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Review Paper

Effect of exogenous carbonaceous materials on the bioavailability of organic pollutants and their ecological risks

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ABSTRACT

The presence of exogenous carbonaceous materials (ECMs) in organic contaminated soil is widespread because of their intentional application as carbonaceous amendments (e.g. biochar and activated carbon) or unintentional discharge (e.g. carbon nanomaterials). Most research so far has focused on the sorption behaviors of ECMs in soil. However, the impact of ECMs on the bioavailability of organic pollutants (OPs) and their ecological damages remain unclear. This paper presents an overview on how the ECMs affect bioavailability of OPs to different organisms, such as microorganisms, plants and earthworms. This is affected by different biological response and properties of ECMs. Moreover, the possible risks of ECMs on soil biota are also discussed at different level. This review presents a unique insight into risk assessment of ECMs. Further researches should focus on possible change in physicochemical characteristics of ECMs when exposed to the natural environment and the consequent influence on their sorption ability and ecotoxicity outcomes.

1. Introduction

Currently, increasing concern has been raised concerning the issue of soil organic contamination resulting from intensive industrial and agricultural activities (Cheng et al., 2016; Liu et al., 2012; Tang et al., 2014b; Yang et al., 2015; Zhou et al., 2016). The coexistence of exogenous carbonaceous materials (ECMs) and organic pollutants (OPs) is possible in the soil environment (Pang et al., 2011; Zeng et al., 2013a). ECMs can be divided into carbonaceous amendments and carbon nanomaterials (CNMs). Carbonaceous amendments, such as activated carbon and biochar, are highly recommended in soil remediation because of their large surface area, high porosity and great sorption capacity (Feng et al., 2010; Ghosh et al., 2011; Tang et al., 2016). In addition, the rapid development of nanotechnology and unique physicochemical properties of carbon nanomaterials (CNMs) have caused a sharp increase in the production and utilization (Tang et al., 2015, 2014a, 2012; Zhou et al., 2017). Various CNMs such as carbon nanotubes (CNTs), graphene and fullerene have been widely applied in numerous fields, which inevitably release into the natural environment (Tang et al., 2014c; Liu et al., 2012; Zhang et al., 2007). Besides, the potential use of CNMs in environmental remediation has also been reported (Gong et al., 2009; Song et al., 2017). Accordingly, whether intentionally (e.g. pollution remediation) or unintentionally (e.g. accidentally spill), the existence of ECMs in soil is possible. In this case, two sides should be considered.

On the one hand, sorption behavior of ECMs would decrease bioavailability of OPs (Fan et al., 2008; Hu et al., 2011; Huang et al., 2008; Yang et al., 2010). The sediments amended with 1% of carbon materials showed a decrease in freely dissolved concentration (C_{free}) of polybrominated diphenyl ethers (PBDEs) in sediments, up to 98.3% with activated carbon, followed by 78.0% and 77.5% with biochar and charcoal, respectively (Jia and Gan, 2014). Towell et al. (2011) also found that extractability of polycyclic aromatic hydrocarbons (PAH) significantly decreased with increasing addition of fullerene soot and CNTs. When evaluating the impact of ECMs on OPs bioavailability, microorganism is mostly studied as it is the primary degrader of OPs and is widely exist in natural environment (Ren et al., 2017; Rodrigues et al., 2013; Zhu et al., 2016a). However, considering the agronomical value and engineering application of plants in soil remediation, it is also important to investigate its impact on plants uptake of OPs (Hamdi et al., 2015; Zhou and Hu, 2017). In addition, earthworms are considered as predominant soil fauna in terrestrial ecosystem and perform many beneficial functions in soil, including soil structure improvement, carbon and nutrient cycling, and bioaccumulation of OPs (Rodriguez-

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Table 1
Mechanisms and possible effects of ECMs to the bioavailability of OPs.

Materials	Concentration	Pollutants	Species	Mechanism	Bioaccumulation or biodegradation	References
MWCNTs	25,50,100 mg/kg	PAH (100 mg/kg)	High tolerant microbial groups	Microbial attachment to adsorbed OPs	No significant effect	Shrestha et al. (2015)
Biochar or CNTs	10, 25, 50 and 100 mg/L	Atrazine (100 mg/L)	Microorganism (*Acinetobacter lwoffii* DNS32)	Cytotoxicity of ECMs	Decrease by 35.9–68.9%	Yang et al. (2017)
Biochar	2.8%	DDT (39 μg/g)	Plant (*Cucurbitapepospp.pepo*)	Root exudates	No significant effect	Denyes et al. (2016)
C_{60}	1000 mg/L	DDE (100 ng/mL)	Plant (*Cucurbita pepo* L., *Glycine* max L. and *Solanum lycopersicum* L.)	Cell membrane damage by CNMs and high availability of CNMs-adsorbed-OPs	Increase by 30–65%.	De La Torre-Roche et al. (2012)
Biochar	10%	PAH (773 mg/kg inherent in soil)	Earthworm (*Eisenia fetida*)	Biochar act as food source for earthworm competing with OPs	Decrease over 40% for the heavier PAHs	Gomez-Eyles et al. (2011)
MWCNTs	0.1 and 1 g/kg	Nonylphenol (5,10 mg/kg)	Earthworm (*Eisenia fetida*)	Cell membrane damage by CNMs and high availability of CNMs-adsorbed-OPs	Increase	Hu et al. (2013)
Biochar	0.05%, 0.5% and 5%	^{14}C-catechol (0.014 mg/mL)	Earthworm (*Metaphire guillelmi*)	Grinding of soil particles in gut passage and surfactant-like substances in gut fluid	Increase by 69.5–112.7%	Shan et al. (2014)

Campos et al., 2014; Shan et al., 2014). Therefore, the influence of ECMs on earthworm accumulation of OPs should also be considered. Meanwhile, due to different uptake route of organisms to OPs, the impact of ECMs can be contradictory. More than by porewater uptake, some species are able to access adsorbed OPs or direct ingest soil particles (Chen et al., 2017; Huang et al., 2017b; Hurtado et al., 2017; Khorram et al., 2016). Therefore, how the ECMs affect bioavailability of OPs to microorganism, plants and earthworm should be separately studied.

On the other hand, although positive role of ECMs in environmental remediation has been recently reported, environmental risks associated with ECMs has become an issue of growing concern (Zeng et al., 2013b). For example, the application of biochar has been questioned due to the release of toxic compounds that are detrimental to soil organism (Buss et al., 2015; Oleszczuk et al., 2013). Altered soil microbial community, reduced seed germination and earthworm avoidance in biochar amended soil have been documented (Buss and Masek, 2014; Masiello et al., 2013; Tammeorg et al., 2014). Moreover, CNMs exhibit stronger toxicity than biochar due to smaller size and higher content of catalyst metals. Potential risks of CNMs have long been studied. Knowledge of CNMs-induced toxicity is diverse, which can be roughly divided into those mediated by inherent toxicity of CNMs (e.g. oxidative stress), or by attachment to organisms and consequently hindering their physiological activity, but also by toxic substances along with CNMs (Barbolina et al., 2016; Rajavel et al., 2014; Tu et al., 2013). Hence, a comprehensive understanding of how ECMs adversely affect soil biota is essential.

Overall, research on the interactions among ECMs, OPs and organisms is important to advance our understanding of the environmental impacts of ECMs and their performance in contaminated soil remediation. In this paper, firstly, we explored the impact of ECMs on the bioavailability (i.e. biodegradation/bioaccumulation) of OPs to various organisms. Subsequently, the possible risk of ECMs on soil biota is discussed at different levels (e.g. cellular, individual and community level). Moreover, future research needs of the risk assessment of ECMs are highlighted.

2. How do ECMs affect bioavailability of OPs?

ECMs are generally thought to decrease bioavailability of OPs in soil due to their strong sorption capacity. A modeling experiment by Marchal et al. (2013b) showed that it was the low desorption rate of PAH rather than bacterial activity that restricted PAHs mineralization. Moreover, microbial debromination ratio of BDE-47 was dropped by 92.8%–98.2% with 5.0% amendment of ECMs (Zhu et al., 2016a). Similar findings have been observed in plant accumulation of OPs. This may be attributed to the decrease of freely dissolved fraction of OPs in soil pore water, which is the primary form to be assimilated by plants roots in soil (Khorram et al., 2016). For example, addition of 5% biochar decreased turnips uptake of PAH by approximately 84% in the study of Khan et al. (2015), and a 2.5% and 5% amendment of biochar reduced concentration of emerging organic contaminants (i.e. bisphenol A, caffeine, carbamazepine, clofibric acid, furosemide, ibuprofen, methyl dihydrojasmonate, tris(2-chloroethyl)phosphate, triclosan, and tonalide) by 34–48% in lettuce roots and 23–55% in lettuce leaves in the study of Hurtado et al. (2017). Hamdi et al. (2015) also reported that CNTs significantly reduced bioaccumulation of pesticide by lettuce, with 88% decrease in roots and 78% in shoots by decreasing pesticide bioavailability. In addition to plant uptake, after amendment of 2% biochar, earthworm accumulation of fomesafen also decreased by 49.5%–52.9% compared to the control treatment (Khorram et al., 2016). Petersen et al. (2009) also proved that soils amended with 3.0 mg/g CNTs significantly decreased pyrene bioaccumulation in earthworms. Moreover, it has been reported that the acute toxicity to earthworm *Eisenia fetida* induced by complex of multi walled carbon nanotubes (MWCNT) and sodium pentachlorophenate was lower than