Environmental Pollution 236 (2018) 626–633

Contents lists available at ScienceDirect

Environmental Pollution

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

Exposure to organophosphate flame retardants of hotel room attendants in Wuhan City, China☆

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ARTICLE INFO

Article history:
Received 11 September 2017
Received in revised form
22 January 2018
Accepted 22 January 2018

Keywords:
Organophosphate flame retardants
Urinary metabolites
Dust
Hand-wipe
Hotel environment

ABSTRACT

Indoor environments provide sources of exposure to organophosphate flame retardants (PFRs), which are artificially synthesized fire-protecting agents used as additives in interior products. As public spaces, hotels are required to meet stricter fire-precaution criteria. As such, room attendants may be exposed to higher levels of PFRs. Our goal was to characterize the exposure of hotel room attendants to PFRs by measuring metabolites in their urine and the corresponding parent PFRs in dust and hand-wipes collected from 27 hotels located in Wuhan City, China. The exposure of the attendants was found to be omnipresent: urinary metabolites of PFRs, such as DPHP (diphenyl phosphate), BDCIPP (bis(1,3-dichloro-2-propyl) phosphate), and DoCP (di-o-cresyl phosphate) & DpCP (di-p-cresyl phosphate) were detected with high frequency (87%, 79% and 87%, respectively). We observed that metabolites in post-shift urine were consistently present at higher levels than those in the first morning voids ($p < 0.05$ for BDCIPP and DPHP). Regarding external exposure, 10 PFRs were determined in both dust samples and hand-wipes, with TCIPP (tris(2-chloroisopropyl) phosphate) being the most abundant compound in both matrices. The levels of PFRs in hand-wipes and dust samples were not correlated. PFRs in dust and their corresponding urinary metabolites were not significantly correlated, while a moderate significant correlation of TDCIPP (tris(1,3-dichloro-2-propyl) phosphate) in hand-wipes and its urinary metabolite, BDCIPP, was observed in both morning void samples ($p = 0.01$) and post-shift urine ($p = 0.002$). Moreover, we found that participants from high-rise buildings (defined as > 7 stories) had significantly higher BDCIPP and DPHP concentrations than those from low-rise buildings. A possible reason is that high-rise buildings may use high-grade fireproof building materials to meet stricter fire restrictions. Overall, these results indicate that PFRs exposure in hotels is a contributor to the personal exposure of hotel room attendants.

1. Introduction

Organophosphate flame retardants (PFRs) are artificially synthesized compounds extensively utilized as fire-protecting agents and plasticizers in the manufacturing industry worldwide (Wei et al., 2015). Numerous consumer products are key sources of PFRs in the indoor environment (Wang et al., 2017). PFRs have been detected in upholstery (polyurethane foam cushions, mattresses, sofas (Stapleton et al., 2009) and textiles (Van der Veen and de Boer, 2012)), electronic equipment (Kajiwara et al., 2011), paints (Van der Veen and de Boer, 2012), and various construction and finishing materials (Wang et al., 2017).

Since PFRs are frequently present as nonreactive additives in commercial manufacturing, which results low affinity to finished

☆ This paper has been recommended for acceptance by Eddy Y. Zeng.

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https://doi.org/10.1016/j.envpol.2018.01.079
0269-7491/

products, these chemicals can gradually migrate from the products into the indoor environment over time (Kanazawa et al., 2010). Consequently, PFRs exist in indoor dust and air in different indoor microenvironments that are closely related to our daily life (Zhou et al., 2017), such as residential buildings (Langer et al., 2016), public buildings (offices (Carignan et al., 2013), schools (Mizouchi et al., 2015), daycare centers (Langer et al., 2016), and hotels (Takigami et al., 2009)) and vehicles (Brandsma et al., 2014). Studies have indicated that exposure to PFRs depends on indoor human activities (Carignan et al., 2013; Cequier et al., 2015; Hoffman et al., 2014, 2015; Kucharska et al., 2015), and such exposure poses potential health risks. Epidemiologic studies have found that the body burdens of TCIPP (tris(2-chloroisopropyl) phosphate), TCEP (tris(2-chloroethyl) phosphate), TNBP (tri-*n*-butyl phosphate), and TPHP (triphenyl phosphate) are associated with elevated DNA oxidative stress (Lu et al., 2017). Exposure to PFRs is also associated with changes in human sphingolipid homeostasis (Zhao et al., 2016), the alteration of thyroid hormone levels and poorer semen quality (Meeker and Stapleton, 2010), and these relations may ultimately result in adverse health effects.

Therefore, monitoring the extent of an individual's internal exposure as well as possible exposure sources of PFRs is a high priority for evaluating potential human health risks. In several recent studies, urinary metabolites were measured and used as biomarkers for PFR exposure (Cequier et al., 2015; Hoffman et al., 2017; Meeker et al., 2013; Ospina et al., 2018). Previous work has suggested that dust is an important source of PFRs exposure (Cequier et al., 2014; Mitro et al., 2016; Mizouchi et al., 2015). Moreover, PFRs in hand-wipes can be seen as an external contributor to exposure through dermal absorption or hand-to-mouth pathways, as well as an indicator of body burden (Hoffman et al., 2015; Liu et al., 2017a). Several studies have reported the detection of PFRs on hands; therefore, skin surfaces are also an alternative contributor to the body's burden (Hoffman et al., 2015; Liu et al., 2017a, 2017b). As such, associations have been observed between certain PFR metabolites in the urine of individuals and the corresponding parent compounds of the PFRs in indoor dust or hand-wipes (Dodson et al., 2014; Hoffman et al., 2015).

The hotel environment, in some ways, is similar to the household environment. In addition, as public spaces, commercial hotel environments must comply with more stringent fire-precaution criteria (Cao et al., 2014; Takigami et al., 2009). Flammability criteria are the main drivers of the use of large volumes of flame retardants (Dodson et al., 2017). Consequently, comparable or even higher PFR levels are expected to be present in hotels compared to the residential environment. Cao et al. (2014) found higher PFR levels in hotels compared to other microenvironments in China, and Takigami et al. (2009) investigated the occurrence of flame retardants in dust in one hotel in Japan. Moreover, the extent of human exposure may increase due to the occupational activities of the attendants, for example, housekeeping, which is presumed to cause more constant contact with dust and other emission sources of PFRs in the environment during the work period. However, because only 4 hotels were included in the former investigations, uncertainties still remain as for the generalizability of those results. Additionally, the lack of biological measures precludes correlating external exposure with the body burdens of PFRs.

In the present study, we hypothesized that the occurrence of PFRs in hotels is a contributor to the personal exposure of hotel room attendants. We determined the levels of DNBP (di-*n*-butyl phosphate), BDCIPP (bis(1,3-dichloroiso-propyl) phosphate), DPHP (diphenyl phosphate), BBOEP (bis(2-butoxyethyl) phosphate) and DoCP (di-o-cresyl phosphate) & DpCP (di-p-cresyl phosphate) in urine samples from 26 attendants and monitored the within-person diurnal variability of urinary metabolites during the workday; the external exposure levels of 10 PFRs were also determined in the dust and hand-wipes collected from 27 commercial hotels. Furthermore, we investigated the correlations between the biological levels and external measures of paired hand-wipe and dust sample. Additionally, we also examined potential predictors of the body burdens of PFRs.

2. Methods

2.1. Sample collection

In the sampling campaign, 27 commercial hotels from the central urban area of Wuhan City were studied (Fig. S1). All sample collection was conducted between December of 2016 and April of 2017. To study the general external occurrence of PFRs in the 27 hotels, we collected 40 dust samples and 38 hand-wipes (27 pairs) (details in Table S2). Since each staff member was assigned to duty on one floor and a limited area, which included guest rooms, corridors, and other public locations, sample collections did not cause interference. Among the room attendants who provided hand-wipes, 23 were willing to provide urine samples, but 3 participants who did not provide hand-wipes were willing to provide urine. The demographic characteristics of the attendants ($n = 26$) are listed in Table S1.

Each room attendant who worked 8 h per day provided a sample of urine on a workday at the time they were about to finish their work shift at the hotel (4:30 p.m.) and a first-morning urine from the next day at home. A total of 52 urine samples were stored in polypropylene containers at −20 °C. All the attendants gave their informed consent, and the study was approved by the Tongji Medical College Ethical Committee.

We sampled dust from vacuum cleaners that had been utilized on hotel floors by the room attendants during daily cleaning jobs. Each sample was collected in a clean paper bag, wrapped in aluminum foil and then finally placed in a polyethylene sealed bag and stored at −20 °C.

The procedure of hand-wipe collection was described in previous study (Hoffman et al., 2015). Briefly, laboratory staff (wearing gloves) collected a sterile gauze pad that had been soaked in 5 mL of isopropyl alcohol prior to use the pad thoroughly wipe the front and back of both hands of each attendant twice from the fingertips to the wrist. The hand-wipes were sampled at the end of a shift, sealed in 50 mL polystyrene centrifuge tubes and stored at −20 °C.

2.2. Analysis of urine sample

Urine samples were prepared following a simple procedure. For each sample, 1 mL of urine was spiked with the internal standards BBOEP-d4, DPHP-d10, and BDCIPP-d10 (1 ng, each). A volume of 3 mL of acetonitrile was mixed with the urine after pH adjustment (350 μL; 1 M sodium acetate buffer, $pH = 5$) and shaken for 30 s. After centrifugation (Eppendorf) for 3 min at 8000 rpm, 5 mL of methyl *tert*-butyl ether was added, and the mixture was vortexed for 30 s and then again subjected to centrifugation at 8000 rpm. The supernatant (organic phase) was removed from the white precipitate and yellowish water phase, concentrated to near dryness and reconstituted with 100 μL of MeOH:H_2O (10:90, v:v). The extract was filtered through a 0.22-μm membrane filter.

The analysis of urinary metabolites of PFRs was performed using an LC-30A UPLC system coupled to a triple-quadrupole LCMS-8050 (Shimadzu, Japan) in the ESI negative mode. The MS/MS parameters were as previously described (Van den Eede et al., 2015).