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Research review paper

# Bioremediation of soils contaminated with polycyclic aromatic hydrocarbons, petroleum, pesticides, chlorophenols and heavy metals by composting: Applications, microbes and future research needs

Ming Chen [a,b], Piao Xu [a,b], Guangming Zeng [a,b,*], Chunping Yang [a,b], Danlian Huang [a,b], Jiachao Zhang [c]

[a] College of Environmental Science and Engineering, Hunan University, Changsha 410082, China
[b] Key Laboratory of Environmental Biology and Pollution Control (Hunan University), Ministry of Education, Changsha 410082, China
[c] College of Resources and Environment, Hunan Agricultural University, Changsha 410128, China

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ABSTRACT

Increasing soil pollution problems have caused world-wide concerns. Large numbers of contaminants such as polycyclic aromatic hydrocarbons (PAHs), petroleum and related products, pesticides, chlorophenols and heavy metals enter the soil, posing a huge threat to human health and natural ecosystem. Chemical and physical technologies for soil remediation are either incompetent or too costly. Composting or compost addition can simultaneously increase soil organic matter content and soil fertility besides bioremediation, and thus is believed to be one of the most cost-effective methods for soil remediation. This paper reviews the application of composting/compost for soil bioremediation, and further provides a critical view on the effects of this technology on microbial aspects in contaminated soils. This review also discusses the future research needs for contaminated soils.

**Contents**

## 1. Introduction

Soil contamination resulting from industrial and agricultural activities has caused high concerns in recent years (Ha et al., 2014). Various pollutants entering the soil or water pose a huge threat to human health and natural ecosystem (Gong et al., 2009; Hu et al., 2013; Kavamura and Esposito, 2010; Tang et al., 2014; Udeigwe et al., 2011; Xu et al., 2012; G.

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* Corresponding author at: College of Environmental Science and Engineering, Hunan University, Changsha 410082, China.
*E-mail address:* zgming@hnu.edu.cn (G. Zeng).

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Zeng et al., 2013; G.M. Zeng et al., 2013). Main soil pollutants include polycyclic aromatic hydrocarbons (PAHs), petroleum and related products, pesticides, chlorophenols and heavy metals.

Composting, the major process of stabilizing agricultural solid waste and municipal solid waste (MSW) through the degradation of biodegradable components by microbial communities, has been adopted as one of the most cost-effective technologies for soil bioremediation (Alburquerque et al., 2009; Fernandez et al., 2007; Gandolfi et al., 2010; Huang et al., 2008, 2010; Yu et al., 2011; G.M. Zeng et al., 2011). Accordingly, its application for the remediation of contaminated soil is increasing, due to the fact that chemical and physical remediation technologies are either incompetent or too costly. Over the years, large numbers of works have validated its effectiveness for the remediation of soils polluted by a wide range of organic pollutants and heavy metals (de la Fuente et al., 2011; Laine and Jørgensen, 1997; Megharaj et al., 2011; Semple et al., 2001; Tandy et al., 2009). Composting strategies for soil bioremediation are diverse, including direct composting, compost addition, bioaugmentation, incorporation of bulking agent and surfactant application (Fig. 1). Researchers often employed a single or a combination of these strategies to achieve their ends. To reduce costs, organic wastes from industrial and agricultural practices were often selected as the initial composting materials during soil bioremediation. The utilization of these organic wastes for soil remediation is also helpful in decreasing the need for their storage and treatment. Organic matter from composting offers the benefit of improving soil quality and fertility (Pedra et al., 2007). The application of municipal solid waste compost (MSWC) effectively promoted soil organic matter content in Haplic Podzol and Calcic Vertisol (Pedra et al., 2007). The addition of composted sewage sludge (SS) and thermally dried SS to soils induced an increase on the content of available P, total N and total organic C (Fernandez et al., 2009).

Composting is a technology that utilizes microbes to clean up or stabilize the pollutants (Kästner and Mahro, 1996; Lu et al., 2013; G. Zeng et al., 2011; Zhang et al., 2013). A large number of studies showed that many kinds of microbes had strong ability to degrade various organic pollutants and imposed excellent passivation effect on heavy metals (Samanta et al., 2002; Watanabe, 2001; Yu et al., 2011). Bacteria and fungi, the main pollutant-degrading microbes in composts, have been widely considered to be the most crucial factors governing the remediation of contaminated soils. Remediation of contaminated soils by composting or compost addition mainly relies on two mechanisms (Puglisi et al., 2007): (i) adsorption by organic matter and (ii) degradation by microorganisms (Fig. 1). The decomposition of organic pollutants in soil/compost mixture relies mostly on the microbial activity.

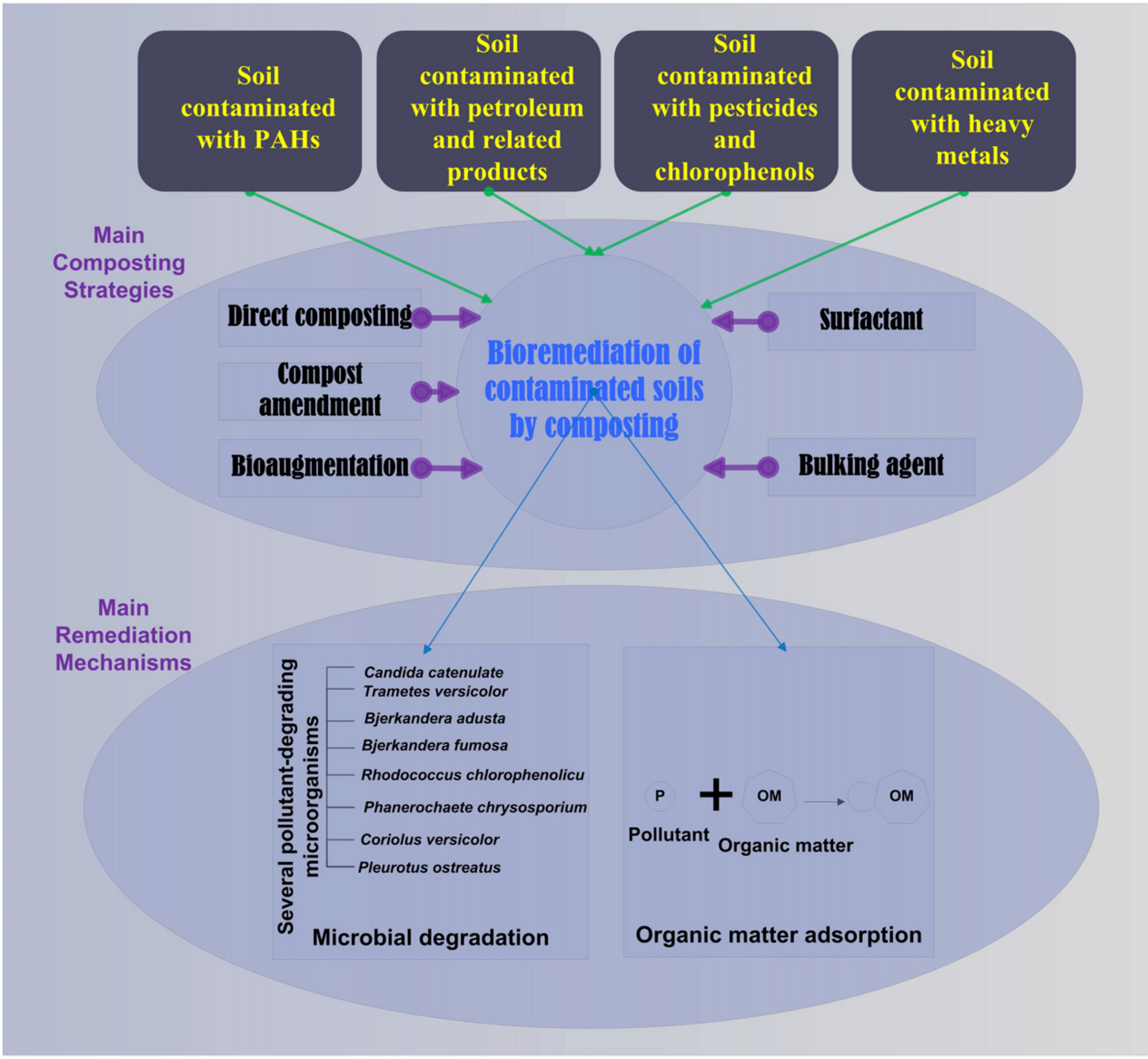

**Fig. 1.** Main strategies and mechanisms for bioremediation of contaminated soils by composting or compost.

Organic amendments from compost are an important source of nutrients, which provide more available carbon sources for indigenous microbes. In addition, organic amendments from compost also directly increase the density of microbes that are responsible for the decomposition and biotransformation of pollutants in soils (Namkoong et al., 2002). Reviewing microbial aspects according to found pollutants in soil/compost mixtures should contribute to broaden our knowledge of microbial community dynamics and pollutants' biodegradation process, enabling one to select potentially suitable microorganisms as amendments to promote microbial activity and pollutant removal.

This review focuses on the application of composting and compost for bioremediation of soils contaminated with organic pollutants and heavy metals (Fig. 2). The present paper also describes the microbial aspects related to this technology in soils. Moreover, other problems such as bioavailability, organic matter mineralization and compost quality are also discussed.

## 2. Composting application

### 2.1. Bioremediation of soils contaminated with organic pollutants

Composting is a promising technology for soil bioremediation due to its advantages over physical and chemical technologies. Composting and mature compost have been successfully applied to the bioremediation of contaminated soils with PAHs, pesticides, petroleum and other pollutants by providing a degrading matrix, available nutrients, and large numbers of active microorganisms (Jaspers et al., 2002; Scelza et al., 2008; Semple et al., 2001).

#### 2.1.1. PAHs

PAHs are known as one of the most widespread organic pollutants in soils due to natural or anthropogenic activities (Ortega-Calvo et al., 2013; Puglisi et al., 2007; Sayara et al., 2011). They are carcinogenic and mutagenic (Rivas, 2006). PAHs, being composed of fused benzene rings, persist in soils because of unique physical properties and high resistance to nucleophilic attack (Zhang et al., 2006). Antizar-Ladislao et al. (2005) investigated the biodegradation of 16 USEPA-listed PAHs from industrial site by in-vessel composting in different types of reactors. Using activation energy value ($E_a$), the authors concluded that the PAH removal mechanism was biological in the standard-composting reactors ($E_a = -6.43$ kJ mol$^{-1}$, $R^2 = 0.99$), whereas chemical reactions caused the removal of PAHs in the soil reactors. The addition of SS compost into PAH-contaminated soil led to an almost complete decomposition of anthracene and pyrene but a weak removal of benzo[a]pyrene after 15 months (Hamdi et al., 2007). As a model substrate of PAHs, phenanthrene is ubiquitously distributed in the environment. Toxic phenanthrene tends to highly bioaccumulate in organisms (Shailaja and Rodrigues, 2003). The interaction between phenanthrene and tricyclazole in medium, soil and the mixture of soil and compost was investigated, showing a negative effect on their degradation (Liu et al., 2008). The authors further attributed this negative effect to their molecular similarity. Molecular similarity of tricyclazole to phenanthrene may cause their competition interacting with the active site of phenanthrene dioxygenase. Interestingly, the inhibited effect of tricyclazole on phenanthrene decomposition could be reduced or eliminated in the soil and compost mixture due to the high microbial density in relation to phenanthrene degradation (Liu et al., 2008). Ma et al. (2003) tested the bioremediation effect on anthracene-contaminated soil by composting, finding that the removal percentages of anthracene for composting materials with old compost and without old compost were 55.3% and 50.5%, respectively. Pig manure which is able to improve the number of microbial population, organic matter and dissolved organic carbon content, and Tween 80 that can increase the pyrene bioavailability by acting as a surfactant, had a beneficial effect on pyrene removal, but none of them could increase the phenanthrene removal in soil vegetated with *Agropyron elongatum* (Cheng et al., 2008). The highest percentage removal was 92% in soil amended with 7.5% pig manure, and 79% in soil amended with 100 mg kg$^{-1}$ Tween 80, respectively. Experiments with different ratios of contaminated soil to green waste (0.6:1, 0.7:1, 0.8:1, 0.9:1 and 1.0:0) and moisture content (MC) (40%, 60% and 80%) were conducted to detect the optimal experimental conditions for maximum PAH removal (Antizar-Ladislao et al., 2008). PAH removal in reactors without green waste was significantly lower than that with green waste at 38 °C. However, this significant difference disappeared at 70 °C. High temperature was considered adverse to microbial activity, and volatilization was the leading mechanism of PAH

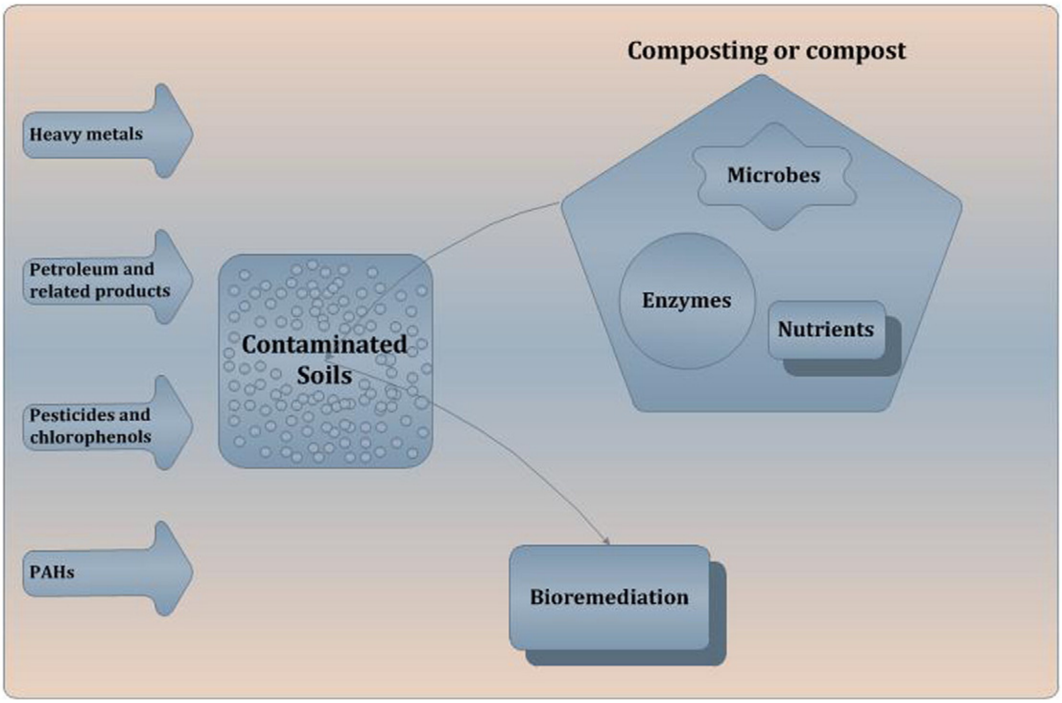

**Fig. 2.** Bioremediation of soils contaminated with organic pollutants and heavy metals by composting or compost.

removal (Antizar-Ladislao et al., 2008). PAH removal in different conditions at 38 °C was found as follows: 76.4% removal with soil to green waste ratio = 0.6:1 and MC = 60%; 82.0% with soil to green waste ratio = 0.7:1 and MC = 60%; 76.7% with soil to green waste ratio = 0.8:1 and MC = 60%; 69.1% with soil to green waste ratio = 0.9:1 and MC = 60%; 68.4% with soil to green waste ratio = 0.8:1 and MC = 80%; 39.2% with soil to green waste ratio = 0.8:1 and MC = 40%; and 9.5% with soil to green waste ratio = 1.0:1 and MC = 0% (Antizar-Ladislao et al., 2008).

Organic matter and soil structure affect the bioavailability of organic xenobiotics. Compost incorporated into contaminated soil is capable of significantly reducing bioavailability of phenanthrene and increasing the remediation rate of phenanthrene by either trapping of organic matter from compost or degrading microorganisms (Puglisi et al., 2007). Soil organic matter is capable of enhancing the adsorption of pollutions and reducing the bioavailability of pollutants, leading to decreased fraction available for microorganism-mediated degradation. However, the water-extractable organic matter (WEOM) from cow manure compost was observed to increase the apparent solubility of phenanthrene, pyrene and benzo[a]pyrene with 8.4, 34 and 89 times higher than their measured water values, respectively, thus promoting their biodegradation (Kobayashi et al., 2009). It was confirmed that the increase of PAH solubility and biodegradation was attributed to the high molecular weight fraction of WEOM (>1000 Da).

Decomposition of organic contaminants in soil is often difficult due to low bioavailability. Incorporation of surfactants into soil can increase the bioavailability of some organic pollutants (Cheng et al., 2008). Wu et al. (2013) explored the impact of compost amendment on the removal and bioavailability of PAHs in soils contaminated with diesel, coal tar and coal ash, finding that over 90% of total PAHs disappeared in all analyzed soils. This disappearance was mainly attributed to degradation and desorption, and degradation exerted a more important role in PAH loss than desorption. In the study, the type and rate of compost did not significantly affect PAH bioavailability, but the bioavailability of PAH with more benzene rings was lower than that with less rings. Plaza et al. (2009) reported that the composting process caused a structural conversion of humic acids from an organic substrate by reducing the aliphatic fraction and increasing polarity and aromatic polycondensation in a PAH-contaminated soil. This conversion decreased the PAH binding affinity of humic acids, and thus improved PAH-degrading microbial accessibility. It has been well known that addition of humic acids accelerated biodegradation of phenanthrene (Ortega-Calvo and Saiz-Jimenez, 1998). Composted materials blended with phenanthrene-contaminated soil could aid in the reduction of phenanthrene (Puglisi et al., 2007).

### *2.1.2. Pesticides and chlorophenols*

Pesticides and chlorophenols entering the soil can bring environmental hazards, and influence soil properties involved in biochemical and microbial aspects. Thus, bioremediation of soils contaminated with pesticides and chlorophenols has drawn considerable research interest.

Polychlorinated dibenzo-p-dioxins, dibenzofurans (PCDD/Fs) and chlorophenols are important components of wood preservative KY-5 that was extensively used in Finland, and thus they usually simultaneously appear in soils. Although chlorophenols can be effectively degraded during composting of contaminated sawmill soil, large amounts of PCDD/Fs were not significantly decomposed (Laine et al., 1997). More than 90% of the chlorophenols disappeared in composting pile composed of straw compost and chlorophenol-contaminated soil (Laine and Jørgensen, 1997). Pentachlorophenol (PCP), a highly chlorinated organic pollutant with stable aromatic ring structure is persistent in the soil (Scelza et al., 2008). The percent removal of PCP was 76% in contaminated soil amended with MSWC as a supplemental nutrient in a laboratory-scale study (Miller et al., 2004). The treatment with compost provoked the enlargement of bacterial and fungal populations. Scelza et al. (2008) performed a laboratory-scale study to explore the impact of PCP contamination and incorporation of dissolved organic matter (DOM) or compost on an agricultural soil. The PCP removal was found to be most pronounced for the soils with 0.27% of compost, and with 0.07% or 0.2% of DOM. Straw compost and remediated soil have been applied to treat chlorophenol-contaminated soils, showing that 56% of the [$^{14}$C]PCP was mineralized without harmful side reactions (Laine and Jorgensen, 1996). Several organic amendments including compost, corn fermentation byproduct, corn stalks, manure, peat and sawdust have been used to improve the herbicide removal of atrazine, trifluralin and metolachlor in contaminated soils (Moorman et al., 2001). The incorporation of 0.5% manure, 5% cornstalk and 5% peat in soils enhanced the atrazine removal, and the addition of sawdust, corn fermentation byproduct, manure and cornstalk at a rate of 5% increased the metolachlor degradation (Moorman et al., 2001). However, all amendments proved to be ineffective in enhancing the trifluralin degradation. Another study done by Delgado-Moreno and Peña (2009) has found that the addition of compost, vermicompost and olive cake in soils did not increase the overall removal of simazine, cyanazine, terbuthylazine and prometryn. Employing composted manure and biosolid manure, Gan et al. (1998) enhanced the degradation of methyl isothiocyanate (MITC) and methyl bromide (MeBr) from soils, and thus reduced the emission or volatilization of MITC and MeBr by almost 100% and 12%, respectively. Noteworthy, comparison of degradation kinetics between nonsterile and sterile amended soils indicated that the degradation mechanism of these two compounds differed. MITC degradation mainly relied on soil microorganisms, while MeBr decomposition was mainly achieved chemically. Kadian et al. (2008) investigated the effect of different amendments (mushroom spent compost, biogas slurry, farmyard manure and sodium citrate) on the removal of atrazine in contaminated soils. The addition of biogas slurry showed the highest atrazine removal (34.14%), followed by a combination of sodium citrate and farmyard manure (31.8%), mushroom spent (29.17%) and farmyard manure (22.07%). Two organic amendments (MSWC and composted straw) have been applied to bioremediate atrazine-contaminated soil during laboratory incubations (Houot et al., 1998). The last residual atrazine concentrations were higher in soils treated with these two organic amendments, due to the fact that both amendments promoted the production of bound residues of atrazine. MSWC accelerated the atrazine sorption, and thus reduced its bioavailability for its degrading microorganisms, while composted straw was correlated with the formation of hydroxyatrazine (Houot et al., 1998).

### *2.1.3. Petroleum and related products*

Human activities and emergencies (for example oil spills) lead to release of large amounts of petroleum and petroleum hydrocarbons into the environment (Khamforoush et al., 2013; Zhu et al., 2001). The unexpected petroleum from anthropogenic factor could persist in soils, and destroy the ecosystem. Fortunately, an eco-friendly technology, known as composting technology, has been successfully used for bioremediation of petroleum hydrocarbon-contaminated soil. The influence of organic amendments on bioremediation of diesel-contaminated soil has been revealed. There was evidence that incorporation of organic amendment from SS or compost into contaminated soil significantly enhanced the degradation of petroleum hydrocarbons and n-alkanes with the optimal soil/amendment ratio of 1:0.5 (Namkoong et al., 2002). Bioremediation of Tarpley clay soil contaminated with petroleum hydrocarbons was carried out with biosolid addition, supplement of organic fertilizers and monitored natural attenuation (Sarkar et al., 2005). After incubation for 8 weeks, approximately 96% and 93.8% of total petroleum hydrocarbon were removed from the soils treated with biostimulation methods and monitored natural attenuation, respectively. Mineral nutrient, sawdust, hay and compost were selected as amendments to remediate heavy mineral oil contaminated soils, resulting in decontamination percentages of 18–40% (Lee et al., 2008). However, only 9% of the initial hydrocarbon disappeared in unamended soils. A mathematical modeling relying on the reaction kinetics, mass and energy balances, the Haug's model (Haug, 1993), has been introduced to predict the effect of composting on remediation of petroleum hydrocarbon-contaminated soil (Khamforoush et al., 2013). The predicted results are comparable with experimental data, validating the reliability of this model. Khamforoush et al. (2013) further investigated the impact of wood chips (a kind of bulking agent), amendment types (food waste, yard waste, biowaste and sludge), the ratio of amendment/bulking agent to soil and experimental conditions on the soil bioremediation. The optimal remediation was observed when bulking agent/soil ratio, amendment/soil ratio, airflow and initial moisture content were 2.25 kg $kg^{-1}$, 2.5 kg $kg^{-1}$, 0.520 $m^3$ $day^{-1}$ $kgBVS^{hnu}$ and 62.5%, respectively. The composting mixture consisting of soil spiked with diesel oil and biowaste (e.g. garden waste, fruit and vegetable) had a greater first-order rate constant of degradation for diesel than the soils at room temperature (4-fold) and at composting temperature (1.2-fold) (Van Gestel et al., 2003).

Sometimes, compost addition in contaminated soils may be inefficient in enhancing pollutant degradation due to decreased bioavailability caused by organic matter adsorption, slow composting process and excessive nutrients (Gallego et al., 2001; Ros et al., 2010).

### *2.2. Biorestoration of metal contaminated soils*

Heavy metal contamination in post-industrial sites such as mine soils is a worldwide environmental issue warranting consideration. How to solve this issue is a great challenge. The fate and behavior of heavy metals in soils are governed by several reactions, including precipitation, adsorption, complexation, methylation, demethylation, oxidation, reduction, and so on (Park et al., 2011). In general, remediation of metal-contaminated soils can be carried out by "ex situ" techniques (e.g. extraction) and "in situ" techniques (e.g. stabilization) (Mora et al., 2005). Bioremediation of heavy metal contaminated soils can be achieved by adding compost (a type of stabilization technique) which is capable of complexing, absorbing and (co)precipitating heavy metals (Burgos et al., 2010; Park et al., 2011). The mechanisms for the enhancement of remediation of metal-contaminated soils by organic amendments have been reviewed, and include immobilization and reduction (Park et al., 2011). Heavy metal fractions by the sequential extraction procedure include water soluble fraction, residual fraction, organically bound fraction, carbonate bound fraction, exchangeable fraction and Fe and Mn oxide bound fraction (Achiba et al., 2009; Illera et al., 2000; Tessier et al., 1979). Somewhat differently, Planquart et al. (1999) divided heavy metal fractions into five fractions: exchangeable fraction, acid-soluble fraction, humic fraction, reducible fraction and residual fraction. Heavy metals often behave differently in various soils. It is difficult to assess the metal pollution levels by determining the metal concentration in soils. Several factors affected the distribution of heavy metals in soils, including physico-chemical properties of soils, redox potential, ligand, and so on (Achiba et al., 2009; Kabala and Singh, 2001; Narwal et al., 1999). Thus, many studies focused on the fractionation of heavy metals associated with their mobility and availability.

The effect of compost addition on the remediation of metal-contaminated soils depended on the compost types, pollution level and soil types in the practical application (van Herwijnen et al., 2007). An investigation by Farrell and Jones (2010) into the effect of several types of composts on the remediation of metal-contaminated soil found that all types of composts decreased the levels of heavy metals in soil solution. Pérez-de-Mora et al. (2006) stated that compost application increased soil pH, and reduced the solubility of heavy metals. These authors thought that soil pH was the most important factor for the reduction of heavy metal solubility.

Comparison between co-composting of metal polluted mine soil with organic wastes and addition of mature compost into mine soil contaminated with Cu, Pb, As and Zn, showed that these two methods have a very similar effect on remediation of soil polluted by heavy metals (Tandy et al., 2009). The application of MSWC and manure changed the soil features (pH, organic matter content, electrical conductivity and the total nitrogen content) and distribution of heavy metals (Achiba et al., 2009). This study indicated that the distribution of heavy metals (Cd, Cu, Pb, Zn, Cr and Ni) in amended soils exhibited a similar pattern: the residual fraction was predominant, followed by Fe and Mn bound fraction. Another study done by Pérez et al. (2007) showed that Fe, Mn, Cd and Ni were mainly present in the residual fraction in soils amended with 0.0, 12.5, 25.0, 50.0 and 100.0 t/ha of compost, respectively. By contrast, the majority of Cu existed in the organically bound fraction. Municipal waste compost (MWC) and biosolid compost (BC) as amendments improved the soil pH, hydrosoluble carbon and the content of organic carbon, and reduced the concentrations of soluble Cd, Cu and Zn (Mora et al., 2005).

The effects of compost addition on heavy metal distribution in the neutral soil and the acidic soil were different (Alburquerque et al., 2011). In the neutral soil, compost addition mainly led to increase of the NaOH-extractable Fe fraction, and decrease of EDTA-extractable Zn and Cu fraction. Mn species was almost not affected by compost application with >99% of the total Mn belonging to the residual fraction in the neutral soil. In the acidic soil, the NaOH-extractable, EDTA-extractable and residual Zn fractions were significantly increased after compost addition. $CaCl_2$-extractable Cu fraction in the unamended soil was significantly higher than in compost-treated soil. Variation in soil pH and the interaction between organic matter/compound and heavy metals were believed to be responsible for the change in heavy metal distribution in these two types of soils (Alburquerque et al., 2011; Clemente and Bernal, 2006). The pH was identified as a critical factor controlling the mobility and availability of Zn in amended soils (Illera et al., 2000).

A combination of three amendments including compost, lime and phosphate resulted in a decreased phytoavailability of Pb and Mn (Padmavathiamma and Li, 2010). This combination reduced the exchangeable Pb in soil for *Poa pratensis* L., and Mn in soil for *Lolium perenne* L. (Padmavathiamma and Li, 2010). Compost and SS application caused an increase of labile Zn and Pb in soil, suggesting that these organic amendments have a potential to increase metal mobility in soil (Santos et al., 2010). Clemente et al. (2006) determined the heavy metal fractionation in contaminated soil from the mining area at La Unión (Murcia, Spain) amended with mature compost. Compost showed a positive impact on the immobilization of Zn and Pb, but enhanced the Cu solubility due to organic matter chelation. Humic acids obtained from mature compost resulted in significant Zn and Pb fixation, and slight Cu and Fe mobility in soil (Clemente and Bernal, 2006). The effect of compost and pig slurry as amendments on heavy metal solubility has been evaluated in soils from a mining area located in Spain (Pardo et al., 2011). These organic amendments raised the EDTA-extractable concentrations of Zn and Pb. Role of humic acids from BC in Cu bioavailability was assessed (Soler-Rovira et al., 2010). The Cu(II) complexing capacity of humic acids increased as the humification degree, aromaticity and COOH groups increased. The bioavailability and solubility of Cu were largely affected by the pH of soil/amendment system, followed by soil organic matter and humic acid fraction (Soler-Rovira et al., 2010). Composting by solid olive husk is a more appropriate and safe method for bioremediation of metal-contaminated soil than that using fresh olive husk (de la Fuente et al., 2011). Soil organic matter from the compost did not induce significant effects on the change of heavy metal solubility, while fresh olive husk could improve Mn bioavailability and thus bring phytotoxicity.

There has been effort to reduce the availability of Cd, Cu, Mn and Zn by incorporating BC, sugarbeet lime (SL) and fresh "alperujo" in acid soil (Burgos et al., 2010). The maximum reduction of these heavy metals was found in acid soils treated with BC and SL. Increase in pH was considered to be the major reason for this reduction, based on the fact that most heavy metals are less available in alkaline environment (Burgos et al., 2010). However, in neutral soil, the presence of these amendments almost did not result in changes in pH and content of extractable heavy metals. Lagomarsino et al. (2011) demonstrated that chemical speciation of Cu was diverse in soil: acid-soluble fraction (34%), the fraction bound to organic matter (12%), reducible fraction (32%), residual fraction (16%), and soluble and exchangeable fraction (4%). The application of dolomitic limestone and compost increased the fraction bound to organic matter by 3-fold as compared to untreated soil, and reduced the exchangeable Cu fraction, but almost had no effect on residual, reducible and acid-soluble fractions. The reduction of exchangeable Cu fraction caused by these amendments was attributed to Cu precipitation and complexation. Efforts have been made to maximize the bioremediation effects of composting or compost amendments by adding biochar. It has been reported that both green waste and biochar amendments increased Cu concentration in soil pore water 30 times or more, but significantly reduced Cd and Zn concentrations (Beesley et al., 2010). van Herwijnen et al. (2007) suggested that heavy metals could form complexes with organic matter, being responsible for metal immobilization and bioavailability. The authors indicated that green waste compost application decreased Zn leaching in metal-polluted soil. Contrastly, composted SS improved Zn leaching. However, the use of composts in metal-polluted soil could not provide a permanent immobilization of metals because of the natural occurrence of organic matter degradation. It was found that the percent of acid soluble Zn was increased by the application of compost (Planquart et al., 1999). Solubility and mobility of Pb were low in the soil, regardless of whether the soil was neutral or acidic (Alburquerque et al., 2011). This situation was not changed significantly by compost addition. The formation of insoluble organic matter–Pb complexes, variation in soil pH, mineralization and high salt content of compost jointly favored the Pb immobilization (Alburquerque et al., 2011; Castaldi et al., 2005; Walker et al., 2003). Bhattacharyya et al. (2008) added MSWC into submerged rice paddies, and detected the Co and Ni fractions in MSWC using a sequential extraction method. It was showed that Co and Ni were significantly bound to several components in MSWC including Fe and Mn oxides and organic matter.

Winter wheat (*Triticum aestivum* L.) potted soils treated with 0–50 mg $kg^{-1}$ of Cd were amended with 0–120 mg $kg^{-1}$ of compost to explore the influence of compost application on the immobilization and phytotoxicity of Cd in metal-contaminated soils, leading to 71.8–95.7% decrease of soluble/exchangeable Cd and 0.4–18.4 times increase of organic-bound and inorganic precipitates Cd (Liu et al., 2009). Redistribution of Cd species alleviated the Cd phytotoxicity, and increased the seed yield and growth of wheat. Three factors for the variation in Cd species after the compost application were discussed by Liu et al. (2009), including organic matter, soil pH and P content. Increase of organic matter, soil pH and P content after the addition of compost lowered the bioavailability of heavy metals by improving the formation of stable complexes with humic substances, the adsorption on soil particles and the occurrence of insoluble precipitates, respectively (Liu et al., 2009; Sauve et al., 2000; Shuman, 1999; Walker et al., 2003). Composted pine bark (PB) with organic matter content = 81% and a pH of 5.6, composted pruning waste and biosolids (BS) with organic matter content = 47% and pH = 6.9, and spent mushroom compost (SM) with organic matter content = 75% and a pH of 8.0 were treated using 80 and 200 mg $kg^{-1}$ of Cd during 4 weeks, exhibited different abilities to immobilize Cd (Tapia et al., 2010). The total Cd extracted from the compost was 0.2% for the BS, 4.0% for the PB and 0.7% for the SM, supporting that BS had greater capacity to immobilize Cd than PB and SM (Tapia et al., 2010). Tejada (2009) recorded a lower percentage of inhibition in soil enzymes in Cd contaminated soils amended with crushed cotton gin compost (CC) than in Cd contaminated soils amended with SS, poultry manure (PM) and organic MSW. The author explained that there was a higher humic acid concentration in CC-amended soils than in other soils. Humic substances containing a variety of carboxyl, alcohol, phenolic and carbonyl could bind to heavy metals, form the metal–humate complexes, resulting in that the bioavailability of Cd dropped (Dar, 1996; Datta et al., 2001; Tejada, 2009). CC with higher humic acid content than other organic amendments was thus more beneficial to bioremediation of Cd contaminated soils.

## 3. Microbes in contaminated soils treated by composting or compost

Microbial community dynamics and microbial interactions in contaminated soils treated by composting or compost were shown in Fig. 3.

### 3.1. Microbial aspects related to PAH-contaminated soils

Microbial community composition is dynamic during composting. Microbial community composition shifts during in-vessel composting of an aged coal-tar soil polluted with PAHs were explored using phospholipid fatty acid (PLFA) analysis (Antizar-Ladislao et al., 2008). The results showed that temperature significantly influenced fungal to bacterial PLFA ratios and Gram-positive to Gram-negative bacterial ratios. The extent of PAH losses was associated with the Gram-positive to Gram-negative bacterial ratios ($P < 0.005$) at 70 °C. Some microorganisms which can transform toxic contaminants into nutrients for assimilation would become the dominant species (MacNaughton et al., 1999; G.M. Zeng et al., 2011). The predominance of bacterial community changed during composting of pyrene-contaminated soil. Peng et al. (2013) performed an investigation on the dynamics of bacterial community in an in-vessel composting bioremediation of soils contaminated with $^{13}C_{4-4,5,9,10}$-pyrene by DNA-based stable isotope probing (SIP), noticing that dominant bacterial communities related to pyrene decomposition were α-, β-, γ-Proteobacteria, and Actinobacteria at 38 °C during composting of 14 days, and *Streptomyces* at 55 °C, respectively. Besides, after 42 days of composting, *Ralstonia*, *Acinetobacter* and *Thermobifida* were predominant at 70 °C, while after 60 days of composting, *Thermobifida* and *Streptomyces* dominated. Ma et al. (2003) performed microbial enumeration and assessed microbial diversity by the Shannon–Weaver index during composting of anthracene-contaminated soil mixed with kitchen waste. They found that mesophilic bacteria dominated at the beginning, thermophilic bacteria and thermophilic actinomycetes became predominant at the stage of high temperature. Microbial diversity drastically raised with Shannon–Weaver index increasing from 0.52 at 38 °C to 2.13 at 56 °C in composting materials inoculated with old compost and from 0.18 at 38 °C to 1.49 at 56 °C in composting materials without old compost, respectively (Ma et al., 2003). In a study done by Gandolfi et al. (2010), it proved that compost amendments caused a complete change of predominant microbial community composition from Alpha- and Gammaproteobacteria to Bacteroidetes and Firmicutes. Interestingly, at the end of this experiment, Bacteroidetes became the only component of microbial community. The use of vermicompost from olive-mill wastes for bioremediation of PAH-contaminated soils proved to be effective for removal of pollutants. Based on the Shannon ($H$) diversity index, Di Gennaro et al. (2009) observed that vermicompost addition in PAH-contaminated soil increased the microbial diversity after 7 days and declined at time 30 days. They further concluded that the introduction of vermicompost stimulated the metabolically activity of microbial community by inducing the expression of biodegradation indicator genes in native microorganisms and supplementing new PAH-degrading microbes.

To date, many investigations of PAH decomposition by fungi and bacteria have been performed in contaminated soil composting system. It was suggested that in addition to microbial degradation, PCP adsorption mediated by *Phanerochaete chrysosporium* mycelia was a factor governing the removal of PCP according to the studies of our group and other groups (Logan et al., 1994; Shim and Kawamoto, 2002; Yu et al., 2011). The presence of *Trametes versicolor* was capable of enhancing PCP removal (Walter et al., 2005). A consortium of three basidiomycetes from compost which we believed to be *T. versicolor*, *Bjerkandera adusta*, *Bjerkandera fumosa* and/or *Lopharia spadicea* on the basis of rDNA sequencing and sequence comparison was assessed for pyrene removal, achieving a pyrene removal of about 56% in soil (Anastasi et al., 2009). Kobayashi et al. (2009) have discussed the role of WEOM from cow manure compost in PAH degradation mediated by *Sphingomonas* sp. AJ1, showing WEOM was helpful in enhancing the PAH biodegradation by

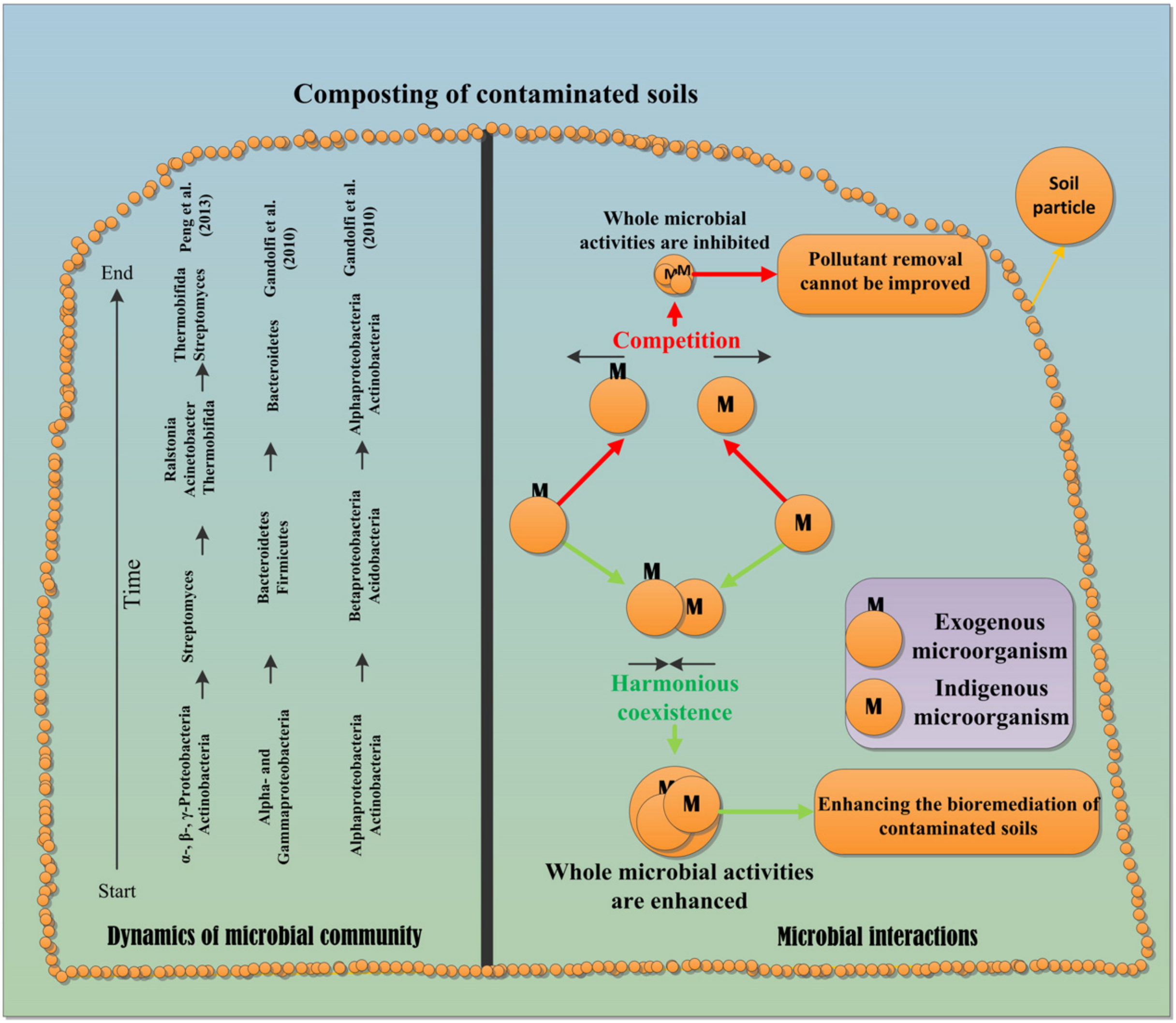

**Fig. 3.** Dynamics of microbial community and microbial interactions in contaminated soils treated by composting or compost.

increasing PAH solubility. Liu et al. (2008) found that the growth of phenanthrene-degrading microorganism *Sphingomonas paucimobilis* ZX4 could be inhibited by tricyclazole. Increasing concentration of tricyclazole would enhance the inhibition on the growth of *S. paucimobilis* ZX4 and decomposition of phenanthrene.

Bioaugmentation was often used to accelerate the decomposition of pollutants. However, the inoculants of foreign microorganisms were not always effective in enhancing the PAH degradation during composting. Canet et al. (2001) achieved a conclusion that fungal inoculation including four well-known PAH-degrading microorganisms (*P. chrysosporium* IMI 232175, *Coriolus versicolor* IMI 210866, *Pleurotus ostreatus* IMI 341687 and Wye isolate #7) in the mixture composed of non-sterile coal-tar contaminated soil and wheat straw was unsuccessful to improve the PAH removal. By contrast, native microorganisms could utilize wheat straw as organic substrate, being the main degraders for the PAH by their metabolism. The foreign microorganisms did not die during this experiment but could not serve an effective role in PAH degradation due to the fact that they were still in a metabolically inactive form. The authors speculated that the heavily contaminated environment and experimental conditions might be the reasons for the inhibition of foreign microbial activity (Canet et al., 2001). Similarly, inoculation with *P. chrysosporium* in soil composting system was previously confirmed to be ineffective in significantly improving the benzo(a)pyrene removal during 95 days (McFarland and Qiu, 1995). Another study from Sayara et al. (2011) reported that the introduction of the white-rot fungi *T. versicolor* ATCC 42530 did not significantly improve the decomposition of PAHs, but the additions of compost and rabbit food led to 89% and 71% removal of the total PAHs after 30 days, respectively, as compared to only 29.5% removal in the unamended soils. The bioaugmentation process can be affected by several factors such as physico-chemical properties of pollutants and soils, the competition between native and foreign microorganisms for nutrients, their antagonistic interactions, and death caused by protozoa and bacteriophages (Mrozik and Piotrowska-Seget, 2010). It was also found that biodiversity of indigenous microorganisms could act as a barrier to the invasion of exogenous microorganisms (Kennedy et al., 2002). Combination of these factors determined the removal effect of pollutants in composting of contaminated soils.

### *3.2. Microbes associated with soils polluted by petroleum and related products*

Single microorganism or microbial community played a key role in hydrocarbon elimination, and impacted the stability of composting. Soil contaminants often have an adverse effect on soil microorganisms. Gasoline exhibited a toxic effect on microorganisms in soils, resulting in a dynamic change of soil microbial population. Fortunately, the application of PM, CC, organic MSW and SS could decrease the inhibited effect of gasoline on the soil microorganisms (Tejada et al., 2008). Compost addition increased the total number of heterotrophic microorganisms in soil contaminated with diesel oil (Gandolfi et al., 2010). 16S rDNA sequence analysis showed that compost addition led to a change of dominant microbial community from Alphaproteobacteria and Actinobacteria to Betaproteobacteria and Acidobacteria in this contaminated soil. As the treatment process continued, the number of Alphaproteobacteria and Actinobacteria increased (Gandolfi et al., 2010). However, Ros et al. (2010) reported that compost addition did not significantly increase the level of microbial populations in an aged recalcitrant hydrocarbon contaminated soil, because this level was similar between compost-amended soil and unamended soil. The denaturing gradient gel electrophoresis (DGGE) analysis indicated that the phylum Actinobacteria composed of Actinomycetales was most predominant in compost-amended soils, including Arthrobacter, Microbacterium, Nocardia and Mycobacterium species (Ros et al., 2010).

Baheri and Meysami (2002) assessed the feasibility of fungi bioaugmentation during composting of total petroleum hydrocarbon contaminated soils, and tried to determine the best strain for the removal of total petroleum hydrocarbon. Then, they selected *B. adusta* BOS 55 as the promising strain for their experiments. The density of indigenous microorganisms responsible for the degradation of petroleum hydrocarbon in soil is insufficient in some cases. To address this problem, *Candida catenulate* CM1, a petroleum-degrading microorganism, was used as amendments during composting of soil contaminated with 2% (w/w) diesel (Joo et al., 2008). Inoculation with *C. catenulate* CM1 exhibited a higher removal of petroleum hydrocarbon compared with that without inoculation (84% vs. 48%). Re-inoculation with microorganisms isolated from the mazut-contaminated soil and the supply of nutritional elements increased the number of hydrocarbon-degrading microorganisms with more than 20 times the untreated soil (Beskoski et al., 2011). This treatment also led to a decrease of petroleum hydrocarbon content from 5.2 g $kg^{-1}$ to 0.3 g $kg^{-1}$. However, the percent removal of total petroleum hydrocarbon was only 10% in an untreated control pile (Beskoski et al., 2011). Bioaugmentation with B-2-2 strain belonging to the genus *Acinetobacter* in a hydrocarbon-contaminated antarctic soil enhanced the bioremediation efficiency with a hydrocarbon removal of 75% (Ruberto et al., 2003). Similarly, the presence of the microbial consortium comprising six bacteria species also enhanced the biodegradation of heavy crude oil in soils amended with sugar cane bagasse wastes and spent compost (Trejo-Hernandez et al., 2007). These bacteria species belong to *Bacillus*, *Pseudomonas*, *Klebsiella* and *Serratia* genera. Bioremediation of diesel-contaminated soils from Long Beach (California, USA) and Hong Kong (China) by bioaugmentation, biostimulation and natural attenuation after 12 weeks was compared (Bento et al., 2005). The maximum degradations found in Long Beach soil treated with bioaugmentation in the light (C12–C23) and heavy (C23–C40) fractions of total petroleum hydrocarbon were 72.7% and 75.2%, respectively. Bioaugmentation of the Long Beach soil and natural attenuation of the Hong Kong soil showed the highest microbial activity in the form of dehydrogenase activity, reaching 3.3 fold and 4.0 fold, respectively (Bento et al., 2005).

### 3.3. Microbes in soils contaminated with pesticides and chlorophenol

Soil microorganisms and exogenous microorganisms are responsible for the degradation of pesticides in soils during the composting process. The presence of pesticides affected the population and activity of microbes in contaminated soil with composting. The interaction between pollutant and microbes also influences the effect of composting on soil remediation. Pollutants' properties such as toxicity affect the biodegradation of some pollutants. The undesirable properties of pollutants are adverse for the growth of microorganisms, interfering with their normal metabolic function. Our group conducted PCR-DGGE to explore the dynamics of microbial community during composting of PCP-contaminated soils, finding that microbial abundance was inhibited by PCP stress (G.M. Zeng et al., 2011). In a study carried out by Scelza et al. (2008), the soil microflora was difficult to recover from its inhibitory state induced by toxicity of PCP. Composting exhibited a strong effect on the removal of chlorophenol with a degradation rate about 85.85% (decomposed from 212 mg $kg^{-1}$ to 30 mg $kg^{-1}$) in four summer months; the second summer of composting resulted in that the concentration of chlorophenols became only 15 mg/kg (Valo and Salkinoja-Salonen, 1986). Bacterial populations which occurred in compost-, manure- and cornstalk-amended soils at a rate of 5% were increased compared to nonamended soils, but these amendments had no effect on fungal or actinomycete populations (Moorman et al., 2001).

The addition of pollutant-degrading microorganisms can improve the degrading progress, although there have been large numbers of microbial degraders in composting of contaminated soil. In the presence of *Rhodococcus chlorophenolicus* (a microorganism that can degrade several types of chlorophenols) as amendment, the decomposition of chlorophenols was faster than that without this microorganism inoculated (Valo and Salkinoja-Salonen, 1986). *P. chrysosporium*, known as a type of basidiomycete, exhibited high degradation ability to lignin, PCP, and so on by secreting lignin peroxidase (LiP) and manganese peroxidase (MnP) (Chen et al., 2011; Martinez et al., 2005; Yu et al., 2011). Thus, *P. chrysosporium* inoculants could increase lignocellulose biodegradability and ultimately improve the quality of compost products. Our group showed that inoculation with *P. chrysosporium* had a strong positive effect, both on the composting efficiency and the removal of PCP (Yu et al., 2011). In the study, our group also observed that inoculation time influenced the removal of PCP in soil based on composting, because the inoculation on the fifteenth day of composting had a better effect on PCP removal than that from the start. We cannot neglect the positive effect of microorganism addition on remediation of polluted soil with composting.

Noteworthy, mineralization of pollutants in soil by microbes in composting also may bring ecotoxicological risks to soil environment, if incomplete pollutant mineralization occurs (Laine and Jørgensen, 1997). As suggested by Laine and Jørgensen (1997), safe, effective and fast composting methods with complete mineralization needed to be constructed. White rot fungi have been shown to have a high capacity to remove various organic pollutants during composting. However, unfortunately, sometimes, white rot fungi also convert organic contaminants to harmful metabolites. For example, white rot fungi may transform chlorophenols into PCDD/Fs by peroxidase enzymes (Öberg and Rappe, 1992).

The application of composting technology as a remediation strategy for contaminated soil requires an understanding of microbes involved in pollutant biodegradation and biotransformation. The knowledge about the effect of more types of pollutant-degrading microorganisms as amendments during composting of contaminated soil is still poor despite the availability of various microorganisms.

### 3.4. Microbes in metal-contaminated soils

Heavy metals have a direct or indirect impact on microbes in metal-contaminated soils. Low concentrations of heavy metals are beneficial to microbial growth, but they would be toxic to microorganisms if they are in excessive concentrations (Giller et al., 1998). The population and activity of soil microbes were useful indicators reflecting the fluctuation of soil quality (improvement or degradation) (Pérez-de-Mora et al., 2006). The presence of heavy metals in soils influenced the key microbial processes such as nutrient cycling and organic matter transformation, and improved the microbial tolerance or resistance to heavy metals due to fast adaptation to environmental changes (Hassen et al., 1998; Obbard, 2001). Long-term exposure to heavy metals may increase the tolerance and resistance of microorganisms (Ellis et al., 2003). Significantly elevated levels of organic contaminants and heavy metals coexisted in agricultural and industrial soils, making it difficult to entirely remediate (Beesley et al., 2010). The use of composting or compost amendments has multiple benefits of improving the soil fertility and decreasing the bioavailability and toxicity of heavy metals. Industrial waste often is toxic and hazardous to human and soil microbes due to high concentrations of heavy metals in them. Tannery waste is one of them. Accordingly, a 50-day composting treatment could increase the removal of Cr, Pb, Cd, Cu and Zn in this waste, but significantly reduced the microbial density (Haroun et al., 2007). The organic matter of composts was capable of transforming free $Cu^{2+}$ into complexed $Cu^{2+}$ in the soil water, and thus contributed to a reduction of toxicity to bacteria (Clemente et al., 2003; Kiikkila et al., 2001; Kiikkilä et al., 2002). Accumulation of heavy metals in soils reduced the content of microbial biomass, limiting the functional diversity of ecosystem (Chander et al., 1995; Kandeler et al., 1996). Initially, the microbial accounts of total aerobic mesophile, aerobic bacilli and yeast and molds were $5 \times 10^6$, $8 \times 10^9$ and $8.3 \times 10^6$ CFU $g^{-1}$, respectively (Haroun et al., 2007). After 50 days, the microbial density became $<10$ CFU $g^{-1}$ of total aerobic mesophile, $<10$ CFU $g^{-1}$ of yeast and molds and $9.5 \times 10$ CFU $g^{-1}$ of aerobic bacilli. MWC and BC treatments in metal-contaminated soil significantly improved the average values of microbial biomass C with the highest microbial biomass carbon found in the soil amended with MWC (443.9 mg $kg^{-1}$) (Pérez-de-Mora et al., 2006). Average values of microbial biomass C are higher in soils amended with MWC and BC than in the control (Mora et al., 2005). SS addition led to a decrease in Cd toxicity to microbial activity in soils (Moreno et al., 2002). The application of organic amendments such as compost in metal-contaminated soils could cause the variation in the soil microbial population by changing pH, decreasing the solubility of heavy metals, and increasing allochthonous microbial biomass and available nutrients (Alburquerque et al., 2011). MWC and BC addition induced shifts in bacterial and fungal communities in metal-contaminated soils (Pérez-de-Mora et al., 2006). Composts may introduce new species that were lack in the contaminated soils, and further improved organic matter turnover, mycorrhizal symbionts and microbial functionality (Farrell and Jones, 2010; Pérez-de-Mora et al., 2006). It must be noted that only some studies are available concerning microbial aspects in metal-contaminated soils treated by composting or compost.

## 4. Conclusions and future research needs of composting for contaminated soils

Potential risks of soils contaminated with organic pollutants and heavy metals to human health and natural ecosystem have attracted high attention. Many methods have been developed to minimize these risks over the last few years. Nowadays, composting is believed to be one of the most-effective methods for simultaneously increasing waste disposal, soil organic matter content and soil fertility besides bioremediation. Composting is also a valuable technology to minimize the possible deleterious impact from the direct addition of organic wastes due to insufficient mature and a lack of stability.

The present composting approach for soil bioremediation suffers from the following limitations:

(1) Bioavailability. Bioavailability of a pollutant in soil is determined by its accessibility for adsorption, toxicity and utilization. Many organic compounds including various pesticides and PAHs enter the soil, and may be bound to soil in a strongly sorbed state. In long-term bioremediated soils contaminated by spiking with PAHs, the bioavailability of PAHs is a more limited factor for the bioremediation efficiency rather than the density of PAH-degrading microorganisms. Bioavailability of contaminants varies as time goes on. This process is called aging. The occurrence of aging brings a great challenge to the remediation of soil pollution. Thus, finding an appropriate method to investigate the bioavailability of a variety of pollutants in soil is potentially important before selecting a remediation technology for soil pollution control.

(2) Organic matter mineralization. Bioremediation of contaminated soils by the adsorption of organic matters from composting or compost is a promising and sustainable technology. However, as time went on, organic matter may be mineralized and soil pH would vary, leading to the reversion of pollutant stability.

(3) The poor adaptiveness of exogenous microorganisms in contaminated soils. This poor adaptiveness often results in that bioaugmentation is ineffective in enhancing the degradation of pollutants. Appropriate methods should be explored to solve this problem.

(4) The understanding of microbial progress during composting of soils contaminated with organic pollutants and heavy metals is still poor, although various pollutant-degrading microorganisms have been isolated and their degradation pathways have been well documented. A deeper understanding of microbial lifestyle and dynamics of communities found in soil/compost mixtures is thus necessary to further increase the effect of composting or compost on remediation of contaminated soils, using molecular biology such as high-throughput sequencing.

(5) Little is known about enzymatic aspects in contaminated soils remediated by composting or compost. Therefore, a detailed survey of enzymatic aspects in these soils is still necessary.

(6) The influence of interaction between pollutants from the same soil/compost mixture on their degradation is unclear. Further studies are still needed to address this issue.

(7) Compost quality. We must assure compost quality, because low quality composts often have high amounts of toxic organics and metals. High-quality composts require high organic matter content and low levels of heavy metals in corresponding materials.

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