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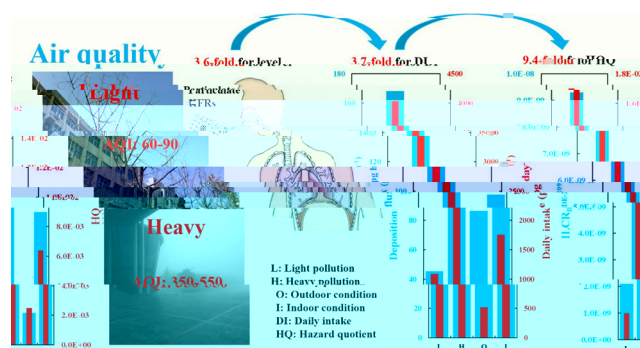
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GRAPHICAL ABSTRACT

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ABSTRACT

The health impact of haze is of great concern; however, few air quality stuogy2hc43935IsaAa(I)uhav(is281(investig/T12saA 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-209) values were approximately 1.7×10^{-2} and

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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

analytical methods were developed on based on a previous report [32]. Detailed information on the full names of the compounds and the analysis procedure is provided in the Supplementary material and Table S1.

2.4. Quality assurance and quality control (QA/QC)

Quality assurance and quality control (QA/QC) was conducted by analysing the field and procedural blanks, standard spiked recoveries and certified dust samples (SRM 2585, NIST, Gaithersburg, MD). One field blank (n = 6), one procedural blank (n = 6) and one certified dust sample (SRM 2585) (n = 6) were run with each detection batch (12 samples). The results showed that only BDE-209 and DBDPE were detected in the blanks at mean amounts less than 1% of that in the samples. The mean values of the blanks were subtracted from the analysed values of BDE-209 and DBDPE in all particulate samples. The standard spiked recoveries of BDE-77 (mean ± SD: 94 ± 18%), BDE-128 (101 ± 24%) and ^{13}C-BDE-209 (106 ± 21%) ranged from 76% to 127%. The results for the HFRs in SRM 2585 showed good repeatability (the relative standard deviation (RSD) for individual HFRs ranged from 3.1% to 28.7%) and were consistent with certified or indicative values [32] (the mean SRM value/certified SRM value ranged from 85.3% to 118.6%). Detailed QA/QC information is provided in Table S2. Most target compounds were detected in all particulate samples except for BDE-28, BDE-100, BDE-183 and TBB, which were only detected in less than 10% of the samples; hence, these target compounds were not reported in the discussion section.

3. Results and discussion

3.1. Atmospheric concentration and composition of HFRs

Fig. 1 shows the individual the HFR concentration ranges, and Table S3 exhibits the mean atmospheric HFR concentrations under different conditions. Among the 8 batches of samples, regardless of the air quality and spatial conditions, the total mean concentration (sum of the 9 stages) of $\Sigma_{10}HFRs$ was 378.1 pg m^{-3}. The HFR concentrations decreased in the order of PBDEs > NBFRs > DPs, which accounted for 44.9 ± 5.6%, 39.6 ± 3.9% and 15.5 ± 2.5% of the total HFRs, respectively. Among the individual PBDEs, NBFRs and DPs, BDE-209, DBDPE and anti-DP were the dominant components (Table S3). For the heavy pollution conditions, the mean $\Sigma_{10}HFR$ concentration was 590.1 pg m^{-3}, which was 3.6-fold higher than that for the light pollution conditions (166.2 pg m^{-3}). Moreover, the mean $\Sigma_{10}HFR$ concentration in the indoor environment was 416.9 pg m^{-3}, which was 1.2-fold higher than that for the outdoor environment (339.4 pg m^{-3}) (Table S4).

3.1.1. PBDEs

For the eight batches of samples, the mean Σ_5PBDE concentration in PM$_{10}$ was 169.9 pg m^{-3}. For the heavy pollution conditions, the mean Σ_5PBDE concentration was 272.8 pg m^{-3}, which was 4.0-fold higher than that for the light pollution conditions (67.8 pg m^{-3}). The concentrations of Σ_5PBDEs and BDE-209 in Xinxiang represented a moderate level in China but were slightly higher than most data from other countries (Fig. 2 and Table S5), where the highest Σ_8PBDE and BDE-209 concentrations of 106 pg m^{-3} and 16 pg m^{-3} were detected in Ispra, Italy [33]. In the present study, BDE-209 was the dominant component in all samples and presented a mean concentration of 162.2 pg m^{-3}, and it accounted for 95.5% of the lower molecular weight Σ_5PBDEs, with the concentrations and proportions of other lower molecular weight congeners presenting considerably lower values.

3.1.2. NBFRs

For the eight batches of samples, the mean concentration of Σ_3NBFRs in PM$_{10}$ was 149.6 pg m^{-3}. For the heavy pollution

conditions, the mean Σ_3NBFR concentration was 222.7 pg m^{-3}, which was 2.9-fold higher than that for the light pollution conditions (76.6 pg m^{-3}). Among the individual NBFRs, DBDPE was the dominant component in all samples, with a mean concentration of 117.0 pg m^{-3}, and it accounted for 78.2% of the Σ_3NBFRs. TBPH accounted for 20.7% of the Σ_3NBFRs, with a mean concentration of 30.9 pg m^{-3}. Conversely, the corresponding proportion of BTBPE was much smaller. The NBFR profiles in this study were similar to those in previous studies [6,34,35]. Although the available data on NBFRs in atmospheric particles are much fewer than those for PBDEs, the total mean concentrations of NBFRs and DBDPE in Xinxiang exhibited a moderate level in China but were higher than most data from abroad (Fig. 2 and Table S6), where the highest Σ_2NBFR and DBDPE concentrations of 23.2 pg m^{-3} and 22 pg m^{-3} were detected in Cleveland, IL, USA [36].

3.1.3. DPs

For the eight batches of samples, the mean concentration of Σ_2DPs was 58.1 pg m^{-3}, and anti-DP was the dominant component, accounting for 71.3% of the Σ_2DPs. For the heavy pollution conditions, the mean Σ_2DP concentration was 95.4 pg m^{-3}, which was 4.4-fold higher than that for the light pollution conditions (21.8 pg m^{-3}). According to a relatively small database, the total mean atmospheric concentration of DPs in Xinxiang exhibited a slightly higher level than that in most research data (Fig. 2 and Table S7).

The isomeric ratio of DPs is a significant tool for assessing the movement and environmental process of these substances [37]. The fractional abundance (f_{anti}) is defined as follows: f_{anti} = anti-DP/(anti-DP + syn-DP). In summary, the f_{anti} value in this study ranged from 0.59 to 0.81, with a mean of 0.72, which was slightly higher than that in the existing limited domestic studies but similar to that in many studies abroad (Fig. 2 and Table S7). Reports have shown that commercial DP products include approximately 35% syn-DP and 65% anti-DP, which results in an f_{anti} value of 0.65; however, previous studies have reported that f_{anti} values in different commercial products were not consistent with the value of 0.65 [37], which may have bene related to different manufacturing techniques and production lots [38]. Furthermore, the physicochemical properties of syn-DP and anti-DP (e.g., solubilities and log K_{OA}) may also contribute to this difference [39].
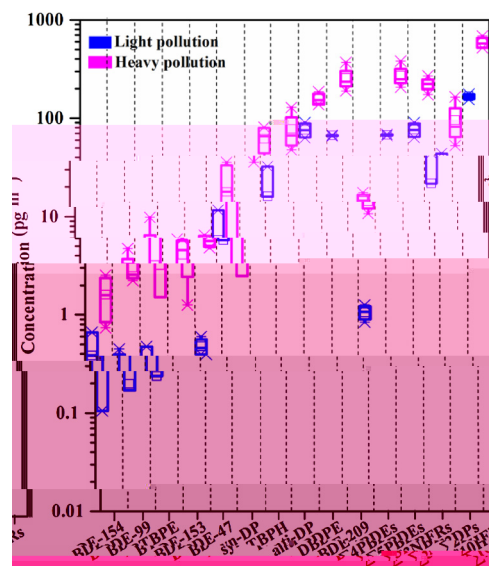

Fig. 1. Concentration ranges (sum of 9 stages) of individual HFRs. Σ_4PBDEs is the sum of BDE-47, 99, 154 and 153; Σ_5PBDEs is Σ_4PBDEs plus BDE-209; and $\Sigma_{10}HFRs$ is the sum of Σ_5PBDEs, Σ_3NBFRs and Σ_2
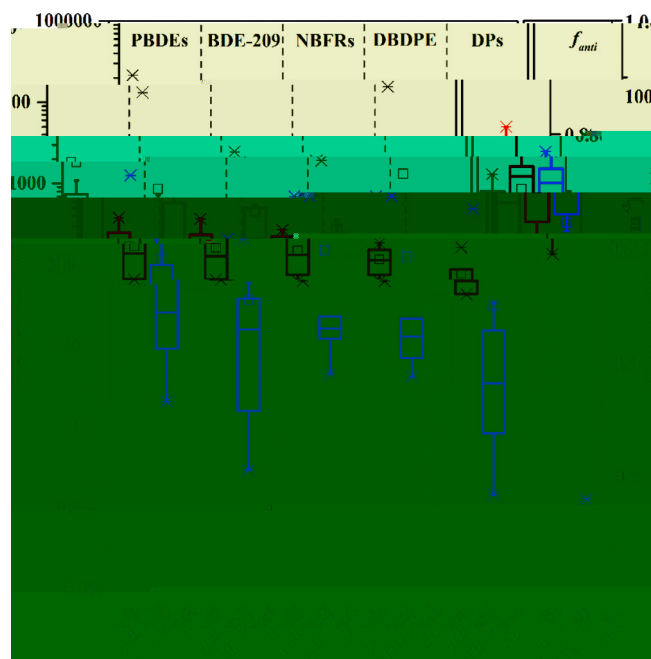
Fig. 2. Comparison of HFR concentrations in the present study with literature data from around the world.

3.2. HFR concentrations under different haze conditions

3.2.1. HFR concentrations under light and heavy pollution conditions

Previous studies have investigated the atmospheric occurrence of HFRs without considering the air quality of the sampling sites over time [6,7,9], which weakens the comparability of data from different studies. Under light pollution conditions, the mean atmospheric PBDE, NBFR, DP and HFR concentrations were 67.8, 76.6, 21.8 and 166.2 pg m^{-3}, respectively, and under heavy pollution conditions, these values were 272.8, 222.7, 95.4 and 590.1 pg m^{-3} (Fig. 3 and Table S3-S4), respectively, indicating that the HFR concentrations in heavy pollution were significantly higher than those in light pollution conditions (Fig. 3). Similarly, Wu et al. found that VOCs (108 species) accumulated at the beginning of haze episodes, and during haze pollution, VOC concentrations were 2- to 5-fold higher than that under non-haze pollution conditions in Beijing, China [19]. Han et al. reported that VOC (26 species) concentrations during haze pollution were ~1.5-fold higher than that on non-haze days in Shanghai, China [17]. To the best of our knowledge, this study is the first to compare the HFR occurrence in atmospheric particles between different air pollution conditions.

Furthermore, the concentration ratios of heavy pollution/light pollution were much higher for lower molecular weight HFRs (ranged from 5 to 25) than for higher molecular weight HFRs (Fig. 3), indicating that lower molecular weight HFRs can massively accumulate in particulate matter during heavy haze processes. Accordingly, because of their relatively lower K_{OA} values, lower molecular weight HFRs in the gas phase can crucially partition into the particulate phase during the process of haze formation when concentrations of particles in the air increase remarkably. Therefore, air quality represents a significant factor influencing particulate HFR concentrations, especially for lower molecular weight HFRs in the atmosphere, whose abundance in the particulate phase can increase significantly when the air quality decreases sharply.

Fig. S1-S5 show the Pearson correlations among the PBDE, NBFR, DP and HFR concentrations and meteorological parameters, including AQI, PM$_{10}$, PM$_{2.5}$, wind speed and temperature. In summary, the PBDE, NBFR, DP and HFR concentrations were positively correlated with the AQI level, with Pearson correlation coefficients of 0.802 (p < 0.05), 0.983 (p < 0.0001), 0.839 (p < 0.01) and 0.942 (p < 0.0001), respectively (Fig. S1). Similar positive correlations also occurred among the PBDE, NBFR, DP and HFR concentrations and PM$_{10}$ and PM$_{2.5}$ levels (Fig. S2-S3). Conversely, wind speed and temperature presented significantly negative correlations with PBDE, NBFR, DP and HFR
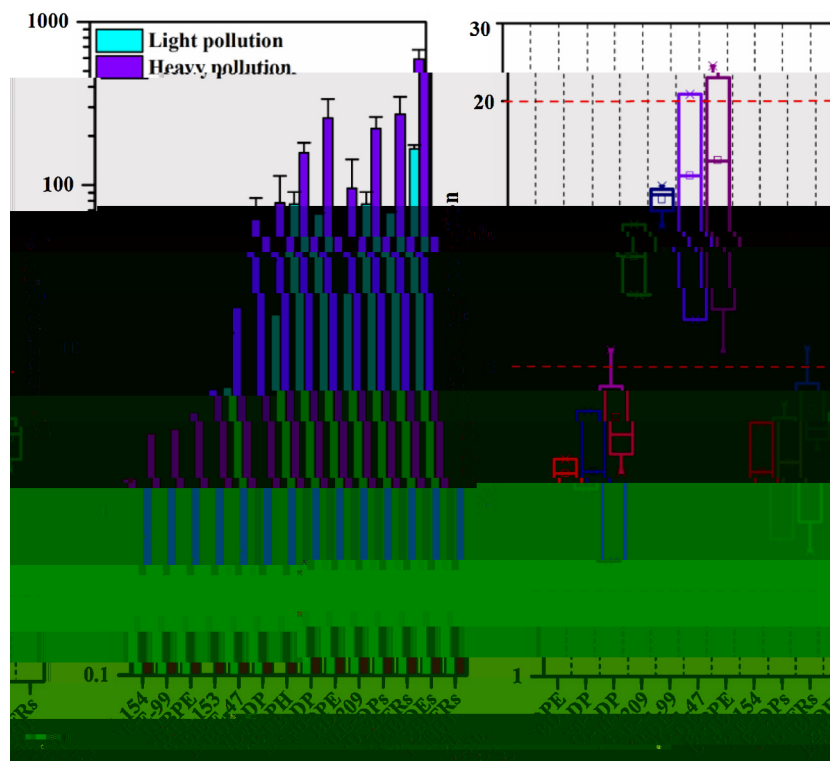

Fig. 3. Comparison of HFR concentrations between different haze conditions and the concentration ratios of heavy pollution/light pollution. Note: The ratios of BDE-153 and TBPH were not included because they were not detected in light pollution conditions.

concentrations (Fig. S4-S5). Consistent with our results, several previous studies have also indicated that lower wind speed, lower temperature, and higher PM_{10} and $PM_{2.5}$ levels were important factors for improving the partitioning of HFRs into the particulate phases [6,28]. As a result, because values of these meteorological parameters can vary greatly (especially in China) under different sampling conditions, the specific values of these parameters should be considered and noted fully in relevant research in the future.

3.2.2. Cause for significant variation of HFRs under different haze conditions

The remarkable variations of HFR concentrations between light and heavy pollution conditions might be explained by the following reasons. First, local HFR emissions are continual and HFRs tend to accumulate in the ambient atmosphere without diffusion under stationary synoptic conditions during a haze episode. Various household decoration materials and electronic equipment are well known as the major HFR emission sources in urban air. Studies have reported substantial concentrations of HFRs in vehicle air and dust, thus, vehicles may also constitute a significant HFR emission source in outdoor conditions [40–42].

Second, the height of the ABL plays a crucial role in controlling atmospheric exchange between the free atmosphere and surface atmosphere, which can affect the mixing and dispersion of atmospheric contaminants [43]. A previous study reported that a distinct decline of the height of the ABL was found at the beginning of a haze pollution event, and the lower ABL height was unfavourable for vertical dispersion of atmospheric contaminants, thereby maintaining the contaminants in a compressed atmospheric space during the haze episode [19]. The diffusion space for atmospheric pollutants, including HFRs, under heavy pollution conditions would be much smaller than that under light pollution conditions [18]. Furthermore, the relatively low wind speed during haze pollution periods causes unfavourable diffusion of atmospheric particles and HFRs [44]. Therefore, during heavy haze episodes, HFRs accumulate in the compressed atmospheric space without diffusion.

Third, stationary synoptic conditions have distinctive features during heavy haze processes. The highly increased number of fine particles during haze episodes contributes to increasing the surface area of the particles, ultimately improving the capacity of particulate matter to adsorb HFRs. Additionally, with relatively high humidity during a

haze episode, the process of hygroscopic growth of the particle nucleus intensely promotes the partition of gaseous HFRs into particulate matter and ultimately aggravates HFR pollution in the particulate phase [45].

According to the above discussion, we can deduce that continuous HFR emissions, stationary synoptic conditions, low ABL height and elevated atmospheric particle concentrations are crucial factors controlling HFR concentrations in atmospheric particles under heavy pollution conditions.

3.3. Size distribution of HFRs

For the eight batches of samples, the total mean concentrations and percentages of individual HFRs in each size fraction are exhibited in Fig. S6-S7 and Table S8. Overall, both PBDEs and NBFRs were enriched in the 2.1–3.3 μm fraction and accounted for 20.0% and 15.7% of the total HFRs in all fractions, respectively. DPs were enriched in the 0.7–1.1 μm fraction and accounted for 19.1% of the total HFRs. BDE-209 and DBDPE were the two major individual HFR components in all fractions (Table S9).

The total mean normalized size distributions of PBDEs, NBFRs, DPs and individual HFRs are presented in Fig. S8. In summary, the results showed that most individual PBDEs and DPs congeners were characterized by trimodal distributions in the 0.7–1.1, 2.1–3.3 and 9.0–10 μm fractions, which is similar to the findings of previous studies on PBDEs [14,46]. However, NBFRs were characterized by a bimodal distribution in the 4.7–5.8 and 9.0–10 μm fractions.

The normalized size distributions of PBDEs, NBFRs and DPs during light and heavy pollution conditions are presented in Fig. 4, and the normalized size distributions of individual HFR congeners are shown in Fig. S9. Overall, the distribution patterns of PBDEs NBFRs, and DPs all presented significant differences between light and heavy pollution conditions (Fig. 4). Specifically, NBFRs were characterized by a bimodal distribution in the 4.7–5.8 and 9.0–10 μm fractions during heavy pollution conditions, whereas PBDEs and DPs were characterized by a trimodal distribution in the 0.7–1.1, 2.1–3.3 and 9.0–10 μm fractions during heavy pollution conditions, which is similar to the findings of previous studies [14,46]. DPs were characterized by a bimodal distribution in the 0.7–1.1 and 2.1–3.3 μm fractions during light pollution conditions. However, neither PBDEs or NBFRs presented significant variations among the different size fractions during light pollution
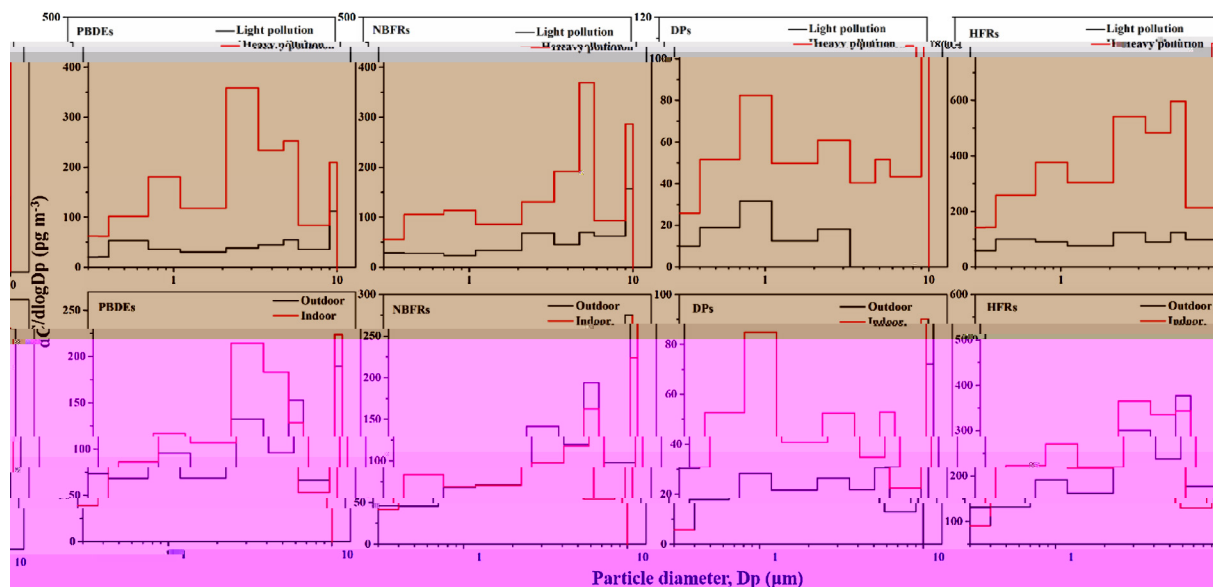

Fig. 4. Mean normalized size distributions of PBDEs, NBFRs and DPs, where dC is the mass concentration on each filter, and dlogDp is the logarithmic aerodynamic diameter (Dp) size interval for each impactor stage.

conditions.

The percentages of HFRs in fine ($< 2.1\,\mu$m), coarse (2.1–$5.8\,\mu$m) and large (5.8–$10\,\mu$m) particles in indoor and outdoor environments and the influence of haze levels are provided in Fig. S10 and Table S10-S11. Overall, the HFR percentage in fine particles in the indoor environment was remarkably higher than that in the outdoor environment during light pollution episodes. Conversely, the HFR percentage in large particles from the outdoor environment was remarkably higher than that in the indoor environment during light pollution conditions. However, the above variations were not obvious during heavy pollution conditions. A previous study demonstrated the important contribution to atmospheric particles of brake dust and tire dust with aerodynamic diameters of 0.9–$11.5\,\mu$m [47], and the resuspension of road dust induced by traffic activities or wind has been recognized as a potential source for atmospheric particle-bound HFRs [42,48].5.12tial
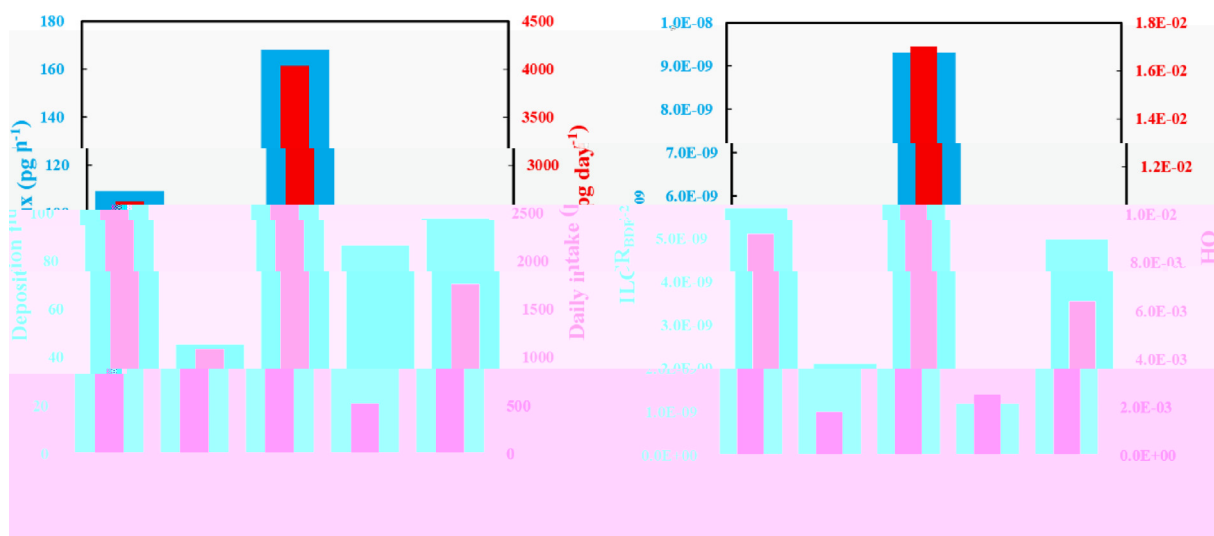
Fig. 6. Deposition fluxes, daily intake, hazard quotient (HQ) and ILCR$_{BDE-209}$ values of HFRs under different conditions. M is the total mean value, L is light pollution conditions, H is heavy pollution conditions, O is outdoor conditions, and I is indoor conditions.

correct the exposure assessment results of contaminants penetrating into human lungs [57,58]. According to our results, most of the inhaled HFRs (81.5%) deposited in the HA while only a small portion (18.5%) deposited in the TB and AR (Table S13). If only the portion deposited into the AR is directly hazardous to human health, then the bioaccessible fraction constituted only 7.4–10.0% of the total inhaled HFRs in the present study (Table S14), which is much lower than the empirical value of 75%. Hence, the human exposure assessments performed based on the total contaminant concentrations by previous studies probably have considerable bias. Moreover, the ratio of the deposition fraction/ inhalable fraction of inhaled HFRs between heavy conditions and light conditions is 0.8–2.3 (for different compounds), whereas that between outdoor conditions and indoor conditions is 1.0–3.6 (for different compounds) (Table S15), indicating that much higher proportions of particle-bound contaminants can deposit into the human respiratory tract on serious haze days and in traffic environments. To the best of our knowledge, this study is the first to report variations in the deposition of compounds in the human respiratory tract under different air quality conditions.

3.5. Health risks and the importance of air quality in assessments of human exposure to HFRs

Noncancer risk (hazard quotient, HQ) and incremental lifetime cancer risk (ILCR) were evaluated based on the estimation of human inhalation exposure to HFRs. Detailed information on the HQ and ILCR from the ICRP model is provided in the Supplementary material.

We evaluated only the cancer risk induced by BDE-209 because BDE-209 is the only target with congener carcinogenic potency reported by previous studies [31,59]. For general adults, the HQ and ILCR$_{BDE-209}$ values of HFRs were 1.8×10^{-3} and 2.1×10^{-9} in light pollution conditions and 1.7×10^{-2} and 9.3×10^{-9} in heavy pollution conditions, respectively (Fig. 6b and Table S16). Even under heavy exercise conditions (assuming a high breathing rate of $3\,m^3\,h^{-1}$) [52], the HQ and ILCR$_{BDE-209}$ values of HFRs were still much lower than 1 and within the safe acceptable range (1.0×10^{-6}-1.0×10^{-4}) established by the USEPA [60]. Thereby, particle-bound HFRs posed a relatively low noncancer and cancer risk in the present study. Nevertheless, the ratio of heavy pollution/light pollution was 3.6 for the HFR concentration level, 3.7 for the DI$_{total}$, 9.4 for the HQ and 4.4 for the ILCR$_{BDE-209}$, indicating that haze episodes can amplify the atmospheric HFR level, human inhalation exposure and eventual human health risk.

The above assessment indicated that the noncancer risk and

incremental lifetime cancer risk in indoor conditions (6.4×10^{-3} and 5.0×10^{-9}) was not significantly higher than that in outdoor conditions (2.5×10^{-3} and 1.2×10^{-9}), whereas the risks in heavy pollution conditions (1.7×10^{-2} and 9.3×10^{-9}) were much higher than those in light pollution conditions (1.8×10^{-3} and 2.1×10^{-9}). According to our results, air quality is a crucial factor influencing the particulate HFR level in the atmosphere as well as human inhalation exposure and the corresponding health risks. As discussed above, additional attention should be paid to air quality, and it must be considered and labelled when investigating SVOC contamination characteristics in the atmosphere.

4. Conclusions

Haze plays a distinctive role in controlling HFR occurrence in the atmosphere, and HFRs can accumulate in the atmosphere synchronously with particulate matter during haze episodes. Particle-bound HFR levels under heavy haze pollution conditions were approximately 3 times higher than those under light haze pollution conditions. Apparent differences were found for the particle size distribution of HFRs under light and heavy haze pollution conditions. Human inhalation exposure assessments based on total contaminant concentrations in previous studies likely have considerable bias because not all inhaled HFRs can deposit into the human respiratory tract. The HQ and ILCR$_{BDE-209}$ values in heavy haze pollution conditions were significantly higher than those in light pollution conditions. Because meteorological parameter values can vary significantly (especially in China) spatially and temporally, air quality should be considered and labelled when investigating organic contamination in the atmosphere in future research. If air quality is not considered during particulate matter sampling, the results and conclusions of relevant studies may suffer from prominent contingencies and poor universality.

Conflicts of interest

The authors declare that they have no competing financial interests to disclose.

Appendix A. Supplementary data

Supplementary material related to this article can be found, in the online version, at doi:https://doi.org/10.1016/j.jhazmat.2018.07.109.

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