USING COMPOST, ZEOLITE AND CALCIUM OXIDE TO LIMIT THE EFFECT OF CHROMIUM (III) AND (VI) ON THE CONTENT OF TRACE ELEMENTS IN PLANTS

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Abstract

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The research aimed to determine the influence of Cr(III) and Cr(VI) in doses of 0 (control), 25, 50, 100 and 150 mg Cr kg⁻¹ of soil as well as compost, zeolite and CaO additives on the content of heavy metals in the spring oilseed rape (Brassica napus var. oleifera) and yellow lupine (Lupinus luteus L.). The contents of heavy metals (Cr, Zn, Ni, Cu) in plants, were determined using the method of spectrophotometry. All of the investigated element contents in the tested plants differed significantly in the case of applying amendments to the soil, as well as increasing concentrations of Cr(III) and Cr(VI). In the series without additives in soils containing Cr(III) and Cr(VI), rapeseed was characterized by a higher average content of Cr than yellow lupine. Compost most noticeably limited the negative effect of chromium. Cr(VI) modified the contents of trace elements in the tested plants more significantly than Cr(III).

Keywords: chromium contamination, heavy metals, compost, zeolite, calcium oxide

INTRODUCTION

The observed changes in the chemical composition of soil and plants are the consequence of the contamination of individual components of the natural environment by progressing and degrading anthropogenic activity (Adamcova et al. 2016; Sas et al. 2015). Changes in the chemical properties of soil have a direct effect on plants, indirectly affecting humans and animals. Heavy metals can be included among the main factors influencing the contamination of the natural environment (Alvarez et al. 2017).

Chromium is a highly reactive element; at both levels of oxidations (III and VI), it creates numerous complexes with water particles as well as organic compounds, e.g. [Cr(NH₃)₆−n−m(H₂O)ₙRₘ⁺](3−m)+ (Liu et al. 2016). The toxicity of Cr(III) compounds is approximately one hundred times lower than that of Cr(VI), with the mutagenic effect being a characteristic of Cr(VI) compounds (Lukina et al. 2016). Cr(III) occurring in a contaminated natural environment is insoluble, and characterized by a low level of mobility; however, in the presence of organic ligands, the complexation of Cr(III) takes place, which increases its solubility, thus influencing its higher mobility and availability to living organisms (Yilmaz, Soylak 2016). Cr(VI) is considered to be more available due to its solubility, strong oxidizing properties and permeability through cellular membranes (Hu et al. 2016).

Basic natural reducers of Cr(VI) include many organic compounds, reduced sulfur compounds and Fe(II) compounds, which play the most important role. Their source in soils and natural waters are minerals, such as: biotite [K(Mg,Fe,Mn)₃(OH,F)₂AlSi₃O₁₀], hematite [Fe₂O₃],...
magnetite [Fe₃O₄], siderite [FeCO₃] and pyrite [FeS₂] (Zhu et al. 2016). The level of toxicity of tri- and hexavalent chromium ions on microorganisms is greatly dependant on the pH level of the soil; in acidic soils, ions of trivalent chromium have a strong toxic effect, whereas in neutral soils – hexavalent chromium ions (Huang et al. 2009). The toxic effect of chromium on plants is connected not only with the level of its oxidation but also depends on the type of compounds present in the natural environment. The same content of chromium in the compounds Cr₂O₇ and Cr(SO₄)₃ results in the compound in the first case being very toxic to plants, whereas the second does not cause damage (Kabata-Pendias, Pendias 2011).

Chromium compounds are essential to the proper functioning of the organisms of mammals; their role in the metabolism of lipids and cholesterol has been documented. Chromium is found in the composition of the so called Glucose Tolerance Factor (GFT), essential for the proper metabolism of glucose and aiding insulin activity (Guimarães et al. 2016). Human and animal organisms take up chromium mostly with food and water, and from the atmosphere, as well as through the skin. Regardless of the means of absorption, Cr(VI) is absorbed more efficiently as compared to Cr(III) because chromium Cr(III) compounds are very poorly absorbed from the intestines, whereas the absorption of Cr(VI) is significantly higher (Mathebula et al. 2017).

Cr(VI) is listed among the priority pollutants, therefore the remediation of soil by chromium contamination is very important for the scientific community (US EPA 1998). Furthermore, the effects of chromium in the +3 and +6 oxidation state vary so much, that each of these compounds should be analyzed separately. Taking into the above into account, research aimed at determining the influence of zeolite, compost and CaO on limiting the negative influence of Cr(III) and Cr(VI) compounds on spring rapeseed and yellow lupine was assumed. The contents of chromium, copper, zinc and nickel as well as the crop yield of above-ground parts of the tested plants were determined in the plant matter.

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<td>Nickel</td>
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1: Pot experiment from the plant growth facility of the University of Warmia and Mazury in Olsztyn.
MATERIALS AND METHODS

The pot experiment was carried out in the plant growth facility of the University of Warmia and Mazury in Olsztyn (Fig. 1). The soil placed in polyethylene pots with a volume of 9.5 kg, with a granulometric content of loamy sand: (fractions in mm: <0.002−1.27 %; 0.002 − 0.005 − 1.95 %; 0.005 − 0.010−2.74 %; 0.010 − 0.020 − 4.52 %; 0.020 − 0.050 − 9.37 %; 0.050 − 0.100 − 8.36 %; 0.100 − 0.250 − 30.55 %; 0.250 − 0.500 − 31.20 %; 0.500 – 1.000 – 9.93 %; 1.000 – 2.000 – 0.11 %), was characterized by properties presented in Tab. I.

The soil was artificially contaminated with water solutions of Cr(III) in the form of KCr(SO 4)2·12H2O and Cr(VI) in the form of K 2Cr2O7 in the following doses: 25, 50, 100 and 150 mg .kg−1. The experiment was carried out in four series: without additives and with the application of compost, natural zeolite and CaO. Each treatment was replicated fourfold. Compost and zeolite were applied in the amount of 3 % of the mass of the soil in the pot, while CaO in an amount equivalent to 1 hydrolytic acidity (HAC). Some of the values of chemical additives have been provided in Tab. II. The control objects to which alleviating substances had not been added and to which Cr(III) and Cr(VI) compounds had not been introduced into the soil were also accounted for. Moreover, aqueous solutions of macro- and microelements in the following amounts in mg.kg−1 soil: N-110, P -50, K -110, Mg -50, M- 5, Mo- 5, B-0.33 were added to each pot. The soil samples were thoroughly mixed and were allowed to stabilize under natural conditions for three weeks before using as a growth experiment.

The experiment covered the farming of spring rapeseed (Brassica napus var. oleifera) of the Hunter variety (main crop) and yellow lupine (Lupinus luteus L.) of the Polo variety (aftercrop). The plants were watered every other day with distilled water to 60 % of the maximum water holding capacity of the soil by adding deionized water. Spring oilseed rape and yellow lupine were grown at respective densities of 25 and 18 plants per pot. The crops were harvested in the flowering phase.

Laboratory and statistical analysis

In the collected plant material, the crop yield of above-ground plant parts was determined, as well as indicating the contents of chromium, copper, zinc, and nickel using the method of flame atomic absorption spectroscopy (FAAS) on an atomic absorption spectrometer on a SpectrAA 240FS type apparatus (VARIAN, Australia) in extracts obtained following the “wet” mineralization of soil in nitric acid (HNO3 p.a.) at a concentration of 1.40 g.cm−1 in a MARS 5 type microwave oven (CEM Corporation, USA), in HP500 teflon dishes. The conditions of the process, such as the sample weight, volume of nitric acid and temperature of mineralization were assumed in accordance with MARS 5 methodology (CEM Corporation, USA). 0.300±0.1 mg of dried and homogenized plant samples was decomposed in the microwave oven using 10 ml HNO3. The plant digests were adjusted to the final volume of 100 ml with deionized water. Deionized water with an electrical conductivity of (0.055 µS.cm−1), purified using the Crystal 10 system (Adrona Laboratory System) was used for the analyses. Certified reference material (Sigma Aldrich Chemie GmbH, No. BCR142R) was used for analyses. The calibration of measurement apparatuses was carried out with the use of certified standard solutions of the Fluka and Merc company. Stock solutions of metals (1000 mg/L) were prepared from their nitrate salts.

The obtained results were subjected to statistical calculations using three-way ANOVA and Duncan’s test. The Statistica 8.0 program was used to carry out the calculations.

RESULTS AND DISCUSSION

Effect of Cr(III) and Cr(VI) on the content of trace elements in plants

The concentration of Cr(III) and Cr(VI) in plants is connected with a set of chemical and geological properties of soil; however, the main role is played by the content of chromium compounds in the soil (Wyszkowski, Radziemska 2010; Radziemska et al. 2016a). This means that the content of Cr in plants is dependent on the content of this element in soils, as well as the species of plant.
In the conducted experiment, the content of Cr in spring rapeseed (main crop) and yellow lupine (aftercrop) was influenced by both the form as well as the dose of chromium, as well as additives in the form of compost, zeolite and CaO (Fig. 2). In the series without additives in the soils containing Cr(III) and Cr(VI), the main plant—spring rapeseed was characterized by a higher average content of Cr than the interplant—yellow lupine. In the control series (without additives), the above-ground parts of spring rapeseed in pots with Cr(VI) were characterized by over three times higher average contents of Cr than in pots with...
Cr(III). The accumulation of Cr in above-ground parts of spring rapeseed was positively correlated with the increasing contamination of the soil with Cr(III) – ($r = 0.743$) and Cr(VI) – ($r = 0.774$), with Cr(VI) exerting a stronger influence on Cr content, causing a nearly eight-fold increase in the amount of Cr as compared to the control sample – without contamination when applied in doses of 100 and 150 mg·kg$^{-1}$.

Chromium compounds are passively taken up by plants, collecting, above all, in their above-ground parts and roots, but are not transported to the seeds and beans. Based on a review of Polish and international literature on the subject, it can be concluded that the natural content of chromium in plants in generally low (Ahemad 2015; Ding et al. 2016; Islam et al. 2016). Cr(III) and Cr(VI) in the series without additives had a smaller influence on Cr content in the aftercrop – yellow lupine. Its content in the above-ground parts of the discussed plant was higher than in the case of samples with Cr(III) than Cr(VI). In the series without alleviating additives, Cr(III) and Cr(VI) in the highest doses (150 mg·kg$^{-1}$ soil) caused a respective over four-fold and two-fold increase in Cr content of yellow lupine as compared to pots without contamination. The above-described results confirm the studies of other authors such as Scoccianti et al. (2016), and Filipek-Mazur and Gondek (2000), who confirmed that Cr(VI) has a strong toxic effect on plants in the main crop. A reduction of crop yield in the case of aftercrop, however, was not confirmed, which can be explained by the reduction of Cr(VI) to Cr(III). Radziemska et al. (2013) report that, along with the increasing contamination of soil with heavy metals, a significant decrease in the amount of minerals taken up by plants is reported. According to Rezajcka et al. (2005), in the grain of wheat, the content of chromium was found to be in the range of 0.002 to 0.183 mg·kg$^{-1}$ d.m. (dry mass), whereas Lavado et al. (2001) determined this content to be the level of 1.77 mg·kg$^{-1}$ d.m.

Increasing doses of Cr(III) and Cr(VI) as well as additives in the form of compost, zeolite and CaO significantly influenced the contents of Cu in the above-ground parts of the tested plants (Fig. 3).

In the series without alleviating additives, the above-ground parts of spring rapeseed and yellow lupine in pots with Cr(III) were characterized by nearly 3-times higher average contents of Cu than in pots with Cr(VI). In the series without additives, the contamination of Cr(VI) had a stronger influence on the content of Cu in the above-ground parts of the main crop (spring rapeseed) than Cr(III). The highest dose of this contamination (150 mg·kg$^{-1}$ soil) caused a nearly 9-fold decrease ($r = -0.70$) in Cu content in the above-ground parts of the analyzed as compared to the control sample (without Cr). Studies by Zheljazkova and Nielsen (1996) found the accumulation of copper in the roots of the analyzed plant to be higher than in leaves and inflorescences. This phenomenon can be explained by the defense mechanisms of plants against the excessive accumulation of copper in their above-ground parts (Ernst et al. 1992). The accumulation of Cu in above-ground parts of yellow lupine (aftercrop) in the series without alleviating additives was negatively correlated with...
increasing doses of Cr(III) \((r = -0.91)\) and Cr(VI) \((r = -0.84)\). In this series, the highest dose of Cr(VI) 150 mg·kg\(^{-1}\) soil resulted in an almost 13-fold decrease in Cu content in the above-ground parts of yellow lupine as compared to the plants in the control pots (without contamination). In an analogical sample with Cr(III), the decrease in copper content was only 15%.

Zinc expresses a high affinity to mineral colloids and is characterized by a low susceptibility to creating complex connections with organic matter (Asensio et al. 2016). It is characterized by high mobility in soil, as well as high assimilability by plants due to the speed at which the complexes in which it occurs dissolve, especially in an acidic environment (Ming et al. 2016). By changing the pH of the soil environment, its assimilability can be limited, e.g. by the addition of calcium fertilizers (Kabata-Pendias 2011). Factors of the experiment, such as: the dose and form of chromium Cr as well as alleviating additives (compost, zeolite, CaO) significantly affected the content of Zn in the above-ground parts of the grown plants (Fig. 4).

In the series without alleviating additives, spring rapeseed grown as the main crop, as well as yellow lupine grown as the aftercrop, were characterized by higher average contents of Zn in the soils with Cr(III) than in Cr(VI). A more targeted influence on the content of Zn in the above-ground parts of the main plant (spring rapeseed) in the series without alleviating additives was exhibited by Cr(VI) than in Cr(III). The accumulation of Zn in the above-ground parts of the discussed plant was negatively correlated with increasing soil contamination with Cr(VI) \((r=-0.98)\), where the highest dose of this contaminant (150 mg·kg\(^{-1}\) soil) resulted in an almost two-fold decrease in the content of Zn as compared to the sample without contamination (control). Cr(III) caused irregular changes in the content of the above component in spring rapeseed in the series without additives. In the case of the aftercrop – yellow lupine, Cr(III) applied in the highest dose – 150 mg·kg\(^{-1}\) soil caused an increase in the content of Zn by 59% \((r = -0.751)\) as compared to the variant free of contamination. In the same study series, the lowest dose of Cr(VI) (25 mg·kg\(^{-1}\) soil) caused an increase in Zn content by 36%, while its following doses decreased its content in relation to the control combination. Along with an increase in soil acidity, Cr(VI) sorption decreases, while the sorption of Cr(III) increases (Kabata-Pendias 2011). As a result of liming soil, the solubility of chromium decreases (Shaheen, Rinklebe 2016), which limits its negative influence on plants. Moreover, liming increases the amount of non-organic chromium compounds (hydroxides and phosphates), and decreases the amount of organic connections of trivalent chromium (Otabbong 1990).

The content of Ni in the above-ground parts of spring rapeseed and yellow lupine was influenced by the dose and type of contamination, as well as compost, zeolite and CaO introduced in order to neutralize the negative effect of chromium on the analyzed plants (Fig. 5). Crops vary in terms of their ability to assimilate nickel, though it is generally easily taken up, and at a level proportional to its concentration in the soil. In the case of limited amounts of nickel in soil, this element is found in plants in complexes with organic compounds,
whereas in excessive amounts, it may occur in plants in a cation form [Ni²⁺] (Radziemska et al. 2016b, Li et al. 2015). In the series without alleviating substances, the average content of Ni in the above-ground parts of spring rapeseed was at a similar level in soils containing Cr(III) and Cr(VI), in contrast to the aftercrop – yellow lupine, where a higher average content of Ni in the plant occurred in pots with Cr(III). The content of Ni in the above-ground parts of spring rapeseed in the series without additives was negatively correlated with increasing contamination of soil with Cr(III) (r = −0.849) and Cr(VI) (r = −0.881). The highest doses of Cr(III) and Cr(VI) (100 and 150 mg·kg⁻¹ soil) had the greatest negative influence on the contents of the discussed element, under the influence of which Ni decreased by 59-63 % as compared to the control combination (without chromium). Cr(VI) in the dose of 150 mg·kg⁻¹ soil caused a decrease in Ni in the above-ground parts of yellow lupine – the aftercrop – by 84 % as compared to the control sample (without contamination). Cr(III) had a similar influence, though lesser, i.e. 52 %, though its first dose caused an increase in Ni content in yellow lupine.

Effect of compost, zeolite and CaO on the content of trace elements

Metals accumulated in soil, including chromium compounds, can easily move to above-ground parts of plants if factors decreasing their assimilability by plants are not used (Wyszkowski, Radziemska 2013a). Studies prove that soil contaminated with Cr(III) and Cr(VI), as well as the applied alleviating additives, significantly affect the chemical composition and crop yield of plants grown in it (Wyszkowski, Radziemska 2010). PH is treated as an indicator of changes in the properties of soil, because under the influence of external factors, it is the first to undergo changes. It influences (directly and indirectly) many mechanisms leading to the mobilization heavy metals, and determines the strength of binds of heavy metals by organic and mineral matter, and their bioavailability to plants (Gusiatin, Kulikowska 2016, Imtiaz et al. 2016; Zhang et al. 2016). In the carried out studies, compost, zeolite and CaO significantly affected the content of Cr in the above-ground parts of plants – spring rapeseed and yellow lupine (Fig. 6). An especially visible effect in the form of a decrease in the average content of Cr in the above-ground parts of the main crop – spring rapeseed, was observed in the samples with Cr(VI) following the use of compost as an additive. In this series, compost caused a 5-fold decrease in Cr content as compared to the control series (without additives). Zeolite and CaO significantly affected the content of Cr in the above-ground parts of the aftercrop – yellow lupine (Fig. 6). An especially visible effect in the form of a decrease in the average content of Cr in the above-ground parts of the aftercrop (yellow lupine) as compared to the series without additives (control). In pots with Cr(III), CaO caused a 64 % decrease in chromium

6: Effect of CaO, compost and zeolite on chromium and copper content in spring rapeseed (Brassica napus L.) and yellow lupine (Lupinus luteus L.) (mg kg⁻¹ dry mass).

LSD for: a – chromium form; b – chromium dose; c – kind of neutralizing substance;
** – significant at p = 0.01; * – significant at p = 0.05; n.s. – not significant
content as compared to the control series. Zeolite had a similar effect, though to a lesser extent, i.e. 47%. The influence of zeolite on Cr content in yellow lupine was not positive in pots with Cr(VI), as it caused an increase in its accumulation in the above-ground parts of the plant. Only CaO was found to have a beneficial limiting effect.

Neutralizing additives in the form of compost, zeolite and calcium oxide significantly influenced Cu content in the above-ground parts of spring rapeseed and yellow lupine (Fig. 6). The accumulation of Cu in the above-ground parts of spring rapeseed grown as the main crop in pots to which Cr(VI) had been added was positively influenced by all introduced additives, especially zeolite and compost. The influence of CaO was somewhat lesser, though also positive. In pots with Cr(III), all three additives, i.e. compost, zeolite and CaO, caused a decrease in the average content of Cu in the above-ground parts of the analyzed plant as compared to the control series (without additives). The addition of CaO to the soil had the strongest limiting effect, causing a decrease in average Cu content by 74 % as compared to plants grown in soils without the addition of alleviating substances (control). In the case of the aftercrop – yellow lupine, the additives functioned in a similar manner, causing a decrease in the average content of Cu in plants grown in pots with Cr(III), while an increase in the analyzed element in pots with Cr(VI) as compared to control samples (without additives). CaO had the strongest limiting effect on Cu content in above-ground parts of yellow lupine in pots with Cr(III), as the content of Cu decreased by 75 % upon its application as compared to the control variant. In pots with Cr(VI), the highest, as much as 152 %, increase in Cu was caused by compost. According to Blaziak et al. (1996), the application of both straw with the addition of nitrogen as well as manure caused an increase in the copper content of grains.

Amongst the alleviating substances used in the experiment, only compost and zeolite in soils with Cr(III), and compost grown in pots with Cr(VI) increased Zn content in the above-ground parts of spring rapeseed as compared to the series without additives – control. CaO caused a decrease in Zn content in above-ground parts of the above plant in pots with Cr(III) by 30 %, and by 39 % in their Cr(VI) counterparts as compared to the control series (Fig. 7). Zeolite had a similar effect (~15%), but only in pots contaminated with Cr(VI). The aftercrop – yellow lupine, contained the most Zn in its mass after applying compost to the soil, both in pots with Cr(III) and with Cr(VI), as compared to the control series. CaO, and to a small extent zeolite, influenced a decrease in the content of Zn in the above-ground parts of the above plant by 38 % and 5 % respectively in pots with Cr(III) and 35 % and 4 % respectively in pots with Cr(VI). In another experiment carried out by Radziemska et al. (2013), the addition of mineral-based amendment caused a significant decrease in the nickel content of maize (Zea Mays L.).
CONCLUSION

The content of trace elements (Cr, Ni, Zn, and Cu) in spring rapeseed and yellow lupine depended on the dose of the Cr(III) and Cr(VI) contaminant and the application of alleviating substances incorporated into the soil. The more severely the soil was contaminated with Cr compounds, the higher the content of Cr(III) and Cr(VI) in test plants became. In the control series (without additives), the main crop – spring rapeseed – was characterized by a higher average content of Cr in pots with Cr(III) and Cr(VI), than the aftercrop – yellow lupine. Compost most significantly counteracted the negative effects of chromium, causing a significant decrease of its content in the above-ground parts of both tested plants. The influence of CaO and zeolite, although often beneficial, was smaller than that of compost. Cr(VI) modified the contents of trace elements in plants more strongly than Cr(III). Calcium oxide, more than zeolite and compost, limited the content of zinc Zn in the tested plant species. The substances applied to the soil tended to positively influence copper Cu content in plants. Of the applied substances, compost had the strongest effect on the average content of Ni in the above-ground parts of spring rapeseed. However, despite the promising results, the immobilization effects of amendments (zeolite, compost, calcium oxide) in polluted soils with Cr(III) and Cr(VI) need to be further assessed in the future, especially the case of soil contaminants under field conditions.

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