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Studying the Distribution of Toxic Elements in Ash-and-Slag Wastes from the Enterprises of the Fuel-and-Energy Complex of the Kemerovo Region

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Abstract

Toxic element distribution in ash-and-slag wastes from the fuel-and-energy complex of the enterprises of Kemerovo Region was studied. The mobile species of copper, nickel, zinc, lead are revealed to migrate into the groundwater. It is demonstrated that the ash-and-slag wastes in the case of contacting with water become a source of toxic elements (vanadium, molybdenum, arsenic, nickel, zinc, manganese and chromium). The content of these elements in groundwater in the territory of ash-and-slag dumps and adjacent natural water sources significantly exceeds the maximum permissible concentration thereof in water.

Key words: ash-and-slag waste, toxic elements, gross forms of elements, mobile forms of elements, water-soluble forms of elements

INTRODUCTION

Energy engineering is the leading branch of modern industry, but at the same time it represents a source of a significant biosphere contamination. Alongside with gaseous emissions, the combustion of solid fuels under the conditions of thermal power plants and boiler plants is accompanied by the formation of ash and slag stored as ash-and-slag dumps. In the Kemerovo Region in 2010 there have been 3 318 517 thousand tons of ash-and-slag wastes accumulated, whose prevailing amount is stockpiled [1]. Among the nine TPP in Kuzbass, only three ones have installed units for dry ash withdrawal (the Tom'-Usinsk TPP, with dry ash withdrawal capacity of 20 thousand tons/year, the West Siberian TPP with the capacity of 100 tons/year, the Kemerovo TPP with the capacity of 30 tons/year). More than 2 million tons of wastes is transported to dumps by water together with slag in the form of ash-and-slag mixtures [2].

The chemical composition of ash-and-slag wastes is by 80–90 % presented by oxides such as SiO_2, Al_2O_3, FeO, Fe_2O_3, CaO, MgO. In addition, the waste composition involves the residues of unburned coal particles (0.5–20 %), and trace impurities such as manganese, vanadium, lead, chromium compounds *etc*. [3, 4]. The elements whose content in coal and coal ash is higher than Clarke values are called typomorphic, or characteristic ones. They include gold, selenium, mercury, germanium, arsenic, *etc*. [5, 6]. The literature is mainly devoted to studying the total content of trace elements, whereas mobile and soluble species remain unexplored. However, just the mobile and soluble special forms of trace elements are important for assessing the environmental impact of the elements with respect to the territory.

A reasonable assigning the list of trace elements for environmental monitoring in the territory of a particular coal basin is of great practical importance. In particular, for an objective assessment of the impact of ash-and-slag

TABLE 1

Content of gross, mobile and soluble special forms for the toxic elements in the ash-and-slag wastes and boiler slag of the Kemerovo Region, mg/kg

Elements	Boiler-plant slag		Ash-and-slag wastes		MPC
	Sample No. 1	Sample No. 2	Sample No. 3	Sample No. 4	
			Gross forms		
Vanadium	22.40	44.80	67.20	50.10	150.0
Cadmium	<0.05	<0.05	<0.05	<0.05	0.5*
Cobalt	6.02	10.60	6.26	8.30	–
Manganese	2246.0	1250.0	6585.2	920.0	1500.0
Copper	29.00	41.00	20.70	16.30	33.0*
Molybdenum	4.10	4.80	2.10	3.30	–
Arsenic	<0.1	5.00	6.00	13.00	2.0
Nickel	30.00	40.00	39.30	20.20	20.0
Tin	<0.1	<0.1	2.20	1.50	–
Mercury	0.91	0.29	<0.1	0.60	2.1
Lead	26.00	92.00	66.00	16.00	32.0*
Antimony	1.31	1.13	2.10	0.72	4.5
Chromium	77.60	83.00	69.00	60.00	–
Zinc	21.00	41.00	16.10	28.40	55.0*
			Mobile forms		
Cobalt	0.08	0.12	0.16	0.10	5.0
Manganese	620.0	190.0	3150.0	460.0	300–700
Copper	10.00	26.00	20.00	12.00	3.0
Molybdenum	0.20	0.80	0.60	1.00	–
Nickel	24.00	29.00	12.00	11.00	4.0
Lead	18.00	85.00	32.00	6.20	6.0
Chromium	0.43	0.52	1.34	1.90	6.0
Zinc	12.00	35.00	5.10	10.00	23.0
			Water-soluble forms		
Vanadium	0.0940	<0.0005	<0.0005	0.0200	0.001
Cadmium	<0.00001	0.00009	0.00008	<0.00001	0.005
Cobalt	<0.0002	0.0007	0.0002	<0.0002	0.01
Manganese	0.0020	0.0270	0.1400	0.0510	0.01
Copper	0.0025	0.0080	0.0030	0.0035	0.001
Molybdenum	0.1900	0.0210	0.0018	0.0350	0.0010
Arsenic	0.0130	0.0081	<0.005	0.0100	0.05
Nickel	0.0090	<0.0002	0.0100	<0.0002	0.01
Mercury	<0.0001	<0.0001	<0.0001	<0.0001	0.00001
Antimony	0.0056	0.0055	<0.0005	0.0102	0.05
Lead	<0.0002	<0.0002	<0.0002	<0.0002	0.006
Chromium	0.0019	0.0053	0.0021	0.0105	0.02
Zinc	<0.005	0.0110	0.0490	0.0132	0.01

Notes. 1. For gross and mobile forms the MPC values for soil (MPC_s) are presented, mg/kg [18–20], for water-soluble species the MPC values for water (MPC_w) are used, mg/dm^3 [21]. 2. Dash means that there are no standard data required for MPC.

* Approximately permissible concentration.

wastes on the environment in the territory of the Kemerovo Region it is necessary to develop programs for eco-analytical monitoring, including a definite list of toxic elements.

The purpose of this work consisted in studying the distribution of toxic elements in ash-and-slag wastes presented by gross, mobile and soluble special forms as well as in assessment of a potential hazard of ash-and-slag dumps.

EXPERIMENTAL

In accordance with the purpose posed, as the objects of investigation we chose: ash-and-slag wastes (a mixture ash and slag), slag taken from the boiler plants of the Kemerovo Region, groundwater and surface water in the territory of ash dumps, as well as soil taken from the areas adjacent to ash dumps.

Sampling the ash-and-slag wastes was performed in accordance with a normative document [7] that establishes general requirements for sampling a representative portion of mineral wastes. Soil sampling was carried out according to the techniques described in [8, 9]. Sampling the surface water and groundwater was performed according to [10].

The concentration of the elements in the samples was determined by means of the following analytical methods: atomic emission spectroscopy with inductively coupled plasma (Liberty Series II, (Varian, USA)), iCAP 6300 Duo (Thermo Scientific, UK); atomic absorption spectroscopy with electrothermal atomization (SpectrAA-640z, (Varian, USA)); atomic absorption spectroscopy with the use of a “Hg cold vapor” technique (Mercury analyser M-6000A, (CETAC Technologies Inc., USA)) according to certified methods [11–16]. Sample preparation for the determination of the total content of the elements in the samples of ash-and-slag wastes and soil was carried out within sealed plastic containers for the decomposition with a mixture of nitric and hydrochloric acids using a Hot-Block heating platform (Environmental Express, UK). The determination of the mobile species of metals was carried out after conditioning the sample under investigation during 24 h in ammonium acetate buffered solution at pH 4.8 at a room temperature. Prior to the determination of elements in water the sample under investigation was filtered, preserved with nitric acid and concentrated *via* evaporation.

All the analytical investigations were performed at West Siberian Test Center, accred-

TABLE 2

Content of the gross forms of toxic elements in the ash-and-slag wastes and boiler slag of the Kemerovo Region, mg/kg

Elements	Ash-and-slag wastes	Boiler-plant slag	MPC_s with background taken into account [18, 19]	Clarke values for the elements in coal ash [22]	Ash content range for the Kuzbass power-generating coal [23]
Vanadium	22.4–156.8	56.0–145.6	150.0	180±20	95.2–301.6
Cadmium	<0.05–0.4	<0.05–0.09	0.5*	1.50±0.30	1.8–8.8
Cobalt	7.1–24.7	7.35–15.08	–	37±2	28.2–77.4
Manganese	239.6–600.0	184.6–600.0	1500.0	480±30	1515–2704
Copper	14.0–24.0	16.7–52.6	33.0*	110±10	57.1–114.2
Molybdenum	4.0–8.0	4.00–40.0	–	14±1	6.1–13.8
Arsenic	<0.1–15.0	<0.1–13.0	2.0	~50.0	157.2–843.9
Nickel	16.2–65.4	15.0–84.2	20.0	96±5	36.2–132.6
Tin	<0.1–60.0	<0.1–42.0	–	ND	10.7–18.6
Mercury	<0.1–5.0	<0.1–9.88	2.1	0.1	0.06–12.0
Lead	8.8–110.6	6.0–236.0	32.0	56±7	126.2–416.6
Antimony	<0.1	<0.1–15.0	4.5	4–7	8.3–31.7
Chromium	38.4–247.9	20.0–958.9	–	120±5	129.7–290.4
Zinc	16.0–60.2	14.0–50.0	55.0*	170±10	171.4–454.3

Notes. 1. Dash means that there are no standard data required for MPC. 2. ND – No data available.

* Approximately permissible concentration.

TABLE 3

Content of toxic elements in the mobile and soluble forms in the ash-and-slag wastes and boiler slag of the Kemerovo Region, mg/kg

Elements	Ash-and-slag wastes	Boiler-plant slag	MPC
		Mobile forms	
Copper	3.5–20.4	3.0–30.0	3.0
Nickel	3.0–55.0	2.5–65.0	4.0
Zinc	3.6–60.0	3.6–50.0	23.0
Lead	2.0–104.0	2.0–101.6	6.0
Manganese	20.0–586.0	26.0–510.8	–
Chromium	<0.2–4.7	<0.2–2.8	6.0
Molybdenum	0.2–6.0	0.2–5.0	–
Cobalt	<0.4	<0.4	5.0
		*Water-soluble forms**	
Vanadium	<0.0005–0.17	<0.0005–1.77	0.001
Cadmium	<0.00001	<0.00001–0.0016	0.01
Cobalt	<0.0002–0.0009	<0.0002–0.03	0.01
Antimony	<0.0005–0.006	<0.0005–0.05	0.05
Copper	<0.001–0.016	<0.001–0.03	0.001
Molybdenum	<0.001–0.60	<0.001–1.05	0.0012
Nickel	<0.0002–0.27	<0.0002–0.50	0.01
Arsenic	<0.005–0.50	<0.005–0.60	0.05
Lead	<0.0002–0.03	<0.0002–0.025	0.1
Mercury	<0.0001	<0.0001–0.0002	0.00001
Manganese	<0.005–0.13	<0.005–0.44	0.01
Chromium	<0.0005–0.6	<0.0005–0.18	0.001
Zinc	0.0075–0.136	<0.005–0.0078	0.01

Notes. 1. For mobile forms, there are MPC_s presented with background taken into account, mg/kg [20], for water-soluble forms there are MPC_w presented, mg/dm^3 [21]. 2. Dash means that there are no standard data required for MPC.

* The aqueous extract was made according to the ratio of Solid/Liquid = 1 : 1, the content of water-soluble forms of elements in mg/kg corresponds to the value expressed in mg/dm^3.

ited in the State Standard System of the Russian Federation (No. ROSS RU. 21AYa07 0001). For the investigation, we chose the list of selected toxic elements, whose content in environmental objects is standardized [17]. The systematization of the data obtained was carried out according to the following number of samples: 182 samples for ash-and-slag wastes and slag wastes of boiler plants, 35 samples for groundwater, 17 samples for surface water and 20 samples for soil.

RESULTS AND DISCUSSION

Ash-and-slag wastes and slag wastes of boiler plants

Typical distribution for toxic elements in the gross (total content), mobile and soluble forms inherent in the samples of the ash-and-slag wastes and slag wastes of boiler plants is presented in Table 1. Data concerning the content of gross, mobile and soluble special forms of toxic elements in the ash-and-slag wastes and slag wastes of boiler plants located in the Kemerovo Region are presented in Tables 2 and 3.

As far as the studied list of the total content of toxic elements in the ash-and-slag wastes is concerned, there is a significant discrepancy with early published data [23] for zinc, cadmium, antimony, cobalt and arsenic in particular. The authors of [23] mainly systematized the data of a semi-quantitative spectral analysis, whereas we used the method of atomic emission spectroscopy with inductively coupled plasma. The authors of [24] examined the distribution of trace elements such as Zn, Cd, Pb, As, Cu, Ni, Be, Sb in coal taken from the

TABLE 4

Excess order for the concentration of toxic elements with respect to the corresponding values of MPC_s for the gross and mobile elemental forms inherent in the ash-and-slag wastes and boiler slag of the Kemerovo Region

Elements	Ash-and-slag wastes		Boiler-plant slag	
	MPCs excess order	% of samples with respect to the total number thereof	MPCs excess order	% of samples with respect to the total number thereof
		Gross forms		
Chromium	2.7	20.0	9.0–10.7	25.0
Lead	2.0	90.0	3.2–4.3	50.0
	3.4	2.0	7.4	2.0
Mercury	2.4	4.0	1.0–5.0	4.0
Tin	13.3	4.0	9.0	4.0
Antimony	–	–	1.5–3.3	8.3
		Mobile forms		
Copper	2.0–4.0	60.0	1.6–3.2	57.0
	6.8	40.0	6.6–10.1	43.0
Nickel	2.0	31.0	1.3–1.5	28.5
	13.6	35.0	7.8–16.3	28.5
Zinc	2.6	30.0	1.7–2.2	28.6
Lead	17.3	40.0	1.2–16.9	43.0

Note. Dash means that the data does not exceed the MPCs value.

TABLE 5

Excess order for the concentration of toxic elements with respect to the corresponding values of MPC_w in the aqueous extract of the ash-and-slag wastes and boiler slag of the Kemerovo Region

Elements	Ash-and-slag wastes		Boiler slag	
	MPC_w excess order	% of samples with respect to the total number thereof	MPC_w excess order	% of samples with respect to the total number thereof
Vanadium	20.0–81.0	30.0	8.0–20.0	50.0
	110.0–170.0	53.0	700.0–920.0	15.0
			1770.0	2.0
Manganese	4.0–6.0	73.0	5.0–42.0	34.0
	11.5–13.0	12.0		
Copper	4.0–9.0	58.0	2.0–10.0	60.0
	10.0–16.0	35.0	20.0–30.0	12.0
Molybdenum	10.0–67.5	77.0	1.0–58.0	62.0
	100.0–500.0	15.0	83.0–333.0	4.0
			641.0–875.0	2.0
Arsenic	10.0	4.0	3.0–12.0	10.0
Nickel	1.0–5.0	65.0	1.0–6.0	14.0
	27.0	4.0	50.0	2.0
Zinc	1.0–8.0	69.0	–	–
	10.0–13.6	5.0	–	–
Chromium	5.0–8.0	27.0	180.0	10.0
	10.0–30.0	50.0		
	200.0	11.5		
Cobalt	–	–	3.0	2.0

Note. Dash means that the data does not exceed the MPC_w value.

TABLE 6

Content of toxic elements in the groundwater within the area of ash dumps and natural water sources near the ash dumps

Elements	Piezometric wells		Surface water	
	Content, mg/dm^3	MPC_w excess order	Content, mg/dm^3	MPC_w excess order
Vanadium	<0.0005–0.08	0.5–80.0	<0.0005–0.15	0.5–150.0
Cadmium	<0.0005–0.001	–	<0.0005–0.0006	–
Cobalt	<0.0002–0.002	–	<0.0002	–
Antimony	<0.0002	–	<0.0002–0.002	–
Copper	<0.001	–	<0.001	
Molybdenum	0.001–0.16	0.8–133.0	0.002–0.160	2–133.0
Nickel	<0.0002–0.03	0.02–2.8	<0.0002–0.002	0.02–1.8
Arsenic	<0.005–0.021	0.1–0.42	0.007–0.029	–
Lead	<0.0002–0.03	0.002–0.3	<0.0002	–
Mercury	<0.0001	–	<0.0001	–
Manganese	0.10–3.80	10.0–380.0	ND	–
Chromium	0.004–0.008	4.0–8.0	0.004–0.012	4.0–12.0
Zinc	0.005–0.11	0.5–11.0	<0.002–0.028	0.5–2.8

Notes. 1. Dash means that the data does not exceed the MPC_s value. 2. ND – No data available.

Podmoskovye, in flue ash, in furnace slag. With the help of the method of atomic absorption spectrometry for coal ash, they found that the content of some trace elements (Zn 454 mg/kg, Cd 12.66 mg/kg, Pb 95.4 mg/kg, Ni 160.0 mg/kg, Cu 90.67 mg/kg) are higher than our data for the Kuzbass coal. The authors of [5] also noted that the total bulk of the Kuznetsk coal basin is characterized by a low content of trace elements, exception for separate layers.

Typically, the environmental monitoring is based on the comparison of the results of chemical analysis with the standardized values of the concentrations of substances under monitoring. It has been found that concentrations of chromium, lead, mercury and arsenic in the ash-and-slag wastes in the gross form exceed the values of MPC_s [18, 19]. For the 70 samples of boiler slag under investigation there were high levels of chromium, lead, mercury, tin and antimony revealed those are many times greater than the MPC_s values (Table 4). For mercury, one of the most environmentally hazardous elements, the fact of exceeding the MPC_s was revealed for a small number of samples. This could be, to all appearance, caused by the fact that that the main amount thereof under combustion is transferred into the gas phase to be dissipated in the environment [25].

A different picture is observed for mobile species of heavy metals (see Table 4). So, the maximum concentrations of the mobile forms of heavy metals in the ash-and-slag and boiler slag have been found for copper, nickel, zinc and lead.

TABLE 7

Content of toxic elements in soils adjacent to the ash dump areas determined by different methods

Elements	Content, mg/kg	MPC_s, mg/kg
	Gross forms	
Cadmium	<0.05–0.40	0.5*
Copper	16.40–42.05	33.0*
Nickel	28.00–43.00	20.0*
Lead	20.80–42.00	32.0*
Zinc	52.00–151.60	55.0*
Arsenic	6.00–21.00	2.0*
Mercury	<0.1–1.40	2.1
	Mobile forms	
Copper	3.00–19.00	3.0
Zinc	14.00–62.00	23.0
Nickel	8.70–30.00	4.0
Lead	15.00–23.00	6.0

Note. The content of gross forms was determined in accordance with [11], the content of mobile forms was determined according to a technique described in [13].

*Approximately permissible concentration.

TABLE 8

Excess order for the concentration of toxic elements in mobile forms with respect to the corresponding values of MPC_s in soil adjacent to ash dumps

Elements	MPC_s excess order	% of soil samples with respect to the total number thereof
Copper	1.5	67.0
	6.0	33.0
Nickel	1.6–2.2	33.0
	2.5–7.5	67.0
Zinc	2.0–2.7	44.4
Lead	1.7–3.8	100.0

The toxicity of industrial wastes should be evaluated not only from their impact on the soil, but also basing on the ability of toxic elements to be transferred into the water. From the results obtained it follows that the aqueous extract of the ash-slag wastes and boiler slag exhibit a high concentration of vanadium, manganese, copper, molybdenum, arsenic, nickel, zinc, chromium (Table 5).

Aquatic objects

The influence of ash dumps upon groundwater and surface water was analyzed. According to Tables 3 and 6, the high concentration values of vanadium, molybdenum, arsenic, nickel, zinc, manganese and chromium are inherent both in water-soluble forms of ash-and-slag wastes, and in groundwater and surface water from naturally occurring water sources located near the ash dumps. For these elements, there are no geochemical barriers in the course of the infiltration of naturally occurring waters through the bed of the ash dump. Cadmium, cobalt, antimony, lead and mercury were found neither in an aqueous extract of wastes, nor in the ground and surface water.

Soils

Systematization was performed for data obtained from studying the soil adjacent to the territories of ash dumps (Table 7). It is seen that copper, nickel, lead, zinc and arsenic exhibit exceeding the MPC_s values for the gross forms of elements. High concentration values are revealed for the mobile species of zinc, lead, copper and nickel to occur (Table 8). This indicates the transition of these metals into the soil solutions, despite the low concentration values of nickel and copper in the gross form. The data obtained indicate that just the mentioned metals contaminate the territory near the ash dumps.

CONCLUSION

1. Ash-and-slag waste in the case of contacting with water represent a source of toxic elements (vanadium, molybdenum, arsenic, nickel, zinc, manganese and chromium) in groundwater within the areas on ash-and-slag dumps and nearby naturally occurring water sources; the content of these elements in the observation wells is significantly greater than the maximum permissible concentration thereof in water.
2. A significant migration of motile forms of heavy metals (copper, nickel, zinc, lead) from the ash-and-slag waste dumps into the soil is revealed to occur, in spite of a low concentration of nickel and zinc in the gross form.
3. The analysis of the distribution of toxic metals in the system of wastes–ground (surface) water–soil allows one to describe quantitatively supplying the heavy metals to the environment from ash and slag, although the distribution of separate metals in the system is different.
4. These data should be used in the planning of environmental activities on the sites of storing the ash-and-slag wastes.

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