

# **Compost as a Soil Amendment to Remediate Heavy Metal-Contaminated Agricultural Soil: Mechanisms, Efficacy, Problems, and Strategies**

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Abstract Compost or composting has been widely investigated under the background of heavy metal pollution of agricultural soils and rapid growth of organic wastes. Compost is rich in nutrients, humic matter, and microorganisms; it may be added to agricultural soil as a fertilizer to improve soil fertility and promote the growth of crops and microorganisms, and as a soil amendment to relieve heavy metal pollution. However, the effectiveness and security of compost application in agricultural soil continue to generate concern. In this review, the efficacy and mechanisms of compost remediation technologies for heavy metal-contaminated agricultural soil are presented. Poor quality, unsuitability for multiple heavy metal-contaminated soils, and potential longterm risks are the main limitations of the effectiveness and security of compost application to soils. Therefore, improving the quality of the compost, adding amendments, or combining with phytoremediation may be considered when adopting compost to remediate

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polluted agricultural soil. In addition, we propose several approaches to optimize these strategies and render the remediation of heavy metal-contaminated agricultural soil using compost safer and more effective. The findings of this review will help support the large-scale application of compost in agriculture in the future.

Keywords Soil amendment · Heavy mental pollution · Mechanism · Bioavailability · Biochar · Phytoremediation

# **1** Introduction

Soil pollution is an important factor restraining the sustainable development of agriculture and threatening grain security. High concentrations of heavy metals, such as Cd, As (arsenic is a metalloid, but is usually classified as a type of heavy metal (Järup 2003; Park et al. 2011)), Hg, Pb, Zn, Ni, and Cu, in agricultural soils are observed in many countries (Bhuiyan et al. 2010; Khaokaew et al. 2012; Liu et al. 2011; Maas et al. 2010; Wei and Yang 2010). For example, Alvarez-Ayuso et al. (2012) found that concentration ranges of As, Pb, and Sb in the northwestern regions of Zamora Province, Spain, were 246-758, 757-10,660, and 14.1-324 mg kg<sup>-1</sup>, respectively. Such values considerably exceed maximum tolerable levels in agricultural soils. Human activities, such as long-term use of sewage irrigation, extensive use of fertilizers and pesticides, and unreasonable storage of industrial and mining solid wastes, are recognized as some of the main pollution sources of soils (Bhuiyan et al. 2010; Liu et al. 2005a, b). In agriculture, heavy metals in soils can cause aquatic environment pollution through runoff into surface water or leaching into groundwater (He et al. 2004; Rattan et al. 2005). Plant growth inhibition, lower biomass production, and heavy metal accumulation result in the reduction of grain yield and cause serious economic losses on account of plants growing in polluted agricultural soils (Nagajyoti et al. 2010). Moreover, high human health risks have been observed as a result of heavy metal exposure to agricultural soils (Arao et al. 2010; Khan et al. 2008). Therefore, heavy metal pollution of agricultural soils should be effectively controlled and ameliorated.

Heavy metals, unlike organic contaminants, cannot be degraded by microorganisms. Thus, the use of soil amendments may be a highly practical and low-cost approach to remediate soils through changing the mobility and bioavailability of heavy metals (Bolan et al. 2014; Udeigwe et al. 2011). In previous studies, sewage sludge (Kidd et al. 2007), coal fly ash (Muir et al. 2007), compost (Paradelo et al. 2011), and biochar (Lucchini et al. 2014) have been used as soil amendments.

Composting refers to the process of humification and stabilization of organic wastes (e.g., sewage sludge, manure, municipal solid waste, and green waste) (Awasthi et al. 2015; Xi et al. 2012). Under the action of microorganisms and enzymes, organic wastes are degraded and transformed into CO<sub>2</sub>, H<sub>2</sub>O, mineral ions, and humic substances through different phases (i.e., mesophilic, thermophilic, and maturation stages) (Bialobrzewski et al. 2015; Lu et al. 2014). Mineral ions, humic substances, and microbes in compost considerably influence the immobilization of heavy metals and reduction of the ecological and environmental risk of heavy metals in agricultural soils (de la Fuente et al. 2011; Udovic and McBride 2012). Heavy metal immobilization can involve one or a combination of the following reactions: adsorption, complexation, precipitation, and redox reactions (Huang et al. 2010; Lagomarsino et al. 2011; Park et al. 2011; Vaca-Paulin et al. 2006). Generally, reducing the risk of crop failure and economic losses, and decreasing human health risks from heavy metals may be achieved by using compost.

Compost is regarded as a great alternative for waste management. After undergoing the composting process, the volume and weight of organic wastes are reduced, the phytotoxicity of heavy metals and organic pollutants is released, and most of pathogens and parasites are removed from the wastes (Chefetz et al. 1996; Kapanen et al. 2013; Kulikowska and Gusiatin 2015). Additionally, compost can be used as an organic fertilizer to replace chemical fertilizers in agriculture. With compost application, the physical structure and fertility of soil are improved, microbial activity is enhanced, crop biomass is increased, and crop growth is promoted (Calleja-Cervantes et al. 2015; Fagnano et al. 2011; Pérez-de-Mora et al. 2006; Proietti et al. 2015; Tian et al. 2015). Therefore, composting is a low-cost, highly practical, and environmentally friendly approach to repair heavy metal-contaminated agricultural soil. Considering these merits, compost has received widespread attention for its use as a soil amendment.

With the increasing production of compost in many countries (such as the USA, the UK, France, Germany, Australia, Iran, and China), its application in agricultural soils has been gradually implemented (Hargreaves et al. 2008). For example, in Iran, some farmers used compost on agricultural land as a soil conditioner or fertilizer (Sharifi and Renella 2015). Unfortunately, some researchers remain wary of the large-scale application of compost in agriculture because compost may also bring about several adverse effects, such as heavy metal accumulation, soil salinization and alkalization, and the pollution risk of groundwater (Achiba et al. 2009; Carbonell et al. 2011; Korboulewsky et al. 2002; Sharifi and Renella 2015). From the perspective of heavy metal remediation, most previous studies found that compost is a promising strategy to immobilize heavy metals in soils through changing the physicochemical property of soils and reacting with heavy metals (Bolan et al. 2014; Liu et al. 2009). However, the extensive use of compost may introduce additional heavy metals into soils and increase the risk of heavy metal pollution (González et al. 2012; Sharifi and Renella 2015). Thus, the risk and security of using compost in agriculture cannot be ignored.

This review describes the effects of compost application alone in polluted agricultural soils from the two aspects (i.e., mobility and bioavailability) of heavy metals. Based on effects, this review gives an analysis of some problems which hinder the application of compost as a soil amendment. On the basis of these problems, effective strategies from the existing research are also discussed. This review provides a valuable reference for the effectiveness and security of the large-scale application of compost in agriculture.

# 2 Single Application of Compost to Remediate Heavy Metal-Contaminated Agricultural Soil

2.1 Mechanisms in the Remediation of Heavy Metal-Contaminated Soils Using Compost

Compost is the product of microbial degradation of organic wastes; hence, it is rich in humus substance, microorganisms, and inorganic components. Compost addition to soil alone can change the mobility and bioavailability of heavy metals in soil environment, as well as the toxic effects on plants and animals. These actions are attributed to various processes, including adsorption, complexation, precipitation, and redox reactions.

#### 2.1.1 Adsorption and Complexation

Adsorption plays an important role in immobilizing heavy metals in soil. On one hand, compost has a potential role as a biosorbent for adsorbing heavy metals (Anastopoulos and Kyzas 2015; Soares et al. 2016), and the adsorption capacity of compost has been assessed in many studies using adsorption kinetics and adsorption isothermal experiments (Paradelo and Barral 2012; Venegas et al. 2015; Zhang 2011). Compost can also reduce heavy metal contents in water by 85-89 % through chemical absorption, and the removal ability of compost is superior to zeolite (Simantiraki and Gidarakos 2015). On the other hand, compost can influence the physicochemical properties (e.g., pH, oxidationreduction potential (Eh), and organic matter content) of soil, thereby indirectly enhancing the adsorption of heavy metals by soils (Kargar et al. 2015; Vaca-Paulin et al. 2006). Compost mainly immobilizes heavy metals through its humus substance, microorganisms, and inorganic components. Abundant humus substances in compost contain a large number of organic functional groups, such as carboxyl, carbonyl, and phenols, which can bind metal ions through complexation (Caporale et al. 2013; Chang Chien et al. 2007; Tsang et al. 2014). In addition, compost exhibit different affinities to different heavy metals. Chang Chien et al. (2006) showed that the affinity of humics with heavy metals followed the order: Pb>Cu>Cd>Zn. Humics can also act as surfactants because they have both hydrophobic and hydrophilic components (Kulikowska et al. 2015). In addition, some microorganisms in compost can immobilize heavy metals through biosorption and biomineralization (Lloyd 2002; Sudha Bai and Abraham 2003). For example, del Carmen Vargas-Garcia et al. (2012) found that more than 90 % of the available Pb was removed by some isolates (*Graphiumputredinis*, *Fusarium solani*, and *Penicillium chrysogenum*) selected from the composting process. Furthermore, iron, manganese, and aluminum oxides in compost often have potential to retain heavy metals in soil (Hettiarachchi et al. 2003; Tapia et al. 2010). The adsorption of heavy metals in these inorganic components is almost an irreversible process (Hettiarachchi et al. 2003).

# 2.1.2 Precipitation

Precipitation is an immobilization process of heavy metals because it results in low-soluble content in soil. Humic substances are an important and abundant component of compost. After compost addition to soil, humic substances react with heavy metals to produce humate or metal-humic complexes which are insoluble or too stable to be hydrolyzed, and eventually precipitated (Garcia-Mina 2006). Taking Cu for example, compost addition to soil can reduce the acid-soluble and reducible fractions of Cu by enhancing precipitation and complexation (Lagomarsino et al. 2011). The abundant phosphorus (P) content in some compost also contributes to the immobilization of heavy metals. Insoluble precipitates may form from the reaction of inorganic phosphorous compounds with soil mobile heavy metals (Katoh et al. 2014; Liu et al. 2009). The precipitation of heavy metals by humics and phosphate strongly depends on soil pH (Garcia-Mina 2006; Matusik et al. 2008). Fe compounds in compost also play an important role in the retention of heavy metals, as proven by Hettiarachchi et al. (2006) using µ-XANES and µ-XRF.

#### 2.1.3 Redox Reactions

For some variable valence heavy metals (e.g., As and Cr), different valence states mean different phytotoxicity, bioavailability, and mobility. Reaction occurs between heavy metals and organic matter in compost can turn the toxic state of heavy metals to non-toxic state. Dissolved organic matter (DOM) contained in compost promotes the redox reaction of heavy metals, such as the reduction of Cr(VI) into Cr(III) (Banks et al. 2006; Chiu et al. 2009). DOM contains redox-active functional groups (e.g., quinone moieties) which accept electrons from electron donors and present electron transfer capacities, thereby promoting the redox reaction of heavy metals (Huang et al. 2010; Yuan et al. 2011). In addition, because the dissolved oxygen is consumed by microbes thus forming anaerobic condition after the addition of compost, the reduction of sulfate may be enhanced and some heavy metals are translated into less bioavailable species of metal sulfide (Hashimoto et al. 2011; Paul 2014). Therefore, the bioavailability and toxicity of heavy metals are reduced through direct or indirect redox reactions.

# 2.2 Efficacy of Compost for Remediation of Heavy Metal-Contaminated Agricultural Soils

#### 2.2.1 Effect of Compost on the Mobility of Heavy Metals

The mobility of heavy metal in soil is controlled by its natural properties as well as those of the soil, including pH, organic matter, metal oxides content, and mineral composition (de Matos et al. 2001; Gadd 2004). For example, low pH could increase the leaching of Cr, Cu, and Zn, but the remobilization of As mainly depends on Eh condition (Kumpiene et al. 2007). Compost addition to contaminated agricultural soil can also alter the mobility of heavy metals (Table 1).

Achiba et al. (2009) showed that the mobility of heavy metals is slightly affected by 5 years of municipal solid waste (MSW) compost application in a field pot. By contrast, Clemente et al. (2010) used green waste compost to assess the mobility of heavy metals and found that compost could increase the solubility and mobility of both As and metals because of increase in pore water concentration of organic carbon and Fe in soil. Therefore, further attention can be given to the change of mobility of heavy metals in contaminated agricultural soil when applying compost.

After compost addition, different heavy metals may exert different responses. Arsenic, in particular, presents characteristics different from those of other cationic metals, such as Cu, Cd, Pb, and Cr. For example, As is easily mobilized but other cationic metals are easily fixed under high pH (Bissen and Frimmel 2003; Kumpiene et al. 2007; Temminghoff et al. 1997). Furthermore, the affinity of organic matter for As is lower than that of cationic metals (Fleming et al. 2013). In a compost experiment, Tsang et al. (2013) found that the mobility and leachability of Cu decreased whereas those of As increased through assaying the concentration of heavy metals in soil pore water and the TCLP test. Some studies also indicate that compost can increase the mobilization of As. Hartley et al. (2009, 2010) found that green waste compost increased soil pore water As concentrations in contaminated soils, which might be associated with the increased available phosphate, dissolved organic carbon, and Fe concentrations in soil pore water. Besides of As, the mobility of some heavy metals (e.g., Sb and Cu) may also increase in compost-amended soils, whereas the mobility of other heavy metals decreases in same case (Clemente et al. 2010; Zeng et al. 2015). Nevertheless, in real environment, agricultural soils are not commonly polluted by a single metal but by various metals (Cai et al. 2012). Therefore, it is worth taking great care of the remediation of multiple heavy metal (particularly in the presence of As) contaminated agricultural soil when applying compost.

Table 1 Effect of compost application on the mobility of heavy metals in soils

Feedstock	Heavy metals	Effect	Effective mechanisms	Reference
MOW, MSW, DOM	Cu, Zn, Ni, Pb, Cd	DOM and MOW compost can absorb heavy metals	Adsorption	Venegas et al. (2015)
MSW	Cd, Cu, Pb, Ni, Zn, Cr	Not significant influence on mobility factor of metals	Formation of both soluble and insoluble complexes with organic compounds	Achiba et al. (2009)
GW	As, Pb, Cu	Increased solubility and mobility of both As and metals	Increase in organic carbon and Fe in soil pore water	Clemente et al. (2010)
GW, AM	Cu, As	Reduction of the mobility and leachability of Cu but not As	Surface complexation	Tsang et al. (2013)
GW	As	Increased the concentration of As in soil pore water	Increase in phosphorous availability and increase in water-soluble Fe and carbon in pore water	Hartley et al. (2009, 2010)

MOW municipal organic waste, MSW municipal solid waste, DOM domestic waste, GW greenwaste, AM animal manure

# 2.2.2 Effect of Compost on the Bioavailability of Heavy Metals

Bioavailability is a key indicator to measure the ecological impact of heavy metals and plays an important role in ecological geochemical evaluation. However, researchers from different backgrounds may have different interpretations of bioavailability. In general, bioavailability is defined as the process for chemicals to cross an organism's cellular membrane from food (i.e., oral) or from the abiotic environment (i.e., external) to living organisms (Bolan et al. 2014; Semple et al. 2004). Therefore, both organism and soil properties, including soil pH value, cation or anion exchange capacities, soil type, soil texture (clay content), total organic carbon, and soil organic matter (Perez-Esteban et al. 2014; Naidu 2011; Zhou et al. 2014), should be considered when studying heavy metal bioavailability.

A number of recent studies have shown that compost application in contaminated agricultural soils can reduce the bioavailability of heavy metals (Table 2), thus reducing harm to plants, soil animals, and microorganisms. The effect of fresh solid olive husk and composted solid olive husk on the bioavailability of Pb and uptake by plant was studied by de la Fuente et al. (2011). Results showed that fresh solid olive husk is toxic to plant, and composted solid olive husk could reduce the bioavailability of Pb by complexing the action of humic substances. Zhou et al. (2012) also reported that green waste-based compost addition in alkaline soil can reduce the concentration of extractable Pb and its availability by using a modified BCR sequential fractionation test, and reduce Pb and Zn accumulated in Rhodes grass. Compost usually reduces the available forms of heavy metal to plants (including water-extractable and exchangeableadsorbed fractions) and increases the non-available forms (including organic matter bounded and residual fractions), which can relieve heavy metal accumulation in plants (Paradelo et al. 2011; Rosen and Chen 2014; Zheljazkov and Warman 2004). However, compost addition may also increase the bioavailability of heavy metals (Table 2). Smolinska (2015) noted that compost addition increased the accumulation of Hg in Lepidium sativum L. Carbonell et al. (2011) showed that MSW compost can slightly increase the total concentration of heavy metals and their bioavailability in soil. Although compost can increase heavy metal bioavailability and mobility, the heavy metal concentration in the edible plant parts may remain below established threshold that can damage plant quality or be harmful to human health (Papafilippaki et al. 2015). When the concentration levels in edible plant part exceed the thresholds, compost needs to be combined with some other remediation methods (e.g., phytoremediation) to remove the heavy metals (Smolinska 2015). However, few studies have surveyed the combination technologies applicable to the remediation of agricultural soil, and further research is needed. Additionally, changes in heavy metal bioavailability with compost addition still need to be given further attention.

Different effects of compost on heavy metal bioavailability may result from its stability and maturity (Walker et al. 2004) which are highly related to the content and nature of soluble organic matter in compost (Füleky and Benedek 2010). Mature compost contained more aromatic structures, and organic matter mainly occurred in the humic acids fraction which has higher stability (Huang et al. 2006). The application of stable and mature compost to soil can lead to heavy metals immobilization by forming stable complexes with surface functional groups (e.g., OH and COOH groups) of the organic polymers (Achiba et al. 2009; Madrid et al. 2007). However, unstable or immature compost contains relatively high soluble organic matter content (Madrid et al. 2007). When it applied to the agricultural soil, negative impacts on heavy metal immobilization and crop growth may be observed (He et al. 2011).

Different findings may be obtained as a result of the specific nature of different heavy metals. Many studies found that compost application effectively reduces the bioavailability and phytotoxicity of most heavy metals, except As. Fleming et al. (2013) used two methods (i.e., modified Morgan test and red worm bioassay test) to assess heavy metal bioavailability and observed that compost can reduce Pb bioavailability but fails to reduce As bioavailability. Udovic and McBride (2012) also observed that the bioavailability of Pb is reduced but that of As is increased after compost addition. Consequently, from the bioavailability point, single addition of compost may cause negative effects on the remediation of multiple heavy metal contamination in agricultural soil.

Many studies have concentrated on remedying agricultural soil by using compost through long-term experiments. Baldantoni et al. (2010) observed a rising tendency of total and available metal concentrations in two textured soils after 3-year application of compost. Sato et al. (2010) found that compost addition causes little

SOHMn, Fe, Zn, PbBioaccumulation test (Beta maritima L)GWPh, ZnSingle-step chemical extraction (CaCl2, EDTA and HCI), sequential extraction (modified BCR), bioaccumulation test (Lepidium sativum L)GWHgSingle-step chemical extraction (DTPA), bioaccumulation test (Lepidium sativum L)MSWCr, Ni, Cu, Pb, ZnSingle-step chemical extraction (Water andret)MSWCr, Ni, Cu, Pb, ZnSingle-step chemical extraction (Water andret)MSWCd, Zn, PbSingle-step chemical extraction (Water andret)UKPb, AsSingle-step chemical extraction (MAE), bioascay test (Eisenia andret)GWZnSingle-step chemical extraction (DTPA), bioascay test (Eisenia andret)GWZnSingle-step chemical extraction (MAE), bioascay test (Eisenia bioascay test (EiseniaFWPb, AsNi, Cr, Pb, Zn, Ni, CrMS	Bioavailability assay Effect	ct	Effective mechanisms	Reference
Pb, Zn Hg Cr, Ni, Cu, Pb, Zn Cd, Zn, Pb Pb, As Pb, As Pb, As Pb, As Cu, Cd, Pb, Zn Cd, Cu, Pb, Zn, Ni, Cr		Reduction of the concentration of Pb in shoot tissue of plant	Strong chelating ability	de la Fuente et al. (2011)
Hg Ct, Ni, Cu, Pb, Zn Cd, Zn, Pb Pb, As Pb, As Pb, As V Cu, Cd, Pb, Zn Cd, Cu, Pb, Zn, Ni, Cr		Reduced Pb and Zn accumulated in Rhodes grass	Adsorption and complexation	Zhou et al. (2012)
Cr, Ni, Cu, Pb, Zn cd, Zn, Pb Pb, As Zn Zn Pb, As Pb, As Cu, Cd, Pb, Zn Cd, Cu, Pb, Zn, Ni, Cr	þ	Increase in accumulation of Hg in Lepidium sativum L.	Formation of the organic Hg compounds which are easier to uptake by plants than inorganic Hg	Smolinska (2015)
Cd, Zn, PbSingle-step and CatenhousesPb, AsSingle-stepPb, AsSingle-step(ammon extractic bioascas)ZnSingle-cher bioascaSingle-cher bioasca bioascaPb, AsModified h in vitro scaber)WCu, Cd, Pb, ZnSingle-cher bioascaerCd, Cu, Pb, Zn, Ni, CrBCR test		Slightly increase in the total and available metals concentration		Carbonell et al. (2011)
Pb, As Single-step (ammon extractic bioassay Zn Single-cher sequent bioaccur bioacur bioaccur bioaccur bioaccur bioaccur bioacur bioaccur bioaccur		A different result between the two methods	Formation of the complexes with soluble organic compounds	González et al. (2013)
Zn Single-chei sequenti bioaccur bioaccur L. <i>Cava</i> Pb, As Modified N in vitro <i>scaber</i> ) in vitro <i>scaber</i> ) Cu, Cd, Pb, Zn Ni, Cr BCR test	tial Re	Reduced availability of Pb but increase in As availability by modified Morgan's test and the red worm bioassay; BCR test do not reduce Pb availability	Increasing in the soil pH	Fleming et al. (2013)
Pb, As Modified M in vitro in vitro <i>scaber</i> ) scaber) ;W Cu, Cd, Pb, Zn Single-cher Cd, Cu, Pb, Zn, Ni, Cr BCR test	7	duction of the contents of Zn in plant, but not significantly affect on Zn bioavailability through chemical extraction methods	Reduction of the contents of Zn in plant, but Formation of the metal-humic complexes not significantly affect on Zn bioavailability through chemical extraction methods	Al Chami et al. (2013)
5W Cu, Cd, Pb, Zn Single-cher Cd, Cu, Pb, Zn, Ni, Cr BCR test		Reduction of the bioavailability of Cd in soil, but increased As potential bioavailability	Pb: complexation and precipitate; As: increase in the soil pH and competitive displacement by $PO_4^{3-}$	Udovic and McBride (2012)
Cd, Cu, Pb, Zn, Ni, Cr BCR test		The rising tendency of total and available metals concentrations during 3 years	The soluble fraction of organic matter may increase metal availability by chelation	Baldantoni et al. (2010)
	Incr Incr Incr Incr Incr Incr Incr Incr	Increase in the total metals concentration; metals, except Cd and Pb, mainly found in the residual fraction; not significant influence on the exchangeable fraction of metals	Complexation	Ben Achiba et al. (2010)
CM Cd Sequential extraction, bioaccumulation test ( <i>E. vesicaria</i> L. <i>Cavalieri</i> )	lation	Immobilization of Cd in soils and decreased the uptake of Cd by wheat	Increase in the soil pH and formation of insoluble precipitates with inorganic phosphorous compounds	Liu et al. (2009)

accumulation of Cd in soils in a 4-year field experiment. In a 7-year field plot experiment, Ben Achiba et al. (2010) found that the total concentrations of heavy metals, particularly for Cu, Zn, Pb, and Cd, in the amended soil increased with increasing dosages of compost addition. Some researchers believe that long-term application of compost may promote the mobilization or redistribution of heavy metals with pH change and humic mineralization, thus leading to serious pollution (Ben Achiba et al. 2010; Díaz-Barrientos et al. 2003). With the passage of time, compost addition alone may cause potential risks to the environment or humans. Further studies are needed to completely monitor and evaluate the influence of compost on the mobility and bioavailability of heavy metals in agricultural soils in long-term field experiments. Effective strategies should also be developed to solve the problem.

# **3** Problems of Single Application of Compost to Remediate the Contaminated Agricultural Soil

This section reviews the remediation mechanisms of compost for heavy metal-contaminated agricultural soil. The adsorption, complexation, precipitation, and redox reactions among heavy metals, soil, and compost are mainly discussed. Compost can generally immobilize heavy metals in farmland soils through these reactions, thereby reducing the harms to plants, microbes, and humans. However, compost can also cause serious heavy metal pollution by increasing heavy metal content, mobility, bioavailability, and phytotoxicity. Consequently, the value of compost as soil amendment is reduced. The problems on the single application of compost are summarized as follows:

- Compost products worldwide are mostly of poor quality (Fuchs and Cuijpers 2016; Smith 2009). Such quality would not only result in the accumulation of heavy metals from compost in the soil (Alvarenga et al. 2015; Kupper et al. 2014; Saha et al. 2010; Smith 2009) but also affect the remediation capacity of compost in contaminated soils (He et al. 2011), thereby limiting the use of this material in agriculture.
- Different heavy metals may not achieve the same immobilization/mobilization effect with compost application (Ben Achiba et al. 2010; Fleming et al.

2013). Therefore, risk may exist in the remediation of multiple heavy metals (particularly the coexistence of As with other heavy metals)-polluted agricultural soils by using compost.

 Heavy metals can be fixed in soils over a short-term period. However, as a result of external environment changes (e.g., pH), decomposition of organic matter, and other factors (Ben Achiba et al. 2010; Bolan and Duraisamy 2003; Díaz-Barrientos et al. 2003), heavy metals may be slowly released or redistributed as time goes on, hence resulting in serious heavy metal pollution.

# 4 Strategies for Enhanced Remediation of Heavy Metals by Using Compost

4.1 Improvement of Compost Quality

# 4.1.1 Inoculation Methods

Microbes play an important role in the composting process. The addition of inocula during composting process can increase the N and C mineralization; enhance the biodegradation of polysaccharides, proteins, and aliphatics; and increase the humic substance contents and humification degree, thereby accelerating compost maturity and improving the quality of compost (Awasthi et al. 2014; Guo et al. 2015; Jiang et al. 2015; Mupambwa et al. 2016). However, some researchers showed that there was no significant effect with inocula addition (Faure and Deschamps 1991). This result may be related to the type and number of microbial inoculum, the timing of inoculation, and the competition between indigenous microbes and inoculations microbes (Xi et al. 2015). To make full use of microorganisms, multistage inoculation of different microbes in compost is considered as a more efficient strategy than the initial-stage and two-stage inoculation (Xi et al. 2012). Currently, researches begin to focus on the study of multistage inoculation. For example, Zhou et al. (2015) inoculated *Thermoactinomyces* sp.  $GF_1$  and GF<sub>2</sub>, Coprinus cinerea and Coprinus comatus, and Trichoderma harzianum and Rhizopus oryzae in the first, second (after thermophilic phase), and third (after C. cinerea and C. comatus had disappeared) stages of composting, respectively. As a result, the composting process was improved, and the maturity of compost was accelerated (Zhou et al. 2015). Multistage inoculation effectively avoids the competition between inoculations microbes; thus, they can fully use the nutrients in compost to enhance growth and propagation. The inoculations microbes are also protected from competing with indigenous microbes, and the microbial community diversity is increased (Xi et al. 2015).

#### 4.1.2 Addition of Chemical Additives

Chemical additives, such as liming, fly ash, zeolite, earthworm casts, polyethylene glycol, and biochar, are usually used to optimize the composting environment and process, and improve the quality of compost. For example, the presence of biochar in composting can increase the temperatures of composting (Wei et al. 2014), prolong the thermophilic phase (Wei et al. 2014; Zhang and Sun 2014a), and regulate free air space and aeration (Sanchez-Garcia et al. 2015; Zhang and Sun 2014a). Earthworm casts, a peat-like material, have high porosity and a large surface area, and contain many nutrients (e.g., exchangeable phosphorus, potassium, calcium, and magnesium). The presence of earthworm casts in composting can not only result in good water-holding capacity, but also support a great environment for microbial growth and activity (Zhang and Sun 2015). Previous researchers used zeolite and earthworm casts (Zhang and Sun 2015), rhamnolipid (Zhang and Sun 2014b), and biochar (Zhang and Sun 2014a) to aid the composting process. Addition of these chemical additives prolong the high-temperature stage, increase the microbial numbers and enzyme activities, enhance the humification and degradation of organic wastes, and finally improve the finished compost quality (Zhang and Sun 2014a, b, 2015). The results of these studies are consistent with those obtained by some other researchers who used phosphogypsum, fly ash, jaggery, polyethylene glycol, wood shavings, and agricultural and yard waste as additives (Awasthi et al. 2015; Gabhane et al. 2012). It is also noteworthy that some additives can immobilize heavy metals during composting process. For example, natural zeolite, a crystalline mineral, has a strong adsorbability and ion-exchange capacity due to its unique crystalline structure (Zorpas et al. 2008), so that it is capable of reducing the bioavailability of heavy metals in composting. In addition, Lu et al. (2014) used rock phosphate to aid composting, which reduced the Cu and Zn contents, as well as the Cu bioavailability, as a result of the enhanced adsorption and complexation process, increased pH value and promoted the degradation of organic matter. Singh and Kalamdhad (2013c) found that the use of lime in composting process can increase pH, and provide an appropriate Ca content, thus efficiently reducing heavy metal bioavailability and leachability.

#### 4.1.3 Earthworm Composting

Earthworm activities in the composting process result in the high maturity and stability of compost (Yang et al. 2014). The nutrient content is increased, and the toxicity of heavy metals can be relieved during this process (Song et al. 2014). Heavy metal bioavailability is reduced in compost, which is either caused by the formation of stable metal-humus complexes for the high level of humification and mineralization of vermicomposting (Hait and Tare 2012; Song et al. 2014), or the enrichment of heavy metals in the body of earthworm tissue (Dai et al. 2004; Goswami et al. 2014; Song et al. 2014). Soil pH change is also a significant factor in influencing the speciation of heavy metals in soil. With the decreased of pH, bioavailable fractions, such as exchangeable and carbonate-bounded heavy metals, are reduced and some inert fractions (residual) of heavy metals are increased (Singh and Kalamdhad 2013a, b). Therefore, earthworm composting may be a good method to improve compost quality. For example, Maňáková et al. (2014) demonstrated that bioavailable fraction of As in vermicomposting process was reduced more significantly than in composting, and As(V) was more dominant than As(III) in finished vermicompost. Soobhany et al. (2015) also found that earthworm composting significantly could decrease the contents of Cu, Cd, Cr, Co, Zn, and Ni to less degree than the Mauritius standards.

In conclusion, addition of inocula, chemical additives, and earthworms to the composting process can optimize the compost environment, maintain nutrients, promote the degradation of organic matter, and increase the humification level, thereby indirectly enhancing the remediation potential of compost. Furthermore, these additions can also reduce the concentrations and bioavailability of heavy metals in compost, hence lowering the risk of introduction of heavy metals caused by compost application.

# 4.2 Combination of Compost and Other Soil Amendments

A number of studies have concentrated on using compost in combination with other soil amendments to repair soils, including traditional amendments (e.g., zeolite, fly ash and liming) and biochar. These studies showed that compost-amendment mixtures can improve soil fertility, immobilize heavy metals, promote plant growth and yields, increase microbial activities, and reduce ecological risk (Beesley et al. 2014; Chander and Joergensen 2002; Kostov and Van Cleemput 2001; Ye et al. 2000; Zeng et al. 2015). Such effects may be attributed to the interaction of compost with other amendments. For example, Odlare and Pell (2009) suggested that compost can potentially mitigate the toxic effect of wood fly ash in agricultural soil. It is noteworthy that the combination method was commonly applied in non-agricultural soils (e.g., mining and urban soil). For the remediation of agricultural soil, this method may provide a good reference.

# 4.2.1 Traditional Amendments

A lot of traditional amendments have been proposed and tested for the combination with compost to immobilize heavy metals in soil. These amendments include silicon/ calcium materials (e.g., liming and silicate), clay minerals (e.g., zeolite, bentonite, and sepiolite), and metals and metallic oxides (e.g., zero-valent iron and iron oxide). The remediation mechanisms of these amendments may be different. For example, silicon/calcium materials are added to achieve the stabilization of heavy metals through the raising of soil pH and the formation of insoluble silicates and carbonates (Gray et al. 2006). Clay mineral, with high surface area, can immobilize heavy metals by adsorption, complexing, and coprecipitation (Shi et al. 2009; Sun et al. 2015). Because of these different characteristics and remediation mechanisms, the combination of compost with these soil amendments has a synergistic effect or a complementary effect with a greater efficacy for stabling heavy metals in soil (Brown et al. 2003; Tsang et al. 2013). The effect of combined of compost and traditional amendments on the mobility and bioavailability of heavy metals is concluded in Table 3. Alvarenga et al. (2008) applied different dosages of MSW compost combined with calcium oxides in an acidic mine soil. This treatment not only improved soil quality (e.g., increased soil organic matter and available P and K contents) and accelerated plant growth (i.e., *perennial ryegrass*), but also reduced the mobile and bioavailable fractions of Cu, Pb, and Zn. Hwang and Neculita (2013) showed that food waste compost in combination with zeolite, which was added to tailings, could significantly increase soil pH. The mixtures could also benefit the immobilization of heavy metals through adsorption and precipitation processes.

For As-contaminated soils, the use of compost combination with other amendments is a good strategy. The mobility and availability of As in soil are mainly dependent on adsorption/desorption processes and coprecipitation with metal oxides (Kumpiene et al. 2008). In general, iron salts, including Fe oxides and zero-valent iron, are commonly used for As immobilization purposes (Hartley and Lepp 2008; Kumpiene et al. 2008). Iron salts react with arsenic to reduce the stabilization of As in soil by the formation of insoluble secondary oxidation minerals and amorphous iron(III) arsenate (FeAsO<sub>4</sub>·H<sub>2</sub>O) (Kumpiene et al. 2008). Therefore, the combination of iron salts and compost can be beneficial to make up for the lack of compost used in arsenic-contaminated soil. In a previous research, the application of green waste or manure compost reduced the Cu concentration in pore water and leachability, but fails to reduce the mobility of As in soils (Tsang et al. 2013). Nevertheless, combination of compost with zero-valent iron or acid mine drainage sludge can simultaneously reduce the leachability of Cu and As. González et al. (2012) have studied the result of applying compost alone or the combination of compost, marble sludge, and synthetic Fe oxides. They found that the mixtures considerably reduced the Pb, Cd, and Zn concentrations in both soil lixiviate solution and pore water, as well as the As content in pore water. Compost applied together with traditional amendments (e.g., zeolite and iron oxide) in different proportions can also reduce the bioavailability and plant uptake of As (Gadepalle et al. 2008). Gadepalle et al. (2008) indicated that the remediation of heavy metal-polluted soil is effective when high-dosage compost is mixed with low-dosage zeolite or iron oxide. The different results of remediation using compost combination with other amendments may be related to the types and characteristic of amendments. Tsang et al. (2014) concluded that the leachability and concentration of As in pore water can be reduced by addition of green waste compost and fly ash mixture, rather than the

Table 3 Effect of the contract o	Effect of the combination of compost with		other amendments application on the heavy metals in soils		
Group	Metals	Source of soil	Effect	Effective mechanisms	Reference
MSWC + CaO	Cu, Pb, Zn	Mining soil	Reduction of the mobility and bioavailability of Cu. Db. and Zn	Increase in the soil pH and formation of the insolution contrastes and/or phosphates	Alvarenga et al. (2008)
Compost + lime + phosphate	Pb, Mn	From the backyard of Surrey Fire	More efficient in decreasing the exchangeable metals fraction and mobility and bioavailability of metals by combined application than addition alone	Increase in the soil ph and formation of the complexes and precipitates	Padmavathiamma and Li (2010)
GWC + biochar	Cu, Pb	From mature woodland	Immobilization of Cu and Pb for the mixture addition is much more efficient than commost sincly	Increase in the soil organic matter content, soil and pore water pH; high content P and Fe in commost and biochar	Karami et al. (2011)
Compost + biochar	Cd, Cu, Zn, Pb	Wetland soil	Metals bioavailability, mobility, and ecological risk reduced more in amended soil with compost-biochar mixture don compost sindly	Free ions form complex with organic ligands and heavy metals exchange with $Ca^{2+}$ , $Mg^{2+}$ , and other cation associated	Zeng et al. (2015)
Compost + biochar	Cu, Ni, Pb, Zn	Mining soil	Reduction of the mobility factor for Ni, Pb, and Cu and the content of the CaClextractable of heavy metals	Increase in the soil pH and formation of the stable complexes between organic matter and heavy metals	Rodríguez-Vila et al. (2014, 2015)
GWC + biochar	Zn, Cd, As, Cu	From an embankment	Decrease in phytotoxicity of heavy metals, and contents of Zn and Cd in pore water, but increased the pore water	Increase in the soil pH and dissolved organic carbon; effect of soluble phosphorous on As	Beesley et al. (2010)
OMWC + biochar	Cd, Cu, Pb, Zn, As Mining	Mining soil	concentration of As and Cu Increase in As mobility but not increase in the mobility of other heavy metals $(i, -C_1, D_1, C_2, -C_2)$	Increase in the soil pH, dissolved organic carbon, and soluble phosphorous	Beesley et al. (2014)
Compost + AMDS, Compost + zero-valent iron	Cu, As	From a former timber treatment facility	u.e., eu, ro, eu, zu) Reduction of the mobility of As	Adsorption and co-precipitation of inor- ganic materials (AMDS and zero-valent iron)	Tsang et al. (2013)
CM + MS, CM + BF, CM Zn, Cd, Cu, Pb, As Mining + MS + BF	Zn, Cd, Cu, Pb, As	Mining soil	Decrease in the Pb, Cd, Zn mobility and As content in pore water, but not decrease in As concentration in lixiviate solution	and surface complexation of compost Zn, Cd, and Pb: Increase in the soil pH and organic matter; As: increase in the pH, dominance of negatively charged arsenate species, and competition from soluble	González et al. (2012)
Compost + zeolite, Compost + iron oxide GWC + fly ash, GWC + limestone, GWC + bentonite	As As, Cu	Mining soil From a former timber treatment facility	Reduction of the bioavailability and plant uptake of As GWC + fly ash could reduce As leachability and pore water contents of As, but not reduced by GWC + limestone or GWC + bentonite	organic compounds Adsorption and pH effect Adsorption on the Fe/Al coprecipitates or incorporation into calcium-silicate- hydrate	Gadepalle et al. (2008) Tsang et al. (2014)
MSWC municipal solid waste compost marble sludge, BF synthetic Fe oxides	'aste compost, <i>GWC</i> 'tic Fe oxides	7 greenwaste compost,	<i>MSWC</i> municipal solid waste compost, <i>GWC</i> greenwaste compost, <i>OMWC</i> olive mill waste compost, <i>AMDS</i> acid mine drainage sludge, <i>CM</i> agricultural greenhouse wastes compost, <i>MS</i> marble sludge, <i>BF</i> synthetic Fe oxides	pueses cid mine drainage sludge, CM agricultural g	reenhouse wastes compost, MS

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combination of compost with limestone or bentonite. This difference may be the result of the effect of amendments on negligible Fe dissolution or Fe-colloid mobilization in soil because As transport was mainly controlled by colloidal Fe oxides at pH above 4.5 (Neubauer et al. 2013; Tsang et al. 2014).

### 4.2.2 Biochar

Biochar is a carbon-rich material. Some researchers have observed good remediation effects by combining compost with biochar (Table 3). Generally, biochar has a neutral to alkaline pH and large surface area, both of which contribute to the stabilization and adsorption of heavy metals (Ahmad et al. 2014; Chen et al. 2008; Lehmann et al. 2011). The use of biochar in soil has been considered as a significant strategy to immobilize heavy metals (Zhang et al. 2013). In recent years, researchers have been increasingly focusing on combining biochar with compost to remediate the polluted soils. In a 4-month pot experiment, combined application of compost and biochar was observed to increase the soil organic matter and available N and P, thereby offering sufficient nutrition for plant growth promotion. This treatment also significantly reduced the concentration of Cu and Pb in soil and pore water (Karami et al. 2011). Zeng et al. (2015) studied the total and available heavy metal content in soil pore water and soil microbial biomass to assess the bioavailability, mobility, and ecological risk of Cu, Cd, Zn, and Pb in compost combined with biochar-amended soil. These researchers found more reductions in metal bioavailability, mobility, and ecological risk in amended soil with compost-biochar mixture than the single application of compost.

Although there are many related studies in recent years, few studies have investigated the effect of combined biochar and compost to repair multiple heavy metal-contaminated soils containing As. Beesley et al. (2010) applied mixture of green waste compost and biochar to a multi-element-contaminated soil and found this method could reduce the phytotoxicity of heavy metals and Zn and Cd contents in pore water but increase the concentration of As and Cu in pore water. Beesley et al. (2014) showed that the mobility of As increased whereas that of other metals (i.e., Cd, Pb, Cu, and Zn) was reduced by this technology. It in concluded that the effect of compost and biochar mixture in repairing arsenic-contaminated soil is not very clear; thus, further studies on this topic are necessary. Combining compost with other amendments may remediate the polluted soil more efficiently than the compost application alone. Suitable amendment approach should be reasonably selected based on the pollution characteristics of agriculture soil, particularly for As-polluted soils. In addition, the interaction between compost and other amendments is relatively complex. Future research could pay more attention to the interaction effect, mechanism, and influencing factors of amendment mixtures.

#### 4.3 Combination of Compost and Phytoremediation

Compost addition can immobilize heavy metals in soil, which means that it reduces heavy metal mobility and bioavailability but not actually remove them from soil. Moreover, different heavy metals may not achieve the same immobilization/ mobilization effect with compost addition in multimetal-polluted soils. Phytoremediation, particularly the phytoextraction, is an inexpensive and convenient technology (Ali et al. 2013; Meagher 2000; Salt et al. 1998). Phytoextraction, which uses plants to transfer and accumulate heavy metals from soil into roots and shoots, is one of the main mechanisms for phytoremediation of soil polluted with heavy metals (Jing et al. 2007). Phytoextraction includes natural and chelate-induced process (Smolinska 2015). The natural process mainly depends on hyperaccumulator plants. However, they have a limited biomass production and slow growth rate; in addition, they accumulate only certain heavy metal element (Brunetti et al. 2012; Evangelou et al. 2007), examples of which include Pteris vittata for As (Wang et al. 2002), Sedum alfredii Hance for Zn and Cd (Yang et al. 2004), Phytolacca acinosa Roxb. (Phytolaccaceae) for Mn (Xue et al. 2004), and Sesbania drummondii for Pb (Sahi et al. 2002). The chelate-induced process mainly depends on soil amendments (e.g., nitrilotriacetic acid, EDTA, and compost) to increase the accumulation effect by plants (Evangelou et al. 2007; Meers et al. 2005; Smolinska 2015). Compost contains a large number of organic matters, and its application in soil can facilitate the uptake of heavy metals by accumulator plants or simple higher plants. This may be resulted from the positive effect of compost on plant biomass and biometric parameters and the formation of soluble heavy metal-organic complexes (Businelli et al. 2009; Smolinska 2015; Zheljazkov and Warman 2004). Smolinska (2015) reported that green waste compost could efficiently extract Hg through *L. sativum* L. in Hg-contaminated soil. Brunetti et al. (2012) found that compost and mixtures of compost and *Bacillus licheniformis* can promote the accumulation of heavy metals (Cr, Cu, Pb, and Zn) in plants. The mobility factor of Ni, Pb, and Cu and the content of the CaCl<sub>2</sub>-extractable metals in the soil can be significantly decreased and Ni can be efficiently extracted by mustards (Rodríguez-Vila et al. 2014, 2015). Hence, the combination of accumulator plants or simple higher plants with compost to remediate heavy metals which cannot be immobilized by compost can be a feasible strategy.

# **5** Conclusions and Future Research Directions

Single application of compost can potentially fix heavy metals in soil. However, the maturity degree of most compost is low, and compost generally contains a large amount of heavy metals. Application of low-quality compost in soil can not only harm the growth of plants and microbes but also reduce its effectiveness as a soil amendment. Additionally, compost addition alone may not be suitable for remediating multiple heavy metalpolluted agricultural soils because of the characteristic of the soil, compost, and heavy metals. Compost only immobilizes heavy metals in soil and reduces their availability and uptake by plants, while heavy metals still retained in the soil. With the passage of time, serious heavy metal pollution may occur because of environmental changes and organic matter decomposition. Thus, large-scale use of compost is limited in agriculture in many cases. The negative impacts of compost applied in agriculture should be reduced to broaden its application.

This paper summarizes some methods to improve the quality of compost based on the related literature. Strategies, such as the combination of compost and other soil amendments, as well as the combination of compost and phytoremediation, are recommended based on related studies in repairing heavy metalcontaminated non-agricultural soils. The use of these methods can mitigate the problems of multiple heavy metal pollution and long-term risk in agriculture. However, differences exist between non-agricultural and agricultural soils in terms of soil structure, fertility, and contamination level. To date, few research studies have used the above mentioned to repair heavy metals contaminated agricultural soils. To enhance the effectiveness and security of compost application in agriculture, these methods can be applied to agricultural soil, and the following research areas can be further studied.

- 1. The multistage inoculation process of composting is optimized by using some microbes which can immobilize or adsorb heavy metals. Therefore, these microbes are needed to research in further studies.
- 2. Little is known about the repair mechanism of compost-amendment mixtures in contaminated soils. Further studies are still needed to clarify the interaction between the compost and other amendment and the mechanism among amendments, soil, and heavy metals.
- Single application of compost or/and a combination of compost and other amendments do not remove heavy metals from soil. Heavy metals remaining in the soil may cause serious heavy metal reactivation. Therefore, advanced methods and technologies are needed for long-term monitoring.
- 4. Modified (such as acid/base activation, graft, and load) materials may undergo excellent adsorption, complexation, precipitation, and redox reactions with pollutants. The application of modified materials in compost-amended soil may exert significant effects on the promotion of composting technology in agriculture.
- 5. The addition of compost in combination with phytoremediation is an efficient technology to mitigate the long-term risk of compost application. Nevertheless, few literature reports have studied these aspects such as plant selection and the proportion of applied compost and rhizosphere biochemistry in relation to this technology. These deficiencies should also be further studied.
- 6. The effect of compost combination with other amendments on enzymatic and microbial aspects in contaminated soils should also be explored in future studies.
- 7. Agricultural measures, including water management and land use pattern change, are important activities in agriculture. The influence of these measures on heavy metal remediation using the combination of compost and other amendments is still unclear. Further studies are necessary to address these problems.

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