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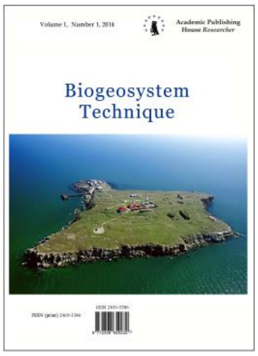

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Effect of Natural and Technogenic Factors on the Mobility and Transformation of Metal Compounds in Soil

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Abstract

The effect of natural and technogenic factors on the Cu, Ni, Mn, Pb, Cd, and As compounds mobility and transformation were studied from an analysis of the fractional–group composition of Cu, Zn, and Pb compounds in the soils of areas adjacent to the Novocherkassk power station. Novocherkassk Power Station is one of the largest power plants at the South of the Russian Federation. The soils sampled from the non-arable sites at different distances from the Power Station. The investigated soils were meadowy chernozem, calcareous chernozem, and alluvial-meadowy soil. The total concentration of metals was determined by the X-ray-fluorescent method. Mobile forms of metals include exchangeable, complex and specifically adsorbed compounds were determined. The fractionation method by Tessier (1979) was applied for metal compounds determination. Changes in the composition of Cu, Zn, and Pb compounds in the soils of technogenic landscapes were estimated. The effect of aerosol technogenic emissions on the mobility of metal compounds was revealed; a higher metal mobility was found in soils with low buffering capacity. Common and specific features of the formation of Cu, Zn, and Pb compounds in soils were determined. The role of individual soil components in the retention of metals in clean and contaminated soils was established.

Keywords: availability, fractions, heavy metals, power station.

1. Introduction

The natural and technogenic factors influence biogeochemical cycle of biosphere (Kalinitchenko, 2016). Novocherkassk Power Station is one of the largest power plants of the Russia Federation; it is the principal source of contaminants in Novocherkassk, because it is located only 7.5 km southeast of the city. The Novocherkassk Power Station (NPS) produces 1 % of the emissions across the territory of the Russia Federation and more than 50 % over the Rostov Region (Mandzhieva et al., 2016). The power station emits annually 250000 tons of contaminants, which include heavy metals (HMs), which are deposited on the soil (Ecological Bulletin of the Don

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Region, 2015; Public report, 2015; Orlović-Leko et al., 2010). The major components of NPS emissions include ash, sulfurous anhydride, nitrogen oxide, soot (more than 30 t/year), vanadium pentoxide (about 8 t/year), iron oxide (more than 5 t/year), chromic anhydride (about 0.1 t/year), hydrogen fluoride (7 kg/year), etc. Ash retains up to 85 % of the initial content of chemical elements from coal (Kizil'shtein et al., 1990). During the combustion of fuel, most chemical elements get into aerosols; therefore, the content of HMs in the volatile ash of emissions is higher than that in the raw materials by a factor of several.

Heavy metals and hazardous elements play a significant role among the pollutants (Il'in et al., 2003). The pollution of soils with metals due to the aerotechnogenic emission is a worldwide issue (Moon et al., 2013). Metals are released during various processes, such as combustion or component wearing. Lead, copper, zinc and cadmium are considered as major inorganic pollutants (Kizilkaya et al., 2012; Minkina et al., 2015; Minkina et al., 2011; Motuzova et al., 2014; Šeda et al., 2017). The aim of this work was to study the heavy metal content in the soils and plants around Novocherkassk Power Station.

2. Materials and methods

The monitoring plots were laid out in 2000. The monitoring plots were located in non-arable patches of soils at different distances from the Power Station (Fig. 1). Most of them lie to the north-west of the station and in the direction of the prevailing wind (Minkina et al., 2013). The soils investigated were meadowy chernozem, calcareous chernozem, and alluvial-meadowy soil. The majority of the soils of monitoring plots were ordinary chernozems; the soils of plots 9 and 10 +were used as control soils.

The low-humus calcareous sandy alluvial meadow soil (plot 2), which had a light texture and a low cation exchange capacity (CEC), and the low-humus silty clayey meadow-chernozemic floodplain soil (plot 3) with a high CEC that differed from the control soils (Minkina et al., 2009; Tishchenko, Bezuglova, 2012).

Soil samples for the determination of soil properties and the contents of Cu, Ni, Mn, Pb, Cd, and As compounds were taken from a depth of 0–20 cm. Basic physical and chemical properties of the studied soils were determined (Methodological guidelines for the integrated monitoring of soil fertility of agricultural lands, 2003). Separated extractions for determination of HM forms including exchangeable (NH_4OAc, pH 4.8), complexed (from the difference between the contents of metals in the 1 % EDTA in 1 M NH_4OAc and NH_4OAc extracts) and specifically adsorbed (from the difference between the contents of metals in the 1N HCL and 1 M NH_4OAc extracts) (Minkina et al., 2008; Mandzhieva et al., 2014); and the fractionation method by Tessier (1979) were applied. The total contents of HMs in the soil were determined by the X-ray fluorescent scanning spectrometer "SPECTROSCAN MAKC-GV". Mobile concentrations were determined by atomic absorption spectrophotometer.

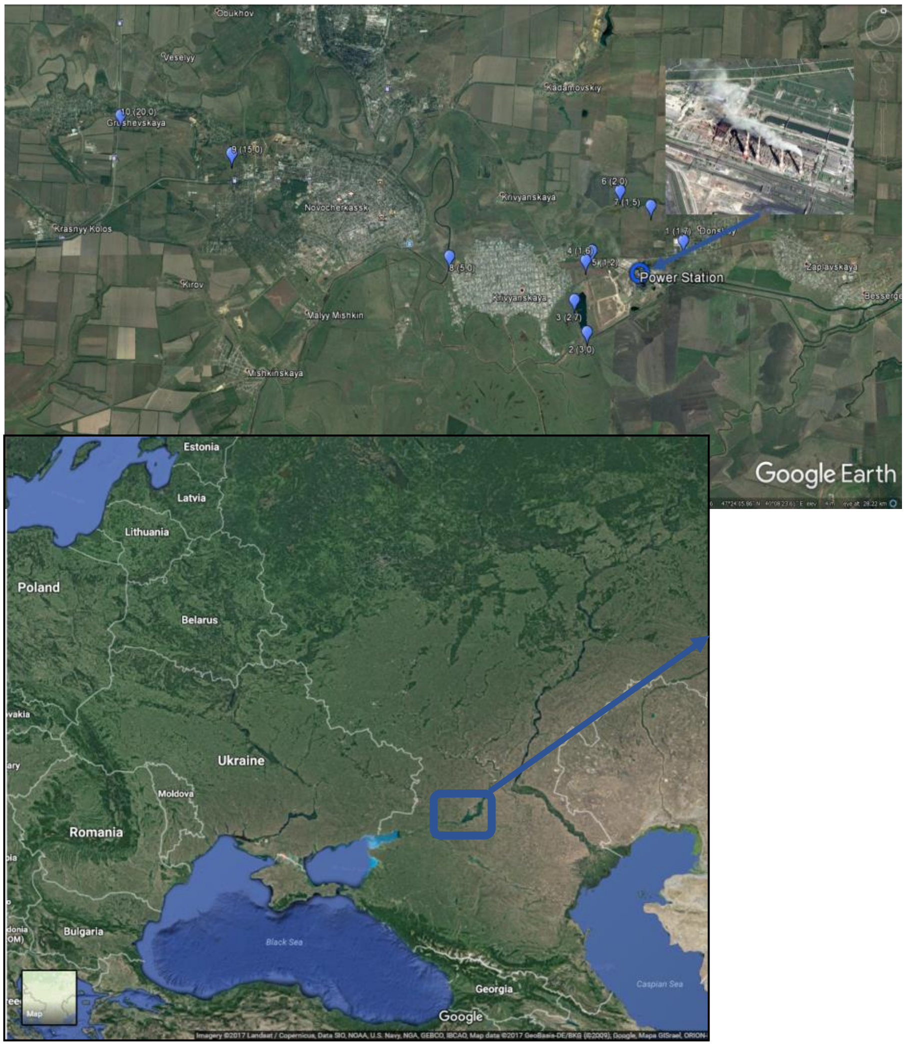


Fig. 1. Schematic map of monitoring plots in the area affected by the Novocherkassk power Station

Plot no. – direction and distance from the Power Station: 1–1 km to the northeast; 2–3 km to the southwest; 3–2.7 km to the southwest; 4–1.6 km to the northwest; 5–1.2 km to the northwest; 6–2.0 km to the north-northwest; 7–1.5 km to the north; 8–5 km to the northwest; 9–15 km to the northwest; 10–20 km to the northwest.

The total contamination index was calculated as:

$Zc = \sum Kc - (n-1)$,

were Zc – the total contamination index; Kc – concentration index ($Kc = Ci/Cbi$, Ci – concentration of elements; Cbi – background concentration of elements); n – number of elements (Il'in, 2000).

3. Results and discussion

The local areas within a 5 km distance to the north-west of the Power Station have become contaminated with total and mobile forms of Cu, Pb, Zn and mobile Ni (Table 1, 2). The HM contents gradually decrease with distance; their contents in remote soils (plots 9, 10) approach the background level (Belousova, 2001; Bezuglova et al., 2016; Minkina et al., 1998; Nikityuk, 1998; Samokhin, 2003; Samokhin et al., 2005; Sobornikova, Kizilshtein, 1990).

In the soils of the background plots, the total content of Cu was 37 mg/kg and that of Zn was 72–80 mg/kg (Table 1). The soil of plot 9 characterized the background content of Pb (25 mg/kg), because plot 10 was located 500 m from a highway, which affected (increased) its Pb content.

The background contents of the above metals in soils were higher than their Clarkes – 20, 50, and 10 mg/kg for Cu, Zn, and Pb, respectively (Vinogradov, 1957), which was related to the mineralogy of soil-forming rocks in Rostov region (Russia). The yellow-brown loesslike loams and clays of the Ciscaucasian Plain partially inherited stable minerals from the original rocks enriched in heavy metals. Some metal ions released during weathering and pedogenesis were strongly fixed in the structure of clay minerals (Minkina et al., 2009; Mandzhieva et al., 2014).

The increase in the total metal content due to Power Station emissions was the most greatly manifested in the soils located near the source of contamination along the main wind direction (plots 4, 5, 8) and in the soils of plot 6. The contents of total Cu, Pb, and Zn in these soils exceeded their MPC values (55, 32, and 100 mg/kg for Cu, Pb, and Zn, respectively). At the same time, the soils of plot 1 and, especially, plots 2 and 3 were less contaminated, although they are located near the source of contamination. This is related to the fact that these plots are beyond the zone of the predominant wind direction. HM total and mobile content lessened with depth, with maximum concentrations found in the 0–5 cm soil layer.

Table 1. Total heavy metals concentrations in monitoring plot soils and their buffer ability

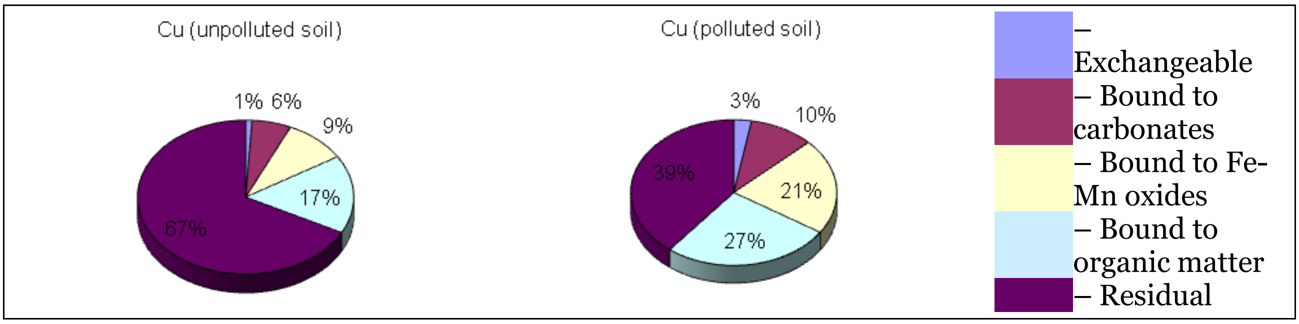
Distances (km) and directions from the Power station	Soil	The total HM concentration in 0-5 cm layer, mg/kg								The total contamination index	Soil HM buffering capacity
		As	Pb	Zn	Cu	Ni	Mn	Cd	Cr		
1,0 NE	Calcareous chernozem	11.0	45.9	114	54.5	69.7	879	0.5	134	11.9	High
3,0 SW	Alluvial-meadowy soils	7.6	10.9	110	48.5	32.9	861	0.5	66	6.1	Middle
2,7 SW	Meadowy soils	7.5	28.0	103	48.3	50.8	361	0.3	113	6.1	Middle
1,6 NW	Calcareous chernozem	8.1	38.5	116	80.6	56.7	566	0.5	134	9.7	High
1,2 NW	Calcareous chernozem	10.2	53.3	146	66.5	55.4	827	0.5	157	12.3	High
2,0 NNW	Meadowy soils	8.1	45.5	120	60.1	68.7	928	0.5	136	10.6	High
1,5 N	Calcareous chernozem	9.1	28.0	99	42.7	56.7	849	0.7	136	10.0	High
5,0 NW	Meadowy soils	9.4	31.9	120	65.8	67.8	983	0.3	131	9.7	High
15,0 NW	Calcareous chernozem	9.4	31.1	100	58.2	57.0	808	0.3	104	8.3	High
20,0 NW	Calcareous chernozem	10.0	28.6	119	46.1	50.3	744	0.3	117	8.3	High
Elements background concentration for present soil		1,8	18.0	72	39.0	32.0	624	0.2	110	-	

The mobility of metals in the contaminated soils varied analogously to their total content (Endovitskii et al., 2009). In the soils closest to the Power Station, the mobile forms of Cu, Pb, and Zn increased by 2.7, 2.5, and 3.8 times, respectively. The content of exchangeable compounds exceeded the metal MPC in the soils of plots 4, 5, 6, and 8 for Cu; plots 5 and 6 for Pb; and plot 5 for Zn (the MPCs are 3, 6, and 23 mg/kg for the mobile compounds of Cu, Pb, and Zn, respectively). Distribution of mobile Cu, Pb, Zn, Cd, Cr, Mn, Ni within non-contaminated soils is characterized by the following trend: complexed < exchangeable < specifically adsorbed. The same succession is relevant for metals contained within contaminated soils, with the exception of Cu, Pb and Ni, in which the exchangeable forms content becomes less than complexed forms.

Table 2. Concentration of exchangeable heavy metals in soil of monitoring plots, mg/kg

Distances (km) and directions from the Power station	Depth, cm	Pb	Zn	Cu	Ni	Mn	Cd	Cr
1,0 NE	0–5	6.5	27.0	3.7	4.4	76.7	0.2	5,4
	5–20	4.4	11.2	1.9	3.1	57.6	0.2	5,2
3,0 SW	0–5	5.3	23.8	4.0	3.8	61.5	0.9	5,6
	5–20	4.5	25.7	3.5	3.4	40.7	0.8	5,6
2,7 SW	0–5	7.1	14.4	1.5	3.6	54.1	0.4	4,1
	5–20	6.3	12.8	0.7	2.3	32.3	0.4	3,8
1,6 NW	0–5	13.2	22.4	7.1	3.4	70.2	0.6	4,9
	5–20	9.5	14.1	4.9	2.2	57.4	0.4	3,8
1,2 NW	0–5	10.7	21.8	5.6	5.9	69.1	0.9	6,4
	5–20	8.4	16.9	4.0	4.4	57.2	0.7	5,3
2,0 NNW	0–5	14.2	23.5	5.2	5.8	88.7	0.4	6,3
	5–20	12.9	19.3	2.7	4.5	73.2	0.2	5,5
1,5 N	0–5	10.1	12.6	3.7	5.7	76.2	0.5	5,3
	5–20	7.5	9.8	1.6	3.7	62.7	0.3	4,3
5,0 NW	0–5	13.1	14.8	4.7	4.3	104.2	0.8	5,7
	5-20	9.9	12.3	2.4	2.9	85.4	0.2	4,7
15,0 NW	0–5	10.5	11.1	7.3	3.6	57.6	0.8	4,6
	5–20	7.2	7.1	2.2	2.5	41.8	0.7	3,5
20,0 NW	0–5	6.5	23.0	4.6	3.3	59.7	0.7	4,7
	5–20	3.2	18.1	1.9	2.3	47.9	0.4	3,5
The maximum concentration limits (MCLs)		6,0	23.0	3.0	4.0	700.0	–	6.0

The fractionation of soil compounds revealed the role of individual soil components in the retention of metals in clean and contaminated soils (Ladonin, Karpukhin, 2011). The main tendency in the changes in the fractional compositions of metals was the increase in the portion of more mobile compounds with increasing accumulation of metal (Fig. 2). The strong fixation of Cu and Pb that has reached the soil is caused by organic matter and nonsilicate Fe minerals, and nonsilicate Fe minerals are responsible for the fixation of Zn. The greatest amount of metals is concentrated within the minerals' crystal lattices in unpolluted soils (Fig. 2). A significant decrease in the most residual fraction can be used to diagnose anthropogenic contamination.



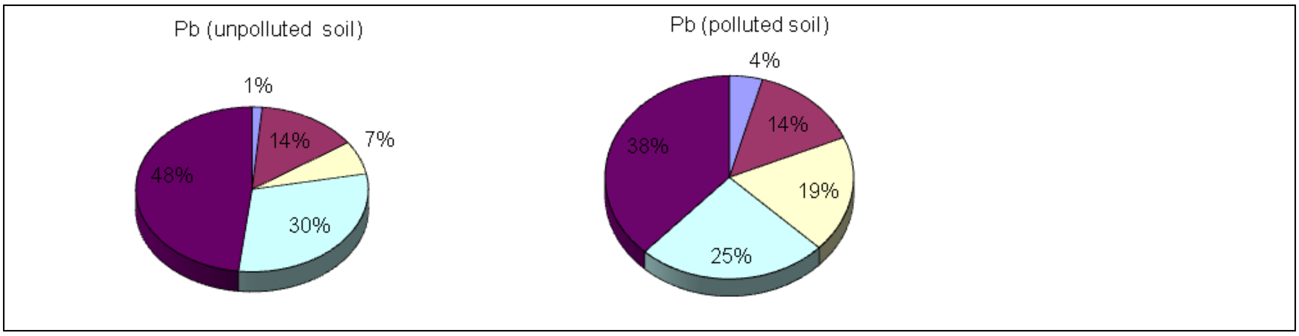


Fig. 2. Fractionation of Cu and Pb in soils around the Power Station

The mobility of metals in soils depended on the buffering properties of the soils (Table 1). The largest differences in buffering properties were revealed for soils strongly differing in texture. The buffering capacity of the soils investigated towards HMs decreases in the following order: calcareous chernozem > meadowy chernozem > alluvial-meadowy soils (Table 1). The highest buffering capacity was observed for the silty-clayey meadowchernozemic soil (plot 3); the lowest buffering capacity was found for the sandy alluvial meadow soil (plot 2). In the meadow-chernozemic soils (plots 3, 6, 8), organic substances actively interacted with HMs (especially Cu and Pb); the highest content of organomineral complex compounds of metals was found in these soils. This was especially true for copper and lead, which are active complexing agents. The combined input of metals had a higher effect than individual input. According to the danger of contamination under similar metal loads, the soils formed the following decreasing series: meadow-chernozemic soils > ordinary chernozems > alluvial meadow soils. In the sandy alluvial soil, the highest content of exchangeable metals was found in comparison to other soils, as well as an insignificant content of their complex compounds. This was related to the low sorption capacity of those soils characterized by a low content of the clay fraction and organic matter and, hence, the surface sorption of ions by soil components as the main mechanism of metal binding. The high mobility of metals in the sandy alluvial meadow soil was responsible for their hazardous migration from the soil into ground and surface waters.

The total contamination index is higher in the monitoring plots which are located closest to the Power Station (Table 1).

4. Conclusion

1. Aerosol emissions from the Novocherkassk Power Station are the major agents of technogenic impact on the soils under study in the Rostov region (Russia); exhaust gases of automobile engines are additional sources of Pb emission. The highest degree of contamination with metals is observed for soils located within a radius of 5 km from the Power Station along the predominant wind direction. The contents of Cu, Zn, and Pb in these soils exceed their MPC levels.

2. In the contaminated soils, the increase in the total metal content (above the MPC) is accompanied by changes in the proportions of metal compounds. In contrast to clean soils, where the metals strongly retained in the structure of silicate minerals are predominant (48–78 % of the total amount), the content of loosy bound metal compounds increases in the contaminated soils. These changes are proportional to the HM load.

3. The group composition of metal compounds varies among the contaminated soils. Copper and lead arriving with technogenic emissions are retained in the mobile forms, predominantly as organomineral complexes, and zinc occurs in exchangeable form and as compounds specifically sorbed by Fe–Mn (hydr)oxides. Organic substances and nonsilicate Fe minerals are the most involved in the strong fixation of Cu and Pb; nonsilicate Fe minerals are also involved in the strong fixation of Zn. The major hazard of contamination of the ecosystem with HMs is related to the increase in metal mobility.

4. The environmental contamination hazard increases with the decreasing metal-buffering capacity of soils. According to the buffering capacity with respect to HMs, the soils in the vicinity of the NPS form the following increasing sequence: silty clayey meadow-chernozemic soil < clay loamy meadow-chernozemic soil < clay loamy ordinary chernozem < sandy alluvial meadow soil.

5. There is dependence between the HM accumulation and location around the Novocherkassk Power Station as well as soil adsorption properties.

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Влияние природных и техногенных факторов на подвижность и трансформацию соединений металлов в почве

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Аннотация. Влияние природных и техногенных факторов на подвижность и трансформацию соединений металлов изучена на основании анализа фракционно-группового состава соединений Cu, Zn, Pb в почвах территорий, прилегающих к Новочеркасской электростанции. Новочеркасская электростанция одна из крупнейших на юге России. Почвы отобраны на участках залежи или целины на разном расстоянии от ГРЭС. Исследуемыми почвами были чернозем обыкновенный, аллювиально-луговая почва и лугово-черноземная. Общее содержание металлов определено методом рентгенфлюоресцентной спектроскопии. Были определены мобильные формы металлов, включающие обменные, комплексные и специфически сорбированные соединения. Для определения соединений металлов в почве был использован метод фракционирования по Tessier (1979). Оценены изменения в составе соединений Cu, Zn, Pb в почвах техногенных ландшафтов. Выявлено влияние аэрозольных техногенных выбросов на подвижность соединений металлов; более высокая подвижность металлов была обнаружена в почвах с низкой буферной способностью. Определены общие и специфические особенности формирования соединений Cu, Zn, Pb в почвах. Установлена роль отдельных почвенных компонентов в сохранении металлов в естественных и загрязненных почвах.

Ключевые слова: доступность, фракции, тяжелые металлы, электростанции.