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TRACE METALS' MIGRATION FROM SOIL TO SEEPAGE WATER

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Abstract. In this paper the rate migration of trace metals (Pb, Cd, Cu, Zn) in polluted turf-podzol sandy loam soil was investigated in lysimetric experiment. Koller's model was applied to obtain the ranking of the metals according to their rate of migration in soil to seepage water. Among the studied metals, the most intense migrant in the soil to seepage water was lead. Lead had highest and earliest maximum concentration in seepage water. Cooper had weak migration intensity. According to metals migration ability, they can be ranked in the following descending order: Pb > Cd > Zn > Cu. Findings are relevant to assessing the metal as dangerous.

Keywords: trace metals, migration, soil profile, pollution, lysimeter

INTRODUCTION

According to the World Health Organization, among pollutants that have a negative impact on humans, toxic metals are in the second place after pesticides and far ahead of such known environmental pollutants as carbon dioxide and sulfur (Kabata-Pendias 2010, Houben and Sonnet 2014). The metals such as cadmium, lead and mercury are common soil pollutants, being emitted mainly as a result of various industrial and agriculture activities (Soetan *et al*. 2010,

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Titov *et al*. 2014, Chen *et al*. 2016). Although in some case their levels are low, they contribute to the deposition and build-up in soils. Toxic metals are persistent in the environment and are subject to bioaccumulation in food-chains. Cadmium exposures are associated with kidney and bone damage (Kutsenko 2004, Sarkar 2018). This metal has also been identified as a potential human carcinogen, causing lung cancer. Lead exposures have developmental and neurobehavioral effects on fetuses, infants and children, and raise blood pressure in adults. Despite the fact that microelements such as copper and zinc are essential for life, in excess they can also cause acute toxicity (*Health*… 2007). Additionally, through their insidious mode of action metals are notorious for promoting many chronic conditions including carcinogenesis (Mamtani *et al*. 2011).

Various studies show that over the past decades, the metal content in the environment – in air, water, and soil – has been steadily increasing (Sherameti and Varma 2015, Jaishankar *et al*. 2015, Singh *et al*. 2016, Tutic *et al.* 2015, *Trace*... 2001). This is due to the rapid development and active work of industrial enterprises, the sharp increase in the number of vehicles, the annual introduction of fertilizers to the soil (Chang *et al*. 2014). Metal pollution can arise from many sources but often arises from metal purification processes, such as the smelting of copper and the preparation of nuclear fuels. Electroplating is the primary source of chromium and cadmium. Soil metal concentrations are dynamic; these concentrations depend on variety of natural and anthropogenic sources (such as agricultural land management, atmospheric deposition, fertilization, and irrigation), as well as the output pathways that take elements out of the soil layer (Xia *et al*. 2013). Therefore, metals concentration in the environment as a result of human-made incomes will increase. According to the "National Report of Ukraine", 27,900 tons of metal compounds came into the atmosphere as a result of the work of 3,643 enterprises in 2014 (*National*… 2016). In general, in Ukraine the soils are most polluted with lead, zinc, copper, and less with manganese, cadmium and nickel. A few polluted sites were formed around the enterprises as a result of metals emissions during many years.

Metals have a long half-life in soil with the preservation of toxic properties; accumulate in living organisms (Jaishankar *et al.* 2014, Mukti 2014, Hazrat *et al*. 2019). Thus, the half-life (T_{50}) of mobile forms in a 0–20 cm layer of soddy-medium podzolic soils and black soil is, respectively, for: Cd – 0.87–1.0 years, Pb – 0.96–1.1 years, Cu – 0.53–0.6 years, Zn – 0.65–0.8 years (Ryzhenko and Kavetsky 2013). Through precipitation of their compounds or by ion exchange into soils and mud, metal pollutants can localize and lay dormant. Unlike organic pollutants, metals do not decay and thus pose a different kind of challenge for remediation (*Adverse*... 2011). It is believed that the most mobile elements in soil samples are Mn, Cd and Pb, whereas the least mobile elements are Ni, Zn and Cu (Sungur *et al*. 2014). In studies by Xueqi Xia *et al*. (2013) for Cd, Ni, Pb, Zn, and Cu, almost 100% of the output fluxes were from seepage water.

The presence of trace metals in ground waters is usually related to human industrial activities. Vertical displacement and leaching of metals may occur in the soil profile and contaminate the ground water. Urban runoff may contain significant concentrations of iron, lead, zinc, and copper, and to a lesser extent, cadmium, chromium, mercury and nickel (Amin *et al*. 2011). To evaluate metals contamination in seepage water, it is advisable to investigate this issue with the use of a lysimetric experiment to understand their migration properties and their influence on the ecosystem and the quality of agricultural crop production. It is very important for soils with weak buffering capacity such as turf-podzol sandy loam soil. However, turf-podzol soils make up nearly one third of the Ukrainian arable lands. The assessment of the fact that metals are dangerous to human health and ecosystem is not possible without revealing their migration properties in the soil profile. Mobility is a concept frequently used to estimate the risk of contamination of other environmental compartments (Domergue and Vedy 1992). The highlighting of metals migration peculiarity in soil allows predicting the process of increasing of metal present in ground water and in lower horizons of the soil profile after the occurrence of impact pollution and also makes it possible to measure the quality of the soil for crop growth.

The aim of this investigation was to assess the rate migration of metals in polluted turf-podzol sandy loam soil in a lysimetric experiment.

MATERIALS AND METHODS

Research on migration in the soil profile can be carried out by different methods. Lysimetric method is one of the best for studying the pollutants' speed migration as well as for researching the amount of toxicants in seepage water. Very often lysimetric method is used for highlighting the period of the appearance of pollutants in seepage water (or in different soils horizons up to parent rock) and transformation of trace elements in soil. The experimental investigation was conducted at Chernigiv Institute of Agrotechnical of the Ukrainian Academy of Agrarian Science (now Institute of Agricultural Microbiology and Agro-Industrial Production). Sod podzolic soil has the following physic chemical characteristics: pH salt 5; organic matter by Turin, Walkley-Black 1.1%, CEC 6.3 cmol/kg. Control concentration of trace metals in soil (1 MHCl, mg kg^{-1}) was: Cd – 0.1; Pb – 0.3; Cu – 0.92; Zn – 2.4; Ni – 1.1; Co – 1.5. Control concentration of metals in seepage water was (mg kg^{-1}): Cd – 0.005; Pb – 0.035; Cu – 0.2; Zn – 2.5. Determination of heavy metals was conducted using thin layer chromatography (TLC). The method is based on the extraction of metal ions from solutions by diphenyldiithiocarbazone (ditizon). Complex compounds of metals (dithizonates) are formed in a certain range of pH. Further, the colored dithizonates of metals are identified by chromatography in a thin layer of the adsorbent with qualitative and quantitative determination. This method is officially recognized in Ukraine (Kavetsky *et al.* 2001). The experimental scheme was: 1 – Control; Cu – 500, Zn – 1,500, Pb – 150, Cd – 15 mg kg^{-1}. We used the following salts of metals: $Pb(NO_3)_2$, $ZnSO_4$, H_2O, $CuSO_4$, $7H_2O$, $CdSO_4$. The investigation was conducted in the conditions of stationary soil lysimeters. The rate of migration of lead, zinc, copper, and cadmium in the soil profile was studied. Seepage water was selected step by step seasonally. An average sample was sampled for analysis during the periods of March–May (spring filtration), June–August (summer filtration), September–November (autumn filtration) during the six-year period. The mixed metal compounds were once added to the soil surface in the spring of the first year of the study. Mean standard deviations, variance, and minimum, maximum, standard errors were calculated in four replicates. The experimental results were interpreted using standard statistical methods.

Stationary soil lysimeters consist of 48 sections in two parallel blocks. Under each lysimetric section (cells) there are receptacles for collecting filtrate. Lysimeters are of a concrete, bulky type. The lysimetric cells are filled with soil consistently, starting from the parent rock, taking into account the power of the genetic horizons. The cut-off scheme of a lysimetric cell with soil (Sod podzolic soil) is shown in Fig. 1.

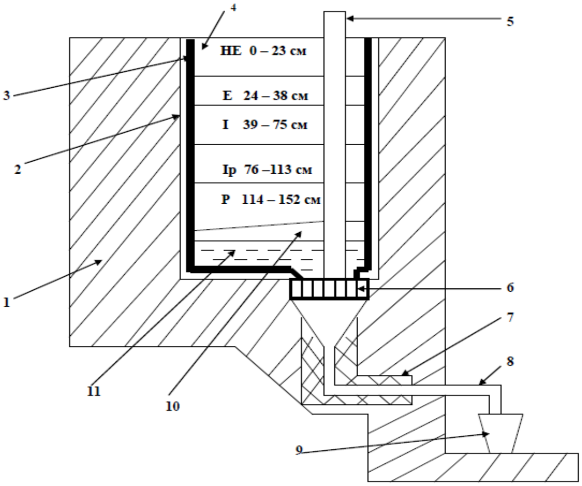

Fig. 1. Slice-scheme of a lysimetric cell with soil

1 – external wall (concrete); 2 – waterproofing 3-cm plaster + 2 layers of ruberoid on bitumen, gluing with sheet polyisobutylene and staining it with bituminous varnish; 3 –waterproof; 4 – the soil according to the genetic horizons that are in the field; 5 – drainage vertical pipe (for washing the disk); 6 – drainage polyvinyl plated disc; 7 – mineral wool; 8 – discharge filtration pipe; 9 – receiver for collecting filtration water; 10 – river sand (10 cm); 11 – bit glass (8 cm).

RESULTS AND DISCUSSION

The dynamics and character of metal filtration in seepage waters are presented in Table 1.

Table 1. Metals' filtration in seepage water

	Period of seepage in lysimeter	Cd	Pb	Zn	Cu
1	1* s.	0.030** 0.005±0.001	0.033 0.050±0.01	0.020 0.300±0.05	0.010 0.050±0.05
2	1 – sum.	0.040 0.006±0.001	0.043 0.065±0.01	0.034 0.510±0.10	0.020 0.100±0.10
3	1–2 a–w	0.680 0.102±0.05	0.700 1.05±0.20	0.550 8.250±0.34	0.300 1.500±0.30
4	2 – s.	0.400 0.060±0.02	0.410 0.615±0.01	0.280 4.200±0.23	0.130 0.650±0.10
5	2 – sum.	0.400 0.060±0.01	0.440 0.66±0.01	0.300 4.500±0.22	0.150 0.750±0.10
6	2–3 a–w	1.420 0.213±0.05	1.500 2.25±0.20	0.700 10.50±0.56	0.77 3.85±0.40
7	3 – s.	0.90 0.14±0.05	1.00 1.50±0.10	0.700 10.50±0.42	0.50 2.50±0.30
8	3 – sum.	1.00 0.15±0.05	1.200 1.80±0.10	0.850 12.75±0.63	0.55 2.75±0.30
9	3–4 – a–w	1.500 0.225±0.07	1.500 2.25±0.07	1.00 15.00±0.65	0.70 3.50±0.40
10	4 – s.	1.550 0.233±0.06	1.900 2.85±0.10	1.280 19.2±0.45	0.80 4.00±0.56
11	4 – sum.	1.670 0.251±0.07	2.100 1.95±0.20	1.350 20.25±1.9	0.90 4.50±0.50
12	4–5 – a–w	1.80 0.270±0.05	2.15 3.225±0.30	1.450 21.75±2.50	1.00 5.00±0.55
13	5 – s.	1.80 0.27±0.04	2.130 3.195±0.30	1.600 24.00±4.4	1.20 6.00±0.75
14	5 – sum.	1.850 0.278±0.07	2.050 3075±0.3	1.600 24.00±3.5	1.30 6.50±0.70
15	5–6 – a–w	1,850 0.278±0.08	1.90 2.85±0.03	1.700 25.50±5.0	1.30 6.50±0.80
16	6 – s.	1.80 0.27±0.05	1.800 2.70±0.03	1.740 26.1±4.1	1.30 6.50±0.80
17	6 – sum.	1.740 0.261±0.05	1.770 2.66±0.03	1.750 26.25±5.0	1.40 7.00±0.95
18	6–7 – a–w	1.700 0.255±0.06	1.680 2.52±0.04	1.750 26.25±4.5	1.40 7.00±0.90
19	7 – s.	1.650 0.248±0.06	1.500 2.25±0.04	1.780 26.70±3.2	1.45 7.25±0.90
20	7 – sum.	1.550 0.233±0.07	1.430 2.15±0.0	1.70 25.5±2.1	1.45 7.25±0.95
	S^2	*0.38*	*0.46*	*0.37*	*0.25*

* 1, 2 … – researching year; sum.– summer filtration, a–w – autumn–winter filtration, s. – spring filtration.

** in the numerator: metals concentration, % of the applied amount; in the denominator: metals concentration, mg l^{-1}. S^2 was obtained for metals concentration, % of the applied amount.

Among the studied metals, the most intense migrant in the soil was lead. Its amount was highest in the seepage water. At the same time, copper was characterized by the lowest amount in seepage water (Table 1). The change of the metals concentration in seepage water is presented in Fig. 2.

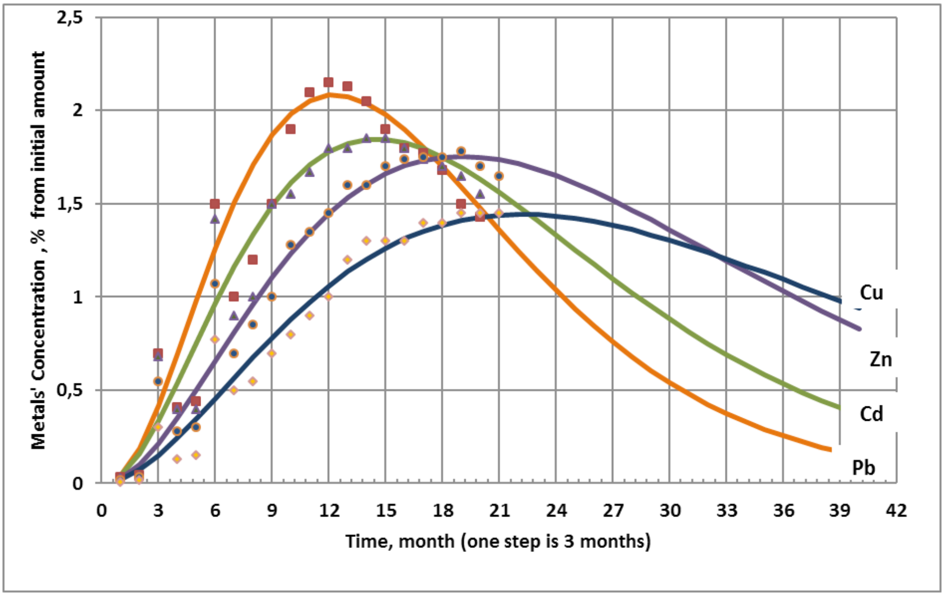

Fig. 2. The dynamic of metals' filtration in seepage water

The dynamics of metal filtration in seepage water was formalized using the Koller equation:

$$y = a_0 \times x^{a1} \times e^{a2x} \tag{1}$$

where: a_0, a_1, a_2 are regression parameters (Ryzhenko and Kavetsky 2013). They are presented in Table 2.

Table 2. Parameters of regression in the Koller equation describing the migration of metals, P_{05}

	Pb	Cd	Zn	Cu
a_0	0.0525	0.0478	0.0300	0.022
a_1	2.4550	2.1795	2.0916	1.994
a_2	-0.2000	-0.1500	-0.1100	-0.090
R^2	*0.88*	*0.92*	*0.94*	*0.96*

Equation (1) allows to predict the metal concentration in seepage water (R^2 = 0.97–0.99). For example, 13 years after the contamination of seepage water with Cd, it will present about 1% of the initial amount of Cd. This simulation model also makes it possible to develop a strategy for safe agriculture on polluted soils as well as for environment control. In addition, the prediction of the time of metals' maximum concentration appearance in seepage water allows to assess the dangerousness of pollutants' dangerous. On the one hand, the faster the metal maximum concentrations in the seepage water appear – the more intensive the metal's migration into the soil profile, and, therefore, metal is more dangerous to the environment. After all, the metal on the way of its migration can come into contact with other components of the ecosystem and negatively affect them. On the other hand, high-speed migrant could soon leave the ecosystem, and the metal content for uptake by plant roots will decrease. As a result, plants could accumulate less metal. Metal's maximum concentration in seepage water could be calculated as global maximum (extrema) of the function (1) (Guryanova *et al.* 2014). Maximum concentration for each metal was different as well as time of appearance of maximum concentration in seepage water (Table 3). Pb was characterized as the most intensive migrant. Lead had the highest and earliest maximum concentration in seepage water. Cooper had weak migration intensity: its predicted maximum concentration was lowest and appeared last. An extremum of the function was calculated as follows:

$$a_0 > 0, a_1 > 0, a_2 < 0. \tag{2}$$

We should find the maximum of this function in the interval $[0, \infty)$. To do this, we used a differential calculation:

$$y' = a_0 \left(a_1 x^{a_1 - 1} e^{a_2 x} + a_2 x^{a_1} e^{a_2 x} \right) = a_0 a_2 x^{a_1 - 1} e^{a_2 x} \left(x + \frac{a_1}{a_2} \right) \tag{3}$$

Investigating the change of the sign of the derivative when passing through a critical point, we come to the conclusion that this is the point of maximum:

$$x = -\frac{a_1}{a_2} \tag{4}$$

The maximum concentration is calculated as follows:

$$y_{\max} = y\left(-\frac{a_1}{a_2} \right) = a_0 \left(-\frac{a_1}{a_2} \right)^{a_1} e^{-a_1}. \tag{5}$$

The extrema for Pb were obtained by experimental and theoretical methods. The experimental data is presented in Table 1. The calculating of Pb extrema (concentration and time) by the theoretical method was obtained as follows:

$$x = -\frac{1.8455}{-0.09} = 12.28 \tag{6}$$

$$y = 0.0525\,(-\frac{2.455}{-0.2})^{2.455}\; e^{-2.455} = 2.13 \tag{7}$$

Thereby, the maximum Pb concentration was 2.13% from the initial amount, and time of its appearance was 36.84 months (3.1 year). Similarly, the extrema for other metals were calculated (Table 3).

Table 3. Extremas of function describing the migration of metals from soil to seepage water

	Pb	**Cd**	**Zn**	**Cu**
T_{extr} (x)	12.28	14.53	19.02	22.16
$Conc_{extr}$ (y)	2.13	1.84	1.75	1.48

The metal's dissipation constant (*k*) in soil (0–20 cm) as well as the half-life period (years^{-1}) was presented in previous papers (Ryzhenko and Kavetsky 2013, Ryzhenko *et al*. 2005). This constant was obtained by the exponential model:

$$\left(k \quad \frac{2{,}303}{t} \lg \frac{C_0}{C_t} \right) \tag{8}$$

where: C_t – final concentration; C_o – initial concentration; *k* – dissipation constant of Metal; *t* – time or period, days.

Metals' dissipation constants (*k*) were: Pb – 0.6381, Cd – 0.6987, Zn – 0.9116, Cu – 1.2158. Metals' dissipation in the soil increased with the value of the constant (Ryzhenko and Kavetsky 2013). The metal's dissipation constant (*k*) in soil (0–20 cm), first of all, depends on bioavailability of metals for the investigated plants growing in the field. In soils, the majority of toxic metals is in the upper soil layer and binds to humus substances due to the biological transfer of metals from the lower layers of the soil to microorganisms and root systems of plants, as well as to the modes of soil evaporation. There are many factors which have influence on decreasing the mobile (or available for plants) forms in 0–20 cm layer in soil: the content of organic matter of the soil, pH, plants species, etc. Also many factors contribute to the speed migration of metals in deeper soil profile. The soil texture plays an important role in mobility of metals. Texture reflects the particle-size distribution of the soil and, thus, the content of fine particles like oxides and clay. These compounds are important adsorption media for metals in soils. The clay soil retains high amount of metals when compared to sandy soil. The fine textured soils contain higher amounts of Pb and coarse textured soil contains lower amount of lead (Chambers *et al*. 1991, Sherene 2010). One of the important factors of metals' migration rate is the peculiarity of distribution in the soil profile. It depends on structure features of the soil profile. For example, the black soils are characterized by radial type of vertical migration, while turf-podzol sandy loam soil – by "eluvially-illuvial" type of trace metals' vertical migration. Organic matter is important for the retention of metals by soil solids, thus, decreasing mobility and bioavailability. The pH of the soil solution maintained at neutral to slightly alkaline condition showed low mobility of all metals. In general, sorption increases with increasing pH. That is, the lower the pH value, the more metal can be found in solution and thus more metal is mobilized. When pH falls to below 5, mobility is enhanced as a result of the increased proton concentration. With pH values above 7, some heavy metals tend to form hydroxy – complexes which will increase the solubility of the metal in question. Adsorption was greater at pH 6.5 than at pH 4.5 (Sherene 2010). The amount of metals adsorbed decreases with increasing ionic strength for minerals with permanent surface charge density (Tsvetkova and Gunko 2015). When the temperature raises, the metal activity in the soil solution may be more active and has faster absorption rates (Sherene 2010). The amount of metals adsorbed decreases with increasing ionic strength for minerals with permanent surface charge density (Tsvetkova and Gunko 2015). However, physical and chemical properties of metals as well as other pollutants also play an important role in their migration characteristic (Ryzhenko *et al*. 2017, Ryzhenko 2019).

Similarly, to the dissipation constant, we assumed that the parameters of regression in Koller's equation could present the rate of migration of metals. Metals mobility to seepage water increased with the value of a_1 and a_2. We obtained the close connection between a_1, a_2 and k (R^2 = 0.72–0.79) (Fig. 3). However, descending order of metals according to their dissipation in 0–20 cm soil layer was reversed to descending order of metals according to their migration to seepage water. For example, cooper had least T_{50} (0.6 years^{-1}) in the 0–20 cm soil layer and weak migration intensity to seepage water.

According to a_1 and a_2, metals can be ranked in the following descending order: Pb > Cd > Zn > Cu.

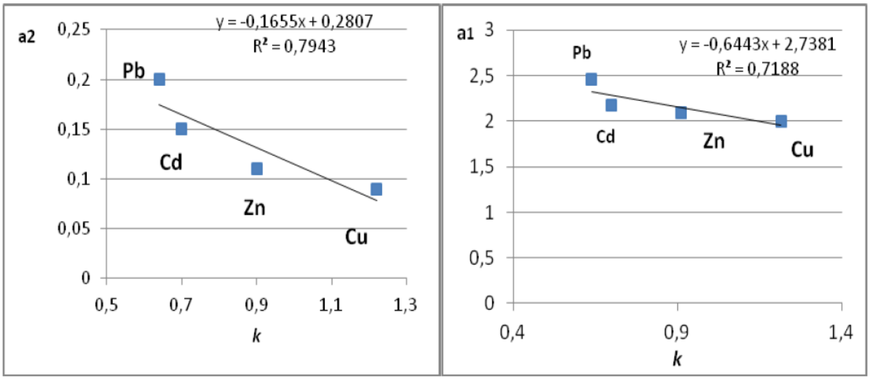

Fig. 3. Correlation between a_2, a_2 and *k*

CONCLUSIONS

Pb was characterized as the most intensive migrant. Lead reached the highest and earliest maximum concentration in seepage water. Cooper had weak migration intensity: its maximum concentration was lowest. The migration of Cd, Cu, Zn, Pb in the soil profile to seepage water was described using the Koller model. This model allows predicting the evolution of the amount of metal present in seepage water after the occurrence of impact pollution. The parameters of regression in Koller's model (a_1 and a_2) were proposed as indicators of metal migration rate. We obtained the following ranking of the metals Pb > Cd > Zn > Cu according to the values of a_1 and a_2. We also obtained the quite close connection between a_1, a_2 and dissipation constant (k) ($R^2 = 0.72$–0.79). It can be concluded that the parameters of regression in Koller's equation not only characterize the migration ability of the metal to seepage water, but also make it possible to measure the quality of the soil for crop growth and environment control.

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