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Review article

Soil contamination and sources of phthalates and its health risk in China: A review☆

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ABSTRACT

Phthalates (PAEs) are extensively used as plasticizers and constitute one of the most frequently detected organic contaminants in the environment. With the deterioration of eco-environment in China during the past three decades, many studies on PAE occurrence in soils and their risk assessments have been conducted which allow us to carry out a fairly comprehensive assessment of soil PAE contamination on a nation-wide scale. This review combines the updated information available associated with PAE current levels, distribution patterns (including urban soil, rural or agricultural soil, seasonal and vertical variations), potential sources, and human health exposure. The levels of PAEs in soils of China are generally at the high end of the global range, and higher than the grade II limits of the Environmental Quality Standard for soil in China. The most abundant compounds, di-*n*-butyl phthalate (DBP) and di-(2-ethylhexyl) phthalate (DEHP), display obvious spatial distribution in different provinces. It is noted that urbanization and industrialization, application of plastic film (especially plastic film mulching in agricultural soil) and fertilizer are the major sources of PAEs in soil. Uptake of PAEs by crops, and human exposure to PAEs via ingestion of soil and vegetables are reviewed, with scientific gaps highlighted.

1. Introduction

Phthalic acid esters (PAEs, or simplier phthalates) are extensively used as plasticizers in industrial products such as plastics and polyvinylchloride to enhance their plasticity and versatility, and also widely used in non-polymeric products, such as solvents in adhesives, cosmetics, paints, (Staples et al., 1997; Zolfaghari et al., 2014; Steinmetz et al., 2016). Global annual production of solid plastics was about 300 million tones each year (Garcia1and Robertson, 2017) with China one of the largest consumers. PAEs are not chemically bound to the polymeric and non-polymeric products, and hence, they can easily migrate from products and have been released as xenobiotic and hazardous compounds into the environment (Kong et al., 2012; Benjamin et al., 2015).

Many studies have reported the ubiquitous nature of PAEs in air and water (Philip et al., 2018), sludge (Cai et al., 2007; Zolfaghari et al., 2014), soil (Cai et al., 2008a; Niu et al., 2014; Net et al., 2015a; Wang et al., 2017; Sun et al., 2018), and biota (Mo et al., 2009; Wang et al., 2015). The wide occurrence of PAEs in the environment can result in human potential exposure to PAEs via ingestion, inhalation and dermal absorption from different sources (e.g., food, water, air, dust and soil) (Guo et al., 2012; Net et al., 2015a), as confirmed by the high detection frequencies of phthalate metabolites in human matrice samples (e.g., urine, blood) from different countries (Guo et al., 2014; Net et al., 2015a; Bui et al., 2017; Polinski et al., 2018). Numerous epidemiological and toxicological studies have demonstrated that some PAE compounds are endocrine disrupting chemicals (Bui et al., 2017; Chen et al., 2017a). Butyl benzyl phthalate (BBP) and di-(2-ethylhexyl) phthalate (DEHP) are classified as probable and possible human carcinogens by US Environmental Protection Agency (USEPA), respectively (USEPA, 2007). The global occurrence of PAEs and their various health effects have raised increasing concern of public, scientific and governmental organizations. The environmental protection agencies of various countries including US, Canada, China, and EU (European

☆ This work does not deal with research or studies on human subjects or experimental animals.
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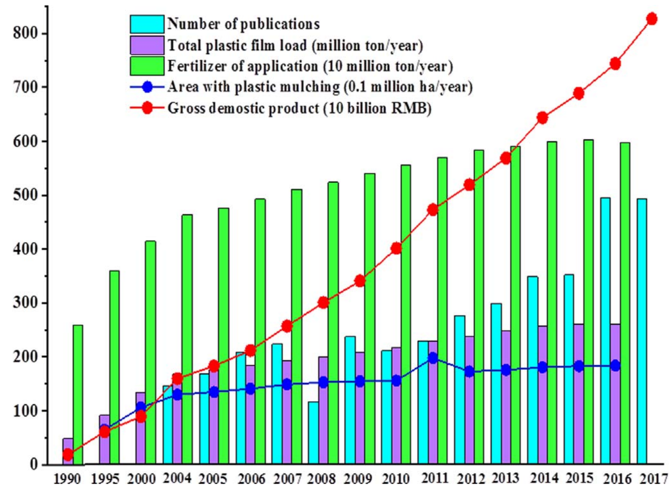

Fig. 1. Gross domestic product (GDP) in Chinese currency (Ren Min Bi, or simply RMB), fertilizer amounts of application, total plastic film load (million ton/year), area with plastic mulching (0.1 million ha/year), and the number of scientific publications related to phthalate (searching "phthalate" in Title of publication within *Web of Science* of Thomson Corporation).

Union) have classified some PAE compounds (e.g, DEHP) as priority pollutants. The published papers about PAEs have increased from 147 to 212 from 2005 to 2010, and up to 494 in 2017 (searching "phthalate" in titles of publication within *Web of Science* of Thomson) (Fig. 1).

China has experienced rapid urbanization, industrialization and agricultural modernization in the past decades. These activities have released large amounts of harmful metals and organic contaminants, which have severely deteriorated the environmental quality (Cai et al., 2008a; Ministry of Environmental Protection and Ministry of Land and Resources of China, 2014; Sun et al., 2018). A recent nationwide survey of soil pollution in China show that 16.1% of the soil samples, 19.4% of agricultural soil samples, are contaminated by heavy metals (e.g., cadmium, arsenic) and/or organic pollutants (e.g., PAEs, polycyclic aromatic hydrocarbons (PAHs)) if compared to soil environmental quality limits of China (GB15618-1995) (Ministry of Environmental Protection and Ministry of Land and Resources of China, 2014). As mentioned above, China is one of the largest producers and consumers of PAEs, the plastic products in 2016 was up to 77 million tonnes (CPPIA, 2016). In particular, agricultural boom in the past decades has been coupled with widespread applications of plastic film (Fig. 1 and Table 1), leading to higher residues of PAEs in the environment. PAE contamination in soil, water, and air of China is relatively severe when compared with other countries or regions (Cai et al., 2008a; He et al., 2015; Sun et al., 2016).

Recently, various soil survey and monitoring programs have been conducted in China (Niu et al., 2014; Zhang et al., 2015a; Sun et al., 2016; Wang et al., 2018). Abundant data on the occurrence of PAEs in soil and human health risk have been published in the past, which provides the opportunity to assemble in an integrated report on the occurrence and health effects of soil PAEs. Hence, the present work aimed to (1) gather all the existing data of soil PAEs published after 1996 to provide a critical analysis of PAE spatial and temporal distribution in soil, (2) analyze their potential sources, (3) discuss the uptake and accumulation of PAEs by crops; (4) assess the human health risk of exposure to soil PAEs. Finally, data gaps are highlighted, and recommendations for further research are given.

The literature cited in this review was mainly searched from the databases of Web of Science and China National Knowledge Infrastructure (http://www.cnki.net/). It should be noted that the number of PAE compounds determined in the literature varied from 3 to 16 (Table 1). Most studies have reported the occurrence of six PAE priority pollutants classified by USEPA (namely, BBP, DEHP, dimethyl phthalate (DMP), diethyl phthalate (DEP), di-*n*-butyl phthalate (DBP), and di-*n*-octyl phthalate (DOP)). Nevertheless, the total concentrations of PAEs (Σ_{PAEs}) may be the sum of six PAE priority pollutants classified by US EPA (Σ_{6PAEs}), or the sum of sixteen PAE compounds (Σ_{16PAEs}), or others. The difference in summation of PAE compounds has complicated comparison of Σ_{PAEs} among different studies. Thus, all comparisons of PAE levels attempted among regions or cities are based on the reported results, especially based on DBP and DEHP levels which normally account for the largest part of the Σ_{PAEs} (Cai et al., 2008a; Niu et al., 2014; Zhang et al., 2015a; Sun et al., 2016; Wang et al., 2018). In this review paper, it is assumed that no big differences exist between the results produced by all reporting scientists/laboratories due to analytical uncertainties/biases. The authors are aware of the fact that differences between laboratories are common because various extraction procedures (microwave-assisted extraction, supercritical fluid extraction, ultrasonic or accelerated solvent extractions) as well as gas/liquid chromatography and mass spectrometry (GC, GC-MS, LC-MS) detection techniques are used for analyzing soil PAEs (Net et al., 2015b). In addition, all mean values mentioned in the present review are arithmetic mean values based on dry weight, unless otherwise mentioned.

2. Occurrence and distribution of PAEs in soil

2.1. Regional variation of soil PAEs

Numerous studies have been conducted to investigate the levels of soil PAEs in China. The main regions and cities studied are located in North China (e.g., Beijing, Tianjin, Jinan), Northeast China (e.g., Heilongjiang, Liaoning, Jilin, Sanjiang plain), East China (e.g., Yellow River Delta, Nanjing, Xiangyang, Hangzhou, Taizhou, Nanchang) and South China (e.g., Pearl River Delta, Guangzhou, Dongguan, Huizhou, and Guilin), particularly in the Yellow River Delta and the Pearl River Delta (Fig. 2). The soil types investigated include urban soil (e.g., residential, roadside, park), agricultural soil (e.g., vegetable, orchard, farmland), natural and reclaimed wetland soil. In this work, data of PAEs in soil were aggregated based on the regions or location sampled (Table 1).

Big variations were found among different provinces/regions/cities, even within the same city (e.g., Beijing, Guangzhou) (Table 1 and Fig. 2). The total concentrations of 16 PAEs (Σ_{16PAEs}) varied from 0.001 to 1232 mg/kg (Table 1), with the maximum concentrations, being considerably higher than the respective concentrations reported in soils from the Netherlands (Peijnenburg and Struijs, 2006), Denmark (Vikelsøe et al., 2002), France (Tran et al., 2015), or Serbia (Škrbić et al., 2016). Extremely high Σ_{6PAEs} (sum of six PAEs: 124–1232 mg/kg) were observed in soils of cotton field in South Xinjiang (Guo and Wu, 2011). High Σ_{16PAEs} (322 mg/kg) was also found in a site of urban soil of Guangzhou (Zeng et al., 2009). Relatively high Σ_{6PAEs} (referred to the sum of six USEPA PAE compounds) was also detected in the soil of non-industrialized region (from 2.1 to 158 mg/kg with a mean of 19.1 mg/kg) and an electronics manufacturing area (from 8.6 to 172 mg/kg with a mean of 35.3 mg/kg) in Xiangyang (Wu et al., 2015). PAE concentrations in the contaminated area were significantly higher than the non-industrialized area (Wu et al., 2015). The maximum value of Σ_{PAEs} in other studies was all less than 50 mg/kg, generally less than 10 mg/kg (Table 1).

High mean or median Σ_{PAEs} were recorded in the soils of the Pearl River Delta region (21 mg/kg; Cai et al., 2005; Cai et al., 2008a), at the non-industrialized region and electronics manufacturing area of Xiangyang (Wu et al., 2015), being significantly higher than in other regions (< 10 mg/kg in general) (Table 1). The average levels of Σ_{6PAEs} in Pearl River Delta region (Cai et al., 2005) and Xiangyang (Wu et al., 2015), and the maximum of Σ_{PAEs} in some regions such as Beijing (Zhang et al., 2014) and Tianjin (Kong et al., 2012) were higher than the stringent grade II limits for PAEs (10 mg/kg) in arable soils recommended by the

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