Pedosphere **27**(2): 193–204, 2017
doi:10.1016/S1002-0160(17)60310-2
ISSN 1002-0160/CN 32-1315/P
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Published by Elsevier B.V. and Science Press

PEDOSPHERE

www.elsevier.com/locate/pedosphere

Remediation of Heavy Metal-Polluted Agricultural Soils Using Clay Minerals: A Review

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(Received July 27, 2016; revised January 23, 2017)

ABSTRACT

Heavy metal contamination of agricultural soils poses risks and hazards to humans. The remediation of heavy metal-polluted soils has become a hot topic in environmental science and engineering. In this review, the application of clay minerals for the remediation of heavy metal-polluted agricultural soils is summarized, in terms of their remediation effects and mechanisms, influencing factors, and future focus. Typical clay minerals, natural sepiolite, palygorskite, and bentonite, have been widely utilized for the *in-situ* immobilization of heavy metals in soils, especially Cd-polluted paddy soils and wastewater-irrigated farmland soils. Clay minerals are able to increase soil pH, decrease the chemical-extractable fractions and bioavailability of heavy metals in soils, and reduce the heavy metal contents in edible parts of plants. The immobilization effects have been confirmed in field-scale demonstrations and pot trials. Clay minerals can improve the environmental quality of soils and alleviate the hazards of heavy metals to plants. As main factors affecting the immobilization effects, the pH and water condition of soils have drawn academic attention. The remediation mechanisms mainly include liming, precipitation, and sorption effects. However, the molecular mechanisms of microscopic immobilization are unclear. Future studies should focus on the long-term stability and improvement of clay minerals in order to obtain a better remediation effect.

Key Words: bentonite, bioavailability, immobilization, liming effect, palygorskite, sepiolite, soil pH

Citation: Xu Y, Liang X F, Xu Y M, Qin X, Huang Q Q, Wang L, Sun Y B. 2017. Remediation of heavy metal-polluted agricultural soils using clay minerals: A review. *Pedosphere*. **27**(2): 193–204.

INTRODUCTION

The accumulation of heavy metals from emissions of nonferrous metal smelting, disposal of high-metal waste, and wastewater irrigation have led to the heavy metal pollution of soils. Excessive heavy metals in soils may pose risks and hazards to humans and the ecosystem through their presence in the food chain, which has attracted academic attention and public concerns (Ahmad *et al.*, 2015).

Approximately 2×10^5 km^2 of cultivated land in China is contaminated by cadmium (Cd) and lead (Pb) (Xue *et al.*, 2014), with cultivated land contaminated by Cd covering over 1.3×10^5 km^2 (Yu *et al.*, 2006). It has been estimated that about 1.5×10^5 t farm products, including 5×10^4 t rice, are polluted by Cd each year (Yu *et al.*, 2006; Wang *et al.*, 2011). In Japan, agricultural land designated as "Cd contaminated" now exceeds 6 000 ha throughout the country, according to a survey by the Ministry of the Environment, Japan (Arao *et al.*, 2010). Some Australian-grown and Bangladeshi vegetables contain levels of heavy metals that are higher than the Australian standard maximum limits (Rahman *et al.*, 2014). Mercury (Hg) pollution in soil from Spain has attracted local attention (Higueras *et al.*, 2015).

The Ministry of Environmental Protection and the Ministry of Land and Resources of China issued a joint report on the status of soil contamination in China in 2015. Soils in some areas, especially those surrounding mining and industry activities, have been seriously polluted. Contamination by heavy metals accounts for the majority of the soils classified as being contaminated. Among the heavy metals, Cd is present at the highest percentage in soils (Zhao *et al.*, 2015).

To restore soil properties, soils polluted with heavy

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metal require remediation. Immobilization, soil washing, and phytoremediation techniques are frequently used for the remediation of heavy metal-polluted soil. Soil washing is one of the few permanent treatment alternatives used to separate heavy metals from soils, and involves physical separation and chemical extraction (Dermont *et al.*, 2008). Phytoremediation refers to the use of plants and associated soil microbes to reduce the concentrations or toxic effects of heavy metals in soils (Ali *et al.*, 2013). Techniques of phytoremediation include phytoextraction, phytofiltration, phytostabilization, phytovolatilization, and phytodegradation (Alkorta *et al.*, 2004). Immobilization of heavy metals refers to the use of *in-situ* techniques that utilize immobilization agents or amendments to reduce the bioavailability of heavy metals in soil (Rahman *et al.*, 2016). This can be achieved through adsorption, precipitation, and complexation reactions, which result in the redistribution of heavy metals from the solution phase to the solid phase, thereby reducing their bioavailability and transport in the environment (Bolan *et al.*, 2014). The most widely applied amendments include lime and limestone, clay minerals, zeolites, phosphates, and organic composts (Kumpiene *et al.*, 2008).

The function of agricultural production is the main difference between agricultural fields and metal-polluted industrial fields. Soil washing can remove heavy metals from soils, which is of great advantage. However, it is difficult to complete large-scale engineering over large areas of agricultural fields due to the high operation cost. Phytoremediation is a green solution for the remediation of heavy metal-polluted soils; however, agricultural production is affected by new plants by varying degrees. Furthermore, owing to the variety of the large areas of agricultural fields, the selected plants are affected by the local climate and other factors, which limit the application of phytoremediation over large areas. Compared with soil washing and phytoremediation, immobilization has the advantage of having a negligible impact on agricultural production.

In recent years, clay minerals have been utilized as amendments for the remediation of heavy metal-polluted soil owing to their low cost, abundant reserves, and high performance. Clay minerals are one of the most abundant materials in the earth. Clay minerals have played an important role in the development of human civilization. For environment protection, clay minerals have been used in the disposal and storage of hazardous chemicals, including heavy metals (Ismadji *et al.*, 2015). In this review, the remediation of heavy metal-polluted agricultural soils using clay minerals is summarized.

CLAY MINERALS USED AS AMENDMENTS FOR HEAVY METAL-POLLUTED SOILS

Clay minerals are hydrous aluminosilicates, broadly defined as minerals that make up the colloid fraction of soils, sediments, rocks, and water. Clay minerals play an important role in the environment by acting as a natural scavenger of pollutants, through the uptake of cations and anions either through ion exchange or adsorption (Yuan *et al.*, 2013). In the remediation of agricultural soils polluted by heavy metals, sepiolite, palygorskite, and bentonite have often been utilized as amendments.

Sepiolite

Sepiolite, $Mg_8Si_{12}O_{30}(OH)_4(H_2O)_4{\cdot}8H_2O$, is a porous fibrous hydrated magnesium silicate. Its structure is composed of blocks of two tetrahedral silica sheets, which sandwich an octahedral sheet of magnesium oxide/hydroxide (Suárez and García-Romero, 2011). Among the different clay minerals studied, sepiolite has received the most attention, especially in its sorption of heavy metals such as Cd, zinc (Zn), copper (Cu), and Pb (Álvarez-Ayuso and García-Sánchez, 2003; Keller *et al.*, 2005; Lazarević *et al.*, 2007; Kocaoba, 2009; Padilla-Ortega *et al.*, 2013).

In recent years, natural sepiolite has been applied for the immobilization of heavy metal-polluted soils as show in Table I. The remediation effects of sepiolite have been confirmed in many field demonstrations and pot trials. The advantages of sepiolite have been shown in the remediation of Cd-polluted acid paddy soils on a field scale, including its high performance, universal applicability, low cost, and simplicity of use. Sepiolite alone or in combination with other materials such as limestone can significantly reduce the Cd content of brown rice, regardless of its use in pot trials or in field demonstrations. For example, sepiolite reduced the Cd content of brown rice to 0.18 mg kg^{-1}, which is below the maximum levels proposed in the Chinese national standard "Maximum Levels of Contaminates in Foods" (GB 2762-2012) and by the Codex Alimentarius Commission (CAC 153-1995) of the World Health Organization (WHO). Additionally, sepiolite was able to increase the pH of acid paddy soils and decrease the phytoavailable fraction of heavy metals in soils (Liang *et al.*, 2014, 2015).

The addition of sepiolite decreases the bioavailable fraction of Cd in soil and then restrains Cd uptake by

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