

Mobility of trace elements in arable soils from the Bobov dol valley, Bulgaria

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Abstract

Soils in the Bobovdol valley are influenced by a complex anthropogenic impact that increases the geochemical and pedo-morphological diversity in the region. In order to assess the mobility of some essential trace elements and toxicants three soil types used for agriculture were studied. They are located at a different distance from the coal fired Bobovdol Thermoelectric Power Plant (Bulgaria) and thence diversely enriched with Cu, Zn, Pb and Mn. The local geochemical particularities, profile distribution of elements and key soil features are used to assess the mobility of Cu, Zn, Pb and Mn.

According to the results obtained studied soils are characterized by low total and mobile content of Pb, Cu, Zn, and Mn. Their total contents are lower than the background values in Bulgarian soils but lithic discontinuity or fly ash admixtures entail higher amounts. All elements are mainly concentrated in the silt fractions but river sediments (in Gleyic Fluvisol – profile 7) are richer in clays and clay-metal complexes. Organo-metal complexes of the elements are more typical of the strongly altered anthropogenic horizons in Pretic-Carbic Luvisol (profile 6).

Despite of all various geochemical and physicochemical conditions in soils, studied elements form stable associations with soil components which determine their slight mobility and strong competition.

There is a well-pronounced antagonism among the mobile forms (water-soluble and ion-exchangeable) of Cu, Zn, Mn and Pb and the most competitive ions are: Cu-Pb and Zn-Mn in reducing environment of river's sediments while in the case of preferential adsorption of lead onto the mineral colloids: Zn-Pb and Mn-Pb. Copper is the least mobile element in the studied soils regardless of soil type and features while the bioavailability of lead is the most dependent on the soil type.

The content of mobile forms of the elements in studied soil types is non-toxic and decreases in the following order of average concentrations: Mn (0.72 mg/kg) > Pb (0.55 mg/kg) > Zn (0.09 mg/kg) > Cu (0.05 mg/kg).

Key words: trace elements, agricultural soils, physicochemical conditions, antagonism

The Bobovdol valley is simultaneously agricultural and industrial region due to electricity production in the Bobovdol Thermoelectric Power Plant. The impact of these activities on soil properties is different, but both are a source of chemical elements that enter the soils (Tsolova et al., 2016). Among them heavy metals are recognized as very toxic even they occur in the small amounts.

Heavy metals in soils show typical ion exchange behaviour, demonstrating essentially the same affinity for exchange as do alkaline earth metals having the same charge and similar ionic radius. Trace elements in cationic form are probably not dominantly adsorbed on phyllosilicates or other layered silicate clays because they are always vastly outnumbered by other cations with which they compete (e.g. Ca). They are strongly adsorbed on the phyllosilicates if chemisorption takes place but also occur as readily mobile species. Humus matter and metal oxides are much more efficient sorbents of heavy metals in cationic form than even the most efficient sorbent among phyllosilicates, indicating that specific adsorption and other complexation processes are the dominant binding mechanisms (Violante et al., 2010).

The identification of trace elements compounds in soils and their bonds with different soil components are the basic tools for studying the small geochemical cycles of elements in the anthropogenically modified landscapes, revealing the mechanisms of their adverse effects on the environment, and evaluating the resistance of the soil system to the external impacts (Marinkina, 1999; Tsolova et al., 2013; Andreeva et al., 2015). There is a published information in Bulgaria of the concentrations of different compounds of trace elements in soils obtained by the methods of chemical fractionating (Marinkina, 1999; Atanasova, 2012; Dinev, 2012; Hristova, 2013; Yossifova et al., 2016). They allow answering some ecologically important questions and revealing the not understood problems. At least two aspects of the problem of characterizing the fractional composition of the trace elements compounds in soils are topical. One of them is connected with the improvement of methods for their determination, and the other one concerns

the evidence that these results can be used for assessment and forecasting of the ecological status of soils. The capability of fractionating methods to reveal the regional particularities of soils is also important (Minkina et al., 2010; Minkina et al., 2014; Ladonin, 2002).

Still, in the anthropogenically modified landscape the mobility of trace elements is one of the basic tools for assessing the soil toxicity. **The aim of this study is to assess the mobility of trace elements in agricultural soils located at a different distance from the coal fired Bobovdol Thermoelectric Power Plant (Bulgaria) and thence diversely enriched with Cu, Zn, Pb and Mn.**

Materials and methods

Three soil types, located at the different distances from the Bobovdol Thermoelectric Power Plant (TPP), were studied. The location of profiles and the view of localities are shown in Fig. 1.

Profile № 5. Chromic Endocalcaric Cambisol (Clayic)

Soils are located in a plain-hilly relief (over the village of Dolistovo), at the upper part of a south-eastern slope with an incline of 6-70, at 592 m a.s.l. These soils are formed on calcaric clay materials and are prepared for sowing the next crop at the time of the study.

Profile № 6. Pretic-Carbic Luvisol (Differentic, Hypereutric, Bathyruptic)

It is located in the South of the Thermal Power Plant (TPP), in a plain terrain, with a slight incline to the Southeast, near the temporary tailing pond and the water tower of the TPP, at 505 m a.s.l. The relief is flat-hilly, formed by non-calcaric clay materials. Cultivated crop is wheat.

Profile № 7. Gleyic Fluvisol (Cleyic, Humic, Anthrotoxic)

Soil from the overflowing terrace of the river Razmetanitsa, located at 500 m.a.s.l. The soil is cultivated with sunflower as a rotation crop. Because the groundwater is shallow (150 cm) the



Figure 1. Location of profiles and landscape view of studied soils

soil odor is strong and typical of rotting processes and methane formation. Soils are formed on alluvial non-calcaric parent materials. The relief of the surrounding area is flat-hilly.

Methods

The local geochemical particularities, element fractionation, and profile distribution were simultaneously used to assess the mobility of Cu, Zn, Pb, and Mn.

Soil survey in the Bobovdol valley was performed by advanced pedological methods for morphogenetic diagnosis of soils (Yolevski and Hadjiyanakiev, 1976; Yolevski et al, 1980; IUSS Working Group WRB, 2006) and the actual sampling standards (BDS¹ ISO 10381-1, 2, 4.). The methodology in Guidance for Soil Description

(FAO, 2006) is also used for description of soil profiles.

The following methods of analysis are applied:

1. Pre-treatment of soil samples for physico-chemical analysis according to BDS ISO 11464.
2. pH (H₂O) determination - potentiometrically according to ISO 10390.
3. Determination of the total content of trace elements – soil mineralization with “aqua regia” (ISO 11466) and measuring the contents by AAC (ISO 11047) on a Perkin-Elmer 2100.
4. Determination of the content of easily soluble trace elements – by extraction of soil with 1M NH₄NO₃ (ISO/DIN 19730:2012).
5. Cation exchange capacity – method of Ganev and Arsova (1980). This method determines the contribution of the permanent, preferential charges (on basal surfaces, T_{CA}) and variation charges (basically pH dependent exchange includ-

¹ Bulgarian State Standard

ing the lateral surfaces, T_A) of soil colloids in the cation exchange capacity of soils by titration of soil extract (4 g of soil and several portions of 8 cm³ of 1.0 n sodium acetate and 0.2 n potassium maleate with pH 8.25 to reach 100 cm³) with 0.04 n sodium hydroxide solution in the presence of phenolphthalein to determine TA and subsequent titration of the above eluate with 0.04 n complexon after dilution up to 200 cm³ deionized water and addition of 10 cm³ of triethanolamine (1:1) and 2 cm³ of 5.0 n potassium hydroxide solution (non-carbonate) to achieve pH 12-13 in the presence of chromium-blue to determine T_{CA} .

6. Particle size distribution – method of Kachinsky (1958). This method differs from the method described in ISO 11277:2009 mostly in samples pre-treatment (NaOH is used here as a dispersing agent) and sieves mesh sizes. The following size of major fractions are recognized by Kachinsky: Sand – this fraction involves particles with 1.00-0.05 mm equivalent spherical diameter; Silt fraction – from 0.05 to 0.001 mm equivalent diameter and Clay <0,001 mm.

Results and Discussion

The data in table 1 show that the contents of total and mobile forms of the microelements vary considerably, but some of the soil-typical tendencies are standing out. Copper has the most homogeneous distribution both in spatial (horizontal) and vertical (in the depth) direction. This element is the least mobile and its water-soluble and exchangeable forms do not exceed 0.08 mg/kg. The contents of the rest analysed elements varies in a wider range not only as a result of the pedogenesis but also due to the anthropogenic activities: presence of ash in Ap2 horizon of profile 6, which is the reason for the increase of zinc content; lithological heterogeneity below 80 cm, especially in the Ck horizon, which sharply increases the manganese content and morphologically contrasts with the next horizons; hydromorphous genesis and reducing conditions in profile 7, which increase the mobility of copper and zinc. The deeper accumulation of lead and manganese (in profiles 5 and 7) seems also inherited from the soil forming materials and

might exceed the background value of 26 mg/kg for Pb (Regulation № 3). Still, these concentrations are non-toxic as well as the total contents of the elements in the rest horizons – they are lower than their background values in the Bulgarian soils (34 mg/kg is the background of Cu and 88 mg/kg of Zn, Regulation № 3). Concerning the mobile concentrations of the elements, we should also note that the mobility of zinc and manganese is higher in the surface horizons while the mobility of the lead becomes higher in reducing conditions. As known, the mobility of Zn and Mn in soils is also low but the weathering may increase their contents and mobility (Schulin et al., 2010), which is also confirmed by this research. Although, according to Prüeb's (1992) phytotoxic thresholds based on 1M NH₄NO₃ extract (1 mg/kg for Pb and Cu, and 30 µg/g for Zn), the mobile concentrations of Pb, Cu and Zn are non-toxic.

There is a strong positive correlation between the contents of Cu, Zn, and Pb in profile 5 which reveals their simultaneous accumulation in soils (Table 2). They are mainly concentrated in the silt fractions (0,01-0,001 mm), which, regardless of their weaker adsorption ability than clays, firmly fix them and determine the low values of element mobile concentrations. The strong negative correlations of these elements with the organic carbon are also determined by the silt fractions, where the content of organic matter is low, considering the strong negative correlation among these soil components ($r_{\text{Corg-0.01}} = -0.88$). The organo-mineral complexes are typical of clay fractions ($r_{\text{Corg-0.001}} = 0.86$) and determine the mobile concentrations of Zn and Mn which is also demonstrated by the strong correlation between elements ($r_{\text{Zn}_M\text{-Mn}_M} = 0.98$). There is a slight antagonism between the mobile concentrations of Cu and Mn in this profile, and very strong between the mobile Zn-Pb and Mn-Pb. This is partially connected to the strong nexus between Zn and Mn as we already noted. The slightly acid reaction in profile 5 does not determine the contents and the mobility of Cu but influences the biochemistry of the rest elements, especially Pb's. The preferential adsorption of Pb presumably also takes place especially in the metamorphic horizon due to the

Table 1. Content of trace elements in the studied soils (mg/kg, %)

Hori-zons	pH	Cu		Zn		Mn		Pb					
		Total	Mob.*	%**	Total	Mob.	%	Total	Mob.	%			
Profile 5. Chromic Endocalcaric Cambisol													
Ap	5.0	24.00	0.03	0.13	65.00	0.18	0.27	245.00	2.75	1.12	24.00	0.33	1.38
B	5.0	26.50	0.05	0.19	70.00	0.13	0.18	240.00	0.85	0.35	30.00	0.70	2.33
Ck	7.5	25.50	0.03	0.12	70.00	0.10	0.14	300.00	0.58	0.19	29.50	0.78	2.64
M***	5.8	25.33	0.04	0.14	68.33	0.13	0.20	261.67	1.39	0.56	27.83	0.60	2.12
Profile 6. Prettic-Carbic Luvisol													
Ap1	8.0	23.00	0.08	0.33	58.50	0.10	0.17	600.00	0.30	0.05	26.50	0.25	0.94
Ap2	8.2	25.50	0.05	0.20	118.00	0.08	0.06	735.00	0.20	0.03	21.00	0.45	2.14
Bt	7.7	24.50	0.05	0.20	66.50	0.05	0.08	1210.00	0.20	0.02	25.00	0.60	2.40
Ck	8.1	25.50	0.03	0.12	58.50	0.03	0.05	2400.00	0.10	0.00	22.50	0.45	2.00
C1	8.0	17.50	0.08	0.43	50.00	0.08	0.15	395.00	0.15	0.04	16.50	0.65	3.94
M	8.0	23.20	0.06	0.25	70.30	0.07	0.10	1068.00	0.19	0.03	22.30	0.48	2.29
Profile 7. Gleyic Fluvisol													
Agp	8.0	24.50	0.01	0.04	63.50	0.05	0.08	475.00	0.70	0.15	17.50	0.60	3.43
Ag1	7.9	20.00	0.05	0.25	58.50	0.05	0.09	550.00	0.60	0.11	17.00	0.55	3.24
Ag2	7.9	28.00	0.05	0.18	69.00	0.08	0.11	780.00	0.40	0.05	20.00	0.55	2.75
M	7.9	24.17	0.04	0.16	63.67	0.06	0.09	601.67	0.57	0.10	18.17	0.57	3.14

Mob.* – mobile element content, mg/kg; %** – relative element content; M*** – arithmetic mean

Table 2. Correlations between the content of elements and mobility sources contents in profile 5

Parameter	CuT.	CuM.	ZnT.	ZnM.	MnT.	MnM.	PbT.	PbM.
CuT.	1.00							
CuM.	0.80	1.00						
ZnT.	0.92	0.50	1.00					
ZnM.	-0.74	-0.19	-0.94	1.00				
MnT.	0.04	-0.56	0.43	-0.70	1.00			
MnM.	-0.87	-0.40	-0.99	0.98	-0.53	1.00		
PbT.	0.94	0.56	1.00	-0.92	0.36	-0.98	1.00	
PbM.	0.84	0.35	0.99	-0.99	0.58	-1.00	0.97	1.00
pH	0.11	-0.50	0.50	-0.76	1.00	-0.60	0.43	0.64
<0.001, %	-0.58	0.03	-0.85	0.98	-0.84	0.91	-0.81	-0.93
<0.01, %	0.60	0.00	0.87	-0.98	0.82	-0.92	0.83	0.94
Corg, %	-0.91	-0.48	-1.00	0.95	-0.45	1.00	-1.00	-0.99
T ₈₂ (CEC), cmol/kg	0.82	0.32	0.98	-0.99	0.60	-1.00	0.96	1.00

Table 3. Correlations between the mobile content of elements and mobility sources contents in profile 6

Parameter	CuT.	CuM.	ZnT.	ZnM.	MnT.	MnM.	PbT.	PbM.
CuT.	1.00							
CuM.	-0.75	1.00						
ZnT.	0.54	-0.27	1.00					
ZnM.	-0.39	0.87	0.11	1.00				
MnT.	0.60	-0.90	-0.15	-0.87	1.00			
MnM.	0.04	0.62	0.13	0.82	-0.61	1.00		
PbT.	0.64	-0.15	-0.02	0.07	0.24	0.60	1.00	
PbM.	-0.43	-0.07	-0.13	-0.44	-0.04	-0.61	-0.66	1.00
pH	0.12	-0.14	0.47	0.15	0.04	-0.18	-0.36	-0.38
<0.001, %	-0.01	0.57	0.10	0.63	-0.62	0.89	0.55	-0.23
<0.01, %	0.03	0.53	0.26	0.64	-0.67	0.86	0.47	-0.19
Corg, %	0.54	0.08	0.61	0.41	-0.32	0.75	0.65	-0.43
T ₈₂ (CEC), cmol/kg	-0.38	0.70	0.13	0.62	-0.85	0.62	0.02	0.23

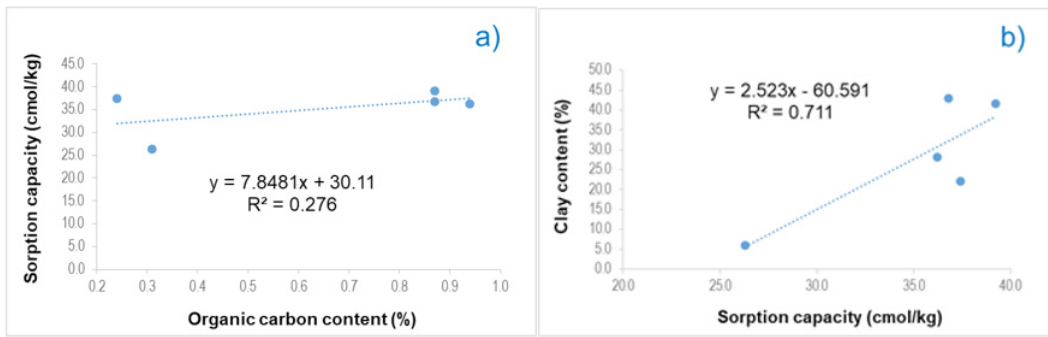


Figure 2. Correlations between organic carbon and adsorption potential (a) and between the adsorption potential and clay fraction (b) in profile 6

Table 4. Correlations between the mobile content of elements and mobility sources contents in profile 7

Parameter	CuT.	CuM.	ZnT.	ZnM.	MnT.	MnM.	PbT.	PbM.
CuT.	1.00							
CuM.	-0.07	1.00						
ZnT.	1.00	0.03	1.00					
ZnM.	0.83	0.50	0.88	1.00				
MnT.	0.67	0.69	0.74	0.97	1.00			
MnM.	-0.60	-0.76	-0.68	-0.94	-1.00	1.00		
PbT.	0.90	0.36	0.94	0.99	0.92	-0.88	1.00	
PbM.	0.07	-1.00	-0.03	-0.50	-0.69	0.76	-0.36	1.00
pH	0.07	-1.00	-0.03	-0.50	-0.69	0.76	-0.36	1.00
<0.001, %	0.95	0.24	0.98	0.96	0.87	-0.82	0.99	-0.24
<0.01, %	0.39	0.89	0.48	0.84	0.94	-0.97	0.74	-0.89
Corg, %	0.36	-0.95	0.27	-0.22	-0.44	0.53	-0.07	0.95
T _{8.2} (CEC), cmol/kg	0.42	0.87	0.51	0.86	0.96	-0.98	0.77	-0.87

process of fertilization established here (Tsoleva et al., 2016). This phenomenon is observed from O’Day (1999) with iron oxides.

Anthropogenic influence in profile 6 changes some correlations (Table 3). The most significant of them are related to the ash content which increases the nexus of Cu, Zn, and Pb with organic matter. The adsorption potential in this profile is owed not only of the clay fraction but also of the silt fraction, which are the main source of the studied elements and define their mobility. An exception is noticed for lead, which mobility does not depend

on the studied factors. Copper to some extent is also an exception – it is mainly associated with organic matter but mineral colloids are the source of its ion-exchangeable forms, according to the calculated statistical correlations (fig. 2).

The distribution of manganese does not depend on any of the studied parameters, which shows that sources external to the soil are responsible for manganese occurrence in this profile too. Still, during the soil genesis Mn sorption onto the mineral and organic colloids is well pronounced. This is probably the reason of which the antagonism

between the mobile concentrations of Mn and Pb, and Zn and Pb is not so strong.

The reducing conditions in profile 7 increase the significance of the organic fractions as a source of mobile forms of Pb and Mn – this is expressed in the strong correlations with pH and organic carbon. Clay fraction and its sorption potential are the main sources of total and mobile forms of the rest of the elements. Copper again is an exception – although it is concentrated in the clay fraction, it occurs in the organic and silt fraction and this fact increases the variety of factors that impact its accumulation and mobility in profile 7. There is a well-pronounced antagonism in Gleyic Fluvisol among the mobile forms (water-soluble and ion-exchangeable) of Cu, Zn, Mn and Pb as the most competitive ions are: Cu-Pb and Zn-Mn.

Conclusion

Studied soils are characterized by low total and mobile content of Pb, Cu, Zn, and Mn. Their total contents are lower than the background values in Bulgarian soils but lithic discontinuity or fly ash admixtures entail higher amounts. All elements are mainly concentrated in the silt fractions but river sediments (in Gleyic Fluvisol – profile 7) are richer in clays and clay-metal complexes. Organo-metal complexes are more typical of the strongly altered anthropogenic horizons in Pretic-Carbic Luvisol (profile 6).

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The content of mobile forms of the elements in studied soil types is non-toxic and decreases in the following order of average concentrations: Mn (0.72 mg/kg) > Pb (0.55 mg/kg) > Zn (0.09 mg/kg) > Cu (0.05 mg/kg).

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