Journal of Hazardous Materials 359 (2018) 491–499

Contents lists available at ScienceDirect

Journal of Hazardous Materials

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

Amplification effect of haze on human exposure to halogenated flame retardants in atmospheric particulate matter and the corresponding mechanism

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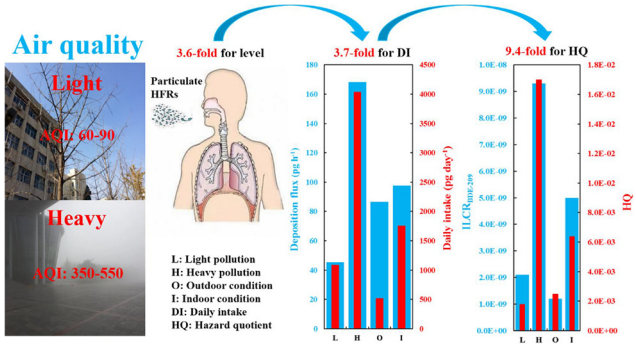
GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Halogenated flame retardants
Haze
Size distribution
Health risk

ABSTRACT

The health impact of haze is of great concern; however, few air quality studies have investigated trace pollutant contamination in the air. Size-segregated atmospheric particles (nine size fractions derived from PM_{10}) were collected in dwelling (indoor) and traffic (outdoor) environments in Xinxiang, China, during light pollution conditions (air quality index (AQI), 60–90) and heavy pollution conditions (AQI, 350–550), and they were analysed for halogenated flame retardants (HFRs), including polybrominated diphenyl ethers (PBDEs), novel brominated flame retardants (NBFRs) and Dechlorane Plus (DP) isomers. HFR occurrence levels generally decreased in the order of PBDEs > NBFRs > DPs. The total mean abundance ratios of heavy pollution/light pollution were 4.0, 2.9, 4.4 and 3.6 for PBDEs, NBFRs, DPs and HFRs, respectively. Meteorological conditions played distinctive roles in the HFR distribution in the air. Apparent differences were found for the particle size distribution of HFRs under light and heavy pollution conditions. In general, for adults, the estimated hazard quotient (HQ) and incremental lifetime cancer risk ($ILCR_{BDE\text{-}209}$) values were approximately 1.7×10^{-2} and

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https://doi.org/10.1016/j.jhazmat.2018.07.109
Received 15 March 2018; Received in revised form 21 July 2018; Accepted 30 July 2018
Available online 02 August 2018
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9.3×10^{-9} in heavy pollution conditions, respectively, which were significantly higher than those in light pollution conditions (1.8×10^{-3} and 2.1×10^{-9}, respectively).

1. Introduction

Halogenated flame retardants (HFRs), particularly polybrominated diphenyl ethers (PBDEs), novel brominated flame retardants (NBFRs) and Dechlorane Plus (DP) isomers, have been widely used in consumer products, such as electronic products and household materials, to reduce flammability [1]. PBDEs are the dominant compounds in HFRs and include the commercially available Penta-BDE, Octa-BDE and Deca-BDE products [2,3]. PBDEs fall under the Stockholm Convention list of persistent organic pollutants (POPs) because of their persistence and toxicity and the threat they pose to the environment and human health [4]. Therefore, NBFRs have emerged as alternatives to PBDEs. The European Union recommended syn-DP and anti-DP as possible substitutes for Deca-BDE commercial materials in several applications [5]. Because of the intensive application and toxicity of HFRs, their occurrence, fate, and behaviour and the consequent human health risks have caused increasing concern in recent years [6,7].

In ambient air, HFRs are partitioned between the gaseous and particulate phases similar to other semi-volatile organic compounds (SVOCs) [7]. The particulate phase has crucial importance for HFRs because many of these compounds have relatively low vapour pressures and high octanol-air partition coefficients (K_{OA}). In the atmosphere, HFRs tend to bind to particulate matter and could easily be resistant to metabolic breakdown and photodegradation [6]. One of the significant factors determining the atmospheric transport of particulates is their aerodynamic diameter [7,8]. The particle size distribution of HFRs is of significant importance because human inhalation exposure to particle-bound HFRs is a function of particle size [8,9]. In recent years, studies have mainly investigated particulate PBDEs in individual size fractions, such as PM_{10} and $PM_{2.5}$ [6,10–12]. However, only limited data on the particle size distribution of PBDEs in megalopolises or e-waste recycling areas are available [7,13,14]. The particle size distribution of HFRs in the atmosphere is still poorly understood, and further investigations are urgently needed to identify the behaviours and health effects of these compounds [7,13].

In recent years, severe haze pollution has occurred frequently over China, and the health impact of haze is causing increasing concern [15–17]. Under different haze levels, the concentrations of particulate matter in the air and meteorological factors, including the height of the atmospheric boundary layer (ABL), wind speed, humidity and temperature, vary greatly, often by orders of magnitude [18]. Whether these parameters have a meaningful effect on the HFR distribution and occurrence in the air and whether they have an influence on the comparability among data from different studies have not been determined. To date, only limited studies regarding volatile organic compounds (VOCs) and polycyclic aromatic hydrocarbons (PAHs) have considered the influence of haze pollution on contamination by these compounds [17,19,20]. Little is known about the variation of HFRs in different haze pollution conditions, and how haze pollution affects human exposure to HFRs is still unknown. Moreover, previous studies on HFRs in particulate phases mostly focused on indoor environments, and they seldom performed comparisons between indoor and outdoor environments [21,22]. Outdoor environments, especially traffic environments, have been found to produce an unhealthy effect on human lung function, and exposure to traffic environments caused an immediate and transient increase in arterial stiffness in healthy volunteers [23–25].

The present study addresses these gaps by determining the size distribution of HFRs in particulate samples from dwelling and traffic environments under light and heavy haze pollution conditions. Hence, the major objectives of the present study were (1) to determine the contamination characteristics of HFRs in atmospheric particles in dwelling and traffic environments in Xinxiang, China; (2) to examine the extent to which haze may influence the occurrence and particle size distribution of HFRs; and (3) to assess the health risk of human exposure to HFRs under different air quality conditions.

2. Materials and methods

2.1. Experimental design

The representativeness of samples is essential in the field of environmental monitoring and assessment because it determines the validity of the data. For spatial variation investigations, dozens of sampling sites should be set to guarantee the representativeness of the samples. For other research goals, including temporal variation investigations, the spatial dimension should be ignored, and additional samples should be collected in the temporal dimension [26–29]. Similarly, to explore the influence of haze on HFR contamination in atmospheric particles, the spatial dimension can be ignored and the sampling density under different haze conditions should be the focus. To better eliminate accidental errors, repeated sampling (a total four rounds) was carried out at each indoor and outdoor sampling site under different haze conditions in the present study. Because of the long duration of sampling and limitations on the number of instruments, sample size is naturally limited for atmospheric sampling [8,27,30]. Nevertheless, due to the long sampling time and strong fluidity and homogeneity of air, especially in outdoor environments, the samples in the present study are representative; thus, the results and conclusions obtained from these samples can be generalized [6,8,14,30,31].

2.2. Sampling information

One indoor (dwelling environment) sampling site was set in one newly remodelled apartment building, and one outdoor (traffic environment) sampling site was set at an important main road intersection. The height of the indoor and outdoor sampling sites was approximately 0.6 m above the floor or road surface, respectively. A total of 72 particulate samples (8 sample batches with 9 size fractions in each batch) was obtained through two rounds of sampling at the two sampling sites under light pollution (January 2017, air quality index (AQI) ranging from 60 to 90) and heavy haze pollution conditions (December 2016, AQI value ranging from 350 to 550). Each sample was collected on a Whatman quartz fibre filter (preheated for 4 h at 450 °C) with a diameter of 81 mm using an Anderson eight-stage cascade impactor (Tisch Environmental Inc., Cleves, OH, USA) with a flow rate controlled at 28.3 L min^{-1}. The cutoff aerodynamic diameters for each stage were 9.0–10 (inlet), 5.8–9.0, 4.7–5.8, 3.3–4.7, 2.1–3.3, 1.1–2.1, 0.7–1.1, 0.4-0.7 and < 0.4 μm (backup filter), and the sum of these particles constituted the PM_{10}. The sampling duration was 48 h for heavy pollution conditions and 120 h for light pollution conditions. After sampling, the filter samples were carefully wrapped in aluminium foil and stored at −20 °C until analysis. In addition, meteorological data were recorded during the sampling process, including the AQI, PM_{10}, $PM_{2.5}$, temperature and wind speed.

2.3. Sample analysis

In total, 8 PBDEs (BDE-28, BDE-47, BDE-99, BDE-100, BDE-154, BDE-153, BDE-183 and BDE-209), 4 NBFRs (TBPH, TBB, BTBPE and DBDPE) and 2 DP isomers (syn-DP and anti-DP) were analysed. The

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