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ORIGINAL PAPER

EFFECT OF SALINITY ON THE MOBILITY OF TRACE METALS IN SOILS NEAR A SODA CHEMICAL FACTORY*

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Abstract

This study has been conducted to evaluate the mobility of trace metals and their phytoavailability in soils exposed different salinity levels caused by waste deposition from soda production in the vicinity of the Ciech Soda Poland S.A. plant. The study was carried out on twelve soil samples from the depth 0-20 cm. Total N, total C and inorganic C content were determined in all the samples. Analyses of total metal content and concentrations of their available fractions were also carried out. All metals were determined by the atomic spectrometry absorption method, using a Philips 2100X spectrometer. In order to perform soil salinity, electrical conductivity (EC) and chloride ions (Cl^-) content were measured in selected samples, and ESP (exchangeable sodium percentage) was calculated. The concentration of chloride ions was determined by the Mohr method. In order to cover a large range of soil salinity, measured by EC values, slightly saline samples up to soils with high salt concentrations have been chosen. All analyses were performed with three replications to give arithmetic mean values. Statistical correlation was evaluated using the Statistica computer program. An increase in soil salinity measured by electrical conductivity (EC) altered the mobility and phytoavailability of metals. The content of phytoavailable, DTPA-extractable zinc, lead, copper, manganese and iron varied, but salinity did not influence their mobility in a clear way. It is a consequence of the complexicity of processes that govern the behaviour and mobility of trace metals.

Keywords: trace metals, mobility, salt affected soils, industrial wastes.

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INTRODUCTION

Salt toxicity is one of the major edaphic factors limiting plant growth (Khatar et al. 2017). Apart from natural salinization, human induced secondary salinization of soils occurs. In the temperate climate zone, salt-affected soils are formed under the impact of salt springs during the natural soil forming processes (Piernik, Hulisz 2011) but also due to anthropogenic saline habitat, as in areas where deicing salts are used for the removing of snow and ice from roads in the winter season (Backstrom et al. 2004), near mining waste deposition sites and in the vicinity of soda industry plants (Cieśla et al. 1981, Gonet, Hermann 1995).

There are waste ponds in the vicinity of the Ciech Soda Poland S.A. plant, located in Mątwy, which are a source of soluble salts causing soil salinity. This soda producing industrial plant has deposited alkaline slurry for more than ninety years. The easily soluble salts (mainly $CaCl_2$ and NaCl) are dominant components of soda waste disposed of into the inadequately sealed ponds (Cieśla et al. 1981). The effects of the accumulation of soluble salts on physical and chemical properties of soil are well recognized (Lemanowicz et al. 2018, Lemanowicz 2019). Excess of sodium ions negatively affects soil structure, aggregate stability and soil hydraulic properties. A high sodium concentration leads to degradation of soil due to the dispersion of clay and slaking. This can affect the flow of water and air as well as root penetration (Crescimanno et al. 1995). The accumulation of salts reduces plant growth and the activity of microorganisms. Microbial processes such as nitrogen cycling, emission of greenhouse gases, including carbon dioxide and methane, are affected (Adviento-Borbe et al. 2006). High salt concentrations may have a negative effect on plants because of direct toxicity or through osmotic stress. High concentrations of salts may also change the chemical behaviour of the metals accumulated in soils, causing an increase or decrease in their availability. Thus far, not much research has been conducted on the effect of soil salinity on the mobility of trace metals. Salinity is known to change the ionic strength of the soil solution, but little quantitative information is available regarding the impact on concentrations of microelements readily available for plants.

The aim of this study has been to assess the influence of high accumulation of soluble salts on metal mobility in salt-affected soils in the vicinity of sedimentation ponds located at the Ciech Soda S.A. plant in Mątwy (the Province of Kujawy and Pomerania, central Poland). In particular, the research has been undertaken in order to asses to what extent mobility and phytoavailability of trace metals (Zn, Cu, Pb, Mn, Fe) could be influenced by the excess of chloride ions in soils. Soils affected by anthropogenic salinization caused by the waste stored in ponds, are among the most productive soils (Gleyic Phaeozems, IUSS WRB 2014) and the increase of metals phytoavailability may be hazardous.

MATERIAL AND METHODS

The study was carried out on twelve soil samples from the vicinity of the Soda-Mątwy deposition tanks collecting waste from soda production. Soils for the study were sampled in the direct vicinity of the deposition tanks on May 2017, and the sampling sites were located 5 to 8 m from the base of the tanks. Soil sampled from the depth 0-20 cm was air dried and passed through a 2 mm sieve before analysis. In order to cover a large range of soil salinity, measured by EC values, slightly saline samples up to samples of soil with high salt concentrations were chosen. Measurements of soil salinity were performed on the basis of electrical conductivity (EC), chloride ions (Cl^-) content and the ESP index.

Electrical conductivity (EC) was measured in a 1:5 soil-water solution with a conductivity meter.

The exchangeable sodium percentage (ESP) index was calculated for assessing soil sodicity. The ESP was defined as:

$$ESP = 10 \cdot cNa^+/CEC,$$

where:
c – denotes the concentration of exchangeable Na^+ (mmol kg^{-1}),
CEC – cation exchange capacity (mmol kg^{-1}).

Total C was determined by dry combustion at 900°C, and inorganic C was measured by dissolution of carbonates in 10% HCl (Santi et al. 2006). Total N was determined using the Kjeldahl method on a Kjeltec analyzer. Concentration of Cl^- ions was determined according to the Mohr method. Total metal contents were measured after mineralization of samples in HNO_3 and $HClO_4$ and analyzed with an atomic absorption spectrometer Philips 2100X. The available fractions of metals were evaluated with the use of a DTPA extraction technique according to Lindsay, Norvell (1978). The accuracy of the method was verified through analysis of a standard reference sample (Till 3).

All analytical measurements were performed with three replications to give arithmetic mean values. The maximum allowable standard deviation between replicates was set at 10%.

The central tendency is given as a median and the variation is given as min and max values. Statistical correlation between soil parameters was evaluated using the Statistica computer program.

RESULTS AND DISCUSSION

The soils were characterized by pH values ranging from 7.10 to 8.15, and the clay content ranged between 4.2 and 9.8% – Table 1. The soil pH

Table 1

Selected soil properties

No	Fraction <0.002 mm	pH H_2O	pH KCl	SOC	N	C/N	CEC	EC	ESP
	(%)			(g kg^{-1})			(mmol kg^{-1})	(dS m^{-1})	
1	4.23	7.93	7.80	6.65	1.30	5.24	89.90	0.75	0.31
2	4.20	8.15	7.81	5.38	1.40	3.85	90.40	5.17	0.52
3	7.86	7.74	7.67	10.47	2.80	3.70	152.00	0.75	0.41
4	7.17	7.69	7.69	7.65	2.90	2.68	129.40	0.80	0.45
5	5.64	7.63	7.66	8.12	3.30	2.47	113.40	0.78	0.36
6	5.62	7.41	7.75	8.63	4.00	2.18	139.10	2.86	0.59
7	4.84	7.46	7.70	4.66	1.80	2.63	174.30	2.67	0.79
8	4.76	7.58	7.73	3.63	1.9	1.90	156.20	2.50	1.09
9	6.62	7.10	7.35	30.16	4.40	6.92	700.80	43.91	1.49
10	6.60	7.24	7.38	27.23	3.60	7.65	448.80	38.18	1.97
11	8.19	7.95	7.27	2.22	2.00	1.13	158.50	2.68	0.26
12	9.80	7.66	7.20	2.49	2.00	1.24	163.40	6.68	0.22
Min.	4.20	7.10	7.20	2.22	1.30	1.13	89,90	0.75	0.22
Max.	9.80	8.15	7.81	30.16	4.40	7.65	700.80	43.91	1.97
Mean	5.98	7.63	7.62	10.44	2.67	3.67	213.89	9.19	0.75
SD	1.34	0.30	0.18	8.92	1.02	2.00	180.28	15.12	0.52
CV	22.50	3.94	2.39	85.44	38.06	54.65	84.29	164.61	69.98

was only slightly higher than in soils beyond the impact of the soda industry (Kobierski, Dabkowska-Naskręt 2012). Other authors reported a similar effect after adding NaCl to soil (Garcia, Hernandez 1996).

The soils submitted to our study are rather poor in organic matter as the content of soil organic carbon (SOC) ranged from 2.22 to 30.16 g kg^{-1}, with the mean value 10.44 g kg^{-1} (Table 1).

Salt-affected soils usually exhibit low organic matter content due to insufficient plant growth leading to low inputs of organic materials. According to Wong et al. (2010a), several contradictory processes occur in saline soils, among which humification is often inhibited. Saline soils are also subjected to increased losses of organic matter due to dispersion and leaching. As a result of these processes, the organic matter content is usually smaller in salt-affected soils than in adjacent non-degraded soils.

The content of total N ranging from 1.30 to 4.40 g kg^{-1} was low as compared to the soils of the Inowrocław region beyond the salinity impact. The C/N ratio was relatively narrow, ranging from 1.13 to 7.65 (Table 1).

Cation exchange capacity (CEC) of the soils ranged from 89.9 to 700.8 mmol kg^{-1}, with the mean value 213.89 mmol kg^{-1}. The composition

Table 2

Content of exchangeable cations and chlorides (Cl^-)

No	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-
	(mmol(+) kg^{-1})				(mg kg^{-1})
1	2.83	2.98	83.53	0.60	44.00
2	4.73	3.18	82.22	0.35	39.00
3	6.21	5.79	4.07	8.20	65.00
4	4.07	2.79	105.89	0.56	100.00
5	8.20	2.75	124.14	4.00	58.00
6	13.79	4.74	154.25	1.49	113.00
7	16.97	4.32	133.25	1.65	432.00
8	104.31	5.28	590.32	0.87	493.00
9	88.57	5.24	353.69	1.32	19570.0
10	4.10	10.32	143.63	0.46	17275.0
11	3.57	10.18	148.76	0.84	73.00
12	2.83	2.98	83.53	0.60	18.00
Min.	2.83	2.75	4.07	0.35	18.00
Max.	104.31	10.32	590.32	4.00	19570.0
Mean	29.61	5.23	174.89	1.85	3190.0
SD	34.86	2.71	154.28	2.34	7133.60
CV	117.73	51.67	88.21	126.71	228.71

of exchangeable cations is dominated by calcium ions; the content of Na^+ ions varied from 2.83 to 104.31 mmol kg^{-1}, although an elevated concentration of these ions was observed in most samples (Table 2).

The content of chloride ions varied in a wide range from 18.0 to 19570.0 mg kg^{-1}, with the mean value 3190.0 mg kg^{-1}. Samples rich in sodium also contained a high concentration of chloride. An extremely high concentration of chloride may affect the soil's organic matter composition and properties. It has been found (Oberg, Sanden 2005) that chlorine derivatives of soil organic matter, induced microbially or enzymatically, are formed. Chlorine substituents are covalently bound to aromatic and aliphatic structures in large molecules comprising organic matter. The increased chlorine content makes organic matter more hydrophilic and consequently more mobile.

The data presented in Table 2 indicate that ions which contribute to soil salinity are Ca^{2+}, Na^+ and Cl^-. However, saline soils which occur in the surroundings of the soda plant do not present distinct features of salt accumulation.

Soil salinity was estimated by the salt content measured as electric conductivity (EC). Over all the sites, EC varied from 0.75 to 43.91 dS m^{-1} (Table 1). FAO regulations classify salinity of soils on the basis of EC as nonsaline 0 - 4.5 dS m^{-1}, slightly saline 4.5 - 9 dS m^{-1}, medium saline 6 - 18 dS m^{-1} and highly saline >18 dS m^{-1}. A level of EC above 8 dS m^{-1} affects all but very tolerant crops and also alters the mobility of trace elements (HATJE et al. 2003, ACOSTA et al. 2011).

The ESP index referred to the concentrations of sodium ions on cation exchange sites (CEC) ranged from 0.22 to 1.97. Such a high concentration of sodium ions in soil is characteristic for saline soils (WONG et al. 2010*b*). The content of Na^+ ions is highly correlated with the EC value ($r = 0.985$; $p<0.05$) and the concentration of Ca^{2+} ($r = 0.946$; $p<0.05$) – Table 3. Thus, salt-affected soils in the vicinity of the soda plant represent the sodium chloride type of salinization.

Table 3

Correlation coefficients between soil properties ($n = 12$; $p<0.05$)

Variable		*r*
Dependent (Y)	independent (X)	
pH H_2O	Mn_{DTPA}	0.654
pH KCl	Pb_{DTPA}	-0.622
pH KCl	Fe_{DTPA}	0.632
pH KCl	Mn_{DTPA}	-0.594
Corg	EC	0.934
Corg	ESP	0.818
Corg	Na^+	0.949
Corg	Ca^{2+}	0.899
Corg	Cl^-	0.960
N	EC	0.596
N	Na^+	0.622
N	Ca^{2+}	0.651
N	Cl^-	0.960
EC	Na^+	0.985
EC	Ca^{2+}	0.946
ESP	Ca^{2+}	0.758
ESP	Cl^-	0.871
Na^+	Cl^-	0.995
Zn_{tot}	Pb_{tot}	0.944
Zn_{tot}	Fe_{tot}	0.856

Table 4

Total trace metal concentrations in soils

No	Zn	Cu	Pb	Mn	Fe
	(mg kg^{-1})				
1	105.13	101.48	155.23	417.75	43085.0
2	122.28	85.73	127.85	396.00	43440.0
3	175.05	108.63	155.93	320.25	37495.0
4	117.95	67.18	102.98	476.00	47237.5
5	88.43	66.73	86.28	463.25	42907.5
6	86.61	59.81	72.13	567.75	31945.0
7	18.05	4.51	16.35	205.25	19660.0
8	18.30	5.12	15.55	200.50	13512.5
9	26.03	3.59	35.54	229.50	13295.0
10	35.55	8.73	27.25	209.25	21777.5
11	38.53	10.12	32.20	217.75	24075.0
12	144.50	103.62	167.75	315.25	43417.5
Min.	18.05	3.59	15.55	200.50	13295.0
Max.	175.05	108.63	167.75	567.75	47237.5
Mean	81.37	52.10	82.92	334.88	31820.0
SD	53.43	43.05	58.15	126.96	12707.6
CV	65.66	82.63	70.12	37.91	39.93

Total metal concentrations in soils are shown in Table 4. The range of concentrations was as follows: Zn: 18.05 - 175.05 mg kg^{-1} , Cu: 0.18 – 108.63 mg kg^{-1}, Pb: 15.55 - 167.75 mg kg^{-1}, Mn: 200.50 - 567.75 mg kg^{-1}, Fe: 13295.0 - 47237.5 mg kg^{-1}. Soils contained higher content of zinc, copper and lead than the geochemical background values (KOBIERSKI, DĄBKOWSKA--NASKRĘT 2012). Elevated concentrations of metals, such as Zn, Cu and Pb, and high accumulation of soluble salts may conicide, resulting in a change in the mobility and bioavailability of metals. Concentrations of some metals are strongly statistically correlated, e.g. Zn and Pb ($r = 0.944$; $p<0.05$) or Zn and Fe ($r = 0.856$; $p<0.05$) – Table 3. The correlations reveal associations between metals in soil and in the raw materials used in the Solway process of soda production. According to the European Commission (*Integrated Pollution…* 2007), approximately 73% of the polluting metals linked to Na_2CO_3 production are derived from limestone (raw material), and 21% originate from the fuel used for combustion of the raw materials.

The mobility and content of bioavailable trace metals in soils with different salinity levels were evaluated with the use of DTPA (1M diethylenetriaminepentaacetic acid) extraction. The bioavailable metal concentrations were compared with the total metal concentrations in order to determine the contribution of of phytoavailable fractions of metals in the soils. Table 5

Table 5

DTPA – extractable metals

No	Zn		Cu		Pb		Mn		Fe	
	(mg kg^{-1})	(%)	(mg kg^{-1})	(%)	(mg kg^{-1})	(%)	(mg kg^{-1})	(%)	(mg kg^{-1})	(%)
1	1.35	1.30	1.23	1.21	1.26	0.80	b.d.l.	b.d.l.	30.08	0.07
2	1.27	1.00	1.17	1.36	1.64	1.30	b.d.l.	b.d.l.	44.40	0.10
3	16.42	9.40	7.35	6.77	0.53	0.30	4.12	1.29	7.64	0.02
4	11.46	9.70	3.36	5.00	2.55	2.50	4.58	0.96	48.48	0.10
5	8.25	9.30	5.41	8.11	1.77	2.00	4.78	1.03	24.08	0.06
6	12.39	14.30	5.96	9.96	2.06	2.90	4.62	0.81	34.16	0.11
7	5.20	28.70	1.80	39.91	3.33	20.30	4.86	2.37	15.52	0.08
8	4.32	23.60	1.58	30.86	2.90	18.60	4.58	2.29	18.40	0.14
9	5.65	21.70	0.96	26.74	3.38	9.50	4.92	2.14	23.04	0.17
10	3.14	8.80	0.66	7.56	2.85	10.40	5.16	2.46	21.48	0.10
11	12.37	32.10	1.44	14.23	3.30	10.20	5.01	2.30	2.64	0.01
12	10.39	7.20	1.30	1.25	3.21	1.90	5.46	1.73	3.12	0.01
Min.	1.27	1.00	0.66	1.21	0.53	0.30	n.d.	n.d.	2.64	0.01
Max.	16.42	32.10	7.35	39.91	3.38	20.30	5.46	2.46	48.48	0.17
Mean	7.68	13.92	2.68	12.75	2.40	6.72	4.01	1.45	22.75	0.08
SD	4.92	10.27	2.28	12.83	0.94	7.02	1.90	0.90	14.79	0.05
CV	63.97	73.76	85.03	100.64	39.02	104.42	47.43	62.07	65.02	63.11

b.d.l. – below detection limit

presents the results for soils with different salinity levels. The amounts of the mobile, bioavailable fractions of Zn, Cu, Pb, Mn and Fe varied, accounting for 1.27 - 16.42 mg kg^{-1}, 0.66 - 7.35 mg kg^{-1}, 0.53 - 3.38 mg kg^{-1}, 0.00 - 5.46 mg kg^{-1} and 2.64 - 48.48 mg kg^{-1}, respectively. It can be seen from Table 5 that the DTPA extractable zinc (expressed in % of total) followed an increasing trend due to elevated salinity, expressed by the EC values. This is in agreement with results of other authors. KESHARZ et al. (2006) reported that bioavailable zinc increased by 1 to 6 % with increasing levels of salinity. Other trace metals followed various trends. Other authors have reported that a relationship between salinity and mobility of heavy metals is not linear. Such a phenomena is connected with several factors influencing metal mobility. The fate of trace metals in soil is for example governed by the chloride content. A high concentration of soil chloride can change mobilization of metals and promote the metal uptake by plants (NORVELL et al. 2000, USMAN et al. 2005). It was observed that plants accumulated more lead at high soil salinity rather than at low salinity (KADUKOVA, KALOGERAHIS 2007). Other results showed that marine water derived salts, rich in chlorides and sodium ions, added to soil caused an increase in the release of trace metals, including Zn (WONG et al. 2010*b*). It was reported that adding NaCl promoted a high release of Pb to the soil solution (Acosta et al. 201l). According to these researchers, the main mechanism regulating Pb and Cu mobility was the competition with Ca and Mg ions for sorption sites, followed by metal chloro-complexation. Others have also reported that the presence of complexing ligands may increase metal mobility in soil (Zovko, Romic 2011). The common inorganic ligand complexed with trace metals are Cl^- ions. The salinity related to increasing mobility of Zn, Pb and Cu may be attributed to the formation of stable, dissolved chloride complexes. Because of the high solubility and high mobility of chloride ions in soil, one of the factor affecting the solubility and immobilization of trace metals in soils is the formation of a complex with inorganic ligands like Cl^{-1}. Chloride ion complexes are often more mobile than free metal cations. Chloro-complexes are particularly strong when formed with metals such as Pb, Zn, Cd and Cu, and chloride ions are regarded as selective ligands for Pb (Usman et al. 2005). The formation of complexes could also lead to enhanced metal uptake through faster diffusion to roots and larger uptake. Such complexes have a lower positive charge than free metal ions or may be uncharged, thereby they present higher mobility (Usman et al. 2005). Thus, chloride ions, particularly in high concentration, contribute to the release of trace metals in soil.

Another factor affecting metal mobility in the studied soils is the high ionic strength of the soil solution. The increased metal concentrations in the available form due to the soil's high salinity led to a high ionic strength of the liquid soil phase.

An increase in the concentration of phytoavailable forms of a metal which follws an increasing ionic strength caused by as high soil salinity can be attributed to:

- cation competition for exchange sites of soil due to the increased cation content.
- a decrease in the initial activity of metal cations in the solution – formation of uncharged ion pairs and complexation with anion ligands present in the soil solution.

An increase in the content of trace metals in the phytoavailable concurrent with increasing salinity may be also attributed to the exchangeable process of Ca and Na ions with trace metals. The high ionic strength of the soil solution causes displacement of trace metals adsorbed on the soil's colloid particles.

Moreover, other mechanisms caused by chloride-sodium salinity of the soils might alter metal solubility. For instance, a change in soil acidity can influence the solubility and phytoavailability of Mn, Fe and Zn. Soil pH is the one of the most important factors affecting trace metal mobility. It is a well-known fact that higher pH values result in lower mobility and availability of cationic species like Zn, Cu, Pb, Mn and Fe (Shaheen at al. 2013). The mobility and bioavailability of the trace metals depends on a combination of dissolution – precipitation reactions as well as adsorption –

desorption and complexation processes, particularly in a saline environment. The effect of pH, redox status, complex character of the colloid matrix (mineral and organic), the nature of trace metals and anions, together with the aforementioned factors influence the mobility and phytoavailability of trace metals in saline soils (Shaeen, Frohne 2017). Those processes are complex and can lead to a fate of trace metals different than expected or reported by other authors. In general, DTPA concentrations of Zn, Cu, and Pb increased with the increasing salinity of soils in our study, although this trend was was not confirmed by statistical analysis. The increased ionic strength of the soil solution causes displacement of trace metals adsorbed in the soil (Kopittke et al. 2006). Therefore, accumulation of easily soluble salts containing a high level of Cl^{1-} might aggravate the problem of pollution. The above data indicate that the mobility and bioavailability of trace metals in the analyzed soils with different salinity levels are controlled not only by the concentration of chloride ions and salinity, but also by a range of processes and soil properties, including the nature and homogeneity of the soil components, surface charge of the soil colloids, content of organic matter, variation in soil pH, redox status and ionic strength of the soil solution. Although this study focused on a small number of samples, the results provide the information on very complex phenomena of the mobility of trace metals in soils in the presence of excessive amounts of soluble salts. Thus, the consequences of metal accumulation (due to their phytoavailability) in saline soils are hardly predictable.

CONCLUSIONS

1. The results of the study indicate that the high concentration of easily soluble salts from the waste of soda industry changes the mobility of metals in the analyzed soils.
2. The process of salt accumulation is associated with a change in the mobility and phytoavailability of trace elements in a complex way. Generally, an increase in the phytoavailable fractions of zinc and copper, as well as iron and lead was observed.
3. The above study confirmed that the ecological risk is higher when the accumulation of metals is accompanied by an increase in the salinity in soil.

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