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Transfer of Copper, Lead and Zinc in Soil–Grass Ecosystem in Aspect of Soils Properties, in Poland

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Abstract The total metal concentrations in soil samples from polluted area (roadside soils) ranged from 13.87 to 195.76 mg/kg for Cu; 13.56–310.17 mg/kg for Pb and 18.43–894.11 mg/kg for Zn and they were, respectively about 5, 2 and 13 times above the corresponding values in soil samples from country area. The mean values of EDTA-extractable concentrations in soil samples at unpolluted sites were: 2.47 mg/kg for Cu, 6.33 mg/kg for Pb and 4.94 mg/kg for Zn. The highest concentrations of Cu, Pb and Zn in grass were measured in soils from polluted area. Higher values of proportions of EDTA-extractable metals (24% for Cu, 40% for Pb and 38% for Zn) indicate that anthropogenic metals were more mobile and bioavailable than the same metals in soils from unpolluted area (20, 16 and 20% for Cu, Pb and Zn, respectively). The availability of Cu, Pb and Zn are affected by soil properties such as pH, organic matter content and cation exchange capacity. Correlation between the EDTA-extractable forms concentrations of metals and the total concentration in the various soils was observed. The coefficients of determination (R^2) varied between 0.809 for Cu; 0,709 for Pb and 0.930 for Zn in polluted soils and they are higher than corresponding values in unpolluted soils.

Keywords Metals · Availability forms · Soil · Grass · Transfer factor

The normal abundance of metals in earth material is commonly referred to by the geochemists as background and for any particular element this value or range of values is likely to vary according to the nature of the material. Trace elements in soils are derived from parent materials and anthropogenic inputs. Emission of metals may come from domestic waste, chemical industry and transportation. Anthropogenic impacts are extreme and urban soils develop within a wide range of different ecosystems (Norra and Stüben 2003). Urban traffic is one of the major sources for urban soil pollution. Roadside soils are an important reservoir for the pollution directly from vehicle sources, which could come easily in contact with pedestrians and people residing within the vicinity of the roads either by suspended dust or by direct contact (Plak et al. 2010). Urban soils present peculiar characteristics compared with agricultural soils, resulting from high anthropogenic pressure, which complicates investigations (De Kimpe and Morel 2000). In several studies, the contamination levels of urban soils have often been shown to be higher than those of their agricultural counterparts (Biernacka and Małuszyński 2006; Pruvot et al. 2006; Błoniarz and Zaręba 2006). Metals can be present in soils forming chemical-physical associations of different availability: simple or complex ions in soil solution; exchangeable ions; linked to organic substances; occluded or co-precipitated with oxides, carbonates and phosphates or other secondary minerals and ions in the crystalline lattices of primary minerals (residual fraction). The assessment of environmental risks requires not only the total amount of the metals in soils to be measured but also the amounts of metals in each association form, i.e. the availability of metals. Only a small portion of trace elements in soils is bioavailable. Water soluble and exchangeable fractions are considered to be bioavailable, oxide-, carbonate- and organic matter–bound fractions may be potentially bioavailable but residual fraction is mainly not available to plants. The proportions

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of each fraction vary between soils and largely determine the availability and mobility of metals in soils. Many chemical processes are involved in the transformation of trace elements in soils but precipitation–dissolution, adsorption–desorption and complexation are the most important processes controlling bioavailability and mobility of trace elements in soils. A widely used method for the evaluation of the availability forms of metals in soils is the leaching of soils by means of chemical extractants (Maiz et al.1997; Quevauviller et al. 1994; Rauret et al. 1999). Extraction tests are widely used to assess the release metal contaminants from soils, sludges and sediments. The choice of extracting agent depends on specific aims of a certain investigation. EDTA, DTPA and acetic acid exactions are often used for studies on physicochemical processes in soils like trace metal mobility The two main extraction approaches to soils are single and sequential extraction methods (Peijnenburg et al. 2007; Rao et al. 2008; Ure 1996). Estimation of the migration ability of metals in the natural environment is considered to be a necessary stage for predicting the ecological situation. The process of metal accumulation in agricultural plants is especially interesting since they contribute toxic element into human food chain. The pattern of metals accumulation in plants, regarding the type soil, the biological peculiarities of plants, the nature of elements and concentrations is discussed by many investigators (Grytsyuk 2006; Keller et al. 2002; Krolak 2003).

The principle objective of the work was to evaluate the degree of contamination of soils-grass ecosystem by Cu, Pb and Zn, determine and compare the potential effect of soil properties (pH, organic matter, cation exchange capacity CEC) on the availability of studied metals and uptake by grass in polluted and unpolluted soil ecosystem. Kabata-Pendias (1994) suggests that anthropogenic metals in soils are more mobile and bioavailable than the same metals which derive ultimately from the soil parent material, presumably because of the differences in forms. The results of Chłopecka et al. (1994) for contaminated soils around Tarnowskie Góry support this concept. It has been found that residual (nonavailable) forms of metals dominate in uncontaminated soils.

Materials and Methods

The total of 29 topsoil (0–20 cm) and grass samples were taken from two areas under different impact of anthropo pressure in May–June 2011 in central part of Poland. Fifteen sampling sites (roadside soils samples) were located along the highway E7 with high traffic intensity from Warsaw to Gdańsk (region 1.) and fourteen sampling sites were located in the region 2. Regarded as unpolluted (country area 10 km distant from route E7, natural pastures). The soils sampled were dried in air circulation room and then treated to remove stones and plant residues and passed through a 2 mm nylon sieve. The sieved samples were stored in plastic bags for physic-chemical analysis. The grass samples were washed in tap water and deionized water in the order and dried in an oven at 60°C to constant weight and milled into powder for measurement of metal content. Physicochemical analysis of soils: pH, hydrolytic acidity (H_h), content of organic matter (OM), the sum of exchangeable cations (S), cation exchange capacity (CEC)] was carried out by standard techniques (Ostrowska et al. 1991). Cation exchange capacity (CEC) is calculated by adding the sum of exchangeable bases (S) to hydrolytic acidity (H_h): $CEC = S + H_h$. The values of pH of the water extract is detected potentiometrically. For analysis of total metals concentrations in soil and grass, 1 g of dried sample was mineralized by a mixture containing 3 mL HNO_3 and 1 mL $HClO_4$ and then filtered. For analysis of available metals in soil, air-dried soils were extracted by 0.05 mol/L ethylene-diamine-tetra-acetic acid disodium (EDTA-Na_2 at $pH = 7.0$). 100 mL of the EDTA solution was added to 10 g of soil sample placed in polypropylene tubes. The tubes were shaken on a rotating shaker for 1 h and then were centrifuged. Metal concentration in the supernatant liquid were measured with a flame atomic absorption spectrometry (FAAS). The same procedure without samples was used as control. Three replications were conducted for each sample. Quality assurance and quality control (QA/QC) for metals in soil samples were estimated by determining metal concentrations in the Standard solutions Merck (Merck, Darmstadt, Germans). The detection limit is calculated based on estimated instrumental detection limit assuming that 1 g of sample is digested or diluted to 100 mL. Limits of detection (mg/kg dry matter) for Cu, Pb and Zn were: 0.001; 0.003 and 0.001, respectively. The soil-to-grass factor (transfer factor, TF) was defined as follows:

$$\begin{aligned} \mathrm{TF} = {} & \text{concentration of metals in grass } (\mathrm{mg/kg})/ \\ & \text{concentration of metals in soil } (\mathrm{mg/kg}). \end{aligned}$$

Data analysis (e.g., mean, SD, coefficient of variation) and statistical analyses were conducted in this study. To test the relationships between the EDTA-extractable metal concentrations values and the soil total concentrations in soils, linear regressions were performed. The basis statistics, the correlation significance and regression analysis were performed using Microsoft Office Excel 2007 for windows. Statistical pattern of soils and grass contamination by metals was analyzed on the basis of measurement results.

Table 1 Physicochemical properties of soils from two regions

Location	Soil properties				
	pH	OM (%)	H_h (cmol(+)/kg)	S (cmol(+)/kg)	CEC (cmol(+)/kg)
Region 1					
n = 15					
Mean	6.50	5.47	1.35	15.36	16.71
Range	4.38–7.92	2.08–13.98	0.92–1.94	5.54–23.20	6.76–24.12
SD	0.95	3.25	0.27	4.84	4.74
Region 2					
n = 14					
Mean	5.42	9.61	2.18	16.73	18.90
Range	3.35–7.98	5.60–25.00	0.83–3.79	11.60–26.50	14.21–27.33
SD	1.40	5.25	0.85	5.37	4.73

Mean mean value, *Range* range value, *SD* standard deviation

Results and Discussion

The physicochemical analysis of studied soils (region 1.-polluted and region 2.-unpolluted), summarized in Table 1, showed large differences in soil characteristics. The values of pH ranged from strong acid to mild alkaline and it was location dependent. In detail, fifteen soil samples collected from polluted area and fourteen soil samples from unpolluted area ranged from 4.38 to 7.92 and from 3.35 to 7.98, respectively. The content of organic matter in soil samples from polluted and unpolluted areas varied: 2.08%–13.98% and 5.60%–25%, respectively. Other soil properties studied, such as: Hh, S and CEC ranged: 0.92–1.94 cmol(+)/kg; 5.54–23.20 cmol(+)/kg; 6.76–24.12 cmol(+)/kg, respectively at polluted sites. The corresponding values in soil samples from country area varied: 0.83–3.79 cmol(+)/kg; 11.60–26.50 cmol(+)/kg; 14.21–27.33 cmol(+)/kg, respectively.

Total and EDTA-extractable metals concentrations in soils are presented in Table 2. There were distinct differences among the two sampling locations in accumulation and availability of studied metals. Variation metals concentrations in roadside soils (region 1.) were larger than the fluctuations observed in soil samples from country area (region 2.). The total metal concentrations in soil samples from polluted area ranged: 13.87–195.76 mg/kg for Cu; 13.56–310.17 mg/kg for Pb and 18.43–894.11 mg/kg for Zn. The mean values of Cu, Pb and Zn concentrations were: 65.23, 86.57 and 301.17 mg/kg, respectively. Similar concentrations were observed in other polish soils neighbouring chosen communication routes (Plak et al. 2010). Level of contamination of roadside sols in China was lover (Chen et al. 2010). The total concentrations of Cu, Pb and Zn in soils from region 1. Were above the critical ranges given by Kabata-Pendias and Pendias (1984). The critical soil total concentration is defined as the range of values above which toxicity is considered to be possible. The total concentrations of Cu, Pb and Zn in soils in roadside were, respectively about 5, 2 and 13 times above the corresponding values in soil samples from country area, probably due to the atmospheric emissions from the traffic that significantly increases the concentrations of metals in the upper horizons of the neighbouring soils (Finster et al.

Table 2 Total and EDTA-extractable heavy metals concentrations (mg/kg dry matter) in soils

Location	Total concentration			EDTA-extractable		
	Cu	Pb	Zn	Cu	Pb	Zn
Region 1						
n = 15						
Mean	65.23	86.57	301.17	13.18	26.35	96.41
Range	13.87–195.76	13.56–310.17	18.43–894.11	3.68–36.10	4.71–59.08	3.63–263.91
SD	58.50	90.47	317.73	9.65	18.76	93.38
Region 2						
n = 14						
Mean	12.55	40.08	24.34	2.47	6.33	4.94
Range	5.66–21.30	34.65–45.71	16.54–39.46	1.11–3.98	4.22–8.14	2.52–8.32
SD	5.16	3.42	7.61	0.92	1.46	1.77

2004; Kim and Fergusson 1994). This pollution results in a high level of contamination of plants grown of these soils.

The EDTA-extractable Cu, Pb and Zn concentrations in roadside soils samples were higher than corresponding values in soil samples from country area. For example, the EDTA-extractable heavy metals concentrations at polluted sites ranged: 3.68–36.10 mg/kg for Cu; 4.71–59.08 mg/kg for Pb and 3.63–263.91 mg/kg for Zn with the mean values of 13.18; 26.35 and 96.41 mg/kg, respectively. The EDTA-extractable concentrations in soil samples at unpolluted sites ranged from 1.11 to 3.98 mg/kg for Cu; 4.22–8.14 mg/kg for Pb and 2.52–8.32 mg/kg for Zn with the mean values of 2.47, 6.33 and 4.94 mg/kg, respectively. Zeng et al. (2011) noted similar values for EDTA-extactable metals concentrations in China (1.38–5.66 mg/kg for Cu, 2.04–9.74 mg/kg for Pb and 1.40–12.23 mg/kg for Zn).

The availability difference of the same metals was observed between the studied areas. The proportion of each element extractable with EDTA gives information about metal availability. The quantities of Cu, Pb and Zn extracted from soils (Table 3) indicated, on average, 24, 40 and 38% availability forms, respectively, in roadside soils and 20, 16 and 20% availability forms, respectively, in soils from country area. Therefore, it is indicated that anthropogenic metals in soils were more mobile and bioavailable than the same metals in soils from unpolluted area. It has been found that for the soil samples from 1. Region the order of metal availability was: Pb > Zn > Cu. Similar results are reported by Maiz et al. (1997). The availability for Cu, Pb and Zn in soil samples from 2. Region generally formed the order of Cu = Zn > Pb. In these soils availability showed wide variations, which can be explain by the variability of the physicochemical soil parameters but also by the contamination route of these soils (dust deposition, emissions from the traffic). The specificities of roadside soils complicate the generalization of the approach.

Table 3 The proportion of EDTA-extractable metals in soils

Location	EDTA-extractable metal [%]		
	Cu	Pb	Zn
Region 1			
n = 15			
Mean	24.28	39.87	37.72
Range	11.79–37.10	15.00–64.15	19.69–49.34
SD	7.63	16.09	8.65
Region 2			
n = 14			
Mean	20.15	15.97	20.14
Range	14.68–27.01	9.58–22.72	15.24–25.14
SD	3.39	4.22	3.06

Table 4 presents the concentrations of Cu, Pb and Zn measured in grass (in dry weight). It shows a great variability of the concentrations according to the metal elements and the studied area. The highest concentrations of Cu, Pb and Zn in grass were measured in soils from polluted area (up to 4, 2 and 8 times above the corresponding values in soils from unpolluted area according to the element). This difference of contamination between grass samples from two regions can be caused by the dust emission due to motor vehicle traffic. The concentrations of lead in grass from polluted area were lower than corresponding values reported by Pruvot et al. (2006) (these data referred only to unwashed grasses).

The transfer factor values ranged: 0.08–0.76 for Cu; 0.01–1.29 for Pb and 0.13–3.42 for Zn in roadside soil samples with the mean values of 0.39; 0.33 and 0.77, respectively (Table 4). They were higher than corresponding

Table 4 Heavy metals concentrations (mg/kg) in grass and transfer factor values (TF)

Location	Metal concentration			Transfer factor (TF)		
	Cu	Pb	Zn	Cu	Pb	Zn
Region 1						
n = 15						
Mean	17.14	11.13	109.74	0.39	0.33	0.77
Range	5.31–34.43	2.88–32.33	35.86–287.97	0.08–0.76	0.01–1.29	0.13–3.42
SD	8.81	7.90	80.57	0.20	0.35	0.83
Region 2						
n = 14						
Mean	4.15	5.28	13.91	0.34	0.13	0.59
Range	1.22–7.38	3.95–7.54	8.62–22.24	0.15–0.55	0.10–0.17	0.41–0.74
SD	1.69	1.02	3.66	0.11	0.02	0.13

Table 5 Correlation coefficients between the proportion of EDTA-extractable metals in total concentration, transfer factor values and soil properties (correlation is significant at the 0.05 probability level)

Location	Soil properties		
	pH	OM	CEC
Region 1			
%EDTA-Cu	−0.815	−0.369	−0.690
%EDTA-Pb	−0.321	−0.324	−0.477
%EDTA-Zn	−0.777	−0.086	−0.420
TF[Cu]	−0.229	−0.701	−0.445
TF[Pb]	−0.034	−0.425	−0.230
TF[Zn]	−0.057	−0.401	−0.353
Region 2			
%EDTA-Cu	−0.752	0.433	−0.664
%EDTA-Pb	−0.596	0.420	−0.377
%EDTA-Zn	−0.346	0.450	−0.237
TF[Cu]	−0.769	0.405	−0.701
TF[Pb]	0.229	−0.196	0.133
TF[Zn]	−0.311	−0.074	−0.142

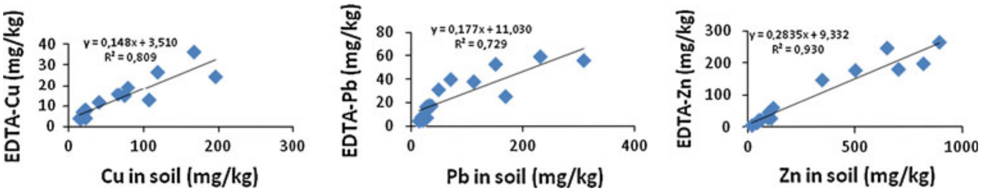

Fig. 1 Linear correlation between soil EDTA-extractable metals concentrations and total concentrations of metals in soils from region 1

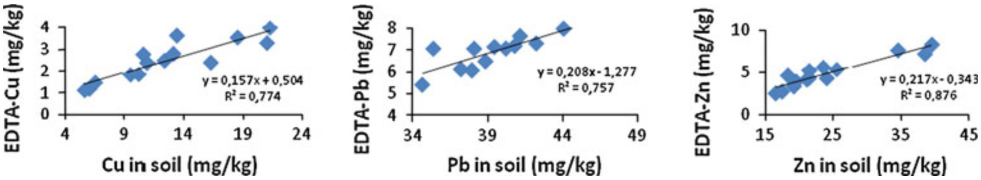

Fig. 2 Linear correlation between soil EDTA-extractable metals concentrations and total concentrations of metals in soils from region 2

values in soil samples from unpolluted area. The highest transfer factor value was for Zn and the lowest for Pb. Similar results was determined by Grytsyuk (2006). The average values of TF(Cu), TF(Pb) and TF(Zn) in country soils was measured at 0.34 (with a range of 0.15–0.55), 0.13 (0.10–0.17) and 0.59 (0.41–0.74), respectively.

Correlation analysis (Table 5) has shown that proportion of EDTA-extractable studied metals concentrations in soils are affected by soil properties such as pH, organic matter content and cation exchange capacity (CEC). This was in agreement with the research results reported by Zeng et al. (2011) and Keli et al. (2009). Many researchers have investigated factors influencing metals uptake by plants (Jung and Thornton 1997; Tang 2007). Generally in natural systems mobility end availability of Cu, Pb and Zn usually decreased with the increasing soil pH and decreasing organic matter content. A negative correlation between soil pH and metal mobility and availability to plants has been well documented in numerous studies. For example, with decreased soil pH, the dramatic increases in metal desorption from soil constituents and dissolution in soil solution were observed for Cd, Pb and Zn (Sukreeyapongse et al. 2002; Bang and Hesterberg 2004). In this study, organic matter was negatively correlated with the proportion of EDTA-extractable all three metals in soil samples from polluted area, but in soil from country area this relationship was positive. Organic matter plays a significant role in determining the availability and mobility of metals in soils in the two aspects. One is that organic matter could reduce the metal availability by adsorption or forming stable complexes. On the other hand, organic matter is also involved in supplying organic chemicals to the soil solution, which may serve as chelates and increase metal availability to plants (Vega et al. 2004). This could partially explain the positive correlation of EDTA-extractable metals and organic matter in the current and previous reports (Pappaffilippaki et al. 2007). No significant correlations were observed between transfer factor values and pH in roadside soil samples. Therefore, it indicates that metals accumulation in grass (especially Pb, Zn) from polluted area depends not only on soil pollution but also on air metal pollution and transfer factor values cannot reliably estimate the availability of heavy metals to grass. Among studied metals, only TF values for Cu were significantly correlated with soil properties. In soil samples from region 2. The TF[Cu] was correlated with: pH ($R = -0.769$), OM ($R = 405$) and CEC ($R = -0.701$). The obtained results showed good linear relationships between EDTA-extractable Cu, Pb, Zn concentrations and total metals concentrations in all soil samples from two areas (Figs. 1, 2). For the three elements, the coefficients of determination (R^2) varied between 0.809 for Cu, 0.709 for Pb and 0.930, for Zn, in soils from polluted area, and they are higher than in soils from unpolluted area. The correlations observed were strong, however Cu, Pb and Zn availability could also be affected by other physicochemical parameters of soils. Relationships between the proportion of EDTA-extractable metal and selected soil parameters were examined.

Metals pollution levels in soil samples from the two studied areas shoved significant differences, both in total concentrations and in EDTA-extractable metals concentrations. Total concentrations of Cu, Pb and Zn in roadside soil samples were, generally, above the critical ranges of values above which toxicity is considered to be possible. Level of contamination by Cu, Pb and Zn was, respectively about 5, 2 and 13 times above the corresponding level in soil samples from unpolluted area, probably due to the atmospheric emissions from the traffic that significantly increases the concentrations of metals in the upper horizons of the neighbouring soils. This pollution results in a high level of contamination of plants grown of these soils. Higher values of proportions of EDTA-extractable metals indicate that anthropogenic metals were more mobile and bioavailable than the same metals in soils from unpolluted area. The availability of Cu, Pb and Zn are affected by soil properties such as pH, organic matter content and cation exchange capacity. Metals accumulation in grass (especially Pb, Zn) from polluted area depends not only on soil pollution but also on air metal pollution and transfer factor values cannot reliably estimate the availability of heavy metals to grass. Although individual studies showed significant correlations between available concentration of metals in soils and their concentrations in plants, often including soil properties like, pH, organic matter, exchangeable in the regression, it is difficult to predict metal availability for one area using relationships obtained for another area.

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