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Relationship between soil properties and potentially toxic element content based on the dataset of the Soil Information and Monitoring System in Hungary

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The aim of this paper is to examine the relationship between soil properties and potentially toxic element contents of arable soils based on the dataset of the Soil Information and Monitoring System in Hungary. Nine potentially toxic elements (As, B, Cd, Co, Cr, Cu, Ni, Pb, Zn) were compared with selected soil parameters. We carried out grouping of related soil properties by principal component analysis. The method was suitable to describe the relationship within groups of the soil properties. We studied correlations of the resulting components and potentially toxic elements. The change of Ni content was influenced by the physical properties of the soil (e.g. clay content, field capacity, $R = 0.67$). Boron is the only one of the examined elements that indicates significant positive correlation with saline-alkali ($R = 0.21$) parameters. Zn, Co and Cr behaved very similarly; their correlation with components 1 and 2 were the closest.

Key words: principal component analyses (PCA), Soil Information and Monitoring System (SIMS), soil properties, potentially toxic elements (PTE), Pearson correlation

Introduction

Soils may vary considerably according to their chemical, physical and biological characteristics. Thus, metal solubility and extractability may be affected by the total metal contents or by simultaneous variation in soil properties (pH, CEC, organic matter, mineral composition, and soil texture) (Hlavay et al. 2004). The elements are pres-

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ent in soils in various forms and these can strongly affect their behavior in terms of biological availability, potential toxicity and mobility within the profile (Alloway 1995).

In the case of most cation metals the retention capacity of soil for trace metals increases with pH value, while other metals indicate maximum value in the neutral domains (for example: B), and yet the adsorption of most anion metals decreases with the growth of pH value (Adriano 2001).

A significant amount of the toxic metal content in natural and uncontaminated soil is inherited from the soil parent material (Csathó 1994). According to Kádár (1995) we considered potentially toxic elements which have a deleterious effect on the soil–plants–animal–human system. Adsorption of potentially toxic elements (PTEs) in the soil occurs by different methods for each element; thus the mobile contaminants of the soil (e.g. Cd, Zn, Pb, Cu) and the elements rapidly becoming insoluble (Cr) can be distinguished (Fodor 2000).

Based on the assessments of Fügedi et al. (2006) there is no characteristic element group in most parts of Hungary (this statement refers mostly to the lowland areas of Hungary). Practically all examined elements accumulate and leach out jointly, depending on local sedimentological, pedological and agricultural factors.

The fate and transfer of metals are complex and depend on the soil's mineralogy and the physical transport processes involved (Navarro et al. 2008).

In this study we analyzed the relationship between the pseudo total PTE contents and soil property scores derived from soil parameters performing principal component analyses (PCA).

Analytical method

Data source

Data of the Hungarian Soil Information and Monitoring System (SIMS) was used for statistical analyses. This system was created in 1992 and provides yearly data regarding the condition of the Hungarian soils. Soil samples are annually taken for analyses between 15th September and 15th October. The nation-wide monitoring system contains 1235 sampling points. From the 1235 points 864 can be found on arable land (Juhász 2006). To get the information on baseline conditions, all the chemical, physical and biological soil parameters were determined for each horizon in the laboratory. We evaluated soil data of 842 sampling points from the top soil on arable land. We used the data of 2010 for the evaluation of descriptive statistics, and the data of 1992 for the evaluation of principal component analysis.

Laboratory methods of the used soil parameters

Pseudo total content of the potentially toxic elements was determined by digestion with a mixture of cc. nitric acid and hydrogen peroxide at 105 °C and measured with an ICP device from the filtrate by the emissive spectrometric method (MSZ 21470-50: 1998). Characteristic basic properties of the soils were measured according the Hun-

garian standards. The upper limit of plasticity, according to Arany (KA) was determined by mechanical mixing (MSZ 08-0205:1978). Soil texture determination was performed by the pipette method (MSZ 08-0205:1978). The water retention measurements were performed with distilled water according to the MSZ and the ISO standards (MSZ 08-0205:1978; ISO 11274:2009). The total water capacity (pF 0), the field capacity (pF 2.5) and the wilting point (pF 4.2) were determined on undisturbed soil samples. At pF values of 0 to 2.5, 100 cm³ of water-saturated soil core samples were exposed to suction in boxes containing sand and kaolin plates. pF 4.2 values were determined on re-saturated soil pastes in a pressure membrane apparatus with cellophane membranes. The pF 6.2 values were calculated from hygroscopic measurements applying a relative humidity of 34% above the samples (using CaCl₂ · 6 H₂O as humidity regulator) (Várallyay 1973). Bulk density was calculated from dry soil weights (105 °C, 48 h) of undisturbed samples. Ca(CH₃-COO)₂-extractable acidity (y1) and KCl extractable acidity (y2) was calculated from the acidimetric titration (Buzás 1993). Calcium carbonate content was determined with the Scheibler method (Nelson 1982). Organic matter content was determined by wet combustion with the Tyurin titrimetric method (Tyurin 1931). Soil pH was determined in 1:2.5 soil/distilled water and soil/KCl suspensions using the potentiometric method (McLean 1982). Cation exchange capacity (CEC) and exchangeable Ca²⁺, Mg²⁺ determinations were performed with the modified Mehlich method (Buzás 1993). Total soluble salt content was calculated based on the measured electric conductivity of the saturated soil paste. Soda-alkalinity was determined in salt-affected soils measuring the phenolphthalein alkalinity (Buzás 1993).

Statistical analysis of data

Two datasets were used for statistical analysis. The most recent data are available from 2010 for the descriptive statistical analysis and we used the 1992 datasets for PCA, because the physical properties of the soil were tested only in the initial year.

We examined all those possible soil parameter effects which, based on previous experience, could influence the transport processes in the soil. The following soil properties were examined for the PCA: pH(H₂O), pH(KCl), cation exchange capacity (CEC, meq/100 g), soil organic matter (OM, %), soil texture (clay, silt, sand, %), Arany-type texture index (KA), bulk density (BD, g/cm³), saturated water retention value [pF 0, log(cm)], field capacity value [pF 2.5, log(cm)] and wilting point [pF4.2, log(cm)], exchangeable Ca²⁺, Mg²⁺ (m/V, %), total salt content (TSC, %), soda-alkalinity (S, %), hydrolytic acidity (y1), exchangeable acidity (y2). The following PTEs were involved in the statistical analyses: Cd, Cu, As, Ni, Co, Cr, B, Zn, and Pb.

SPSS 17.0 for Windows was used for statistical analyses. The PTEs were analyzed first with descriptive statistics. Thereafter we performed the adequacy test of the soil properties for PCA. The adequacy test consists of four main parts. In the first one a correlation matrix was used to identify a possible relationship among the soil properties. The second test was the anti-image process in which the variance of the variables was

split into an explained (image) and unexplained (anti-image) one (see ‘Results and discussion’). The third test was the Barlett test which investigates the hypothesis that the variables do not correlate with each other. The fourth test was the Kaiser–Meyer–Olkin (KMO) criterion, one of the most important measuring methods to decide the suitability of variables for PCA (Sajtos and Mitev 2007). Variances of the first five components were more than 1 which were used for later examinations. Thus, these components met the criteria based on Kaiser Normalization (Sajtos and Mitev 2007). Finally we studied the relationship between the received components (scores) and the PTEs with Pearson correlation.

Results and discussion

Descriptive statistics of examined PTE's content

We compared values of the examined metal and metalloid content by their basic statistic indicators (Table 1).

Table 1
Pseudo total metal content of soil samples (mg kg^{-1} , $n = 842$, 2010)

Statistical indicators	As	B	Zn	Cd	Co	Cr	Ni	Cu	Pb
Mean	6.7	6.47	54	0.23	9.07	29.1	22.8	17	17.8
Lower bound*	6.46	6.14	52.5	0.21	8.8	28.3	22	16.2	17
Upper bound*	6.93	6.8	55.5	0.24	9.34	30	23.5	17.8	18.6
5% Trimmed mean	6.47	6.27	53	0.21	9.01	28.6	22.5	16.7	17.1
Median	6.4	5.84	53.3	0.2	8.92	28.3	22.3	15.2	17.5
Std. D.	3.5	4.88	22.2	0.24	3.98	13.1	10.2	11.4	11.8
Minimum	0.29	0.5	7.25	0.1	0.9	3.27	1.95	0.23	0.99
Maximum	52.3	57	208	5.55	24.3	90.1	67.6	115	92.3
CV%	52.3	75.4	41	105	43.9	44.8	44.9	67.1	63.4
Skewness	3.22	3.08	1.21	13.3	0.24	0.67	0.49	2.81	1.18

Note: * marked means the 95% confidence interval for mean

Looking at the means and the 5% trimmed mean of the elements we can see that in each case values of the 5% trimmed mean are less than the arithmetic mean. Furthermore it can also be seen that between the maximum values and upper bound values the difference is greater than between minimum values and lower bound values. The above findings demonstrate that outliers are in the top 2.5% of the distribution. This may be caused by the influence of human activities or be of geologic origin (e.g. high arsenic content of the peat soils in Hungary). The interval between the lower and upper bound is considered to be the characteristic domain. We compared the characteristic domain and 5% trimmed mean of the PTEs with limit values of the geologic background concentration (“A”) of the 6/2009 KvVM-EüM-FVM ministerial joint decree.

Among the examined elements Cr ($30 \text{ mg}\cdot\text{kg}^{-1}$) and Ni ($25 \text{ mg}\cdot\text{kg}^{-1}$) approached the limit values, followed by Pb ($25 \text{ mg}\cdot\text{kg}^{-1}$). The lowest concentration compared to the limit value was measured in Cd ($0.5 \text{ mg}\cdot\text{kg}^{-1}$), Zn ($100 \text{ mg}\cdot\text{kg}^{-1}$) and Cu ($30 \text{ mg}\cdot\text{kg}^{-1}$). Concentration of the As ($10 \text{ mg}\cdot\text{kg}^{-1}$) and Co ($15 \text{ mg}\cdot\text{kg}^{-1}$) were about two thirds of the limit values (the data in brackets correspond to the “A” limits). The coefficient of variation (CV) provides information about the value of the relative standard deviation expressed in the percentage of average. Table 1 shows that the CV value of the Cd was extremely high (105%), which is associated to a 13.3 skewness value. These values indicate that the distribution of Cd differs from its normal distribution. The CV value of B, Cu and Pb was quite significant, unlike the CV values of the Zn, Co, Cr, and Ni, which were the lowest, and their skewness values were less than 1. The distribution of these elements (Zn, Co, Cr, and Ni) complied with the terms of normal distribution.

Our results correspond with evaluation of Kuti (2009) who studied the content of PTEs by their median values.

Soil parameters evaluated with PCA

The adequacy test of soil properties shows that the dataset is adequate for PCA. We are unable to show the correlation matrix due to its large size; however, we have concluded that there is correlation among the variables; there were a significant 130 (85%) of the 153 correlation values; furthermore the highest correlation value of 0.8 was between clay % and KA. Outside the diagonal elements of the anti-image covariance matrix the portion of variance is independent of other variables; therefore these values should be low. In our case less than the quarter of these values were higher than 0.09 that corresponds to the criteria (Sajtos and Mitev 2007). The main diagonal elements of the anti-image correlation matrix are adequate to the expected measures of sampling adequacy (MSA) value. In the case the MSA values have changed between 0.52 and 0.95. The Null hypothesis of Barlett test can be rejected, as the level of significance was less than 0.05. The KMO value in this example was 0.77 which means that the variables are suitable for PCA.

Table 2 shows the component matrix of the examined parameters and their communality. Communality indicates the amount of common variances of the variables. CEC and pF0 showed the highest communality, and y2 the lowest. It can be seen (Table 2) that the PC1 group includes the main soil physical properties, where the sand content was in inverse ratio to the remaining variables. The OM's matrix weight values are very close in groups PC1, PC2 and PC4; however its highest value was in PC2 compared to PC1 and PC4. This shows that the soil's organic matter plays an important role in its physical properties, as well as in the creation of negatively charged places, which is correlated slightly with CEC and exchangeable cation contents. The PC3 component consists of soil acidity indication factors, where the y1 and y2 parameters (acidity values) show an inverse relationship with pH (H₂O) and pH (KCl). The PC4 (BD and pF0) equally indicates the soil compaction and soil structure, and accordingly the extent of hydraulic conductivity of the soil or the rate of infiltration as

well. The PC5 group consists of salinity parameters such as total salt content (TSC) and soda-alkalinity. The cumulative % of variations shows the total explained variance at the bottom of Table 2. This means that five components explain 81.12% of common variances among all variables. Components 1, 2, and 3 show the highest weight of the explained variances (24.89%, 17.59% and 17.26%, respectively) in the component analysis.

Table 2
Rotated component matrix (Meaning of notations provided in the text)

Variables	Components					Communalities
	PC 1	PC 2	PC 3	PC 4	PC 5	
Sand (%)	-0.924	-0.205	0.024	-0.085	-0.017	0.904
pF2.5	0.850	0.234	-0.024	0.252	0.087	0.849
Silt (%)	0.817	-0.016	0.064	0.083	-0.034	0.680
pF4.2	0.754	0.291	0.051	0.167	0.112	0.697
KA	0.736	0.462	0.021	0.285	0.027	0.838
Clay (%)	0.732	0.422	-0.129	0.068	0.096	0.746
CEC	0.298	0.902	0.117	0.109	0.111	0.940
ecCa	0.236	0.863	0.222	0.107	0.000	0.860
ecMg	0.227	0.800	0.069	0.093	0.120	0.719
OM	0.435	0.503	0.032	0.405	-0.067	0.612
y1	0.117	-0.057	-0.925	0.047	0.010	0.876
pH (KCl)	-0.085	0.150	0.909	0.060	0.184	0.894
pH (H ₂ O)	0.035	0.183	0.891	0.036	0.224	0.880
y2	-0.140	0.013	-0.717	0.083	0.087	0.549
BD	-0.153	-0.156	0.036	-0.922	-0.054	0.901
pF0	0.334	0.124	-0.032	0.883	0.028	0.909
Soda	-0.021	-0.034	0.125	0.006	0.937	0.895
TSC	0.162	0.203	0.071	0.060	0.883	0.856
Eigenvalues	6.915	3.355	1.637	1.376	1.344	14.603
Cumulative % of variations	24.887	42.475	59.737	71.037	81.126	

Extraction method: principal component analysis. Rotation method: Varimax with Kaiser normalization. Rotation converged in 6 iterations. *Note:* Bold denotes the parameters of a component

Figure 1 shows the scatter matrix plot of the components in rotated space (Table 2) which represents the horizontal and vertical distances of variables among the components in the range between -1 and 1. Within the component variables their proportional and reverse relationship with each other by their component weight matrix is indicated. Figure 1 shows that the coherent variables are separated in both horizontal and vertical direction, depending on the size of the component matrix weight and direction. Those variables that are close to the edge of the graph have an absolute value of variables approaching the value 1.

Presentation of correlation between PTE's and groups of the soil parameters

Table 3 shows the possible relationship between PTEs and calculated principal components indicated by Pearson correlation. Arsenic is most closely related to PC2, probably because the sorption reactions on the surfaces of soils influence the mobility, toxicity and availability of arsenic in the subsurface environment (Pigna et al. 2006). However, the correlation is not high ($R = 0.32$) because PC2 informs mainly about negatively charged surfaces and arsenic is usually present in anionic form as arsenate and arsenite in the soils, and therefore adsorbed in positively charged surfaces by Fe/Al oxides (Xu et al. 2009).

Table 3
Correlation between principal components and the PTEs

PTE	PC1	PC2	PC3	PC4	PC5
As	-0.063	0.319**	0.192**	-0.151**	-0.022
B	0.024	0.366**	0.206**	0.048	0.215**
Zn	0.453**	0.481**	-0.162**	0.026	0.025
Cd	0.335**	0.143**	-0.069	0.109**	-0.054
Co	0.467**	0.427**	-0.167**	-0.007	0.016
Cr	0.471**	0.463**	-0.134**	-0.159**	-0.050
Ni	0.670**	0.319**	-0.177**	-0.141*	-0.139**
Cu	0.256**	0.360**	0.047	-0.007	-0.009
Pb	0.188**	0.049	-0.268**	0.159**	0.010

*, ** Correlation is significant at the 0.05 and 0.01 levels, respectively (2-tailed).

The correlation of boron is highest with PC2 ($R = 0.37$), PC3 ($R = 0.2$) and PC5 ($R = 0.215$), and it is the only one of the examined elements which indicates significant correlation with saline-alkali (PC5) parameters. This observation is explained by the fact that the boron content accumulates better in salty soils of arid areas. The other two components (PC2 and PC3) are also important. This observation is similar to the findings of other authors that boron adsorption is predictable using the soil chemical properties (pH, soluble salt content, surface area, organic and inorganic carbon content, and aluminum oxide content). Saline-alkali properties (PC5) as well as the amount of the exchangeable cations (PC2) commonly influence the boron mobility in the saline-alkaline soil (Goldberg et al. 2008).

The relationships of Zn, Co, Cr and Cu are strong with clay, silt and sand fractions, water retention (PC1), CEC, and exchangeable cation (PC2) components. These PTEs are characterized by their mobility that depends mostly on the clay, silt and sand fractions, soil organic matter and CEC, the exchangeable cation contents of the soil. Correlations of Zn, Co and Cr are almost in similar strength with PC1 and PC2. According to the literature the sorption and mobility of these elements is influenced by several factors in soils, such as pH, clay mineral content, CEC and soil organic matter. During the sorption process, metal surface precipitates may also form at the surfaces of phyllo-

silicate minerals (Shuman 1975; Ford and Sparks 2000; Voegelin et al. 2002; Bradl 2004). The effect of soil pH on the pseudo total PTE content was weak (Zn, Co and Cr) or non-detectable (Cu) in this study.

Similarly, the clay, CEC, OM, pH and CaCO₃ contents were the main factors regulating Ni retention in the studied soils (Mellis et al. 2004; Adhami et al. 2008; Ramachandran and D'Souza 2013). Our study shows that in the case of nickel this relationship is twice stronger with PC1 than with PC2. In our opinion, Ni enriches better in fractions of fine-textured soils.

Cd shows a relatively close correlation with PC1, weak correlation with PC2, PC4 components, and no detectable relation with the PC3 component. One possible explanation is that the presence of humic substances, mineral components, and proportion of macropores (Camobreco et al. 1996; Bergkvist et al. 2003) regulate the movement (leaching) of cadmium in soil. Our results could not prove the fact that any relationship between pH (PC3) and Cd content exists. Likely the effect of pH prevails in the quantity of Cd accessible for plants, and not in the pseudo total Cd content examined by us.

The relationship between PC2 and Pb ($R = 0.049$) was not able to prove that the presence of soil OM also plays an important role in Pb adsorption (Pinheiro et al. 1999). It has already been mentioned that the OM's matrix weight values show no significant differences in groups PC1 (0.435), PC2 (0.503) and PC4 (0.405) (see Table 2). Accordingly we assume that the OM content can be a part of group PC1 or PC4 (but not a part of PC2, which has the highest matrix weight); thus the correlation between OM and Pb can be detected. The Pb content in the soil is in significant negative correlation with PC3, which shows that Pb solubility and concentration depends on pH (Elkhatib et al. 1991).

Conclusions

This article deals with a comparative evaluation of soil properties in their mutual relationship to potential toxic elements. In our study we represented eighteen different variables, which were divided into five major groups by PCA based on their properties, which correspond to accepted professional knowledge. The separated components were the soil physical properties associated with soil texture (PC1), CEC and exchangeable cation contents (PC2), parameters associated with soil acidity (PC3), the variables indicating soil structure and compaction as well as determining fluid flow (hydraulic gradient) (PC4), and alkaline-saline properties (PC5). We found an important relationship between the calculated components and the examined PTEs' concentration in correlation, which are the following:

1. Soil physical variables had no effect on As.
2. There was a weak relationship between the boron concentration and the alkaline-saline variables.
3. CEC and exchangeable cation contents had no effect on Pb; there was only a negative correlation with pH.

4. The Ni content showed the closest correlation ($R = 0.67$) with clay, silt and sand fractions, water retention (soil physical parameters); Co behaved very similarly to Ni ($R = 0.47$).
5. Zn showed a stronger relationship with CEC and exchangeable cation contents and clay, silt and sand fractions, water retention (soil physical parameters), than Cd and Cu did.

We found that the applied method (PCA) was suitable to investigate the effect of different soil parameters on the pseudo total PTEs' concentration. There is a possibility of further development of this method if the analysis of bioavailable content is involved in addition to the pseudo total content of the elements.

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