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Dynamics of migration property of some heavy metals in soils in Kharkiv region under the influence of the pyrogenic factor

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Abstract. In soils after fires trace metals sharply change their migration ability and can form poorly-soluble hydroxides which are hazardous chemical formations, the nature of which has not been fully explored until now. In addition, in interstitial water, there is a probability of the formation by metals of hydroxocomplexes with different amounts of hydroxide ions. We studied the range of dynamics of migration capacity of sedimentation of hydroxides and the region of predominance of soluble hydroxocomplexes by developing logarithmic concentration diagrams (LCD). We developed logarithmic concentration diagrams, the equation of formation of prevailing forms, using which it is possible to clearly determine the regions of maximum sedimentation (accumulation) of hydroxides and hydroxocomplexes of heavy metals after the influence of the pyrogenic factor. The obtained calculations of the results of the predictive modeling of the dynamics of migration capacity and postpyrogenic migration geochemical processes in ecogeosystems have been organized and systematized. The determined patterns can be useful for the analysis of possible geochemical migration (accumulation) of heavy metals in ecological systems in the study of technogenic and ecological situation after fires. Based on the calculations made, mathematical models of heavy metals` behaviour are developed, which are useful for drawing up a forecast estimation of the dynamics of their geochemical migration and accumulation in ecological systems as a result of the influence of the technogenic loading of the pyrogenic factor. The conditions of concentration and migration of compounds of heavy metals were determined, and the equation for calculating the concentration of mobile forms of trace metal compounds has been developed. The developed map of the activity of geochemical migration of heavy metals under the influence of the technogenic loading of pyrogenic factor will make it possible to elaborate the migratory capacity of trace metals and provide a forecast of their behaviour in ecological systems after fires. This will allow preventive measures to be taken to ensure environmental safety and prevent adverse effects on human health and the condition of the components of the environment. The creation of similar cartographic material may be extrapolated to other regions of Ukraine, affected by technogenic loading of pyrogenic factor. The development of logarithmic concentration diagrams allows us to predict the capacity of compounds of lead, nickel, chromium,and copper for migration or accumulation of heavy metals due to changes in the acidity of soils under the influence of the pyrogenic factor. Having used the map of the soils of the Kharkiv region, we analyzed and provided a forecast of the migration ability of lead compounds in cases of fire in different types and subtypes of different environmental conditions.

Key words: natural fires, heavy metals, migratory properties of chemical element, map of soils.

Динаміка міграційної здатності деяких важких металів у ґрунтах Харківського регіону під дією пірогенного чинника

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Анотація. У ґрунтах після пожеж важкі метали різко змінюють свою міграційну здатність і можуть утворювати важкорозчинні гідроксиди, які є небезпечними хімічними утвореннями, природа яких до цього часу повністю не досліджена. Крім того, у ґрунтовому розчині є ймовірність утворення металами гідроксокомплексів з різною кількістю гідроксид-іонів. Діапазон динаміки міграційної здатності осадження гідроксидів і області переважання розчинних гідроксокомплексів вивчені нами за допомогою побудови концентраційно-логарифмічних діаграм (КЛД). Побудовано концентраційно-логарифмічні діаграми, виведено рівняння утворення переважаючих форм за допомогою чого можна чітко визначити області максимального осадження (акумуляції) гідроксидів та гідроксокомплексів важких металів після впливу пірогенного чинника. Отримані розрахунки результатів прогностичного моделювання динаміки міграційної здатності та постпірогенних міграційних геохімічних процесів в екогеосистемах нами були упорядковано та систематизовано. Виведені закономірності можуть бути корисними для аналізу можливої геохімічної міграції (акумуляції) важких металів в екогеосистемах при вивченні техногенно-екологічної ситуації після пожеж. На підставі проведених розрахунків розроблено математичні моделі поведінки важких металів, які корисні для складання прогнозної оцінки динаміки їх геохімічної міграції та акумуляції в екогеосистемах в результаті впливу техногенного навантаження пірогенного походження. Встановлено умови концентрування і міграції сполук важких металів, виведено рівняння для розрахунку концентрації рухомих форм сполук важких металів. Побудована карта активності геохімічної міграції важких металів під впливом техногенного навантаження пірогенного походження дозволить деталізувати міграційну здатність важких металів та надати прогноз їх поведінки в екогеосистемах після пожеж. Це дозволить вжити превентивних заходів щодо забезпечення екологічної безпеки та негативного впливу на здоров'я людини та стан компонентів навколишнього природного середовища. Створений подібний картографічний матеріал можливо екстраполювати на інші регіони України, що зазнають техногенного навантаження пірогенного походження. Побудова концентраційно-логарифмічних діаграм дозволяє прогнозувати здатність сполук плюмбуму, нікелю, хрому, купруму до міграції або акумуляції важких металів унаслідок зміни кислотності ґрунтів під дією пірогенного чинника. Використавши карту ґрунтів Харківського регіону, нами проаналізовано та надано прогноз міграційної здатності сполук плюмбуму при виникненні пожежі у різних типах та підтипах різних екологічних умов.

Ключові слова: природні пожежі, важкі метали, міграційні властивості хімічних елементів, карта ґрунтів.

Introduction. The dynamics of geochemical peculiarities of migration of heavy metals under the influence of the pyrogenic factor has been studied in our previous works (Buts et al., 2018, Buts, Y., Kraynyuk, O. 2018), where we proved that after a fire, acidity of soil increases. No doubt, first of all, this is related to increase in the amount of ash and remains of combustion products which have alkaline reaction (Suchikova Y., et al., 2017). Also, an important role in migration or accumulation of elements belongs to the condition of flammable materials, particularly – moisture of forest litter. Increase in alkalinity to 15% was reported by the authors (Burlakova et al., 2002). This study is a continuation of our study published earlier (Buts, 2018, Asotskyi V., 2018).

The **objective** of this study was analysis of the dynamic of migrational ability of heavy metals in soils of Kharkiv Oblast under the impact of the pyrogenic factor. **For this purpose, we solved the following tasks**: suggesting a mathematical model of behaviour of heavy metals for predictive assessment of their geochemical migration and accumulation in ecogeosystems as a result of impact of technogenic load caused by the pyrogenic factor; determining conditions of concentration and migration of compounds of heavy metals and their dynamic, finding a mathematical description of concentration of mobile forms of compounds of heavy metals.

Materials and methods. Reduction of acidity of the soil column contributes to increase in oxidizing-restorative potential, which reflects in maintaining of exchangeable Ca^{2+} and Mg^{2+}. Change in soil pH also affects the migration property of heavy metals.

As we know, different plants differently accumulate various microelements, including heavy metals. That is, one should take into account also the pattern of distribution of heavy metals in the above-ground organs of plants. This pattern is related to various parameters of dynamics of geochemical migration of chemical elements during and after fire .The most characteristic parameter is radial distribution of most heavy metals (HM) in soil section, including the upper soil horizons with interlayers of fragments of plant litter and forest litter. In this case, HM concentration in radial differentiation in the soil column significantly fluctuates, which is well described in scientific publications (Neshovorova, 2014).

Obviously, heavy metals in soils can form poorly-soluble hydroxides. Furthermore, in soil sections, there is a possibility of metals to form hydroxocomplexes with different amounts of hydroxide-ions. We studied the range of sedimentation of hydroxides and the area of predominance of soluble hydroxocomplexes by developing logarithmic concentration diagrams (LCD) (Buts, 2018).

The obtained calculations can be used for predicting dynamics of geochemical migration of heavy metals in soils after technogenic consequences of disasters caused by pyrogenic factors.

Using such calculations, we have developed a logarithmic concentration diagram, according to which one can clearly designate areas of maximum sedimentation of hydroxides of metals.

Predicted model of migration of compounds of lead in ecogeosystems. We have conducted a study attempting to determine the probability of mathematical regularity and its dynamic in formation of migration of poorly-soluble compounds of heavy metals in soils after technogenic impact caused by the pyrogenic factor. For lead, the following pattern is observed: at pH<7, all lead in the soil will be present in soluble form; at pH=8, concentrations $[Pb^{2+}]$ will equal higher than 0.01 mol/l, i.e. at shift of pH to more alkaline area, solubility of plumbum compounds steeply decreases. At pH=9, concentrations $[Pb^{2+}]$ will not exceed 10^{-4} mol/l. At pH=9–13, in the soil environment, particles of $[Pb(OH)^{+}]$, $Pb(OH)_2$ and $Pb(OH)_3^-$ will be present, solubility of which is very low, i.e. in low-alkaline and alkaline environment, compounds of lead accumulate, and therefore their migration is not possible (Fig. 1).

Developing dependency of concentration of most probable ions $[Pb(OH)_n^{2-n}]$ on pH environment and line of trend (Fig. 1) allows one to make a predictive assessment of migration or accumulation of compounds of lead in the soil column. Dependency of soluble compounds of lead on pH can be described by the following pattern:

$$lg[Pb(OH)_n^{2-n}] = 0.0109 \cdot pH^3 - 0.0888 \cdot pH_2 - 1.891 \cdot pH + 13.064 \quad (1)$$

Reliability of approximation R^2=0.99.

Thus, among all particles $[Pb(OH)_n^{2-n}]$, Pb^{2+} are dominant in acidic and neutral environment. Their concentration in interstitial water is described by the equation (1) which is applicable for pH>6.5.

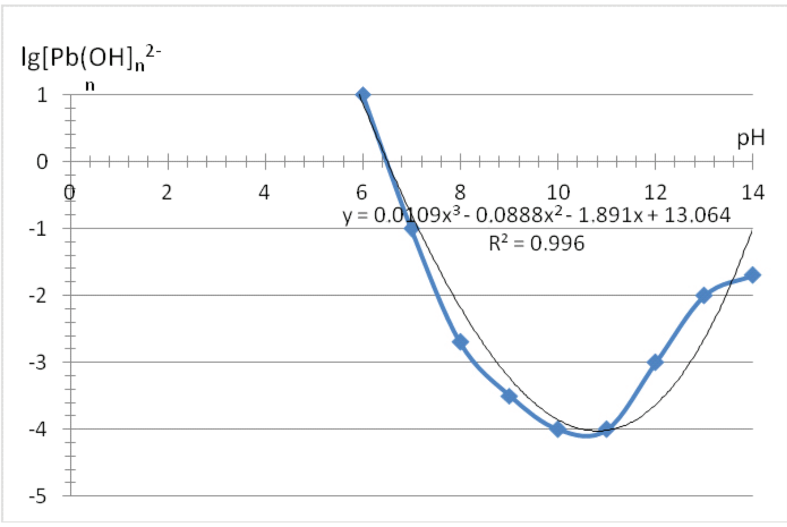

Fig. 1. Dependency of formation of particles $[Pb(OH)_n^{2-n}]$ on pH of soil environment

At pH<6.5 all lead in interstitial water will be present in soluble form as Pb^{2+}. At pH=9–13, non-soluble particles $[Pb(OH)_n^{2-n}]$ form, mainly $Pb(OH)_2$, migration of lead compounds does not take place, and its accumulation is observed.

Predictive model of migration of compounds of chrome in ecogeosystems. In acidic environments, all chrome present in the soil section will be in soluble form (Fig. 2): at pH=4, concentration $[Cr^{3+}]$ can reach 1 mol/l, but at increase in pH to 5, concentration of this chemical element $[Cr^{3+}]$ will equal only 0.01 mol/l; at pH=7–10, non-soluble $Cr(OH)_3$, $Cr(OH)^{2+}$ predominate. In alkaline environments at pH>9, soluble hydroxocomplexes $Cr(OH)_4^-$ begin to form, concentration of which at pH=11 will equal 0.001 mol/l, and at pH=12 increases by 10 times to 0.01 mol/l.

Development of dependency of concentration of most probable ions $[Cr(OH)_n^{3-n}]$ on pH environment and trend line (Fig. 2) allows one to make a predictive assessment of migration or accumulation of chrome compounds in interstitial water. Dependency of soluble compounds of chrome on pH can be described with the following pattern:

$$lg[Cr(OH)_n^{3-n}] = -0.002 \cdot pH^4 + 0.0599 \cdot pH^3 - 0.4087 \cdot pH^2 - 0.9691 \cdot pH + 6.6899 \quad (2)$$

Reliability of approximation is R^2=0.99. Thus, among all particles $[Cr(OH)_n^{3-n}]$, predominating are Cr^{3+} predominate in acidic environments. Their concentration in interstitial water is described by equation (2) applicable for pH>4. At pH<4, all chrome present in soil will be in soluble form as Cr^{3+}. At pH=7–10, non-soluble particles $[Cr(OH)_n^{3-n}]$ form, mainly $Cr(OH)_3$, migration of chrome compounds does not occur, its accumulation is observed, and only in highly alkaline environments will chrome again change to soluble form as negatively charged hydroxocomplexes with a high number of hydroxide ions.

Predictive model of post-pyrogenic geochemical migration of compounds of copper in ecogeosystems. According to the LCD that we developed, one can predict dynamic of mobile forms of compounds of copper depending on pH of environment (Fig. 3). Diagram indicates formation of ions $[Cu(OH)_n^{2-n}]$. In acid environments, Cu^{2+} ions will be present, the quantity of which in

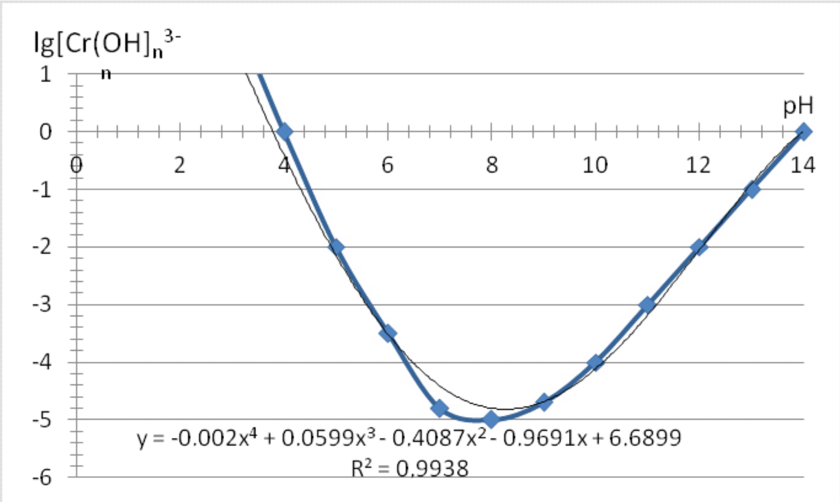

Fig. 2. Dependency of formation of particles $[Cr(OH)_n{}^{3-n}]$ on pH of soil environment

Thus, dependency of soluble copper compounds on pH can be described as follows:

$$lg[Cu(OH)_n{}^{2-n}]=0.0068\cdot pH^3–0.0089\cdot pH^2+2.2185\cdot pH+9.251 \quad (3)$$

This dependency correlates well with the developed logarithmic concentration diagram, which indicates reliability of approximation R^2=0.99.

It should be noted that among all particles $[Cu(OH)_n{}^{2-n}]$, the dominant are Cu^{2+}. Their concentration in interstitial water is described using equation (3), which is applicable at pH=3. At pH<3, all cuprum present in interstitial water is present in insoluble form.

interstitial water will steeply decrease after increase in pH. For example, at pH=4.5, concentration of ions $[Cu^{2+}]$ can reach 1 mol/l; at pH=5, $lg[Cu^{2+}]$= -1, i.e. concentration of ions $[Cu^{2+}]$ will not exceed 0.1 mol/l; and at pH=6, $lg[Cu^{2+}]$=-3, and therefore concentration of ions $[Cu_{2+}]$ will not exceed 0.001 mol/l; further at pH higher than 7, the amount of $[Cu^{2+}]$ becomes insignificant, and non-soluble $Cu(OH)_2$ forms; and only at pH close to 14, do ions of $[Cu(OH)_3^-]$ form in insignificant amount, solubility of which is slightly higher than solubility of $Cu(OH)_2$, but is still low.

For optimizing predicting migration of compounds of copper, we have developed a separate dependence of concentration of most probable ions $[Cu(OH)_n{}^{2-n}]$ on pH of environment and also developed the trend line. Using the latter, let us demonstrate the tendency in this model and make a probable prediction of the dynamics of their behaviour (Fig. 3).

Predictive model of migration of compounds of nickel in ecogeosystems. In acidic and neutral environments, mobile forms of nickel compounds predominate. At pH<5, all nickel will be present in soluble form as Ni^{2+}; at pH=5, $lg[Ni^{2+}]$=0, i.e. concentration $[Ni^{2+}]$ can reach 1 mol/l; at shift of pH to 5.5, concentration $[Ni^{2+}]$ will not be higher than 0.1 mol/l; and at pH=6, amount of $[Ni^{2+}]$ will not exceed 0.01 mol/l (Fig. 4). At pH>8, compounds of nickel will be present in non-soluble form.

For predictive assessment on dynamics of migration or accumulation of nickel compounds in interstitial water, we have specifically developed a dependency of concentration of most probable ions

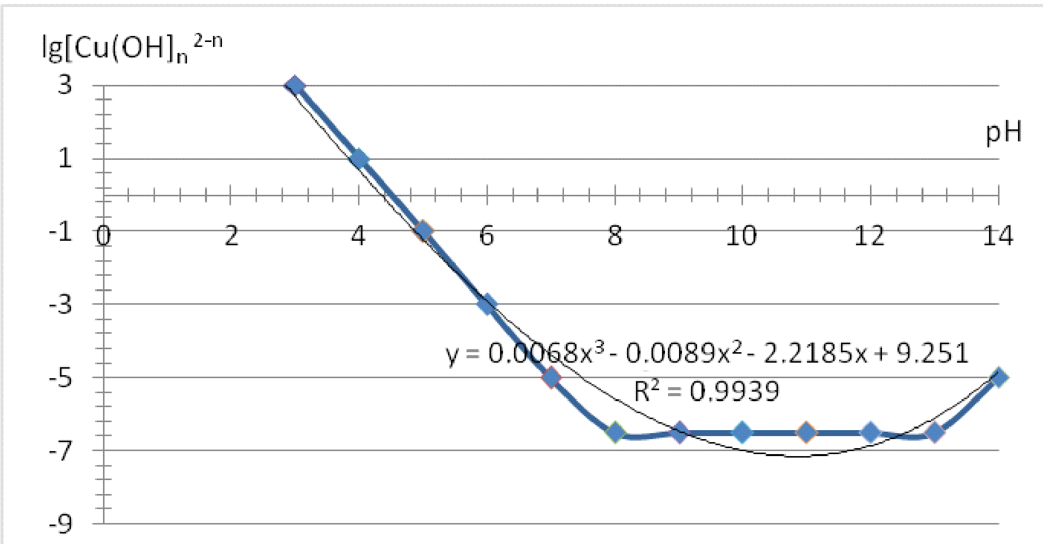

Fig. 3. Dependency of particles $[Cu(OH)_n{}^{2-n}]$ on pH on soil environment

$[Ni(OH)_n{}^{2-n}]$ on pH of environment and developed a trend line (Fig. 4). Dependency of soluble compounds of nickel on pH can be described as follows:

$$lg[Ni(OH)_n{}^{2-n}] = 0.0147 \cdot pH^3 - 0.1968 \cdot pH^2 - 1.1505 \cdot pH + 8.5013 \quad (4)$$

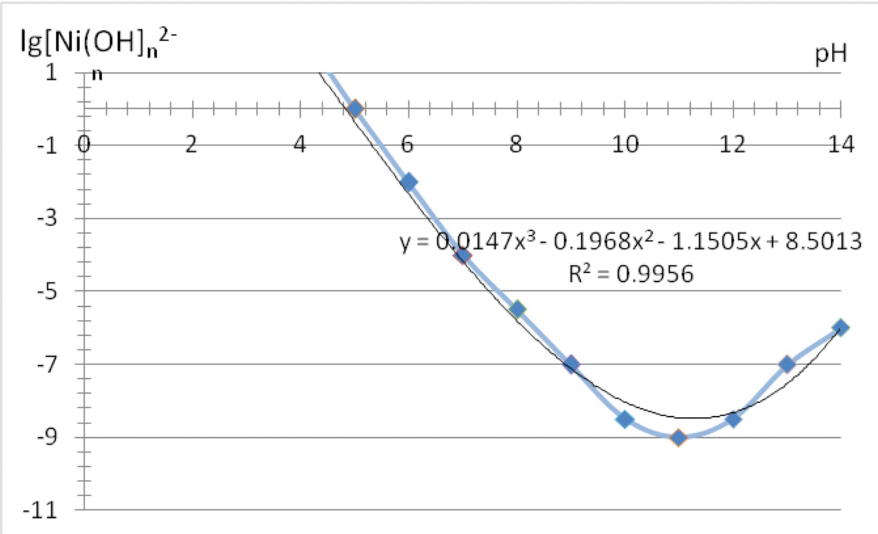

Fig. 4. Dependency of formation of particles $[Ni(OH)_n{}^{2-n}]$ on pH of soil environment

This dependency correlates well with the developed logarithmic concentration diagram, which is indicated by reliability of approximation R^2=0.99.

Thus, amid all particles $[Ni(OH)_n{}^{2-n}]$, Ni^{2+} predominate in acidic and neutral environments. Their concentration in interstitial water is described by equation (4) which is applicable for pH>5. At pH<5, all nickel in interstitial water will be present in soluble form as Ni^{2+}. At pH=8–14, non-soluble particles $[Ni(OH)_n{}^{2-n}]$ form, mainly $Ni(OH)_2$, migration of compounds of nickel is almost impossible. Nickel concentrates.

We have systematized calculations of the results of predictive modeling of post-pyrogenic migrational geochemical processes in ecogeosystems (Table 1), therefore they can be useful for analysis of probable dynamic of geochemical migration (accumulation) of heavy metals in ecogeosystems in the study of technogenic-ecological situations after fires.

Results and their analysis. Predicted activity of dynamics of geochemical migration of heavy metals in ecogeosystems under the influence of technogenic load caused by pyrogenic factors (on the example of Kharkiv region). Let us consider, on the example of Kharkiv region, map of soils, obtained using interactive map of soils of Ukraine (Interactive map of soils of Ukraine), comparing which with an atlas of soils (Krupskij, 1979) allowed us to thoroughly study properties of soil in the region, including their acidity. Parameter of pH of soils in Kharkiv region ranges from 4.5 to 9.6 (Table 2).

Table 1. Table of results of predictive modeling of post-pyrogenic migratory geochemical processes in ecogeosystems

Me	Equation of dependency of concentration of soluble forms of metals on pH of soil environment	Conditions of using equation	Note. Conditions of migration or accumulation of heavy metals
Cu	$lg[Cu(OH)_n{}^{2-n}]=0.007 \cdot pH^3 - 0.009 \cdot pH^2 + +2.22 \cdot pH + 9.25$	pH >3	At pH<3, all cuprum in soluble form Cu^{2+} At pH=7-14, all copper accumulates in soil in non-soluble form
Ni	$lg[Ni(OH)_n{}^{2-n}]= 0.015 \cdot pH^3 - 0.20 \cdot pH^2 - - 1.15 \cdot pH + 8.50$	pH>5	At pH<5, nickel is in soluble form Ni^{2+}. At pH=8–14, nickel accumulates in soil
Pb	$lg[Pb(OH)_n{}^{2-n}]= 0.011 \cdot pH^3 - 0.089 \cdot pH^2 - -1.89 \cdot pH + 13.06$	pH>6,5	At pH<6.5, lead is in soluble form Pb^{2+}. At pH=9–13, all lead accumulates in soil
Cr	$lg[Cr(OH)_n{}^{3-n}]= -0.002 \cdot pH^4 +0.06 \cdot pH^3 - -0.41 \cdot pH^2 - 0.97 \cdot pH + 6.69$	pH>4	At pH<4, all chrome is in soluble form Cr^{3+}. At pH=7–10, it accumulates

Table 1. Soil acidity of Kharkiv region

Soil	pH	Soil	pH
Sod-podzolized soils	4.6-5.7	Chernozems on common loess rocks	
Podzolized soils:		Average humus	6.9-7.2
Grey	4.5-5.1	Low-humus	7.2-7.3
Dark-grey	6.8-7.0	Deposit-solonetzic chernozem soils on loess rocks	6.9-7.6
Podzolized chernozems	7.0-7.1	Meadow-chernozem soils	9.5-9.6
Regradated soils	7.2	Meadow soils	9.5-9.6
Deep chernozems on loess rocks	6.7-6.8	Sod soils	5.7–6.7

For meadow-chernozem and meadow soils, characterized by pH>9, migrational property of most HM after effect of a pyrogenic factor will not change in any way(Fig. 5). There is observed accumulation of HM compounds in soil.

Most likely, in soils of podzolized dark-grey, podzolized chernozems (with pH close to neutral), there will be observed formation of soluble compounds of HM, which will lead to activation of migration in soil, or introduction or accumulation of them in plants. As a result of fire, pH can increase up to 7.5-7.8, which indicates decrease in solubility of HM compounds and their accumulation in soil. These soils are mostly located in Zhovtneve forestry.

A similar situation is characteristic of regradated chernozems (Fig. 5) located in the north and north-east of Kharkiv Oblast (Hutiansky, Kupiansky, Zhovtnevy forestries).

Such prediction can be made also for common chernozems on loess parent rocks which dominate in the northern part of Kharkiv Oblast (Balakliisky, Krasnohradsky, Iziumsky, Blyzniukivsky forestries).

Most likely, for soils of podzolized dark grry chernozems (with pH close to neutral), formation of soluble compounds of lead will be observed, leading to its migration in soil and introduction and accumulation in plants. After fire, pH can increase up to 7.5-7.8, causing reduction in solubility of compounds of lead and their accumulation in soil. These soils are mostly located in Zhovtneve forestry.

Conclusions. According to the conducted assessments, we have developed mathematical models of dynamic of behaviour of heavy metals, useful for making predictive assessments of their geochemical migration and accumulation in ecogeosystems as a result of impact of technogenic load caused by the pyrogenic factor. We determined conditions of concentration and migration of compounds of heavy metals, and formulated an equation for calculation of mobile forms of heavy metal compounds (Table 1).

The mathematical models were developed on the basis of logarithmic concentration diagrams which take into account formations of equal-weight concentrations of different compounds of heavy metals in conditions of technogenic load caused by the pyrogenic factor.

The developed interactive map of activity of geochemical migration of heavy metals under the impact of technogenic load caused by pyrogenic factors will help provide more details for the assessment of migrational properties of HM and prediction of the dynamic of their behaviour in ecogeosystems after fires. Similar cartographic material can be extrapolated for other regions of Ukraine, which are affected by technogenic pyrogenic load.

Development of logarithmic concentration diagrams allows prediction of the property of compounds of lead to migrate or accumulate as a result of acidity of soils under the impact of the pyrogenic factor. We analyzed the map of soils in Kharkiv region and made a prediction of the migrational property of heavy metals in the event of fire in different forestries.

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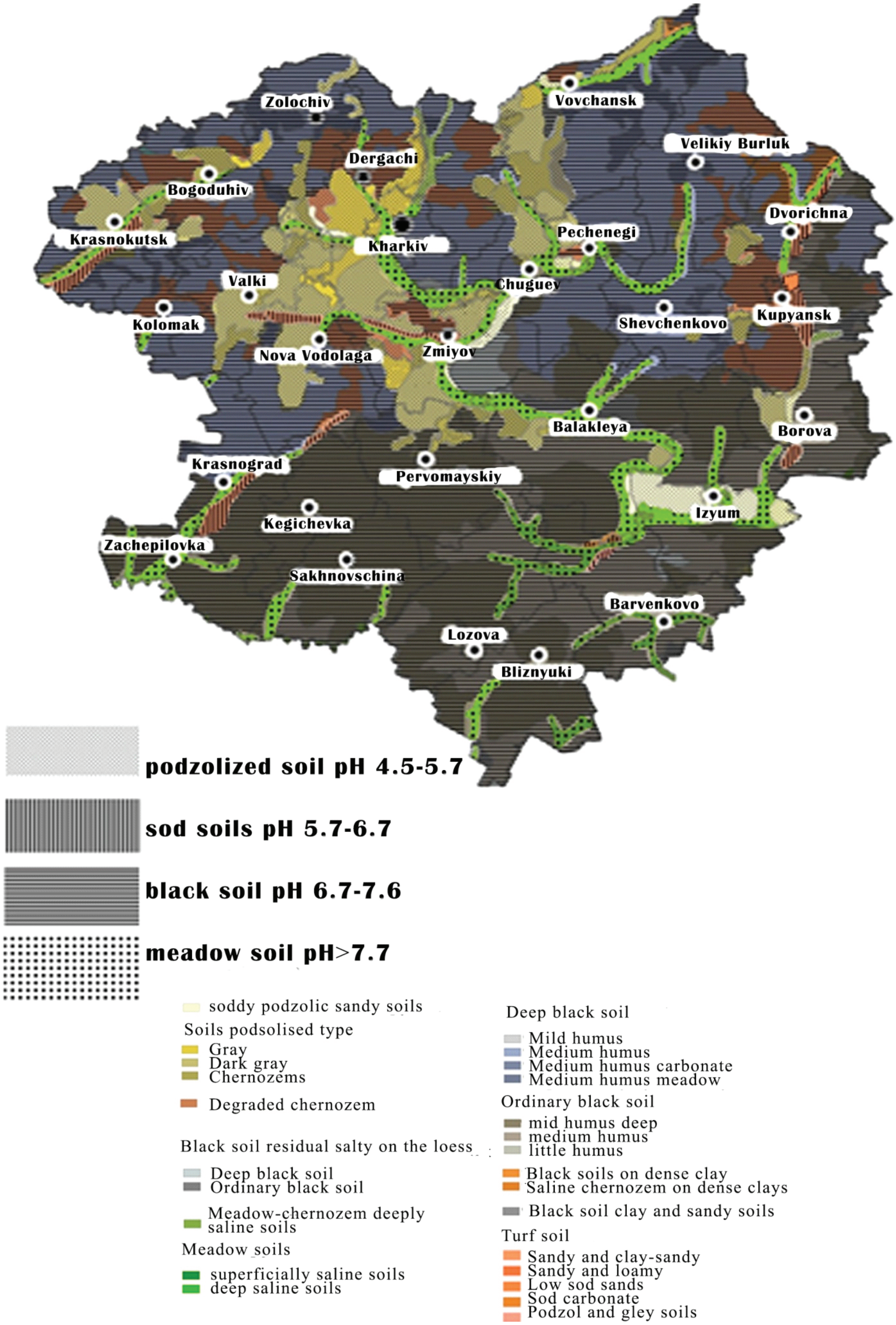

Fig. 5. Activity of geochemical migration of heavy metals under the influence of technogenic load caused by the pyrogenic factor (on example of Kharkiv region)

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