA), the global total release of uranium and thorium during coal combustion amounts to approximately 37.3 t per year [4]. As reported in [5], more than 750 million tonnes of ash-and-slag waste have accumulated in ash disposal sites of Kazakhstan over several decades of coal-fired power plant operation. It can therefore be readily inferred that existing ash-and-slag materials are effectively transforming into quasi-technogenic deposits of radioactive elements and numerous rare metals, including toxic ones.

Ash-and-slag disposal sites of coal-fired power generation represent long-term sources of anthropogenic impact, capable of forming elevated radiation backgrounds in adjacent areas and affecting the atmosphere, soils, and surface waters. The radioecological hazard of such facilities is determined not only by the absolute values of radionuclide specific activity, but also by the scale of waste accumulation, storage conditions, and the potential for secondary dust formation and migration of radioactive components into the environment [6].

The degree of trace element enrichment in combustion products depends on their modes of occurrence in coal, the volatility of their oxides, and other compounds formed during the combustion process. Low-volatility compounds tend to accumulate in ash-and-slag wastes, whereas more volatile species are preferentially concentrated in emissions, particularly in fly ash [7].

Particular attention should be given to establishing the relationship between the contents of rare-earth elements (REEs) and naturally occurring radionuclides (NORs). Many REE-bearing minerals, such as monazite, xenotime, and bastnäsite, simultaneously incorporate uranium and thorium into their crystal structures [8]. Consequently, enrichment of coal and ash-and-slag materials with REEs is often accompanied by increased radioactivity. In addition to natural radionuclides, coals and ash-and-slag wastes contain a wide spectrum of metals, including rare-earth elements of both environmental and geochemical significance. During coal combustion, redistribution of chemical elements occurs, whereby rare-earth elements, as well as uranium and thorium, are preferentially concentrated in the mineral phase and subsequently accumulated in ash-and-slag wastes. The co-occurrence of rare-earth elements and naturally occurring radionuclides is controlled by their common geochemical hosts, primarily aluminosilicate, phosphate, and oxide minerals.

In this context, analysis of the relationship between the distribution of rare-earth elements and indicators of natural radioactivity provides deeper insight into element migration within the “coal–ash–environment” system and enhances the informativeness of radioecological assessments of coal-based power generation.

Despite the existence of individual studies addressing the natural radioactivity of coal and ash-and-slag wastes, there remains a need - under the conditions of Kazakhstan - for a comprehensive analysis of radiometric indicators specifically from the perspective of assessing the environmental impact of fuel-based power generation. This need is driven by both the scale of coal-fired electricity production and the specific characteristics of the high-ash coals utilized, which result in substantial accumulation of combustion wastes.

Accordingly, the use of natural radioactivity indicators of coal and ash-and-slag wastes as a tool for radioecological assessment is of practical importance for monitoring coal power facilities, analyzing environmental risks, and substantiating measures aimed at reducing anthropogenic impacts on the environment.

1. Materials and methods

Coal and ash-and-slag waste samples were collected at the Temirtau power plant and at the power plant located in the settlement of Topar, taking into account zones of technological impact. The Temirtau power plant utilizes coal from the Ekibastuz basin, while the Topar power plant operates on coal from the Borly deposit; accordingly, the samples in Tables 1 and 2 are labeled by the source coal deposit (Borly and Ekibastuz) rather than by the sampling site.

Gamma-spectrometric analysis of coal samples and ash-and-slag wastes with a mass of approximately 1 kg was performed at Ecoexpert LLP (Karaganda) using a CANBERRA spectrometer and Genie-2000 software. The specific activities of the radionuclides U-238, Ra-226, Th-232, and K-40 were determined. The measurement duration was approximately 5 h, ensuring minimal statistical uncertainty. Uranium and thorium activities were calculated based on the characteristic gamma lines of their daughter decay products. For the given measurement geometry (Marinelli beaker, sample mass of about 1 kg) and counting time, the minimum detectable activity (MDA) for K-40 was 23 Bq/kg; specific activities below this threshold are reported as '<23' in Table 1.

For a subset of samples, additional measurements were carried out using an MKS-01A “MULTIRAD” scintillation gamma spectrometer with PROGRESS software. The analysis was conducted for samples with a mass of about 1 kg over an energy range of 60–2000 keV. Instrument calibration was performed using standard Cs-137 and K-40 sources. The calculation of radionuclide specific activity was carried out in an automated mode in accordance with methodologies applied in radiation monitoring.

The recorded natural gamma-radiation spectra of coal and ash clearly exhibit the main photopeaks of radionuclides from the uranium and thorium decay series, as well as the single gamma line of K-40, confirming the reliability of identification and quantitative assessment of the natural radioactivity of the studied samples.

For a subset of samples, high-sensitivity neutron activation analysis was additionally applied (Laboratory of Tomsk Polytechnic University, Tomsk, Russia), enabling quantitative determination of naturally occurring radionuclides and associated trace elements. The samples were irradiated in a thermal neutron flux at a research nuclear reactor, followed by spectrometric measurement of the induced gamma activity and calculation of elemental concentrations through comparison with reference standards. The experimental workflow is illustrated in Figure 1.

Coal ash yield was determined using standard laboratory methods in accordance with applicable regulatory documents and ranged from 32.8 to 38.8%. To assess the redistribution of radionuclides during combustion, concentration factors were calculated as the ratio of the specific activity of a radionuclide in ash-and-slag wastes to its activity in the parent fuel.

Radioecological assessment was performed based on the calculation of the effective specific activity of ash-and-slag wastes, taking into account the contribution of the main naturally occurring radionuclides. In addition, a potential dose burden was evaluated using the calculated specific activity values, which made it possible to characterize the potential environmental impact of coal-based power generation [9]:

$$D=0.462A_{Ra}+0.604A_{Th}+0.042A_{K40}, \quad (1)$$

where D is the absorbed dose rate (nGy/h^{-1}), and A_{Ra}, A_{Th}, and A_{K40} are the specific activities of the corresponding radionuclides (Bq kg^{-1}).

The total content of light rare-earth elements (∑LREE) was calculated as the sum of the concentrations of La, Ce, Nd, and Sm. In addition, concentration factors and correlation coefficients between ∑LREE, Th, and U were calculated, with the level of statistical significance (p) being evaluated. These calculations enabled an analysis of the potential co-migration and mineralogical association of the elements, as well as the identification of radiogenic sources of REEs in the coal matter.

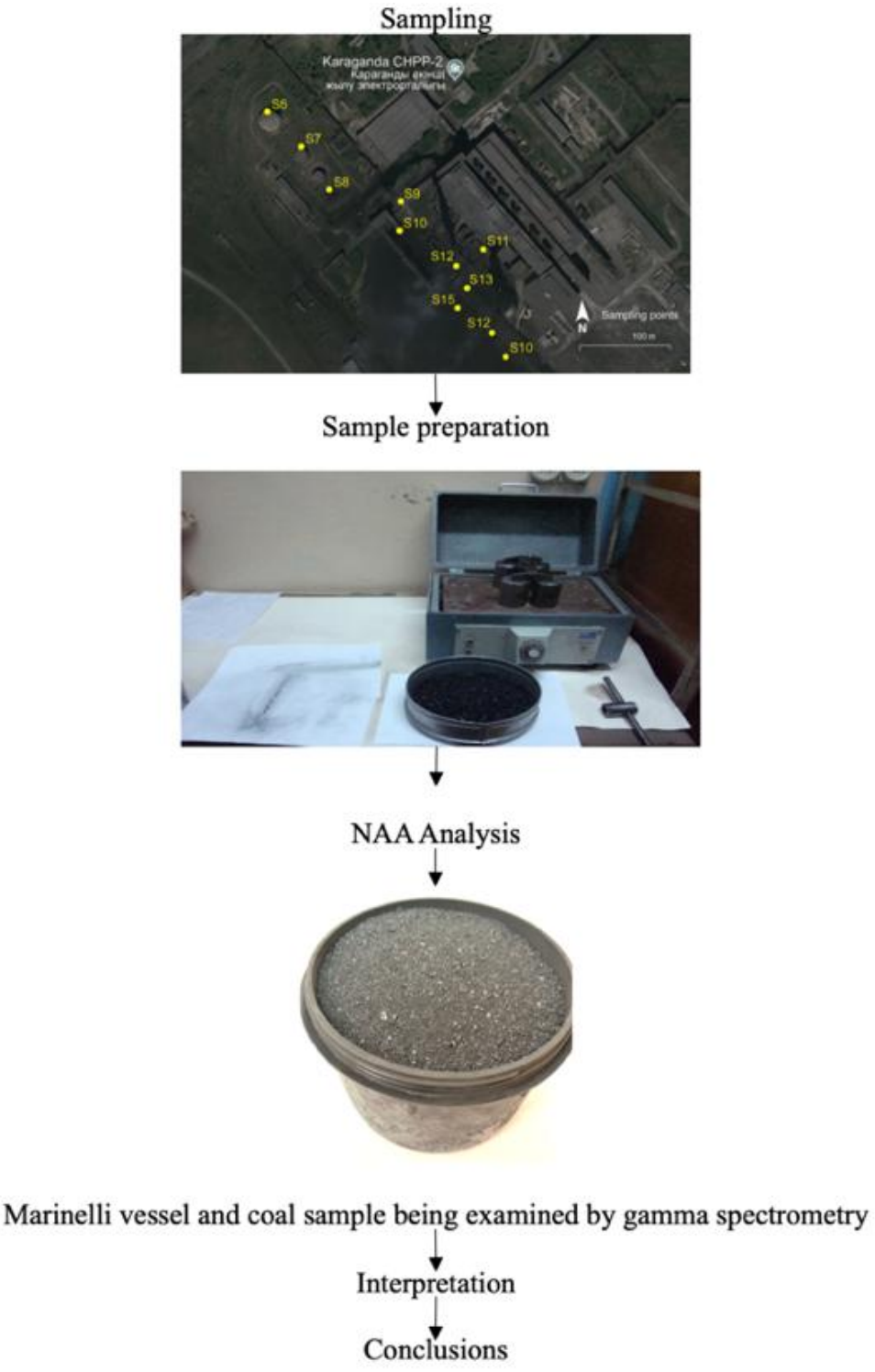

Fig. 1. – Schematic of the research workflow

2. Results

Gamma-spectrometric investigations showed that coals from the Ekibastuz and Borly coal basins are characterized by relatively low specific activities of naturally occurring radionuclides of the uranium and thorium decay series, as well as of the K-40 isotope. In most cases, the obtained values are comparable to background and global average levels typical of fossil coals, which allows the studied coals to be classified as low-radioactivity materials.

Data on the radioactivity of coals and ash-and-slag wastes obtained in this study are presented in Tables 1 and 2.

Table 1. Results of gamma-spectrometric analysis of natural radioactivity

Series	Radionuclide	Borly	Borly	Ekibastuz	Ekibastuz
		Bq/kg			
		Coal	Ash-and-slag materials	Coal	Ash-and-slag materials
U^{238}	U^{238}	26,2	68,2	17,5	33,5
	Th^{234}	26,2	68,2	17,5	33,5
	Pb^{214}	26,3	65,2	24,1	34,5
	Pb^{210}	43,3	75,1	18,3	24,3
	$Ra-^{226}$	22,0	64,2	18,4	32,9
	Bi^{214}	23,4	59,7	24,6	33,0
Th^{232}	Th^{232}	21,8	45,4	15,2	30,4
	Ac^{228}	21,8	45,4	15,2	30,4
	Pb^{212}	20,9	44,3	16,3	32,2
	Ra^{224}	20,1	42,8	17,0	30,2
	Bi^{212}	16,2	46,1	18,5	34,3
	Tl^{208}	7,2	15,2	5,3	11,1
	K^{40}	<23	113,0	97,0	144,0

Note: Note: '<23' denotes a K-40 specific activity below the minimum detectable activity (MDA = 23 Bq/kg) for the applied measurement conditions.

Table 2. Results of instrumental neutron activation analysis

Deposit	Number of samples	Sample	INAA			
			U, g/t	Th, g/t	K^{40}, Bq/kg	Th/U, g/t
Ekibastuz	11	Coal	0,8-1,5	2,6-3,6	61,4-87,6	3,3-2,4
	9	Ash-and-slag materials	4,2-8,2	11,8-17,9	112,1-243	2,8-2,9

The results of gamma-spectrometric and neutron activation analyses of coal indicate low levels of natural radioactivity. Radionuclide concentrations in coals from the studied deposits are close to global average values. At the same time, coal combustion products–ash-and-slag materials–exhibit significantly higher specific activities of naturally occurring radionuclides compared to the parent fuel. For all investigated sites, a consistent increase in the concentrations of U-238, Th-232, and K-40 in ash-and-slag materials was observed, which is attributed to the burnout of the organic matter of coal and the redistribution of radionuclides into the mineral phase. This effect confirms the well-established pattern of natural radionuclide concentration in solid combustion residues previously reported for coal-based power generation in other regions.

Radionuclide concentration factors, defined as the ratio of a radionuclide's specific activity in ash-and-slag materials to that in the parent coal, were calculated from the gamma-spectrometric data (Table 1). They amount to 2.6 (Borly) and 1.9 (Ekibastuz) for U-238, 2.9 and 1.8 for Ra-226, and 2.1 and 2.0 for Th-232; for K-40, the factor is 1.5 for the Ekibastuz samples, while for Borly only a lower-bound estimate (>4.9) is possible due to below the MDA in the parent coal (<23 Bq/kg). These values are consistent with the ash yield of the studied coals (32.8–38.8%), corresponding to a theoretical maximum enrichment factor of 2.6–3.0. Higher apparent factors derived from the non-paired INAA data (Table 2; ~4.5–5.5 for U and Th) should be regarded as indicative only.

A comparison with the worldwide average specific activities of naturally occurring radionuclides in soils (U-238 – 33 Bq/kg, Ra-226 – 32 Bq/kg, Th-232 – 45 Bq/kg, K-40 – 420 Bq/kg [1]) shows that the U-238 and Ra-226 activities in the Borly ash-and-slag materials exceed the average soil levels by approximately a factor of two, while the Th-232 activity is comparable to that of soils and the K-40 activity remains below the soil average. Although the absolute specific activities of ash-and-slag materials are only moderately elevated relative to soils, their radioecological significance is determined primarily by the large scale of waste accumulation, long-term open storage, and the potential for secondary dust formation and migration of radionuclides into adjacent environments.

The total content of light rare-earth elements (∑LREE) was determined as the sum of the concentrations of La, Ce, Nd, and Sm. In addition, concentration factors and correlation coefficients between ∑LREE, thorium, and uranium were calculated, with evaluation of their statistical significance (p). These calculations made it possible to analyze potential patterns of co-redistribution of the investigated elements and their mineralogical associations, as well as to assess the role of radiogenic components in shaping the rare-earth element composition of coals.

A significant correlation was observed between ∑LREE and uranium content ($r = 0.68$, $p = 0.022$), as well as between ∑LREE and thorium content ($r = 0.63$, $p = 0.038$). An even stronger correlation was identified between ∑LREE and the combined content of U + Th ($r = 0.75$, $p = 0.008$) (Figure 2).

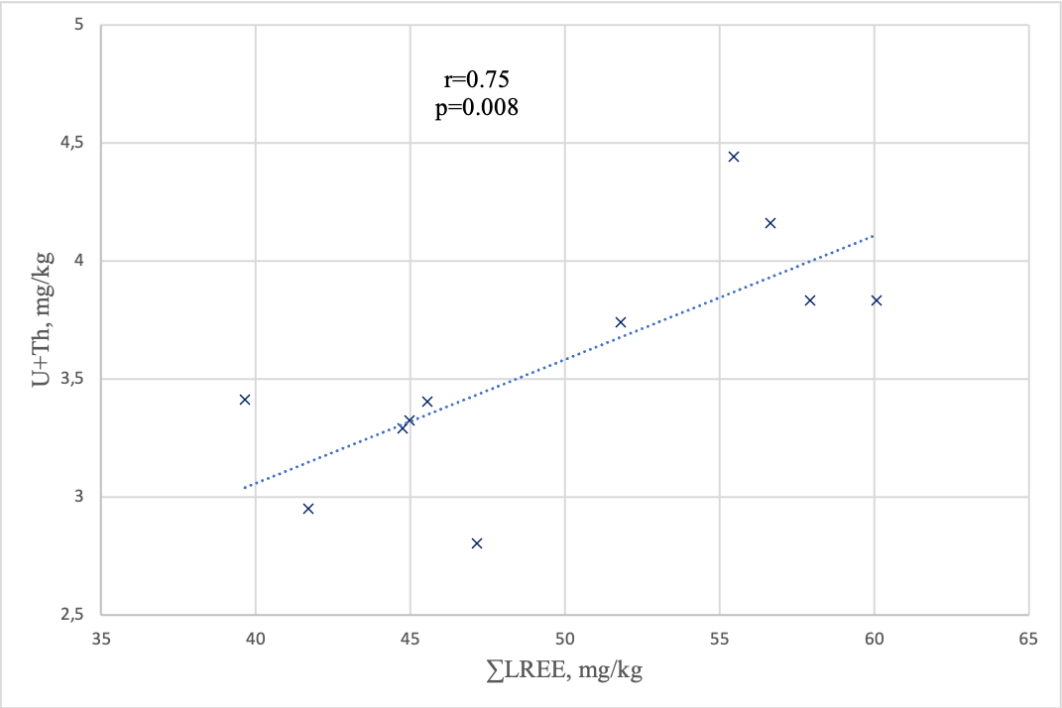

Fig. 2. – Relationship between the total uranium and thorium concentration and the total concentration of light REEs in coal

The assessment of potential dose burden based on the calculated specific activity values indicates that ash-and-slag disposal sites may serve as sources of additional external exposure in adjacent areas. The magnitude of this impact is governed not only by the radioactivity of the wastes but also by their operational conditions, the area of ash disposal sites, and the effectiveness of dust suppression and reclamation measures. Under conditions of long-term accumulation and open storage of ash-and-slag wastes, this factor becomes particularly important for environmental safety. The calculated absorbed dose rate D was 176 nGy h^{-1}, corresponding to an equivalent dose rate of 0.18 μSv h^{-1}, which results in an annual effective dose of 1.6 mSv. The obtained values slightly exceed global average levels and are comparable with the results of radioecological studies of coal in Poland [10], India [11], and Serbia [12].

Thus, regardless of the deposit or the type of coal used, ash-and-slag materials are characterized by significantly higher levels of natural radioactivity compared to the parent fuel. This fact is of fundamental importance for the radioecological assessment of fuel-based power generation, as ash-and-slag materials represent long-term sources of anthropogenic impact on the environment.

Conclusions

The studies have shown that the natural radioactivity of solid fuels and ash-and-slag materials serves as an informative tool for assessing the environmental impact of coal-based power generation. The investigated coals are characterized by relatively low levels of natural radioactivity corresponding to background values, whereas a significant concentration of naturally occurring radionuclides occurs in ash-and-slag materials.

It has been established that rare-earth elements exhibit geochemical affinity with elements of the uranium–thorium series, as evidenced by the identified correlations between the total content of light rare-earth elements and the concentrations of uranium and thorium. This indicates a preferential association of rare-earth elements with the mineral matter of coal and its combustion products.

Dose assessment has shown that ash-and-slag disposal sites can generate additional external gamma radiation in adjacent areas. The obtained dose values are comparable to those reported for coal-mining regions in other countries and indicate the necessity of considering the radiation factor in environmental monitoring of coal-based power facilities.

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