



Review

An overview of phthalate acid ester pollution in China over the last decade: Environmental occurrence and human exposure

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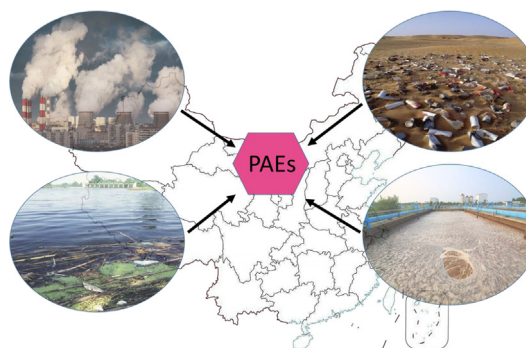
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HIGHLIGHTS

- PAEs were detected ubiquitously and abundantly in China.
- DEHP was the most abundant phthalates in various environmental media.
- PAEs concentrations exhibit a location dependent.
- Limited treatment might cause the widespread contaminations of PAEs.

GRAPHICAL ABSTRACT



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ABSTRACT

The toxicity and bioaccumulation potential of phthalate acid esters (PAEs) make their impact on the environment a matter of considerable concern. Due to China's recent economic development and population growth, it has become one of the largest manufacturers and consumers of PAEs, with an associated contamination threat to several environmental compartments. The aim of this overview is to present a systematic account of PAE occurrence in various environmental media in China in the last decade, including the air, surface water, sediments, soil, sewage, and sludge; human exposure to PAEs is also evaluated. This reveals a location-dependence that can be attributed to regional differences in economic and industrial development as well as specific geographic location. A need for further study into the transportation and transformation behavior of PAEs in different environmental media and into PAE control technologies is identified, as a means of effectively assessing potential ecological and health risks.

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1. Introduction

Phthalate acid esters (PAEs) are widely used as plasticizers to enhance the technical properties of polyvinyl chloride (PVC) (Mackintosh et al., 2004; Staples et al., 1997). Finished plastic products such as plastic films, medical devices and tubing may contain 20–60% PAE by weight (Gómez-Hens and Aguilar-Caballeros, 2003). From 2007 to 2017, the worldwide yield of PAEs increased from 2.7 to nearly 6 million tons per year (Bauer and Herrmann, 1997; Xie et al., 2007). Over that period, restrictive regulations have caused European and North American production to stagnate (EU 2004, 2005, 2007, 2008; CPSI, 2008), and now the majority of output is consumed by developing countries such as Brazil, China, and India. In China, PAEs are extensively applied in daily necessities, food packaging, medical equipment, industrial piping, upholstery, agriculture plastic films, etc. In 2009, it was estimated that 1.3 million tons of PAEs were consumed annually, primarily di(2-ethylhexyl) phthalate (DEHP) and dibutyl phthalate (DnBP) (CPCIA, 2009).

Since PAEs are not covalently bonded to the polymeric matrix in soft PVC and exist in a leachable form, they migrate into the environment easily upon disposal of phthalate-containing products through plastic aging and decomposition (Afshari et al., 2004; Fujii et al., 2003; Loraine and Pettigrove, 2006). PAEs have been demonstrated to be endocrine-disrupting chemicals (EDCs). The US Environmental Protection Agency has identified six congeners, including di-methyl phthalate (DMP), di-ethyl phthalate (DEP), di-*n*-butyl phthalate (DnBP), di(2-ethylhexyl) phthalate (DEHP), di-*n*-octyl phthalate (DnOP) and butylbenzyl phthalate (BBP), as priority pollutants.

The indiscriminate use of these compounds has led to the sporadic occurrence of residues in various environmental compartments, which point to the potential for PAE pollution. Strong concern regarding PAEs has been expressed in recent years due to their ubiquity and occurrence in high concentrations. As a result of their extensive application and the variety of pathways through which they can be transported, a significant amount of PAE residue reaches even remote environments through atmospheric transport, landfill leaching, agricultural run-off, and sewerage discharge (Thuren and Larsson, 1990; Xie et al., 2007; Gao et al., 2014). The compounds and their metabolites have been detected in many environmental media, including the air, rainwater/storm water, surface water, sediments, soils, and biota (Gasperi et al., 2009; Loraine and Pettigrove, 2006). Some widely used PAEs such as DnBP and DEHP have been detected at relatively high levels worldwide, even in regions where governmental restrictions on the production and use of PAEs are already in place such as Europe and the United States (Clark, 2003; Dargnat et al., 2009a; Hutzinger, 1980; Peijnenburg and Struijs, 2006).

In addition, there is increasing evidence suggesting that some PAEs have an acute and chronic toxic effect on both aquatic and terrestrial organisms, including humans (Staples et al., 1997). The typical symptoms of phthalate toxicity in the embryos of aquatic organisms are death, tail

curvature, necrosis, cardio edema, and no touch response. BBP, DnBP, and DEHP have estrogenic endocrine disrupting effects on intact organisms (Chen et al., 2014). Human exposure to PAEs could potentially cause adverse effects on the reproductive, hepatic and renal systems (Hauser and Calafat, 2005; Swan, 2008; Liu et al., 2012). There is increasing concern about uncertainties regarding PAE exposure and the risks that PAEs may pose to human health and the environment.

Meta-analysis into spatio-temporal trends in the fate of environmental PAEs has shown a location-dependent effect, with differences between Europe and other countries such as USA and Canada (Bergé et al., 2013). However, there is lack of systematic information on PAE occurrence in environmental media in China. Therefore, this overview aims to summarize the current status of PAE contamination in both the non-natural and natural environments of China, elucidate the distribution, behavior, and fate of these compounds in various environmental compartments, analyze geographical trends, and assess possible threats to ecosystem and human health. The data are extracted from published literature in research journals and national reports published over the last decade.

2. Environmental occurrence of PAEs in China

2.1. Atmosphere

Due to the low volatility of PAEs, their participation in the atmospheric environment can be expected to be limited. They have, however, been found in both the air and rainwater. PAEs enter the atmosphere mainly through volatilization in waste combustion (Salapasidou et al., 2011). In urban areas, industrial emissions and automobile exhaust have been found to be the primary sources of atmospheric PAEs (Staples et al., 1997; W. Wang et al., 2013; He et al., 2015). In Tianjin, a mega-city adjacent to the capital of China, DEHP has been shown to be the predominant PAE compound in both PM_{2.5} (particles up to 2.5 µm in size) and PM₁₀ (particles up to 10 µm in size), with concentrations up to 98.29 ng/m³ (Table 1), followed by DnBP, with a maximum concentration of 12.9 ng/m³. The concentrations of other PAEs were all below 1 ng/m³ (Kong et al., 2013). In Nanjing, the air was moderately polluted by DEHP (up to 357 ng/m³), followed by DnBP (up to 89 ng/m³) in aerosols. DnBP was the most commonly detected abundant PAE in both urban and suburban areas of the city (P. Wang et al., 2008; Wang et al., 2012). The total trend of PAEs occurrence in atmosphere are shown in Fig. 1, the average levels of DEHP was significantly higher than other PAEs in China.

Research in Greece showed significantly higher DEHP concentrations in an urban traffic area (from 4.63 to 45.0 ng/m³) than on an urban industrial site (up to 6.50 ng/m³), implying that PAEs may have entered the air from vehicular emissions (Salapasidou et al., 2011). Similarly, PAE concentrations have been shown to be about 5.8 times higher at industrial sites than background sites in the Netherlands (Peijnenburg and Struijs, 2006). PAEs have also been detected in

Table 1
Occurrence of PAEs in atmosphere (range/mean value, ng/m³).

Location	Remark	Media	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Tianjin	Urban area	PM ₁₀	0.88	0.73	12.90	0.15	98.29	0.83	(Kong et al., 2013)
		PM _{2.5}	0.54	0.3	8.72	0.08	75.68	0.33	
Nanjing	Summer	Aerosols			1.8–89		95–357		(Wang et al., 2007)
	Winter				3.4–34		25–333		
Nanjing	Urban center	Gas-phase	3.9–21.8	1.1–9.3	28.1–66.7	1.0–7.1	6.8–28.7	0.3–2.4	(P. Wang et al., 2008)
		Particle phase	0.7–3.3	0.1–0.9	4.1–17.9	0.15–1.7	1.1–11.4	0.05–0.5	
	Suburban	Gas-phase	1.9–7.3	0.08–3.7	4.1–20.0	0.01–2.1	3.4–12.0	n.d.	
		Particle phase	0.01–0.5	0.03–0.4	0.7–7.8	0.02–0.7	0.1–2.9	0.01–0.7	
Nanjing	Urban area	Suspended particulates	n.d.–75.0		22.0–150.0		4.9–52.0		(Wang et al., 2012)
Taizhou	Summer	PM _{2.5}	0.02		7.99–13.74		80.62–164.87		(Gu et al., 2010)
	Winter		0.18–0.49		45.71–52.54		97.68–191.82		
Norwegian Sea			0.01–0.22	0.18–0.90	0.16–0.43	0.02–0.07	0.08–0.46		(Xie et al., 2007)
Netherlands	Background locations				2–31		0.73–57		(Peijnenburg and Struijs, 2006)
	Industrialized area				4–70		0.73–333		
	Populated area				5–17		0.73–70		
France	Urban area of Paris	Total	0.5	10.7	22.2	4.6	18.9	0.5	(Teil et al., 2006)
Greece	Urban-industrial site		n.d.	n.d.	1.20–3.36	0.11–0.80	n.d.–6.50	n.d.–0.11	(Salapasidou et al., 2011)
	Urban-traffic site		n.d.	n.d.	0.43–2.40	0.04–0.98	4.63–45.0	n.d.–0.11	(Clark, 2003)
U. S.							5.00–132		
U.S.	Urban area of New York	Gas-phase			3.73–3.59				
U.S.	California	Gas-phase		n.d.–610	n.d.–18.0	n.d.–8.5	n.d.–230		(Rudel et al., 2010)
PAEs in rainwater (range/mean value, µg/L)									
Gansu, China	(Semi-) arid Loess Plateau	Rainwater			>1		>1	<1–>1	(Zhu et al., 2004)
Netherlands		Rainwater	0.008–0.018	0.24–0.43	0.28–0.88	0.14–0.26	0.69–1.7	0.038–0.25	(Vethaak et al., 2005)
France	Urban area of Paris	Rainwater	0.116	0.333	0.592	0.081	0.423	0.010	(Teil et al., 2006)

rainwater in China, with DEHP and DnBP both at levels of over 1 µg/L (Zhu et al., 2004). Ambient concentrations of DEP and BBP have been reported in the United States that are substantially higher than those in suburban and urban locations in China and in Paris (Rudel et al., 2010). Comparable PAE concentrations in rainwater have also been detected in Europe (Teil et al., 2006; Vethaak et al., 2005).

2.2. Surface water

Most PAE compounds display stable chemical properties in this environment due to their low volatility, low water solubility, and high lipophilicity. The contribution of hydrolysis and photolysis in water to the overall environmental degradation of PAEs, including DnBP and BBP, is usually low (Wolfe et al., 1980), and DEHP and DnOP do not hydrolyze in water (Thomas, 2010). Thus, abiotic hydrolysis is not expected to play a significant role in determining the environmental fate of phthalate acid esters. The hydrolysis rate of PAEs, which relates to their structural characteristics and environment, usually ranges from approximately 4 months for DMP to 100 years for DEHP (Wolfe et al., 1980). The photo-oxidation process of PAEs is also relatively slow, with half-lives ranging from 2.4 to 12 years for DEP and DnBP and 0.12 to 1.5 years for DEHP (Howard, 1991).

Surface water is considered to be the component of the natural world that is most affected by human activity. The occurrence of PAEs in surface water has been monitored around the world, including in rivers, lakes, groundwater, and even tap water (Table 2). In China, the occurrence of PAEs in surface water has been documented more widely in southern cities than northern cities, with the highest levels, 41 µg/L DnBP and 101 µg/L DEHP, being found in the Haihe river (Chi, 2009). As the largest tributary in the Heilongjiang river basin of Northeastern China, there was cause for concern regarding PAE contamination of the Songhua River. The authors of the current review detected all six PAEs in the water of the Harbin section of the river. Concentrations of both DnBP and DEHP exceeded 10 µg/L, with most of the highest concentrations occurring downstream of sewage treatment plants (STPs)

(Gao et al., 2014). This study indicated that STP discharge is a potential source of surface water pollution by PAEs. In addition, Wen et al. showed that DEHP was found as the primary PAE congener in Songhua River mainstream and its tributaries, the PAEs concentration was found to be influenced by branch inflow and land usage (Wen et al., 2018).

PAEs have also been detected in drinking water sources, ranging from 0 µg/L to 6.6 µg/L in the Mopanshan Reservoir (which serves as the source of drinking water for Harbin city) and the Yangtze River (which serves as the source of drinking water for Nanjing city) (Wu et al., 2009). DnBP and DEHP were the major pollutants in the Yangtze River, with concentrations of up to 35.65 µg/L and 54.73 µg/L (F. Wang et al., 2008). The total trend of PAEs occurrence in surface water in China are shown in Fig. 1, the average levels of DnBP and DEHP was higher than others, it is consistent with the widespread use of plasticizers. For instance, 134,000 and 394,000 tons of DnBP and DEHP were produced globally (Xu et al., 2008). PAE-levels in surface water in China are comparable with other areas of the world, including Europe (Bendz et al., 2005; Dargnat et al., 2009b; Fromme et al., 2002; Vethaak et al., 2005), the US (Solis et al., 2007), Canada (Keil et al., 2011; Mackintosh et al., 2006) and Malaysia (Santhi and Mustafa, 2013), where concentrations are generally under 100 µg per liter. The surface water of Nigeria and South Africa, however, has been shown to be highly contaminated by PAEs, with a concentration of DEHP exceeding 2300 µg/L being reported, much higher than in any other published data (Adeniyi et al., 2011; Fatoki and Noma, 2002).

2.3. Soil

PAE contamination in soil is mainly caused by the application of plastic agricultural films, municipal biosolids, agricultural chemicals, and wastewater irrigation (He et al., 2015). The study of PAE uptake and metabolism by plants shows that some PAE compounds can migrate through soil and reach groundwater or enter the food supply through accumulation in crops or plants (Sun et al., 2015). PAEs have been detected in several soil types in China, at levels ranging from

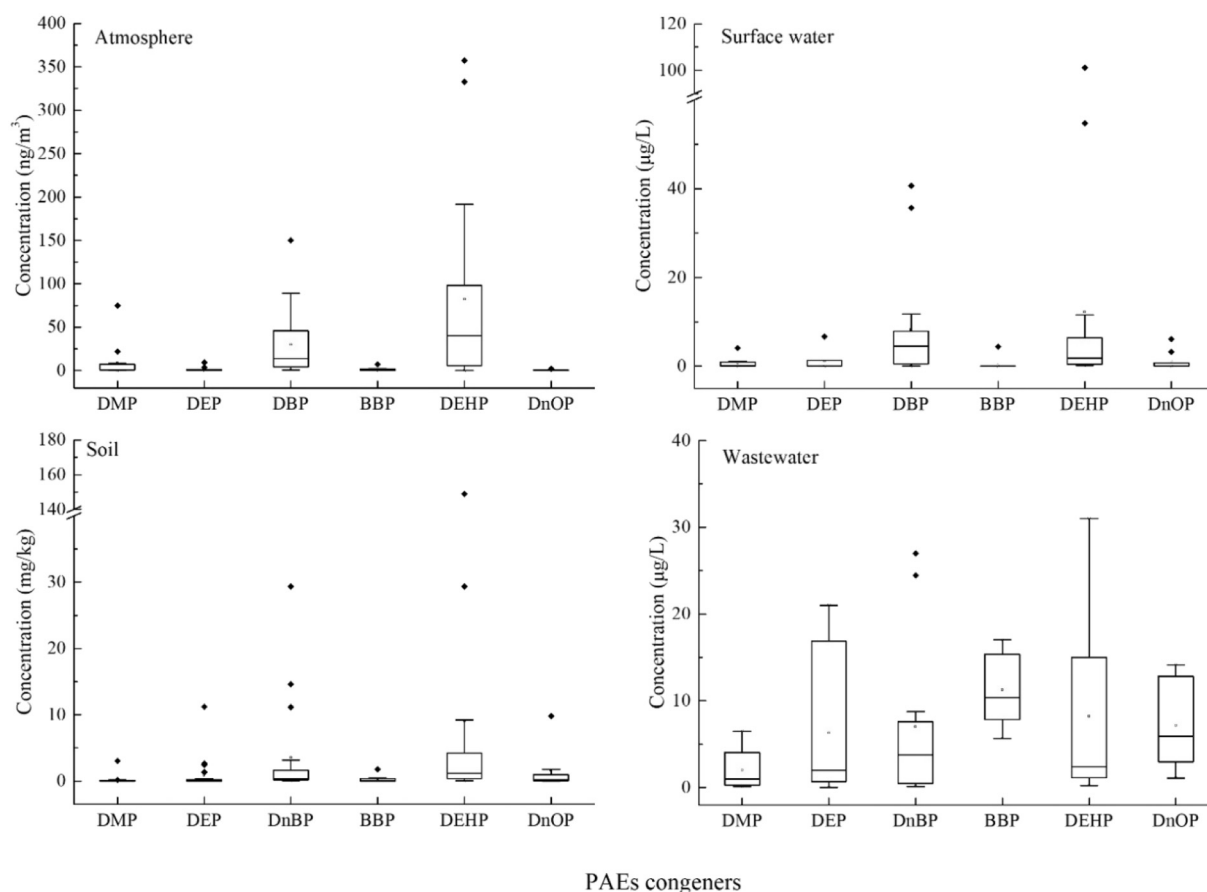


Fig. 1. PAEs concentrations in difference phases.

thousandths of a milligram (mg) to several milligrams per kilogram (Table 3). Higher levels have been reported from northern China than southern China (Hu et al., 2003). The highest concentrations of PAEs in soils were found in cotton fields south of Xinjiang, with DnBP ranging 11.15–57.7 mg/kg and DEHP ranging 103.5–149 mg/kg (Guo and Wu, 2011). Xu et al. measured the concentrations of DnBP and DEHP in two types of arable soil (fluvo-aquic soils and black soils). The results demonstrated that DnBP (2.75–29.37 mg/kg) was the major pollutant rather than DEHP (0.49–7.99 mg/kg) and that PAE concentrations in

the fluvo-aquic soils from Handan are higher than in black soils from Harbin (Xu et al., 2008). Kong et al. showed much higher PAE levels in arable soils than in wasteland and that there was a clear link between the contamination of vegetable soils by DEHP and DnBP and the consumption of plastic films (Kong et al., 2012).

Sun et al. reviewed the priority organic contaminants including PAEs across China, founding that the PAEs concentrations in North and Central China were higher, but since 2005, PAEs pollutants in South China had been on the rise (Sun et al., 2018). All of this research suggests

Table 2

Occurrence of PAEs in surface water (range/mean value, µg/L).

Location	Types	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Songhua River watershed	Songhua River	0.065–0.208	0.140–0.334	0.190–4.762	–	0.364–2.682	ND–0.621	(Wen et al., 2018)
Harbin	Mopanshan Reservoir	n.d.–0.042	n.d.–0.055	0.053–4.498	n.d.	0.129–6.571	n.d.–0.448	(Liu et al., 2013)
Tianjin	Haihe River	–	–	0.35–40.68	–	3.54–101.1	–	(Chi, 2009)
Nanjing	Yangtze River	0.003–0.499	0.0073–0.051	0.01–6.165	n.d.–0.044	1.197–6.205	–	(Wu et al., 2009)
Nanjing	Taihu lake	0.037–0.097	0.017–0.021	3.35–7.88	n.d.–0.025	0.446–0.752	–	(Gao et al., 2011)
Wuxi	Wujing River	n.d.	n.d.	0.528–5.067	n.d.–0.07	0.513–1.483	–	(Zhang et al., 2011)
Hefei	Chaohu Lake	0.123–1.106	0.133–0.157	0.924–5.43	0.066–0.076	0.162–0.231	0.032–0.04	(He et al., 2013)
Wuhan	Yangtze River	n.d.–0.1	n.d.	n.d.–35.65	–	3.9–54.73	0.07–3.2	(F. Wang et al., 2008)
Other countries								
Sweden	Surface water	–	0.01–0.03	0.02–0.06	–	0.01–0.04	–	(Bendz et al., 2005)
Germany	Surface water	–	–	0.12–8.8	–	0.33–97.8	–	(Fromme et al., 2002)
Netherlands	Surface water	<0.0045–0.19	<0.07–2.3	<0.066–3.1	<0.01–1.8	0.9–5	<0.002–0.078	(Vethaak et al., 2005)
France	Marne River	0.030–0.050	0.025–0.092	0.140–0.220	0.023–0.035	0.307–0.708	n.d.	(Dargnat et al., 2009b)
Canada	False Creek Harbor	0.0035	–	–	0.0035	0.275	–	(Mackintosh et al., 2006)
U. S.	Eleven Point River	–	–	0.16–1.36	0.07–0.14	–	–	(Solis et al., 2007)
	White River	–	–	0.14–4.14	0.04–0.35	–	–	
Malaysia	Selangor River	0.002–0.028	0.006–0.113	0.028–0.306	0.002–0.021	0.051–0.507	0.0002–0.014	(Santhi and Mustafa, 2013)
Nigeria	Ogun river catchments	n.d.	1480–1755	2080–2705	–	255–480	–	(Adeniyi et al., 2011)
South Africa	Port Elizabeth harbor	0.03–350.8	0.03–398.3	1–1028	–	2.1–2306	–	(Fatoki and Noma, 2002)

Table 3
Occurrence of PAEs in soils (range/mean value, mg/kg).

Location	Types	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Northeast China	Arable soils (3 cities)	n.d.	0.18–1.36	0.16–1.56	–	0.51–2.15	–	(Hu et al., 2003)
Northwest China	(2 cities)	n.d.	0.18–0.25	0.38–0.39	–	1.67–2.17	–	
Northern China	(4 cities)	n.d.–0.2	0.15–2.61	0.14–0.98	–	0.51–2.18	–	
Eastern China	(7 cities)	n.d.	n.d.–1.29	0.21–1.38	–	0.2–5.98	–	
Southern China	(4 cities)	n.d.	n.d.–0.17	n.d.–0.26	–	0.54–3.42	–	
Southwest China	(3 cities)	n.d.	n.d.–0.37	0.51–0.64	–	1.02–2.08	–	
Xinjiang	Cotton fields	n.d.–3.01	n.d.–2.42	11.15–57.7	–	103.5–149	–	(Guo and Wu, 2011)
Sanjiang Plain	Topsoil	0.0266	0.0349	0.0285	–	0.0279	–	(X.L. Wang et al., 2013)
Harbin	Black soils	–	–	2.75–14.62	–	0.49–4.20	–	(Xu et al., 2008)
Handan	Fluvo-aquic soils	–	–	3.18–29.37	–	1.15–7.99	–	
Beijing	Greenhouse soils	<0.01–0.02	<0.01–0.05	0.34–1.66	–	0.22–0.74	<0.09	(Ma et al., 2003)
Tianjin	Farmland	0.003–0.088	0.003–0.081	0.007–0.189	0–1.79	0.039–2.37	n.d.–0.647	(Kong et al., 2012)
	Vegetable	0.002–0.101	0.002–0.114	0.013–0.285	0–0.358	0.028–4.17	0–9.78	
	Orchard	0.003–0.032	0.003–0.03	0.02–0.138	0–0.125	0.026–0.358	n.d.–0.728	
	Wasteland	0.003–0.073	0.005–0.059	0.009–0.147	0–0.471	0.051–0.494	n.d.–1	
Binzhou, Yellow River Delta	Urban soil	0.002–0.060	n.d.–0.004	0.245–2.058	n.d.	1.465–6.320	n.d.–0.044	(H. Yang et al., 2013)
	Suburb	0.001–0.065	0.001–11.24	0.166–1.450	n.d.	0.710–4.473	n.d.–0.142	
	Rural	0.001–0.005	n.d.–0.001	0.136–1.039	n.d.	0.431–2.449	n.d.–0.068	
Yangtze River Delta	Agriculture soil	0.0002–0.071	0.0005–0.0906	n.d.–1.5	n.d.–0.0122	n.d.–9.19	n.d.–0.273	(Sun et al., 2016)
Nanjing	Vegetable soils	n.d.–0.012	n.d.–0.007	n.d.–0.046	n.d.–0.002	0.204–0.704	0.002–0.019	(J. Wang et al., 2013)
	Using plastic film	n.d.–0.016	n.d.–0.027	n.d.–1.41	n.d.–0.041	0.034–7.033	n.d.–1.739	
Guangzhou	Agricultural soil	0.001–0.157	0.001–0.178	0.009–2.74	n.d.–0.18	0.107–29.37	n.d.–0.084	(Zeng et al., 2008)
China (national scale)		n.d.–0.016	n.d.–11.24	n.d.–2.74	n.d.–1.79	n.d.–29.37	n.d.–9.78	
Other countries								
Denmark	Agriculture soil	n.d.	0.009	0.061	n.d.	0.182	n.d.	(Laternus and Grøn, 2007)
U. K.	Brickearth	0.0001	0.0002	0.008		0.022	0.012	(Gibson et al., 2005)
	Gault clay	0.0001	0.0009	0.0079		0.076	0.014	
Netherlands				0.006				(Peijnenburg and Struijs, 2006)

that PAE pollutants in soils are largely derived from human agricultural activities. However, urbanization may also enhance PAE contamination. Yang et al. found that PAEs occur ubiquitously in the soil of the city of Binzhou in the Yellow River Delta, and that DEHP was the dominant type in urban areas, with concentrations ranging from 1.465 mg/kg to 6.3 mg/kg, while DnBP was predominant in suburban soil, with concentrations ranging from 0.001 mg/kg to 11.24 mg/kg (H. Yang et al., 2013). Higher PAE levels in agricultural soil have been reported in the Yangtze River Delta (YRD), which accounts for >15% of China's GDP, than any other region (Sun et al., 2016). Few studies have reported the level of PAEs in the soils of other countries in the last decade, but where data is available, it indicates much lower concentrations than the Chinese average (Laternus and Grøn, 2007; Gibson et al., 2005; Peijnenburg and Struijs, 2006). In China, total trend of PAEs occurrence in soil are shown in Fig. 1, the average levels of DEHP and DnBP were significantly higher than other PAEs, the observations that there is more soil contamination by PAEs in the south than in the north and that soil PAE concentrations have exhibited an increasing trend from 2003 to date (Table 3), together with regional variations in the reported PAE concentration, indicate that this pollution is related to rapid urbanization and economic growth (Niu et al., 2014).

3. Occurrence and fate of PAEs in the sewage treatment process

The fate of PAEs in STPs has received much attention because the ubiquity and persistence of the compounds mean that they may contaminate the environment after discharge from STPs. The transportation, transformation and ultimate environmental fate of PAEs depend upon the persistence of the compounds and their biodegradation. The occurrence of PAEs in STPs has been investigated in many countries all around the world. However, in contrast with developed countries such as those in Europe and the United States, data concerning PAEs in Chinese STPs are relatively scarce. Investigations have mostly been conducted in densely populated cities.

3.1. PAEs in sewage

The most common group of PAEs, DEHP and DnBP, have been found ubiquitously in STP sewage. DEP and DMP also frequently occur, while DnOP and BBP are less commonly reported (Table 4). In Harbin, the largest provincial capital in Northeast China, PAEs were detected in the influents at detection frequencies ranging from 42% to 100% and DEHP and DnBP concentrations of up to 30.99 µg/L and 24.46 µg/L, respectively (Gao et al., 2014). This work also reported that the efficiency with which three of the studied STPs removed some PAEs was limited, implying a potential risk that sewage-sourced PAEs were being discharged into the Songhua River. DnBP, DEP, and DEHP were found in sewage in Beijing at mean concentrations of 27 µg/L, 21 µg/L and 15 µg/L, respectively (Zhou and Liu, 2013). The level of PAEs in Beijing was higher than in Tianjin, where DnBP and DEHP were found in sewage at concentrations of up to 0.6 µg/L and 2.4 µg/L, respectively (Zhang and Wang, 2009). Generally, PAEs occurred in sewage shown no significant difference in sewage in China (Fig. 1). Moreover, the PAE levels in sewage in China are similar with those in other areas of the world, such as Europe (Clara et al., 2010; Dargnat et al., 2009b; Roslev et al., 2007; Sanchez-Avila et al., 2009; Vogelsang et al., 2006), the US (Jackson and Sutton, 2008), Canada (Barnabe et al., 2008) and Australia (Tan et al., 2007a), where reported values are generally below 100 µg/L. In contrast, concentrations over 100 µg/L have been detected in two Nordic countries, Finland (Marttinen et al., 2003a; Marttinen et al., 2003b) and the Netherlands (Vethaak et al., 2005), much higher than in most published data for China.

3.2. Removal efficiency and the fate of PAEs in STPs

The efficiency with which STPs remove PAEs and their ultimate fate have been extensively documented, with studies suggesting that performance relates to the treatment process and variations in conditions. The removal efficiencies for DEP and DEHP in a Norwegian STP were in the range of –300%–>90% and –60%–>90%, respectively (Vogelsang

Table 4

Occurrence of PAEs in wastewater (range/mean value, µg/L).

Location	Media	DMP	DEP	DnBP	BBP	DEHP	DnOP	Reference
Harbin	Influent (3 STPs)	4.05–6.49	2.25–16.86	8.73–24.46	n.d.–5.67	2.42–30.99	4.63–12.84	(Gao et al., 2014)
	Effluent	n.d.–1.52	n.d.–1.37	3.47–4.13	n.d.–17.03	1.7–25.4	1.11–14.15	
Harbin	Influent	0.779	1.977	6.411	n.d.	n.d.	7.152	(Kong et al., 2008)
	Effluent	0.111	0.022	2.674	n.d.	n.d.	2.953	
Beijing	Influent	0.98	21	27	n.d.	15	n.d.	(Zhou and Liu, 2013)
	Effluent	0.27	0.69	5.7	n.d.	2.5	n.d.	
Tianjin	Influent			0.21–0.583		1.12–2.382		(Zhang and Wang, 2009)
	Effluent			0.14–0.32		0.24–0.478		
Other countries								
Finland	Influent (4 STPs)	n.d.–1	<1–74	3.0–9.0	<1–5	28–122	n.d.–2	(Marttinen et al., 2003a)
	Effluent	n.d.–<1	n.d.–4	n.d.–6	n.d.–<1	2–8	n.d.–1	
Norway	Influent (5 STPs)		1.3–4.9			<0.5–21		(Vogelsang et al., 2006)
	Effluent		<0.5–9.9			<0.5–34		
Netherlands	Untreated municipal sewage	0.39–6.2	0.0041–44	<0.38–51	0.56–4.9	<13–100	0.26–2.4	(Vethaak et al., 2005)
	Sewage	<0.002–1.3	<0.35–5.2	<0.69–21	<0.17–1.3	1–150	0.012–2.8	
	Effluent (2 STPs)	<0.003–0.32	<0.3–0.93	<0.42–0.48	<0.07–0.29	<0.47–2.4	<0.002–0.19	
Denmark	Influent	0.269–4.31		15.34–24.67	9.41–80.74	53.23–84.1		(Roslev et al., 2007)
	Effluent	n.d.–0.237		1.83–2.73	1.99–4.33	2.08–9.93		
Austria	Influent (2 STPs)	0.26–0.81	1.2–2.7	<0.1–0.47	n.d.–0.26	4.1–13	n.d.–<0.1	(Clara et al., 2010)
	Effluent	n.d.	n.d.–<0.1	n.d.	n.d.	<0.2–1.3	n.d.	
France	Influent	0.82 ± 1.13	7.71 ± 5.2	11.10 ± 0.37	1.12 ± 0.54	22.46 ± 13.22	0.10 ± 0.16	(Dargnat et al., 2009b)
	Effluent	n.d.	0.78 ± 0.22	0.15 ± 0.12	0.30 ± 0.12	5.02 ± 1.53	n.d.	
Spain	Influent	0.6 ± 0.5	50.7 ± 19	46.8 ± 15	0.67 ± 0.8	47.9 ± 25		(Sanchez-Avila et al., 2009)
	Effluent	0.13 ± 0.02	49.8 ± 2.8	n.d.	0.01 ± 0.03	1.28 ± 0.5		
Canada	Influent					70 ± 9.1		(Barnabe et al., 2008)
	Effluent					54 ± 7		
U. S.A	Influent		n.d.–10	n.d.–3.6	14	33	4.2	(Jackson and Sutton, 2008)
	Effluent		<1	<0.36	0.84	2.9	0.39	
Australia	Influent		1.08	0.201	0.134	0.716		(Tan et al., 2007b)
	Effluent		0.0049	0.0344	0.0757	0.589		

et al., 2006). Gao et al. found that long alkyl chain PAEs adsorb onto suspended organic matter in sewage due to their low solubility and the high log K_{ow} , hindering further degradation of the compounds. After going through processing, the suspended particles undergo hydrolysis, and eventually the residual PAEs are released in the effluents (Gao et al., 2014).

The processes affecting the fate of organic pollutants in STPs include volatilization, solid settlement, hydrolysis, biodegradation, and adsorption (Rogers, 1996). Biodegradation and adsorption are considered to be the major pathways responsible for the fate of PAEs in the treatment process. Both mechanisms can be affected by several factors, including the properties of the treatment process, temperature, and the initial concentration of target compounds. At the laboratory scale, Huang et al. investigated the fate of DnBP in the Anaerobic/Anoxic/Oxic (A/A/O) treatment process and found that under the optimal process parameters, about 72.66% of DnBP was biodegraded, 24.44% was accumulated in the system, and 0.5% remained in the waste sludge; only 2.4% was discharged with the effluent (Huang et al., 2010). However, such satisfactory performance was not observed in the full-scale A/A/O treatment process. Gao et al. provided a performance comparison of the Cyclic Activated Sludge Technology (CAST) process and Biological Nutrient Removal (BNR) treatments including the Anoxic/Oxic (A/O) and Anaerobic/Anoxic/Oxic (A/A/O) processes. PAE-removal by the BNR treatments was limited, while CAST performed better. This may be attributable to differences in the ambient temperature and recirculation mode. The results also indicated that biodegradation was the major pathway for PAE removal in the CAST process, while in the A/A/O process more PAE-removal occurred by adsorption by sludge; the short alkyl chain groups (DMP, DEP, and DnBP) tended to be removed by biotransformation rather than adsorption to the excess sludge, while the opposite held true for the longer alkyl chain groups (BBP, DEHP, and DNOP) (Gao et al., 2014). Marttinen et al. also found that at 20 °C, in SBRs with optimal aerobic/anoxic periods, the removal efficiency of DEHP from the water phase was able to reach >95% and the average biotransformation removal reached 42% (Marttinen et al., 2004).

Conversely, in a full-scale biological process operating at 18.5 °C, only 14% of the DEHP load in the influent was removed by biotransformation, based on an overall removal efficiency of 97%, and 68% by adsorption to the primary and excess sludge (Marttinen et al., 2003b).

It can be concluded that biodegradation and adsorption are the main PAE removal mechanisms in STPs. The proportion of PAEs adsorbed onto sludge increases with their alkyl chain length and the lipophilicity of the substance. These studies reveal that PAEs can be removed by adsorption without being completely metabolized or degraded, a vitally important finding in light of the potential environmental risks posed by the discharge of pollutants from STPs.

4. Ecological effects of and human exposure to PAEs

4.1. Effect of PAEs on environmental ecosystems

4.1.1. Aquatic toxicity

PAEs have been shown to affect reproduction in annelids (both aquatic and terrestrial), molluscs, crustaceans, insects, fish, and amphibians, to impair development in crustaceans and amphibians and to induce genetic aberrations (Oehlmann et al., 2009). Numerous studies have focused on the endocrine-disrupting effects of PAEs in different species of fish and mammals. A recent study reported that long-term exposure to DEHP disrupts endocrine function in marine fish, with males being more sensitive than females (Ye et al., 2014). The aquatic toxicity of PAEs varies widely across the class of compounds, with only the lower alkyl chain length (C1 to C4) consistently posing toxicity (Parkerton and Konkel, 2000). Literature data show that DMP, DEP, DnBP, and BBP consistently have both acute and chronic effects in freshwater and saltwater species of algae, invertebrates, and fish (Staples et al., 1997; Adams et al., 1995). PAEs with alkyl chain lengths equal to C6 (DEHP, DnOP) are not acutely toxic to aquatic organisms, with acute toxicity reported only at concentrations significantly above their water solubilities (Bradlee and Thomas, 2003).

4.1.2. Uptake by plants

Research has also focused on PAE bioaccumulation/bioconcentration in soil ecosystems. Cai and Mo surveyed the bioconcentration factors (BCF) of plants grown in PAE-contaminated soils and found values that were generally below 1 (Cai et al., 2008; Mo et al., 2009). Sun et al. carried out a cultivation study using lettuce, strawberry, and carrot plants to determine the potential of plant uptake of DnBP and DEHP and their primary metabolites mono-*n*-butyl phthalate (MnBP) and mono (2-ethylhexyl) phthalate (MEHP). BCF values for the four compounds ranged from 0.16 ± 0.01 to 4.78 ± 0.59 (Sun et al., 2015). Some research has reported that, though various vegetable plants have the ability to take up PAEs from soil, the translocation from roots to above-ground tissues is relatively weak. Once inside the plant, phthalate diesters are readily metabolized into their monoesters, highlighting the potential risk of human exposure via consumption of vegetables grown in PAE-contaminated soils (Wu et al., 2013; Sun et al., 2015).

The potential health risks of environmental PAEs are of grave concern because human beings are inevitably exposed to PAEs from contaminated surface water and organisms (Adeniyi et al., 2011). Reproductive developmental effects of PAEs have been observed in humans, including shortened anogenital distance in newborn boys and, in adults, shortened pregnancy, lower sex and thyroid hormone levels, and reduced sperm quality (ECB, 2008). Thus, PAEs in the ecosystem have attracted considerable attention due to the potential threat they pose to the environment and human health.

4.2. Human exposure to PAEs

4.2.1. Exposure pathways

Humans can be exposed to PAE compounds mainly through dietary intake, inhalation, and skin contact from the use of pharmaceuticals and personal care products (PPCPs) (Koniecki et al., 2011). Dietary intake is a major factor in exposure to PAEs because PAEs such as DEHP and DnBP are utilized as plasticizers in food plastic packaging (Clark, 2003). Observations have indicated that food uptake is the predominant source of DEHP intake in adults, making an approximately 98% contribution to total DEHP intake in adult populations (Fromme et al., 2007; Koch et al., 2003). For DnBP, a greater percentage of the daily intake comes from other sources than food, mainly the air and personal care products (Wormuth et al., 2006). Inhalation and dermal absorption are also considered to be the main pathway for exposure to DEP, which is frequently detected in PPCPs such as soap or shampoo (Chen et al., 2012; Latini, 2005).

In a study in China, DMP, DEP, DnBP, and DEHP were frequently detected (>60%) in food, with detection rates of up to 82%, 81%, >94% and 82%, respectively, whereas DNHP, DCHP, and DnOP were found in fewer than 16% (Guo et al., 2014). In the UK, Bradley et al. found 15 PAEs in retail foodstuffs, of which DEHP was the most frequently detected, in 66 of 261 samples (Bradley et al., 2013). Chang et al. used the hazard index (HI) to evaluate simultaneous exposure to PAEs in foodstuffs, finding that the highest exposure (0.443 mg/kg) and highest average daily dose (ADD) was to DEHP (Chang et al., 2014). Guo et al. estimated total PAE exposure doses for a population in Shanghai based on the assumption that the body weight of an average Chinese adult is 60 kg, and calculated that the total daily intakes of DMP, DEP, DnBP, and DEHP were 49.0 µg/day, 91.1 µg/day, 310 µg/day and 131 µg/day, respectively (Guo et al., 2014).

In 2011, the Taiwan Food and Drug Administration reported that DEHP and di-isononyl phthalate (DiNP) had been illegally added to clouding agents used in food and beverages. 965 contaminated products were identified, of which 206 were exported to 22 countries. DEHP concentrations ranged from 2.4 µg/g to 34.1 µg/g in contaminated beverages, and 2.3–2108 µg/g and 1.7–527 µg/g in contaminated food supplement capsules and powders, respectively (J. Yang et al., 2013). Yang et al. estimated that drinking 500 mL of sports drinks would result in an average DEHP intake of 140 µg/kg/day, which is several times that

recommended by government guidelines (20–60 µg/kg/day). The study suggested that consuming tainted drinks and food would lead to a daily intake of DEHP that greatly exceeds established safety guidelines, which should raise concerns about potential adverse effects (J. Yang et al., 2013).

PAEs in the atmosphere and dust present a potential hazard of exposure by inhalation and dermal absorption. The process by which indoor DEHP is emitted has been shown to be controlled by the external mass transfer, with the emission rate affected by environmental parameters such as temperature, relative humidity, and air flow rate (Xu et al., 2009). The contribution of indoor air has been shown to account for 3.1% and 1.7% of human exposure to DnBP and DEHP, respectively (Clark, 2003). DMP, DEP in the gas phase, and DEHP in the particle phase have been shown to be the most abundant compounds in indoor air (Pei et al., 2013). Guo et al. measured concentrations of nine PAEs in indoor dust samples collected from six cities in China and Albany, New York, USA. The results indicated that, in dust samples collected from China, DEHP, DnBP, and DIBP were the predominant compounds; concentrations of DEHP (100% detected) ranged 9.9–8400 µg/g dw, 10 times higher than the concentrations of DnBP (100% detected, 1.5–1160 µg/g dw) and DIBP (100% detected, 2.6–299 µg/g dw), while others were all below 50 µg/g dw. The extent to which indoor dust contributes to human exposure has been reported to vary from 2% to 5% of the estimated total daily intake (DI) in China (Guo and Kannan, 2011). Cao et al. applied an exposure scenario approach to show that the general population of China was exposed to DEHP and DnBP at 3.80 and 3.02 µg/kg bw/day, respectively, and that food intake was the largest contributor of the different sources of exposure (Cao et al., 2016). Plastic materials, cosmetic and PPCPs are important PAE sources in house dust. Zhang et al. showed that DnBP, DEHP, and BBP can all come from the same source and are significantly influenced by the use of solid-wood floor wax in flooring material (Zhang et al., 2013). For comparison, in Europe, conclusions based on a scenario approach showed that consumer products and various indoor sources dominate exposure to DMP and DEP, whereas food has a major influence on exposure to DnBP and DEHP (Wormuth et al., 2006).

4.2.2. Body loading

The different exposure pathways result in the presence of PAE metabolites in human blood and urine. These thus have the potential to be used as biomarkers for the early effects of environmental exposure on humans (Jiang et al., 2018; Zhang et al., 2016). Mono-2-ethylhexyl phthalate (mEHP) has been found in maternal blood, cord blood, and meconium specimens in 201 mother-newborn pairs from Shanghai, and mEHP levels in meconium specimens were highly correlated with those in cord blood (Li et al., 2013). Furthermore, urinary PAE metabolites can be used as biomarkers of human exposures to phthalates. Guo et al. analyzed 14 PAEs metabolites in 183 urine samples collected from Shanghai, Guangzhou, and Qiqihaer, China in 2011 (Table 5). All urine samples contained PAE metabolites, of which the total concentrations ranged from 18.6 to 3160 ng/mL. Mono-*n*-butyl phthalate (mBP) was the major metabolite, with a median concentration of 61.2 ng/mL, followed by mono-2-isobutyl phthalate (miBP) (56.7 ng/mL), indicating widespread exposure to the parent compounds of these two metabolites, DnBP and DIBP (Guo et al., 2011). Research into phthalate metabolites in the urine of Chinese young adults also showed that mBP and miBP were the dominant compounds, with concentrations of 67.0 and 57.2 ng/mL, respectively, and that the concentration of mMP (31.8 ng/mL) is higher than those of other metabolites, indicating a high exposure to DMP for Chinese young adults compared with adults in China, Japan, and the US (Gao et al., 2016).

The urinary concentration of mEP was 324 ng/mL for females in Spain and 207.9 ng/mL for the general population in the US. This is one order of magnitude higher than that found in China and Japan (Casas et al., 2011). Earlier studies showed that urinary mEP concentrations were usually higher in females than in males (Silva et al., 2004).

Table 5

Concentrations (ng/mL) of urinary phthalate metabolites in China, Japan, USA and Spain.

Country	Population types	mMP	mEP	mCPP	mBP	miBP	mECP	mCMHP	mEHHP	mEOHP	mCHP	mBzP	mEHP	mOP	mNP	Reference
China	General population (n = 183)	12	21.5	1.3	61.2	56.7	30	18.5	11.3	7	–	0.6	2.1	–	–	(Guo et al., 2011)
China	General population (n = 40)	7.1	19.5	1.2	59.7	50.8	15.7	14.2	7.1	4.9	nd ^a	nd	2.3	nd	nd	(Guo et al., 2011)
China	Young adults (n = 108)	31.8	37.5	3.18	67	57.2	24.3	15.2	14.5	9.73	–	0.14	1.53	–	–	(Gao et al., 2016)
Japan	General population (n = 35)	17.5	14.8	0.8	15.7	8.4	14.7	8.6	6.9	4	nd	nd	2	nd	nd	(Guo et al., 2011)
USA	General population (n = 2350)	1.4	207.9	3.3	25.9	4.9	35.3	–	23.1	15.9	–	17.4	2.1	–	–	(Colacino et al., 2010)
Spain	Female adults (n = 118)	–	324	1.5	27.5	29.9	32.2	–	17.3	15.7	–	10.5	4.4	4	2.8	(Casas et al., 2011)

^a nd = Not detectable.

Many studies suggested that personal care products are a major source of DEP exposure (Wormuth et al., 2006). The high level of DnBP exposure in China is likely to reflect differences in exposure sources compared with other countries. In another piece of research, six PAE metabolites, monomethyl phthalate (mMP), monoethyl phthalate (mEP), monobutyl phthalate (mBP), monobenzyl phthalate (mBzP), mono-2-ethylhexyl phthalate (mEHP), and mono-2-ethyl-5-oxohexyl phthalate (mEOHP) were monitored in spot urines from 150 individuals recruited from Chongqing, China. This study observed a significant positive correlation between mEP and the straight-line velocity of sperm motion (Liu et al., 2012).

Following uptake into the body, phthalates are rapidly absorbed, metabolized by hydrolysis and subsequent oxidation, and excreted in urine and faeces (Hartmann et al., 2015). Although PAEs are rapidly excreted (Bekö et al., 2013; Lee et al., 2014), their endocrine-disrupting effects and toxicity to reproduction will already have been achieved. A recent study investigated the relationship between prenatal phthalate exposure and child obesity by measuring maternal urinary phthalate metabolite concentrations and the weight and height of 4–7-year-old children. It revealed that mono-3-carboxypropyl phthalate (MCP), a non-specific metabolite of several forms of phthalate, was positively associated with the children being overweight or obese (Buckley et al., 2016). For adults, Campbell et al. showed that overweight individuals have higher levels of DEHP metabolites in their urine due to higher energy intake and concomitant higher DEHP exposure (Campbell Jr. et al., 2018).

Previous studies have documented the occurrence of phthalate metabolites in the human body, including maternal blood and cord blood, which implies a possible impact on infants. For a full understanding of the potential risks associated with PAE exposure, a comprehensive investigation is needed into the contribution of different exposure routes.

5. Conclusions

Research conducted over the last decade indicates that PAEs are ubiquitous and abundant in numerous environmental compartments in China and that they may therefore pose a significant threat to environmental safety. The reported levels of PAE contamination are in the µg range per liter in surface water, in the mg range per kilogram in soil, and the ng/m³ to µg/m³ range in the atmosphere. DEHP is reported to be the most abundant phthalate in various environmental media due to its widespread utilization and persistence. However, in terms of human exposure, DnBP has the highest number of exposure pathways in China, and the level of its metabolite in humans has been shown to be high. The reported observations exhibit a location dependence that can be attributed to variations in level of economic and industrial development as well as specific geographical location.

The widespread contamination by PAEs in China today is mainly caused by a huge discharge volume and limited treatment efficiency. However, there is insufficient information about the level of occurrence of PAEs in contamination sources such as sewage and sludge. There is a need for better monitoring of PAE occurrence in STPs and optimization of wastewater treatment to reduce the direct discharge. There is severe lack of information regarding the level of PAE contamination in areas

outside certain hotspots in China. Meanwhile, the transportation and transformation behavior of PAEs in different environmental media and the technologies by which they can be controlled need to be investigated so that their potential risks to human health and the environment can be better-assessed.

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