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Occupational exposure assessment of phthalate esters in indoor and outdoor microenvironments

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ABSTRACT

Phthalate esters (PAEs) are widely used as plasticizers in consumer products. PAEs are a group of environmental hormone which disrupts human and animals' endocrine systems. Different occupational groups are exposed to various levels of PAEs. In the present study, four typical occupational groups were chosen, including doctors, college teachers, college students, and drivers who worked in public traffic system. In order to understand the exposure levels to PAEs *via* inhalation, air samples were collected from multiple microenvironments including indoor and outdoor in Hangzhou to measure the gas and particle concentrations of six PAEs, together with time spent in different microenvironments of these four groups. A comprehensive PAEs exposure model was built to estimate the daily PAEs exposure through inhalation, oral and dermal pathways. The Monte Carlo simulation results show that doctors were exposed to the highest level of PAEs, and consequently had the highest health risk among these four occupational groups. In contrast, college students had the lowest health risk. By setting the exposure level of staying in residences as the baseline, doctors and drivers were two occupations exposed to high PAEs health risk. Di-(2-ethylhexyl) phthalate (DEHP) was the largest contributor among the six phthalates, posing moderate health risk (10^{-5}–10^{-6}) to every occupation. For traffic microenvironments alone, the total exposure levels for different transportation modes were in the descending order of busses, cars, cabs, tubes, motor bikes, and walking.

Introduction

Phthalate esters (PAEs) are typical semi-volatile organic pollutants that have been widely used in daily necessities especially soft-polyvinyl chloride (PVC) as plasticizers, and also can be used as raw materials for consumer products (Hauser and Calafat, 2005; Guo et al., 2014), *e.g.*, pesticide carriers, repellents, cosmetics, fragrance products, and lubricants (Zeng et al., 2008). However, some studies showed that PAEs as a kind of environmental hormone can disrupt human and animals' endocrine system (Mankidy et al., 2013; Hart et al., 2014), *e.g.*, decreased immunity, disorders of the nervous system, and reproduction dysfunction (Mankidy et al., 2013). Specifically several rodent experiments have proved that PAEs can cause the fetuses in the wombs of the female rodent's deformity and even death (Ema et al., 1996). While it has been confirmed that PAEs have carcinogenicity like high doses of di-(2-ethylhexyl) phthalate (DEHP) can lead to the proliferation of stem cells, and the formation of liver tumors (Elcombe et al., 2002). Also DEHP, di-*n*-butyl phthalate (DBP), and butyl benzyl phthalate (BBP) produced reproductive and developmental toxicities (Lyche et al., 2009) in rodents. Therefore, people would get chronic injury through prolonged exposure to PAEs especially sensitive populations, such as infants and pregnant women. The United States Environmental

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Protection Agency (EPA) has classified DEHP as a kind of probable human carcinogen (Bu et al., 2016).

Currently more and more studies focused on exposure assessment of PAEs. Human exposure pathways to PAEs in air included inhalation, oral ingestion, and dermal absorption (Diaz-Barriga, 1994), and daily PAEs intake for people has been estimated by indirect calculation or combining with biomonitoring (Hines et al., 2009, 2011). Researchers sampled foodstuffs, ambient particles, drinking water, soil, and indoor particles in Tianjin, concluded that contaminated air was the important source of people's daily PAEs intake, accounting for 45% of total PAEs exposure (Ji et al., 2014). In soil and street dust studies, recent researches demonstrated children were inclined to exposure to PAEs by getting in touch with some products containing PAEs (Hu et al., 2003; Skrbic et al., 2016). At the same time, the estimation results showed daily PAEs intake for children was about four times higher than that for adults (Skrbic et al., 2016). Wang et al. (2015b) collected drinking water samples from 19 main water source areas in Zhejiang Province, drawing a statement that human activities and industrial emissions were the most important reasons for PAEs pollution which increased the health risk for humans especially citizens. Sources of PAEs in air were complex, and it was important for us to find out people's exposure level of PAEs in air.

Compared with outdoor environments, humans spent over 80% of their time in indoor environments in their daily lives, and indoor environment quality had a significant impact on human health. For indoor PAEs pollution, there was a certain relationship between children's asthma and high concentration of indoor PAEs which has been proved by several papers (Callesen et al., 2014; Shu et al., 2014; Beko et al., 2015). Most traditional exposure assessments to PAEs only considered general environment or a specific microenvironment alone (Wang et al., 2014), but studying all kinds of microenvironments during personal daytime was essential because exposure time and concentration of contaminants were quite different in various microenvironments. Especially, traffic microenvironments (Nasir and Colbeck, 2009) were also a kind of important exposure environment besides conventional daily exposure microenvironments such as offices, residences, schools. A paper (Schnell et al., 2016) which classified urban environments into different types suggested that studying different types of microenvironments separately would provide a better understanding of human exposure assessment than results estimated based on all mixed measures. Moreover, not only the total environment should be categorized, but also population should be classified into different occupational groups. Some studies concentrated on this point found that occupational groups from various sectors shared different exposure levels of PAEs (Hines et al., 2009, 2011; Kolena et al., 2014), but they did not calculate complete daily PAEs exposure for different occupational groups in various microenvironments. According to time distribution model in microenvironments which was based on random model, whole microenvironments people experienced every day could be divided into five parts: residence, work environment, other indoor environment (except living and work environment), transport environment, and other outdoor environment. To calculate completed daily PAEs exposure for different groups, relevant exposure data in the above five kinds of microenvironments should be synthetically considered at the same time.

Also the uncertainty should be considered in exposure assessment. Combining some random model can assign parameters determined by input information of exposure models to a probability distribution before model simulating; then, get whole exposure level distribution of target population. Relevant typical studies included: Stochastic Human Exposure and Dose Simulation (SHEDS) (Mokhtari et al., 2006; Liu et al., 2009; Baxter et al., 2011; Jiang et al., 2011), Simulation of Human Activity and Pollution Exposure (SHAPE) (Ilie et al., 2016), Air Pollution Exposure (AirPEX) (Freijer et al., 1998). These studies built models to calculate some chemicals' exposure levels based on measuring or model predictive data to complete human specific exposure assessment. However, uncertainty runs through whole process of the exposure assessment (Stanek, 1996), including model framework, and the parameters in models. Thus, results given by models need uncertainty analysis. Monte Carlo simulation (Cely-Garcia et al., 2017; Pelletier et al., 2017) is a common analysis method in confirmation of parameters' uncertainty in quantitative risk analysis, and its core is repeating sampling from the probability distribution of several input variables to establish the distribution of output results. This paper chose Monte Carlo simulation for the sensitivity and uncertainty analyses.

In this study, we focused on four kinds of important occupational groups in Hangzhou, China, including doctors, college teachers, college students, and drivers. Among these groups, drivers were specified to cab, bus, tube, or other drivers that worked in the public transportation system. Reasons for choosing these four kinds of occupations were that high concentrations of PAEs have been detected in doctor offices, hospital wards, and some public transportation carriages (Pei et al., 2013; Wang et al., 2015a; Chi et al., 2017), so exposure levels of PAEs for occupational groups who worked in those microenvironments needed to be discussed. Furthermore, people exposed to classrooms for a long time have proven the existence of inhalation health risk from PAEs (Wang et al., 2017); therefore, these four kinds of occupational groups were selected in this research. During 2011–2017, indoor PAEs concentrations in more than ten kinds of microenvironments in Hangzhou were measured, which provided the basis for our occupational daily PAEs exposure assessment research. This study aims to evaluate the above occupational groups' daily PAEs exposure levels by different pathways in five kinds of microenvironments on the basis of Monte Carlo simulation to provide completed occupational exposure assessment of PAEs, and according to exposure assessment, optimize human routine transportation mode in urban area of Hangzhou. We hope to build holonomic exposure assessment to different groups, especially to some high exposure risk groups, and provide healthier traffic choices.

1. Materials and methods

1.1. Sampling and measuring of indoor PAEs

From 2011 to 2017, we simultaneously monitored the gas and particulate concentration of 6 main PAEs including dimethyl phthalate (DMP), DBP, BBP, diethyl phthalate (DEP), DEHP, and dinoctyl phthalate (DOP). All the indoor microenvironments we chose were located in Hangzhou, China. We tested 6 kinds of