Chemosphere 205 (2018) 71–79

Contents lists available at ScienceDirect

Chemosphere

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

Human exposure to brominated flame retardants through dust in different indoor environments: Identifying the sources of concentration differences in hair from men and women

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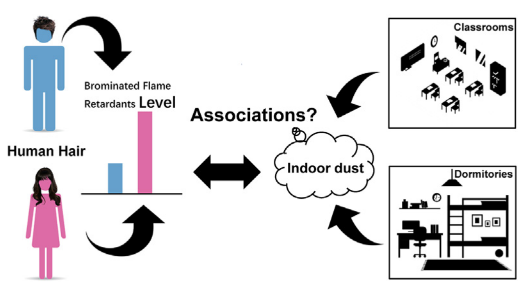
HIGHLIGHTS

- PBDE concentrations were significantly higher in female hair than in male hair.
- BFR profile differences in hair were affected by indoor environments.
- Academic and residential environments were considered in combined exposure assessment.
- BFRs were measured in human hair and dormitory and classroom dust collected from Beijing, China.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 January 2018
Received in revised form
15 March 2018
Accepted 19 March 2018
Available online 16 April 2018

Handling Editor: J. de Boer

Keywords:
Brominated flame retardants
Hair
Gender disparity
Indoor dust
Human exposure

ABSTRACT

Brominated flame retardants (BFRs) can accumulate in humans and are associated with adverse health effects. The study was conducted to determine the differences in Polybrominated diphenyl ethers (PBDEs) and alternative brominated flame retardant (Alt-BFR) concentrations between men and women. We analyzed hair samples from 14 male and 20 female university students, paired dust samples from their dormitories (10 for males and 8 for females), and six dust samples from university teaching buildings. The total PBDE concentrations in hair from females were significantly (three times) higher ($p = 0.012$) than that from males (means 372 and 109 ng/g, respectively). The mean total PBDE concentrations in classroom and dormitory dust were 36100 and 2012 ng/g, respectively. The PBDE patterns were different in the male and female hair samples, as were the patterns in the classroom and dormitory dust. There are no reports concerning human exposure to BFRs through dust that was assessed considering academic and residential environments simultaneously. The differences between BFR exposure for males and females and the differences between BFR concentrations in hair samples from males and females were consistent for 71.4% of the compounds. However, using only dormitory dust in the calculations gave consistent differences only for 28.6% of the compounds, suggesting that the BFR concentration differences in hair were mainly because females spent much more time than males in classrooms.

1. Introduction

Brominated flame retardants (BFRs) are used in various

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https://doi.org/10.1016/j.chemosphere.2018.03.133
0045-6535/

commercial and consumer products, e.g., electronics, furniture, and upholstery (Hoh et al., 2005; Wang et al., 2010). Polybrominated diphenyl ethers (PBDEs) were some of the most widely used BFRs (Harrad et al., 2008b; Zhu et al., 2015) but now have mostly been withdrawn and replaced with alternative flame retardants. For example, the commercial mixture penta-BDE was predominantly replaced by Firemaster 550, which contains di(2-ethylhexyl)tetrabromophthalate (TBPH) (Venier et al., 2012). Other replacements include hexabromobenzene (HBB), pentabromoethylbenzene (PBEB), pentabromobenzene (PBBZ), pentabromotoluene (PBT), pentabromobenzyl acrylate (PBBA), and 2,3,5,6-tetrabromo-*p*-xylene. Some of these replacement flame retardants have also become environmentally ubiquitous (Salamova et al., 2014; Venier et al., 2014; Liu et al., 2016). BFRs may disrupt the endocrine and reproductive systems, have neurotoxic effects, and cause learning disabilities and obesity (Dingemans et al., 2011; Patisaul et al., 2013; Yanagisawa et al., 2014).

Many studies of BFR concentrations in human tissues have recently been published (Lee et al., 2007; Sjodin et al., 2008; Zheng et al., 2014). The chemical structures of PDBEs are similar to those of thyroid hormones triiodothyronine and thyroxine (Zhang et al., 2008), and exposure to polyhalogenated aromatic hydrocarbons, especially PBDEs, may increase thyroid cancer incidence (Zhang et al., 2008; Berg et al., 2017). Thyroid cancer is relatively common, but males and females have different incidence rates. The age-adjusted thyroid cancer incidence rate was 11 per 100000 people in 2006, but the incidence rate was 2.9 times higher for women than for men (Rahbari et al., 2010). This disparity has also been found in China. Shaanxi Provincial People's Hospital recorded 849 thyroid cancer cases over 10 y and male:female ratios of 1:2.88 and 1:3.33 for all patients and for patients aged <25 y, respectively (Wu et al., 2015). The average age for thyroid cancer diagnosis in China has decreased. This trend was first found in Qingdao (Guo et al., 2016), and the number of thyroid cancer patients aged 20–29 y in Kunming has been increasing each year since 2008 (Zhao et al., 2012).

Disparities in thyroid cancer incidence, aggressiveness, and prognosis between males and females are well known, but the causes of these disparities are poorly understood (Rahbari et al., 2010). The morbidity of thyroid goiter, the precursor of thyroid cancer, showed an annual increase in South China Agricultural University, Guangzhou, and it increased with the increase in age of college students. The morbidity among females was 12.6 times higher than that among males in the college (Mingyi et al., 2012), which provided a more considerable gender disparity than nonspecific population. Different BFR concentrations have been found in tissues from men and women (Król et al., 2014; Zheng et al., 2014), but the causes have not been identified. It is necessary to determine whether men and women, college students more importantly, have different BFR exposure patterns and if so why. Such data would allow us to determine whether different BFR exposure patterns cause the disparity in thyroid cancer incidence in males and females. Exposure to BFRs can be reflected in BFR concentrations in hair (Poon et al., 2014; Yuan et al., 2016). BFRs are effectively accumulated in hair, which contains 88% protein and 3–4% lipids. Hair sampling is noninvasive and harmless (Poon et al., 2014). We therefore used hair to monitor BFR exposure in this study.

Indoor dust is generally the main source of human exposure to PBDEs (Johnson-Restrepo and Kannan, 2009). Dust exposure accounted for 82% of the estimated total PBDE intake in North America (Lorber, 2008). Indoor dust has been linked to human exposure to BFRs in previous studies (Chen et al., 2008; Harrad et al., 2010). The risks posed to infants, toddlers, children, teenagers, and adults by exposure to BFRs through dust have been determined (Wang et al., 2014a; Zhu et al., 2015). No distinction has been found previously between males and females when calculating the risks posed by BFR exposure. It is important to determine whether differences in indoor dust exposure patterns cause differences in BFR exposure for males and females and to determine how exposure calculations match reality.

The aims of this study were (1) to investigate the exposure of men and women to BFRs by analyzing hair samples from students of the Minzu University of China and (2) to estimate the exposure to BFRs in indoor dust for men and women and investigate the role indoor dust plays in BFR concentration differences in hair from men and women.

2. Materials and methods

2.1. Sampling

Hair and dust samples were collected from the Minzu University of China, Beijing. A 1 g hair sample was collected from the occipital area of each of 34 students (14 males, 20 females), and a 1 g dust sample was collected from each of the 8 male dormitories and 10 female dormitories. Each dormitory dust sample was paired with 1 or 2 hair samples. Each volunteer completed a lifestyle questionnaire. If two persons sharing a dormitory gave different answers about their dormitory, the average was used. The questionnaire is shown in Table A.1 and the results are given in Table A.2.

The female dormitory 1 and male dormitory 1 dust samples were labeled FD1 and MD1, respectively. The paired female and male hair samples were labeled FH1-1 and FH1-2 and MH1-1 and MH1-2, respectively. Dust samples were collected from three classrooms on each of the six floors of the main teaching building where academic classes and daily study of students were held. The samples from each floor were combined, thus giving six pooled samples (one for each floor). The classroom dust samples were labeled TB-F1 (teaching building first floor), TB-F2, etc. No classroom contained an air purifier. Each dust sample was collected from the floor and furniture using a vacuum cleaner containing a dust unit that was easily removed and emptied. A new filter was used for each sample. Each sample was wrapped in an aluminum foil, sealed in a plastic bag, and transported to the laboratory at 5 °C. The samples were stored at −20 °C in the laboratory. The samples were collected in June and July 2016.

2.2. Chemicals

Pesticide grade acetone, *n*-hexane, and dichloromethane were obtained from J.T. Baker (Phillipsburg, NJ, USA). High-purity nitrogen was obtained from Cheng Wei Xin (Beijing, China). Ultra-pure water was produced using a Milli-Q system (EMD Millipore, Billerica, MA, USA). Analytical grade anhydrous sodium sulfate was baked at 450 °C for 5 h before use. Silica gel (100–200 mesh; Merck, Darmstadt, Germany) was activated at 105 °C for 12 h, cooled, and deactivated by adding 3% of the sorbent weight of deionized water.

Individual unlabeled PBDE standards (BDE-28, BDE-47, BDE-66, BDE-85, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183, BDE-190, and BDE-209) were purchased from Cambridge Isotope Laboratories (Andover, MA, USA). A standard containing unlabeled PBEB, PBBA, PBT, and TBPH was purchased from AccuStandard (New Haven, CT, USA). An unlabeled HBB standard was purchased from Dr. Ehrenstorfer (Augsburg, Germany). Unlabeled 2,3,5,6-tetrabromo-*p*-xylene and PBBZ standards were purchased from Wellington Laboratories (Guelph, Canada). Internal standards ($^{13}C_{12}$-labeled BDE-139, $^{13}C_{12}$labeled-BDE-209, and $^{13}C_6$-labeled HBB) were purchased from Cambridge Isotope Laboratories.