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SOIL CHEMISTRY

Assessment of Soil Contamination by the Content of Heavy Metals in the Soil Profile

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Abstract—A new soil-ecological definition of the maximal permissible concentration (MPC) of heavy metals in soils is suggested that regulates the sampling in contaminated territories. Instead of the shallow pits usually used for collecting surface samples for soil-hygienic and other investigations, it is proposed to fulfill a detailed analysis along the entire soil profile including not only the determination of the heavy element content in certain horizons but also the soil density in these horizons. For the polyelemental contamination Zc (according to the Saet equation) based on the background (clarke) excess, the established Zc values ranging from 1 to 128, may reach absurd values of 800–900 upon taking into consideration only one surface layer. At the same time, the use of the weighted average content of the metals in the soil profile adjusts the Zc values for the existing natural conditions. Upon aerial impact, the consideration of the heavy metal contents along the soil profile instead of their contents in the surface horizon only leads to a decrease in the indices of the soil contamination degree. Upon the hydrogenic impact, the transition from the heavy metal contents in the surface horizon to their contents in the soil profile gives higher values of the soil contamination.

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INTRODUCTION

Upon the study of soil contamination with heavy metals, we have to face many unsolved problems. For example, the content of heavy metals is usually compared to the maximal permissible concentration (MPC). However, there is no single definition of the MPC. According to Orlov and coauthors [11, p. 187], "the MPC is the content of a hazardous substance in the environment that virtually neither affects human health nor causes unfavorable consequences for their progeny for a certain time period or upon permanent contact." The term *hazardous substance* causes objections as applied to soils. This expression, which is true for air and water, cannot be automatically applied to soils. As a matter of fact, the MPC value is compared to the total content of a heavy element in the soil including the undividable introduced and natural parts. It is a serious drawback that this definition ignores the background content of the element in the soil.

The definition given by Bol'shakov and coauthors [1, p. 30] appears to be more precise: The *MPC is a hygienic standard, i.e., the concentration of a substance in water, air, soil, and food, which should not exert a negative effect on the adjacent media and humans....* This definition makes allowance for the total concentration of the substance in the soil rather than its anthropogenic portion.

Moreover, the unification of the MPCs for air, water, and soil turned out to be poorly efficient in general. The behavior of pollutants differs significantly in water, air, and soil. In the air (or water), the pollutants spread quickly in substantial volumes, and the substance concentration in these media is averaged because of the diffusion. As a result, a single analysis characterizes the average content of a substance in the air (water). On the contrary, an aerogenic pollutant (a solid-phase pollutant, in particular) spreads extremely slowly in the soil. The great difference between the pollutant concentrations in the different soil layers contrasts with its homogenous distribution in the water and air. The uneven contamination of a soil profile is very acute upon aerial pollution with the maximal contamination being observed in the uppermost horizon (the litter in forests) and much weaker (or almost absent) contamination, in the underlying layers. Thus, the problem of the heavy metal content in a soil profile should be solved taking into account the uneven distribution in the horizons, and the analysis of a single sample is not enough in this case.

The concentration of heavy metals in the upper soil layer has an important consequence. The metal concentration in the thin litter layer is less hazardous for plants and biota than its spreading into the entire pedon (the A + B horizons). Meanwhile, it is the state of the upper soil horizons that the contamination degree is recommended to be estimated from while taking samples not deeper than 0–2, 0–5, or 0–10 cm [10]. As is specified in another publication [15, p. 235], soils should be sampled at a depth of 0–10 cm for virgin land; 0–20 cm, for cropland; from the litter in forests; and from the uppermost 0–20 cm peat layer in bog soils. Thus, it is recommended to consider the contamination in the uppermost genetic horizon: the O, T, or any A horizon. In fact, these horizons appear to be maximally contaminated upon aerial pollution, although the underlying horizons may be weakly contaminated.

However, aerial pollution is not the only kind of technogenic contamination of soils. Rivers collecting industrial sewage pollute alluvial soils. The contamination occurs in the depth rather than on the surface in the alluvial soils. This is seen, for example, from the sulfur contamination of the medium horizons of alluvial soils in the floodplains of minor rivers in Ivanovo oblast [14]. We also revealed the maximal hydrogenic contamination with heavy metals in the middle horizons of alluvial soils in Perm [3]. The recommendation to assess the contamination degree according to the condition of the upper soil layer for hydrogenically polluted alluvial soil is inconsistent.

The monitoring of contaminated soils is evidently based on the landscape–geochemical approach, which recognizes only the aerial source of contamination. Neither hydrogenic contamination nor the intricate behavior of pollutants in the soil profile are taken into account in this case.

The soil approach that takes into consideration the heavy metal distribution along the soil profile rather than in a single (often very thin) O or AO horizon appears to be more valid. Plants will not feel the contamination in the case when pure layers underlie the polluted horizon; they develop normally and do not accumulate an excessive amount of hazardous metals. Ecologists often observe this situation at the sites that soil scientists identify as contaminated. This contradiction should be first recognized and then eliminated.

Soil contamination requires another approach based on the average-weighed content of heavy metal in the soil profile (i.e., the A + B horizon). In this case, the insignificant contribution of the contaminated uppermost layer will be balanced by the pure A and B horizons, and the average concentration of the metal in the soil may turn out to be low.

The suggested approach will make the contamination assessment more adequate not only for forests but also for urban soils, which are now sampled only at low depths. This gives only limited data diverging from the average information for the whole soil profile.

The uneven distribution of pollutants by the soil profile is connected with the different density of the horizons. Upon a similar concentration of a metal, its mass accumulated in peat or litter of low density may be several times lower than in the mineral horizon of the same thickness. Since it is the toxicant's mass that determines its impact on plants and biota, the different density of the biogenic and mineral soil horizons should be taken into consideration in order to obtain comparable results.

The aim of this study is to propose a procedure for determining the weighted average content of heavy metals in soil profiles for assessing the degree of the soil contamination according to the MPCs and clarkes and to test this procedure using the example of contaminated soils with different contamination sources.

OBJECTS

Soils contaminated with the emissions from the Mid-Urals copper smeltery. The copper-smelting plant is located within the Pervoural'sk–Revda industrial cluster in Sverdlovsk oblast; it was put into operation in 1940. Two main production departments are running now, i.e., the copper-smelting shop, which makes the largest contribution to the air pollution (87%) and the sulfuric-acid department. The enterprise's emissions to the atmosphere contain sulfur dioxide and anhydrous hydrogen fluoride, whereas the aerosols comprise a number of heavy metals, i.e., Cu, Zn, As, Cd, etc. [6]. The territory is situated in the southern taiga subzone of the Middle Urals with the percentage of forest land exceeding 60%. The soils are gray forest ones, clayey, and heavy clayey.

The territory was subdivided according to its response to the technogenic load according to the data obtained by ecologists from the vegetation status assessment. In the zone of technogenic devastation (the technogenic desert), the wood stand has been totally lost and the herb canopy is either absent or consists of horsetail, cereals, and widely spread mosses. There are eroded places with almost completely washed off litter and the humus soil horizon. In the impact zone, the tree leaves are burnt, and the tops of the crones are drying. In the buffer zone, the vegetation is suppressed to a low and medium degree, and top-dry coniferous trees are common. Finally, the vegetation is not disturbed in the background territory [6].

In accordance with these gradations, we analyzed the soil in three profiles. The samples were collected in the year of 2000. Profile 1 was made in the technogenic desert 0.5 km to the east of the plant in compliance with the predominating wind direction; profile 2 was studied in the impact zone 1 km to the west of the plant; and profile 3, in the background area 30 km to the west of the plant. In the zone of the technogenic desert, the soil has been significantly acidified (the pH_{water} is equal to 4.6 in the upper soil layer). With the growing distance from the plant, the soil's acidity decreases. The chemical composition of the soils is described in [5].

Soils contaminated with the emissions from the Noril'skii Nickel enterprise. The investigated region is located on the southern part of the Taimyr Peninsula. The soils develop under the conditions of permafrost occurring close to the surface, which results in the weak evaporation of moisture and the development of gley. The gas and dust exhausts of the enterprise influence the soils in the town's vicinity [16].

We studied the northeastern part of the Noril'sk region in the direction of the town of Talnakh. We sampled soils at a distance of 4 and 10 km from Noril'sk in the summer of 2004. The chemical composition of the soils is described in [5].

The soils contaminated with the emissions of the Chusovskoi metallurgical works. The town of Chusovoi is one of the metallurgical centers in Perm krai. The total area of the town covers about 58 km^2. Districts and settlements with lawn-and-garden sites with low-rise buildings occupy the bulk of the town's territory. In 2005, the total emissions to the atmosphere of more than 70 pollutants from the industrial enterprises of the town of Chusovoi constituted 24.7 thousand tons. The metallurgical enterprise Chusovskoi Metallurgical Works is the main source of heavy metals penetrating into Chusovoi's environment. The particles emitted from this enterprise to the atmosphere contain heavy metals, which are accumulated in the soil cover of the town [12]. In addition, the soils are contaminated with slags of the metallurgical plant, which were scattered somewhere in the town to form chemo-technozems.

The soils were studied in six profiles. Three alluvial gray-humus soils were analyzed in the floodplain of the Chusovaya River on Zakur'e Island. The soils developed on carbonate deposits were investigated on the terraces of the Chusovaya River. The soddy–podzolic soil in the forest–park of the new town on the left bank is one of them. Two other soils were studied on the right bank of the river in the old town: the soddy soil in the square and the chemo-technozem near the Metallurgists' club. The chemical composition of the soils was described in [4].

The alluvial soils in Perm. We analyzed the alluvial soils of the minor rivers in Perm, which were flooded during the high-water season and covered with warp. The profiles were situated in the zone influenced by the Perm–Krasnokamsk industrial center in the floodplains of the left tributaries of the Kama River, i.e., the Danilikha and Mulyanka rivers. The profile of a gray-humus gley soil was located in the Danilikha River floodplain 500 m away from the Perm-2 railway station. The profile of the gray-humus gley soil was determined on the Mulyanka River's floodplain 200 m away from the place of the river's crossing by a bridge of the Kosmonavtov highway.

All the studied soils are subjected to anthropogenic impacts. The worst water quality is registered in the lower reaches districts and settlements of the Danilikha River, where the water is unsuitable for drinking. According to the report by the Department of Environment Control for the year of 2004, the average quality of the water in the Mulyanka River is referred to the 2–3 quality classes, and to the 4th class in the upper reaches due to the high content of nitrates and iron [13]. The chemical composition of the soils is described in [3].

METHODS

The total chemical composition of the soils was analyzed using an energy dispersion X-ray fluorescent analyzer (Tefa-6111 (Ortec)). The following heavy elements were determined: Mn, Ni, Cu, Zn, Ga, As, Pb, Rb, Sr, Y, and Zr.

The procedure. The contents of the heavy metals in the upper horizons were compared to their contents in the entire soil profile, where the metals influence the vegetation and the macro- and microbiota. The content of the metals in the pedon ($C_{pedon)}$ was calculated as the weighted average amount of the metals including the B horizon (down to the C horizon) while also taking into consideration the differences in the horizons' density:

$$C_{pedon} = \Sigma Ci\, hi\, \gamma i : \Sigma hi\, \gamma i,$$

where Ci is the metal content, hi is the thickness, and γi is the density of the i-th soil horizon.

However, in the urban environment, lithozems and technozems are formed showing obliterated features of the initial genetic horizons. For these soils, the anthropogenic layers should be analyzed to a certain limited depth, which we assume to be 1.0 m. For soils without an illuvial B horizon, we also recommend restricting the analysis to a certain fixed depth of sampling, e.g., 1.0 m.

The excess of the heavy metal content over the MPC is calculated from the following equation:

$$K_{MPC} = C_{pedon} : MPC$$

The Saet equation is used for the calculation of the polyelement soil contamination with heavy elements [10]:

$$Zc = \Sigma(C_{soil} : C_{background}) - (n - 1),$$

where n is the number of analyzed elements.

The following categories of the total soil contamination are distinguished [10]:

Contamination category of soils	Zc
Permissible	1–8
Low	8–16
Average	16–32
High	32–64
Very high	64–128

The simplified method of digging shallow pits is widespread. The calculation based on the Saet formula uses the data on the heavy metal content in the surface soil layer. Since, in our opinion, the contamination should be analyzed in the entire soil profile, the Saet equation should be modified as follows:

$$Zc = \Sigma(C_{peredon} : C_{background}) - (n - 1).$$

Table 1. Coefficients of MPC (K_{MPC}) and clarke (K_{clarke}) in the surface horizon (K_{upper}, above the line) and in the soil profile ($K_{profile}$, below the line), and the total contamination index Zc. Soils of Revda technogeochemical anomaly

Object, coefficient	Ni	Cu	Zn	As	Pb	Zc	Contamination category
				K_{MPC}			
Technogenic desert	0.42/0.85	26.8/3.7	9.7/2.8	25.7/5.2	38.3/6.1	–	Unknown
Impact zone	0.52/0.66	159/10.5	25.3/2.1	111/1.7	152/3.5	–	–
Background	0.55/0.75	1.5/0.73	4.0/0.80	1.2/0.80	4.3/0.82	–	–
				K_{clarke}			
Technogenic desert	0.85/1.80	74/10.2	19.4/5.6	52/10.4	46/7.3	188/31	Out of category/medium
Impact zone	0.52/1.4	436/29	51/4.2	221/3.5	182/4.2	886/38	Out of category/strong
Background	1.2/1.6	4.2/2.0	7.9/1.6	2.4/1.6	5.2/1.0	17/4	Medium/available

Note: Here and in Tables 2–4, dash means that the index was not determined.

For checking the two approaches to the description of the soil contamination (with account for the surface horizon or the soil profile), we used the assumed values of the clarkes and MPCs of the heavy metals. The clarke values were mainly taken according to Vinogradov (cited after [8]): 40, for nickel; 20, for copper; 50, for zinc; and 5 mg/kg, for arsenic. Two elements are an exception. The chromium clarke was substantially lowered recently: from 200 (as suggested by Vinogradov) to 70 mg/kg (as introduced by Bowen [17]). On the contrary, the clarke value for lead was raised from 10 (after Vinogradov) to 25 mg/kg (according to Kabata-Pendias and Pendias [9]).

The MPC values were taken for all the elements except for arsenic. We took 85, 55, 100, and 30 mg/kg for nickel, copper, zinc, and lead, respectively [1]. For arsenic, we took the APC value equal to 10 mg/kg [1]. As for chromium, although its role in the soil contamination is important, there are no standards for this element in Russia. For the elements with no adopted MPC (APC) values, we used the well-known empirical dependence: MPC = (3–5) × background [7]. Substituting the average coefficient (4) and the clarke value (70 mg/kg), we came up with the conventional value for the chromium MPC equal to 280 mg/kg. It is close to the standards applied abroad [2]. Further, we use this value.

RESULTS AND DISCUSSION

The soils of the Revda geochemical anomaly. In the case when the contamination is assessed in only in the upper 2-cm-thick A0A1 horizon, the MPC is substantially exceeded in the technogenic desert: 26 (As), 27 (Cu), and 28 (Pb). However, this is mainly the surface contamination, and the MPC excess decreases by 4–6 times if the soil profile's thickness is taken into account (Table 1). The difference in the contamination degree is still more pronounced upon the different approach to the evaluation of the metal content in the impact zone. The K_{MPC} values reach 111 (As), 152 (Pb), and 159 (Cu) in the case when only the litter's contamination is taken into consideration, and the MPC excess is 2–10 times lower in the case when the entire soil profile is considered. The contamination parameters differ even for the background territory. The K_{MPC} values reach 4 for Zn and Pb in the case when only the litter contamination is regarded, whereas no MPC excess for these metals is registered in the entire pedon (K_{MPC} = 0.8).

The application of the Saet equation permits us to assess the degree of the total soil contamination with several heavy elements. It turns out that the Zc value significantly exceeds the boundary value (128) and reaches absurd values of 188 and 886 in the technogenic desert and the impact zone, respectively, in the case when only the surface horizon is considered. However, the consideration of the entire soil profile's contamination gives adequate indices, and the soils are classified as medium and highly contaminated according to Zc = 31–38. For the litter only, Zc = 17, and the contamination is estimated as that of the medium degree, whereas, for the soil profile, Zc = 4; hence, the background territory can be in fact considered as not contaminated.

The soils near the town of Noril'sk. A highly contaminated though thin (3 cm) horizon of forest litter was studied at a distance of 4 km from the town. If we consider the contamination solely in this layer, the MPC excess reaches a very high level, i.e., 193 for Cu and 132 for Ni. However, this is mainly surface contamination, and the MPC excess is much lower (4–5 times) in the case when the contamination of the entire soil profile is considered (Table 2).

At a distance of 10 km from the town, the 2-cm-thick forest litter layer is also considerably contaminated. The assessment of only this layer results in a substantial excess over the MPC value: 96 and 32 times for Cu and Ni, respectively. The MPC excess decreases to 2–3 times in the case when the contamination in the entire soil profile is taken into account.

In the case when only the forest litter is considered, the application of the Saet equation for the calculation

Table 2. Coefficients of MPC (K_{MPC}) and clarke (K_{clarke}) in the surface horizon (K_{upper}, above the line) and in the soil profile ($K_{profile}$, below the line), and the total contamination index Zc. Soils in the Norilsk suburb

Distance from the town	Ni	Cu	Zn	As	Pb	Zc	Contamination category
K_{MPC}							
4 km	132/2.2	193/4.6	0.35/0.93	1.2/0.32	3.9/0.5	–	Unknown
10 km	32/3.2	96/2.1	0.48/0.82	0.90/0.32	1.5/0.42	–	–
K_{clarke}							
4 km	280/4.7	531/12.6	0.70/1.8	2.4/0.72	4.6/0.8	815/17	Out of category/medium
10 km	68/6.8	264/5.8	1.0/1.6	0.9/0.32	1.8/0.50	332/12	Out of category/low

Table 3. Coefficients of MPC (K_{MPC}) and clarke (K_{clarke}) in the surface horizon (K_{upper}, above the line) and in soil profile ($K_{profile}$, below the line), and the total contamination index Zc. Soils in Chusovoi town

Object	Ni	Cu	Zn	As	Pb	Cr	Zc	Contamination category
K_{MPC}								
Alluvial gray-humus soil. Profile 3.	0.65/0.63	0.78/1.0	1.3/1.1	0.60/0.49	0.67/0.71	0.62/1.0	–	Unknown
Alluvial gray-humus soil. Profile 5.	0.63/0.59	1.04/0.75	1.68/1.24	0.50/0.41	0.77/0.70	0.93/0.57	–	–
Soddy–podzolic soil. Profile 1.	0.71/1.41	0.78/0.94	1.0/1.30	0.40/0.76	0.83/0.55	0.60/0.54	–	–
Urbolithozem. Profile 4.	1.06/1.11	1.73/1.22	2.82/1.72	0.80/0.52	1.10/0.50	3.54/1.63	–	–
Technozem. Profile 6.	1.07/0.76	2.13/2.48	3.62/2.90	1.10/0.71	2.03/2.71	7.1/1.95	–	–
K_{clarke}								
Alluvial gray-humus soil. Profile 3.	1.38/1.33	2.14/2.75	2.6/2.26	1.2/1.0	0.8/0.9	2.48/4.0	6/7	Permissible/Permissible
Alluvial gray-humus soil. Profile 5.	1.33/1.25	2.86/2.06	3.36/2.48	1.0/0.82	0.9/0.84	3.72/2.28	8/5	Permissible/Permissible
Soddy–podzolic soil. Profile 1.	1.5/2.35	2.14/2.58	2.0/2.6	0.8/1.52	1.0/0.6	2.4/2.16	5/7	Permissible/Permissible
Urbolithozem. Profile 4.	2.25/2.35	4.76/3.35	5.64/3.44	1.6/1.04	1.32/0.60	14.2/6.52	25/13	Medium/weak
Technozem. Profile 6.	2.27/1.61	5.86/6.82	7.24/5.80	2.2/1.42	2.4/3.2	28.4/7.8	43/21	Strong/medium

of the total soil contamination at a distance of 4 km from the town brings us to an abnormally high value (815), which is far beyond the permissible upper limit (128). However, the consideration of the entire soil profile's contamination puts the results in order, and the soil is classified as medium contaminated according to the index Zc= 17. At a distance of 210 km from the town, the total contamination also reaches an absurd value (332) for the litter only. However, for the soil profile, the Zcvalue decreases to 12 designating that the soil is classified as low-contaminated.

The soils in the town of Chusovoi. In the weakly contaminated alluvial (profiles 3 and 5) and soddy–podzolic (profile 1) soils, moving from the consideration of the upper horizon's contamination to that of the entire soil profile insignificantly influenced the MPC's excess and the total contamination index. The Zc index, for example, changes by 1–3 units (Table 3).

However, the difference appeared to be significant in the polluted urbolithozem and technozem. In the urbolithozem, the transition from the upper horizon's contamination to that of the soil profile lowered the clarke's excess for copper, zinc, and chromium; as a result, the total contamination index Zcdecreased from 25 to 13, which converted the medium contamination to the low category. In the technozem, the transition from the consideration of the upper horizon's contamination to that in the soil's profile lowered the clarke's excess for zinc and chromium and also decreased the index of the total contamination Zcfrom 44 to 22, which resulted in the conversion of the high contamination category to the medium one.

The alluvial soils in the city of Perm. These soils are hydrogenically contaminated with the maximal contamination being observed in the middle part of the profile. The consideration of the entire soil profile's

Table 4. Coefficients of MPC (K_{MPC}) and clarke (K_{clarke}) in the surface horizon (K_{upper}, above the line) and in the soil profile ($K_{profile}$, below the line), and the total contamination index Zc. Alluvial soils in Perm

River floodplain, coefficient	Ni	Cu	Zn	Pb	Zc	Contamination category
			K_{MPC}			
Danilikha	3.1/4.8	2.2/3.7	3.5/3.8	3.0/2.2	–	Unknown
Mulyanka	0.85/0.78	1.0/1.2	1.0/1.3	0.77/0.69	–	–
			K_{clarke}			
Danilikha	6.6/10.2	6.0/10.2	7.0/7.6	3.6/2.6	20/27	Medium/medium
Mulyanka	1.8/1.65	2.75/3.3	2.0/2.6	0.93/0.8	5/6	Permissible/Permissible

contamination as compared to the contamination of the upper layer only raises the contamination degree. This is particularly noticeable in the soil of the Danilikha River's floodplain. For this soil, the MPC's excess for nickel, copper, and zinc is significantly higher in the case when the contamination is assessed for the entire soil profile rather than for the upper horizon only (Table 4). This is also true for the clarke excess. As a result, the total contamination index Zcis equal to 27 (for the entire soil profile) versus Zc= 20 (for the upper horizon only). Thus, unlike the aerially contaminated soils, the maximal contamination in the alluvial hydrogenically contaminated soils is registered in the middle and the lower horizons, which is taken into account upon the calculation of the weighted average value of the heavy metal content.

CONCLUSIONS

The new soil approach to the determination of the MPCs of the heavy metals in the soils implies that the MPC is the weighted average according to the horizon's thickness and the density concentration of the heavy metal in the soil's profile including the A and B horizons that should not exert a negative influence on the plants, biota, and soil and ground water, as well as humans.

The new soil-ecological definition of the MPCs regulates the soil sampling in contaminated areas. Instead of collecting surface samples, we suggest performing a comprehensive analysis of the entire soil profile including not only the determination of the heavy element content in each horizon but also measurement of the soil's density in these horizons. For urbozems and technozems with distorted initial genetic features of their horizons, the anthropogenic layers should be analyzed to a depth of 1.0 m. For contaminated alluvial soils, the layers should also be studied to a depth of 1.0 m.

The indices of the MPCs' excess according to the heavy metals, i.e., the K_{MPC} values, do not have an upper limit. However, the index of the total contamination Zc for the polyelement contamination Zcobtained from the Saet equation proceeding from the background (clarke) excess is characterized by an upper limit of 128. As was found, the Zc values may become absurd (800–900) in the case when only the surface layer is considered. At the same time, the use of the weighted average content of the metals in the soil profile normalizes the Zc values.

Thus, for the aerial contamination, the consideration of the heavy metal content in the soil profile instead of its content in the surface horizon lowers the soil's contamination degree. On the contrary, for the hydrogenic contamination, the transition from the heavy metals' assessment in the surface horizon to that in the entire soil profile raises the soil's contamination degree.

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