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The geoecological analysis performed for the geochemical composition of ash and slag waste obtained at Zmiiv thermal power plant

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Abstract. The objective of the study was the composition of ash and slag waste from the Zmiiv TPP (thermal power plant) and the peculiarities of migration of heavy metals (HM) from the place of storage of ash and slag waste into the ecosystem. To achieve this goal, the following tasks were solved: chemical analysis of ash and slag waste of the Zmiiv TPP;

identification of the probability of HM migration into the soil environment in the places of ash and slag waste storage. Ash and slag of the Zmiiv TPP contain Cu, Cr, As, Cd, Ni, Pb in quantities several times higher than the threshold limit value (TLV). For ash and slag wastes, the total pollution rate was $Z_c = 43$, which corresponds to a high level. That is, this artificial horizon is dangerous. HMs migrate to groundwater and soils near the ash stockpiles as a result of infiltration of precipitation waters, leaks from water-bearing communications, water filtration through the base of the ash stockpiles of the Zmiiv TPP. To determine soil contamination near the ash stockpiles, we analyzed soil at the distances of 0, 5, 10, 50 and 100 meters. The contents of the HM decreased further away from the stockpiles. At the distance of up to 100 meters from the dump, there were excesses of the threshold limit values for Ni, Cu, As, Cr in the soil. The concentration factor exceeded one for Cr, As, Cu, Cd, Ni. Only at the distance of 100 meters did the contents of Pb and Zn reached the background values. The calculation of the total rate of soil contamination allowed us to classify these soils as moderately dangerous and acceptable. However, the Zn indicator has several significant disadvantages, particularly it does not take into account the differences in the potential hazards of the elements, as well as, most importantly, the synergistic effects of polymetallic pollution. The coefficient of synergistic effect of heavy metals was 26.64 (in the soil of the ash stockpiles), then decreased, but even at the distance of 100 meters it equaled 11.23, i.e. at the distance of 0... 100 m from the ash stockpiles, the overall actions exceed the norm. The study revealed that Cu, Ni, Zn and Cr had low mobility in the soil near the ash stockpiles and therefore accumulated near the stockpiles, which may be explained by neutral and slightly alkaline soil pH values. The ratio of mineral phases and glass varied, but we should note the predominance of aluminosilicates, calcium silicates and glass in the ash and slag wastes. Heavy metal compounds are confined mainly to amorphized clay aggregates and soot-coal ash formation, to a lesser extent to slag glass and even less to grains of quartz sand. Since ash contains such fractions that can be easily carried by the wind, it should be assumed that ingress of HM into the ecosystem occurs by air, which also contributes to air pollution. The solution to the problem of ash and slag waste disposal can be found in their utilization in the production of construction materials, in road construction, but it is necessary to study the composition of ash and slag and the probability of migration of HM depending on the conditions of use.

Key words: geoecological analysis, ash and slag waste, heavy metals, man-made load, ecosystem pollution

Геоекологічний аналіз геохімічного складу золошлакових відходів Зміївської теплоелектростанції

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Анотація. Метою дослідження є аналіз геохімічного складу золошлакових відходів Зміївської теплоелектростанції (ТЕС) та виявлення особливостей міграції важких металів (ВМ) від місця складування золошлакових відходів у екосистему. Для досягнення встановленої мети було вирішено наступні завдання: проведення геохімічного аналізу золошлакових відходів

Зміївської ТЕС; вивчення вірогідності міграції ВМ у грунтах місць зберігання золошлакових відходів. Золошлаки Зміївської ТЕС містять Cu, Cr, As, Cd, Ni, Pb у кількостях, що в декілька разів перевищують гранично-допустиму концентрацію (ГДК). Для золошлакових відходів сумарний показник забруднення становить $Z_c = 43$, що відповідає високому рівню. Тобто цей штучно створений горизонт є небезпечним. ВМ мігрують у підземні води і у ґрунти поряд із золовідвалом за рахунок інфільтрації атмосферних опадів, викиди з водопровідних комунікацій, фільтрації вод через основу золовідвалу Зміївської ТЕС. Для визначення забруднення ґрунтів поблизу золовідвалу здійснено аналізи ґрунту на відстані 0...100 метрів. Встановлено, що зменшення концентрації ВМ у ґрунті з відстанню від золошлаковідвалу. На відстані до 100 метрів від відвалу спостерігається перевищення у ґрунті ГДК за вмістом сполук Ni, Cu, As, Cr. Коефіцієнт концентрації перевищує одиницю для Cr, As, Cu, Cd, Ni. Лише на відстані понад 100 метрів вміст Pb та Zn досягає фонових значень. Розрахунок сумарного показника забруднення ґрунтів дозволяє віднести дані ґрунти до помірно небезпечних та допустимих. Однак, є декілька суттєвих недоліків у показника Z_c . Насамперед, він не враховує відмінностей потенційної небезпеки хімічних елементів, а також, що найбільш важливо, синергетичні ефекти поліметалічного забруднення. Коефіцієнт синергетичного впливу важких металів становить 26,64 (у ґрунті золовідвалу), далі зменшується, але навіть на відстані 100 метрів становить 11,23, тобто на відстані 0...100 м від золошлаковідвалу не виконується умова не перевищення коефіцієнту сумарної дії одиниці. Встановлено, що Cu, Ni, Zn і Cr характеризуються низькою рухливістю у ґрунті поблизу золовідвалу, через що вони акумулюються в екосистемі поряд з золовідвалом, що пояснюється нейтральними і слаболужними значеннями рН ґрунту (рН=8,0...8,5). Співвідношення мінеральних фаз і скла нестійке, проте слід зазначити переважання у золошлаку алюмосилікатів, силікатів кальцію і скла. Сполуки ВМ приурочені в основному до аморфізованих глинистих агрегатів і сажисто-вуглецевим утворенням золи, в меншій мірі до шлакового скла та ще менше до зерен кварцового піску. Оскільки золошлак містить такі фракції, що можуть легко розноситися вітром, слід припустити, що надходження ВМ у екосистему відбувається і повітряним шляхом, що також сприяє забрудненню атмосферного повітря. Вирішення проблеми утилізації золошлакових відходів слід віднайти у виробництві будівельних матеріалів, у дорожньому будівництві, але необхідно вивчати склад золошлаків і вірогідність міграції ВМ залежно від умов використання.

Ключові слова: золошлакові відходи, важкі метали, техногенне навантаження, забруднення екосистеми

Introduction

The work of TPPs (thermal power plants) generates wastes: ash and slag (heavy fraction) and ash (volatile fraction). Wastes most often are deposited in the open-air. Subject to atmospheric precipitations, the components of ash and slag may migrate into the environment.

Stockpiles of ash occupy large areas, having a negative impact on the environment. A number of studies have focused on danger of ash-and-slag wastes, and the opinions on the subject vary. A number of researchers report the great threat these wastes pose and the possibility of using ash and slag in production of construction materials, road construction, which would reduce the expenditures of raw material, and even propose using them as fertilizers (Bushumov S.A., 2020; Snikkars P.N., 2020; Cristina T. A., 2019). Other researchers draw attention to the environmental pollution with wastes from TPPs (Sokolov A.V., 2019, Ochur-ool A. P., 2019, Turhan Ş., 2020 3-7). Fly ash and ash and slag wastes contain As, Pb, Cr, Cu, Ni, Co, V, Cd, Zn, Se, Mn, Fe, K, Ba, Na, Ca, Mg, Be, F (Pribilova V.M., 2013, Krainiuk E. V., 2004), which may be dangerous to human health (Kornus A., Kornus O., Shyshchuk V., 2020). Ash and slag have high concentrations of zinc, plumbum, cuprum, nickel, vanadium, cadmium, barium, sodium, beryllium, cobalt (Ochur-ool A. P., 2019). There is research confirming significant soil contamination with Ni, Cr with Hg near the stockpiles of ash (Turhan Ş., 2020).

A number of European countries are processing ash and slag wastes, for example Denmark and Germany effectively use ash and slag in production of construction materials. At the same time, the share of recycled ash and slag accounts for up to 100%. For instance, in Germany, it is prohibited to deposit ash and slag wastes. Poland, China and the USA recycle around 60% of generated ash and slag (Snikkars P.N., Zolotova I.Yu., Osokin N.A., 2020).

Provision of the needs of industry and the population requires great resources of electric energy. Therefore, for example, generation of electric energy in Ukraine in 2019 accounted for 141.2 B kW, over a third of this energy was generated by TPPs and CHP (combined heat and power) stations (Statistichnij shhorichnik Ukrainy, 2019).

During the generation of thermal energy, thermal power was 129,045 GCal/year, and the main providers of the thermal energy were thermo-regulating units, heating plants, TPPs and CHPs, the work of which left ash and ash-and-slag waste.

The greatest activity of TPPs is concentrated in eastern Ukraine (Fig. 1). The Zmiiv TPP is among the five largest TPPs, with power over 2,000 MW (Fig. 1,2).

Objective of the study

The objectives of the study were the composition of ash-and-slag wastes of the Zmiiv TPP and specifics of the migration of heavy metals from place of

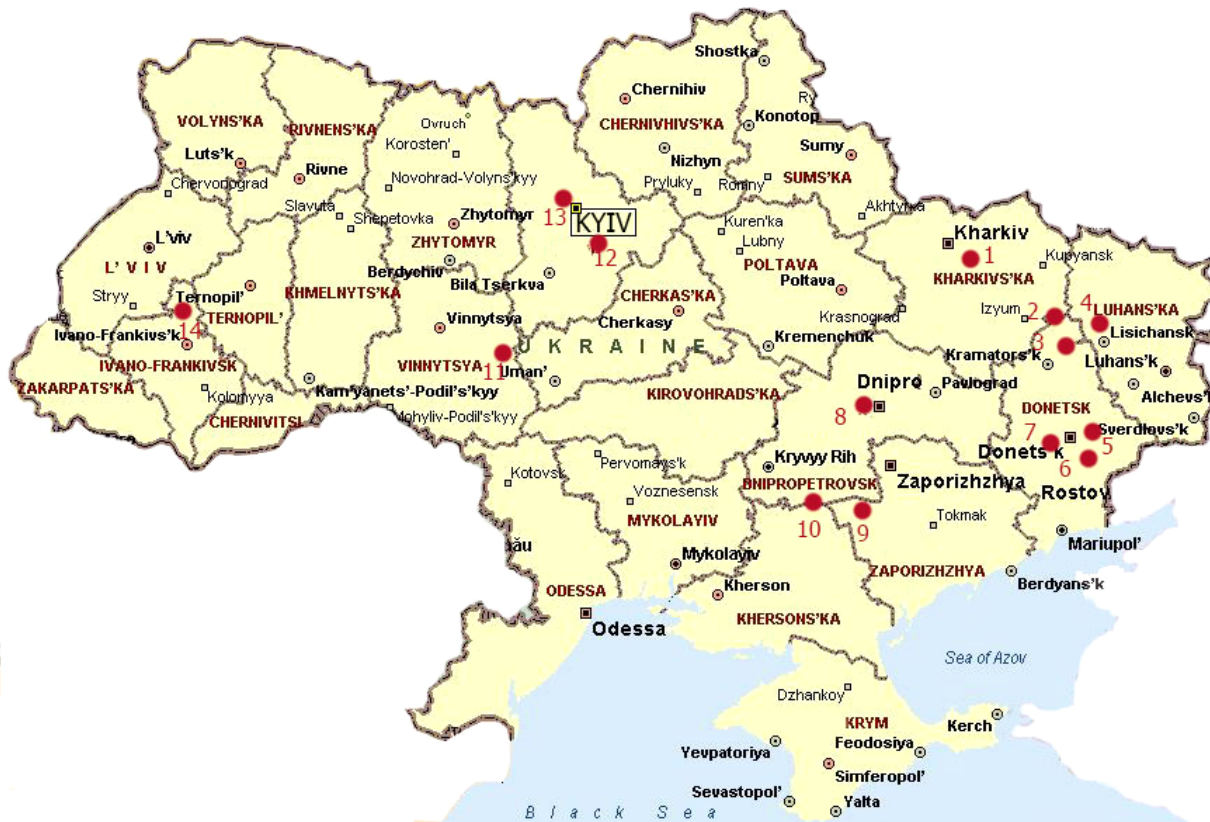


Fig. 1. TPPs in Ukraine

1 – Zmiiv, 2 – Slovianska, 3 – Vuhlehrska, 4 – Luhansk, 5 – Zuivska, 6 – Starobeshevska, 7 – Kurakhivska, 8 – Prydniprovsk, 9 – Zaporizka, 10 – Kryvyi Rih, 11 – Ladyzhyn, 12 – Trypilska, 13 – Dobrotvir, 14 – Burshtyn

deposition of ash slag wastes to the components of the ecosystem. To achieve the goal, we solved the following tasks: performing geochemical analysis of ash and slag wastes of the Zmiiv TPP; finding probability of migration of HM to soil in places of storage of ash and slag wastes.

The Zmiiv TPP is the leading polluter in Kharkiv Oblast, its impact assessed as 60% of the total

pollution in the Oblast (Pribilova V.M., 2013). The TPP has operated since 1960, having the electric capacity of 2,150 MW. The TPP produces 18 B kW per h of electric energy annually. As fuel, the Zmiiv TPP uses low-grade coal with addition of mazut or gas of the Shebelinka gas field. The output of ash and slag accounts for 1.2 M T/year. A total of 200 thousand tons is processed annually, 1 M T is stored in the

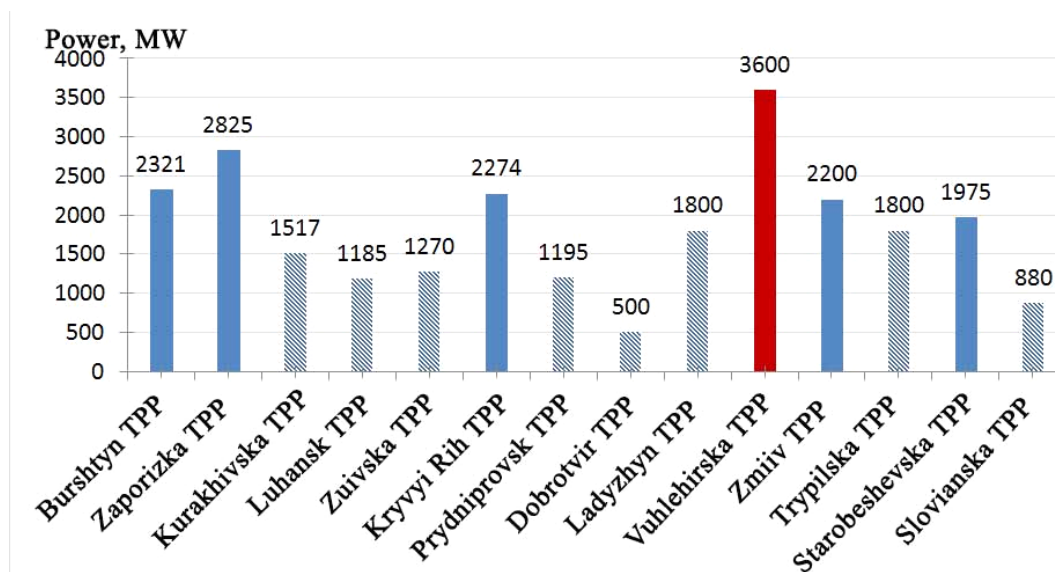


Fig. 2. Power generated by TPPs in Ukraine, MW



Fig. 3. Polygon of ash and slag wastes of the Zmiiv TPP

stockpiles. Currently, the ash stockpiles of the Zmiiv TPP comprise 30 M T of ash and slag. The system of ash and slag removal is hydraulic.

The polygon of ash and slag wastes of the Zmiiv TPP occupies an area of 350 ha (Fig. 3). The ash stockpiles cause a number of ecological problems. Ash and ash and slag wastes are solid non-burned residuals of solid fuel, which are transferred to ash stockpiles. The main mass (965 - 98%) of ash and slag wastes comprises oxides – 45... 60%; CaO – 2.5...9.6%; MgO – 0.5...4.8%; Fe₂O₃ – 4.1-10.6%; Al₂O₃ – 10.1...21.8% i SO₃ – 0.03... 2.7%.

The most important component of ash and slag wastes is SiO₂ (over 40%), which together with Al₂O₃ takes part in formation of calcium aluminosilicates. Up to 15-80% ash and slag wastes are composed of crystalline phase, the rest – poorly soluble quartz, mullites, hematite, magnetite, etc. (Krainiuk E. V., 2004). Furthermore, the ash and slag contain Zn, Tl, Pb, Cr, Mn, Co, Ni, Hg, As, Sb, V, Sr, Ge, B, Be, F and other (Pribilova V.M., 2013).

The stockpiles of the Zmiiv TPP are of a hydrotechnical structure, which also has an effect on the environment (Fig. 4). The stockpiles of the Zmiiv TPP have an impact on groundwater and chemical

composition in the area of Lyman village, the Lyman and Chaika Lakes. For the TPP and coal CHP, the storage of wastes, particularly ash and slag, is a relevant ecological problem.

Technogenic load requires constant control of the condition of the components of geological environment, the main of which are soil and water-bearing horizons, for the expedience and efficiency of the environment-protective measures depend on them. Therefore, determining the scales of pollution of constituents of the geological environment, detection of abnormality, designation of ranges of pollution is a relevant task of geological-ecological studies.

Materials and methods

The content of heavy metals in ash, ash and slag wastes and soil was studied using atomic absorption analysis (AAA) on a C-115 spectrometer. For this method, the lower limit of detection was 0.2 µg/mL of extraction solution. Identification was not hindered by the presence of other metals in the samples.

To determine the composition of solid non-organic part of ash and slag, we performed X-ray analysis. Studies were performed on an X-ray diffractometer DRON -1.5 at the regimes of analysis:

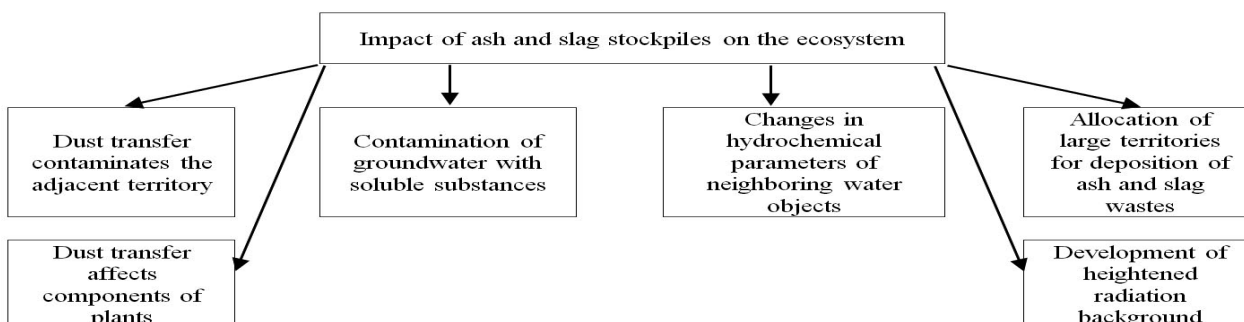


Fig. 4. Impact of ash and slag wastes and deposition sites of ash and slag wastes of TPP on the environment

voltage $U = 35\text{kV}$, power of anode current $I_{\text{anode}} = 20\text{ mA}$, velocity of circulation of a sample was 2 degrees/min; the speed of tape of the recorder was 600 mm/h. Copper electrode was used. To avoid β -emission, the analysis was performed using Ni filter and the method of powder diffraction. Specially selected regimes of analysis allowed us to obtain high resolution, especially when examining small amounts of samples.

Results and their analysis

Content of separate toxic elements and heavy metals, determined using AAA method, is presented in Table 1.

Table 1. Chemical composition of ash and slag wastes of the Zmiiv TPP (mean values), mg/kg

	Fe	Cu	Mn	Cr	As	Cd	Ni	Pb	Zn
Concentration of ash and slag, C mg/kg	2800	55.4	34	16	27	0.8	135	16	12
Background content	1510	5.7	792	1.7	1.5	0.3	14.4	13.9	18.7
TLV		3	1500	6	2	3	4	30	23
Coefficient of contamination (excess of TLV), K		18.5	0	2.7	13.5	0.3	33.8	0.5	0.5
Concentration coefficient, K_c	1.9	9.7	0	9.4	18	2.7	9.4	1.2	0.6

Despite the fact that the samples contained Cu, Cr, As, Cd, Ni, Pb in concentrations that were several times higher than the TLV (Table 1), the chemical analysis of the samples of ash and ash and slag wastes of the TPP indicated that these values may be dozens of times higher. Therefore, for example, As in ash may account for up to 58 mg/kg, Ni even up to 56 g/kg, and Cr to 43 g/kg. In those samples, compared with ash and slag wastes of other TPPs, the amounts of Fe and Mn were small (Krainiuk E. V., 2004). We determined that the acidity of aquatic environment equaled $\text{pH} = 8.8$.

For the aforesaid conditions, we determined the coefficient of contamination according to the formula:

$$K = \frac{C_{\text{sample}}}{\text{TLV}}, \quad (1)$$

where C_{sample} is actual concentration of the metal in the soil (ash and slag wastes).

However, because in this case K does not take into account regional specifics of the content of HM, we calculated the concentration coefficient:

$$K_c = \frac{C_{\text{sample}}}{C_b}, \quad (2)$$

where C_b is regional background content of element.

Contamination usually is poly-elemental, and so assessment of it requires calculation of parameter of overall contamination (Z_c), which reflects the additive sum of the values of concentration coefficients that exceed the background level (Buts, Yu., Krainiuk, O. 2006). Overall parameter of contamination (Z_c) was determined according to formula:

$$Z_c = \sum_1^n K_c - (n - 1), \quad (3)$$

n – number of examined chemical elements.

According to the level of Z_c , the extent of contamination was as follows: moderately dangerous level at $Z_c < 16$; average (allowable) level of

contamination at $Z_c = 16\text{--}32$; high level at $Z_c = 32\text{--}128$; extremely high level at $Z_c > 128$.

For ash and slag wastes, the overall parameter of contamination was $Z_c = 43$, which corresponds to high level. That means that this man-made horizon is dangerous.

Migration of elements from ash and slag wastes is determined by the properties of rocks embedded beneath the ash stockpiles. Within the ash stockpiles, the basis for the ash and slag layer comprises silt loams, but they do not provide complete isolation of groundwater from hydro alluvium. Thus, HM migrate to groundwater and soils around the stockpiles through infiltration of water from atmospheric precipitations, drainage from water communications, filtration of water through the base of ash stockpile of the Zmiiv TPP.

To determine the contamination of soils near ash stockpiles, we analyzed soil at the distance of 0, 5, 10, 50 and 100 meters from the stockpiles.

We determined decrease in concentration of HM in soil further away from the ash and slag stockpiles (Fig. 5). At the distance of 100 meters from the stockpiles, we saw excesses of TLVs of Ni, Cu, As, Cr (Fig. 6). Concentration coefficient was more than 1 for Cr, As, Cu, Cd, Ni. Only at the distance of 100 meters, did the concentrations of Pb and Zn reach the

background values. Increase in Fe concentration at the distance of 100 meters from the ash stockpiles, compared with the sample at the distance of 50 meters, had no pattern, and may be explained by error, for its increase was only 2.4%, or by dust transfer (Fig. 4).

Calculation of total parameter of soil contamination (Fig. 7) allowed us to identify these soils as moderately dangerous and acceptable. However, there are several significant disadvantages to the Zc parameter. In particular, it does not take into account differences between the potential dangers from chemical elements, and also, most importantly, synergic effects of polymetallic contamination.

Coefficient of synergic impact of heavy metals, which was determined using the formula:

$$\sum_1^n \frac{C_i}{TLV_i} \leq 1, \quad (4)$$

equaled 26.64 (in soil of ash stockpiles), further decreased, but even at the distance of 100 meters it was 11.23, i.e. at the distance of 0-100 m from the ash stockpiles, the condition of absence of excess of coefficient of total action of the unit was not satisfied.

Petrographic analysis using the method of immersive preparations determined presence of poorly melted quartz grains in the ash with distinctive parameters of refraction. In the periphery, isotropically with $N=1.470-1.490$, and in the central part similarly to quartz with $N_e=1.554$, $N_o=1.543$. There also occur non-transparent brown ashes with semi-metal gloss of grain of iron hydroxides, and also white, translucent, poorly fibered grains of, probably, wollastonite $CaSiO_3$, $N_q=1.632$, $N_p=1.619$. Often, there occur yellowish brown grains with $N_m=1.645$ represented probably by compounds of aluminosilicates of iron. Glass is also composed of mostly iron- aluminosilicates with $N=1.625-1.638$.

Ratio of mineral phases and glass varied, though we have to note the dominance of aluminosilicates, silicates of calcium and glass in ash and slag wastes (Fig. 8).

Compounds of HM are mostly confined to amorphous clayey aggregates and soot-carbon formation of ash, to a lower degree to slag glass and even less to grains of quartzite sand. "Ash: and slag particles: quartzite filling" was in the ratio 8-12: 61-64: 20-31%.

Migration of HM from ash and slag to the components of the ecosystem depends on many factors, one of the essential being the acidity of the environment. Earlier, we examined migration properties of heavy metals by developing concentration logarithmic diagrams (Buts Y., Kraynyuk O., Asotskyi V., 2020; Y. Buts, V. Asotskyi, O. Kraynyuk, 2019; Buts, Y., Asotskyi, V., Kraynyuk, O, 2018; Krainiuk O., Buts Yu., 2018; Krainiuk O., Buts Yu., Nekos A., 2019). Thus, one may assume that Cu, Ni, Zn and Cr are characterized by low mobility in soil, thus accumulating near the ash stockpiles, which is due to neutral and poorly-alkaline values of soil pH ($pH=8,0...8,5$).

Usually, the contamination of territory around stockpiles of ash and slag takes place only due to migration of HM from ash and slag, and due to wind transfer of dry ash and slag from the surface of ash stockpiles, dust.

Conclusions

Soils are depositing environments, their condition may be considered an integral parameter of the long term process of contamination of ecosystems. Furthermore, contamination of soils is related to contamination of the troposphere – the layer closest to Earth’s surface, surface waters and groundwater.

We determined ash and slag of the Zmiiv TPP to have poorly melted grains of quartz, iron hydroxides, wollastonite, aluminosilicates of iron and slag glass.

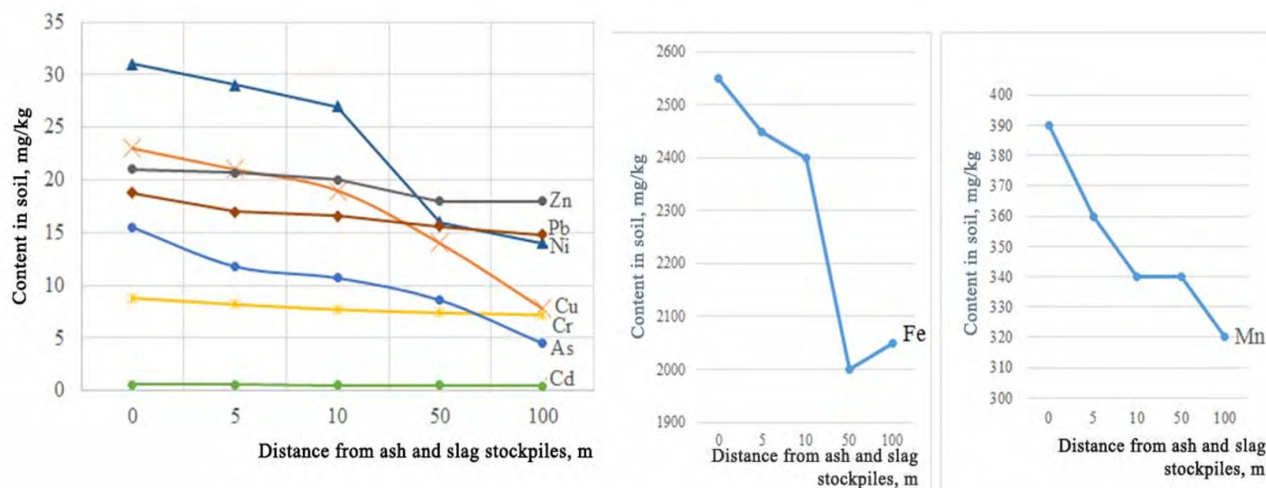


Fig. 5. Dependence of HM content in soil on distance from stockpiles of ash and slag wastes

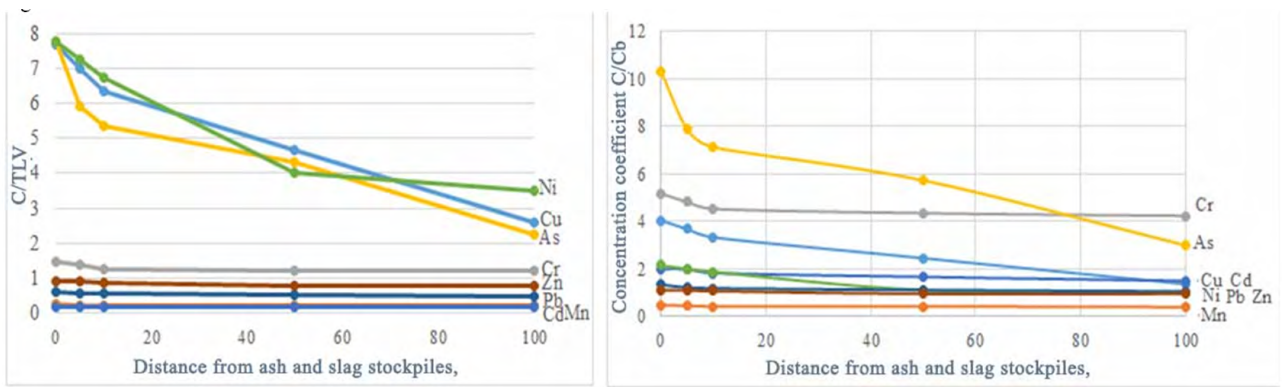


Fig. 6. Dependence of HM content in soil on distance from stockpiles of ash and slag wastes

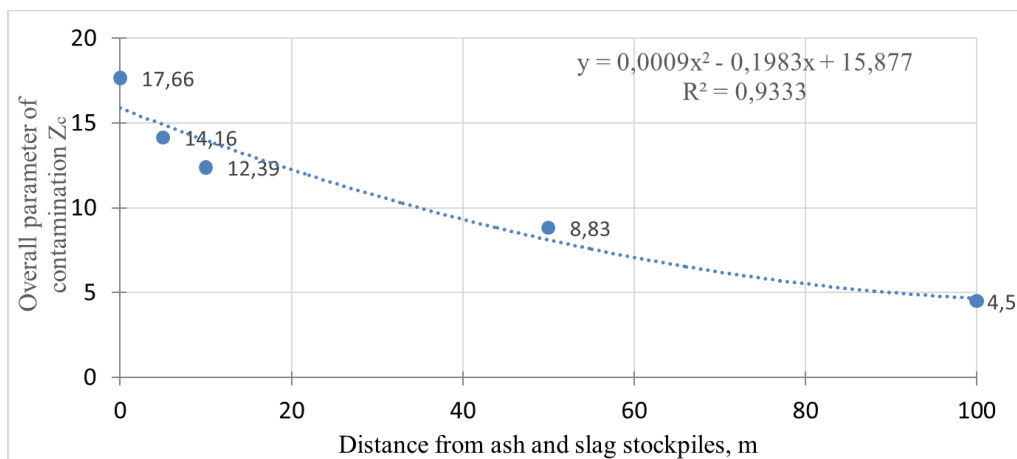


Fig. 7. Dependence of total parameter of contamination of soils on distance from stockpiles of ash and slag

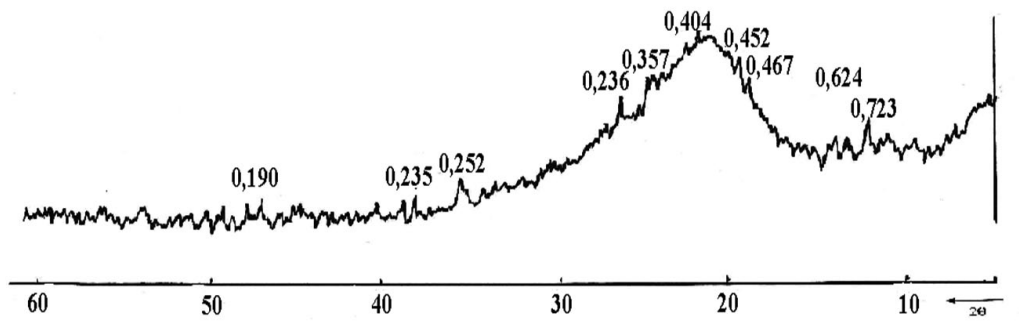


Fig. 8. X-ray diagram of ash and slag wastes of the Zmiiv TPP

Ash and slag of the Zmiiv TPP contain Cu, Cr, As, Cd and Ni in concentrations that several times exceeded the TLV.

As the ash and slag wastes of the Zmiiv TPP are deposited, the contaminated water infiltrates, adversely affecting the geochemical properties of the soil and the hydrochemical properties of surface waters and groundwater.

Even, at the distance of 100 meters from the stockpiles, there were observed excesses of TLVs of Ni, Cu, As, Cr in soil. Concentration coefficients exceeded one for Cr, As, Cu, Cd, Ni. Only at the

distance of 100 m did the contents of Pb and Zn reach the background values.

Because ash and slag wastes contain such fractions that may be easily transferred by wind, we may presume that introduction of HM to the ecosystem takes place by wind, which also contributes to the pollution of the atmospheric air. The problem of utilization of ash and slag wastes should be solved by producing construction materials, and using them in road building, but the composition of ash and slag and probability of migration of TH depending on conditions of use should be studied.

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