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## Combined effects of dissolved humic acids and tourmaline on the accumulation of 2, 2′, 4, 4′, 5, 5′- hexabrominated diphenyl ether (BDE-153) in *Lactuca sativa*<sup> $\frac{1}{5}$ </sup>



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#### ABSTRACT

In order to investigate the effects of dissolved humic acid (DHA) and tourmaline on uptake of 2, 2', 4, 4', 5, 5'- hexabrominated diphenyl ether (BDE-153) by Lactuca sativa, different fractions of DHA, including DHA<sub>1</sub> and DHA<sub>4</sub>, as well as different doses of tourmaline were introduced into BDE-153 contaminated solutions for plant growth. The levels of BDE-153 in L. sativa tissues were positively correlated with the Fe levels ( $R^2 = 0.9264$ ) in seedings of the treatments with different doses of tourmaline. However, when adding DHA<sub>1</sub> and DHA<sub>4</sub> into the system, the correlation coefficients ( $R^2$ ) decreased to 0.6976 and 0.5451 from 0.9264, respectively. In contrast with the Fe contents, the presence of DHAs didn't affect the  $R^2$ between the levels of BDE-153 and the lipid contents in plant tissues, Our results indicated that both DHA<sub>1</sub> and DHA<sub>4</sub> could severely alter the BDE-153 uptake by L. sativa through reducing the Fe uptake instead of the lipid contents. Additionally, DHA<sub>4</sub> exhibited much stronger abilities to alter the BDE-153 accumulation than DHA<sub>1</sub>. Transmission electron microscopy (TEM) observations indicated that either DHA<sub>1</sub> or tourmaline or co-treatment with DHA and tourmaline had no negative impact on *L. sativa* at the cellular level. The present study provides important information for the impacts of different fractions of DHA extracted from soil on the BDE-153 migration in plant systems. Moreover, we elucidated the importance of the iron in tourmaline for migration of the polybrominated diphenyl ethers (PBDEs) in plant systems.

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#### 1. Introduction

Polybrominated diphenyl ethers (PBDEs), which have been widely used in the domestic and industrial materials to reduce their flammability (Abbasi et al., 2015), are emerging contaminants and have been detected in various environmental matrices, including water, sediment and soil, air, as well as living organisms such as

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plant and animal tissues, even in human milk and blood samples (Guo et al., 2016; Kim et al., 2016; Lake et al., 2011; Marchitti et al., 2013; Wang et al., 2014a,b,c). PBDEs in the environment may pose potential risks to living organisms and human health. 2, 2′, 4, 4′, 5, 5′- hexabrominated diphenyl ether (BDE-153) is the most widely investigated congener among PBDEs due to its environmental abundance and toxicity (Birnbaum and Staskal, 2004). Its high hydrophobicity (log  $K_{\rm ow}=7.6$ ) determines that BDE-153 is easily to be adsorbed into the soil matrix, which is one of major reservoirs for accumulating environmental pollutants (Wu et al., 2015). Approximately 3900 ng/g of PBDEs was found in sludge amended agricultural soil in Sweden (Sellström et al., 2005).

It is highly possible that the accumulated PBDEs in plant tissues could transfer via food chains and eventually end up in human body. Previous studies demonstrated that the agricultural plants

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can take up and accumulate PBDEs in both shoots and roots (Bizkarguenaga et al., 2016; Deng et al., 2016; Huang et al., 2011). For example, a concentration gradient of PBDEs accumulation was observed in pumpkin, maize and ryegrass with the highest concentrations in the roots followed by the stems and lowest in the leaves (Huang et al., 2010). Therefore, the concern over the PBDEs migration in agricultural plants has drawn increasing attention.

PBDEs accumulation in agricultural plants might be impacted by soil organic matters (SOM), which play a key role in altering environmental behaviors of organic pollutants. Humic acid (HA) is one of the most important components of SOM, and can affect the speciation, transfer, and bioavailability of heavy metals (Lamelas and Slaveykova, 2007) and hydrophobic organic contaminants (HOCs) (Haitzer et al., 1998). HAs have different elemental compositions, functionalities, and conformations. Previous work investigated the sorption mechanisms between different fractions of dissolved humic acid (DHA) and organic compounds such as phenanthrene, lindane, and atrazine (Wang et al., 2011). Their sorption was affected by surface and bulk alkyl carbon contents of the sorbents, rather than aromatic carbon, which is closely dependent on different fractions of DHA. The results implied that with sequential HAs extraction, the surface and bulk particles polarity of HA both decreased. However, the effects of different fractions of DHA on the migration of PBDE in plants are still unclear. Guo et al. (2001) compared the difference between low molecular weight (LMW, 1 kDa) and high molecular weight (HMW, 1 kDa-0.2 µm) of dissolved organic matter (DOM) fractions extracted from seawater on trace metals (Cd, Co, Hg, Cr, Ag, Zn) uptake by American Osyters (Crassostrea virginica). The results suggested that low concentrations (0.5 ppm) of DOM significantly decreased bioavailability of colloidally complexed metals, but a generally enhanced uptake at higher DOM concentrations was evident (Guo et al., 2001). Thus, it is important to figure out the mechanistic processes of different fractions of DHA to change organic pollutant uptake by plants, which could provide the direct evidence for establishing the efficient phytoremediation technique.

Minerals were commonly used in the soil amendment or pollutant removal from aqueous solutions. Also, DHA or DOM is ubiquitous in soil and water. However, there are very few studies regarding the impacts of DHA or DOM combined with minerals on the organic pollutants uptake by plants. Jones and Tiller (1999) compared phenanthrene sorption between dissolved and claybound HA, and found that sorption values for the clay-bound HA were lower than that of the bulk HA prior to adsorption onto clay mineral surfaces. The results implied that the conformation of the humic substance on the surface of the minerals determined the level of phenanthrene sorption. On the other hand, Fe, an abundant element in soil, could alter the arsenic (As) removal by binding with humic substances (Giasuddin et al., 2007) and subsequently result in changes of As bioavailability to plants. Thus, it is critical to figure out the jointed effects of DHA and Fe from the mineral, both of which are considered as soil amendments, on the environmental pollutant migration in the agricultural plant. Tourmaline is a hydrous siliceous material and its general formula is expressed as XY<sub>3</sub>Z<sub>6</sub>Si<sub>6</sub>O<sub>18</sub> (BO<sub>3</sub>) W<sub>4</sub>, where the X site is occupied by Na<sup>+</sup>, K<sup>+</sup>, Ca<sup>2+</sup> or Mg<sup>2+</sup>; and the Y sites are occupied by Li<sup>+</sup>, Fe<sup>2+</sup>, Fe<sup>3+</sup>, Al<sup>3+</sup>, Mg<sup>2+</sup> or Ti<sup>4+</sup>; and the Z sites are occupied by Fe<sup>3+</sup>, Cr<sup>3+</sup>, Al<sup>3+</sup>, Mg<sup>2+</sup> or Fe<sup>2+</sup>. Tourmaline itself has some specific properties, including producing an electrostatic field and releasing macroelements such as K, Mg and Ca ions, and generating spontaneous and permanent poles (Nakamura and Kubo, 1992). Due to its unique physicalchemical properties, tourmaline has been widely applied in health care products. Our previous studies indicated that tourmaline could also efficiently remediate historic heavy metals and PBDEs and PAHs and OCPs contaminated soils (Li et al., 2016; Wang et al., 2014a,b,c). However, the previous studies of the effects of tourmaline on removing environmental pollutants from either soil or water by plants have not taken into considerations the factors of DHA or DOM, both of which could notably determine the organic pollutant uptake by plants. Therefore, beside the tourmaline, we also included different fractions of DHA into the plant system to figure out whether the interactions between tourmaline and DHAs could significantly alter the fate and behaviors of PBDEs in ecosystem.

Different fractions of DHA play the different roles in governing organic pollutants sorption in the aqueous solutions. Tourmaline could decrease the contents of organic pollutants in soil remediation. We proposed that the interactions between different fractions of DHA and tourmaline could significantly affect the organic pollutant migration in plant. Thus, we chose *Lactuca sativa*, widely planted in the China, as a target plant. Different fractions of DHA were isolated from the same soil collected from an electronic waste recycling plant located in Jinghai county in Tianjin, China. Tourmaline was purchased from Hebei province, China. We aimed to investigate the roles of different fractions of DHA as well as different doses of tourmaline in BDE-153 uptake by L. sativa and to further elucidate the correlations of the levels of BDE-153 with the Fe contents released from different concentrations of tourmaline as well as with the total lipid contents in plant tissues. Comprehensive understandings of the roles of different fractions of DHA and tourmalines in altering PBDE transport in plant could help to evaluate PBDEs distribution in plant tissues as well as at each trophic level in food chains. Furthermore, we may use the knowledge to examine the benefits of tourmaline applications in in situ remediation.

#### 2. Materials and methods

#### 2.1. Materials

BDE-153 (50 mg/L) in isooctane was purchased from Accustandard (New Haven, CT, USA). Acetone, dichloromethane and hexane were of analytical grade. Granular anhydrous sodium sulfate and silica gel of 100–200 mesh were purchased from Jiangtian Chemical Cooperation (Tianjin, China).

Iron-rich black tourmaline (particle size: 1000 mesh) was custom-made by Lingshou Minerals Processing Co., Ltd. (Hebei, China). The chemical compositions of the tourmaline (w/w, %) were as follows: SiO<sub>2</sub>, 37.44%; Al<sub>2</sub>O<sub>3</sub>, 29.79%; Fe<sub>2</sub>O<sub>3</sub>, 20.2%; CaO, 0.71%; Na<sub>2</sub>O, 2.18%; MgO, 0.55%; K<sub>2</sub>O, 0.14%; TiO<sub>2</sub>, 0.49%; B<sub>2</sub>O<sub>3</sub>, 0.78%, and others 7.72%. The methods of the tourmaline characterizations were described previously (Yu et al., 2014).

#### 2.2. Preparation and characterization of different fractions of DHA

Soil (0–10 cm in depth) was sampled from the area near an electronic waste recycling plant located in Jinghai county (116°46′30.07″ W, 38°49′22.55″ N) in Tianjin, China. The collected soil was air-dried and passed through a 2-mm sieve. Different fractions of DHA were sequentially extracted from the soil by following the methods suggested by Kang and Xing (2005). Briefly, 25 g soil was placed into 500 mL flasks containing 250 mL of 0.02 mol/L Na<sub>4</sub>P<sub>2</sub>O<sub>7</sub> solution. The mixture was shaken on a shaker (HYG-A, Jiangsu, China) for 4 h and was centrifuged at 150 rpm for 15 min. The supernatant was collected, adjusted pH to 1.0 using 6 mol/L HC1 and then sit on bench for 12 h. The precipitate was collected by centrifugation and the pH of the supernatant was adjusted to 13.0 using 1 mol/L NaOH. The adjusted solution was centrifuged again and the collected supernatant was the first fraction of DHA (DHA<sub>1</sub>). The extraction procedure was repeated by

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