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ECOLOGICAL AND GEOCHEMICAL ASSESSMENT OF THE SOIL CONTAMINATION LEVELS IN THE AREAS OF METALLURGICAL ENTERPRISES OPERATION

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ЕКОЛОГО-ГЕОХІМІЧНА ОЦІНКА РІВНЯ ЗАБРУДНЕННЯ ҐРУНТІВ У РАЙОНАХ ФУНКЦІОНУВАННЯ МЕТАЛУРГІЙНИХ ПІДПРИЄМСТВ

Purpose. Determination of regularities of accumulation and distribution of heavy metals in environmental objects in the zone of metallurgical enterprises influence.

Methodology. Mineralogical and grain size for selected soil samples is defined. X-ray analysis is performed for selected samples of dust emissions, soil and plants determined, the concentrations of heavy metals are defined using atomic emission and atomic absorption spectral method. Ion exchange, back and fixed forms of the heavy metals in soils are determined using not selective extractants.

Findings. The levels of soil contamination with heavy metals and plants in areas adjacent to the metallurgical enterprise are determined. It is detected that in areas of metallurgical enterprises functioning the mobility of the heavy metals is increasing compared to background areas – Zn in 4,8, Ni – in 5,6, Cu – 2.6 times. In the territory of metallurgical enterprises contents of ion exchange forms of heavy metals in the decreasing order create the following series: Zn> Pb> Ni> Cu> Cr; for backup forms: Pb> Cu> Zn> Ni> Cr.

Originality. The mechanism of transformation and migration activity of man-made forms of heavy metals in the system "emissions of the company - soil - vegetation" is fixed. The increase of heavy metals in soils and change of their physicochemical properties (pH, cation exchange capacity, organic matter content) activates their migration activity.

Practical value. Installed migration activity of heavy metals makes it possible to assess their ability to move from the solid phase of the soil in the soil solution, and then to navigate to the profile with the soil solution and to fall into the human body by trophic chain. The features of absorption and woody herbaceous plants of heavy metals allow to use them as bioindicators of soil contamination.

Keywords: *metallurgical enterprises, heavy metals, soils, vegetation*

Introduction. The last decades saw the technogenesis to become the main process describing the conditioning of ecological and geochemical state of a territory. The high concentration of environmentally dangerous production processes, the lack of the proper nature conservation systems as well as outmoded production technologies and equipment cause dramatic degradation of environmental conditions. The intensive industrial application of natural resources caused the significant changes of allocation of some chemical elements in the surface layer of aeration zone. First of all it concerns the heavy metals (HM); the accumulation of high concentration of them is connected to the anthropogenic activity [1–3].

The heavy metals, as a special group of elements, are notable for their poisonous action on living organisms in case of high concentrations exceeding the background or permitted values. The wastes of metal industry enterprises may become the sources of pollution of environmental objects by HM.

Ukraine is a Top-10 country of ferrous metal producers. Metal manufacture is the main source of pollution by HM for the southwestern region in Ukraine. Dneprodzerzhinsk is a city in the region; the PGSC "Dneprovsky Iron&Steel Integrated Works named after F. Dzerzhinsky" (DMKD named after F. Dzerzhinsky) has worked here since 1887. Its annual atmospheric emission exceeds 100 thousand tons of harmful substances; 14,7% of them are solid substances, 3,8% – nitric oxides, 8,0% – sulphur dioxide, and 72,8% – carbonic oxide [4, 5].

Dneprodzerzhinsk is in the Top-10 of the most polluted cities in Ukraine according to Derzhkomstat. The enterprises emits more than 120 thousand tons of polluting substances into city atmospheric air (it accounts for 14% of total wastes in Dnipropetrovs'k region). The pollution agents emission density from the stationary sources is 870–930 tons per 1 square kilometer which is 120 and 25 times more than total volume in Ukraine and region respectively. There is about 500 kg of harmful substances per head. The indicator considerably exceeds the average level in the country (almost 5 times) and the region (almost 1,5 times) [4, 5].

Dust emissions is a significant source of harmful substances emission to the environment resulting in anthropogenic geochemical anomalies.

Review of the related researches. The HM migration processes in natural and anthropogenic systems were studied by D.V. Ladonin, Yu.Ye. Sajet, B.A. Revitch, A.I. Samchuk, L. O. Petrova, I.A. Avessalomova, Ye.P. Yanin and others. To induce the law of HM migration and magnification under the conditions of technogenesis it is needed to study their fractions in the soils, especially on the territories of industrial agglomerations of ferrous metallurgy in Ukraine. The associations of metals typical for the territories of ferrous metallurgy enterprises affected zone differ according to various authors. It is important to take into consideration the biochemical markers for geochemical study of anthropogenic landscapes as they are the major indicator of the distinction between the natural and anthropogenic HM anomalies.

The allocation of unsolved issues. The only study of HM total content is insufficient for the issues of modern geochemistry. The studies show only the direction of the processes, e. g. migration (loss or magnification). In this case it is quite difficult to come to a conclusion about the possible mechanisms of HM anthropogenic fractions transformations in the soils and their migratory activity. The existence of HM fractions which differ both according to mobility, bioavailability and mechanisms of fixation in soil causes the need for complex investigations.

Under the conditions of city ecosystems the study of HM fractions existence in environment components deserves the special consideration [6]. This mainly refers to the moving fractions as their study enables the assessment of different chemical elements and their compounds capability to move from hard soil into soil solution, and then move through the line like soil solution and get into human body by means of trophic chain.

Nowadays there is no general methodological approach to the problem of reliable estimate of the influence of industrial enterprises wastes and particularly of metallurgical complex wastes on the soils ecological condition. Considering the prevailing magnification of polluting substances in soils there is the possibility to use them as the indicator of the secondary pollution of the territory and determination of the degree of environmental ecological condition worsening.

The Purpose of the work is to induce the law of HM allocation and magnification in soils on the area of metallurgical enterprises operation.

The main part. The researches were carried out in Dneprodzerzhinsk, where DMKD named after F. Dzerzhinsky is sited. As a background plot with the similar landscape and geochemical properties the territory Dnipro-Oril Nature Reserve was chosen. As the influence of the industrial complex on the background plot was not ruled out, it was mentioned as the relatively clean zone.

The soil sampling was carried out according to the State Standard (GOST 17.4.4.02-84). The sampling of dust wastes and different plants species and soil sampling were carried out simultaneously in the enterprise site.

The granulometric composition of the soil was tested according to the State Standard (DSTU 2.1-19:2009). X-ray phase analysis of the studied samples of soils, dust, and plants was carried out by means of the diffractometer DRON-2, automatic diffractometer DRON-3M, on cupric radiation (CuKα=1,54178 Å). The chemical elements concentrations in the samples were determined by atomic emission spectral analysis using the spectrograph STE-1. The HM fractions in the soils and dustfalls were studied by the method of successive extractions. The HM content in the extracts was determined by the atomic absorption spectrometry method using the spectrometer KAS-115. The ecological and geochemical assessment of the territory conditions was carried out according to the total pollution index applying the methodology by Yu.Ye. Sajet. The methodology by I.A. Avessalomova was applied to characterize the biogenic migration of HM in the soils caused by the enterprise wastes and biogeochemical properties of the plants of the adjacent territory.

The ion-exchange (acetate and ammonium buffer (pH 4,8), reserve (1 n HCl) and fixed fractions of HM and their residuum after all extractions were determined using the nonselective extractive agents.

The results of the research. As a result of the soils mineralogical makeup study it was established that such minerals as illite, kaolin, montmorillonite, chlorite, prevail in the clay fraction composition. The light fraction of soils is represented by quartz, quartzite, potash feldspar, there are also muscovite, illite, glauconite, opal. The heavy fraction conent of soils is characterized by prevailing ferric hydroxides, leusoxene, zircon, garnet, granatite, ilmenite, rutile. The pecularity of the studied soils is the significant content of ferruginous minerals such as goethite, hematite, magnetite, franklinite.

As a result of the study of soils geochemical indicators it was established that the city area is characterised by the spread of black soils, ordinary, middle-deep, thin-humous, on loess.

The chemical content of the soils from the studied territories is tabulated in table 1.

Table 1

The chemical content of the soils

Indicators	**The territory of sanitary protection zone of DMKD named after F. Dzerzhinsky, %**	**The background plot, %**
SiO_2	50,4	59,5
TiO_2	0,1	0,6
Al_2O_3	4,3	10,7
Fe_{gen}	17,9	4,2
MnO	0,4	0,1
MgO	1,5	1,6
CaO	7,9	3,9
Na_2O	0,4	0,8
K_2O	0,7	2,4
P_2O_5	0,1	0,1
H_2O	1,9	4,5
l.o.i.	13,9	11,2
Total	99,5	99,6

Note: l.o.i. – loss on ignition

The analysis of the data from Table 1. demonstrated that the higher content of ferric oxide (4,2 times), manganous oxide (3,5 times), and calcium oxide (2 times) in the DMKD named after F. Dzerzhinsky affected zone compared to the background territories soils is observed.

As a result of the study of the chemical content of the dustfalls from the sanitary protection zone (SPZ) of DMKD named after F. Dzerzhinsky it was determined that the content of zinc is 10000 mg/kg; of chromium, 2000 mg/kg; of copper, 1000 mg/kg; of lead, 400 mg/kg, which exceeds the values from the background territories significantly.

To estimate the influence of enterprises wastes on the condition of the adjacent territories the total content of HM in the soils from the studied territories was determined (see table 2).

In the table 2: M_{ed} is an average value for the element; Min, minimum value for the element; Max, maximum value for the element; "σ", standard deviation; "–", maximum permissible limit (MPL) is not stated.

The analysis of the data from table 2. demonstrated that the maximum values of HM content are observed on the territories which are adjacent to the integrated iron-and-steel works. The determined anthropogenic maximum values of HM content in soils correspond to the wind rose and refer to the densely populated city areas.

As for the elements of the first danger class (*Cr, Pb, Zn*) the highest maximum values are referred to the content of zinc (2000 mg/kg), which is fortyfold higher compared to the background value. The high index of the background value overriding is observed for lead (25 times). The element on account of its toxic and mutagenic properties constitute the significant danger in the soils from the enterprise territory.

Table 2

The total content of HM fractions in the soils (mg/kg)

Element	**SPZ of DMKD named after F. Dzerzhinsky**		**Background territory**	**MCL**
	M_{ed} **Min-Max**	**σ**		
Mn	1984 100-6000	875	315	1500
Ni	36 5-50	7	20	20
Co	4 2-8	2	3	–
V	82 20-180	34	50	–
Cr	113 6-1000	85	55	100
Mo	4 1-10	2	2	–
Cu	60 8-400	58	20	33
Pb	119 5-300	47	12	32
Zn	373 30-2000	332	52	55
Sn	5 2-20	3	2	–

As for the elements of the second danger class the highest index of the background values overriding referred to tin (200 times), chromium (18 times), nickel (2,5 times), and cobalt (2,7 times).

To release the geochemical association of HM the rates of their concentrations in soils were calculated by means of Yu.Ye. Sajet method. According to the rates values the HM were ranged as following: $Pb_{(9,9)}>Zn_{(7,1)}>Mn_{(6,3)}>Cu_{(3)}>Sn_{(2,5)}>Cr_{(2)}$.

The total pollution index of soils (Z_t) on the industrial territory, calculated by means of Yu.Ye. Sajet method, vary within 0 to 36; mean of sample is 28. The pollution level of soils on the DMKD named after F. Dzerzhinsky SPZ territory according to the mean values Z_t is referred to moderate dangerous category.

Soil buffer capacity is one of the most important ecological and geochemical properties of soils; it depends on physical and chemical properties of soil adsorption complex (SAC) and the content of soil organic substance. The buffer capacity index (K_b) is directly proportional to the SAC sorptive capacity (E) and inversely proportional to pH change. To estimate the HM migration processes in the soils the content of SAC was studied (table. 3).

In the table 3: C_{org} is the quantity of organic substance; E_{tot} is the sum of exchangeable cations; K_b is buffer capacity index.

The experimental testing of SAC sorptive capacity and soils buffer capacity both on the territory affected by the ironworks and background territory was carried out by means of A. I. Samchuk method. It was observed that on the anthropogenically polluted territories adjacent to DMKD named after F. Dzerzhinsky, the soils buffer capacity decreased and the ecological condition changed for the worse.

Table 3

Physical and chemical properties of soils

Indicators		**The territory of sanitary protection zone of** DMKD named after F. Dzerzhinsky	**The back-ground plot**
Exchangeable cation, mg·eq/ 100 g	H^+	10,1	10,8
	Ca^{2+}	2,3	49,3
	Mg^{2+}	1,1	18,4
	K^+	0,2	0,8
	Na^+	1,2	0,6
	E_{tot}	14,9	79,9
K_b		4,5	87,7
C_{org}, *mg/kg*		4,6	7,8
pH		6,2	6,4

The assessment of the HM total content in the soils on the territory of the enterprise sanitary protection zone and outside the territory enabled to determine the HM migration mechanisms in the system of “enterprise wastes – soils”. The environment pollution processes represented on the studied territory by lead and zinc (the first danger class) are not limited by soil contamination. The high migration capacity of the metals and their significant toxic potential make them be involved into “soil-plant” migration chain. The assessment of the process was carried out by means of HM extraction from the samples of the studied soils by means of specific chemical agents according to the extraction methodology. The fig. 1 and 2 show the distribution of HM according to their fractions in the soils.

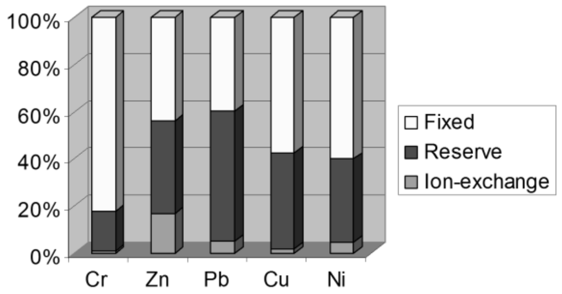

Fig. 1. The HM fractions in the soils of the territory of sanitary protection zone of DMKD named after F. Dzerzhinsky (interval 0–5 sm)

The percentage of HM ion-exchange fractions in the soils of the Dneprodzerzhinsk plot decrease as ranged (%): $Zn_{(16,9)}>Pb_{(5,2)}>Ni_{(4,5)}>Cu_{(1,8)}>Cr_{(0,8)}$. The percentage of HM reserve fractions in the soils of the studied plot decrease as ranged (%): $Pb_{(55,2)}>Cu_{(40,7)}> Zn_{(39,2)}>Ni_{(35,4)}> Cr_{(16,9)}$. According to the percentage of fixed forms decrease the range can be established as follows (%): $Cr_{(82,3)}>Ni_{(60,1)}> Cu_{(57,5)}>Zn_{(43,9)}>Pb_{(39,6)}$.

The background plot demonstrates the following distribution of HM (%): ion-exchange fraction $Pb_{(5,2)}>Zn_{(3,5)}> Ni_{(0,8)} = Cu_{(0,8)}>Cr_{(0,6)}$; reserve fraction $Pb_{(30)}>Ni_{(29)}>Cu_{(19)}> Cr_{(16)}>Zn_{(15)}$; fixed fraction $Cr_{(83,4)}>Zn_{(81,5)}>Cu_{(80,2)}> Ni_{(70,2)}>Pb_{(64,8)}$.

The research of the processes of HM fractions formation in the soils caused by the anthropogenic factors demonstrated that the percentage of moving HM fractions is higher compared to relatively clean soils. The moving capacity of zinc ions decreases 4,8 times; nickel, 5,6 times; copper, 2,6 times.

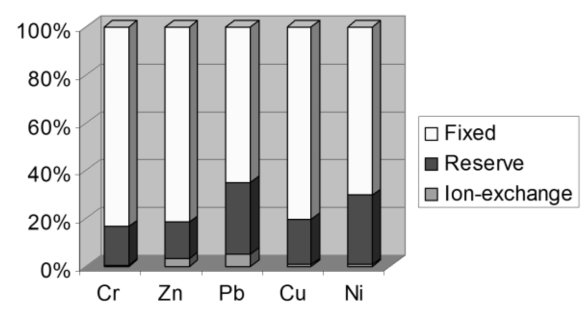

Fig. 2. The HM fractions in the soils of the background plot (interval 0–5 sm)

So, in the soils contaminated anthropogenically the quantity of HM in moving fractions decrease compared to the background plots.

To assess the influence of the ironworks on the distribution of lead and zinc content on the territory of Dneprodzerzhinsk the monoelement maps were created by means of MapInfo 9 (fig. 3, 4).

The content of lead in soils on the background territory is 12 mg/kg, the average total content on the territory of Dneprodzerzhinsk is 119 mg/kg (the horizon of 0–5 sm) which means the tenfold exceed of the background value and fourfold exceed of MCL.

The maximum value of lead content on the studied territory is 300 mg/kg which 25 times exceeds the value for the background territory and 9 times exceed the MCL value. Thus the range of anthropogenic anomalies of lead in the soils of residential city area was determined. The localization of the anthropogenic anomalies of lead content is observed not only near the DMKD named after F. Dzerzhinsky, but also in the city center, which demonstrates the role of motor transport in the soils contamination (fig. 3).

It was determined that the content of zinc on the background territory is 52 mg/kg, the average total content in the soils on the territory of Dneprodzerzhinsk is 373 mg/kg (the horizon of 0–5 sm) which means the sevenfold exceed of the region background and MCL values. The maximum value of zinc content is 2000 mg/kg which 38 times exceeds the value for the background territory and 36 times exceed the MCL value; it was observed in the western area of DMKD named after F. Dzerzhinsky. The western and southern parts of the city are less contaminated, the total zinc content is the same as the for the background soils (fig. 4).

It was determined that the anthropogenic anomalies of the elements are mainly local.

The research included the study of the next after the soils stage of HM migration, the vegetation, which is an intermediary between the soils and human organism.

The intensity of HM magnification by the vegetation is described by the ratio of biological absorption (RBA) and is determined by the ratio of element content in ash is content in soil. The sum of the factors is the biogeochemical activity ratio (BGA).

As a result of the geochemical study of herbs and trees in the DMKD named after F. Dzerzhinsky affected zone the laws of biological absorption of HM from the hard soil by the vegetation were established (table 4). Ni, Mo, Co and Cu were absorbed by the herbs the most intensively (quack grass); Cr, V and Pb were absorbed the least effectively. The mean value of BGA ratio of the species characterizing the intensity of the elements absorption by the plants is.

Fig. 3. The Pb distribution in the soils (interval 0–5 sm) city of Dneprodzerzhinsk

Fig. 4. The Zn distribution in the soils (interval 0–5 sm) city of Dneprodzerzhinsk

Table 4

The ratios of HM biological absorption by the vegetation in the DMKD named after F. Dzerzhinsky affected zone

Leaves and needles	**Mn**	**Ni**	**Co**	**V**	**Cr**	**Mo**	**Cu**	**Pb**	**Zn**	**Sn**	**BGA**
Elytrigia repens (L.) Nevski	0.4	4	1.7	0.2	0.2	3.8	0.9	0.2	0.5	0,8	12.7
Robinia pseudoacacia L.	0.3	2.2	2	0.2	0.1	5	0.6	0.1	0.3	0,8	11.6
Acer platanoides L.	0.4	1.1	1	0.2	0.1	0.8	0.7	0.2	0.2	0,8	5.5
Picea abies (L.) H.Karst.	0.2	0.3	0.8	0.1	0.1	2.5	0.1	0.3	0.3	0,6	5.3
Castanea sativa Mill.	0.3	5.6	2	0.1	0.1	1	0.8	0.2	0.2	0,6	10.9
Betula pendula Roth.	0.3	1.4	1.3	0.2	0.1	1.5	0.3	0.3	1.6	0,8	7.8
Tilia europaea L.	0.3	0.6	1	0.1	0.1	1.3	1.3	0.3	0.2	0,8	6

The trees absorb *Mo, Ni, Co* from soils the most effectively; less effectively, *Cu, Sn;* the least intensive, *V* and *Cr*. The highest value of BGA was determined for *Robinia pseudoacacia* and *Castanea sativa*; the least, for pendent *Picea abies.*

High RBA of *Cu* is reasoned by the ability of the element to create the hard complexes with organic substances.

The conclusions and further development prospectives. As a result of the study of the ironworks activities influence on the soils it was observed:

- the decrease of soil surface layer pH, their cation exchange capacity and buffer value;

- the increase of ferric oxide, manganous oxide, and calcium oxide content as compared to background plots;

- the formation of anthropogenic association of the following HM: lead, zinc, manganese, copper, tin, chromium;

- the exceed of the background values in soils of such iron-containing minerals as goethite, hematite, magnetite, franklinite;

- the contamination level of the soils from the territory of sanitary protection zone of DMKD named after F. Dzerzhinsky according to the mean values of the total index of soil contamination is referred as “moderate dangerous”;

- the trees absorb *Mo, Ni, Co* from soils the most effectively; less effectively, *Cu, Sn;* the least intensive, *V* and *Cr.*

To provide the complex assessment of the ecological situation of the anthropgenically polluted territories it is recommended to carry out the geochemical studies of the soils and vegetation using the quantative indices of anthropogenic migration and magnification of chemical elements.

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Мета. Встановлення закономірностей накопичення та розподілу важких металів в об’єктах навколишнього середовища в зоні впливу металургійних підприємств.

Методика. Для відібраних проб ґрунтів визначено мінералогічний та гранулометричний склад. Для відібраних проб пилових викидів, ґрунтів та рослин виконано рентгенофазовий аналіз, визначено концентрації важких металів з використанням атомно-емісійного та атомно-абсорбційного спектральних методів. З використанням неселективних екстрагентів визначено іонообмінну, резервну та фіксовану форми знаходження важких металів у ґрунтах.

Результати. Встановлено рівні забруднення важкими металами ґрунтів та рослин на територіях прилеглих до металургійного підприємства. Виявлено, що в районах функціонування металургійних підприємств підвищується рухомість важких металів порівняно з фоновими ділянками – Zn у 4,8, Ni – у 5,6, Cu – у 2,6 рази. На території металургійного підприємства вміст іонообмінних форм важких металів у порядку убування утворює наступний ряд: Zn>Pb>Ni>Cu>Cr; для резервних форм: Pb>Cu>Zn>Ni>Cr.

Наукова новизна. Встановлено механізми трансформації та міграційну активність техногенних форм важких металів у системі «викиди підприємства – ґрунти – рослинність». Збільшення вмісту важких металів у ґрунтах та зміна їх фізико-хімічних властивостей (pH, ємність катіонного обміну, вміст органічної речовини) активізує їх міграційну активність.

Практична значимість. Встановлена міграційна активність важких металів дає можливість оцінити їх здатність переходити із твердої фази ґрунту у ґрунтовий розчин, а після цього переміщуватися по профілю із ґрунтовим розчином і по трофічному ланцюгу потрапляти до організму людини. Визначені особливості поглинання деревними та трав'яними рослинами важких металів дозволяють використовувати їх для біоіндикації рівня забруднення ґрунтів.

Ключові слова: *металургійні підприємства, важкі метали, ґрунти, рослинність*

Цель. Установление закономерностей накопления и распределения тяжелых металлов в объектах окружающей среды в зоне влияния металлургических предприятий.

Методика. Для отобранных проб почв определен минералогический и гранулометрический состав. Для отобранных проб пылевых выбросов, почв и растений выполнен рентгенофазовый анализ, определены концентрации тяжелых металлов с использованием атомно-эмиссионного и атомно-абсорбционного спектральных методов. С помощью неселективных экстрагентов определено ионообменную, резервную и фиксированную формы нахождения тяжелых металлов в почвах.

Результаты. Установлены уровни загрязнения тяжелыми металлами почв и растений на территориях, прилегающих к металлургическому предприятию. Выявлено, что в районах функционирования металлургических предприятий повышается подвижность тяжелых металлов по сравнению с фоновыми участками – Zn в 4,8, Ni – в 5,6, Cu – в 2,6 раза. На территории металлургического предприятия содержание ионообменных форм тяжелых металлов в порядке убывания образует следующий ряд: Zn>Pb>Ni>Cu>Cr; для резервных форм: Pb>Cu>Zn>Ni>Cr.

Научная новизна. Установлены механизмы трансформации и миграционная активность техногенных форм тяжелых металлов в системе «выбросы предприятия – почвы – растительность». Увеличение содержания тяжелых металлов в почвах и изменение их физико-химических свойств (pH, емкость катионного обмена, содержание органического вещества) активизирует их миграционную активность.

Практическая значимость. Установленная миграционная активность тяжелых металлов дает возможность оценить их способность переходить из твердой фазы почвы в почвенный раствор, а затем перемещаться по профилю с грунтовым раствором и по трофической цепи попадать в организм человека. Выявленные особенности поглощения древесными и травяными растениями тяжелых металлов позволяют использовать их для биоиндикации уровня загрязнения почв.

Ключевые слова: *металлургические предприятия, тяжелые металлы, почвы, растительность*

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