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EFFECT OF NEUTRALIZING SUBSTANCES ON THE CONTENT OF TRACE ELEMENTS IN SOIL CONTAMINATED WITH COBALT

The aim of the study was to determine the effect of increasing doses of cobalt (0, 20, 40, 80, 160, 320 mg/kg d.m. of soil) on the total content of trace elements in soil after application of manure, clay, charcoal, zeolite and calcium oxide. The neutralizing substances were applied at 2% of the soil weight, and calcium oxide at a dose corresponding to one hydrolytic acidity. The content of the cobalt, cadmium, lead, chromium, nickel, zinc, copper, manganese and iron was determined in soil. The contamination of soil with cobalt and the application of neutralizing substances had significant effects on the total content of trace elements in soil In the series without substances, the soil contamination with cobalt increased the content of cobalt, lead, chromium, nickel and zinc in soil. All the neutralizing substances reduced the content of cobalt, manganese and iron in soil. The highest decrease in the cobalt content was observed in the series with manure, whereas the highest decrease in zinc occurred after addition of charcoal. The decrease in the content of the other metals (except nickel and lead) was observed in the pots with CaO and zeolite. The effect of other neutralizing substances depended on the trace element.

1. INTRODUCTION

The presence of heavy metals in soil results from both natural and anthropogenic phenomena. The effect of anthropogenic factors on the environment is becoming increasingly significant due to human activities, mainly associated with the development of industry, transport and agriculture. The mobility of heavy metals in soil from anthropogenic sources is much larger compared to metals from natural sources [1]. The presence of high concentrations of metals in soil potentially has an adverse effect on the growth and development of plants, animals and humans [2, 3].

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Cobalt is one of numerous heavy metals present in soil. Its content depends mainly on the nature of the bedrock and the type of soil. The highest content of cobalt is observed in fen soils and limestone soils and the lowest – in sandy podzolic soils. The increase in the content of cobalt in soil is mainly due to human activities [4]. The natural cobalt content in the Earth's crust and in the soils (also in Polish soils) are lower than 12 mg/kg [5]. In Poland, the cobalt content in arable layer of soil should not be higher than 20 mg/kg of dry matter [6]. The permissible content of cobalt in Polish soils is exceeded sporadically. This is a problem in some other countries of the world [7].

Cobalt is a very important element for people, plants and other living organisms. The main sources of cobalt in soil include its mining and use in the production of steels, alloys, paints and catalysts as well as oil and coal combustion [8, 9]. Both its deficit and excess in soil can contribute to a decrease in the nutritional value of plants, which are the initial link in the food chain. The first symptoms of an excessive content of cobalt in soil include chlorosis and necrosis [10]. For leguminous plants, growth of nodules is inhibited, which reduces nitrogen fixing [11]. Therefore, it is important to reduce cobalt content in soil.

The aim of the study was to determine the effect of neutralizing substances on the content of trace elements in soil contaminated with cobalt.

2. MATERIAL AND METHODS

This study was based on a pot experiment carried out in a plant growing room at the University of Warmia and Mazury in Olsztyn, in soil with the following properties: pH in 1 M KCl – 5.05, hydrolytic acidity – 28.40 mmol(+)/kg, total exchangeable bases (TEB) – 46.50 mmol(+)/kg, cation exchange capacity (CEC) – 74.90 mmol(+)/kg, base saturation (BS) – 68.08%. The effect of contamination of soil with cobalt (as $CoCl_2$) at 0, 20, 40, 80, 160 and 320 mg Co/kg was tested on spring barley (*Hordeum vulgare* L.), cultivar Mercada (the principal crop) and white mustard (*Sinapis alba* L.) cultivar Bamberka (the successive crop). Substances which neutralise the effect of cobalt such as manure, clay, charcoal, zeolite and calcium oxide were also added to the soil. They were applied at 2% of the soil weight in a pot along with calcium oxide at a dose corresponding to 1 unit of hydrolytic acidity (HAC). Apart from the neutralizing substances, similar amounts of nutrients were added to the soil: 100 mg N, 35 mg P, 100 mg K, 50 mg Mg, 0.33 mg B, 5 mg Mn and 5 mg Mo/kg of soil. Soil (9 kg) was transferred to polyethylene pots and the plants were sown. Moisture content was maintained at 60% of the capillary water capacity during the plant growth. Spring barley was harvested during the ear formation phase and white mustard was harvested during the blossoming phase and soil samples were then taken for analyses.

The soil was dried and sieved through a 1 mm mesh sieve. Subsequently, wet mineralisation of the soil samples was carried out in concentrated nitric acid (HNO_3 analytically pure) at a concentration of 1.40 g/cm^3 in a MARS 5 microwave oven (CEM Corporation, USA) in HP 500 vessels, made of Teflon, by the US-EPA3051 method [12]. The following were determined in the prepared soil by flame atomic absorption spectrometry in an air-acetylene flame: total cobalt, cadmium, lead, chromium, nickel, zinc, copper, manganese and iron. The assays were carried out with Fluka standard solutions with the symbols: Co 119785.0100, Cd 51994, Pb 16595, Cr 02733, Ni 42242, Zn 188227, Cu 38996, Mn 63534 and Fe 16596. The results were compared with the certified analytical reference material – soil A-1 from the AGH University of Science and Technology in Kraków and were worked out statistically with an ANOVA two-way analysis of variance using the Statistica software package [13]. The effect of cobalt and the neutralizing substances on the content of trace elements in soil was assessed by the principal component analysis (PCA); the correlation coefficients have also been calculated.

3. RESULTS AND DISCUSSION

The content of trace elements in soil depended on dose of cobalt and the addition of neutralizing substances (Tables 1–3). In a series without neutralizing substances, the highest contamination with cobalt caused a significant increase in the content of cobalt (r = 1.000), zinc (r = 0.857) and nickel (r = 0.744) in the soil. The content of these elements increased upon increasing the dose of Co to 320 mg/kg of soil. The highest increase was observed for cobalt (25-fold), compared to the soil non-contaminated with cobalt. Zinc and nickel content increased by 108% on average and 25%, respectively, compared to the soil without cobalt. The largest increase in lead (by 63% on average) and chromium (by 58% on average) content was observed in the soil contaminated with 80 mg of Co/mg of soil. Contamination of soil with the largest dose of cobalt significantly reduced the content of cadmium (r = –0.865) in soil (by 23% on average), compared to the soil non-contaminated with cobalt. The largest decrease in manganese content (by 14% on average) was observed in the soil contaminated with 160 mg of Co/kg of soil. Increasing doses of cobalt had a small effect on the content of copper and iron, compared to the content in soil without cobalt.

Introducing the neutralizing substances (except for manure) to soil reduced the content of manganese and iron in soil, compared to soil without their addition (Tables 1, 3). The largest decrease in cobalt content (by 33% on average) was observed in the series in which manure was added and the decrease was much smaller (by 6–12% on average) after the other substances were added. Application of calcium oxide and zeolite to the soil significantly reduced the content of chromium (by 78 and 75% on average), manganese (by 41% and 34%), iron (by 42% and 37%) and cadmium (by 34% and 31%).

However, the content of zinc in soil decreased (by 84%) in the series with an addition of charcoal (Table 2).

Table 1

Total content of cobalt, cadmium and lead in soil [mg/kg]

Dose of cobalt [mg/kg]	Kind of substance neutralizing the effect of cobalt						
	None	Manure	Clay	Charcoal	Zeolite	CaO	Average
Cobalt							
0	10.47	10.47	11.49	13.24	8.31	5.13	9.85
20	29.04	21.86	24.63	28.02	19.50	21.55	24.10
40	40.13	32.64	41.05	31.82	33.36	40.95	36.66
80	76.26	52.65	74.61	71.12	62.71	69.38	67.79
160	131.68	88.67	107.56	128.60	117.92	133.01	117.91
320	259.56	159.69	254.63	227.64	239.13	231.64	228.71
Average	91.19	61.00	85.66	83.41	80.16	83.61	80.84
r	1.000[b]	0.999[b]	0.993[b]	0.998[b]	1.000[b]	0.998[b]	1.000[b]
LSD	*a* = 8.22[b], *b* = 8.22[b], *ab* = 20.13[b]						
Cadmium							
0	1.23	1.20	1.05	0.93	0.95	0.87	1.04
20	1.31	1.10	1.17	1.10	0.92	0.84	1.07
40	1.23	1.20	1.02	0.92	0.77	0.84	1.00
80	1.17	0.99	0.95	0.83	0.80	0.75	0.91
160	1.25	1.01	2.70	0.69	0.86	0.65	1.19
320	0.95	0.95	0.89	0.84	0.63	0.74	0.83
Average	1.19	1.07	1.30	0.88	0.82	0.78	1.01
r	–0.865[b]	–0.795[b]	0.125	–0.537[a]	–0.812[b]	–0.678[b]	–0.443
LSD	*a* = 0.51[b], *b* = 0.51[b], *ab* = 1.24[b]						
Lead							
0	6.28	10.53	10.98	12.99	8.08	9.66	9.75
20	5.72	11.13	12.45	10.08	10.73	11.61	10.29
40	8.34	13.37	11.57	11.48	12.23	11.53	11.42
80	10.26	11.22	9.55	10.78	14.78	10.71	11.22
160	9.90	10.33	16.41	11.83	10.10	10.24	11.47
320	7.62	11.22	13.39	11.37	10.34	10.34	10.71
Average	8.02	11.30	12.39	11.42	11.04	10.68	10.81
r	0.286	–0.166	0.496[a]	–0.048	–0.027	–0.272	0.291
LSD	*a* = 1.65[b], *b* = 1.65[b], *ab* = 4.03[b]						

[a]Significant at $P = 0.05$.
[b]Significant at $P = 0.01$.
LSD for: *a* – cobalt dose, *b* – kind of neutralizing substance, *ab* – interaction, *r* – correlation coefficient.

The addition of all neutralizing substances to the soil significantly increased the content of lead in the soil, compared to soil without additions (Table 1). The content of this element increased the most (by 55% on average) in the soil with clay. It this soil the content of cadmium and chromium increased by 9% and 191% on average, respectively (Tables 1, 2).

Table 2

Total content of chromium, nickel and zinc in soil [mg/kg]

Dose of cobalt [mg/kg]	Kind of substance neutralizing the effect of cobalt						
	None	Manure	Clay	Charcoal	Zeolite	CaO	Average
Chromium							
0	58.47	100.48	223.88	217.07	11.76	21.94	105.60
20	67.05	115.79	231.93	226.09	29.90	24.41	115.86
40	81.91	168.51	234.14	218.75	21.85	11.06	122.70
80	92.26	191.33	233.34	228.74	20.96	16.54	130.53
160	91.20	209.28	234.76	224.85	16.45	16.36	132.15
320	87.30	225.65	235.73	225.38	18.04	13.71	134.30
Average	79.70	168.51	232.30	223.48	19.83	17.34	123.52
r	0.613[b]	0.839[b]	0.632[b]	0.406	–0.201	–0.495[a]	0.777[b]
LSD	*a* = 9.79[b], *b* = 9.79[b], *ab* = 23.99						
Nickel							
0	2.94	2.71	2.80	4.32	4.41	5.19	3.73
20	2.99	2.20	3.49	3.63	4.46	4.92	3.61
40	3.35	3.17	2.99	5.05	4.69	4.69	3.99
80	2.85	5.05	2.80	3.35	4.69	4.18	3.82
160	3.08	2.71	4.59	4.00	4.04	5.01	3.90
320	3.67	2.62	4.59	4.36	4.82	5.56	4.27
Average	3.15	3.08	3.54	4.12	4.52	4.92	3.89
r	0.744[b]	–0.101	0.822[b]	0.069	0.244	0.526[a]	0.860[b]
LSD	*a* = 0.67[b], *b* = 0.67[b], *ab* = 1.65						
Zinc							
0	6.14	11.67	10.26	2.85	2.41	6.09	6.57
20	4.21	13.23	10.62	0.58	3.27	7.74	6.61
40	8.25	11.61	11.22	0.70	2.54	8.04	7.06
80	9.81	10.58	7.60	0.49	4.27	7.04	6.63
160	8.78	10.90	10.15	0.23	5.70	8.08	7.31
320	12.76	10.98	3.68	3.06	6.22	8.69	7.56
Average	8.33	11.49	8.92	1.32	4.07	7.61	6.96
r	0.857[b]	–0.514[a]	–0.852[b]	0.361	0.915[b]	0.711[b]	0.878[b]
LSD	*a* = 1.43[b], *b* = 1.43[b], *ab* = 3.51						

[a]Significant at $P = 0.05$.
[b]Significant at $P = 0.01$.
LSD for: *a* – cobalt dose, *b* – kind of neutralizing substance, *ab* – interaction, *r* – correlation coefficient.

All of the substances, except for manure, had a similar effect on the content of nickel in the soil. The addition of calcium oxide to soil increased the content of nickel (by 56%

on average) and copper (by 49% on average). The addition of zeolite increased the content of copper (by 53% on average) and the addition of manure increased the content of zinc (by 38% on average) (Tables 2, 3). The effect of these neutralizing substances varied from one element to another.

Table 3

Total content of copper, manganese and iron in soil [mg/kg]

Dose of cobalt [mg/kg]	Kind of substance neutralizing the effect of cobalt						
	None	Manure	Clay	Charcoal	Zeolite	CaO	Average
Copper							
0	32.89	33.83	32.72	32.97	52.77	41.01	37.70
20	33.14	34.17	32.03	32.37	46.32	46.24	37.38
40	33.06	34.17	33.83	32.54	38.04	47.11	36.46
80	32.03	33.49	33.83	32.80	48.59	61.58	40.39
160	33.49	33.66	35.03	33.57	63.41	55.65	42.47
320	34.09	32.97	33.66	32.80	55.13	44.58	38.87
Average	33.12	33.71	33.52	32.84	50.71	49.36	38.88
r	0.688[b]	–0.867[b]	0.475	0.312	0.519[a]	0.065	0.424
LSD	*a* – 3.89[b], *b* – 3.89[b], *ab* – 9.54[b]						
Manganese							
0	407.30	363.42	363.08	312.93	243.13	228.56	319.74
20	359.02	345.63	364.95	298.02	272.78	211.95	308.72
40	350.71	354.27	362.40	281.76	233.47	218.22	300.14
80	355.63	437.12	314.63	284.13	250.58	214.16	309.37
160	350.21	329.20	298.19	280.23	222.63	210.94	281.90
320	360.54	345.63	319.54	252.78	214.83	196.03	281.56
Average	363.90	362.55	337.13	284.98	239.57	213.31	300.24
r	–0.349	–0.237	–0.676[b]	–0.909[b]	–0.744[b]	–0.888[b]	–0.853[b]
LSD	*a* – 29.67[b], *b* – 29.67[b], *ab* – 72.69[b]						
Iron							
0	11625.59	11363.36	11576.80	9019.51	7644.76	7145.74	9729.29
20	11104.62	11014.74	11397.88	8808.86	7862.96	6578.65	9461.29
40	11358.60	11879.57	12073.72	8710.03	7050.97	6639.74	9618.77
80	11599.31	11651.03	10464.28	8566.48	7687.67	6695.79	9444.09
160	11861.81	10878.60	10573.17	8607.15	7033.50	6572.64	9254.48
320	11498.10	11734.61	10513.49	7789.58	6460.96	6232.00	9038.12
Average	11508.01	11420.32	11099.89	8583.60	7290.14	6644.09	9424.34
r	0.301	0.175	–0.683[b]	–0.955[b]	–0.850[b]	–0.794[b]	–0.939[b]
LSD	*a* – 357.49[b], *b* – 357.49[b], *ab* – 875.66[b]						

[a]Significant at $P = 0.05$.
[b]Significant at $P = 0.01$.
LSD for: *a* – cobalt dose, *b* – kind of neutralizing substance, *ab* – interaction, *r* – correlation coefficient.

Table 4

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

Factor	Co	Cd	Pb	Cr	Ni	Zn	Cu	Mn	Fe
pH_{KCl}	–0.219	–0.315	–0.065	–0.421[a]	0.379[a]	0.117	0.411[a]	–0.448[b]	–0.503[b]
HAc	0.282	0.303	0.015	0.356[a]	–0.389[a]	0.007	–0.333[a]	0.438[b]	0.506[b]
TEB	–0.318	–0.240	–0.068	–0.386[a]	0.291	0.120	0.360[a]	–0.393[a]	–0.423[a]
CEC	–0.316	–0.202	–0.086	–0.379[a]	0.235	0.166	0.352[a]	–0.354[a]	–0.367[a]
BS	–0.381[a]	–0.252	–0.056	–0.338[a]	0.292	0.072	0.290	–0.361[a]	–0.416[a]
Co		–0.176	0.053	0.046	0.227	0.051	0.113	–0.208	–0.127
Cd			0.227	0.292	–0.251	0.313	–0.372[a]	0.410[a]	0.503[b]
Pb				0.352[a]	0.290	0.020	–0.026	–0.173	–0.103
Cr					–0.361[a]	0.005	–0.717[b]	0.515[b]	0.550[b]
Ni						–0.409[a]	0.493[b]	–0.668[b]	–0.749[b]
Zn							–0.128	0.437[b]	0.531[b]
Cu								–0.740[b]	–0.743[b]
Mn									0.956[b]

[a]Significant at $P = 0.05$.
[b]Significant at $P = 0.01$.
pH_{KCl} – pH in 1 M KCl, HAc – hydrolytic acidity, TEB – total exchangeable bases, CEC – cation exchange capacity, BS – base saturation.

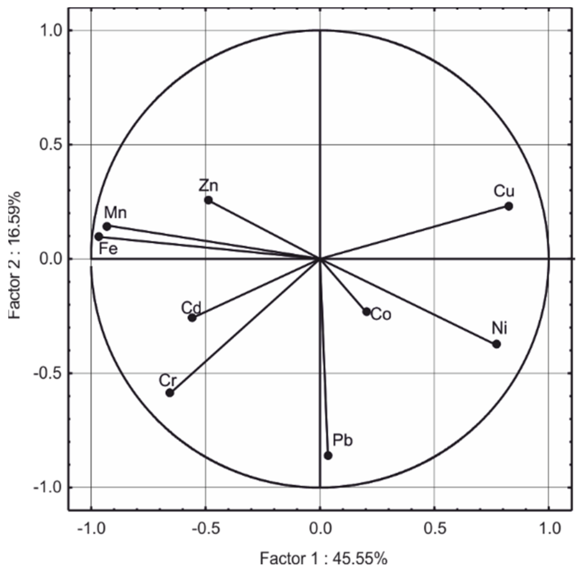

Fig. 1. Total content of elements in the soils illustrated by the PCA method; vectors represent analyzed variables (contents of Co, Cd, Pb, Cr, Ni, Zn, Cu, Mn and Fe)

The correlation coefficients and the PCA analysis (Table 4, Fig. 1) indicate relationships between the total trace elements under study, especially between manganese, iron, as well as copper, nickel and chromium and the other trace elements. The highest positive correlation coefficients were observed between iron and manganese and between iron and cadmium as well as between iron, manganese and chromium, zinc; copper and nickel. Between iron and manganese, nickel and copper, copper and chromium a negative correlation was observed. Significant correlations were found between the content of some trace elements and acidity and sorptive properties of soil, including the content of iron and manganese and pH and hydrolytic acidity. The PCA analysis indicates that zeolite and calcium oxide had the greatest effect and charcoal had the least effect on the content of trace elements in soil (Fig. 2).

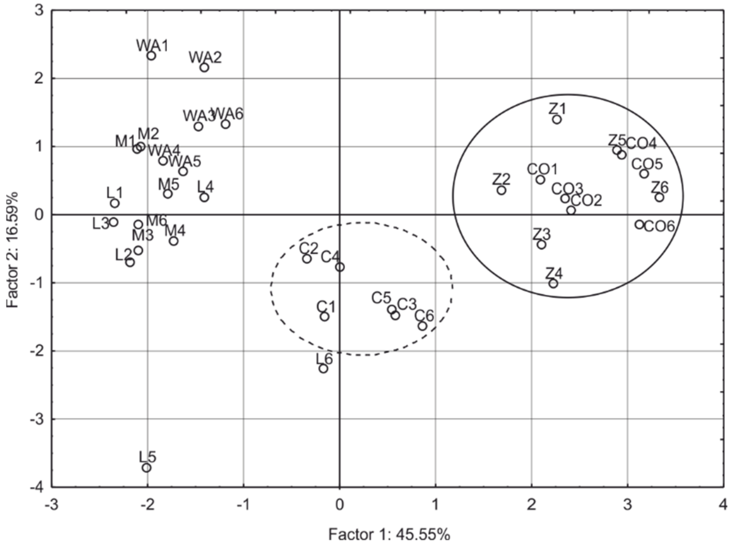

Fig. 2. Effect of neutralizing substances on total content of trace elements in the soils illustrated by the PCA method; points show soil samples with trace elements (WA – without additions, M – manure, L – clay, C – charcoal, Z – zeolite, CO – CaO, 1 – 0, 2 – 20, 3 – 40, 4 – 80, 5 – 160, 6 – 320 mg Co/kg of soil)

The content of trace elements in soil largely depends on the type and chemical composition of soil. Coarse soils have a much smaller tendency to absorb metals compared to fine-grain soils. Moreover, the soil pH is also important [14]. The absorbability of heavy metals from soil increases with decreasing pH [15, 16]. The content of heavy metals as observed in this study was affected by the dose of cobalt and the content of the neutralizing substance. Therefore, it can be concluded that these factors brought about a pH change, thereby affecting the content of the trace elements in soil. The effect of contamination with cobalt and the type of neutralizing substance of the soil pH was confirmed by Kosiorek and Wyszkowski [17]. Changes in the physicochemical properties of soil are accompanied with a competition among trace elements for its sorptive area [14]. A high content of cobalt in soil can affect other elements by decreasing or increasing their availability to plants. According to Li et al. [18], the content of cobalt in soil is strongly correlated to that of manganese and iron. Bradl [19] indicated a correlation between the content of zinc and cadmium in soil solution. The content of trace elements in soil and plants depends on fertilisers [20]. A beneficial effect of magnesium and calcium in reducing the availability of cobalt to plants was found by Micó et al. [9] and Li et al. [18]. A significant decrease in the content of cobalt in soil following the application of manure was also observed in this study. Zeolite has a beneficial effect related to the availability of heavy metals due to a large sorption area and their consequent adsorption [21, 22]. Wyszkowski and Sivitskaya [23] found zeolite to reduce the content of cadmium and lead in soil. According to Dickinson [24], zeolite addition to soil increased pH, which reduces the mobility of heavy metals in soil. A considerable decrease in the content of cadmium, chromium, zinc, manganese and iron in soil observed in this study confirmed the specific properties of zeolite demonstrated by these authors. The addition of calcium oxide to soil reduced the content of these metals in soil much more strongly. Abd El-Azeem et al. [25] and Guodong [26] report that calcium application to soil reduces the availability of heavy metals by increasing the soil pH [27]. On the other hand [23, 28], substances such as compost, bentonite and calcium oxide did not reduce the content of manganese, cadmium, chromium or lead [23], although they did with copper and iron in soil [28].

4. CONCLUSIONS

The total content of trace elements in soil depended on the soil contamination with cobalt and addition of neutralizing substances. In the series without additions, the cobalt soil pollution resulted in an increase in the concentrations of cobalt, lead, chromium, nickel and zinc and in a decrease in the content of cadmium and manganese in soil.

All the neutralizing substances caused some decrease in the content of cobalt, manganese and iron in soil.

The highest decrease in the cobalt content in soil was observed after addition of manure, whereas the highest decrease in zinc occurred in the pots with charcoal added to soil. The decrease in the content of the other metals (except nickel and lead) was verified in the pots with addition of CaO and zeolite.

The influence of the other neutralizing substances depended on the type of an element. Reverse relationships were noticed regarding some trace elements, for example the content of lead in soil increased after application of all neutralizing substances.

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