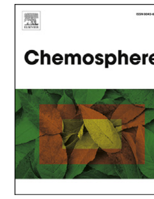
Chemosphere 198 (2018) 130–138

Contents lists available at ScienceDirect

Chemosphere

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



Efficacy of cheap amendments for stabilizing trace elements in contaminated paddy fields

Tai-Hsiang Huang [a], Yun-Jie Lai [a], Zeng-Yei Hseu [b, *]

[a] *Apollo Technology Co., LTD, Taipei 10595, Taiwan*
[b] *Department of Agricultural Chemistry, National Taiwan University, Taipei 10617, Taiwan*

HIGHLIGHTS

- Cheap amendments were applied at paddy field for two cropping seasons.
- Lime and oyster shell increased soil pH over a prolonged period.
- Lime and oyster shell were stronger than sugarcane compost in stabilizing metals.
- Oyster shell was the only amendment in effectively decreasing metal levels in rice.

ARTICLE INFO

Article history:
Received 19 October 2017
Received in revised form
27 December 2017
Accepted 22 January 2018

Keywords:
Bioavailability
Trace element
Stabilization
Rice
Soil remediation

ABSTRACT

In situ stabilization of trace elements by adding cheap amendments is an emerging technology for large-scale soil remediation. Various amendments have been examined well in the literature, but related have focused predominantly on short-term laboratory scale incubation or pot experiments. This study applied dolomitic lime at 40 ton ha^{-1}, oyster shell (OS) at 80 ton ha^{-1}, and sugarcane bagasse compost (SC) at 60 ton ha^{-1} to a paddy field in Taiwan for two rice (*Oryza sativa* L.) cropping seasons. The aims of study were to gain an understanding of the bioavailable concentrations of Cr, Ni, Cu, and Zn in the amended soil and the metal uptake of rice for practical amendment use in field-scale remediation of contaminated soils. The treatments of lime and OS significantly ($p < 0.05$) decreased the 0.1 N HCl−extractable metals in the soil. The increase in soil pH was the key factor in decreasing the bioavailable pool of metals in the soil by using lime and OS. The concentrations of Cu, Zn, and Ni in the brown rice were substantially reduced only through the addition of OS, and thus OS met the requirement of being a cheap, locally available, and environmentally compatible amendment for field-scale soil remediation. The translocation of Cr in rice plants is heavily restricted, and thus no significant differences in Cr uptake by rice grain were observed between the different amendment treatments. However, SC is not recommended as an immobilization agent because it caused a pH decrease in the amended soil.

1. Introduction

Water resources are limited for rice (*Oryza sativa* L.) production globally and are sometimes contaminated with trace elements, and thus paddy soil quality and rice safety are adversely affected by trace elements (Hseu et al., 2010). In industrialized areas throughout Asia, paddy fields for rice production are often close to industrial sites that discharge chemical waste into irrigation channels used to flood paddy fields. Therefore, accumulation of trace elements in rice grains has increasingly raised health concerns in many Asian countries, including Taiwan, Japan, China, Korea, and Thailand (Chaney et al., 2004; Römkens et al., 2009; Rahman et al., 2014). Because rice is a notable trade commodity and a staple food for most people in these countries, reducing the amounts of trace elements to safe levels is a priority (Chen, 2007; Rahman et al., 2014). Conventional techniques aimed at removing trace elements from contaminated soil, known as remediation approaches, have long been applied. Examples include excavation, landfilling, and soil washing; however, these techniques are considered inefficient in terms of energy, a waste of natural resources, and environmentally incompatible (Chang et al., 2013). Therefore, a cost-effective and less disruptive alternative for soil

* Corresponding author.
E-mail address: zyhseu@ntu.edu.tw (Z.-Y. Hseu).

https://doi.org/10.1016/j.chemosphere.2018.01.109

remediation is required (Hodson, 2010). Among the emerging technologies, in situ stabilization of trace elements by adding immobilizing agents has received increasing attention as a promising solution for large-scale soil remediation (Zhou et al., 2014; Lahori et al., 2017). The aim of in situ stabilization is not to remove trace elements from soil but rather to reduce their ability to spread into water or biota, thereby reducing their toxicity and transport potential (Dermont et al., 2008; Kumpiene et al., 2008; Radziemska, 2018).

In situ stabilization relies on adding an amendment to soil to increase the proportion of the total soil metal burden within the soil solid phase either through precipitation or increased metal sorption, thereby reducing metal solubility (Basta et al., 2005). Thus, immobilizing agents are categorized into two groups: pH change–inducing amendments and sorption agents (Kim et al., 2012; Feizi et al., 2018). The pH change-inducing amendments reduce the availability of cationic trace elements in soils through increased pH, which not only causes soil surface deprotonation and provides more sorption sites on the soil surface for trace element adsorption (Al-Abed et al., 2006; O'Day and Vlassopoulos, 2010), but also generates OH^- to offset H^+ in the soil solution, thus lowering soil acidity to form metal-containing precipitates (Chang et al., 2013). Lime, dolomite, steel slag, and naturally sourced liming materials such as oyster and egg shell have all been examined for this purpose (Dermont et al., 2008; Ok et al., 2011; Chang et al., 2013). Sorption agents include compost, clay, iron compounds, and zeolite. These materials have a variety of surface area types containing multidentate functional groups where trace elements can be adsorbed and complexed (Cheng and Hseu, 2002; Buss et al., 2012; Bolan et al., 2014; Oustriere et al., 2016; Radziemska, 2018).

Innovative low-cost, low-input, and readily available amendments are required for soil remediation and community acceptance and may provide a long-term remediation solution for reducing trace element bioavailability in contaminated soils (Dermont et al., 2008; Shaheen and Rinklebe, 2015; Kumpiene et al., 2008). Among the available amendments, dolomitic lime, oyster shell (OS), and crop residue compost have been verified as effective amendments for soil contaminated by trace elements. These cheap amendments were not only available for farmers in Taiwan but also were easily applied in the paddy field. In addition, their use has reduced waste disposal through the revalorization of agricultural waste, thereby benefiting the environment (Basta et al., 2005; Chang et al., 2013; Lahori et al., 2017). For instance, Ok et al. (2011) used OS, which contains substantial $CaCO_3$, to improve soil quality and stabilize Cd in contaminated soil containing approximately 11 mg kg^{-1} of 0.1 N HCl–extractable Cd. Treating the soil with 5% OS powder for 30 d significantly increased the soil pH and exchangeable Ca content and decreased the HCl-extractable Cd to <4.0 mg kg^{-1}. The increase in soil pH caused by adding OS powder increased the net negative charge on the soil surface, which in turn enhanced Cd adsorption. Venegas and Vidal (2016) applied vegetable residue compost in acidic soil contaminated with Cd, Cu, Ni, Pb, and Zn. After application, the compost increased in pH and acid neutralization capacity, thereby reducing the extractability of metals at the initial pH of the amended soil. Additionally, the amounts of 0.1 M $CaCl_2$–extractable Cu and Zn markedly decreased in contaminated soil amended with a compost prepared from a mixture of olive leaves and a solid fraction of olive mill wastewater. This amendment clearly reduced metal uptake by radishes (Walker et al., 2003).

Although various immobilizing agents have been examined for contaminated paddy soils, related have focused predominantly on short-term laboratory scale incubation or pot experiments (Yu et al., 2017; Liang et al., 2017), where trace element accumulation by plants can be overestimated because of root-enhanced uptake in confined spaces (Ciadamidaro et al., 2017). Additionally, Kim et al. (2012) determined that bioavailability tests for trace elements in soil are generally limited to juvenile plants and the concentration of the metals accumulated in only the shoots is determined rather than the final concentration of metals in the edible parts of the crop at harvest. One study showed that in comparison to adult plants, seedlings had fewer restrictive root-to-shoot barriers for trace elements (Gasic and Korban, 2006), resulting in greater accumulation of trace elements in the seedlings. Therefore, the lack of knowledge in the current state of in situ stabilization is the long-term effectiveness of the applied amendments in fields in determining the application of any stabilization technique. However, practical field applications of in situ stabilization of trace elements in paddy soils are rare, and thus householders (as well as policy makers, farmers, and rice consumers) need to be convinced of the efficiency of sustainable soil remediation in safe rice production. This study applied dolomitic lime, OS, and sugarcane bagasse compost (SC) to a paddy field to achieve the following objectives: (1) to understand the bioavailable concentrations of Cr, Ni, Cu, and Zn in the amended soils, (2) to illustrate the effects of these cheap amendments on metal accumulation in paddy rice over two cropping seasons, and (3) to provide a theoretical basis for in situ field-scale remediation of contaminated agricultural soils.

2. Materials and methods

2.1. Site description

The field experiment site in this study was the alluvial plain in Homei Township, Changhua County, central Taiwan (Supplemental information Fig. S1). Taiwan's climate is characterized by high temperatures and high humidity. According to the Köppen climate classification, seasonal variation between dry winters and wet summers creates a wet–dry tropical climate. The mean annual rainfall in Changhua County is 1700 mm and the mean annual air temperature is 24 °C (mean monthly temperatures range from 16 to 30 °C) in the period from 2005 to 2014. Industrial wastewater was illegally discharged from chemical, electroplate, and pigment factories in Changhua County in the 1970s, and thus the surrounding soil was contaminated with trace elements from the irrigation canal system close to these factories. Paddy rice, which is cropped biannually, has been produced for approximately a century in the area. In this study, in situ field-scale remediation was conducted in a contaminated paddy field (24°05′59″N, 120°31′37″E) close to a sublateral canal. The area of the test field was 2400 m^2 (40 m × 60 m). Additionally, the alluvial soil of the test field was derived from Holocene-epoch slate sediment classified as Typic Udorthents by the U.S. soil classification system (Soil Survey Staff, 2014).

2.2. Characterization of study soil and amendments

Before the rice cultivation experiment, a composite sample of surface soil at a depth of 0–20 cm from eight randomly selected subsamples was collected in the test field for laboratory analysis. The soil sample was air-dried, grounded, and sieved using a 2-mm mesh for subsequent laboratory analyses. Soil particle size distribution was determined using the pipette method (Gee and Bauder, 1986). Soil pH was measured in a mixture of soil and deionized water (1:1, w/v) by using a glass electrode (McLean, 1982). Total organic carbon (OC) content was determined using the Walkley–Black wet oxidation method (Nelson and Sommers, 1982). Cation exchange capacity (CEC) was determined using the ammonium acetate method (pH 7.0; Rhoades, 1982). The pseudo-total element content in the soil sample was determined by digesting