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

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ABSTRACT

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).