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Biochars change the sorption and degradation of thiacloprid in soil: Insights into chemical and biological mechanisms☆

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ABSTRACT

One interest of using biochar as soil amendment is to reduce pesticide adverse effects. In this paper, the sorption and degradation of thiacloprid (THI) in a black soil amended by various biochars were systematically investigated, and the mechanisms therein were explored by analyzing the changes in soil physicochemical properties, degrading enzymes and genes and microorganism community. Biochar amendment increased THI sorption in soil, which was associated with an increase in organic carbon and surface area and a decrease in H/C. Amendments of 300-PT (pyrolyzing temperature) biochar promoted the biodegradation of THI by increasing the microbe abundance and improving nitrile hydratase (NHase) activity. In contrast, 500- and 700-PT biochar amendments inhibited biodegradation by reducing THI availability and changing NHase activity and THI-degradative *nth* gene abundance, and instead promoted chemical degradation mainly through elevated pH, active groups on mineral surface and generation of •OH and other free radicals. Furthermore, THI shifted the soil microbial community, stimulated the NHase activity and elevated *nth* gene abundance. Biochar amendments also changed soil bacterial community by modulating soil pH, dissolved organic matter and nitrogen and phosphorus levels, which further influenced THI biodegradation. Therefore, the impact of biochars on the fate of a pesticide in soil depends greatly on their type and properties, which should be comprehensively examined when applying biochar to soil.

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1. Introduction

Thiacloprid (THI), a second-generation neonicotinoid insecticide, has been widely used to control agricultural pests (Jeschke et al., 2011). However, due to its high water solubility (184 mg/L) and low octanol-water partition coefficient ($\log K_{ow} = 1.26$) (Morrissey et al., 2015), the potential of THI to migrate into surface or underground water is now a major concern due to its extensive use in agricultural systems (Botias et al., 2015). Moreover, the recently reported adverse effects on non-target organisms have made these concern more serious (Hallmann et al., 2014; Kessler et al., 2015; Morrissey et al., 2015; Rundlof et al., 2015). Hence, effective approaches to mitigate its ecological risk in agricultural fields are clearly needed.

The fate of pesticides in soil depends on various environmental processes, including evaporation, sorption-desorption, transfer and transformation, and bioaccumulation, etc (Yang and Sheng, 2003; Yavari et al., 2015). Among the above processes, sorption occurs immediately upon the introduction of pesticides to soil, which impacts the magnitude of other processes and ecological effects, such as leaching, chemical transformation, bioavailability and ecotoxicity to non-target organisms (Yavari et al., 2015). THI shows low sorption capacity, which leads to its high bioavailability, and a preference for soil pore water (Jablonowski et al., 2013). Chemical and biological degradation are vital processes determining the dissipation of pesticides. The hydrolysis of nitriles into amides is a main chemical degradation pathway for THI, which is favored under alkaline conditions (Zhang et al., 2010). Reports on the microbial degradation of THI have focused on degradation using single bacterial strains isolated from soils, such as *Variovorax boronicumulans* and *Rhodotorula mucilaginosa*, and the nitrile hydratase (NHase) produced by bacterial strains plays a dominating role in its biodegradation (Dai et al., 2010; Liu et al., 2011; Zhang et al., 2012). The main product of the chemical and microbial

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degradations of THI in those studies is THI-$NCONH_2$ (Liu et al., 2011). In our previous study, we found that THI spiking could increase the relative abundance of phyla Proteobacteria and Bacteroidetes in soils, which could be related to its biodegradation (Zhang et al., 2018a).

Biochars have attracted widespread attention as soil amendments. Biochars show strong sorption capacity to various pollutants due to the high content of organic carbon (OC) with a highly aromatic nature, large surface area (SA) associated with abundant meso- and micro-pores, and polar active surface groups (Chai et al., 2012; Song et al., 2012; Yavari et al., 2015). Much work has been done concerning the sorption of various pesticides onto biochars and soils amended by biochars (Jablonowski et al., 2013; Xiao and Pignatello, 2015; Yavari et al., 2015). The sorption process involves multiple mechanisms, including hydrophobic partitioning, pore-filling, hydrogen bonding, p-π and π-π electron donor-acceptor (EDA) interactions, electrostatic effects and cation bridging, and sorption occurs through a combination of several of these mechanisms (Goulson and Kleijn, 2013; Inyang and Dickenson, 2015; Xiao and Pignatello, 2015). The sorption capacity and mechanism depend on the properties of the pesticide involved and biochar used, and hence, a specialized study is needed for any specific pesticide.

It has been well documented that the amendment of biochar in soil can enhance the sorption capacity for pesticides (Yavari et al., 2015; Ren et al., 2016). However, the sorption capacity of biochars may change after they are amended in soil. Soil components, e.g., dissolved organic carbon (DOC) or fine mineral particles, can compete with pesticides for binding sites or block/cover the meso- and micro-pores in biochars, thus decrease the sites available for pesticide sorption (Jin et al., 2016; Ren et al., 2016). Jin et al. (2016) found that the sorption of imidacloprid, isoproturon and atrazine in soil was enhanced by biochar amendment due to the increase in SA and OC contents. The authors also found that the SA and OC contents of the biochar-soil mixtures were lower than predicted by the sum of the corresponding values of soil and biochar, and prediction only based on biochar sorption capacity led to an overestimation of the sorption capacity in biochar-amended soil. Till now, neither the sorption of THI by biochar nor its sorption in biochar-amended soil has been studied, which hinders the application of biochar technology to control the adverse effects of THI in soil.

Moreover, the amendment of biochar not only affects the sorption of pesticides but also influences their transformation via several pathways. First, biochar can affect the soil microbial community composition and increase soil microbial biomass, resulting in enhanced pesticide biodegradation (Lopez-Pineiro et al., 2013; Nielsen et al., 2014; Xu et al., 2014). However, previous studies also revealed that biochar amendment significantly increases the sorption of pesticides and reduces their bioavailability to soil microbes, which weakens biodegradation (Chai et al., 2012; Nielsen et al., 2014; Oleszczuk et al., 2014). Hence, the opposite results for pesticide biodegradation due to the amendment of biochar may occur, which have been found to be associated with the biochar pyrolyzing temperature (PT), feedstock type, applied rate and soil type (Imparato et al., 2016; Jiang et al., 2016; Mitchell et al., 2016).

More recently, it was found that biochar can also influence the chemical degradation of pesticides. In our previous study, the catalytic effects of biochar derived from pig manure on the hydrolysis of carbaryl and atrazine were determined to result from the release of dissolved metal ions, the increase in pH and the presence of active groups on the biochar mineral surface (Zhang et al., 2013). In addition, it was reported that biochar derived from rape straw can promote the chemical and biological dechlorination of pentachlorophenol by enhancing electron transfer rates in contaminated paddy soils (Tong et al., 2014). It was proposed that environmentally persistent free radicals (EPFRs) exist in biochars and can react with organic contaminants directly or indirectly by activating small free radicals such as hydroxyl radical (•OH), sulfate radicals and superoxide radical anion (Fang et al., 2015a, 2015b; Qin et al., 2016; Yang et al., 2016). It was reported that EPFRs in suspensions of maize straw, wheat straw and pine needle biochars can induce •OH generation and promote the degradation of diethyl phthalate (Fang et al., 2015b) and 1,3-dichloropropene (Qin et al., 2016). However, the impacts of these EPFRs associated with biochar on pesticide transformation are unknown.

In summary, biochar may affect pesticide fate in soil via several processes and previous studies only concentrated on one or two processes, which is not propitious for the accurate manipulation of biochar technology on controlling pesticide adverse effects in soil. Therefore, the objectives of this study were to systematically examine the sorption and chemical and biological degradations of THI in a black soil (BS) before and after biochar amendment and to reveal the mechanisms therein. To this end, six biochar samples were obtained from maize straw and pig manure at three PTs: 300, 500 and 700 °C. Chemical degradation in a biochar suspension was designed, and the NHase activity, degradation genes and soil microorganism community in BS with and without biochar/THI were all monitored to explore the possible mechanisms involved in the changes in THI degradation. The results of this paper provide fundamental information regarding the impacts of biochar amendments on the fate of polar pesticides and for supervising the manipulation of biochar technology in the remediation of pesticide contamination.

2. Materials and methods

2.1. Chemicals

Technical-grade thiacloprid (THI, 97.5%) was purchased from Shandong Sino-agriculture United Biotechnology Company (Shandong, China), and its physicochemical properties are shown in Table S1 of the Supplementary Information (SI). THI-$NCONH_2$ (99%) was purchased from Laboratory of the Government Chemist. All other chemicals used in this paper were of analytical or HPLC grade. A stock solution of THI and THI-$NCONH_2$ were respectively prepared at 5000 mg/L in acetonitrile and stored at 4 °C.

2.2. Soil and biochar preparation and characterization

Black soil (BS) was collected at a depth of 0–20 cm from an agricultural field used for production of maize and wheat crops. The soil samples were separated into two parts; one was sieved through a 10-mesh sieve to remove cinders and stones after air drying and used for measurement of soil physicochemical properties and sorption experiments. The other portion of soil was used in degradation experiments directly after removing cinders and stones.

Maize straw and pig manure were air-dried, ground, and passed through a 10-mesh sieve. The dried feedstocks were charred at 300, 500 or 700 °C for 4 h in a closed 300-mL ceramic crucible with a cap under oxygen-limited conditions in a muffle furnace. The produced biochars were ground to pass through 40–200 mesh (0.075–0.425 mm) sieves and stored in 500-mL glass jars. Based on their feedstock and PT, the biochars are designated as Mn (from maize straw) and Pn (from pig manure), with n = 3, 5, 7 indicating PTs of 300, 500 and 700 °C, respectively. When the PT is not indicated, the biochars are designated as MBs and PBs as a whole.

Biochars were added into BS at a quality fraction of 2% with sufficient mixing. The analysis methods of the physicochemical properties of BS, biochars and soil-biochar mixtures are shown in

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