



Degradation of dibutyl phthalate in two contrasting agricultural soils and its long-term effects on soil microbial community

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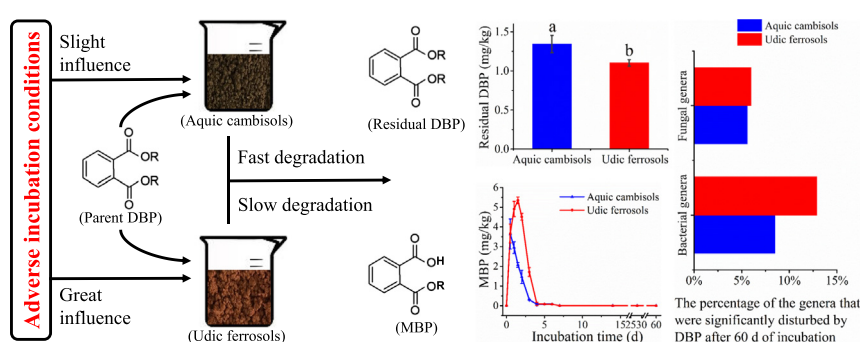
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HIGHLIGHTS

- DBP was degraded faster in aquatic cambisols than in udic ferrosols.
- High levels of DBP residue (1.10–1.34 mg/kg) were detected after 60 days.
- High concentration, low temperature and moisture slowed DBP degradation.
- DBP produced a long-term disturbance of soil microbial community structure.

GRAPHICAL ABSTRACT



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ABSTRACT

Due to its widespread application and large-scale production, dibutyl phthalate (DBP) has become one of the most frequently identified phthalic acid esters (PAEs) in soils. The fate of DBP and its effects on microbial communities in soils with contrasting properties have seldom been studied. In this study, the degradation of DBP and its long-term effects on the soil microbial community were investigated in aquatic cambisols and udic ferrosols. The half-lives of DBP in aquatic cambisols and udic ferrosols were found to be 0.286–1.41 days and 0.870–20.4 days, respectively, indicating that DBP was degraded faster in aquatic cambisols. In addition, the degradation of DBP in aquatic cambisols was less vulnerable to adverse incubation conditions, including high DBP concentration, low temperature and low moisture. These results can be ascribed to the higher microbial abundance and activity in aquatic cambisols than in udic ferrosols. During DBP degradation, the toxic metabolite monobutyl phthalate (MBP) was present only transiently and did not accumulate in the two soils. After 60 days of incubation, the degradation-resistant DBP residue concentrations were as high as 1.10 and 1.34 mg/kg, and the relative abundance of 8.51%–12.9% of bacterial genera and 5.59%–6.02% of fungal genera was significantly disturbed by DBP in both test soils. The results from this study highlight the need to comprehensively evaluate the environmental risks of degradation-resistant DBP residues and the impact of DBP contamination on soil microbial functions.

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1. Introduction

Phthalic acid esters (PAEs) are widely used in various products, such as general plastic products, cosmetics, personal care products, paints, pesticides and medical products (Wang et al., 2013; Zhang et al.,

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2015). The global annual production of PAEs is approximately 6 million tons, more than one-third of which are consumed in China (Wang et al., 2015). Due to their widespread application and large-scale production, PAEs can enter the environment during their manufacture, use and disposal (Kong et al., 2012). Therefore, PAEs have been detected in various environmental matrices worldwide (Niu et al., 2014; Wang et al., 2013). One of the most frequently identified PAE compounds is dibutyl phthalate (DBP), which has been classified as an environmental priority pollutant and an environmental endocrine disruptor chemical by the U.S. Environmental Protection Agency.

DBP concentrations ranging from 0.04 to 29.4 mg/kg have been found in soils across China (He et al., 2015; Niu et al., 2014). According to the allowable concentration in soil (0.08 mg/kg) and the clean-up objective (8.10 mg/kg) of DBP set in the U.S. by the New York State Department of Environmental Conservation (2010), a considerable portion of soils contain high DBP concentrations. It has been proven that high DBP concentrations in soil have toxic effects on microorganisms, animals and plants (M.L. Gao et al., 2017; Ma et al., 2016; Wang et al., 2016). In addition, soil DBP can even harm human health through the food chain (Chen et al., 2017; Sun et al., 2015). For these reasons, knowledge of the fate of DBP is necessary to minimize its environmental and health risks.

Studies using soils inoculated with DBP-degrading bacteria, compost-amended soils or sludge-amended soils have indicated that the half-life of DBP ranges from 1.5 to 10.33 days (Chang et al., 2009; Li et al., 2006; Liang et al., 2008; Zhao et al., 2016). In addition, natural soils have a strong ability to degrade DBP. As reported by Xu et al. (2008), DBP degrades at very high rates, with a half-life of 7.8–8.3 days in natural soils. The focus of most existing studies has been the half-life of DBP. However, although degradation-resistant DBP residues are an important part of the environmental fate DBP, limited information on them is available. There has been growing recognition that the bound organic contaminant residues in soils can be released and pose risks to human and ecosystem health under certain conditions (Y.Z. Gao et al., 2017).

Monobutyl phthalate (MBP) is the primary degradation intermediate of DBP (Sun et al., 2015) and has been reported to be responsible for the adverse effects of DBP (Ema and Miyawaki, 2001). Wang et al. (2012) detected MBP in soil samples at levels of 9.2–57.1 µg/kg. Although a previous study suggested that MBP could be quickly degraded in natural sediments (Otton et al., 2008), there are concerns over whether MBP, formed from the degradation of DBP, could accumulate in soils. Therefore, to comprehensively understand the fate and risk of DBP in soils, it is necessary to investigate the accumulation dynamics of MBP.

As reviewed by Staples et al. (1997) and Liang et al. (2008), microorganisms play crucial roles in the degradation of DBP in soils. Therefore, soil incubation conditions, such as the initial DBP concentration, temperature, and moisture, could affect the degradation rate of DBP by altering the soil microbial activity. For example, increasing the soil temperature or moisture can enhance the degradation rate of DBP (Chen et al., 1997). However, conditions other than temperature and moisture have rarely been studied. In addition, little research has been conducted regarding the influences of soil incubation conditions on the DBP degradation rate in soils with contrasting physicochemical and biological properties.

Although soil microorganisms can degrade DBP, they are also influenced by DBP contamination. Wang et al. (2016) reported that DBP could alter the soil microbial diversity after 25 days of contamination.

However, soil microorganisms have the ability to adapt to environmental disturbances (Li et al., 2013). Therefore, considering only the short-term effects of DBP on soil microorganisms may result in an overestimation of its environmental risks. As reported by Cheng et al. (2014), the influences of organic pollutants on soil microorganisms decrease with contamination time. It is unclear whether microorganisms in different soils could recover in a relatively longer time after DBP contamination.

Hence, systematic studies on the fate of DBP in different soils and its long-term effects on the soil microbial community are urgently needed to obtain a comprehensive understanding of its environmental risks in agricultural soils. The objectives of this study included the following: (1) to elucidate the degradation dynamics, degradation intermediates and degradation-resistant residues of DBP in two contrasting agricultural soils; (2) to evaluate and compare the impacts of initial DBP concentration, temperature, moisture, light and sterilization on DBP degradation; and (3) to reveal the long-term effects of DBP on the soil microbial community.

2. Materials and methods

2.1. Chemicals and reagents

DBP and MBP standards were obtained from Dr. Ehrenstorfer GmbH (Augsburg, Germany). DBP (analytical grade, 99.9%) was purchased from Aladdin Industrial Corporation (Shanghai, China). All organic solvents used, such as acetonitrile, acetone and methanol, were HPLC grade and purchased from Tedia Company (Fairfield, OH, USA). Deionized water (>18.2 MΩ) was prepared using a Milli-Q system (Millipore, USA). Before use, anhydrous magnesium sulfate (MgSO₄) and sodium chloride (NaCl) were baked at 400 °C for 6 h to eliminate potential contamination. Before use, all glassware was dipped in detergent solution, washed ultrasonically for 30 min, oven-dried, immersed in sulfuric acid for 30 min, and washed with tap water and deionized water, each three times in sequence, and finally oven-dried and rinsed with acetone. No plastic items were used in the experiment.

2.2. Soil sampling

Two contrasting soils, aquic cambisols and udic ferrosols, were collected from the top 20 cm of agricultural land in Fengqiu, Henan Province, China (35°00'N, 114°24'E), and Yingtan, Jiangxi Province, China (28°12'N, 116°56'E), respectively. There were no contaminant sources around the sampled lands, and no plastic film was historically used. Field-moist soil samples were transported to the laboratory at a low temperature and immediately sieved through a 2-mm stainless steel mesh. After sufficient mixing, a portion of the homogenized soil samples was stored at 4 °C in a refrigerator for no >1 week for the degradation experiments. The remaining portion of the homogenized soil samples was air-dried and further sieved to determine the chemical and microbial properties of the soils (Table 1), following the methods of Cheng et al. (2014) and Lu (2000). Aquic cambisols have higher organic matter (OM) content, pH value and microbial activity and abundance than do udic ferrosols. The concentrations of DBP in these two soils were below the detection limit.

2.3. Experimental procedures

Soils were taken from the refrigerator and preincubated at 25 °C for 7 days at 50% of their maximum field water-holding capacity (WHC). At

Table 1
Chemical and microbial properties of the test soils.

Soil	pH	Organic matter (g/kg)	Total N (g/kg)	Microbial biomass carbon (mg/kg)	Respiration (mg/kg/h)	Shannon index
Aquic cambisols	8.33	19.3	0.962	211	2.86	4.58
Udic ferrosols	5.15	8.39	0.383	53.6	0.521	4.05

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