Environment International 101 (2017) 80–88

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Food consumption survey of Shanghai adults in 2012 and its associations with phthalate metabolites in urine

Ruihua Dong [a], Tong Zhou [b], Shanzhen Zhao [c], Han Zhang [a], Meiru Zhang [a], Jingsi Chen [a], Min Wang [c], Min Wu [a], Shuguang Li [a], Bo Chen [a,]*

[a] Key Laboratory of Public Health Safety of the Ministry of Education, School of Public Health, Fudan University, Shanghai, China
[b] Shanghai Institutes of Preventive Medicine, China
[c] Shanghai Entry–Exit Inspection and Quarantine Bureau, Shanghai, China

ARTICLE INFO

Article history:
Received 4 August 2016
Received in revised form 10 January 2017
Accepted 10 January 2017
Available online 20 January 2017

Keywords:
Phthalate metabolites in urine
24-hour recall survey
Food Frequency Questionnaire
Shanghai adults

ABSTRACT

Background: Diet is considered to be a significant exposure pathway for phthalates. In this study, we assessed the associations between food consumption and urinary concentrations of phthalate metabolites among Shanghai adults.
Methods: A cross-sectional study involving 2418 participants was conducted in the fall of 2012. Recent food consumption was assessed by a 24-h dietary recall survey, and a Food Frequency Questionnaire (FFQ) characterized long-term dietary patterns. Urinary metabolites of six phthalates were measured.
Results: Both the 24-h recall survey and FFQ identified wheat, dairy, and fruits as being positively associated with the excretion of phthalate metabolites. The 24-h recall data also showed positive associations with processed meats and alcohol. We evaluated the impact of reported consumption of multiple food categories simultaneously (wheat, fruits, meats, etc.) on metabolite excretion and found that, as more food types were consumed, the number of metabolites excreted, as well as their concentrations, increased with high significance (p values < 0.0001). We also evaluated the two survey instruments together. When both surveys reported consumption of fruits and dairy, the numbers of metabolites and their concentrations were significantly higher compared to when both surveys reported non-consumption, (p values < 0.000001). Rice consumption was found to be negatively associated with phthalate excretion; frequent and high levels of rice consumption were found to be associated with lower excretion of metabolites.
Conclusion: Food consumption was associated with phthalate exposure in Shanghai adults. Both 24-h recall and FFQ identified significant associations between consumption of food types and phthalate exposure.

1. Introduction

Public concern has been raised about phthalates due to associations with detrimental health effects, such as allergies, obesity, atherosclerosis, and asthma (Lind and Lind, 2011; Martino-Andrade and Chahoud, 2010; Hatch et al., 2008; Bornehag et al., 2004; Singh and Li, 2012; Caldwell, 2012). Phthalates, which are the diesters of 1,2-benzenedicarboxylic acid, are chemicals that are present in several commercial products. High-molecular-weight (HMW) phthalates (≥250 Da), such as butyl benzyl phthalate (BBzP), bis(2-ethylhexyl) phthalate (DEHP), di-iso-nonyl phthalate (DiNP), and di-iso-decyl phthalate (DiDP), are mostly used in the production of flexible vinyl plastics, flooring, and medical devices (Schettler, 2006; Cao, 2010; Wormuth et al., 2006). Low-molecular-weight (LMW) phthalates (<250 Da), such as dimethyl phthalate (DMP), diethyl phthalate (DEP), di-iso-butyl phthalate (DiBP), and din-butyl phthalate (DnBP), are commonly used in the production of varnishes, paints, lacquers, and personal care products (Cao, 2010; Wormuth et al., 2006; Sathyanarayana, 2008). The major use of phthalates is to impart durability and flexibility to plastics, such as polyvinyl chloride (PVC) (Wormuth et al., 2006). Phthalates are not chemically bound to PVC; therefore, they can easily leach from commercial or industrial products

Abbreviations: BMI, body mass index; BBzP, butyl benzyl phthalate; CIs, confidence intervals; DBP, dibutyl phthalate; DEHP, bis(2-ethylhexyl) phthalate; DEP, diethyl phthalate; DiBP, di-iso-butyl phthalate; DiDP, di-iso-decyl phthalate; DiNP, di-iso-nonyl phthalate; DMP, dimethyl phthalate; DnBP, din-butyl phthalate; FCMs, food contact materials; FFQ, Food Frequency Questionnaires; HMW, high molecular weight; IQR, interquartile range; LC-MS/MS, liquid chromatography tandem mass spectrometry; LMW, low molecular weight; LOD, limit of detection; MBzP, mono-benzylphthalate; MCMHP, mono-2-carboxymethyl-hexyl phthalate; MECPP, mono-2-ethyl-5-carboxypentylphthalate; MEP, monoethyl phthalate; MEHP, mono-2-ethylhexylphthalate; MEHHP, mono-2-ethyl-5-hydroxyhexylphthalate; MEOHP, mono-2-ethyl-5-oxohexyphthalate; MiBP, monoisobutylphthalate; MMP, monomethyl phthalate; MnBP, mono-n-butylphthalate; PVC, polyvinyl chloride; SHFCS, Shanghai Food Consumption Survey.

* Corresponding author at: School of Public Health, Fudan University, 138 Yixueyuan Road, Shanghai 200032, China.
E-mail address: chenb@fudan.edu.cn (B. Chen).

http://dx.doi.org/10.1016/j.envint.2017.01.008
0160-4120/

into food or air, leading to the contamination of foods and the environment (Wormuth et al., 2006).

Diet may be a significant pathway for exposure to phthalates. Certain foods have been associated with phthalate exposure (Schettler, 2006; Colacino et al., 2010; Fromme et al., 2007; Schecter et al., 2013; Martine et al., 2012; Bradley et al., 2013; Sioen et al., 2012; Gonzalez-Castro et al., 2011). For example, the presence of DEHP has been reported in poultry, meat carcasses, cream, eggs, and fish (Serrano et al., 2014). Cross-sectional studies have reported that DEP metabolites in urine are associated with vegetable consumption, and that the metabolites of HMW phthalates and DEHP were positively associated with meat and poultry consumption (Colacino et al., 2010; Trasande et al., 2013). Population-based dietary assessments are of considerable interest due to the presence of phthalates in different types of foods. However, results obtained from epidemiologic and food-monitoring studies have been inconsistent. Phthalates can migrate into food from plasticized PVC food contact materials (FCMs), such as plastic containers, lid gaskets, can linings, tubing, dishware and utensils, gloves, and conveyor belts used in the manufacturing, processing, storage, and transportation of foods (Cao, 2010; National Toxicology P, 2003). Phthalates are also found in the adhesives on food wrappers and printing inks as well as coatings on cookware (Serrano et al., 2014). Foods with high fat have been reported to be contaminated by HMW phthalates that are more lipophilic, such as DEHP (Cao, 2010). Phthalate concentrations may vary based on food production origin, processing, packaging, and lipid content (Schecter et al., 2013; Wittassek et al., 2011).

Most phthalates in human toxicokinetic studies have been reported to have half-lives of <24 h (Koch et al., 2005; Hoppin et al., 2002). It seems that phthalates do not build up in human bodies over days. Based on this, previous studies have primarily used a 24-h recall survey or dietary record method to identify associations between food consumption and phthalate exposure (Zota et al., 2016; Jo et al., 2016; Hartle et al., 2016). In contrast, a Food Frequency Questionnaire (FFQ) is usually used to collect information on long-term dietary behavior and is generally thought not to be well correlated with phthalate excretions in urine. However, some types of food may be consumed at least once in a day, especially main staple foods. We hypothesized that contamination of these types of food with phthalates would result in phthalate excretions in urine would be associated with the long-term dietary behavior. However, such studies linking phthalate exposure to FFQ data are scarce.

Phthalate production and importation in China has been reported to be dominated with more toxic phthalates such as DEHP and DBP than DiNP or DiDP of lesser toxicity (Zhang et al., 2006). DEHP exposure in China has been reported to be similar to that in the U.S. and Japan, but DBP exposure has been shown to be almost ten times higher than in the U.S. and Japan (Guo et al., 2012). Diet was the major source of exposure to DEHP (accounting for >50% of the total DEHP exposure) and an important source of DBP (sum of DiBP and DnBP, accounting for >10% of the total DBP exposure) in China (Guo et al., 2013). The Chinese people have unique dietary habits, and the impact of Chinese diets on phthalate exposure is of special interest in better understanding phthalate exposure in China. To the best of our knowledge, only one study has discussed this topic, with phthalate exposure in school children linked to FFQ data (Guo et al., 2012). In the current study, we used both 24-h recall and FFQ data to identify associations between food consumption and phthalate exposure in Shanghai adults.

2. Methods

2.1. Study population and sampling

The study participants were Shanghai residents who participated in the Shanghai Food Consumption Survey (SHFCS). The SHFCS was performed by Fudan University from September 2012 to August 2014 using a four-time 24-h dietary recall questionnaire to collect the seasonal data of food consumption in the community-based general population (fall 2012, spring and winter 2013, and summer 2014), and a one-time 24-h dietary recall survey to collect the food data in school-based students with ages ≤ 18 years. All participants in the SHFCS also completed the FFQ. However, not all school-based students provided the spot urine samples, and we measured the phthalate metabolites in only the community-based population with ages > 18 years at the time this paper was drafted. The study population in this analysis was therefore community residents of ages > 18 years.

The community-based SHFCS used the multi-stage cluster random sampling method to draw samples from nine of the 18 districts/counties in Shanghai, including Huangpu, Xuhui, Putuo, Hongkou, Jinshan, Pudong, Qingpu, Baoshan, and Chongming. Based on population density, one to six residential communities were randomly selected from each district/county. A total of 25 communities were selected throughout the city. In the first interview (fall 2012), 3322 participants were asked to complete a 24-h dietary recall survey and FFQ in the presence of trained dietitians. Additionally, anthropometric measurements and sociodemographic characteristics were recorded. Spot urine samples were obtained from 3082 participants and stored at −20 °C. After the exclusion of 89 participants lacking weight or height information, 326 with insufficient urine samples for the detection of phthalate metabolites, 25 for unreasonable creatinine concentration (<20 μmol/L or >30,000 μmol/L), and 224 with age ≤ 18 years, 2418 participants with ages > 18 years had complete information on anthropometric measurements, demographic characteristics, food consumption, and phthalate metabolites. Written informed consent was obtained from all participants. The study was approved by the local authorities and the Ethics Committee of the School of Public Health at Fudan University.

2.2. Dietary assessment

Dietary intake was assessed by the 24-h dietary recall survey and FFQ by trained dietitians during face-to-face interviews. The 24-h dietary recall survey gathered information on the types and amounts of foods consumed in the 24 h prior to the spot urine collection. A total of 312 food types were consumed. We grouped these 312 types of foods into the following 19 food categories: rice and products (rice); wheat and products (wheat); other staples; vegetables and products (vegetables); legumes and products (legumes); fungi and products (fungi); fruits and products (fruits); eggs and products (eggs); pork; beef and mutton; poultry; processed meats; visceral products (viscera); fish; other aquatic products (other aquatic); dairy products; beverages; nuts and snacks; and alcohol.

In the FFQ, participants reported the yearly consumption frequency ("never or seldom", "less than once per month", "one to three times per month", "one to three times per week", "four to six times per week", "once per day", "twice per day", and "three or more times per day") and the average mass of each meal for the following 23 food categories: rice and products (rice); wheat and products (wheat); other staples; vegetables and products (vegetables); legumes and products (legumes); fungi and products (fungi); fruits and products (fruits); pork; beef and mutton; viscera; eggs and products (eggs); fish; other aquatic products (other aquatic); processed meats; poultry; cow's milk, yogurt, soy milk; nuts and snacks; beverages; beer; white wine; red wine; and spirits. To simplify the data analysis, we combined cow's milk, yogurt, and soy milk into a "dairy products" category, and beer, white wine, red wine, and spirits into an "alcohol" category.

2.3. Measurement of phthalate metabolites in urine

One spot urine sample from each participant was collected in glass tubes capped with polypropylene lids. Both tubes and lids had been previously washed to remove the background phthalates. We measured 10 phthalate metabolites, including monomethyl