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Phthalates contamination in China: Status, trends and human exposure-with an emphasis on oral intake☆

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A B S T R A C T

Despite the extensive production and use of phthalates in Asian countries, especially China, limited information is available about the current situation of human exposure in this region, and thus identification of further research needs is warranted. This review summarized the current trends of phthalates related to industrial production and human exposure by conducting a comprehensive assessment of phthalates contaminations in air, indoor dust, personal care products (PCPs), foodstuff and internal exposure in China, with comparisons with other countries. The concentrations of phthalates in indoor dust and PCPs in China were moderate, while concentrations in foods and air were among the highest worldwide. Dietary intake of phthalates varied with location, with hotspots in the southern and eastern coastal regions of China which correlated with the extensive industrial production recorded in these regions. This review firstly revealed the significantly differentiated food-type contribution profiles for phthalates in China and in other countries, which were affected by dietary habits and food contamination. The internal exposure for the Chinese population was found to be moderate, however there is a paucity of data available. Knowledge gaps identified concerning phthalates in China include trends in phthalates exposure, sources (e.g. PCPs, pharmaceuticals and medical treatment), and internal exposure derived from biomonitoring, warranting phthalates a research priority.

1. Introduction

Phthalate esters, also known as phthalates, are the predominant type of plasticizers used around the world, as part of many consumer products (Schecter et al., 2013; U.S. EPA, 2012; Zota et al., 2014) (Table 1). High molecular weight phthalates, such as di (2-ethylhexyl) phthalate (DEHP), are mainly used as plasticizers in the manufacture of polyvinyl chloride (PVC), which is used extensively in consumer products, food contact applications (such as plastic packaging film), vinyl wallpaper, flooring, and medical devices (such as medical tubing and blood storage containers) (ATSDR, 2002). Lower molecular weight phthalates, including di-n-butyl phthalate (DnBP) and diethyl phthalate (DEP), are used as solvents and plasticizers for cellulose acetate, in the manufacturing of lacquers, varnishes, personal care products (e.g. perfumes, cosmetics, and lotions), and the coatings of drugs (ATSDR, 1995, 2001), such as in timed-release pharmaceuticals (Hauser et al., 2004). Due to their widespread use (whereby more than 3 million tonnes of phthalates are consumed per year on a global basis) and their ubiquity in the environment, human exposure to phthalates is virtually unavoidable (Bizzari et al., 2000). Six of the phthalate esters are currently listed as priority pollutants by the United States Environmental Protection Agency (USEPA) and are thus regulated. These are dimethyl phthalate (DMP), diethyl phthalate (DEP), butyl-benzyl phthalate (BBzP), di-n-butyl phthalate (DnBP), di (2-ethylhexyl) phthalate (DEHP) and di-n-octyl phthalate (DOP). DEHP and BBP have been classified as a probable and possible

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Table 1
Phthalates and their major metabolites.

Compounds	Abbr.	Primary metabolites	Abbr.
Low molecular weight phthalates			
Dimethyl phthalate	DMP	Mono-methyl phthalate	MMP
Diethyl phthalate	DEP	Mono-ethyl phthalate	MEP
Di-cyclohexyl phthalate	DCHP	Mono-cyclohexyl phthalate	MCHP
Di-n-pentyl phthalate	DNPP	Mono-n-pentyl phthalate	MnPP
Butyl-benzyl phthalate	BBzP	Mono-benzyl phthalate	MBzP
Di-n-hexyl-phthalate	DnHP	Mono-hexyl phthalate	MHP
Di-iso-butyl phthalate	DiBP	Mono-iso-butyl phthalate	MiBP
Di-n-butyl phthalate	DnBP	Mono-n-butyl phthalate	MnBP
High molecular weight phthalates			
Di (2-ethylhexyl) phthalate	DEHP	Mono-(2-ethylhexyl) phthalate	MEHP
		Mono-(2-ethyl-5-hydroxyhexyl) phthalate	MEHHP
		Mono-(2-ethyl-5-oxohexyl) phthalate	MEOHP
Di-n-octyl phthalate	DnOP	Mono-n-octyl phthalate	MnOP
Di-iso-nonyl phthalate	DiNP	Mono-iso-nonyl phthalate	MiNP
Di-iso-decyl phthalate	DiDP	Mono-iso-decyl phthalate	MiDP

human carcinogen, respectively, by the US EPA (US EPA, 2012). In China, DMP, DEP and DnOP have been listed as priority pollutants by the China National Environmental Monitoring Center (Liu, 2013).

China's high production of phthalates in addition to the increasing consumption of phthalate-containing products, may have resulted in the significant occurrence of phthalates in the environment (Guo et al., 2011a, Guo and Kannan, 2011). This review aims to 1) summarize the phthalate related issues, production and trends in China; 2) evaluate the current status of phthalate contamination in different exposure media and food in China; 3) trace the contributions from different food groups to dietary intake of phthalates; and compare the internal exposure and dietary intakes of phthalates for Chinese populations, with other Asian and western countries. Knowledge gaps were identified and research needs in order to better understand the human exposures, pathways and trends of phthalates in China are also suggested.

2. Worldwide trends of phthalates

Currently, there is limited data available to adequately characterize temporal trends regarding phthalate exposures. DiNP (di-iso-nonyl phthalate) and DiDP (di-iso-decyl phthalate) are replacing DEHP [di (2-ethylhexyl) phthalate], the most common plasticizer, in the global market (European Chemicals Agency, 2012), and these two phthalates combined account for 30–60% of the current plasticizer market in the United States and the European Union (European Chemicals Agency, 2012). DiBP (di-iso-butyl phthalate) is structurally similar to DnBP (di-n-butyl phthalate) and may be a substitute for DnBP (Dodson et al., 2012; Wittassek et al., 2007). As expected, a decline in the production of DnBP and DEHP was observed (e.g. Germany, Wittassek et al., 2011), which have been the focus of legislative activities, that include bans on DnBP, BBzP (butyl-benzyl phthalate), and DEHP in children's products. However, the observed trends cannot be elucidated by the legislative activities (Zota et al., 2014).

Biomonitoring studies in U.S. (U.S. population, 2001–2010; Zota et al., 2014) and Germany (predominately university students aged 20–29 years) (Wittassek et al., 2007) revealed pronounced declines of urinary metabolite concentrations of DEP, DnBP, BBzP, and DEHP, whereas for those of DiBP and DiNP, there was an increase. However, significant data gaps and limitations in the identification of specific sources of exposure make it difficult to determine the underlying reasons for the observed trends in phthalate exposure (Just et al., 2010; Koch et al., 2013; Wormuth et al., 2006). Considering the toxicity, adverse epidemiologic evidence and likely increase in exposure of phthalate alternatives (Bertelsen et al., 2013; National Research Council, 2008), future studies should investigate the temporal changes of concentrations of individual phthalates in common exposure sources (such as cosmetics and food). Nevertheless, since the trends in phthalate exposures are not uniform across populations (female and male; children, adolescent and adults), varied trends are expected due to exposure to different products containing phthalates and related legislations (Dickson-Spillmann et al., 2011; Zota et al., 2014).

3. Phthalate-related issues and trends in China

3.1. Production of plastic products in China

The production of phthalate-related products in China has been rapidly increasing in recent years, and, in fact, China has now become the largest consumer of phthalates in the world. As shown in Fig. 1a, from 2003 to 2011, the production of raw plastic products in China had tripled; in 2011, about 50 million tons of raw plastic products were produced. Phthalates account for 90% of the plasticizer used in PVC production, i.e. over one million tons per year (Guo et al., 2011a). Correspondingly, the number of large-scale plastic manufacturers has dramatically increased between 2003 and 2011, from 600 to 1800 (Fig. 1b). Therefore, a prevalence and rise in phthalates contamination in environmental media and corresponding human exposure may be possible, even though there is a lack of evidence revealing such a trend based on both environmental monitoring and biomonitoring. Fig. 1c shows the wide variation in the amount of plastics produced in different regions of China. The highest production are in Jiangsu, Guangdong and Zhejiang Provinces due to the high industrial demands in these regions. Relatively higher environmental and food contamination are expected in the Pearl River Delta and Yangtze River Delta areas of China and future studies on human exposure risk are necessary. On the other hand, as China is among the top three countries, together with U.S. and Japan, with the largest consumption of personal care products (PCPs), (ChinaIRN, 2012), PCPs could contribute to an important phthalate exposure route. The PCPs market in China was the fastest growing in the world, reaching 8% between 2010 and 2013 (BosiData, 2011; ChinaIRN, 2012). In 2014, the total annual industrial value was over USD25 billion with a growth rate of 12.3% (China Cosmetics Market Report, 2015).

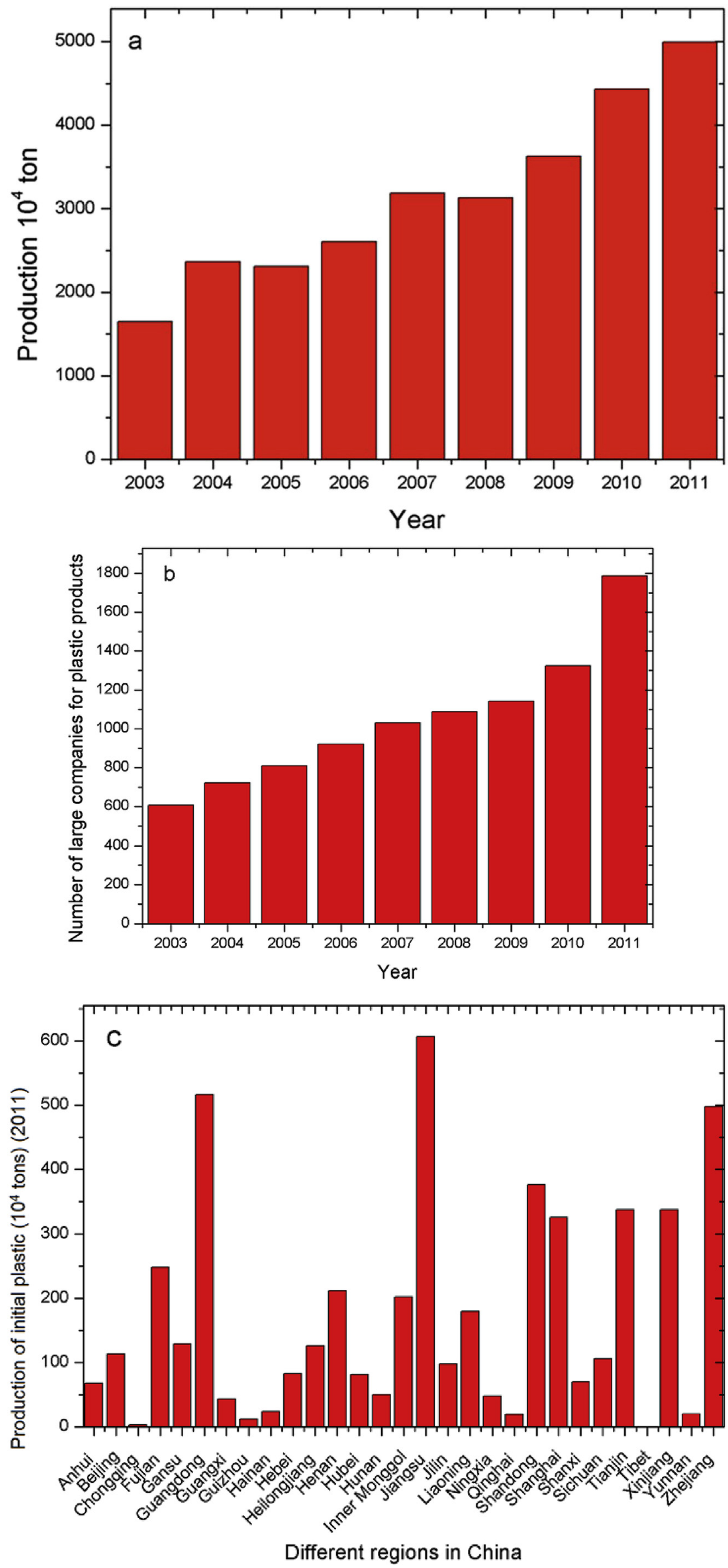

Fig. 1. Variations of plastic production in China from 2003 to 2011 (a. plastic production volume from 2003 to 2011, b. number of large-scale plastic manufacturers from 2003 to 2011, c. plastic production in different regions of China in 2011).

3.2. Trends of contamination and phthalates exposure in China

There is a lack of data derived from both environmental monitoring and biomonitoring, regarding the trend of human exposure data to phthalate in China, Due to limited information on temporal trends, future studies on the internal exposure for China's population determined by biomonitoring studies are required. Environmental monitoring studies on phthalates have increased in China, and phthalates have been found in all environmental matrices and food investigated in China. In light of the wide range of applications and concerns about health effects caused by multiple phthalate exposure sources, Chen et al. (2012) reviewed a time-dependent increase in phthalates and DEHP concentrations in air, based on data on phthalates for the past 10 years in China. However, significant variability was found which might be due to differences in sample sizes, the lack of database and unified sampling or analyses techniques. Nevertheless, no obvious time-dependent trend was found for soil, surface water and sediment.

3.3. Food scandals in China

Following several tainted food scandals in China, namely the melamine-contaminated dairy products scandal in 2008 and the contamination of drug capsules with chromium in 2012, phthalates were detected in alcohol products in China in 2012 which resulted in a major concern regarding contamination by plasticizers. In addition, the illegal use of phthalate plasticizer (DEHP) in clouding agents as additives of foods and beverages was reported in Taiwan in 2011. This incident affected food businesses and aroused a huge concern regarding the use of plasticizers on food safety, especially in China. These serious food safety incidents were widely reported by the Chinese media and thereby drew the public's attention and caused substantial impact on society. Major food safety incidents are handled by governmental agencies, with announcements of decision-making and/or action measures.

4. Exposures and health effects and human exposure

4.1. Human exposures to phthalates

Humans can be exposed to phthalates through multiple pathways, in which the most likely route varies according to individual phthalates. Generally, the major source of human exposure is through ingestion of foods that have been contaminated during production, processing, and packaging. Other significant sources include inhalation of contaminated air, inadvertent ingestion of indoor dust, and via the application of cosmetics and other PCPs (Koo and Lee, 2004; Kavlock et al., 2006). People could also be exposed to high doses of phthalates from medical devices during medical procedures such as blood transfusions and hemodialysis (Calafat et al., 2004). Furthermore, phthalates are able to cross the placenta, and can also be found in breast milk. Hence fetal exposure is closely correlated with maternal exposure (Latini et al., 2003; Lin et al., 2011).

4.2. Health effects

There are many potential public health risks associated with phthalate exposure such as metabolic disorders, carcinogenesis (Lovekamp-Swan and Davis, 2003), reproductive toxicity (Hauser and Calafat, 2005), and endocrine disruption (Latini et al., 2006). Human evidences of adverse health effects associated with plastic additives are limited, suggesting future research trends and needs. Several studies have shown that children's phthalate exposure are closely related to improper sexual development (Colón et al., 2000; Swan et al., 2005), such as premature breast development in girls younger than eight years old (Colón et al., 2000); effects on the immune system (Wang et al., 2014), neuropsychological development (Cho et al., 2010; Téllez-Rojo et al., 2013) and adverse respiratory outcomes including bronchial obstruction and asthma (Hoppin et al., 2004). On the other hand, the effects on adults are considered to be associated with increase in body weight and obesity (Stahlhut et al., 2007; Hatch et al., 2008), diabetes (Svensson et al., 2011), insulin resistance (Trasande et al., 2013), impact on production of reproductive hormones (Duty et al., 2005a; Lottrup et al., 2006; Main et al., 2006), early puberty (Koch et al., 2003), abnormal pregnancy duration (Latini et al., 2003; Whyatt et al., 2009), preterm birth (Meeker et al., 2009), increased risk of breast cancer (due to exposure to DEP) (López-Carrillo et al., 2010), influence on normal pulmonary function for males (Hoppin et al., 2004), and androgen-responsive brain development (Swan et al., 2010).

5. Status of phthalates in different exposure media (air, indoor dust and personal care products) and food

Exposures could be oral (e.g. via food, water and dust), dermal (e.g. via cosmetics) and by means of inhalation (FDA, 2001). Information on phthalates in air, indoor dust PCPs and diet could contribute to a more integrated exposure estimation.

5.1. Occurrence in air

The predominant phthalates in the atmosphere of China are DEHP, DnBP and DiBP (Wang, 2011), suggesting that they are the commonly used plasticizers in China. Based on the studies reported in China during the past 20 years, airborne concentrations of phthalates are normally associated with temperature and spatial variations (Kong et al., 2013; Wang et al., 2011), particle size (Li et al., 2013) and local human activities, including emissions from industrial plants (Zeng et al., 2010). High airborne phthalates concentrations were reported near granulation plants utilized for plastic waste recycling (200–1200 ng/m^3, Huang et al., 2013) and municipal garbage compressing station (415–763 ng/m^3 in particulate phase, Li et al., 2013). Urban air has been investigated frequently for phthalates concentration to trace air pollution in China and the highest fine particulate level (2 μg/m^3) was found in mid-southern China in the city of Chongqing, attributed to its ongoing active industrialization and urbanization (Wang et al., 2006).

The concentration of phthalates in fine particles of 14 Chinese cities (62–2201 ng/m^3) were similar to those in India and Korea (15.8–548 ng/m^3, Park et al., 2006; Choi et al., 2012; Fu et al., 2010), but 1-3 orders of magnitude higher than those reported in other countries e.g. United States (Kansas and Texas: 2.0 ng/m3, Thuren et al., 1990; California: 1.02–9.53 ng/m^3, Rinehart et al., 2006), Belgium (34.5–45.8 ng/m^3, Kubátová et al., 2002) and France (Paris: 5.4 ng/m^3,Teil et al., 2006). Concentrations of airborne phthalates in China were in the high range of worldwide levels, and were in the similar range with other Asian countries (Table S1). However, no maximum acceptable concentration has been set up for airborne concentration of phthalates in China to date. It is necessary to prevent the release of phthalates from human activities, such as industrial emission and municipal waste incineration. Thus, systematic control and management of phthalates release from sources will help to reduce human exposure to phthalates.

5.2. Occurrence in indoor dust

Many studies have reported phthalates contamination in indoor dust in China and other Asian countries (Bamai et al., 2014; Guo and Kannan, 2011; Kang et al., 2012; Kim et al., 2009, 2013; Wang et al., 2013). The sources of phthalates in indoor dust are considered to be related to the presence of various interior materials, such as PVC flooring, vinyl wallpaper, and ceiling materials (Bamai et al., 2014; Bornehag et al., 2005; Xu et al., 2009, 2010). Plastic materials, cosmetics and PCPs were suggested as important sources of phthalates in indoor dusts in China (Zhang et al., 2013). Furthermore, different profiles of phthalates were found in indoor dust in China and U.S. (Guo and Kannan, 2011). Significantly higher median levels of phthalate contamination were found in cities in the Pearl River Delta region, China (mean/median 822–1360 μg/g, Kang et al., 2012; Wang et al., 2013). The highest phthalates concentration was found in houses with toddlers in Nanjing (10900 μg/g) (Zhang et al., 2013). Levels of phthalates in dusts of China were moderate (mean/median: 151–1360 μg/g) compared to other Asian countries (Korea, mean/median: 446–6835 μg/g; Japan, mean/median: 1029–2534 μg/g) (Bamai et al., 2014; Hsu et al., 2012; Kanazawa et al., 2010; Kim et al., 2009, 2013), whereas globally, high levels of phthalates were found in countries including Germany (1273–1320 μg/g), Bulgaria (12240 μg/g) and Denmark (3214 μg/g) (Kolarik et al., 2008a,b; Clausen et al., 2003; BUND, 2011; Fromme et al., 2004) (Table S2).

5.3. Occurrence in personal care products

Phthalate exposures through inhalation and dermal absorption have been reported subsequent to the application of cosmetics and PCPs (Adibi et al., 2003; Duty et al., 2005b; Parlett et al., 2013). To our understanding, information regarding the contribution of phthalate-containing PCPs to total body burden in China's population is limited. However, phthalates have been widely examined (55–72%) in cosmetics and PCPs sold in China. There is a high frequency of occurrence of phthalates in hair gel/hair spray, body lotion, fragrances, nail polish and deodorant, with similar profiles reported in other Asian and Western countries (Koo and Lee, 2004; Houlihan et al., 2002; Hubinger and Harvey, 2006) (Table S3). DEP were detected with the highest frequencies in fragrances and lotions marketed in the United States, Korea and Canada (48–97%), with DBP, BBzP and DEHP lower than 10% (Houlihan et al., 2002; Hubinger and Harvey, 2006; Koniecki et al., 2011; Koo and Lee, 2004), however nail polish was the primary source of DnBP (up to 90% in nail polish) (Koniecki et al., 2011; Koo and Lee, 2004). Similarly, Guo (2013) found DEP to be the most frequently detected phthalate among PCPs from China (detection frequency: 54%, range: ND-937 μg/g), with hand and body lotions being the major contributors to exposures for China's population. The concentrations of the phthalate congeners analyzed in PCPs in China (ND-4348 μg/g in body lotion) were comparable to the range of reported values in Asian (ND-12402 μg/g in fragrance) and western countries (ND-25542 μg/g in fragrance) (Guo et al., 2013; Zheng et al., 2008). However, considering different usage and phthalate profiles of cosmetics and PCPs between Asian region and western countries, the daily exposure dose is expected to be varied between these countries. Despite the significance in human exposure to phthalates, studies that report concentrations of phthalates in PCPs are sparse (Houlihan et al., 2002; Koniecki et al., 2011), especially for China (Liang et al., 2012).

5.4. Occurrence in food

Human exposure to phthalates is mainly via foods, due to both the bioaccumulation of phthalates in the food chain and the use of PVC in food packaging (Chai et al., 2008; Li et al., 2012; Spillmann et al., 2009; Wormuth et al., 2006). In China, DEHP was the most abundant phthalate in all food categories, except for wine, beer, and condiments, in which DBP and DiBP were the predominant compounds (Guo et al., 2012). Chen (2012) found that phthalate concentrations varied with different types of food and locations. In general, higher contamination levels of phthalates in food were found in the Pearl River Delta and Yangtze River Delta areas which corresponded to the high production volume of phthalates in Guangdong, Zhejiang and Jiangsu Provinces (Table S4). Since phthalate concentrations in food in the Pearl River Delta area were much higher than those in the Yangtze River Delta area, people living in the Pearl River Delta area therefore had a higher food phthalate intake (Chen et al., 2012). Furthermore, the highest total concentrations of phthalates in food items in the Pearl River Delta area ranged from 0.43 to 52.4 μg/g ww. Compared with phthalates concentrations in different types of foods from U.S., Norway, and Belgium, the existing studies show significantly higher phthalates contamination in foodstuff such as grains, vegetables and fruits collected in China. The highest contamination of grains (5.79–39.9 μg/g) and meat (2.06–13.6 μg/g) were reported in Guangdong Province (Liu and Huang, 2009 μg/g), while the highest concentrations were found in vegetables in Shanghai (ND-36.4 μg/g) (Cai and Shi, 2005).

The contamination of agricultural lands by phthalates in vicinity of e-waste recycling sites have become human health threats. The concentrations of the six USEfPA priority pollutant phthalates ranged from 0.31 to 2.39 mg/kg in soil to 1.81–5.77 mg/kg in various plants (dry weight) collected 250 m from an e-waste dismantling site at Luqiao township, Taizhou city, Zhejiang Province. The major contaminants found in soil were DEHP and DnBP. Health risk assessment concluded that DEHP may present a high exposure risk to the population (of all ages) in the area due to soil ingestion and the consumption of contaminated vegetables (Ma et al., 2013). Another study showed that the total phthalate concentration of the six priority pollutants in e-waste soil from Taizhou ranged from 12.6 to 46.7 mg/kg (Liu et al., 2009). Plants such as alfalfa and maize have been demonstrated to phytoremediate phthalate compounds from contaminated soil (Li et al., 2014; Ma et al., 2013). On 4 June 2015, the phthalates DEHP, DBP, BBzP and DiBP were added to the list of restricted substances in Annex II of the EU RoHS 2 Directive 2011/65/EU and applies to cables or spare parts for the repair, the reuse or upgrading of electrical and electronic equipment placed on the market on and after 22 July 2019 (EUR-Lex, 2015). Plastic film used for polytunnel greenhouses has also recently become another source of phthalates in food whereby the total phthalates concentration (six priority pollutants) of vegetables at an agricultural area in Nanjing, China ranged from 790 ± 630 to 3010 ± 2130 μg/kg (Ma et al., 2015). In addition, other food categories with high phthalate contamination are listed below:

5.4.1. Aquatic food

In China, high phthalates concentrations were reported in aquatic food in the eastern and southern regions, including Jiangsu Province (0.19–5.0 μg/g), Shanghai Municipality (6.43–7.52 μg/g), Zhejiang Province (3.19–3.20 μg/g), Guangdong Province (10.6–17.3 μg/g), Pearl River Delta area (1.57–7.1 μg/g) and Hong Kong (25.4–52.4 μg/g), with DEHP as the highest contributor. Li reported the phthalate levels in freshwater fish found in Pearl River Delta area, with concentration of DEHP in the range of 11.0–16.3 μg/g in Guangdong while in Hong Kong, the range was 26.1–38.0 μg/g (Huang et al., 2008). In a recent study, He (2015) reported the highest concentration of phthalates in seafood compared with other food groups (cereals, beverages, condiments, snacks and meat products) sold in Yanji, northeast China, with mean and median of 1380 and 1210 ng/g ww, respectively. Based on the Food

Guide Pagoda for Chinese Residents from the Chinese Nutrition Society (2016), seafood was listed as the item with the highest daily consumption rate for China's population, suggesting the significant amount of dietary intake of phthalates.

5.4.2. Bottled and packed food

Phthalate esters applied in the containers of products such as plastic bottles and PVC gloves used for preparation of foods could leach and migrate into the cooking oils or into packed food (Gartner et al., 2009; Tsumura et al., 2001), therefore, it is necessary to conduct a safety evaluation and establish the maximum allowable concentration of phthalates in food. The concentrations of plasticizers analyzed in edible oil in China (Guangdong Province, 8.88 μg/g; Guizhou Province, 52.9–28.9 μg/g; Jiangsu Province, 1.42 μg/g) were significantly higher than those reported in oils marketed in the U.S. (210–7558 ng/g) (Bi et al., 2013). Liu (2009) reported the elevated phthalates concentrations for vegetables and fruits packed with plastic packages in China, which ranged from 0.88 to 28.7 and 1.56–103.7 μg/g, for DBP and DEHP, respectively. Wang (2007) analyzed the concentrations of phthalates in food including steamed breads, seedcakes and some raw vegetables that were packed in plastic bags for 30 min, and recorded level of DEHP, DMP and DBP at 1.920, 0.988 and 0.702 ng, respectively. Thus, the rising trend for consumption of food with disposable packaging and packaged fast food might increase the risk of oral intake of phthalates for Chinese consumers.

6. Human dietary exposure

6.1. Status of dietary exposure of phthalates in China

The European Food Safety Authority (EFSA) established a tolerable daily intake (TDI) of 50 μg/kg body weight/d (bw) for DEHP and 10 μg/kg bw/d for DnBP (EFSA, 2005). The reference doses suggested by the US EPA are 20, 100, 200, and 800 μg/kg bw/d for DEHP, DnBP, BBzP, and DEP, respectively. In comparison to these values, the reported daily dietary exposures to DnBP (0.24–4.76 μg/kg bw/d) and DiBP (0.25–2.51 μg/kg bw/d) of adults in Shanghai and Harbin, China were low (Guo et al., 2012). However, the daily dietary intake estimated for DEHP in Yangtze River Delta and Pearl River Delta areas were slightly higher and three times higher, respectively, than the US EPA's acceptable daily intake dose of 20 μg/kg bw/d) (Chen et al., 2012) (Fig. 2). Furthermore, the estimated intake doses in Shanghai, Harbin (Guo et al., 2012) and Yanjin (He et al., 2015) were in the safe range. When compared with other countries, there are studies in China that have detected relatively high levels of dietary phthalate intake, especially in coastal cities of the Pearl River Delta and Yangtze River Delta regions. Such significant variability of dietary intake doses for populations in different regions in China might be attributed to different levels of industrial development.

6.2. Contribution of food categories to dietary intake of phthalates

To compare the contribution profiles of different foodstuff to dietary intake of phthalates for populations among Asian and western countries, China was taken as an example (Yangtze River Delta and Pearl River Delta, Chen et al., 2012; Shanghai and Harbin, Guo et al., 2012; Yanji, He et al., 2015). Table 2 shows a comparison with western countries (the U.S., Schecter et al., 2013; Norway, Sakhi et al., 2014; Belgian, Sioen et al., 2012) and suggests the top three food categories that have contributed the most to the estimated oral intake for adults for different phthalates. The contributions shown were derived from median and mean results bound probabilistic scenario to represent the most realistic exposure scenario. The food group with the highest contribution to total phthalates from dietary intake source was determined to be cereal (grain) for China's population (37–71%), while for the U.S., they were “other dairy products” excluding milk (30%). Cereals, seafood and beverages were listed as the top three contributors for total phthalates in Yanji, China, and cereals, beverages and meat products for Shanghai and Harbin. The top three contributors were vegetables, meat/egg and aquatic products for populations in Yangtze River Delta, while grain, fruit and vegetables for the Pearl River Delta areas. For Norway and Belgium, the biggest contributor of DEHP was bread, while dairy product was the biggest contributor to the dietary intake of DEHP in the U.S. A review of food monitoring data in the U.S conducted by Serrano et al. (2014) showed high concentrations of DEHP (≥300 μg/kg) were detected in poultry and cream-based dairy products. Thus, dairy products and bread were considered to be the main contributors of DEHP for U.S. and EU countries. Such varied contribution profiles might be explained by the different lifestyle and dietary habits in Asian and western countries. The top contributors and corresponding ratios for different phthalate compounds varied significantly due to different contamination sources and extent of accumulation for these food groups and intake frequencies.

7. Exposure of Chinese population to phthalates

Despite the extensive production and high consumption of phthalates in Asian countries, limited information is available about the exposure of the general population to phthalates in these countries. In recent years, there has been an increase in studies investigating the internal phthalate exposure in Asian populations (general and specific) (China, Guo et al., 2011a,b; Japan Itoh et al., 2007, 2009, and Korea, Ji et al., 2010) by measuring urinary concentrations of phthalate metabolites, which revealed widespread exposure (Table S5). Metabolites of DMP, DEP, DiBP, DnBP, BBzP, DEHP, DiNP and DiDP have been identified as biomarkers of recent human exposure to phthalates (Latini et al., 2005). For DEHP, its metabolite MEHP has been widely used as a biomarker of DEHP exposure assessment, however, more recently, the oxidative secondary metabolites of DEHP, namely mono-(2-ethyl-5-hydroxyhexyl) phthalate (MEHHP) and mono-(2-ethyl-5-oxohexyl) phthalate (MEOHP) have been used to assess exposure to DEHP due to their longer half-lives when compared to MEHP (MEHP, 5 h; MEHHP and MEOHP, 10 h) (Koch et al., 2005). Thus urinary concentrations of these oxidative metabolites are higher than MEHP and are considered better biomarkers (Heudorf et al., 2007; Silva et al., 2006). As shown in Table S5, the concentrations of MEHHP and MEOHP are higher than MEHP. The urinary excretion half-lives of these phthalate biomarkers are short and can only be used to evaluate short-time exposure to DEHP. Their limited stability may also impact repeatability of the results. The metabolites, mono (5-carboxy-2-ethylpentyl) phthalate (5cx-MEPP) and mono [2-(carboxymethyl)hexyl]phthalate (2cx-MMHP) (2cx-MMHP) have been suggested to serve as better parameters to reflect long-term exposure to DEHP due to their longer half-lives (5cx-MEPP, 12–15 h; 2cx-MMHP, 24 h) (Koch et al., 2005).

Variations in profiles of urinary phthalate metabolites have been found among different countries such as the U.S., Germany and in Asia (Silva et al., 2004; Calafat et al., 2006) and were possibly due to different patterns of use of consumer products (Koch et al., 2003; Wittassek et al., 2007). Studies suggest that levels of exposure to phthalates are generally higher in children than in adults due to pica behavior. A study by Wang et al. (2015) (Table S5) showed that children (8–11 years old) living in Shanghai, Jiangsu Province, and Zhejiang Province were widely exposed to phthalates and that children living in manufacturing-intensive regions had a higher risk of cumulative phthalate exposure. A study by Pan et al. (2006) showed exceedingly elevated concentrations of MEHP (562.3 μg/L) and MnBP (548.4 μg/L) in urine of male workers at a factory producing unfoamed PVC flooring in Liaoning Province. The workers were exposed to DEHP via dermal contact and dust inhalation. Occupational exposure to phthalates is a major contributor to elevated body loadings.

Urinary concentrations of phthalate metabolites could be determined using the creatinine correction approach as well as the urine volume approach (Wang et al., 2015). Saravanabhavan et al. (2014) reported that the DEP daily intake calculated using these two approaches were similar and that caution should be exercised when using urinary concentrations to compare trends in intake in sub-populations (such as children vs adults) because additional physiological factors may play a role.

Estimated exposure levels of different phthalates vary among different population groups in different countries (Fig. 3). The estimated daily exposure dose of the four phthalates (DEHP, DnBP, DEP, and DiBP) in the studies in China fell in the ranges reported in Asia, while highest exposures were reported in India and Kuwait. In particular, although higher dietary exposure was expected, phthalates exposure for China's population based on biomonitoring were in the lower range compared with other countries. This difference could be explained by the various pathways contributing to phthalate exposure and the investigated foodstuffs that focus on potentially contaminated areas characterized by heavy industrial input, such as the Pearl River Delta region. Specific population under study will affect the overall exposure dose for the general population and a lack of information to distinguish the various exposure routes would also introduce a certain degree of estimation errors.

The Estimated Daily Intake (EDIs) of populations in China have been calculated via two approaches, namely the biomonitoring estimation approach and the exposure scenario approach for indoor dust ingestion (sampled in Shanghai; Guo and Kannan, 2011), dietary intake (sampled in Shanghai and Harbin; Guo et al., 2012), and PCPs exposure (sampled in Tianjin; Guo et al., 2013) (Fig. 4). Differences in results of estimated exposure of phthalates determined from the two approaches may be due to the omission of additional exposure pathways in the exposure scenario approach whereas the biomonitoring estimation approach inherently takes into account all possible sources and routes of exposure. Consumption of contaminated food is the most important exposure pathway for exposure to phthalates (especially the long-chain phthalates, e.g. DEHP) for the general population, while PCPs is the main contributor to DEP exposure. However, other sources such as contaminated air are also important exposure sources to short chain phthalates, such as DBP, DEP and DMP. Both the biomonitoring estimation approach and the exposure scenario approach have been applied to estimate the exposure levels to DEHP, DnBP and DiBP based on the available monitoring data in China which resulted in both approaches being comparable, but the exposure assessment approach was more comprehensive (Cao et al., 2016). The significant sources of DnBP call for further studies regarding their exposure scenarios. However, exposure studies conducted in different regions could result in different contribution profiles for each exposure route, due to the variation of phthalate distribution, dietary habits, human activities, consumer products and regulatory standards in these different regions.

Over-the-counter medicines made in China is another exposure pathway to phthalate esters. Individual phthalate concentrations ranged from 0.001 μg/g dicyclohexyl phthalate (DCHP) to 5.85 μg/g diethyl phthalate (DEP) and the predominant phthalates in these medicines were DnBP which accounted for more than 65% of the total phthalates in all medicine samples, followed by DEHP and DEP. Phthalates are found in the gastroresistant film coatings of medicines, plastic packing materials and also in Chinese herbal plants used to make medicines (Jia et al., 2017).

8. Knowledge gaps and research needs

Considering a large scale and potentially elevating contamination trend, future research is needed in a variety of fields, including human exposure assessment, exposure trend, source elucidation and health impact assessment. This section presents knowledge gaps, that were identified to the best of our knowledge, for future

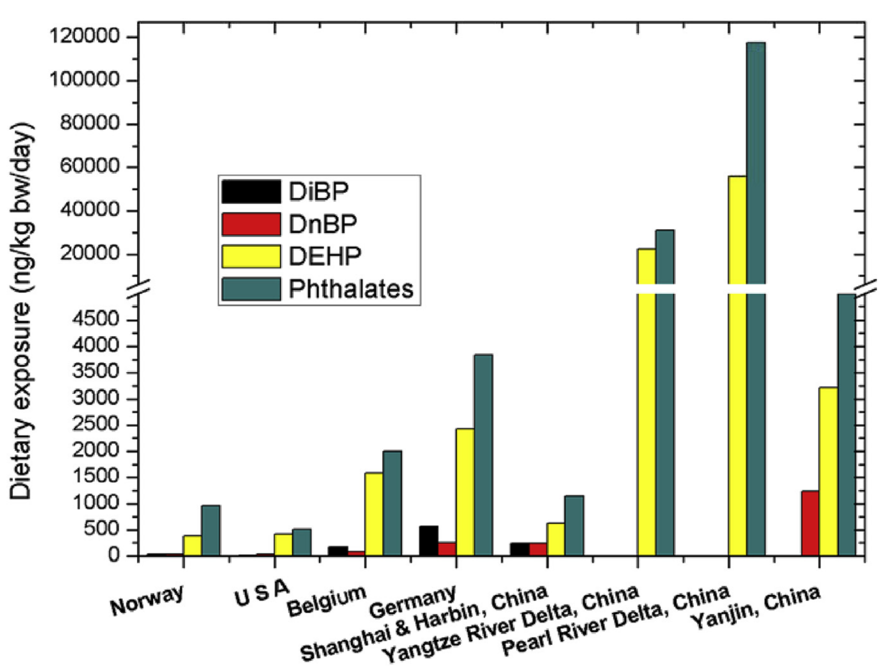

Fig. 2. Daily dietary intake of phthalates for China and other countries (ng/kg bw/day).

research needs. It is noted that additional aspects not included herein may exist, and therefore also require attention and research efforts.

Gap 1. The production and use of phthalates in consumer products and PCPs in developing countries, such as China, could bring parallel widespread exposure to these chemicals among all segments of the population. To understand the extent of exposure to phthalates and its alternatives are of public relevance. Biomonitoring or measuring the concentrations of chemicals or their breakdown by-products in humans are a useful tool. Nevertheless, there is a lack of data concerning nationwide biomonitoring programs to track exposure to phthalates and their possible replacements in China and other developing countries. In addition, limited data is available for phthalate exposure dose for different populations in China, such as different age groups, living area and income-depended lifestyle. For example, there is a need to conduct further research at e-waste recycling areas to elucidate the exposure of workers and the community to phthalates via e-waste processing and disposal and the compounded effects of phthalates with other pollutants such as persistent organic pollutants. Furthermore, case-control studies based on biomonitoring are also encouraged to study the health effects on specific populations with phthalate exposure as a potential health risk factor.

Gap 2. The trends of human exposure to phthalates and their replacements may be affected by changes in production and use of these compounds in the consumer market, prompted, at least partially, by the combined efforts of legislators, industry, and the general public. Biomonitoring data may provide useful insights into these ongoing market changes. However, limited information exists on consistent exposure trends for populations in developing

Table 2
Top three food groups contributing to more than 50% of the dietary exposure to phthalates in adult population in different areas of China and other countries.

	First	Second	Third	% of top 3
Yanji, **China**				
DMP	Cereal (49.1%)	Beverage (39.9%)	Seafood (4.91%)	94
DEP	Beverages (80.1%)	Cereal (11.4%)	Meat Product (5.81%)	97
DnBP	Beverages (54.8%)	Cereal (34.1%)	Seafood (5.51%)	94
BBzP	Cereal (74.4%)	Seafood (5.20%)	Beverage (3.39%)	83
DEHP	Seafood (43.2%)	Cereal (37.0%)	Beverage (11.0%)	91.2
DnOP	Seafood (69.8%)	Cereal (11.6%)	Beverage (11.6%)	93
Total	Cereal (36.5%)	Seafood (30.1%)	Beverage (25.5%)	92
Shanghai & Harbin, **China**				
DMP	Cereal (71.3%)	Beverage (14.4%)	Milk (7.45%)	92
DEP	Beverage (70.3%)	Cereal (16.2%)	Meat Product (6.16%)	93
DiBP	Cereal (80.8%)	Beverage (5.59%)	Meat Product (4.60%)	91
DnBP	Cereal (80.1%)	Beverage (9.07%)	Milk (6.44%)	96
BBzP	Cereal (50.8%)	Cooking Oil (33.7%)	Beverage (10.3%)	95
DEHP	Cereal (65.7%)	Beverage (15.4%)	Meat Product (10.0%)	91
Total	Cereal (70.9%)	Beverage (13.1%)	Meat Product (6.82%)	91
Yangtze River Delta, **China**				
DEHP	Vegetable (53.9%)	Fruit (16.2%)	Aquatic (15.0%)	85
Total	Vegetable (45.5%)	Meat/Egg (16.1%)	Aquatic (13.0%)	75
Pearl River Delta, **China**				
DEHP	Grain (32.3%)	Fruit (25.5%)	Vegetables (18.2%)	76
Total	Grain (44.2%)	Fruit (14.6%)	Vegetables (13.3%)	72
New York State, **USA**				
DMP	Beverage (50%)	Other dairy product (25%)	Grain (25%)	100
DEP	Grain (90.3%)	Other dairy product (6.45%)	Beverage (3.23%)	100
DiBP	Grain (42.1%)	Beverage (21.1%)	Other dairy product (10.5%)	74
DnBP	Other Diary Product (68.9%)	Grain (19.1%)	Beverage (4.92%)	93
DnHP	Meat (40%)	Other dairy product (20%)	Grain (20%)	80
BBzP	Vegetable (73.1%)	Grain (15.9%)	Other dairy product (6.10%)	95
DCHP	Vegetable (85.7%)	Beverage (14.3%)		100
DEHP	Other Diary Product (25.7%)	Grain (20.2%)	Meat (18.1%)	64
DnOP	Beverage (35%)	Meat (20%)	Milk (15%)	70
Total	Other dairy product (30.3%)	Grain (21.6%)	Meat (13.5%)	65
Flanders, **Belgium**				
BBzP	Fruits (28.3%)	Olive oil (14.9%)	Alcoholic drinks (14.0%)	57.2
DnBP	Bread (19.7%)	Processed meat (14.0%)	Biscuits and cakes (10.7%)	44.4
DEHP	Bread (31.4%)	Fruits (8.6%)	Fresh meat (8.1%)	48.1
DCHP	Cheese (21.5%)	Milk (beverages) (11.3%)	Biscuits and cakes (9.5%)	42.3
DEP	Fruits (20.9%)	Bread (11.8%)	Pasta and other grains (10.0%)	42.7
DiBP	Bread (33.5%)	Fruits (17.3%)	Biscuits and cakes (14.9%)	65.7
DMP	Fresh meat (27.2%)	Processed meat (14.6%)	Vegetables (7.2%)	49
DnOP	Margarine (16.6%)	Processed meat (15.2%)	Fresh meat (11.7%)	43.5
Norway				
DMP	Meat and meat products (37%)	Bread (21%)	Fats and oils (10%)	75
DEP	Milk and milk products (25%)	Bread (14%)	Meat and meat products (11%)	61
DiBP	Bread (42%)	Grain and grain products other than bread (20%)	Meat and meat products (14%)	81
DnBP	Beverages (21%)	Bread (19%)	Grain and grain products other than bread (17%)	68
BBzP	Meat and meat products (68%)	Fats and oils (8%)	Bread (7%)	87
DEHP	Bread (24%)	Milk and milk products (19%)	Cheese (17%)	76
DCHP	Meat and meat products (23%)	Fats and oils (23%)	Cheese (12%)	67
DnOP	Meat and meat products (36%)	Fats and oils (21%)	Bread (11%)	76
DiNP	Bread (34%)	Cakes and biscuits (20%)	Meat and meat products (16%)	84
DiDP	Bread (40%)	Beverages (15%)	Meat and meat products (12%)	77

countries, such as China.

Gap 3. Monitoring the sources and pathways of human exposure are needed for different populations in China. Efforts are needed to elucidate compound-dependent dietary and non-dietary exposure pathways and associated daily intakes, particularly for sensitive subpopulations such as children and pregnant women. However, few data is available for non-dietary exposure, including PCPs, cosmetics, medical equipment and pharmaceuticals in China, calling for future studies. With regard to medical exposure, it could result in significant rise in internal levels of phthalates than average population. Thus, there is a pressing need for assessing exposure during medical treatment, especially for people who are undergoing surgery and intensive medical care.

Gap 4. Research is needed to better elucidate the environmental occurrence of phthalates and their replacements regionally or globally. There is still a dearth of information on production and usage as well as distributions of these chemicals and their replacements and fate in the environment. Few investigations have been recorded on some environmental matrices, such as wildlife. The lack of environmental data impedes the evaluation of sources, environmental distribution, and fate of phthalates. Regional or global monitoring studies are critically needed to establish baseline levels and track the spatial and temporal distributions of these emerging chemicals in the environment.

Gap 5. Knowledge gaps should be prioritized in research for better supporting appropriate policy decisions. The three priority topics for phthalates exposure are internal exposure (biomonitoring studies), health effects and exposure sources. Future research are suggested with regard to well-designed, follow-up exposure studies that are relevant to the development of

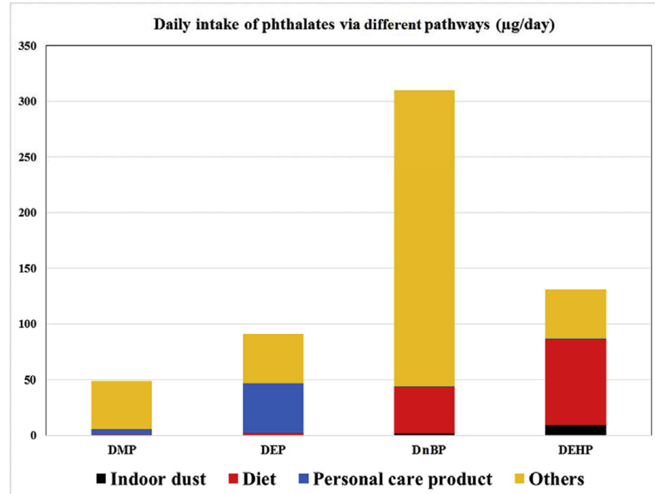

Fig. 4. Comparison of phthalate exposure from different pathways (indoor dust, Shanghai, China; Diet, Shanghai and Harbin, China; PCPs, Tianjin China;) (Guo et al., 2011a,b; Guo et al., 2012, 2013) and the estimated internal exposure based on biomonitoring data (reported in Shanghai, China) (Guo et al., 2012).

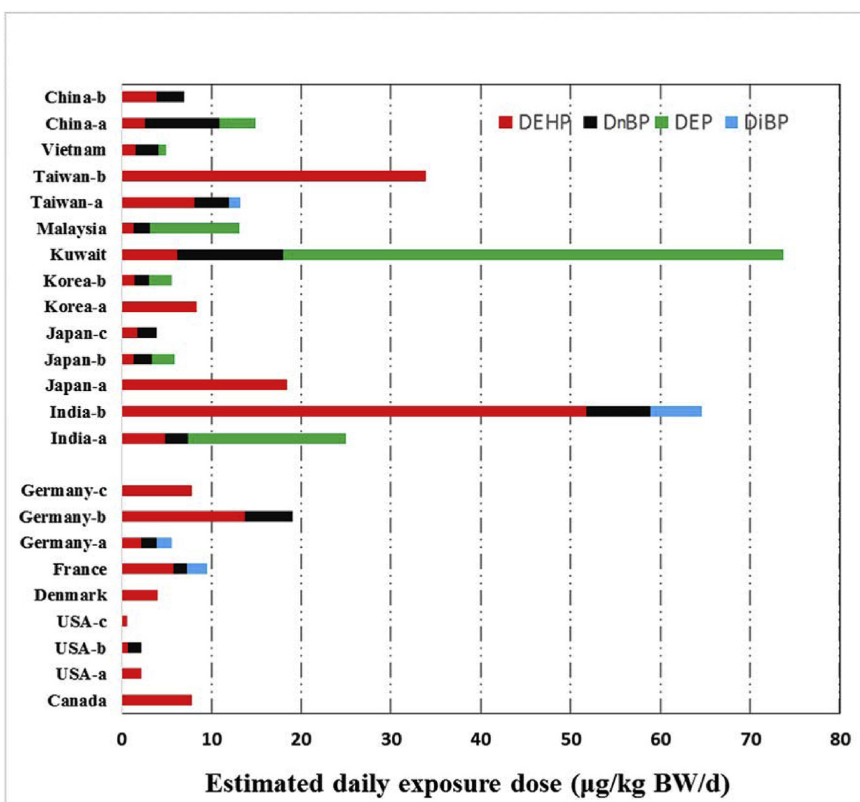

Fig. 3. Comparison of estimated daily exposure dose based on biomonitoring studies in Asian region and other countries. (Germany-a: Fromme et al., 2007; Germany-b: Koch et al., 2003; Germany-c: Wittassek et al., 2007. France: Zeman et al., 2013; Denmark: Frederiksen et al., 2011; USA-a: Calafat and McKee, 2006; USA-b: Kohn et al., 2000; USA-c: David, 2000. Canada: Meek and Chan, 1994; India-a: Guo et al., 2012; India-b: Das et al., 2014; Japan-a: Fujimaki et al., 2006; Japan-b: Guo et al., 2012; Japan-c: Suzuki et al., 2009; Korea-a: Kim et al., 2015; Korea-b: Guo et al., 2012; Kuwait: Guo et al., 2012; Malaysia: Guo et al., 2012; Taiwan-a: Lin et al., 2011; Taiwan-b: Chen et al., 2009; Vietnam: Guo et al., 2012; China-a: Guo et al., 2012; China-b: Cao et al., 2016.).

production systems in highly exposed and vulnerable populations, especially during critical periods of development.

9. Conclusions

Phthalates are ubiquitous in different environmental exposure media and food in China. In this paper, the current state of human evidence, as well as future research trends and needs were discussed. With the consistent rising production of phthalate-related products in China, the human exposure trend for the Chinese population is yet unknown. Indoor levels of phthalates (indicated by dust and PCPs) in China and other Asian countries were moderate, while in air and foodstuffs of Chinese cities, they were lower compared to other parts of the world.

The relatively higher dietary exposure dose for Chinese population were analyzed in this review, however, significant variability was also found based on different areas, with the highest exposure populations in the Pearl River Delta region. In addition, this study is the first to review the contribution of different food categories to the total dietary intake for the populations in China, and different contribution profiles were observed among international nations (China, U.S. and European countries). However, the body loadings reported for Chinese populations were moderate, whereas higher exposure doses were reported in other Asian areas. Nevertheless, since the data of food contamination are mostly derived from areas with high possibility of contamination and a lack of body loadings data in populations in China, discrepancy were reviewed between dietary exposure and biomonitoring estimation.

Human biomonitoring studies for Chinese populations are useful for elucidating potential exposure pathways and exposure trends. Recent advances in the measurement of exposure biomarkers for toxicants such as phthalates, hold much promise in improving the epidemiological database and satisfying a great need for more human studies of adverse health effects. Monitoring the sources of human exposure (PCPs, cosmetics, pharmaceuticals and medical care) and environmental distribution and fate is needed in China.

Compliance with ethical standards

The authors would like to declare that they have no conflict of interest, and there were no human nor animal participants in this research.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.02.088.

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