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DISTRIBUTION OF NATURAL RADIONUCLIDES IN ASHES, COLLECTED BY THERMAL POWER PLANTS ELECTROSTATIC PRECIPITATOR

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Relevance. *In the context of the global trend towards sustainable development, there is a problem of utilization of ash and slag waste from thermal power plants. The use of these wastes in the construction industry is of particular importance, due to the resource intensity of the production of building materials. Fly ash in the building materials composition can influence to a large extent the natural radioactivity of structures and increase the background radiation in the buildings. This is due to the fact that when coal combusting, fly ash is enriched with natural radionuclides.*
Purpose: *to study the radioactivity of fly ash deposited on various fields of the electrostatic precipitator.*
Object: *fly ash deposited on the fields of electrostatic precipitators from the combustion of Kansk-Achinsk brown coal in power boilers BKZ-420-140 PT-2.*
Methods. *Content of Ra-226, Th-232 and K-40 was determined by gamma-spectrometric method, the value of the normalized indicator of specific effective activity for fly ash was calculated. The granulometric composition of the studied samples was determined by the method of laser diffraction analysis. Cluster analysis was used for statistical processing of test results.*
Results. *The presence of two clusters in the content of natural radionuclides was established. In the first cluster, which has high radioactivity, the first and second fields of UGZ-4 electrostatic precipitators are combined, and in the second – the third and fourth fields. A correlation dependence was established between the numbers of fields of electrostatic precipitators, the size of ash particles, the content of Ra-226 and specific effective activity was established. No such dependence was found for Th-232.*
Conclusions. *Particle distribution in the fields of electrostatic precipitators is multimodal. The nature of the distribution of each mode approaches the lognormal law. The content of radium and the value of the specific effective activity depend on the size of ash particles and are described by an exponential function. The highest content of Ra-226 is observed in the ashes deposited on the third and fourth fields of electrostatic precipitators, which indicates the enrichment of finer ash particles with this radionuclide. The conclusions obtained correspond to the well-known thesis about the increased radioactivity of ash particles that are not deposited by ash collection systems and enter the atmosphere together with flue gases. The proposed method for studying the distribution of natural radionuclides over the fields of electrostatic precipitators can be used to predict the radioactivity of particles that are not captured by the flue gas cleaning system and released into the environment.*

Key words:
Fly ash, TPP, radiation hazard, natural radionuclides, building materials.

Introduction

Decree of the Government of the Russian Federation no. 1523-r on June 9, 2020 approved the Energy Strategy of the Russian Federation until 2035 [1]. The Strategy includes an increase in the share of disposed and neutralized waste in the fuel and energy complex industries from 52,6 % in 2018 to 85 % by the end of 2035, including solid fuel combustion products (ash slag), respectively from 8,4 to 50 %. Thus, Russian scientists will have to solve the issue of increasing the use of ash and slag waste by almost six times by 2035.

The largest consumer of waste from solid fuel combustion is the production of building materials, which uses a huge amount of natural resources. In world practice, there is a huge experience in the use of ash and slag. According to the American Coal Ash Association (ACAA), 59 % of the coal ash produced in 2020 was recycled compared to 52 % in 2019, and for the sixth year in a row, more than half of the coal ash produced in the United States is being used in industry. However, the overall level of processing at ACAA enterprises has declined over the past two years from the highest level of 64 % in 2017. This is associated with a decrease in the production of electricity using coal fuel and the deployment of new technological and logistics strategies [2, 3]. Such strategies are likely to develop in the direction of using this material in final products with a large surplus value.

Coal burning is a process of enrichment of combustion products in the form of slag and ash with harmful toxic elements, including natural radionuclides (NRN). Most of the ash is captured by electrofilters (96...99,7 %) and some is carried away together with flue gases into the environment and deposited on the adjacent territory.

It is quite obvious that the radioactivity of ashes and slags primarily depends on the content of radioactive elements in the burned coals. A detailed review and analysis of the results of scientific research in the field of coal radioactivity is presented in [4].

According to the authors [5], the uranium content in the slags remains approximately the same as in the accompanying rocks. Fly ash may contain a large amount of NRN. In [6], based on the analysis of domestic publications, it is stated that even with the combustion of coals with a low content of radioactive elements in their combustion products, the content of uranium and thorium in the fly ash increases by 3–4 times, and sometimes by 9 times in relation to the source fuel.

Studies [7] have established that in the ash collected from the electric filters of the smoke path of the Nazarovo power station, the concentration of uranium increased twice compared to the initial coal, and in thorium – by 4 times. It has also been established that, unlike thorium, which remains in the mineral part of the ash, uranium condenses on fine aerosols that are not captured by electrofilters. The concentration of uranium in the fly ash of electrofilters increases by 6 times; in the ash leaving together with flue gases by almost two orders of magnitude, on average – 71 times. The latter value was obtained in [5] by calculation, based on the balance of the uranium content during coal combustion.

In publication [8], using the example of burning coal in Germany, it is shown that the emission of uranium depends on the brand of coal. Uranium enrichment of the finest fraction of fly ash of black coals turned out to be significantly higher than for brown coals (twice versus 0,8), regardless of the method of their combustion. These data indirectly indicate that the uranium occurrence form in black and brown coals differ significantly [9].

In the research of German authors [10] it is stated that the concentration of radionuclides in brown coal ash is at the level of natural soils due to the very low concentration of radionuclides in coal. The authors state: the irradiation of workers dealing with coal ash at an electric power plant and at a landfill only slightly increased compared to the natural radiation background; the presence of ash dumps does not lead to significant additional irradiation of the population, and the use of by-products in building materials contributes a negligible proportion to the radiation dose received when living in dwellings.

In [11], the nature of the distribution of radionuclides, the coefficients of their relative enrichment and the effect of particle size on the enrichment of nuclides in the ash of electrofilters were studied. For the evaluation they used the relative enrichment factor (REF):

$$REF = \frac{A \cdot B}{C},$$

where A – ash radionuclide concentration, %; B – initial coal ash content, %; C – coal radionuclide concentration, %.

According to the authors, REF values depend mainly on the physical properties of the particle to which the radionuclide is attached after combustion. REF ues >1 indicate non-volatile elements. The coefficient gives the amount of enrichment with radionuclide, normalized for the ash content of the initial coal. Thus, this value gives a more realistic actual value of the enrichment than the value that depends only on the radionuclide. When calculating REF for six Indian thermal power plants, the ash content of the initial coal was assumed by them to be the same and equal to 40 %.

Table 1 presents the results of calculations of the ratio of comparative concentrations of NRN in ash to their content in coal, performed by us according to the data available in literature. Considering the large variability of the ash content of coals of various types and deposits (for example, the average ash content of brown coals of the Kansk-Achinsk basin for dry mass ranges from 6,0 to 12 % [12, 13], for comparability of data from various sources), we calculated REF in relation to the content of radionuclides without taking into account ash content.

Table 1. *Ratio of radionuclides content in ash to their content in coal*

Таблица 1. *Отношение содержания радионуклидов в золах к их содержанию в углях*

Radionuclide ratio Отношение по радионуклидам				Source Источник
U–238	Ra–226	Th–232	K–40	
6,6 (2,6–9,1)	13,1 (12,3–14)	6,1 (4,6–7,5)	5,8 (5,4–6,2)	[14]
9,4 (6,7–11,9)	–	–	–	[8]
–	2,6	2,1	2,8	[15]
4–20	–	4–20	–	[16]
8,0	–	7,4	5,4	[17]
10,7	–	3,5	3,6	[17]
3,8	–	6,6	–	[18]
6,0	–	–	–	[5]
2,0	–	4,0	–	[7]
5,6±2,1 (6,7)*	–	5,7±2,1 (6,4)*	–	[19]
4,98** (2,0–11,3)	4,46** (2,68–6,55)	3,38** (0,85–7,28)	2,36** (1–5,03)	[11]
5,5*	–	5,8*	–	[20]
11	–	9,7	–	[21]
9,4 (7,8–11)	11,7 (11,2–12,1)	9,1 (9,0–9,2)	7,3 (5,7–9,0)	[22] [23]
6,7	–	5,74	–	[24]
7,1±2,86	8,0±4,51	6,61±2,49	4,5±1,76	Average Среднее значение

* *Clark conversion to ash and enrichment coefficient/Пересчет по кларку на золу и коэффициент обогащения;*

** *Recalculation without ash content/Перерасчет без учета зольности.*

The average values of the ratio of specific activities of the NRN in ash and coal, calculated according to the data of the Table 1, were equal to: 7,1±2,86 for U-238; 6,7±4,57 for Ra-226; 6,1±2,66 for Th-232 and 4,0±1,38 for K-40. The Clark content of uranium and thorium for brown coals is 2,9±0,3 and 3,3±0,2 g/t, respectively [20, 21], and in ash: U=16±2 and Th=19±1 g/t. The calculated enrichment index for the Clark content of radionuclides in fly ash and coal turned out to be 11 for uranium and 9,7 for thorium. These indicators are higher than the average and closer to the maximum observed values (Table 1), but do not exceed the latter.

The close values of the ratio of contents for the NRN may indicate the same occurrence form of U and Th in coals. In the brown coals of the Azeyskoe deposit, uraninite was detected both in the organic matter of brown coal and in kaolinite. Single inclusions of coffinite were detected in the organic matter of coal, and the main form of thorium in coal is rare-earth phosphates and monazite. Monazite in ash is represented by amorphous aggregates, cores, films on an aluminosilicate matrix, as well as inclusions in aluminosilicate films [21].

It was convincingly shown in [25] that the bulk of uranium and thorium is accumulated in organic matter in sorbed form and in the form of strong humate complexes, and the main ash-forming aluminosilicate, silicate and carbonate minerals play a secondary role in the concentration of U and Th.

Thus, it can be stated with confidence that as a result of coal burning, the enrichment of the NRN fly ash occurs. The enrichment coefficient can reach one order of magnitude for the ash captured by electrofilters for uranium, radium and thorium. The enrichment coefficient for potassium is almost 1,5 times less than that of uranium (radium) and thorium.

From the point of view of the use of ash and slag materials in the production of the most massive products-cement concretes and mortars, separate selection of ash and slag is preferable. In this case, the additive technologies in concrete and mortar mixtures will not undergo significant changes. For fly ash, the available technological equipment for transportation, storage, feeding and dosing of cement is used, and for slag – inert aggregates.

Currently, work is becoming relevant to replace Portland cement with fly ash in concrete in the amount of 50 % or more [26]. This trend is driven by the need to reduce carbon emissions in cement production, which, according to the World Cement Association, accounts for about 7 % of global carbon emissions. It is quite obvious that in this case the technical and environmental requirements for the quality of fly ash increase significantly.

The separation of ash into fractions of a certain size and composition makes it possible to significantly expand the scope of their application, turning large-tonnage waste of thermal energy into valuable mineral raw materials of technogenic origin. At the same time, the product is enriched with both separate trace elements and NRN [5].

In [27], it is stated that the concentrations of Ra-226, Th-232 and K-40 activity in fly ash samples depend on the concentration of their activity in coal, the operating mode of the station's boiler during coal combustion, its capacity, origin and elemental composition of coal. It was also found that the specific radioactivity of fly ash increases with a decrease in the size of ash particles. Therefore, the released fly ash (having smaller particle sizes, compared to larger particles captured by environmental protection equipment at thermal power plants) has greater radioactivity than ash captured by filters of power plants [15, 28].

Fly ash collected in bunkers of different fields of electofilters has different properties. For example, the particle size decreases, and the specific surface area increases as the storage hopper is removed from the boiler, with an increase in the field number of the electrofilter. In most studies, different sizes were obtained either by air classification or by grinding a combined fly ash sample [29–33]. Only a few studies consider the use of fly ash taken from the fields of electrofilters [11, 33–36]. The NRN distribution research across the fields of the electrofilters has not been studied. When ash is mixed from different fields of electrofilters, as well as during subsequent air separation, obvious differences in the properties of fractions (differentiated by fields) are lost.

The purpose of this investigation was to study the radioactivity of the fly ash deposited on various fields of the electrofilter.

Methods

In this paper, the ashes of thermal power plants obtained by burning coal from the Borodino coal mine are investigated. Samples were taken simultaneously from the pre-chambers and from each field of the electrostatic precipitators of four similar boilers of the BKZ-420-140- PT-2. The designation of the samples is shown in the Fig. 1.

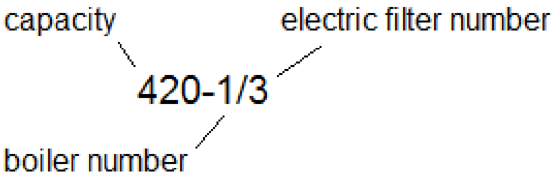

Fig. 1. *Designation of samples from different boiler and electric filters fields*

Рис. 1. *Обозначение образцов из разных котлов и полей электрофильтров*

When marking samples taken from the pre-chamber bunkers, the designation of the field number was replaced with the symbol «F».

The first three boilers are equipped with three-pole electric filters of the UG3-3–1774 type, the fourth is equipped with a six-pole electric filter of the EGBM type. The total number of samples taken is 18.

The content of radionuclides in the material was determined by gamma-spectrometric method using the MKS-AT1315 gamma-beta spectrometer in accordance with the methodology attached to the device. The fly ash samples, previously dried to a constant mass, were placed in a Marinelli vessel, where they were kept closed for 14–15 days. The exposure time for all samples was chosen the same – two hours.

The granulometric composition was determined using a FRITSCH ANALYSETTE 22 MicroTec PLUS laser particle analyzer. The device is certified according to the ISO 13320 standard. The method of determination was as follows: 0,8–1 g of the test material was injected into a closed circuit with 400 ml of water until the required concentration was reached. After its uniform distribution in the circuit, the laser complex sequentially measures the material under study. The fractional composition data is presented according to the fractional diagrams.

For the entire sample (18 samples), the point parameters of the empirical distribution of the normalized index of specific effective activity (A_{eff}) were calculated, a histogram was constructed, and a theoretical distribution law was selected.

The specific effective NRN activity of the product was calculated according to SanPiN 2.6.1.2523-09 (NRB-99) according to the formula:

$$A_{eff} = A_{Ra} + 1.3 \cdot A_{Th} + 0.09 \cdot A_K,$$

where A_{Ra}, A_{Th}, A_K are the specific activities of Ra-226, Th-232 and K-40, Bq/kg, respectively.

The purpose of the cluster analysis was to identify and combine into separate classes (groups) the most closely related in radionuclide composition of the deposited ash particles field numbers of electrofilters. The hierarchical clustering method was used to construct a vertical tree diagram and the K-average method. The measure of proximity determined by the Euclidean distance is a geometric distance in n-dimensional space and was calculated by the formula:

$$d(x, y) = \sqrt{\sum_{i=1}^{n}(x_i - y_i)^2}\ .$$

In order to exclude the influence of the electrofilter type on the results of statistical calculations and to ensure the reliability of the results obtained by the field number, fly ash samples taken from boilers equipped with the same UG3-3 electrofilters were subjected to cluster analysis: 420-1, 420-2, 420-3. Based on the visual analysis of the diagram and the target task of determining the maximum differences in the radionuclide composition of ash deposited by different numbers of fields of electrofilters, the number of clusters for which statistical indicators were calculated was assigned.

Results and discussion

The empirical distribution of the specific effective NRN activity one for four boilers approaches the normal law (Fig. 2).

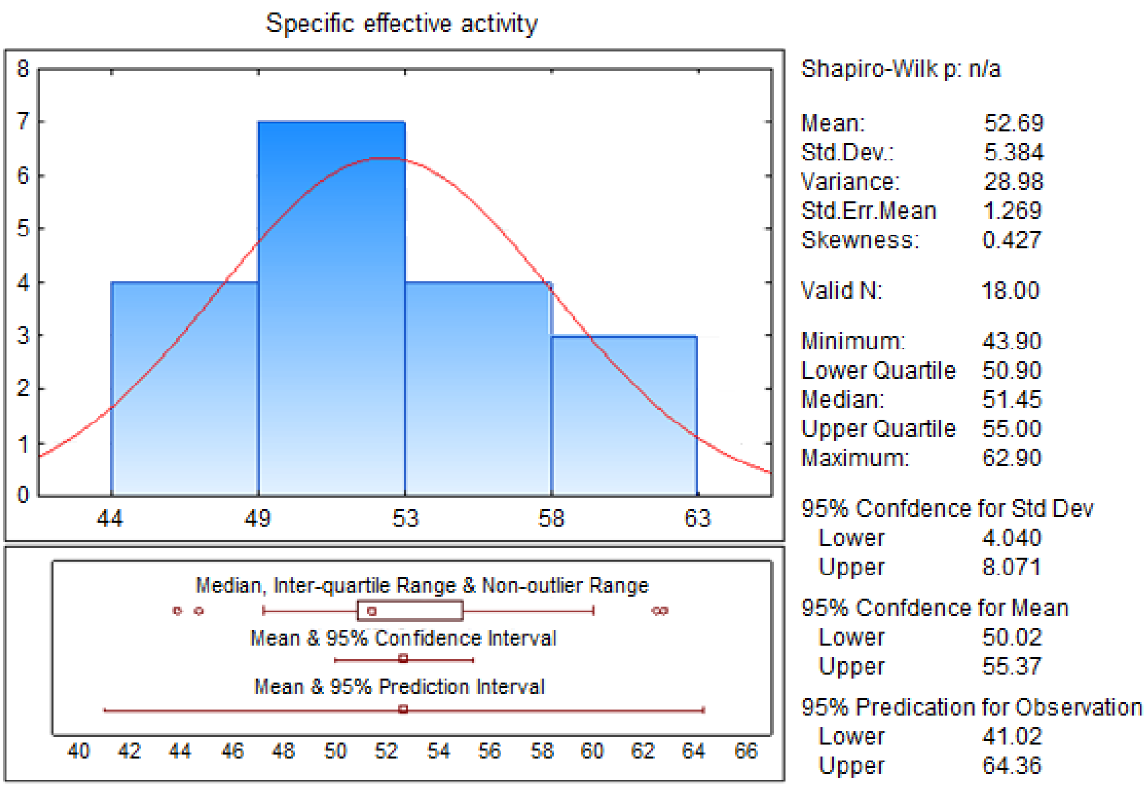

Fig. 2. *Histogram of the empirical and the function of the theoretical normal distribution of the specific effective activity in the ash of electrofilters*

Рис. 2. *Гистограмма эмпирического и функция теоретического нормального распределения удельной эффективной активности в золах электрофильтров*

The average value calculated from 18 samples turned out to be equal to A_{eff}=52,7±5,38 Bq/kg, respectively with the maximum and minimum observed values of the specific effective activity of 62,9 and 43,9 Bq/kg. The proximity of the median value of 51,5 Bq/kg to the average and the value of the asymmetry value with a positive sign indicate an approximation of the empirical distribution to normal theoretical with some deviation towards large values of A_{eff}. The predicted maximum value of the specific effective activity of natural radionuclides with 95 % confidence for the analyzed sample turned out to be 64,4 Bq/kg. The value of the coefficient of variation equal to 10,2 % allows us to state that the variability of the specific effective activity is closer to the insignificant degree.

The dendrogram of hierarchical clustering and the K-means method are presented respectively in Fig. 3 and in Table 2.

The most productive was the implementation of the hypothesis about the presence of the second clusters.

As it can be seen, the indicators of the pre-chambers and the first fields of the electrofilters are combined in Cluster 1, and the second and third fields equipped with the same types of UG3-3 electrofilters are combined in Cluster 2. The specific effective NRN activity was 1,17 times greater for fly ash deposited in the second and third fields. This effect is due to an increased value of the specific activity of the Ra product by 1,34 times.

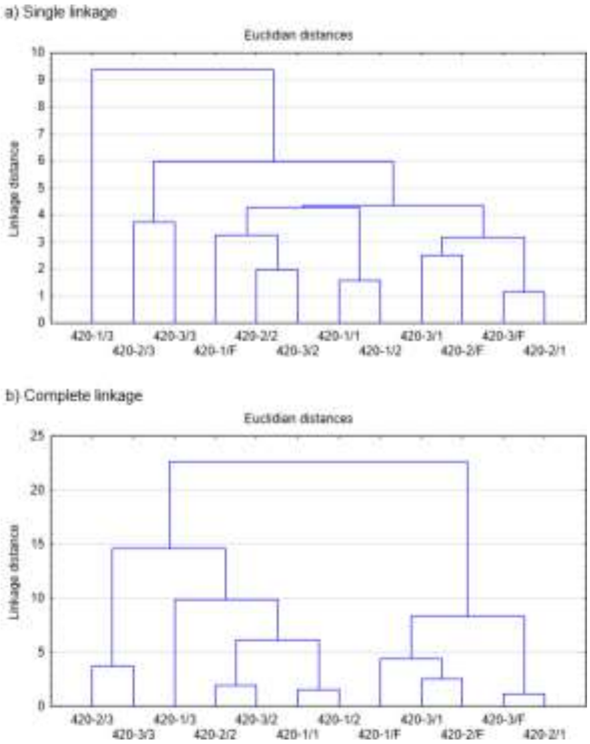

Fig. 3. *Dendrogram of hierarchical clustering according to the NRN specific activity in the ashes captured by various fields of the UG3-3 type electrofilters: a) clustering by single linkage method; b) clustering by complete linkage method*

Рис. 3. *Дендрограмма иерархической кластеризации по удельной активности естественных радионуклидов (ЕРН) в золах, улавливаемых различными полями электрофильтров типа УГ3-3: a) кластеризация методом одиночной связи; b) кластеризация методом полной связи*

The distribution of fly ash granulometric composition for almost all types of electrofilters turned out to be multimodal and approaching the logarithmic distribution law. Fig. 4 shows the size distributions of the diameters of the ash particles of the largest and smallest fraction of the 12 samples studied, taken from three boilers equipped with the same UG3-3 filters.

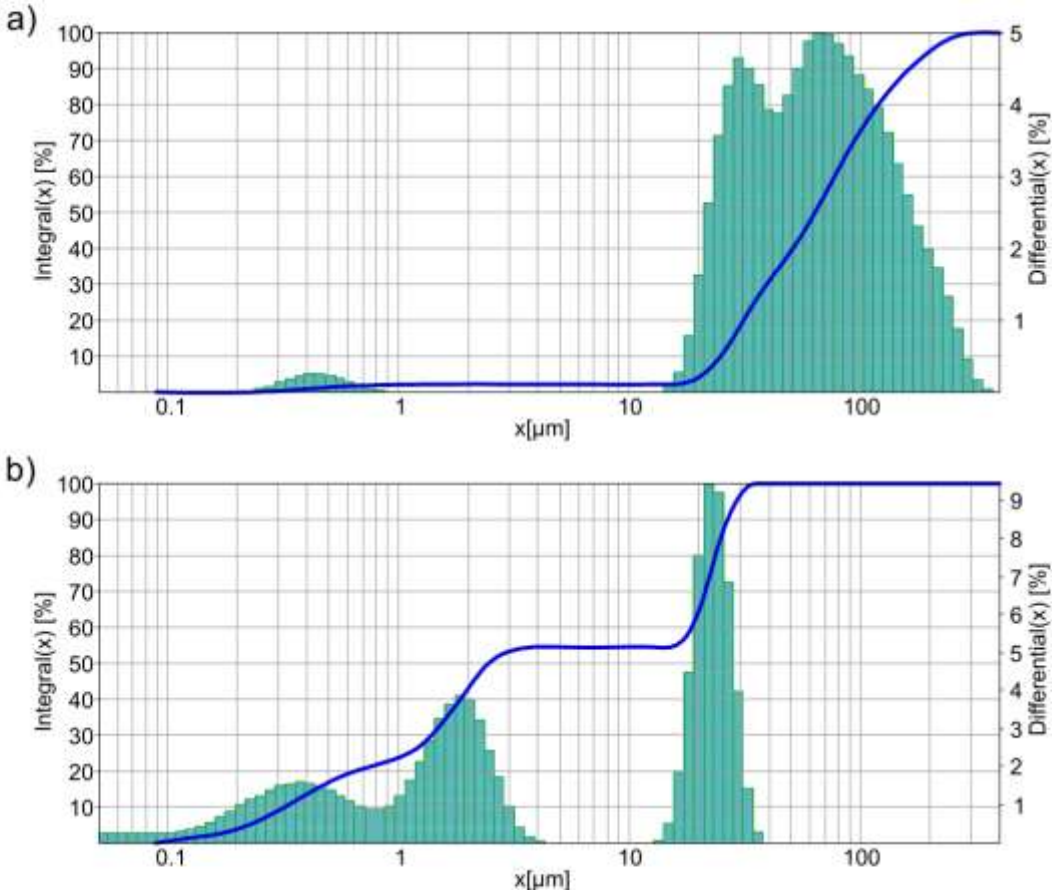

Fig. 4. *Granulometric composition of ash of: a) pre-chamber of the electric filter of the third boiler of the BKZ-420-3/F; b) the third field of the electric filter of the first boiler BKZ-420-1/3*

Рис. 4. *Гранулометрический состав золы: a) форкамера электрофильтра третьего котла БКЗ-420-3/F; b) третьего поля электрофильтра первого котла БКЗ-420-1/3*

The calculated average values of particle sizes for the first and second clusters were equal to 43,0±20,7 and 16,4±4,6 microns, respectively. The pre-chamber and the first field capture particles with an average diameter are 2,6 times larger than of the second and third fields. The variability of the values of the particles captured by the pre-chamber and the first fields is 1,7 times higher than of the second and the third fields. The ash captured by representative samples of the second cluster is much more homogeneous in its granulometric composition.

Table 2. *Selection location and cluster statistics*
Таблица 2. *Место отбора и статистика кластеров*

Indicator Показатель	Cluster 1: 420-1/**F**; 420-2/**F**; 420-2/**1**; 420-3/**F**; 420-3/**1** Кластер 1		
	Average Среднее	Standard deviation Стандартное отклонение	Dispersion Дисперсия
Ra-226	18,1	1,56	2,42
Th-232	21,9	1,22	1,48
K-40	20,0		
Indicator Показатель	Cluster 2: 420-1/1; 420-1/**2**; 420-1/**3**; 420-2/**2**; 420-2/**3**; 420-3/**2**; 420-3/**3** Кластер 2		
	Average Среднее	Standard deviation Стандартное отклонение	Dispersion Дисперсия
Ra-226	24,2	1,68	2,84
Th-232	23,5	3,06	9,34
K-40	21,3	3,40	11,6

The analysis of particle size changes averaged by the field numbers of the electric filters for three boilers shows a confident trend to decrease with an increase in the field number according to the power law (Fig. 5).

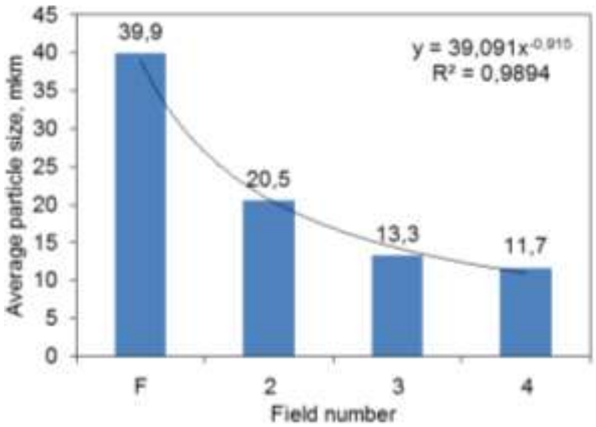

Fig. 5. *Reduction in the size of the ash particles of electric filters with an increase in the field number*

Рис. 5. *Уменьшение размеров частиц золы электрофильтров с повышением номера поля*

The relationship between radioactivity and the size of ash particles captured by electric filters is shown in Fig. 6.

It is clearly seen that a fairly dense correlation is observed between the content of radium and the specific effective activity with the size of ash particles. More radium atoms condense on smaller particles. No such connection is observed for thorium atoms.

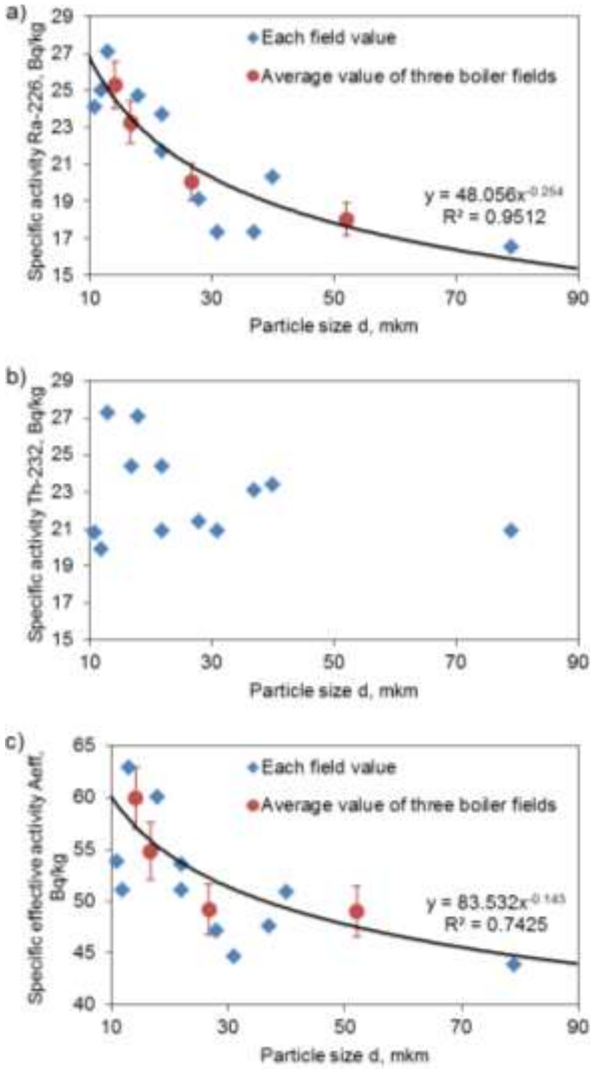

Fig. 6. *NRN content in individual fly ash fractions: a) Ra-226; b) Th-232; c) specific effective activity A_{eff}*

Рис. 6. *Содержание ЕРН в отдельных фракциях золы-уноса: a) Ra-226; b) Th-232; c) – удельная эффективная активность A_{eff}*

Conclusion

The analysis of the radioactivity of ash from the burning of brown coal of the Irsha-Borodinsky section deposited by electric filters of the UG3-3-1774 type and selected simultaneously from three boilers of the BKZ-420-140-PT-2 was carried out.

The empirical distribution of the A_{eff} value approaches the normal law.

The predicted maximum value of the specific effective activity of natural radionuclides with 95 % confidence for the analyzed sample turned out to be 64,4 Bq/kg, which is significantly lower than the normalized level for materials used for any types of construction – 370 Bq/kg.

The distribution of particles in the fields of electrofilters is multimodal. The nature of the distribution of each mode approaches the lognormal law.

The unevenness of NRN precipitation with an electrofilter is established. The radium content and the value of the specific effective activity depend on the size of the ash particles and are described by an exponential function. No such dependence has been established for thorium.

The highest content of Ra-226 is observed in the ash deposited on the third and fourth fields of the electrofilters. This indicates the enrichment of smaller ash particles with this radionuclide. The obtained conclusion corresponds to the thesis about the increased radioactivity of ash particles that are not deposited by ash collection systems and enter the atmosphere together with flue gases.

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РАСПРЕДЕЛЕНИЕ ЕСТЕСТВЕННЫХ РАДИОНУКЛИДОВ В ЗОЛАХ, УЛАВЛИВАЕМЫХ ЭЛЕКТРОФИЛЬТРАМИ ТЕПЛОВЫХ ЭЛЕКТРОСТАНЦИЙ

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Актуальность. *В условиях общемировой тенденции к устойчивому развитию существует проблема утилизации золошлаковых отходов тепловых электростанций. Использование этих отходов в строительной отрасли имеет особое значение в связи с ресурсоемкостью производства строительных материалов. Зола-унос в составе строительных материалов может в значительной степени влиять на естественную радиоактивность конструкций и повышать радиационный фон в зданиях. Это связано с тем, что в процессе сжигания угля летучая зола обогащается природными радионуклидами.*
Цель: *изучение радиоактивности золы-уноса, осаждаемой на различных полях электрофильтра.*
Объект: *зола-унос, осажденная на полях электрофильтров от сжигания Канско-Ачинских бурых углей в энергетических котлоагрегатах БКЗ-420-140 ПТ-2.*
Методы. *Гамма-спектрометрическим методом определено содержание Ra-226, Th-232 и K-40, рассчитано значение нормируемого показателя удельной эффективной активности для золы-уноса. Методом лазерного дифракционного анализа определен гранулометрический состав исследуемых проб. Для статистической обработки результатов испытаний применялся кластерный анализ.*
Результаты. *Установлено наличие двух кластеров по содержанию естественных радионуклидов. В первый кластер, имеющий большую радиоактивность, объедены первые и вторые, а во второй – третьи и четвертые поля электрофильтров УГЗ-4. Установлена корреляционная зависимость между номерами полей электрофильтров, размером зольных частиц, содержанием Ra-226 и удельной эффективной активностью. Для Th-232 такой зависимости не обнаружено.*
Выводы. *Распределение частиц на полях электрофильтров является мультимодальным. Характер распределения каждой моды приближается к логнормальному закону. Содержание радия и значение удельной эффективной активности зависят от размеров частиц золы и описываются показательной функцией. Наибольшее содержание Ra-226 наблюдается в золах, осажденных на третьем и четвертном полях электрофильтров, что свидетельствует об обогащении этим радионуклидом более мелких зольных частиц. Полученные выводы соответствуют известному тезису о повышенной радиоактивности частиц золы, которые не осаждаются системами золоулавливания и попадают в атмосферу вместе с дымовыми газами. Предложенная методика исследования распределения естественных радионуклидов по полям электрофильтров может быть использована для прогнозирования радиоактивности частиц, не улавливаемых системой очистки дымовых газов и выбрасываемых в окружающую среду.*

Ключевые слова:
Золы-уноса, ТЭС, радиационная опасность, естественные радионуклиды, строительные материалы.

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