



Study of applying naturally occurring mineral sorbents of Poland (dolomite halloysite, chalcedonite) for aided phytostabilization of soil polluted with heavy metals



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ARTICLE INFO

Keywords:

Aided phytostabilization
Metal-contaminated soil
Soil reclamation
Risk minimization

ABSTRACT

A greenhouse experiment was carried out for evaluating the effects of mineral sorbents, i.e.: dolomite, halloysite, and chalcedonite on the chemical characteristics of soil contaminated with heavy metals and the uptake of metals by plants. The contents of trace elements in the plants and soil were determined using the method of spectrophotometry. All of the investigated element contents in the tested parts of *F. rubra* differed significantly in the case of applying mineral sorbents to the soil, as well as increasing concentrations of Pb, Cd and Zn. The greatest average above-ground biomass was observed when chalcedonite and halloysite were amended into the soil. Halloysite and chalcedonite also caused significant increases in Pb, Cd and Zn concentrations in the roots. The addition of dolomite significantly increased soil pH. Halloysite and chalcedonite were shown to be the most effective and decreased the average Pb, Cd and Zn contents in soil.

1. Introduction

The contamination of all components of the natural environment has been increasing as a result of progressing urbanization, excessive exploitation of the environment and the continuously increasing population. Solid, liquid and gas contaminants can negatively influence the condition of the soil-water environment, as well as directly and indirectly influencing the development of living organisms (Bojko and Kabala, 2016; Duan et al., 2015; Gworek et al., 2016; Radziemska and Fronczyk, 2015; Sas et al., 2015). In many cases, their negative effects on the environment are of an increasingly globalized character. However, in many cases, their influence is also observed at the local level. The contamination of the natural environment with heavy metals, which can be derived from a wide range of anthropogenic activities, is a particularly important issue (Adamcová et al., 2017). Pressure exerted on the environment and the contamination connected with it, have a real and continuously growing influence on the quality of life.

The progressing degradation of the natural environment, along with an increase in the ecological awareness of society have an influence on the increasing role of the remediation of contaminated areas (Sarwar et al., 2017; Montpetit and Lachapelle, 2017). Returning ecosystems to as close to their natural state as possible appears to be a key task. Intensive enhancement of effective and non-invasive methods of cleaning degraded lands is essential. The rate of innovation and dissemination of new solutions plays a key role in this case. For years, methods used in

order to remediate soils contaminated with heavy metals have been based on one of two strategies: the immobilization of metals in soil or their mobilization and removal from soil (Zhang et al., 2017). Treatments of metal immobilization should be applied especially where there is a need for temporarily restricting ecological risk, e.g. if the removal of excessive metals from soil cannot be applied in a short time interval. The effect of decreasing the mobility of metals can be obtained by introducing organic materials characterized by high sorption capacity to the soil (compost, peat, lignite, saw dust, tree bark), additives rich in clay minerals (bentonite, halloysite, kaolinite, illite, goethite, ferrihydrite, zeolite), materials containing phosphates (apatite, phosphorite) (Molla et al., 2017; Mlekodaj et al., 2014; Ferreira et al., 2015; Radziemska et al., 2016; Gusiatin et al., 2016). Moreover, in order to limit the effects of soil erosion and reduce the spread of contaminants into groundwater, a dense vegetation cover of the soil surface (phytostabilization), which causes the immobilization of heavy metals in the root zone, can be applied directly on the contaminated area (Paz-Ferreiro et al., 2014).

There exist many species of plants which are capable of growing in areas contaminated even with high concentrations of heavy metals (Nadal-Romero et al., 2016; Vaverkova and Adamcova, 2014). Depending on the specific features of the plant, they can prove useful for a given phytoremediation method. In the case of phytostabilization, these plants ought to be characterized, above all, by low habitat requirements, a low translocation factor, fast growth and a well-developed root

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<https://doi.org/10.1016/j.catena.2017.12.015>

Received 5 April 2017; Received in revised form 26 August 2017; Accepted 14 December 2017
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system (Mendez and Maier, 2008). The role of plants in this technique is, above all, reducing the amount of water percolating through the soil matrix, which makes it possible to avoid the formation of toxic and dangerous leachates, additionally stimulating sorption processes of heavy metals in soil. As a result of the effects of plants on the soil environment, bioavailable forms of heavy metals can change over to less easily available ones. Compounds found in the roots (phytosiderophores, organic acids, phenol compounds) react with metal ions and precipitate them in the form of insoluble salts as well as accumulating metals in roots (Zheng et al., 2016). Next, they bind them in the cell wall and accumulate them within the apoplast as well as in vacuoles. Various species of grass, such as red fescue (*Festuca rubra* L.) are the most useful in the process of the aided phytostabilization of heavy metals in soils (Touceda-Gonzalez et al., 2017).

Phytostabilization may be aided by the application of various soil additives (aided phytostabilization). The influence of additives must be analysed in a wide spectrum, thanks to which it will be possible to assess the effects of individual additives on physical and chemical properties of soil, the mobility of heavy metals, the plant, and the effectiveness of the phytostabilization process (Sylvain et al., 2016; Radziemska et al., 2017). The aim of carrying out such experiments in controlled conditions is the concern for the natural environment, decreasing the risk of groundwater contamination, and the possibility to analyse a series of elements before they are applied in field conditions. The novelty of this study is that we examined new amendments which had not been analysed in this perspective.

Poland is among the countries abundant in minerals which, in their raw state, may be great mineral sorbents, with various applications in many branches of environmental engineering. Dolomite ($\text{CaMg}[\text{CO}_3]_2$) – primary dolomites form as a product of sedimentation from lake and sea waters, while secondary dolomites are created as a product of the impact of sea water, thermal waters and others abundant in Mg^{2+} on limestone rocks and deposits (Zhou et al., 2017). Dolomite rocks are found throughout the entire world, including Poland. They create large geological formations, such as the Alps. In Poland, they are found mainly in Upper and Lower Silesia, near Chrzanowo, Jelenia Góra and Złoty Stok. Dolomite formations in the area of the country were created in various geological periods. The oldest of them – Cambrian dolomites, are found in Rędzin in Upper Silesia. They are used mainly for obtaining building and road stone in the form of slabs and architectural details for works on facades (Bok-Badura et al., 2015; Szybilski and Nocuń-Wczelik, 2015).

Halloysite is a mineral in the silicate group, included among clay minerals and occurring in two varieties of hydrohalloysite ($\text{Al}_4[\text{Si}_4\text{O}_{10}](\text{OH})_8 \cdot 4\text{H}_2\text{O}$) and halloysite ($\text{Al}_4[\text{Si}_4\text{O}_{10}](\text{OH})_8$). Halloysites differ in terms of the size of their specific surface area and porosity, as a result of which there are many potential classes of this mineral which can be used for various purposes. The average specific surface area of halloysite is 56.2–58.0 m^2g^{-1} (Levis and Deasy, 2002; Churchman et al., 1995). Halloysite is mined from natural deposits in Brazil, China, France, Japan, the USA, South Korea, Turkey and Poland (Levis and Deasy, 2002). In Poland, halloysite is obtained from the “Dunino” open-cast mine located in the southwestern part of the country. The mine is currently one of three in operation in the world, alongside the mine in the USA and New Zealand, with its deposits estimated at 10–12 million tons (Banaś et al., 2013).

Chalcedonite is a silicate sedimentary rock whose physical and chemical properties create perspectives for wide application in many fields of environmental protection (Michel, 2012). Chalcedonite has high potential for application in environmental engineering, among others as a substrate for gardening, as a sorbent for removing oil spills or as a filtration filler at the last level of sewage treatment, as a raw material for the production of cement, and a silicate filler for the production of paints and varnishes (Kotwa, 2016). The “Teofilów” chalcedonite deposit in Inowódz is the only documented location of such a mine in Poland. The mine has existed since 1970.

The obtained minerals found wide application in many branches of industry and environmental engineering, however, they have not been analysed as sorption materials for purposes of their application in processes of the aided phytostabilization of heavy metals. In connection with the above, we undertook studies on the possibility of their use in aiding immobilization processes of multi-metal contaminated soil using red fescue (*Festuca rubra* L.).

2. Materials and methods

2.1. Experimental system

The greenhouse experiment was carried out in 5.0 kg plastic pots containing soil contaminated with Pb, Cd and Zn, amended with dolomite, halloysite and chalcedonite, and vegetated with *Festuca rubra* L. plants. The plants were watered every other day with distilled water to 60% of the maximum water holding capacity of the soil. The plants were harvested after 40 days, and soil samples were collected. The soil was collected from the top layer (0–20 cm) from a non-contaminated site in an agricultural area, and was characterized by the following physicochemical properties: pH 4.92; hydrolytic acidity ($\text{mmol}\cdot\text{kg}^{-1}$) 31.21; sum of exchangeable bases Ca^{2+} , Mg^{2+} , K^+ , Na^+ ($\text{mmol}\cdot\text{kg}^{-1}$) 61.10; cation exchange capacity ($\text{mmol}\cdot\text{kg}^{-1}$) 94.20; base saturation (%) 65.20; total N ($\text{g}\cdot\text{kg}^{-1}$) 1.22; organic carbon ($\text{g}\cdot\text{kg}^{-1}$) 7.42; N-NH_4^+ ($\text{mg}\cdot\text{kg}^{-1}$) 20.32; N-NO_3^- ($\text{mg}\cdot\text{kg}^{-1}$) 10.01; extractable P ($\text{mg}\cdot\text{kg}^{-1}$) 43.20; extractable K ($\text{mg}\cdot\text{kg}^{-1}$) 8.72; extractable Mg ($\text{mg}\cdot\text{kg}^{-1}$) 31.2; Pb ($\text{mg}\cdot\text{kg}^{-1}$) 16.38; Cu ($\text{mg}\cdot\text{kg}^{-1}$) 8.20; Ni ($\text{mg}\cdot\text{kg}^{-1}$) 4.10; Zn ($\text{mg}\cdot\text{kg}^{-1}$) 23.22; Mn ($\text{mg}\cdot\text{kg}^{-1}$) 208.3. Prior to the experiment, the soil was air-dried at room temperature and sieved through a 2-mm nylon sieve. Simulated contamination of the soil with lead, cadmium and zinc at three levels, as well as a control object without the addition of heavy metals and mineral sorption materials, were applied in the experiments. Pb, Cd and Zn were introduced in the following doses ($\text{mg}\cdot\text{kg}^{-1}$ of soil): lead 0 (control), 200, 400 and 800; cadmium 0 (control), 5, 10 and 20; and zinc 0 (control), 200, 400 and 600. In all of the experimental groups, identical basic mineral fertilization was applied, adding the following to the soil ($\text{g}\cdot\text{kg}^{-1}$): N-26%, K_2O -26%, B-0.013%, Cu-0.025%, Fe-0.05%, Mn-0.25%, and Mo-0.20%. Three mineral sorbents, i.e. dolomite halloysite, and chalcedonite, were mixed in with the soil in the amount of 3.0% (each). Fig. 1 shows SEM photographs of the materials. Each treatment was replicated thrice. The soil samples were thoroughly mixed and were allowed to stabilize under natural conditions for three weeks before being used for the growth experiment.

2.2. Plant chemical analysis

The harvested shoots and roots were washed with ultrapure water, air-dried at room temperature for two weeks, and then ground to powders using an analytical mill (Retsch type ZM 300, Hann, Germany). The roots and shoots were oven-dried at 55 °C to a stable weight, with the dry biomass recorded. The samples were stored at 4 °C, protected from light in clean containers for subsequent chemical analysis. A representative subsample was mineralized in nitric acid (HNO_3 p.a.) with a concentration of 1.40 $\text{g}\cdot\text{cm}^{-1}$ and 30% H_2O_2 using a microwave oven (Milestone Start D, Italy). After filtration, the digestion products were adjusted to 100 mL volume with deionized water. Extracts were analysed for total Pb, Cd and Zn concentrations determined by the Atomic Absorption Spectrometry (AAS) method using an iCE-3000 spectrophotometer (Thermo Scientific, USA). Five-point calibration was performed with standard solutions. Each sample was processed in triplicate.

2.3. Soil analytical methods

PH was measured in water and 0.1 M KCl solution, and electrical

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