Journal of Hazardous Materials 317 (2016) 585–592

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Ecotoxicity of arsenic contaminated sludge after mixing with soils and addition into composting and vermicomposting processes

Jana Vašíčková, Blanka Maňáková, Marek Šudoma, Jakub Hofman*

Research Centre for Toxic Compounds in the Environment (RECETOX), Faculty of Science, Masaryk University, Kamenice 753/5, Brno CZ-62500, Czech Republic

HIGHLIGHTS

- Soil bioassays were used to detect the ecotoxicity of mineral sludge with As content.
- Sludge was mixed with soils and added into composting and vermicomposting processes.
- Soil-sludge mixtures showed inconsistent results depending on the soil, organism and dose.
- Composted and vermicomposted sludge showed high toxicity compared to sludge mixed with soil.
- Chemical analysis including mobile As determination explained only part of the bioassay results.

ARTICLE INFO

Article history:
Received 2 February 2016
Received in revised form 14 May 2016
Accepted 12 June 2016
Available online 14 June 2016

Keywords:
Sludge
Arsenic
Ecotoxicity
Bioavailability
(Vermi)composting

ABSTRACT

Sludge coming from remediation of groundwater contaminated by industry is usually managed as hazardous waste despite it might be considered for further processing as a source of nutrients. The ecotoxicity of phosphorus rich sludge contaminated with arsenic was evaluated after mixing with soil and cultivation with *Sinapis alba*, and supplementation into composting and vermicomposting processes. The *Enchytraeus crypticus* and *Folsomia candida* reproduction tests and the *Lactuca sativa* root growth test were used. Invertebrate bioassays reacted sensitively to arsenic presence in soil-sludge mixtures. The root elongation of *L. sativa* was not sensitive and showed variable results. In general, the relationship between invertebrate tests results and arsenic mobile concentration was indicated in majority endpoints. Nevertheless, significant portion of the results still cannot be satisfactorily explained by As chemistry data. Composted and vermicomposted sludge mixtures showed surprisingly high toxicity on all three tested organisms despite the decrease in arsenic mobility, probably due to toxic metabolites of bacteria and earthworms produced during these processes. The results from the study indicated the inability of chemical methods to predict the effects of complex mixtures on living organisms with respect to ecotoxicity bioassays.

1. Introduction

The remarkable increase in waste production requires the development of sustainable procedures to maximize the recovery of the beneficial properties of these materials. Treatment of groundwater contaminated by industry inevitably generates a high volume of mineral waste sludge with high concentrations of various elements, such as metal(loid)s. At the same time, mineral waste might be considered for further processing as a source of nutrients (N, P, K, Ca) [1,2]. However, the presence of metal(loids) complicated reuse of this material. Arsenic is a frequently detected in waste sludge generated during the treatment process of contaminated groundwater [3]. The levels of arsenic in sewage sludge reach usually into the tens of mg/kg [4], while in industrial sludge the level can be much higher, e.g. 396 ± 1 mg/kg [5]. Arsenic in soil can pose serious risk to soil organisms and plants and via crops to human health. Therefore, variable remediation technologies are proposed to reduce risk of arsenic in waste materials and converted arsenic into less bioavailable forms [6,7].

Bioremediation strategies have been proposed as effective methods for transformation of non-biodegradable metals from more labile and soluble form into less mobile forms [8–12]. Phytostabilization of metals by simple mixing of waste with soil and consequent plant cultivation can result in stabilization and immobilization of toxic metals [13,14]. Biochemical processes taking place in the rhizosphere may influence arsenic speciation and

* Corresponding author.
E-mail address: hofman@recetox.muni.cz (J. Hofman).

http://dx.doi.org/10.1016/j.jhazmat.2016.06.029
0304-3894/

bioavailability to plant [15,16]. Enrichment of organic materials such as compost or vermicompost with mineral materials has also been shown highly beneficial for the immobilization of heavy metals present in both primary materials [1,17,18]. The addition of organic matter can stimulate the activity of indigenous microbes, which may strongly influence arsenic speciation and transform arsenic into soluble and volatile species through the processes of oxidation, reduction and methylation [19]. During vermicomposting, microorganisms are still mainly responsible for biochemical degradation processes, but earthworms enhance microbial activity and diversity [20]. On the other hand, it has been shown that passage of arsenic contaminated soil through the earthworm gut increased water soluble arsenic [21]. These changes affect the possible risks to soil organisms as they are dominantly exposed to mobile species of metals [22,23]. The questions then arise: Does ecotoxicity decrease consequently after application of bioremediation processes?

Determination of the total metal concentration represents an important parameter to assess quality of wastes, but it is inadequate to predict the fraction bioavailable to terrestrial organisms, a measure critical for ecotoxicity and environmental risk [22,23]. Previous studies have demonstrated that soil bioassays can be successfully used to detect the risks of sewage sludge, industrial sludge and composts [24–27]. Bioassays are also useful indicators of the effectiveness of different waste treatment and stabilization processes meant to reduce the mobility of metals in the tested material [23,28,29].

The objective of the present study was to evaluate the ecotoxicity of phosphorus rich mineral sludge contaminated with arsenic after (1) mixing with soil and following cultivation with *Sinapis alba*, and supplementation into (2) composting and (3) vermicomposting processes. *Enchytraeus crypticus* and *Folsomia candida* reproduction tests and the *Lactuca sativa* root growth test were used to identify if these treatments resulted in reduced ecotoxicity. The samples for the ecotoxicity testing were taken from previous studies Maňáková et al. [5,30], where effects of these processes on the evolution of arsenic speciation, mobility and bioavailability have been described. Therefore, the second aim of this study was to explore relationships between the bioassays results and the total and mobile arsenic concentrations measured earlier. Finally, general aim was to evaluate if the bioassay-based approach is suitable for the assessment of this kind of material.

2. Materials and methods

2.1. Sludge and soils

Dewatered industrial sludge with a high arsenic concentration (396 ± 1 mg/kg dry weight) and rich in phosphorus (8.5%) and calcium (12%) was used as the initial material for the study. The sludge was produced by a remediation technology which cleans groundwater contaminated by arsenic from the phosphate industry. Total carbon and organic carbon content were low (1.3% and 0.2%, respectively). Its pH was 8.5. Detailed characterization of sludge and technology of groundwater treatment are shown in Electronic Supplementary material (Table S1) and in Maňáková et al. [5]. The sludge was air-dried, pulverized to a grain size of less than 8 mm and ground.

Two different non-contaminated arable soils (A and B) were sampled as topsoil (0–20 cm) in a rural area near the city of Brno in the Czech Republic. The soils were air-dried and sieved through a 2 mm sieve. Both soils were used for preparing soil-sludge mixtures and as control soils in bioassays. The basic properties of both soils are shown in Table 1.

Table 1
Properties of the soils A and B. As—total arsenic concentration; TOC—total organic carbon content; CEC—cation exchange capacity; N—total nitrogen; HA—humic acids; FA—fulvic acids; Clay—<0.002 mm; Fine silt—0.002–0.01 mm; Silt—0.01–0.05 mm; Fine sand—0.05–0.1 mm; Sand—0.1–2 mm.

	Soil A	Soil B
As (mg/kg)	7.7 ± 0.8	9.3 ± 0.1
pH (H_20)	6.4 ± 0.6	7.2 ± 0.7
TOC (%)	1.2 ± 0.2	4.2 ± 0.6
CEC (meq/kg)	177	525
N (%)	0.24	0.36
HA (%)	0.22	0.51
FA (%)	0.46	0.67
Clay (%)	3.30	5.60
Fine silt (%)	15.7	9.6
Silt (%)	16.9	5.8
Fine sand (%)	13.0	8.4
Sand (%)	54.4	76.2

2.2. Soil-sludge mixtures

The dry sludge was mixed with dry soils A and B to get three rates of the sludge in the mixture: 0.5%, 7.5%, and 50%. These rates correspond to the sludge concentration after virtual application (the As level in the sludge would not allow any real use in the environment) on soil as a soil improver, dredged sediment or inert waste, according to scenarios in the relevant Czech laws (20, 300 and 3750 t/ha, respectively). The ratios were calculated using a topsoil depth of 25 cm and a soil density of 1.5 kg/L. Soil-sludge mixtures were placed in plastic pots (L 50 cm × W 20 cm × H 17 cm) and water was added to 50% of water-holding capacity (WHC). The surface of each pot was sown with white mustard (*Sinapis alba*) seeds into 1.5 cm-deep holes organized in a regular 1.5 × 1.5 cm grid. The pots were equipped with wicks at the bottom enabling continuous replenishment of the moisture from large reservoirs below the pots filled with water. The pots were placed near a large window in a room with sufficient light intensity, a normal day/night regime and at laboratory temperature. During the experiment, tap water was added periodically. Plant cultivation proceeded for 90 days. Samples of the mixtures were collected after 90 days of cultivation. The soil-sludge mixtures were air-dried, homogenized and stored at laboratory temperature before testing. More details on the soil experiment with the sludge are available in Maňáková et al. [30].

2.3. Composting and vermicomposting

The mixture of manure and grass, supplemented with mineral waste sludge was composted. The components were mixed in a dry state (except for the fresh grass) in the volumetric ratio of 3:6:1 (sludge/manure/grass). Water was added equivalent to 50% WHC and maintained during the experimental period. Approximately 290 L of the mixture was composted in a commercial container for home compost production for 90 days outside the laboratory at ambient temperature. Turning was performed weekly to ensure aeration. Representative samples were collected before composting (C-0) and after 90 days of composting (C-90). The samples were air-dried, homogenized and stored at laboratory temperature before testing.

The vermicomposting was carried out in plastic boxes of 10 L capacity. Two different substrates were vermicomposted: (i) the initial mixture of manure and grass, supplemented with mineral waste sludge (C-0) and (ii) the mixture composted for 90 days (C-90). Eight liters of the dry substrate were placed in each vermicomposting box. Water was added to reach 50% WHC. Then, 200 earthworms *E. fetida* per 1 L of dry matter were added. The vermicomposts were kept in the dark at a laboratory temperature of 22 °C for a period of 90 days and the moisture content was maintained at