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Sequential extraction of nickel and zinc in sewage sludge- or biochar/sewage sludge-amended soil

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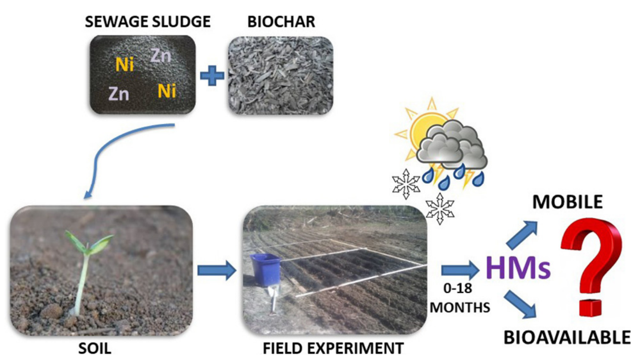
HIGHLIGHTS

- Co-application of biochar with sewage sludge for speciation of Ni and Zn was investigated.
- Application of biochar to sewage sludge decreased mobile fraction of Ni and Zn.
- Biochar can promote the transition of available forms to their residual forms.

GRAPHICAL ABSTRACT



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ABSTRACT

Fractionation of nickel (Ni) and zinc (Zn) in sewage sludge-amended soil and in sewage sludge/biochar-amended soil was investigated. The results were compared with ecotoxicological tests in order to determine the contribution of metals to the toxicity of amended soils. The study was conducted as a long-term field experiment. Sewage sludge (SL) or sewage sludge with a 2.5, 5 or 10% addition of biochar (BC) was added to the soil. Samples for analysis were taken immediately after experiment establishment as well as after 12 and 18 months from the beginning of the study. The fractionation analysis of Ni and Zn was performed using the BCR (Community Bureau of Reference) three-step sequential extraction procedure. The following forms were determined: mobile (F1); bound to Fe—Mn oxides (F2); bound to organic matter (F3) and residual (F4). The soil, SL and BC differed in the contribution of individual forms of the metals. The application of SL into the soil resulted in an increased soil content of mobile forms of Ni and Zn by 180 and 103%, respectively. The mobility index (MI) significantly increased, which evidences the risk related to the presence of these metals. Biochar in the sewage sludge significantly reduced the content of Ni and Zn in F1 fraction. The study also demonstrated that biochar amendment promotes the transition over time of available forms of Ni and Zn into their residual forms (F4), which leads to a further reduction in the environmental risk related to their presence in the environment. The conducted statistical analysis revealed only intermittent relationships between the individual forms of the metals and soil physicochemical properties and toxicity, which may indicate more complex mechanisms that occur in the experimental systems investigated. Therefore, the use of SL in combination with BC can be an effective method for reducing the environmental risk related to the presence of metals in SL.

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1. Introduction

In recent years, in the remediation of heavy metals (HMs) contaminated soils a trend is observed towards reducing HMs mobility by its binding by strong adsorbents (Ahmad et al., 2014; Kumpiene et al., 2008; Mahar et al., 2015). Various adsorbents are used in this method, such as alkaline materials (lime, fly ashes, hydroxyapatites), clay minerals (bentonite and limonite), and organic materials (activated carbon, compost). As a result of adding adsorbents to soils, the mobility and bioavailability of HMs decrease, and thus their bioaccumulation and movement into ground and surface waters is reduced. The studies demonstrated that biochar (Karer et al., 2017; Lahori et al., 2017), which shows strong affinity to heavy metals (Bogusz et al., 2015, 2017; Bogusz and Oleszczuk, 2017), can also be used for HMs immobilization. In the situation when this method performs well for soils, it seems right to apply it to other matrices contaminated with HMs.

Solutions for sewage sludge management have been sought for years, particularly in the context of heavy metal content in sewage sludge (Smith, 1996). HMs content in sewage sludge is one of the criteria that determine its environmental use (CEC, 1986; Dz.U., 2002). Even if the standards for HMs content in sewage sludge, which exist in many countries, are met, its application to soils arouses many controversies. Adding sewage sludge to soils leads to an increased level of HMs in soils (Alloway and Jackson, 1991; Bai et al., 2017; Charlton et al., 2016) and subsequently to their potential accumulation in plants and soil organisms as well as to contamination of ground and surface waters. Additionally, over time changes in sewage sludge composition and soil properties may occur, which affects the chemical forms of HMs determining their mobility and release from sewage sludge.

A certain solution, can be the application of adsorbents together with sewage sludge (Liang et al., 2012; Su and Wong, 2004; Usman et al., 2005). By strongly binding metals with an adsorbent, immobilization of metals in the soil is enhanced and due to this their bioavailability and mobility are reduced. It is important that the added adsorbent should have a long-term effect and moreover it should be neutral to the environment or have a positive impact on the soil. Most studies show (Al-Wabel et al., 2017; Igalavithana et al., 2016) that biochar applied at an appropriate rate improves soil physical, chemical and biological properties, in particular in the case of poor quality soils into which sewage sludge is also incorporated. Thus, combination of sewage sludge and biochar may not only contribute to reduce bioavailability and mobility of HMs, but also to increase soil fertility. Such a solution, that is, combined application of biochar and compost, allows the bioavailability of HMs to be effectively reduced in soils contaminated with these elements (Ali et al., 2017; Karer et al., 2017). Our previous studies also demonstrated that application of sewage sludge with biochar promotes degradation of organic contaminants present in sewage sludge (Stefaniuk et al., 2017), immobilization of organic contaminants (Stefaniuk and Oleszczuk, 2016), and the reduction of the toxicity of sewage sludge-amended soil (Stefaniuk and Oleszczuk, 2016, 2018).

Mobility of HMs, their bioavailability and related ecotoxicity to living organisms largely depend on their specific chemical forms or binding modes. Therefore, estimation of the total content of HMs is insufficient in evaluation of the actual risk related to their presence in the environment. Sequential extraction and fractionation of HMs in environmental matrices are a useful technique for determining chemical forms of HMs. Such information is potentially helpful in predicting the mobility of HMs, their availability to plants or their cycling in the food chain (Sánchez-Martín et al., 2007). In recent years, various methods have been proposed for evaluating different forms of elements. The fractionation protocol starts with the weakest extractant and ends with the strongest and most aggressive one, following the decreasing order of solubility, and it is usually divided into three up to seven fractions. Among different extraction protocols, the BCR (Community Bureau of Reference) three-step sequential extraction procedure is one of the most widely used methods due to its simplicity and due to the fact that results are obtained in a short time, which allows quick risk assessment.

Among HMs mentioned in legislation, relatively the least attention is paid to zinc (Zn) and nickel (Ni) in the context of sewage sludge. Zinc is characterized by low toxicity compared to other HMs, but owing to its much higher concentration in sewage sludge, it may also pose a serious risk to the environment. Similarly to other metals, Zn may adversely affect living organisms, accumulate in plants and soil organisms (Alloway and Jackson, 1991) as well as contaminate ground and surface waters (Bakis and Tuncan, 2011). Fu et al. (2017) assessed that Zn poses a greater ecological threat than other HMs on account of its high content in waters and sediments. On the other hand, Ni is a metal which, similarly to Zn, is toxic and may accumulate and therefore it is included in legal regulations, but in spite of this it is rarely investigated in the context of environmental use of sewage sludge.

The aim of the present study was the application of biochar for Ni and Zn immobilization in sewage sludge applied to non-contaminated soil. The effect of the immobilization was investigated using the modified BCR method (Leśniewska et al., 2016) under a long-term field experiment. To the best of our knowledge, there is no study investigating the bioavailability of Ni and Zn under such conditions, in particular under a long-term field experiment. The lack of information in this regard limits environmental use of sewage sludge and obtaining positive results may contribute to increasing the possibility of using sewage sludge, at the same time reducing the risk associated with the presence of HMs in sewage sludge.

2. Materials and methods

2.1. Materials

Acetic acid, hydroxylamine hydrochloride, hydrogen peroxide (30%) and ammonia acetate were used for ultrasound assisted sequential extraction (UASE) analysis and provided by POCh (Gliwice, Poland). Nitric and hydrochloric acid were Suprapur and purchased from POCh (Gliwice, Poland). Ultrapure water (Milli-Q, Millipore, USA) was used during the study. The quality control of ultrasound assisted sequential extraction was performed using the certified reference material CRM BCR-701 (lake sediment, LGC, Poland). The sewage sludge (SL) was collected from municipal sewage treatment plant localized in Chełm. Sewage sludge was hygienic stabilized. The biochar applied (BC) to soil was produced from willow (*Salix viminalis*) and provided by Mostostal SA (Wroclaw, Poland). Biochar was produced at a temperature 700 °C. The physicochemical properties of S, SL and BC as well as the physicochemical properties of S and it mixture with SL and/or BC (including changes with time) is presented in Tables 1 and S1 (internet Supporting information).

2.2. Field experiment and sample collection

A field experiment was carried out in the growing season at the Experimental Farm in Bezek located in South-East part of Poland (N: 50°20′04″ E: 23°29′49″). The experiment was set up in a completely randomized block design in three replicates (3 plots with area 18.5 m^2 each) on podzolic soil (S) with the granulometric composition of loamy sand.

The SL with or without BC were added to soil before sowing during spring tillage operations. SL and BC (in suitable doses) were mixed together by mechanical (concrete) mixer, added to soil at the dose of 30 t/ha (1%) and next precisely mixed with soil by the rotatory tiller (operating depth – 22 ± 2 cm, width – 185 cm). Following experimental variants were prepared: (1) control soil without amendments (S); (2) sewage sludge (10 t_{dw}/ha) and soil (S + SL); (3) sewage sludge mixed with a 2.5% of biochar and added to soil (S + SL + 2.5% BC); (4) sewage sludge mixed with a 5% of biochar and added to soil