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ORIGINAL PAPER

EFFECT OF NEUTRALISING SUBSTANCES ON THE TOTAL CONTENT OF TRACE ELEMENTS IN SOIL CONTAMINATED WITH ZINC*

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Abstract

Trace elements are particularly important not only for plants but also for soil organisms. However, excessive concentrations of trace elements can be toxic to organisms, in which they can cause various disorders. Application of mineral and organic substances increases the soil sorption capacity and reduces the uptake of heavy metals by plants. A study was undertaken to determine the content of trace elements in zinc-polluted soil after an application of some neutralising substances. Soil was polluted with zinc in doses of 0, 150, 300 and 600 mg kg^{-1} of soil. To reduce the negative impact of zinc, the substrate was amended with compost (3%), bentonite (2%) and zeolite (2% relative to the soil mass). Soil pollution with Zn and application of neutralising substances significantly affected the soil content of trace elements. In the series without neutralising substances, the increasing soil pollution with Zn caused an increase in the content of this metal and Pb, but a decrease in the content of Ni, Cu and Cd, compared with the control treatment (without Zn). Bentonite and compost reduced the content of Zn and Pb, particularly in the treatments most severely polluted with Zn. All the tested substances caused a decrease in the soil content of Ni, Cu and Co compared with the series without neutralising substances. Zeolite had the strongest effect on the content of Cu and Co, whereas compost affected most strongly the content of Ni in soil. Application of bentonite and zeolite contributed to an increase in the soil content of Cr and Fe, but the effect of bentonite was significantly stronger than that of zeolite. Moreover, zeolite resulted in a small significant rise in the soil content of Pb compared with the control.

Keywords: zinc, compost, bentonite, zeolite, soil, trace elements.

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INTRODUCTION

Trace elements are natural components of soil, rock, water, air, plants and living organisms (D`Emilio et al. 2012). Some originate from anthropogenic sources, e.g. industries and agriculture, municipal resources management or road and railroad traffic (Martinez, Motto 2000, Šichorová et al. 2004, Shi et al. 2009*a*,*b*, Zhang et al. 2009, Vodyanitskii 2010, Fijałkowski et al. 2012, Modrzewska, Wyszkowski 2014, Sun et al. 2015). Against the background of other soil-borne elements, trace elements are particularly important not only for plants but also for soil organisms (Zhang et al. 2009, D`Emilio et al. 2012). Excessive concentrations of heavy metals can be toxic to organisms, in which they can cause various disorders (Fawzy 2008, Shi et al. 2009*b*, Wyszkowski, Wyszkowska 2009, Zhang et al. 2009, Saygi, Yiğit 2012). Soil pollution with heavy metals is a growing problem because above a certain concentration in the environment these metals are noxious to plants, animals and, above all, to human health (Kaczor, Brodowska 2007, D`Emilio et al. 2012, Saygi, Yiğit 2012). Excessive soil content of heavy metals can cause water pollution and food contamination (Farrell, Jones 2010). Research on the behaviour of trace elements in soil comprises determination of their mobility and development of adequate methods to prevent contamination of the food chain (D`Emilio et al. 2012). The availability of heavy metals to plants as well as their soil solubility are mostly affected by pH (Sichorova et al. 2004, Fawzy 2008). Moreover, the accessibility of metals to plants is also affected by the quality of soil and activity of microorganisms in soil (Martinez, Motto 2000, Šichorová et al. 2004, Fijałkowski et al. 2012, Tito et al. 2012, Wyszkowska et al. 2013). The forms in which heavy metals appear in soil significantly influence their mobility; in general, Cd, Zn and Mo are considered to be mobile metals, while Cr, Ni and Pb are thought to be immobile ones (Fijałkowski et al. 2012). The mobility of zinc increases when the soil pH falls below 6-6.5. Application of mineral and organic substances (bentonites, zeolites, clays, another aluminosilicates, iron-oxides, phosphates and composts) increases the soil sorption capacity and reduces the uptake of heavy metals by plants (Fawzy 2008, Shi et al. 2009*a*,*b*, Hamidpour et al. 2010, Fijałkowski et al. 2012). In most soils, metals are bound to the clay fraction (Fawzy 2008). As a neutralising substance, zeolite is the most suitable material for remediation of soils polluted with heavy metals, as it is ecologically friendly, it adapts to the pH of the soil substrate, it is easily available and it does not supply other heavy metals to soil (Li et al. 2009, Shi et al. 2009*a*). In addition to this, zeolites immobilise these metals in soil (Antoniadis et al. 2012). On the other hand, as well as neutralising heavy metals, composts enrich soil with many nutrients but sometimes also with substances which are a hazard to the environment (Shi et al. 2009a). Phytoremediation plays a very important role in the neutralisation of heavy metals in soil (Pszczółkowski et al. 2012). Of all trace elements occurring in soil in high concentrations, Zn is the most mobile one and, additionally, it is a toxic element to plants and soil organisms (Vodyanitskii 2010).

Considering the above information, a study was undertaken to determine the content of trace elements in zinc-polluted soil after an application of some neutralising substances.

MATERIAL AND METHODS

A pot experiment was set up in a greenhouse at the University of Warmia and Mazury in Olszyn in 2012. Acid soil originating from sand was submitted to tests. The soil properties prior to the experiment are: pH at 1 mol KCl dm^{-3} – 5.32; hydrolytic acidity (HAC) – 33.6 mmol(+) kg^{-1}; total exchangeable bases Ca^{2+}, Mg^{2+}, K^{+} and Na^{+} (TEB) – 42.1 mmol(+) kg^{-1}; cation exchange capacity (CEC) – 75.7 mmol(+) kg^{-1}; base saturation (BS) – 55.6%; content of $C_{org.}$ – 13.8 g kg^{-1}; content of total-N – 0.79 g kg^{-1}; content of available forms of: phosphorus – 40.3 mg kg^{-1} (very low); potassium – 11.9 mg kg^{-1} (very low) and magnesium – 33.6 mg kg^{-1} (medium); content of total zinc – 35.8 mg kg^{-1}, total cadmium – 0.32 mg kg^{-1}, total lead – 7.22 mg kg^{-1}, total chromium – 10.23 mg kg^{-1}, total nickel – 17.08 mg kg^{-1}, total copper – 13.30 mg kg^{-1}, total manganese – 105.9 mg kg^{-1}, total iron – 3563 mg kg^{-1} and total cobalt – 2.86 mg kg^{-1} of soil. Soil was polluted with Zn (as $ZnCl_2$) in doses of 0, 150, 300 and 600 mg kg^{-1} of soil. To reduce the negative impact of Zn, the substrate was amended with compost (3%), bentonite (2%) and zeolite (2% relative to the soil mass). Once the substrate was prepared as described above, it was enriched with macro- and microelements applied in the same doses in all experimental treatments, i.e. 30 mg N ($CO(NH_2)_2$), 30 mg P (KH_2PO_4), 100 mg K (KH_2PO_4 + KCl), 50 mg Mg ($MgSO_4 \cdot 7H_2O$), 0.33 mg B (H_3BO_3), 5 mg Mn ($MnCl_2 \cdot 4H_2O$) and 5 mg Mo ($(NH_4)_6Mo_7O_{24} \cdot 4H_2O$) per 1 kg of soil, after which yellow lupine (*Lupinus luteus* L.) of the cultivar Mistral was sown. The experiment was conducted at a density of 8 plants per pot filled with 9 kg of soil. Four replications of each variants were used. The soil moisture content was maintained at 60% of the capillary water capacity during the experiment.

Soil material for laboratory analyses was sampled from each pot while harvesting lupine plants in the flowering phase. It was dried and sifted through a 1 mm mesh sieve. Afterwards, it was wet mineralised in nitric acid (HNO_3 p.a. grade) of the concentration of 1.40 g cm^{-3}. The mineralisation was carried out in a microwave oven MARS 5 (CEM Corporation, USA), in Teflon vessels HP500, according to method US-EPA3051 (1994). Afterwards, the total content of metals: zinc, cadmium, lead, chromium, nickel, manganese, copper, iron and cobalt, was determined by flame atomic absorption spectrometry (FAAS) in an air-acetylene flame in a SpectrAA 240FS (VARIAN, Australia) atomic absorption spectrometer. The equipment was calibrated using certified standard solutions by Fluke and Merck. Before the experiment was started, the properties of soil were analysed: pH – by the potentiometric method at 1 mole KCl dm^{-3} hydrolytic acidity (HAC) and the total exchangeable bases (TEB) – by the Kappen method, the content of organic carbon (C_{org}) – by the Tiurin method, available phosphorus and potassium – by the Egner-Riehm method and available magnesium – by the Schachtschabel method (Lityński et al. 1976). Cation exchange capacity (CEC) and base saturation (BS) were calculated from the following formulas: CEC = TEB + HAC; BS = (TEB : CEC) 100 (Lityński et al. 1976). The results underwent statistical processing using two-factorial analysis of variance ANOVA, PCA and correlation coefficients, calculated in a software package Statistica (StatSoft, Inc. 2014).

RESULTS AND DISCUSSION

The results demonstrate that soil contamination with zinc together with an application of a neutralising substance such as compost, bentonite or zeolite significantly affected the soil content of trace elements (Tables 1-3). In the series with no neutralising substances added, the increasing degree of soil pollution with Zn caused an increase in the soil content of this metal ($r = 0.994^{**}$) and Pb ($r = 0.936^{**}$) as well as a decrease in the content of Ni ($r = -0.880^{**}$), Cu ($r = -0.867^{**}$) and Cd ($r = -0.723^{*}$), compared with the control variant (without Zn). The soil content of zinc increased over 12-fold, while that of lead more than doubled. The content of chromium in soil continued to increase until the dose of 300 mg kg^{-1}, after which it decreased. Under the influence of 600 mg kg^{-1} of soil, the content of nickel decreased by 61%, copper by 34% and cadmium by 13%, compared with the variant with no added Zn.

The positive effect on reducing the content of zinc and lead was produced only by compost and bentonite (Tables 1-3, Figure 1). They depressed the content of zinc by an average of 17 and 14%, and that of lead by 24 and 10 %, respectively, compared with the series without any soil neutralising substances. Application of any of the three tested substances reduced the soil content of nickel, copper and cobalt (Tables 1-3, Figure1). However, zeolite had a stronger effect on the content of copper and cobalt, while compost affected more distinctly the content of nickel in soil. The content of copper and cobalt in soil was reduced under the influence of zeolite by an average of 52 and 68%, respectively, while the content of nickel and cobalt decreased by 42% in response to compost. The influence of the other substances on the content of these metals in soil was weaker, reducing them by 27-38%. The soil application of bentonite and zeolite contributed to an increase in the soil content of chromium by 36 and 12%, and that of iron by 22 and 13% , respectively. Zeolite also caused a relatively small (14%) increase in the soil content of lead versus the control treatment. Neither the organic substance,

Table 1

Content of zinc, cadmium and lead in soil (mg kg^{-1} d.m.)

Dose of zinc (mg kg^{-1} of soil)	Type of substance neutralising effect of zinc				
	without additions	compost	bentonite	zeolite	average
Zinc					
0	35.5	35.5	42.2	35.2	37.1
150	142.4	139.6	148.9	161.6	153.1
300	277.2	275.2	243.4	282.3	269.5
600	441.6	292.6	335.9	475.1	386.3
Average	229.2	185.7	192.6	238.6	211.5
r	0.994**	0.902**	0.977**	0.997**	0.983**
LSD for:	Zn dose - 5.26**, type of neutralising substance - 5.26**, interaction - 10.53**				
Cadmium					
0	0.30	0.26	0.28	0.25	0.27
150	0.27	0.28	0.30	0.28	0.28
300	0.25	0.26	0.30	0.25	0.27
600	0.26	0.26	0.29	0.27	0.27
Average	0.27	0.27	0.29	0.26	0.27
r	-0.723**	-0.293	0.255	0.293	-0.398
LSD for:	Zn dose - n.s., type of neutralising substance - n.s., interaction - n.s.				
Lead					
0	7.09	4.69	16.09	12.67	10.14
150	11.37	8.28	11.75	16.82	12.06
300	15.44	11.60	7.62	16.97	12.91
600	17.19	14.26	10.70	11.94	13.52
Average	12.77	9.71	11.54	14.60	12.16
r	0.936**	0.969**	-0.612*	-0.269	0.910**
LSD for:	Zn dose - 1.28**, type of neutralising substance - 1.28**, interaction - 2.56**				

Significant for: ** $P = 0.01$, * $P = 0.05$, n.s. – non-significant, r – correlation coefficient ($n = 12$)

such as compost, nor the mineral ones, such as bentonite and zeolite, had any significant effect on the content of cadmium and manganese in soil.

The PCA and correlation coefficients (Table 4, Figure 2) point up significant correlations between certain trace elements as well as between zinc, chromium and iron versus the soil acidity and sorption properties. The PCA results show that the principal components represent 58.48% of variance, of which 30.13% is represented by the first component and 28.35% – by the second one. The plotted vectors correspond very faithfully to the content of

Table 2

Content of chromium, nickel and copper in soil (mg kg^{-1} d.m.)

Dose of zinc (mg kg^{-1} of soil)	Type of substance neutralising effect of zinc				
	without additions	compost	bentonite	zeolite	average
Chromium					
0	9.80	12.55	14.80	13.50	12.66
150	9.70	9.30	14.20	12.70	11.48
300	11.45	9.05	15.35	10.80	11.66
600	10.55	8.65	12.25	9.50	10.24
Average	10.38	9.89	14.15	11.63	11.51
r	0.524	-0.784**	-0.744**	-0.977**	-0.944**
LSD for:	Zn dose - 1.43*, type of neutralising substance - 1.43**, interaction - 2.85**				
Nickel					
0	16.80	6.10	8.05	7.70	9.66
150	17.80	7.00	8.55	9.35	10.68
300	8.35	7.30	9.35	8.35	8.34
600	6.55	6.35	6.55	6.70	6.54
Average	12.38	6.69	8.13	8.03	8.80
r	-0.880**	0.066	-0.567*	-0.583*	-0.887**
LSD for:	Zn dose - 0.62**, type of neutralising substance - 0.62**, interaction - 1.24**				
Copper					
0	13.35	8.75	5.55	3.45	7.78
150	10.25	6.15	5.35	3.50	6.31
300	9.70	5.90	9.80	6.20	7.90
600	8.85	6.40	5.60	7.30	7.04
Average	10.54	6.80	6.58	5.11	7.26
r	-0.867**	-0.601*	0.120	0.935**	-0.141
LSD for:	Zn dose - 0.59**, type of neutralising substance - 0.59**, interaction - 1.19**				

Significant for: ** $P = 0.01$, * $P = 0.05$, r – correlation coefficient ($n = 12$)

iron, followed by chromium, copper, nickel, cadmium and cobalt, while bearing the least correspondence with the content of zinc and manganese. The highest positive correlation coefficients at the significance level of $P = 0.01$ were achieved between iron versus chromium and manganese, cobalt versus nickel and copper, and nickel versus copper. The results of PCA confirmed that bentonite and zeolite had a stronger effect on the content of trace elements in soil than compost (Figure 3).

The experiment made by Li et al. (2009) indicates that soil addition of zeolite limits the availability of lead to plant and simultaneously improves

Table 3

Content of manganese, iron and cobalt in soil (mg kg^{-1} d.m.)

Dose of zinc (mg kg^{-1} of soil)	Type of substance neutralising effect of zinc				
	without additions	compost	bentonite	zeolite	average
Manganese					
0	109.2	103.5	116.2	106.6	108.9
150	110.9	107.9	111.7	107.5	109.5
300	110.2	107.7	113.4	111.0	110.6
600	108.3	104.3	112.1	119.1	110.9
Average	109.6	105.8	113.3	111.0	110.0
r	-0.525	-0.040	-0.637*	0.979**	0.944**
LSD for:	Zn dose - n.s., type of neutralising substance - 3.21**, interaction - n.s.				
Iron					
0	3600	3783	4572	4318	4068
150	3604	3698	4300	4299	3975
300	3667	3554	4732	3862	3954
600	3740	3095	4291	4059	3796
Average	3653	3533	4474	4135	3948
r	0.976**	-0.981**	-0.351	-0.615*	-0.987**
LSD for:	Zn dose - n.s., type of neutralising substance - 145**, interaction - n.s.				
Cobalt					
0	3.00	1.05	2.60	0.55	1.80
150	2.95	2.55	1.50	1.40	2.10
300	2.20	2.50	1.75	0.65	1.78
600	2.80	0.30	2.10	0.95	1.54
Average	2.74	1.60	1.99	0.89	1.80
r	-0.325	-0.430	-0.210	0.147	-0.696**
LSD for:	Zn dose - 0.81*, type of neutralising substance - 0.81**, interaction - 1.62*				

Significant for: ** $P = 0.01$, * $P = 0.05$, n.s. – non-significant, r – correlation coefficient ($n = 12$)

the quality of soil. Likewise, SHI et al. (2009*b*) demonstrated experimentally that zeolite introduced to soil led to a reduced uptake of cadmium and lead by wheat. An identical relationship, that is a considerable decrease in available cadmium and lead in soil following the soil enrichment with zeolite, was recorded by FAWZY (2008) and by HAMIDPOUR et al. (2010), with the resulting lower content of these metals in plants. SHI et al. (2009*b*) showed that zeolite added to soil caused a substantial decrease in the soil content of copper, cobalt and nickel. A similar relationship was noted by ANTONIADIS et al. (2012), who found that addition of zeolite or lime to soil caused a small dec-

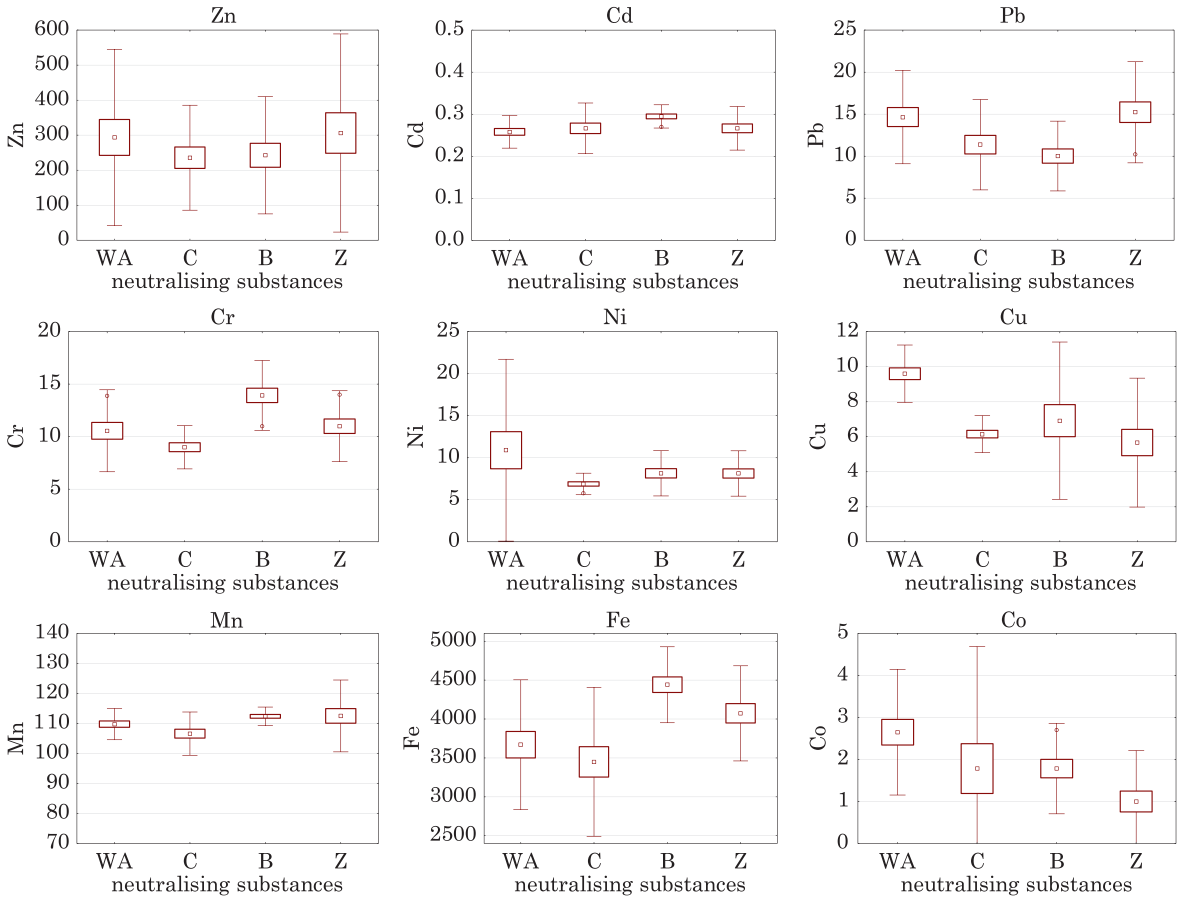

Fig. 1. Average content (±SD) of trace elements in soil contaminated with 150-600 mg Zn kg^{-1} (mg kg^{-1} d.m.) of soil: WA – without additions, C – compost, B – betonite, Z – zeolite

Table 4

Correlation coefficients (r) between content of trace elements and some properties of soil

Variable	Cd	Pb	Cr	Ni	Cu	Mn	Fe	Co
Zn	-0.111	0.365*	-0.344	-0.357*	0.008	0.292	-0.142	-0.137
Cd		-0.239	0.347	0.200	0.101	0.339	0.322	0.235
Pb			0.003	-0.185	-0.405*	0.136	0.038	-0.195
Cr				-0.119	-0.236	0.347	0.844**	-0.077
Ni					0.581**	0.092	-0.103	0.500**
Cu						0.052	-0.280	0.478**
Mn							0.591**	0.165
Fe								-0.047

Significant for ** $P = 0.01$, * $P = 0.05$, r – correlation coefficient ($n = 48$)

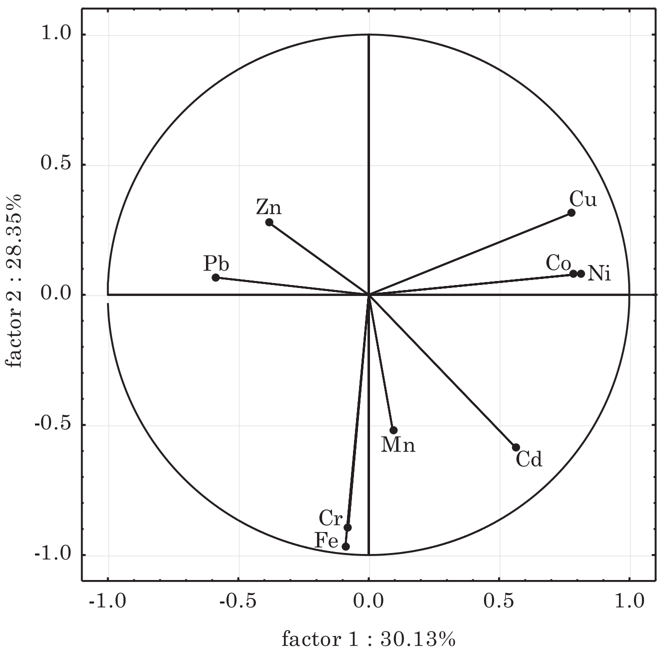

Fig. 2. Content of trace elements in the soils illustrated with the PCA method: vectors represent the analysed variable (content of Zn, Cd, Pb, Cr, Ni, Cu, Mn, Fe and Co)

line in the availability of copper and zinc to plants. In another experiment by Hamidpour et al. (2010), zeolite was distinguished by higher adsorption of cadmium and lead than bentonite and had a higher potential for immobilising cadmium and lead in polluted soil. After some time, trace elements immobilised in soil can again become available to plants, but under natural conditions they may be removed from soil by plants which have incorporated

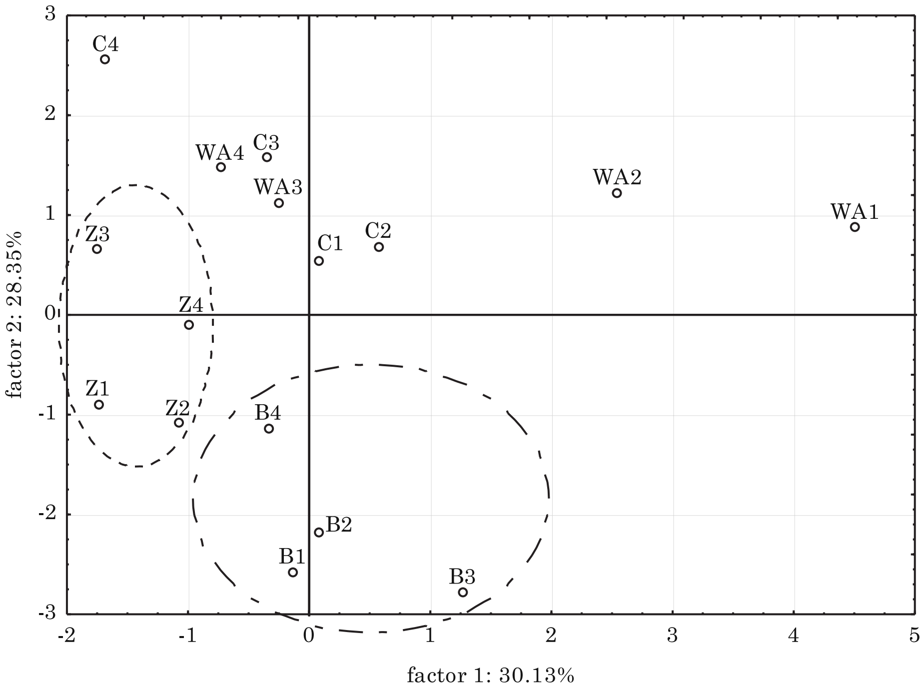

Fig. 3. Effect of neutralising substances on the content of trace elements in the soils illustrated with the PCA method: points show soil samples with trace elements (WA – without additions, C – compost, B – betonite, Z – zeolite; 1 - 0, 2 - 150 mg, 3 - 300 mg, 4 - 600 mg Zn kg^{-1} of soil)

them into plant tissues (Hamidpour et al. 2010). According to Li et al. (2009), elevation of soil pH favours sorption of heavy metals by zeolite and their immobilisation, although they can be arrested in zeolite irrespective of soil pH owing to cation exchange. An increase in the soil pH in the cited study caused immobilisation of lead and decrease of lead availability to plants. Addition of larger amounts of organic matter to soil helps to minimise the availability of heavy metals to plants (Fijałkowski et al. 2012). The experiment conducted by Shi et al. (2009a) proved that a supply of humic substances to soil together with zeolite not only effectively immobilised lead in soil but also contributed to a decrease in the content of this element in oilseed rape. In the research conducted by Wyszkowski and Sivitskaya (2013, 2014), zeolite decreased the content of cadmium and lead while compost and bentonite produced the same effect on the content of copper and iron in soil. The study of Farrell and Jones (2010) indicates positive consequences of using composts for remediation of polluted ground, regardless of the elevated soil pH and adsorption of such trace elements as copper, lead and zinc, which results in their lower mobility and availability to plants. Herwijnen et al. (2007), having added compost to soil, demonstrated immobilisation of lead and zinc in soil and improved phytoavailability of copper, an important element for plants. Sun et al. (2015) showed that an application of bentonite caused decreased bioavailability of heavy metals in soil, especially cadmium and lead, which had a positive effect on the test plants having a lower content of these elements. In their opinion, bentonite can be an effective material in remediation of soil polluted with trace elements. However, should soil conditions change, the previously immobilised heavy metals can be released again and possibly pollute both the soil substrate and waters. Another experiment, by Tito et al. (2012), verifies the positive influence of bentonite added to soil owing to its high porosity, including micropores, which helps to immobilise excessive quantities of zinc and copper in soils.

CONCLUSIONS

1. Soil pollution with zinc and application of neutralising substances significantly affected the soil content of trace elements.

2. In the series without neutralising substances, the increasing soil pollution with zinc caused an increase in the content of zinc and lead in soil but a decrease in the content of nickel, copper and cadmium, compared with the control treatment (without Zn).

3. Among the substances applied to soil, bentonite and compost reduced the content of zinc and lead, particularly in the treatments most severely polluted with zinc.

4. All the tested substances caused a decrease in the soil content of nickel, copper and cobalt compared with the series without neutralising substances. Zeolite had the strongest effect on the content of copper and cobalt, whereas compost affected most strongly the content of nickel in soil.

5. Application of bentonite and zeolite contributed to an increase in the soil content of chromium and iron, but the effect of bentonite was significantly stronger than that of zeolite. Moreover, zeolite resulted in a small significant rise in the soil content of lead compared with the control.

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