
CHANGES IN THE CONTENT OF SOME MICRONUTRIENTS IN SOIL CONTAMINATED WITH HEATING OIL AFTER THE APPLICATION OF DIFFERENT SUBSTANCES*

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Abstract

Petroleum substances enter the environment, where they cause its degradation. Contamination has an adverse effect on the protective function of soil and soil physicochemical properties, leading to disorders in the soil enzymatic activity, depressing soil fertility and impairing the growth, development and chemical composition of plants. We should therefore look for methods which will enable us to reduce the effect of petroleum substances or even to recover the original properties of soil. The objective of the present study has been to determine the effect of substances applied to soil on the content of cadmium, lead, chromium and manganese in soil contaminated with incremental doses of heating oil (0-20 g kg⁻¹ of soil). The effect of incremental doses of heating oil on the content of analyzed trace metals in soil was varied and depended on the application of alleviating substances to soil. The increasing levels of heating oil contributed to a decrease in the content of cadmium and chromium in soil, but had no such regular influence on the content of lead or manganese. None of the applied alleviating substances had a reducing effect on the content of manganese, cadmium, chromium or lead in the tested soil. Zeolite was an exception in that it reduced the content of cadmium and lead in soil. The content of heavy metals in soil, even when contaminated with the highest doses of heating oil, did not exceed the permissible levels defined in the Regulation of the Ministry of the Environment of 9 September 2002 and therefore allowed us to classify the soil as a zero contamination degree according to the IUNG Puławy classification.

Keywords: heating oil contamination, nitrogen, compost, bentonite, zeolite, calcium oxide, soil, trace elements.

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ZMIANY ZAWARTOŚCI NIEKTÓRYCH MIKROPIERWIĄSTKÓW W GLEBIE ZANIECZYSZCZONEJ OLEJEM OPAŁOWYM PO APLIKACJI RÓŻNYCH SUBSTANCJI

Abstrakt

Substancje ropopochodne przedostają się do środowiska, powodując jego degradację. Zanieczyszczenia wpływają negatywnie na funkcję ochronną gleby i jej właściwości fizykochemiczne, zakłócają jej aktywność enzymatyczną, zmniejszają żyzność oraz oddziałują na wzrost, rozwój i skład chemiczny roślin. Należy zatem poszukiwać metod, które umożliwią ograniczenie wpływu substancji ropopochodnych lub nawet odtworzenie pierwotnych właściwości gleb. Celem badań było określenie wpływu zaaplikowanych do gleby substancji na zawartość kadmu, ołowiu, chromu i manganu w glebie zanieczyszczonej wzrastającymi dawkami oleju opałowego (0-20 g kg⁻¹ gleby). Wpływ wzrastających dawek oleju opałowego na zawartość badanych pierwiastków śladowych w glebie był zróżnicowany i zależał od aplikacji substancji łagodzących do gleby. Wzrastające dawki oleju opałowego przyczyniły się do obniżenia zawartości kadmu i chromu w glebie oraz nie miały ukierunkowanego wpływu na zawartość ołowiu i manganu. Żadna z zastosowanych substancji łagodzących nie miała ograniczającego wpływu na zawartość manganu, kadmu, chromu i ołowiu w badanej glebie. Wyjątkiem był zeolit, który spowodował zmniejszenie zawartości kadmu i ołowiu w glebie. Zawartość metali ciężkich w glebie, nawet zanieczyszczonej najwyższymi dawkami oleju opałowego, nie przekraczała dopuszczalnych norm zawartych w Rozporządzeniu Ministra Środowiska z dnia 9 września 2002 r. oraz pozwoliła na ich zaklasyfikowanie do 0 stopnia zanieczyszczenia gleb wg wytycznych IUNG w Puławach.

Słowa kluczowe: zanieczyszczenie olejem opałowym, azot, kompost, bentonit, zeolit, tlenek wapnia, gleba, pierwiastki śladowe.

INTRODUCTION

Heating oil is produced by distillation and used for heating residential houses, greenhouses and small production plants. Some spillage can occur during the recovery, transport, storage and finally consumption of heating oil (NADIM et al. 2000, SYRYGAŁA, ŚLIWKA 2004). As a result, petroleum substances enter the environment, causing its degradation (SURYGAŁA 2006). As the environmental contamination with heating oil is becoming a pressing problem, inexpensive, quick and effective solutions are being searched for to clean the soil and water environment of the contaminant (SURYGAŁA 2006). Contamination has an adverse effect on the protective function of soil and soil physicochemical properties, causing disorders in the soil enzymatic activity (WYSZKOWSKA, WYSZKOWSKI 2010, ZIÓLKOWSKA, WYSZKOWSKI 2010), depressing soil fertility (WYSZKOWSKI, ZIÓLKOWSKA 2007, 2008, 2009c, WYSZKOWSKI, SIVITSKAYA 2012, 2013) and impairing the growth, development and chemical composition of plants (WYSZKOWSKI et al. 2004, WYSZKOWSKA, WYSZKOWSKI 2006, WYSZKOWSKI, ZIÓLKOWSKA 2009ab, 2011, ZIÓLKOWSKA, WYSZKOWSKI 2010). We should therefore look for methods which - under *in situ* conditions - will enable us to reduce the effect of petroleum substances or even to recover the original properties of soil.

The objective of the present study has been to determine the effect of substances applied to soil on the content of cadmium, lead, chromium and manganese in soil contaminated with incremental doses of heating oil.

MATERIAL AND METHODS

The experiment was carried out in a greenhouse at the University of Warmia and Mazury in Olsztyn (north-eastern Poland), on soil whose texture corresponded to loamy sand (fractions in mm: <0.002 – 1.89%, 0.002-0.005 – 2.46%, 0.005-0.010 – 2.87%, 0.010-0.020 – 4.39%, 0.020-0.050 – 8.89%, 0.050-0.100 – 14.08%, 0.100-0.250 – 36.90%, 0.250-0.500 – 22.78%, 0.500-1.000 – 5.74%, 1.000-2.000 – 0%) and which had the following properties: pH in 1 mol KCl dm⁻³ – 4.52, hydrolytic acidity (Hh) – 25.4 mmol(+) kg⁻¹, sum of the exchange capacity of cations Ca⁺⁺, Mg⁺⁺, K⁺ and Na⁺ (S) – 29.1 mmol(+) kg⁻¹, total exchangeable bases (T) – 54.5 mmol(+) kg⁻¹, percent base saturation (V) – 53%, C_{org.} content – 11.3 g kg⁻¹, the content of available forms of phosphorus – 71.9 mg kg⁻¹, potassium – 118.6 mg kg⁻¹ and magnesium – 104.2 mg kg⁻¹ of soil, and the content of total forms of copper – 3.66 mg kg⁻¹, zinc – 14.86 mg kg⁻¹, nickel – 5.08 mg kg⁻¹ and iron – 6.07 g kg⁻¹. The experiment was conducted in three replications, comprising six series: without alleviating substances, with nitrogen as urea (200 mg N kg⁻¹ of soil), compost (3%), bentonite and zeolite (2% in relation to the soil mass) and 50% calcium oxide in a dose corresponding to one full hydrolytic acidity (10.93 g Ca kg⁻¹ of soil). In each series, soil was contaminated with heating oil in doses of 0, 5, 10, 15 and 20 g kg⁻¹ of soil. Additionally, mineral fertilizers were added to each treatment in the following amounts: 100 mg N, 30 mg P, 100 mg K, 50 mg Mg, 0.33 mg B, 5 mg Mn and 5 mg Mo per kg of soil. Nine-kilogram batches of the soil, carefully mixed with heating oil, alleviating substances and supplementary mineral fertilizers, were placed in polyethylene pots. The test crop was cv. Reduta maize (*Zea mays* L.), grown as 8 plants per pot. During the experiment, the soil moisture was maintained at 60% water saturation.

During the harvest, soil samples were taken. They were dried, ground and submitted to flame atomic absorption spectrophotometric assays (FAAS) in order to determine their content of cadmium, lead, chromium and manganese. The equipment was calibrated using certified standard solutions by Fluke and Merc. Soil was wet mineralized in Teflon HP500 vessels placed in a MARS 5 type oven, according to the US-EPA3051 methodology (1994). The soil samples underwent the following determinations: soil reaction (pH) with the potentiometric method in KCl aqueous solution of the concentration of 1 mol dm⁻³ (LITYŃSKI et al. 1976), hydrolytic acidity (Hh) and sum of the exchange capacity of cations (S) with the Kappen method (LITYŃSKI et al.

1976). Based on the hydrolytic acidity (Hh) and the exchange capacity of cations (S), the total exchangeable bases (T) and percent of base saturation (V) were calculated from the following formulas: $T = Hh + S$, $V = S T^{-1} 100$ (LITYŃSKI et al. 1976). Some soil determinations were completed before the experiment, namely the content of organic carbon (C_{org}) with the Tiurin method (LITYŃSKI et al. 1976), available phosphorus and potassium with the Egner-Riehm method (LITYŃSKI et al. 1976) and available magnesium with the Schachtschabel method (LITYŃSKI et al. 1976) as well as the granular composition of the soil with the laser method using a Mastersizer 2000 meter. The results were analysed statistically using a three-factor analysis of variance ANOVA from Statistica software (STATSOFT, INC. 2010).

RESULTS AND DISCUSSION

The increasing doses of heating oil caused different effects on the content of the analysed macronutrients in soil (Table 1). In the treatments without any alleviating substance, a significant decrease in the content of cadmium ($r=-0.865$) and chromium ($r=-0.855$) appeared in soil contaminated with heating oil (versus the treatment without this petroleum substance), with cadmium being depressed more strongly than chromium. Despite the reducing effect of heating oil on the content of chromium, its lowest dose (5 g kg^{-1}) raised this element in soil by 14%. The effect of heating oil on the content of lead and manganese in soil was not so evidently negative, and modified the content of manganese by less than 10%. However, it should be added that the highest dose of heating oil in soil (20 g kg^{-1}) caused a high increase in the content of lead.

Heavy metals contribute to a decrease in the biomass of microorganisms and inhibit their activity in soil; even when they do not diminish microbial counts, they depress the biological diversity of soil microorganisms (WYSZKOWSKA et al. 2008). The concentrations of the analysed trace elements in soil contaminated with heating oil presented in this paper are within the tolerance ranges for toxic elements worked out by the Institute of Soil Science and Plant Cultivation (IUNG) in Puławy, and the degree of soil contamination with cadmium and lead was low enough to classify the soil as zero contamination degree soil. According to the Regulation of the Ministry of the Environment of 9 September 2002 on soil quality standards and earth quality standards in soils contaminated with heating oil, the determined content of the trace elements did not exceed the permissible thresholds set up for any of the three groups of land.

The alleviating substances applied to non-contaminated soil did not cause any significant changes in the content of manganese in soil, but all of them, except nitrogen, raised the level of chromium (Table 1). Calcium oxide

Table 1

Content of trace elements in soil after plants harvest (mg kg⁻¹ d.m. of soil)

| Dose of heating oil (g kg ⁻¹ of soil) | Type of substance neutralizing the effect of heating oil | | | | | | |
|--|--|----------|----------|-----------|---------|---------|---------|
| | without additions | nitrogen | compost | bentonite | zeolite | CaO | average |
| Manganese | | | | | | | |
| 0 | 212.5 | 220.6 | 214.8 | 231.3 | 207.7 | 218.7 | 217.6 |
| 5 | 223.4 | 211.8 | 229.6 | 225.9 | 235.8 | 215.5 | 223.7 |
| 10 | 206.0 | 235.8 | 212.6 | 225.1 | 208.6 | 213.2 | 216.9 |
| 15 | 227.7 | 228.1 | 209.9 | 228.9 | 209.2 | 221.4 | 220.9 |
| 20 | 204.6 | 232.8 | 210.0 | 214.2 | 213.0 | 224.7 | 216.5 |
| Average | 214.8 | 225.8 | 215.4 | 225.1 | 214.9 | 218.7 | 219.1 |
| <i>r</i> | -0.176 | 0.662* | -0.564 | -0.752** | -0.215 | 0.616* | -0.255 |
| LSD | <i>a</i> - n.s., <i>b</i> - n.s., <i>a b</i> - n.s. | | | | | | |
| Chromium | | | | | | | |
| 0 | 2.49 | 2.32 | 3.55 | 3.45 | 3.89 | 4.74 | 3.41 |
| 5 | 2.83 | 1.98 | 3.58 | 3.52 | 4.23 | 4.54 | 3.45 |
| 10 | 2.29 | 2.80 | 3.21 | 3.58 | 4.47 | 5.43 | 3.63 |
| 15 | 1.95 | 2.87 | 3.69 | 4.03 | 4.40 | 4.98 | 3.65 |
| 20 | 1.84 | 2.87 | 3.65 | 4.20 | 4.33 | 5.12 | 3.67 |
| Average | 2.28 | 2.57 | 3.54 | 3.75 | 4.27 | 4.96 | 3.56 |
| <i>r</i> | -0.855** | 0.783** | 0.254 | 0.947** | 0.735** | 0.553 | 0.931** |
| LSD | <i>a</i> - n.s., <i>b</i> - 0.28**, <i>a b</i> - 0.63* | | | | | | |
| Cadmium | | | | | | | |
| 0 | 0.31 | 0.06 | 0.29 | 0.22 | 0.04 | 0.00 | 0.15 |
| 5 | 0.17 | 0.22 | 0.29 | 0.16 | 0.01 | 0.07 | 0.15 |
| 10 | 0.05 | 0.23 | 0.20 | 0.20 | 0.06 | 0.07 | 0.14 |
| 15 | 0.10 | 0.20 | 0.18 | 0.30 | 0.04 | 0.11 | 0.16 |
| 20 | 0.05 | 0.17 | 0.21 | 0.20 | 0.04 | 0.12 | 0.13 |
| Average | 0.14 | 0.18 | 0.23 | 0.22 | 0.04 | 0.07 | 0.15 |
| <i>r</i> | -0.865** | 0.461 | -0.791** | 0.280 | 0.258 | 0.934** | -0.586 |
| LSD | <i>a</i> - n.s., <i>b</i> - 0.06**, <i>a b</i> - 0.13* | | | | | | |
| Lead | | | | | | | |
| 0 | 4.81 | 5.20 | 5.48 | 7.51 | 6.78 | 4.00 | 5.63 |
| 5 | 5.63 | 10.18 | 17.04 | 6.97 | 4.03 | 8.90 | 8.79 |
| 10 | 2.65 | 9.23 | 15.32 | 14.09 | 3.22 | 9.54 | 9.01 |
| 15 | 4.10 | 9.88 | 11.66 | 5.20 | 4.52 | 10.33 | 7.62 |
| 20 | 12.94 | 11.77 | 12.44 | 8.33 | 5.11 | 5.39 | 9.33 |
| Average | 6.03 | 9.25 | 12.39 | 8.42 | 4.73 | 7.63 | 8.08 |
| <i>r</i> | 0.580 | 0.829** | 0.305 | -0.006 | -0.337 | 0.240 | 0.651* |
| LSD | <i>a</i> - n.s., <i>b</i> - 4.17*, <i>a b</i> - n.s. | | | | | | |

LSD (least squares deviation) for: *a* – heating oil dose, *b* – kind of neutralizing substance, *a b* – interaction; significant for: * $-P = 0.05$, ** $-P = 0.01$, n.s. – non-significant; *r* – correlation coefficient

was the most stimulating soil amendment as its application nearly doubled the content of chromium in soil under maize. The influence of the neutralizing substances on the content of cadmium in soil was strongly reducing. The content of cadmium fell to almost zero in the treatment with calcium oxide; the cadmium level was five-fold lower when nitrogen had been applied to soil and eight-fold lower under the influence of zeolite in soil compared to the non-amended soil. In the soils not contaminated with heating oil, it was only calcium oxide that reduced the content of lead, whereas all the other substances contributed to an increase in the content of this element. In soil under maize, bentonite and zeolite particularly stimulated the increasing levels of lead.

All the applied substances contributed to an increase in the average content of cadmium, chromium and lead in the soil contaminated with heating oil compared to the series without soil amendments (Figure 1). Zeolite and calcium oxide acted differently. Zeolite depressed the content of cadmium in soil by two-fold, while calcium oxide did not cause any changes in the soil content of this element. In that soil, nitrogen, compost and bentonite doubled the content of cadmium. Zeolite also reduced the content of lead in soil by 33% on average. The effect of the other substances on lead was stimulating, increasing its content in soil. Nitrogen raised the content of lead in soil by 62%, compost by 123%, bentonite by 37% and calcium oxide by 35%. The biggest changes in the soil content of trace elements were demonstrated with respect to chromium. The content of this element doubled in the soil amended with calcium oxide. Nitrogen and compost raised its soil content by 18 and 59%. Bentonite and zeolite raised its soil content by an average of 72 and 95%, respectively. The content of manganese in soil did not change much under the influence of the tested substances compared to the series without amending substances, except nitrogen (+5%) and bentonite (+4%), which slightly stimulated the increase in the content of manganese relative to the series without soil amendments.

FILIPEK-MAZUR (2004) claims that while it is impossible to remove heavy metals from soil completely, it is possible to alleviate negative effects of their excessive amounts by using chemical methods: liming, application of organic matter, zeolites or phosphates; biological methods, such as phytoremediation, stimulation of crop yields and dilution of elements in yields, selection of cultures tolerant to the uptake of metals; and physical methods, e.g. deep extraction and electrokinetic techniques.

BARAN et al. (2009) observed that the content of lead in soil increases as the amounts of sewage sludge and rockwool added to soil for rehabilitation increased. In the authors' own research, the content of trace elements increased as the dose of heating oil introduced to soil rose. Cadmium, a component of diesel oil, is characterized by high mobility, especially in light and acid soils (BUCZKOWSKI et al. 2002). Lead belongs to the so-called 'death metals'. The metals introduced to soil with fertilizers do not usually pose such

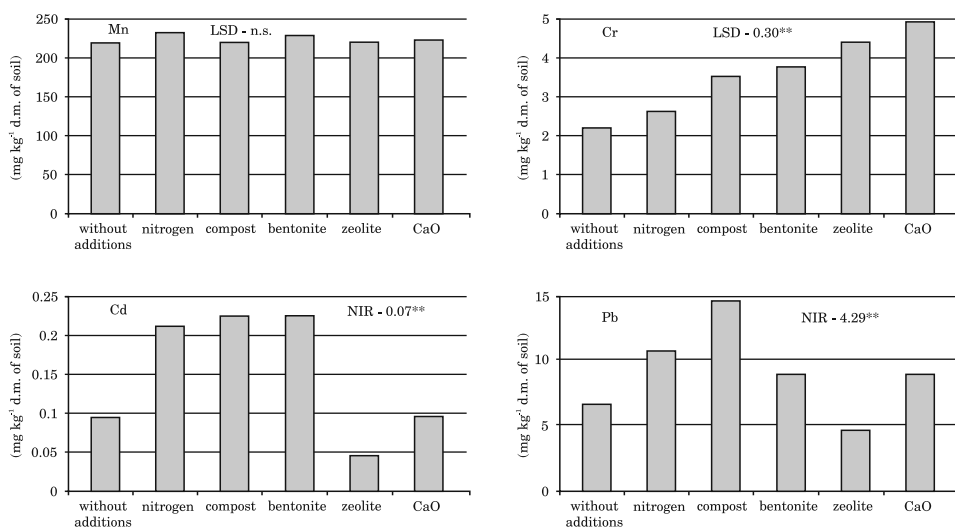


Fig. 1. Content of trace elements in soil after plants harvest (average from series contaminated with 5-20 g heating oil per kg of soil) differences significant for: ** $-P=0.01$, * $-P=0.05$

a severe threat (BARAN et al. 2009). Yet, an elevated concentration of heavy metals in topmost layers of soil make it chemically degraded (TERELAK et al. 2002). Heavy metals belong to the most persistent and slowly migrating soil contaminants.

Organic fertilizers add large amounts of organic matter to soil, have a positive influence on its pH and improve soil absorbing properties (BUCZKOWSKI et al. 2002). Organic matter can form strong bonds with chromium, iron, lead, nickel and cobalt ions, as well as weaker bonds with manganese and zinc ions in soil. Clay minerals are characterized by strong affinity to iron ions, but a weaker one to nickel and cobalt ions, and their oxides form strong bonds with chromium and lead ions (KABATA-PENDIAS, PENDIAS 2001).

Heavy metals are the elements which do not undergo biodegradation to simple compounds. Sorption does not cleanse the soil but causes further accumulation of metals. Minerals which have good adsorption properties improve the soil pH or form insoluble compounds. Zeolite is characterized by good affinity to Cd^{2+} , Pb^{2+} , Zn^{2+} and Cu^{2+} ions, but complete desorption of cadmium occurs at low soil pH, which may explain why zeolite introduced to acid soil under maize had no effect on the content of cadmium, chromium and lead in soil. In order to decrease the content of lead from 500 to 5 mg kg⁻¹ in sandy soil, 5% of zeolite per soil mass should be added; when the contamination reaches 5000 mg Pb kg⁻¹, the required amount of zeolite equals 35% of the soil mass. In fact, much worse sorption of nickel and cadmium by zeolite has been recorded (BUCZKOWSKI et al. 2002). When zeolites are added to soil, the available forms of cadmium are converted into unavailable ones, which diminishes the content of this element in soil (CHEN

et al. 2000). USMAN et al. (2005) achieved positive effects of applying zeolite to soil, as the concentrations of metals in the soil solution fell by 13 to 57%. USMAN et al. (2005) suggest that bentonites are more effective than zeolites in immobilization of metals, which may be attributed to the high content of montmorillonite in bentonites; in zeolites, this mineral makes up just 10% of the total content. In the authors' own investigation, only zeolite had a positive, reducing effect on the content of some of the trace elements in soil.

The soil reaction affects the mobility of heavy metals. All trace metals can form insoluble oxides, hydroxides, carbonates and phosphates only in alkaline environment. Below pH=6, even iron and manganese hydroxides become soluble, which leads to the release of metals adsorbed on their surface (BUCZKOWSKI et al. 2002). KABATA-PENDIAS and SADURSKI (2003) report that soil liming depressed the mobility of metals by rising the soil pH, which favours formation of metal hydroxides and carbonates. In soil of higher pH, bonds between metals and humic acids become stronger. In soils of neutral or alkaline reaction, as well as in the presence of calcium carbonate, metals are slowly transferred to deeper soil horizons KABATA-PENDIAS and PENDIAS (2001).

CONCLUSIONS

1. The effect of incremental doses of heating oil on the content of analyzed trace metals in soil was varied and depended on the application of alleviating substances to soil.

2. The increasing amounts of heating oil contributed to a decrease in the content of cadmium and chromium in soil, but had no such regular influence on the content of lead or manganese.

3. None of the applied alleviating substances had a reducing effect on the content of manganese, cadmium, chromium or lead in the tested soil. Zeolite was an exception as it reduced the content of cadmium and lead in soil.

4. The content of heavy metals in soil, even when contaminated with the highest levels of heating oil, did not exceed the permissible amounts defined in the Regulation of the Ministry of the Environment of 9 September 2002 and therefore allowed us to classify the soil as a zero contamination degree according to the IUNG Puławy classification.

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