The potential impact on the biodegradation of organic pollutants from composting technology for soil remediation

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Abstract

Large numbers of organic pollutants (OPs), such as polycyclic aromatic hydrocarbons, pesticides and petroleum, are discharged into soil, posing a huge threat to natural environment. Traditional chemical and physical remediation technologies are either incompetent or expensive, and may cause secondary pollution. The technology of soil composting or use of compost as soil amendment can utilize quantities of active microbes to degrade OPs with the help of available nutrients in the compost matrix. It is highly cost-effective for soil remediation. On the one hand, compost incorporated into contaminated soil is capable of increasing the organic matter content, which improves the soil environment and stimulates the metabolically activity of microbial community. On the other hand, the organic matter in composts would increase the adsorption of OPs and affect their bioavailability, leading to decreased fraction available for microorganism-mediated degradation. Some advanced instrumental analytical approaches developed in recent years may be adopted to expound this process. Therefore, the study on bioavailability of OPs in soil is extremely important for the application of composting technology. This work will discuss the changes of physical and chemical properties of contaminated soils and the bioavailability of OPs by the adsorption of composting matrix. The characteristics of OPs, types and compositions of compost amendments, soil/compost ratio and compost distribution influence the bioavailability of OPs. In addition, the impact of composting factors (composting temperature, co-substrates and exogenous microorganisms) on the

removal and bioavailability of OPs is also studied.

Keywords: Soil organic contamination; Bioavailability; Amendments; Composting;

Contents

1. Introduction

In recent years, soil organic contamination has become a major problem. Organic pollutants (OPs) of special concern include polycyclic aromatic hydrocarbons (PAHs), pesticides and petroleum (Gong et al., 2009; Tang et al., 2016; Tang et al., 2008; Wang et al., 2017; Zeng et al., 2013a). A number of biological remediation methods have been exploited to organic contaminated soil, either by biostimulation, such as addition of nutrients or organic matter to spur microbial activity, or through bioaugmentation, such as introduction of degrading microbes or organic amendments containing active microorganisms (Kästner and Miltner, 2016; Sayara et al., 2011). However, the effectiveness of biodegradation is restricted by various factors (e.g. oxygen and nutrient limitations, pH and C: N: P ratio) that are important conditions for microbial growth (Hickman and Reid, 2008a). Application of composting or compost amendments for soil remediation is competent to moderate those limitations (Semple et al., 2001).

Compost addition, which is produced from composting of organic wastes, contains abundant microorganisms and nutrients. Many studies have verified the effectiveness of compost addition in bioremediation. The biomass values after compost amendment were one order of magnitude higher than the unfertilized soil (Zhang et al., 2011). Wallisch et al. (2014) also reported that compost amendments stimulted the growth of alkane degrading microorganisms and thus the degradation of alkane. Baldantoni et al. (2017) further demonstrated higher degradation rate of PAHs and enhanced peroxidase activity after compost amendments.

Water-extractable organic matter (WEOM) functions as microbial growth promoters and PAHs mobilizer due to the existence of similar hydrophobic open structures with hydrophobic organic chemicals or the interactions with humic substance-like hydrophobic sites. Its high tendency to adsorb on cell components made the WEOM-associated POPs more likely to be absorbed and degraded (Kobayashi et al., 2009).

In addition to compost addition, soil composting is an alternative technology to the removal of soil OPs. Composting is a biochemical process involving mineralization of organic substrates into more stable, humidified forms and inorganic products. Therefore it can be applied in treatment of soil contaminated with OPs (Houot et al., 2012; Lashermes et al., 2012; Lukić et al., 2016; Peng et al., 2013; Sadef et al., 2014b). The main emphasis of soil composting was laid on the degradation of OPs in contaminated soil. Chen et al. (2016) compared the degradation of 2,2,4,4-tetrabromodiphenyl ether (BDE-47) in soil under composting conditions and natural conditions. The removal rates of BDE-47 increased by 15% in composting soil. Zhu et al. (2017) further studied the benzo(a)pyrene degradation during soil composting with/without co-substrate. About 61% of benzo(a)pyrene was removed in amended composting soil while only 46% was removed in unamended composting soil. Soil composting concerning two ways: one is composting with exogenous raw organic waste materials and another is the full composting process in which feedstocks is soil and biomass (Covino et al., 2016). However, most studies are conducted with the help of exogenous amendments and the amendments could have different effect on the removal of OPs in soil (Loick et al., 2009).

Bioavailability is an important factor affecting the biodegradation of OPs. The definition of bioavailability is varied. Semple et al. (2004) clearly put forward two distinct terms "bioavailability" and "bioaccessibility" to clarify in what circumstances a chemical can be available (Semple et al., 2004). Moreover, Reichenberg and Mayer (2006) proposed the concept of "chemical activity" and "bioaccessibility" from the aspect of chemical kinetics and chemical measurement. However, it is generally acknowledged that OPs bioavailability can be evaluated in terms of sorption/desorption of OPs and the microbial activity (Ehlers et al., 2003; Ren et al., 2017; Semple et al., 2013). It is conceivable that composting matrix influence the bioavailability of OPs in soil, because it can provide nutrients, extra carbon sources and a wide variety of microorganisms, which are beneficial to soil in terms of physical properties, nutrient availability and microbial activity (Adam et al., 2015; Feng et al., 2014; Puglisi et al., 2007; Zhang et al., 2007; Xu et al., 2012). However, the impact of compost amendments or the composting process on bioavailability of OPs is determined by various physicochemical and biological factors (Plaza et al., 2009; Semple et al., 2001). This review will discuss the bioavailability of OPs in soil after compost/composting to better manage this kind of technology and exert its greatest benefit on removal of OPs.

2. How compost addition and composting process affect bioavailability of OPs

2.1. Effects on soil microorganisms

Compost incorporated into soil impact the microbial abundance, microbial community composition and microbial activity (Fig. 1). Some reasons may be responsible for the increase of microbial abundance, such as higher available nutrients, labile organic matter, the increased water retention and aeration (Duong et al., 2012; Hu et al., 2011b; Ros et al., 2010; Schimel et al., 2007; Tejada et al., 2009; Wu et al., 2013).

Organic amendments incorporated into soil provide abundant carbon sources and nutrients. Particularly the labile organic matter, which was readily available to microorganism, contributed a lot to the microbial growth. This was supported by the observation that composting of plant residues with more labile organic matter resulted in higher soil microbial biomass and respiration (Tejada et al., 2009). Besides, Wu et al. (2013) found that compost additions adjusted C: N ratio in soil and thus increased microbial biomass (Wu et al., 2013). However, Tejada et al. (2009) observed optimum C/N ratio (10-12) in mixture of two plant residues composts but not others, indicating that the supplement of N by compost was dependent on sources of compost material. In addition, the animal manure compost was reported to contain struvite that could fix soluble phosphorus and act as slow release fertilizer of phosphorus after land application (Hu et al., 2011b). Duong et al. (2012) also indicated that soil available P concentrations increased by 16-170% with compost addition compared with unamended soils. Both compost-derived P and mobilization of soil P contributed to the marked increase. Compost may indirectly increase P availability by processes including: formation of phospho-humic complexes to decrease P immobilization, substitution of P by humate ions, decreasing potential P binding sites by coating sesquioxide particles with humus (Duong et al., 2012; Zeng et al., 2017).

Furthermore, humidity may largely influence the microbial abundance (Zeng et al., 2013b). Soil of periodic drying decreased water potentials, which would impose physiological stress and induce dormant or even death of microorganisms (Schimel et al., 2007). Compost has high water-holding capacities. Therefore, it could increase the water availability for microbial growth. In addition, compost amendments improved soil aggregate stability, either by flocculating soil particles with organic matter or increasing soil microbial activity accompanied with production of mucilage that could benefit the formation of soil microaggregates (Tejada et al., 2009). The improvement of structural stability increases soil porosity, and subsequently soil aeration, which is favorable for microbial life (Duong et al., 2012). However, not all compost organic matter was responsible for the aggregate binding effect. It was strongly correlated with the concentration of humic acid (HA) (Duong et al., 2012; Tejada et al., 2009).

The microbial community of hydrocarbon-contaminated soils was affected by the compost amendments (Bastida et al., 2016; Gandolfi et al., 2010; Ros et al., 2010). A rise in Gram-positive bacterial biomass and a decrease in the Gram-negative biomass in compost treated soil were observed (Bastida et al., 2016).

Gandolfi et al. (2010) also reported that compost additions completely changed the microbial community composition in PAHs contaminated soil, shifting from *Alpha-* and *Gammaproteobacteria* to *Bacteroidetes* and *Firmicutes*. Noteworthy, only *Bacteroidetes* presented at the end of this experiment, probably due to the abundance of *Bacteroidetes* in compost additions (Gandolfi et al., 2010). The increased soil microbe population was proved to stimulate microbial metabolism and consequently enhance 1,1,1-trichloro-2,2-bis (4-chlorophenyl) ethane (DDT) degradation (Purnomo et al., 2010). However, the increase in microbial numbers not necessarily accelerates the degradation for pollutants. In crude oil contaminated soil, the total petroleum hydrocarbons (TPH) degradation was low after organic additions, especially horticultural waste compost. They concluded that microorganisms preferred readily available compounds rather than the more complex and less degradable crude oil, so the increased microbial respiration was observed but no significant TPH degradation (Schaefer and Juliane, 2007). In view of short intervention time, we should not conclude that compost additions depress degradation of OPs and the impact of contact time will be discussed later.

Apart from microbial community, composts would also affect soil microbial activity. In the study of Bastida et al. (2016), biodegradation of hydrocarbons in natural soil was insignificant and addition of compost promoted its removal by 88% after 50 days. This was driven by reinforced microbial activity and increased abundance of catabolic enzymes by *sphingomonadales* and uncultured bacteria (Bastida et al., 2016). The increased enzyme activities of compost-treated soils has long been found, which is the net effect of stimulated microbial growth and microbial activity, as well as diverse microbial community composition. However, it was found that the increase in enzyme activity was ascribed to soil microorganisms releasing extracellular enzymes but not exogenous enzymes from compost (Iovieno et al., 2009). Di Gennaro et al. (2009) further explained that vermicomposting stimulated metabolically active bacterial community by inducing the expression of biodegradation indicator genes in indigenous microorganisms and inoculation of new PAHs-degrading bacterial. Moreover, compost amendment was characterized as microbiologically active product, which was often overlooked. Sterile compost hindered pentachlorophenol biodegradation as the formation of humus-pentachlorophenol in soil, indicating the importance of compost microflora (Lin et al., 2016). Cattle manure compost contained fungi that were able to mineralize DDT and soil microorganisms could accelerate DDT degradation through synergistic actions (Purnomo et al., 2010). Similarly, basidiomycetes that isolated from straw compost could successfully colonize soil and remove 56% of pyrene after 28 days by production of ligninolytic enzymes (Anastasi et al., 2009). Above researchers threw light on the significance of compost microorganism (Anastasi et al., 2009; Lin et al., 2016; Purnomo et al., 2010).

In addition to microbial activity, composts also act on the accessibility of OPs to microorganism. Different from soil organic matter (SOM), compost organic matters are rich in dissolved organic matter (DOM), which has multiple functions on the binding and release of both OPs and microorganisms. Improving OPs accessibility in soil requires both the mobilization of OPs and degrading-microorganism. Non-aqueous-phase-liquids (NAPL) in soil, such as tar oils, restricted the OPs mass transfer to water phase and for microbial degradation. Composts mixed with soil provide a suitable micro-environment for mass transfer with the help of introduced energy (shear forces and increased temperature) that could destroy the interfacial surface resistance in NAPL (Kästner and Miltner, 2016). Besides, the lack of continuous water films restricts bacterial mobility in soil. Bacteria may be physically connected to DOM and facilitate its direct contact to micelles-associated PAHs (Schaefer and Juliane, 2007). Haftka et al. (2008) proposed that the enhancement of PAHs biodegradation was resulted from faster uptake kinetics of the water-dissolved compounds toward bacterial cells. DOM changed the collector surface charge and competed for the interaction sites between organic matter and bacterial cells, thus aiding the movement of bacteria. Jimenez-Sanchez et al. (2015) further investigated that soil bacterium *Pseudomonas putida* G7 transported under the action of four of representative DOM. Both chemotaxis and decreased interception probability with surrounding soil particles of motile bacteria were accounted for DOM-enhanced mobility. In conclusion, as suggested by Cai et al. (2017), DOM enhance the bioavailability of phenanthrene by increasing the solubility and mass transfer of phenanthrene, facilitating the microbial access to DOM – phenanthrene complex, increasing the microbial activity and uptake kinetics of *Sphingobium* sp.

Besides, it has been reported that the properties of DOM changed during the bioremediation process. The aromaticity of DOM increased and its polar components decreased after microbial transformation, which enhanced its affinity with hydrophobic chemicals (Hur et al., 2011). Banach-Szott et al. (2014) also demonstrated the changes of HA quality parameters in soil added with PAHs. Jednak et al. (2017) further studied the quantitative and qualitative changes of HA during bioremediation of petroleum hydrocarbons from waste mazute. The content of HA increased by 204% in this process and its groups tend to be more aromaticity. Besides, the fulvic acid (FA) carbon content decreased by 44%. A detailed description of DOM change was indicated by Cai et al. (2017). The proteins and tyrosine content in DOM decreased while the content of HA and FA increased. This could be that proteins and tyrosine was utilized for microbial growth while the HA and FA remained in DOM and acted as carriers of phenanthrene towards the degrading bacteria (*Sphingobium* sp). HA can also be co-metabolized by *Sphingobium* sp for phenanthrene degradation.

2.2. Effects on OPs migration and transformation

The physical and chemical properties of soil are essential to understand the migration and transformation of OPs. Compost improve soil physicochemical properties in terms of decreasing soil bulk density and erodibility, increasing water holding capacity, aggregate stability and cation exchange capacity (CEC) (Viaene et al., 2016). Compost additions have a significant influence on organic matter content, soil pores and CEC, which consequently affect OPs bioavailability (Fig. 1).

It has been reported that the combined effect of chemical affinity by organic matter and strengthened physical trapping in mesoporous contributed to chlordecone retention after compost addition (Woignier et al., 2013). Also, Woignier et al. (2016) found that soil–plant transfers of chlordecone were inhibited after incorporation of the compost. The fragile fractal microstructure of allophane clays were strongly altered by incubation of compost. They further proposed that addition of compost caused capillary stresses that led to collapse of the clay microstructure. Pore change induced marked reduction of the hydraulic conductivity and diffusion coefficient by 95% and 70% respectively, which limited the transportation and bioavailability of the pesticide in soil (Woignier et al., 2016). Besides, compost solid would aggregate together with soil particles, blocking the larger soil pores, which increased the difficulties for the initially pores-sequestrated PAHs to be accessed by microorganism (Wu et al., 2014). Unlike in andosols, chlordecone sequestration in nitisol was due to chemical sequestration by the compost particles, and addition of compost did not affect the soil pores structures (Clostre et al., 2013). Sequestration of OPs was strongly affected by soil type (texture, pores, structure). Andosols with organic carbon content up to 100 g/kg presented a fractal structure that could trap chlordecone, thus having 10-fold higher sorption capacities than nitisols (Florence et al., 2015; Levillain et al., 2012). Soil with larger ratio of sand (pore size: 2000–50 μm) has lower loss of high molecular weight (HMW) PAHs compared to soil with higher silt (pore size: 50–2 μm) after compost addition (Wu et al., 2013). The beneficial effects of compost additions to soil are outstanding in sandy soils that are in low water holding capacity and nutrient content and clay soil that are poor aerated and low in available water (Duong et al., 2012). Similarly, Wu et al. (2013) observed over 90% loss of PAHs after compost additions regardless of soil type. However, the contribution of sorption, desorption and degradation to the loss of PAHs vary with soil type. In the diesel spiked soil, strong sorption led to the reduction of PAHs dissipation by 89% regardless of the compost type. As time progress, PAHs removal in compost addition soil was twice of the unamended soil, among which desorption and degradation accounted for 30% and 70% respectively. In coal tar and coal ash contaminated soils, compost addition was beneficial overall for enhancing PAHs removal up to 94%, among which 40% was caused by enhanced desorption. Therefore, functions of compost additions on contaminants sorption are dependent on soil properties.

Compost amendments increase soil CEC either by incorporation of highly decomposed organic matter that has plenty of cation binding sites or stimulating native organic matter decomposition to produce more binding sites (Kodešová et al., 2012). The increment in soil CEC suggested reinforcement of OPs sorption by soil, making it harder to transfer (Duong et al., 2012).

As to soil pH, compost additions generally act as pH adjustor by the advantage of humic substances that were rich in acidic functional groups. In different cases, the effect of compost additions on soil pH was not the same, but had minor impact. Typically, increase in soil pH after compost addition includes the following processes: (i) ammonification; (ii) production of CO_2 due to carbon mineralization and (iii) formation of OH by ligand exchange due to introduction of basic cations such as Ca, K and Mg, while decrease in pH was caused by nitrification that generate acids (Mkhabela and Warman, 2005). Although soil pH is demonstrated to influence OPs sorption/desorption, it has not been documented yet that how the shift in pH caused by compost additions affects OPs biodegradation, due to the negligible effect on soil pH of compost in most times.

3. Contradictory effects of compost additions on bioavailability of OPs

Compost amendments affect bioavailability of OPs by stimulating the indigenous microflora or modifying the soil conditions to be more or less favorable to contaminants sorption/transportation (Fig. 2). There is a dual effect of compost addition to OPs bioavailability in soil. i) Stimulation to soil native microorganisms by compost nutrients and organic matter, together with newly inoculated microorganisms from compost, enhance soil microbial activity, which could promote biodegradation of OPs (Gandolfi et al., 2010; Peng et al., 2013); ii) Introduction of organic matter will increase the adsorption of OPs (Fan et al., 2008; Feng et al., 2010; Kästner and Miltner, 2016; Wu et al., 2013) and iii) DOM in compost will improve the uptake of OPs by microorganisms, increase its solubility and facilitate desorption (Haftka et al., 2008; Hu et al., 2011a; Jimenez-Sanchez et al., 2015; Tang et al., 2014). It is thus important to analyze the evolution of OPs bioavailability for the purpose of optimizing composting process to maximize pollutants degradation.

3.1. The characteristics of OPs

Physicochemical properties such as hydrophobicity, structure and molecular weight of OPs have a bearing on their bioavailability. Bioavailability was negatively correlated with OPs n-octanol/water partition coefficients ($logK_{ow}$) (Wu and Zhu, 2016). Hickman and Reid (2008a) suggested that degradation extents and rates of hydrocarbons varied with different structure, with particular reference to diesel and petroleum contamination. Studies on PAHs showed that bioavailability change of low molecular weight (LMW) PAHs was more time dependent than HMW PAHs, on the grounds that LMW PAHs were more susceptible to the leaching, volatilisation and degradation processes (Wu et al., 2014; Yuan et al., 2014). The presence of multiple PAHs-mixtures decreased the bioavailability of LMW PAHs (naphthalene) by competitive inhibition of the enzymes associated with biodegradation, but increased the bioavailability of HMW PAHs (phenanthrene and pyrene) by inducing catabolic activity of soil microbes (Couling et al., 2010). Therefore, it is important to associate OPs characteristics with the effect of compost on bioavailability. A representative example is the degradation of triazine pesticides (e.g. atrazine), which may be inhibited by compost additions (Alvey and Crowley 1995). High N-containing atrazine served as N source to the degrading microorganisms. Therefore, compost amendments, as an alternative N source, could decrease atrazine degradation (Abdelhafid et al. 2000).

3.2. Quality and nature of compost organic matter

The fate of OPs in soil is dependent on both the quality and nature of compost organic matter. It has been proved that bioremediation of PAHs-contaminated soils with matured compost were more effective than with fresh organic amendments

(Plaza et al., 2009), because fully rotted compost provided available nutrients with low sorption potential for HMW PAHs (Adam et al., 2015). The mechanism and extent of binding are strongly affected by compositional and structural properties of the HA. Compared with soil HA, organic substrate HA is characterized by stronger binding capacity and lower heterogeneity of binding sites. For this reason, if organic substrates are introduced into soil, the affinity of soil HA for PAHs will be increased based on the averaging effect. There may be a decrease in PAHs bioavailability. The composting process is capable of decreasing HA binding affinity and increasing the heterogeneity of binding sites, making it close to soil HA, which is conducive to microbial accessibility to PAHs (Plaza et al., 2009; Senesi and Plaza, 2007). The changes in HA generally lead to lower sorption and thus higher availability to microorganisms. Luo et al. (2015) investigated the change of organic matter during sewage sludge composting and the subsequent influence on its adsorption ability to pentachlorophenol. During composting, HA content was decreased while the FA, humin and total carbon content increased. Higher aromatization of HA and FA caused an increase in their sorption capacity by 54.76 and 36.73 % respectively. However, Ros et al. (2010) found that fresh sludge treatment had the highest percentage of hydrocarbon degradation and bacterial and fungal population than compost sludge treatments, as a result of higher amount of easily degradable substrates and nutrients in fresh sludge. The DOM from mature compost contains a low content of biodegradable organic matter and a higher concentration of organic macromolecules (He et al., 2014), while microorganisms prefer available organics.

This kind of effect is particularly significant if degrading microorganisms does not exist in compost. Another study done by Vieublé-Gonod et al. (2016) observed that isoproturon degradation was negligible in compost but it could be enhanced in compost amended soil. It could be that isoproturon degrading microflora did not colonize the compost organic matter.

Nonetheless, compost may contain active microorganisms for the degradation of pollutants, depending on composition of original materials. Microorganisms could be initiated by a large amount of aromatic compounds, *e.g.*, lignin containing plant material during composting, contributing to highly active compost microflora in comparison to the unfertilized agricultural soil for pyrene degradation (Adam et al., 2015). Concerning the role of compost microorganisms, as we know that each successive stage in composting (mesophilic phase, thermophilic phase and curing phase) is expected to be accompanied by specific populations of bacteria, and different effects on contaminants are found with different stages of compost product. Mesophilic stage compost material exhibited the highest ability to degrade DDT in soil (Purnomo et al., 2010), while maturation microflora showed better performance than the thermophilic microflora in pollutants mineralization in another study (Houot et al., 2012). The contaminants used in those studies may explain the difference. In addition, application of unstable and/or immature organic amendments may cause undesirable consequence on environmentally safety, such as incorporation of pathogens into soil (Senesi and Plaza, 2007). What's more, poorly-decomposed organic matter generally located in coarse sludge compost fractions (> 5 mm) and most humified organic matter generally presented in fine fraction (Doublet et al., 2010). The finest compost size fraction (< 3 mm) with higher surface area volume ratio was demonstrated to be more accessible to microorganism and to release more N and P compared to coarse compost fraction (Verma and Marschner, 2013) .

The different effects of compost amendments depend on the fact that compost materials vary significantly in sources and stages of decomposition, and act on the removal of contaminants through different ways (Li et al., 2015). Houot et al. (1998) used two different compost additions (municipal solid waste compost and composted straw) in soil for atrazine degradation. The addition of municipal compost increased atrazine sorption and decreased its availability to microorganisms. Composted straw with high enzymatic activity or the acidity of the humic components was responsible for the production of large amounts of hydroxyatrazine which favored the opening of the triazine ring and its subsequent mineralization in the soil (Houot et al., 1998). Besides, compost materials determined the properties of the final product, such as relative content of C/N/P. For example, compost made from food residues and animal manure were rich in nutrients while yard waste was deficient of nutrients, thus inorganic fertilizer supplement should be added together (Ilani et al., 2016).

What's more, the effect of compost material type on microbial and fungal biomasses varied with time. Municipal solid waste with more labile organic matter was degraded rapidly and had a short-term impact that lasted only one month. Green waste co-composted with sewage sludge containing more stable organic matter was slowly degraded and had a continuous effect that lasted for 6 months (Vieublé-Gonod et al., 2009). However, Wu et al. (2013) suggested that compost material had little influence on PAHs bioavailability. Duong et al. (2012) also mentioned that the type of compost material was insignificant in changes of soil induced by compost additions, and soil-specific effects (*e.g.*, enhanced aggregate stability for soil with medium to fine texture and nutrient availability for nutrient-deficient soil) appeared to be more important.

3.3. Soil/compost ratio and compost distribution

Apart from inherent properties of compost, how the compost additions are used is important. An inappropriate ratio of compost addition may retard or inhibit microbial activity and bioavailability in soil. Addition of Elliott soil HA (ESHA) within the range of 20-200 μg /g were found to consistently increase pyrene mineralization, while beyond this concentration range it may produce inhibition or present no effects. At high HA concentrations, inhibition can be caused by micelles impeded transportation (Liang et al., 2007). The compost-ratio related effect of compost additions was also investigated by Kodešová et al. (2012) who stated that herbicide mobility decreased with increasing compost content up to 6%, but markedly increased with 7% amendment, and slightly increased with 8% amendment, which was consistent with the adsorption experiment. There is no consistent conclusion about the effect of compost amount on PAHs bioavailability. Puglisi et al. (2007) found no difference in phenanthrene bioavailability after addition of double doses of compost. In contrast, Feng et al. (2014) observed highest PAHs dissipation in soil amended with compost at 10%. Similarly, Hickman and Reid (2008b) tested that compost additions combined with earthworms at a ratio of 1:0.5 to 1:1 (soil/compost, wt/wt) were efficient to the dissipation of extractable petroleum hydrocarbons and PAHs. When higher volumes of compost (1:2 and 1:4) was used, PAHs loss were not advanced, which may indicated that the activity of earthworms were restricted by higher addition of compost (Hickman and Reid, 2008b). In order to find the relationships among the multiple interactions, Wu et al. (2014) investigated multiple factors on PAHs bioavailability in compost amended soils using conjoint analysis and five-way analysis of variance. Soil type and contact time were the most important factors that account for >90% of the bioavailability changes while compost type and ratio of compost addition were insignificant, but their interactions with other factors would make a big difference (Wu et al., 2014).

As to distribution of compost amendments, the ploughing normally created a heterogeneously distribution of organic matter in agricultural practice. Spatial distribution of exogenous organic matter in soil was irrelevant to microbial respiration but it could influence the fate of isoproturon. Compost additions that homogeneously distributed in soil contributed to maximal degradation and mineralization (Vieublé Gonod et al., 2016). Two reasons may account for the enhanced isoproturon mineralization in homogeneous soil-organic matter cores: exogenous carbon from compost promotes growth of isoproturon degrading microorganisms and/or acts as cometabolic substrate. Transportation is important to isoproturon degradation during this process since isoproturon in compost should diffuse into soil and contact with soil microorganisms before degradation, and soil degrading microorganisms should be transported to and proliferate on compost that contained isoproturon. Compared with homogeneous soil-organic matter cores, the heterogeneous compost tend to concentrate together and the contact between carbon, isoproturon and soil microorganism are impeded (Vieublé Gonod et al., 2016). Therefore, proper application of compost should be considered to allow compost components exert the best effect.

4. Variation of OPs bioavailability during composting

Bioavailability of OPs varied during composting, which was affected by a variety of factors. In this part, we focused on composting temperature, co-substrates and exogenous microorganisms (Fig. 3). The enhancement of degradation rates indicated higher bioavailability of OPs. Degradation of compounds during composting proceeds rapidly in the beginning and slows down in the later. A possible reason for this behavior is that easily accessible pollutants molecules which are dissolved in the water phase or adsorbed on the particle surfaces are degraded initially, while the remaining pollutants that are strongly adsorbed to particles or present within the micropores of the particles are difficult to degradation (Sadef et al., 2014a, 2014b).

4.1. Impacts of composting temperature

The temperature during composting is a key parameter controlling degradation rates. There are four distinct successional phases driving chemical and microbial changes through time: (i) the initial mesophilic phase: when the microbial community builds up and adapts to the conditions, with moderate temperatures up to about 45°C; (ii) the thermophilic phase: when metabolism is most intense and temperature increases to peaking at almost 70°C; (iii) the second mesophilic phase: during which the mesophile microorganisms dominate and the temperature decreases to about 40 °C and (iii) the curing phase: when the metabolism slows down and temperature cooling to ambient temperature (Neher et al., 2013; Ryckeboer et al., 2003). Each successive stage in composting is expected to be accompanied by specific populations of bacteria. In a study done by Peng et al. (2013), the predominant bacterial community changed over time during pyrene contaminated soil in-vessel composting. Degradation of pyrene was dominated by α-, β-, γ-*Proteobacteria*, and *Actinobacteria* at 38°C during 14 days of composting, and then *Streptomyces* at 55°C. Later at 70°C after 42 days of composting, *Acinetobacter* and *Thermobifi*da occupied leading position. Finally, *Thermobifida* and *Streptomyces* flourished after 60 days of composting at 38°C (Peng et al., 2013). Composting temperature affects the prevailing of some microbial groups over others. This further determines in which composting stage the maximum degradation rate of OPs occurs. According to Xiao et al. (2011), continuous thermophilic composting promoted the growth of *actinomycetes*, a group of high temperature tolerance microorganisms efficient to degrade organic matter. They proposed that the continuous thermophilic composting contributed to rapid biodegradation of organic matter and shortened the composting cycle.

Therefore, the impact of composting temperature on the OPs bioavailability is, on the one hand, based on the advantageous microbial populations under specific temperature. In the study of Lin et al. (2012), *Oleiphilus* species were found in all composting stage. However, the decomposition of diesel oil mainly occurred in the thermophilic stage. This could be that the *Oleiphilus* bacteria are more active under thermophilic conditions. However, another study done by Arikan et al. (2016) found that higher composting temperature (65 ^{o}C) was not always effective in the removal of monensin relative to ambient temperature (22 ^{o}C), although high composting temperature increased the removal of lasalocid and amprolium than ambient temperature. Similarly, Lukć et al. (2016) claimed that LMW PAHs removal was more favorable in mesophilic phase with 11 and 15% residues in soil than thermophilic phase with 29 and 31% residues. Moreover, Sadef et al. (2014a) studied the influence of composting temperature on the removal rates of 15 key organic micropollutant. The optimal temperatures were compound specific and ranged from 25 to 70 ^{o}C. Thermophilic conditions were optimal for about half of compounds and the remaining half were suited to the second mesophilic conditions. Lashermes et al. (2010) also reported that the highest mineralization of sodium linear dodecylbenzene sulfonate was observed during thermophilic stage, due to the most intense microbial activity in thermophilic temperature, while mineralization of fluoranthene was associated to white-rot fungi that existed in maturation phases, which led to maximal fluoranthene mineralization (Lashermes et al., 2010). In conclusion, the impact of composting temperature was dependent on the type of contaminants because the corresponding degrading microorganisms are specific to temperature.

On the other hand, temperature affects the physicochemical characteristics of compost materials and subsequent bioavailability of OPs. High temperature increased the solubility and mass transfer rates of OPs, thereby making them more available to metabolism (Feitkenhauer et al., 2003). However, lower octanol–water partition of PAHs at high temperature decreased their degradation rates although this kind of effect could have been offset by microorganisms possessing active uptake mechanism (Sandler, 1996; Viamajala et al., 2007). Besides, lower oxygen solubility in higher temperature could also affect the aerobic degradation process. Viamajala et al. (2007) further demonstrated that the elevated temperature during thermophilic phase composting enhance the solubilization rates of phenanthrene, and hence their degradation. Similarly, Zhu et al. (2017) proposed that the enhanced solubility could explain higher removal of benzo(a)pyrene in composting temperatures (46%) treatment than in 22 ^{o}C treatment (29%). However, whether the increased solubility or microbial community changes account for the high temperature impacts need further investigation.

4.2. Impacts of co-substrates

The co-substrates coud adjust the organic carbon availability, C/N ratio, and the moisture of composting environment so as to increase the microbial activity. In the composting of contaminated soil, co-substrates such as manure are often added to provide sufficient readily degradable carbon source and nutrients. No significant degradation of PAHs occurred in treatment S (100% soil) during composting while a dry mass loss of 35±5% was observed in all treatments with organic wastes (Zhang et al., 2011). Sayara et al. (2011) investigated the degradation of PAHs in soil amendment with municipal solid waste compost with rabbit food as organic cosubstrates (biostimulation). Almost 89% of the total PAHs were degraded by the end of the composting period (30 days) compared with only 29.5% in controlled soil. Similarly, the degradation of benzo(a)pyrene in oat straw and ammonium nitrate co-composting soil was higher than in unamended composting soil (Zhu et al., 2017). Mattei et al. (2016) proposed that microorganisms utilize organic matter from green waste and excrete extracellular enzymes for PAHs degradation, which accounted for the improved degradation of PAHs in co-composting of contaminated sediments with green waste treatment than sediments alone. Therefore, it can be proposed that amendments with the higher content of the soluble fraction and protein in composting soil could show better removal efficiency of OPs (Lukić et al., 2016; Sayara et al., 2011). In addition, it was reported that addition of fresh organic matter improved the moisture content in the soil-compost mixture and thus contributed to effective PAHs degradation (Guerin, 2000). An investigation done by Beaudin et al. (1999) also showed that leaf/alfalfa substrate at the C/N ratio of 17 led to the highest degradation of mineral oil and grease. These studies indicated that components of co-substrate appear to be important factor in the removal of OPs during co-composting.

Notwithstanding the benefits of stimulating microbial growth, the co-actions of added organic amendments together with the microbial activities on the dissipation of OPs remain uncertain. Microorganisms have a preference for easily available carbon resources than the resistant pollutants. This is reported by Wang et al. (2011) that treatments with the amendment ratio of 1/1 and 2/1 had average TPH removal rates of 30.7% and 33.3%, but the amendment ratio of 3/1 had a slower net degradation rate of between 11.6% and 26.8%. An excess of readily degradable carbon might overpass the TPH to act as substrate for the metabolism of microbial degraders (Wang et al., 2011). Therefore, a proper amount of amendments should be taken into account in composting to balance the motivating effect on microorganisms and competing effect with pollutants.

4.3. Impacts of exogenous microorganisms

With respect to the role of exogenous microorganisms, 84% of petroleum hydrocarbon was degraded when inoculation of *Candida catenulate* CM1 while only 48% removal ratio was achieved without inoculation in a study carried out by Joo et al. (2008). In some cases when degrading-microorganisms are not enough to heavy contaminated soil, it is useful to introduce active microorganisms in soil remediation, among which white-rot fungi are widely used in composting due to their strong non-specific enzymatic system (Huang et al., 2008; Huang et al., 2017; Wan et al., 2015). However, no significant difference on the dissipation of PAHs between SW (soil/waste mixture) and SWB (soil/waste mixture with inoculation of degrading microorganisms) was found in the study of Zhang et al. (2011). Sayara et al. (2011) evidenced that promotion of the PAHs degradation was not significant when introduction of *T. versicolor*. An interesting approach in cooperative degradation of PAHs by inoculated fungus and the indigenous microbial community was described by Covino et al. (2010), during which inoculum carriers were vitally important. Lignocellulosic residues as inoculum carriers could mitigate the incompetency of white-rot fungi colonization in soil, and this kind of effect was dependent on both substrate and microbial species (Covino et al., 2010). Not all exogenous microorganisms are able to colonize polluted soil due to the following reasons: (i) indigenous and exogenous microorganisms compete for the limited carbon sources; (ii) exogenous microorganisms are preyed by protozoa; (iii) native species diversity resist the invasion of nonnative species and (iv) the implanted exogenous microorganisms may not tolerate to thermophilic temperature (>45^{o}C) during the composting process (Chen et al., 2015; Sayara et al., 2011; Zhang et al., 2011; Zhou et al., 2014).

5. Methods to measure bioavailability of OPs

The most commonly used chemical analytical approaches of bioavailability fall into two types, non-exhaustive extractions, a method based on measuring the rapid desorbed fractions(known as bioaccessible fraction) of OPs by extraction with contaminate sink, such as mild solvent extraction, cyclodextrin extraction, tenax-aided sequential desorption, supercritical fluid extraction and newly developed isotope dilution method, and biomimetic method (or passive sampling), a method determining the freely dissolved concentration of organic chemicals in the aqueous phase and are related to the chemical activity, which has been performed with different devices such as semi-permeable membrane devices, polyethylene devices, and solid phase microextraction fibers (Cui et al., 2013; Cachada et al., 2014; Jia et al., 2014; Riding et al., 2013). Many researchers have reviewed up-to-date technologies used to measure bioavailability of OPs. Cui et al. summarized frequently used bioavailability measurement approaches from the aspect of working principles, advantages and disadvantages and operation protocol (Cui et al., 2013). Likewise, Riding et al. (2013) elucidated the relative strengths and weaknesses of each chemical extraction techniques and the potential influencing factors on PAHs bioavailability measurement. Further, a review about the PAHs bioavailability process in soil and connection of chemical methods with particular organisms are developed by Cachada et al. (2014). Recently, Ortega-Calvo et al. (2015) discussed the probability of integrating bioavailability concepts into risk assessment and regulation, and proposed a system for including bioavailability in risk assessment. Here we will discuss the two typical newly-developed methods [hydroxypropyl-b-cyclodextrin (HPCD) extraction (non-exhaustive extractions) and solid phase microextraction (biomimetic methods)], and their comparation with other methods.

Hydroxypropyl-b-cyclodextrin (HPCD), a kind of cyclic oligosaccharide with hydrophilic surface and hydrophobic cavity that used to entrap OPs, is considered as a reliable predictor of bioavailability to microorganisms (Cachada et al., 2014; Cui et al., 2013; Riding et al., 2013). Mineralized fraction of phenanthrene and its rapidly desorbing fraction extracted by HPCD after 24 h were strongly correlated of approximately 1:1 (Rhodes et al., 2010). However, validated linear regression models with slope from 0.87 to 1.56 and correlations from 0.84-0.98 were developed for 8 of the investigated 12 PAHs, whereas the remaining 4 high molecular weight PAHs (benz[a]anthracene, benzo[a]pyrene, benzo[btk]fluoranthene, benzo[g,h,i]perylene) was not applicaple (Juhasz et al., 2014). Spasojević et al. (2015) demonstrated that cyclodextrin was better correlated with biodegradation of LMW PAHs, but the XAD-4 was suitable for HMW PAHs. Tenax extraction present similar adsorption ability to HPCD or even huger as infinite contaminate sink. Both methods had the potential to assess PAHs and petroleum hydrocarbon biodegradation with different adsorption mechanisms. Contaminants that were dissolved in the water phase were adhered to the apolar surface of Tenax, similar to organic matter, while surface of cyclodextrin molecule was hydrophilic and compounds were entrapped in its hydrophobic cavity, which made Tenax extraction more time-consuming with an extra beads seperation process than HPCD extraction (Bernhardt et al., 2013).

Biomimetic methods are developed on the basis of Equilibrium Partition Theory, which assume that the concentrations of chemicals between organic matter, pore water and the lipids of organisms are proportional to each other (Cachada et al., 2014). Solid phase microextraction (SPME) passive samplers were demonstrated to be a suitable proxy for bioaccumulation in both lab and field studies (Maruya et al., 2015) with inexpensive cost, commercial available of fiber material and solvent free from the extraction stage, thus widely used to evaluate bioavailability in soil. Furthermore, SPME fiber-based bioavailability estimation are unaffected by exposure routes, species density and fiber volume, validating the usefulness in environmental assessments (Harwood et al., 2012). Passive samplers such as SPME are more suitable for in situ detect the bioavailability change by burying it at the site and periodically retrieving it. However, extraction methods such as Tenax extraction and isotope dilution method are more sensitive in monitoring bioavailability than SPME, as reported by Jia et al. (2016).

6. Conclusions and perspectives

Compost amendments or composting process affected soil microorganism and OPs migration and transformation, thus influencing the bioavailability of OPs. Microbial abundance, community composition and activity were changed by composts, during which the DOM in composts play a significant role. DOM act as energy sources for microorganisms and promote bioavailability of OPs. The physicochemical properties of soil were also improved after compost addition or soil composting. Through the deeply discussion, we can see that compost amendments have contradictory effect on bioavailability of OPs, which is dependent on the nature of both OPs and compost additions. Composting temperature, co-substrate and exogenous microorganisms during composting may also affect OPs bioavailability. Finally, the newly developed technology of bioavailability measurement was outlined. Based on our research, we noticed some knowledge gaps that need to be filled in the future.

(1) Compost applications affect OPs bioavailability, which is realized by promoting the indigenous microorganisms, introducing exogenous active microorganism and/or changing the soil properties to be more or less favorable to contaminants sorption/transportation. The performance might be influenced by variable compost quality and composition, compost dose and how the compost additions are applied. Mature compost is more appropriate than unmature compost considering the toxic effect. Different type of compost act on the removal of contaminants through different ways, but their effect on bioavailability is suggested to be insignificant compared to soil type and contact time. The same case is observed for compost ratio. However, more researches are needed to identify the interactions of multiple factors influencing bioavailability since few studies are conducted. Besides, compost should be properly utilized in soil to make good use of its advantage.

(2) Knowledge about the impact factors of composting on bioavailability of OPs may help to control the composting conditions. High temperature typically increases the bioavailability, but the corresponding degrading microorganism is distinctive in temperature tolerance. Besides, a proper amount of composting co-substrate should be considered depending on the motivating effect on microorganisms and competing effect with pollutants. Whether introducing exogenous microorganisms during composting depends on a number of factors, including microbial temperature tolerance, degrading potential and counterpart predator in soil, as well as indigenous microorganisms. It is also difficult to

translate all the aspects of composting, taking into account the variables such as pH, moisture and oxygen variation. Since those variations are rarely discussed in relation to OPs bioavailability and are mostly associated with microbial activity, the detailed mechanisms should be further studied.

(3) Considering the effectiveness of using bioavailability concept in organic contaminated soil remediation, it is important to find an acceptable protocol to investigate the bioavailability of a variety of OPs in soil. Unlike toxic elements, no unified guidelines for OPs are proposed yet. Besides, comparation of different measurement approaches is important to select the most suitable method for evaluation of remediaiton efficiency. Despite various methods to evaluate bioavailability, there still needs extensive researches to find applicability of different bioavailability methods in different cases.

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Figure captions

Fig.1. Effect of compost on microbe and soil physicochemical characteristics.

Fig.2. Contradictory effect of compost on bioavailability of OPs.

Fig.3. Impact factors on bioavailability of OPs during composting process.