THE STUDY OF THE RADIOECOLOGICAL SITUATION IN THE ENVIRONMENT SURROUNDINGS THE COAL POWER PLANT

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The results of analyzing of the radioecological situation near the TPP-4, Ulan Bator, Mongolia are presented in this work. Content of radioisotopes (U- and Th- series, ^{40}K, ^{137}Cs) at the coal, bottom ash, fly ash samples as well as soil and plants using gamma-activation analysis and gamma-spectrometry. Specific effective activity (A_{eff}) of ash wastes was calculated.

Keywords: brown coal, bottom ash, fly ash, thermal power plant, radionuclides, Mongolia, pollution, environment, soil, plant

В работе представлены результаты анализа радиоэкологической обстановки в районе ТЭС-4 г. Улан-Батор, Монголия. Определено содержание радиоизотопов (U- и Th- рядов, ^{40}K, ^{137}Cs) в образцах углей, шлака, золы-уноса, а также пробах почвы и растительности гамма-активационным и гамма-спектрометрическим методами анализа. Рассчитана удельная эффективная активность (Аэфф) золошлаковых отходов.

Ключевые слова: бурый уголь, шлак, зола-уноса, тепловая электростанция, радионуклиды, Монголия, загрязнение, окружающая среда, почва, растительность

INTRODUCTION

It is believed, that the main sources of the radionuclides input to the environment are the nuclear power plants (NPP). The Chernobyl and Fukushima Daiichi nuclear power plants accidents were contributed to negativity position about the NPP [1]. However, the thermal power plants (TPP) are also a source of radionuclides into the environment [2-5] and its can be danger to the health of human living surrounding this plants. Besides carbon and organic matter, coal contains the large number of different elements. Moreover, brown coals are much more enriched with uranium and thorium than the hard ones [6]. Trace elements can be emitted to the environment during coal combustion and become a hazard for the human health. One of the most dangerous radionuclide of the ^{238}U-series is ^{210}Pb. Radioactive ^{210}Pb belongs to the particularly high degree of radiotoxicity group of isotopes. ^{210}Pb is an osteotropic radionuclide that becomes a source of accumulation of equilibrium amounts of ^{210}Bi beta-radiation and ^{210}Po alpha-radiation [7]. The amount of radionuclides depends on their content in coal, and also its preparation and burn technology. Enriched by volatile fine ash fraction is not completely caught by industrial filters. Pollutants contained in the emissions are gradually settled on a surface of soil and plants. Soil is an indicator of the geochemical situation. The upper horizon of soil carries basic information about technogenic impact. Plants are penetrated by heavy metals from roots and surface of leaves. Therefore, chemical composition of the plants reflects element complex of the soils and aerosol pollutants.

The most of the population of the Mongolia (1.4 million people) is concentrated in its capital Ulan Bator which territory with the included in its administrative boundaries aimags is 4704.4km^2. Geoclimatic conditions of Ulan Bator contribute to the accumulation of pollutants in the air [8].

OBJECT AND METHOD

The thermal power plant (TPP-4) that situated in the city Ulan Bator – the largest energy source in Mongolia – was chosen as the object of this study. This power plant uses the brown coal from the nearby Mongolian deposits Baganur and Shive-Ovo. The plant uses about 2.4 million tons of coal per year. Coal contains the natural occurred radionuclides of the U- and Th-series and also the long-lived ^{40}K. During the coal combustion, the bottom ash, as well as, fly ash are producing. Although at the TPP-4 is used the electrostatic precipitator type EGA-2-58-12-6-4 for purification stack gases with a degree of cleaning from fly ash up to 99%, the fine fraction of this fly ash discharge into atmosphere as stack gases. In this way, the thermal power plant (TPP) is can be a source of radionuclides at the environment and can be more dangerous for local population health then nuclear power plants (NPP).

The aim of this work was to analyze radioecological situation in the operating coal power plant surrounding environment. In 2018 year the samples of coal, bottom ash, fly ash were taken from the TPP-4, the soil and plant were sampled from different distances of the object. All samples were air-dried at 105 ^{o}C, grinded, homogenized at agate mortar and prepared for further analysis according to the methodology [9]. Analysis of all samples was carried out at the Flerov Laboratory of Nuclear Reactions Joint Institute for Nuclear Research (FLNR JINR, Dubna) by using gamma activation analyses (GAA) for definition of U and Th contents and gamma spectrometry for specific natural activity registration. The analyze samples were irradiated by the 22 MeV bremsstrahlung (the average electron current 10 μA) for 4 h on the MT-25 microtron. Irradiated samples were measured with use of the HP Ge detector Canberra with FWHM resolution 1.5 keV and efficiency of 1% for the 1332.5 keV photons of ^{60}Co（energy range to 3000 keV). Measurement times were 300, 900 and 3600 s. For determination of thorium each irradiated sample also was measured by a semiconductor Si(Li) detector from XRF spectrometer with 145 eV resolution on Fe line (6.4 keV). Gamma-spectrometric measurement of the samples for determination natural occurred radionuclides and ^{137}Cs were carried out with use of the HPGe and also thin Ge detector Canberra with FWHM resolution 0.55 keV and efficiency of 7% for the 122 keV photon line (energy range to 300 keV).

RESULTS AND DISCUSSION

Proceeding on the coal composition and the characteristics of the gas purification system at the TPP-4 it was calculated that about 2.4 thousand tons of different substances emission into atmospheric air from observed object annually. The results of measuring uranium and thorium concentration in the samples of coal, bottom and fly ash are shown in the table 1.

Table 1. Uranium and thorium concentration in the samples of coal, bottom and fly ash, g/t

Sample	U	Th
coal	5.2 ± 0.1	2.5 ± 0.1
bottom ash	19.3 ± 0.3	10.6 ± 0.3
fly ash	22.5 ± 0.4	14.6 ± 0.4
bottom ash/ coal	3.7	4.2
fly ash/ coal	4.3	5.8

Analysis of the data showed that the using at TPP-4 coals are significantly enriched with uranium twice as much, than accepted average world value for brown coals (2.7 ± 0.3 g/t). The content of thorium in the coal was lower than the clarke value (3.8 ± 0.2 g/t). During coal combustion, uranium and thorium are concentrated in the ash slag. It was found that uranium in concentrations 1.5 times more than global average value for ashes of brown coals (15 ± 1 g/t), at the same time 54% was concentrated in fly ash. Thorium was found in concentrations less than the clarke value (19 ± 1 g/t), 58% was concentrated in the fly ash just like as uranium [10]. It is noted, that uranium and thorium contents in bottom and fly ash at TPP-4 have the same order of values as in work [2]. There are no data on background and maximum allowable concentration of uranium and thorium for the soil of Mongolia, so for comparison were used clarks by Vinogradov [11]: U - 1 mg/kg, Th - 6 mg/kg. Analysis is showed exceeding clarke of uranium by 2.4-3 times and thorium by 1.3-1.9 times at the area around of this power plant. Directly near the TPP-4 were observed maximum uranium and thorium concentrations at the soil (3.0 ± 0.2 mg/kg and 11.4 ± 0.5 mg/kg, respectively). Decreasing of the concentrations of this elements followed by distancing from the object was founded.

Maximum concentration of uranium in herbal mixture (1.1 ± 0.3 mg/kg) was found in sample selected directly near the TPP-4, and then a decrease in concentration occurs. The maximum concentration of thorium (4.3 ± 0.8) was observed at a 10 km distance from the power plant, the minimum – at 5 km distance.

Specific activities of uranium and thorium (Bq/kg) for all samples based on determined content (g/t) were calculated by ^{238}U, whose content in natural uranium is 99.3%, and ^{232}Th, whose isotopic prevalence is almost 100% (table 1). The results of radionuclides specific activity in the samples of coal, bottom and fly ash, as well as, soils and plants sampled at different distances from TPP-4 are shown in table 2.

Table 2. Content of radionuclides in the coal, bottom and fly ash samples and selected from different distances from TPP-4 soil and plant samples, Bq/kg

Sample	Radionuclides					
	calculation		γ- spectrometry			
	^{238}U	^{232}Th	^{226}Ra	^{210}Pb	^{40}K	^{137}Cs
coal	62.5 ± 12.5	8 ± 4	39 ± 4	10 ± 2	38 ± 5	0.5 ± 0.1
bottom ash	237.5 ± 12.5	45 ± 4	305 ± 15	31 ± 4	170 ± 14	≤ 0.1
fly ash	275 ± 12.5	53 ± 4	317 ± 18	118 ± 4	172 ± 15	≤ 0.1
soil, 1 km	37.5 ± 12.5	45 ± 4	18 ± 5	14 ± 3	696 ± 16	1.5 ± 0.2
soil, 5 km	37.5 ± 12.5	33 ± 4	21 ± 6	5 ± 3	567 ± 18	1.8 ± 0.2
soil, 10 km	37.5 ± 12.5	37 ± 4	36 ± 2	14 ± 3	559 ± 11	2.1 ± 0.2
herbal mix, 1 km	7.5 ± 3.8	≤ 4	20 ± 0.5	34 ± 4	212 ± 14	≤ 0.1
herbal mix, 5 km	10.0 ± 3.8	8 ± 4	≤ 0.5	28 ± 6	138 ± 18	≤ 0.1
herbal mix, 10 km	≤ 3.8	16 ± 4	≤ 0.5	6 ± 4	479 ± 12	≤ 0.1

Significant content of long-lived isotopes of the uranium series (^{226}Ra and ^{210}Pb) was found in the fly ash. Specific activity of ^{226}Ra in fly ash was 7.8 times higher than its content in the coal, and ^{210}Pb – 12 times. The gamma spectrum of natural activity in the fly ash sample is shown in figure 1.

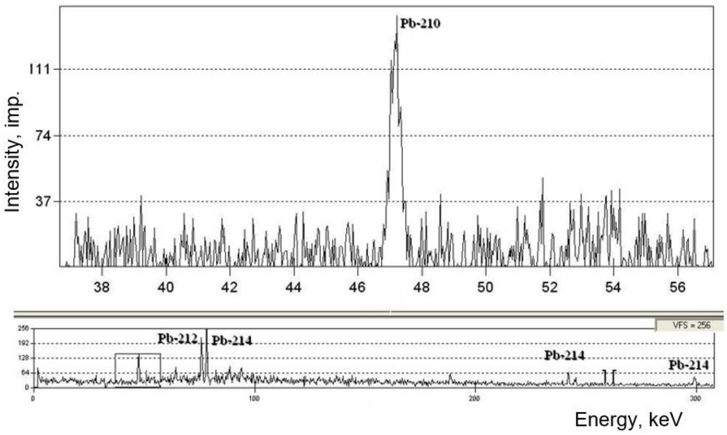

Figure 1. Gamma spectrum of natural activity of fly ash sample measured by 15 h.

Finest particles with high content of radionuclide ^{210}Pb might slip through the gas purification equipment of the TPP and become a cause to an increase ^{210}Pb concentration in the air and possibility pollution the soil. Specific activity of ^{210}Pb at soil samples varies from 5 to 14 Bq/kg while specific activity of ^{210}Pb at herbal mixture samples was 6-34 Bq/kg, higher comparison at soil samples. This fact allows to suppose the aerogenic source of ^{210}Pb release into the environment. Specific activity of ^{137}Cs in the samples of coal, bottom and fly ash and in the samples of plants was at detection limit level. The specific activity of ^{137}Cs in the soils have been determined from 1.5 to 2.1 Bq/kg. ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K contents at soil and herbs are similar to equivalent data reported around in the literature. Mongolian soils and herbs are less polluted with ^{137}Cs than European ones [3-5]. Content of ^{210}Pb obtained in soil samples are lower than at [3-5]; however, its excess in the herbal samples comparing with [4] was noted.

Specific effective activity (A_{eff}) for fly ash, bottom ash and their mixture 3:1 (ash slag wastes) was calculated to assess the possibility of using those waste as a secondary raw material for construction [12]. The specific effective activity for fly ash was 422.6 Bg/kg, for bottom ash – 325.4 Bg/kg and their mixture – 398.3 Bg/kg. Fly ash and ash slag wastes (mixture 3:1) belong to class II materials and cannot use for all type construction. This fact needs to pay attention in when using ash dump materials for construction within settlements.

CONCLUSION

In order to reduce the radiation impact from the thermal power plant № 4 in Ulan Bator it is need to organize close control over the content of natural radionuclides, such as U- and Th-series, ^{40}K and technogenic radionuclide ^{137}Cs, in the using brown coal and their combustion products (bottom ash, fly ash) and also monitoring in soil and plants. It is need to pay attention the content of radionuclides in the fly and bottom ash when using ash slag wastes in construction.

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