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Health Risks Associated with Polycyclic Aromatic Hydrocarbons (PAHs) in Dustfall Collected from Universities in Wuhan, China

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Abstract: Atmospheric dustfall samples from the libraries and dormitories of 14 universities in Wuhan, China, were collected and analyzed for 16 priority polycyclic aromatic hydrocarbons (PAHs). The results showed that the levels of Σ16PAHs in the dustfall from libraries varied from 2.45 to 13.4 μg·g^{-1}, with an average value of 5.06 μg·g^{-1}, while that of dormitories ranged from 2.61 to 10.6 μg·g^{-1}, with an average of 5.19 μg·g^{-1}. The compositions of PAHs in library and dormitory dustfall were similar, and they were dominated by high molecular weight PAHs (HMW-PAHs) (including fluoranthene, benzo(b)fluoranthene, and chrysene), which accounted for 79.2% of the total PAHs. Spatially, the higher levels of PAHs contamination were mainly distributed in areas with high population density and heavy traffic, which suggests that anthropogenic activities made an important contribution to the PAHs emissions. The diagnostic ratios and principal component analyses (PCA) indicated that combustion-related anthropogenic sources, such as petroleum, coal, and biomass combustion, as well as vehicular traffic emission, were the main sources of PAHs bound to dustfall. The toxic equivalent concentrations (BaPeq) of dustfall-bound PAHs ranged from 271 to 2510 μg·kg^{-1}. According to the health risk assessment results, dermal contact was the predominant exposure pathway, followed by ingestion and inhalation. The incremental lifetime cancer risks (ILCRs) of exposure to PAHs in dustfall was 1.230×10^{-6}–1.140×10^{-5}, which indicates that there are low potential carcinogenic risks in the universities.

Keywords: dustfall; polycyclic aromatic hydrocarbons (PAHs); library; dormitory; source identification; risk assessment

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), as a large group of persistent organic pollutants (POPs) composed of two or more fused aromatic rings, have attracted a lot of public attention, due to their potentially toxic, mutagenic, and carcinogenic properties [1]. PAHs in environmental media mainly originate from anthropogenic activities, including the incomplete combustion of fossil fuels, vehicle emissions, and industrial manufacturing [2–4], as well as natural sources, such as volcanic eruptions, forests, and bushfires [5]. PAHs are widely distributed in the environment (including air, soil, sediments, and water) [6], and the deposition on soil and surface waters may lead to bioaccumulations within the food chain, where their presence is ubiquitous [7].

The released PAHs enter the atmospheric environment and can be distributed between gas and particle phases, especially in particulate matter [2]. Atmospheric dustfall refers

to particulate matter that can be delivered to the surface of the earth and water through wet and dry deposition, which is regarded as one of the most complicated and harmful pollutants in atmosphere [8]. Its components and quantity are also potential pollution indicators for urban environments [9,10]. Meantime, dustfall has been regarded as an important carrier of PAHs [7], because of their low vapor pressure and strong affinity for organic pollutants [11]. Epidemiological studies have shown that long-term exposure to PAHs may result in acute or chronic effects on the human body, such as nausea, inflammation, breathing problems, and cardiopulmonary mortality [12,13]. Indoor environments, in which people spend most of their time, are important places for PAHs exposure. PAHs in indoor dust can enter the human body through the inhalation of air (gas phase and particulate phase), ingestion of indoor dust, and dermal contact, which are the three main ways for the chronic toxicity of PAHs [14–16]. Therefore, studying the PAHs in indoor environments is of great significance for human health risk assessment, which has been the scientific research focus for many years.

To date, most studies in the field of PAHs in dustfall have mainly focused on the investigation of outdoor environments [7,17]. However, there has been little discussion about pollution of PAHs in indoor dustfall. Wuhan, as the capital city, engaged in rapid development in urbanization and industrialization during the past decades. The development of various iron and steel smelting and other industrial enterprises have posed a serious threat to the ecological environment of Wuhan. It was reported that regional fine particulate pollution in Wuhan is one of the major air pollutants, raising public awareness [18]. Anyway, as one of the four major centers of science and education in China, Wuhan has 84 regular universities and colleges. It is reported that there are more than 150,000 graduate students and one million undergraduate students in Wuhan [19]. University students spend more than 80% of their time in indoor environments, such as classrooms, library, and dormitories [20]. The existing research articles on PAHs in indoor environments were mainly conducted in European (56%) and Asian (33%) schools, while only 7% and 4% of the works were performed in American and African schools [21]. Only one study has reported PAHs levels in indoor dust on college campuses, showing that the average concentration of Σ16PAHs in indoor dust was 2060 ± 1290 ng g^{-1} dry weight [20]. Currently, the existing research on atmospheric dustfall-bound PAHs in colleges or universities is limited, and the exposure risk of university students to PAHs in dustfall remains unknown.

In the present study, we collected atmospheric dustfall from 14 universities in Wuhan to (1) determine and compare the pollution levels of 16 target PAHs in library and dormitory; (2) explore the composition and distribution characteristics of PAHs in different functional areas in university environment; (3) identify the potential sources of PAHs in dustfall; (4) evaluate the potential cancer risks, using incremental lifetime cancer risk (ILCR) model. This study will provide important data on PAHs pollution in dustfall in different functional areas of colleges and universities, which is of great importance for establishing effective risk prevention mechanisms and risk assessments of human PAHs exposure.

2. Materials and Methods

2.1. Study Area and Sampling

Wuhan is located in the east of the Jianghan Plain, at 113°41′–115°05′ E and 29°58′–31°22′ N. It has a typical north subtropical monsoon-influenced climate, characterized by abundant rainfall, sufficient sunshine, and four distinct seasons, with hot summers and cold winters. Generally, the average annual temperature is 15.8–17.5 °C. Wuhan has a highly vibrant economy, with a gross domestic product (GDP) of 1622.3 billion yuan in 2019 (National Bureau of Statistics). As one of the key higher education hubs, Wuhan has 98 colleges and/or universities, according to the latest statistics from the Ministry of Education, and hosts a huge population of university students, amounting to 1.3 million [22]. With the continuous development of education, there is a certain amount of atmospheric dustfall pollution in campuses, due to dense population, frequent motor vehicles, and complicated human activities.

In the two functional areas (library and dormitory), a total of 36 atmospheric dustfall samples were collected. Locations of the sampling sites for the study are shown in Figure 1. Dustfall was sampled in buildings at least 1.2 m above the ground with small brushes and dustpans. To make the collected samples representative and persuasive, we took samples under stable weather conditions and avoided collecting large particles of gravel, leaves, and other obvious debris. In addition, when collecting samples, we avoided obvious pollution sources, such as trash cans. The collected samples were packaged in sealed bags, later air-dried, and stored at 4 °C until laboratory analysis. After collecting samples at each site, the latitude and longitude, wind speed, weather conditions, wind direction, and humidity were recorded in detail. The corresponding situation of each sampling point and number are shown in Table S1.

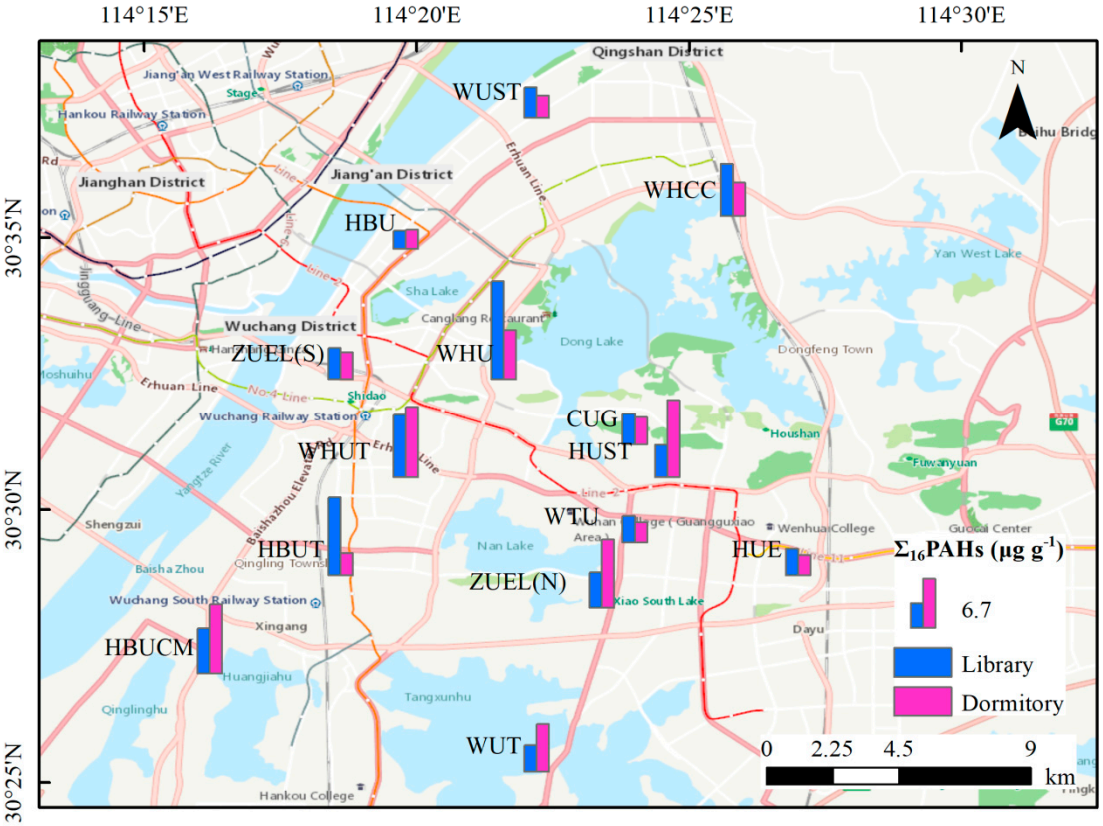

Figure 1. Sampling sites for the atmospheric dustfall samples in Wuhan, China.

2.2. Pretreatment and Chemical Analysis

After sieving, 5 g of dry sample was weighed and spiked with an appropriate amount of anhydrous sodium sulfate and mixed evenly with the sample. Each dustfall sample was spiked with mixed surrogate standards (SS), including naphthalene-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, and perylene-d12. Then, they were extracted at 45 °C for 24 h with dichloromethane (DCM) using a Soxhlet extractor. The extracted solution was evaporated and concentrated to 4 mL, with a rotary evaporator at 35 °C and 60 r min^{-1}. The concentrated extracts were purified with silica gel-alumina (volume ratio 2:1, silica gel deactivated at 180 °C, alumina activated at 240 °C for 24 h, then added 3% ultrapure water to deactivate) chromatographic column. The column was sequentially eluted with 40 mL mixture of DCM and n-hexane (V:V = 2:3) to give fractions enriched in aliphatic hydrocarbons and PAHs at flow rate of ~2 mL min^{-1}. The eluent was concentrated to ~0.5 mL and transferred into a 2 mL cell bottle. Prior to gas chromatograph-mass spectrometry (GC-MS) analysis, hexamethylbenzene (HMB) was added into each vial as internal standard for PAHs quantification purposes.

The 16 target PAHs included naphthalene (Nap), acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fla), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno (1,2,3-cd)pyrene (IcdP), dibenzo(a,h)anthracene (DaA), and benzo(g,h,i)perylene (BghiP). They were determined by GC-MS (7890N-5975, Agilent Technologies, USA). An aliquot of each sample was injected onto a DB-5 capillary column (30 m × 0.25 mm ID × 0.25 μm film thickness). Electron impact (EI) ionization at 70 eV was used with a mass spectrometer operating in the selected ion monitoring (SIM) mode. Injector temperature was 280 °C using the splitless model. The oven temperature started at 80 °C, kept for 2 min, increased to 290 °C at a rate of 4 °C min^{-1}, and kept for 25 min. The carrier gas was helium, and the flow rate was 1 mL min^{-1}.

2.3. Quality Assurance and Quality Control (QA/QC)

The concentrations of PAHs in dustfall samples were quantitatively analyzed by internal standard method. Each batch of samples in the experiment was performed with parallel samples, solvent blanks, and method blanks to monitor the experimental background concentration and extraction efficiency. A mid-point calibration standard was injected after every 10 samples to monitor the instrumental stability. After every batch of 10 samples, n-hexane was injected as a check for the target analyte residuals between the samples. The limits of detection (LODs) and the limits of quantification (LOQs) were evaluated based on the lowest concentration in standards resulting in a signal-to-noise ratio (S/N) $\geq$ 3 and S/N ratio $\geq$ 10, with values ranging from 0.009 to 0.468 and 0.028 to 1.56. The mean matrix spike recoveries of 16 PAHs were 64 ± 9%, 69 ± 11%, 75 ± 10%, 102 ± 5%, and 98 ± 5% for naphthalence-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, and perylene-d12, respectively.

2.4. Source Identification

Diagnostic ratios have been most widely used to distinguish the emission sources of PAHs, due to its simplicity [23,24]. Some scholars have performed similar research before, and the results showed that the concentration of PAHs with same molecular weight and similar structure can be used to determine the source of the pollutants [25–27]. A combination of Ant/(Ant+Phe), Fla/(Pyr+Fla), BaA/(BaA+Chr), and IcdP/(IcdP+BghiP) was applied to identify the potential sources. Principal component analysis (PCA) was used to transform large data sets into less meaningful dimensions, based on the bilinear decomposition, and the purpose of PCA was to obtain a set of linearly uncorrelated components, which explain the total variance in the original data. Anyway, the PCA were conducted on the basis of concentration with varimax rotation, using SPSS 24.0 software for further exploration of the possible source of PAHs in dustfall from different sites [14,28].

2.5. Health Risk Assessment of PAHs in Dustfall

Among these PAHs, BaP can be used as an indicator of carcinogenic risk of exposure to PAHs [29]. In this paper, according to the BaP toxic equivalency factor (TEF) introduced by Nisbet and Lagoy [30], PAHs monomers were conserved into benzo(a)pyrene equivalent (BaPeq) concentration to calculate their exposure level. TEF value of BaP was defined as 1, and the values of other PAHs were based on their carcinogenic levels, compared with BaP, and were shown in Table 1.

The total toxic equivalent can be expressed as:

$$\text{TEQs} = \sum(C_i \times \text{TEF}_i) \quad (1)$$

where C_i and TEF_i are the concentration and TEF of *i* compound, respectively [31].

Health risk assessment is an evaluation method that was developed in the 1980s. It can be used to assess the impact on human health by estimating the probability of harmful factors affecting human and environmental health adversely [12]. The incremental lifetime cancer risk (ILCR) model has been widely used for assessing health risks when humans are exposed to environmental pollutants. This study used the ILCR model to

calculate the carcinogenic risk of exposure to PAHs in the libraries and dormitories of universities in Wuhan. The lifetime carcinogenic risk value of PAHs in atmospheric dustfall through ingestion, dermal contact, and inhalation can be estimated using the following formulas [14].

$$ILCR_{ing} = \frac{CS \times CSF_{ing} \times \sqrt[3]{\frac{BW}{70}} \times IR_{ing} \times EF \times ED}{BW \times AT \times 10^6} \quad (2)$$

$$ILCR_{derm} = \frac{CS \times CSF_{derm} \times \sqrt[3]{\frac{BW}{70}} \times SA \times AF \times ABS \times EF \times ED}{BW \times AT \times 10^6} \quad (3)$$

$$ILCR_{inh} = \frac{CS \times CSF_{inh} \times \sqrt[3]{\frac{BW}{70}} \times IR_{inh} \times EF \times ED}{BW \times AT \times PEF} \quad (4)$$

$$ILCRs = ILCR_{ing} + ILCR_{derm} + ILCR_{inh} \quad (5)$$

$ILCR_{ing}$, $ILCR_{derm}$, and $ILCR_{inh}$ are the values of cancer risks derived from PAHs via ingestion, dermal contact, and inhalation, respectively; CS is the total BaP-equivalent concentration (mg kg^{-1}); CSF_{ing}, CSF_{inh}, and CSF_{derm} are the carcinogenic slope factor through the three pathways ((kg d^{-1} $mg^{-1})^{-1}$); EF is the exposure frequency (d y^{-1}); ED represents exposure duration (d); BW is the body weight (kg); AT is the average life span (d); PEF represents particle emission factor (m^3 kg^{-1}); AF is dermal adherence factor (mg $(cm^2)^{-1}$); SA is exposed skin area (cm^2 d^{-1}); ABS is dermal absorption fraction; ILCRs represents the sum of the risks of the above three exposure routes. Related values are shown in Table S2 [16,28,32].

Table 1. PAHs concentrations (μg·g^{-1}) in atmospheric dustfall from the library and dormitory of Wuhan universities.

Target Compounds	Abbr	Rings	TEF [a]	Library				Dormitory			
				Min	Max	Median	Mean	Min	Max	Median	Mean
naphthalene	NaP	2	0.001	0.09	0.46	0.16	0.19	0.05	0.38	0.12	0.17
acenaphthylene	Acy	3	0.001	0.02	0.14	0.04	0.06	0.02	0.09	0.05	0.05
acenaphthene	Ace	3	0.001	0.01	0.11	0.02	0.03	0.009	0.05	0.02	0.02
fluorene	Flu	3	0.001	0.03	0.29	0.06	0.08	0.02	0.12	0.05	0.06
phenanthrene	Phe	3	0.001	0.36	1.36	0.55	0.63	0.36	1.58	0.53	0.68
anthracene	Ant	3	0.01	0.02	0.10	0.04	0.04	0.02	0.10	0.04	0.05
fluoranthene	Fla	4	0.001	0.34	1.94	0.57	0.70	0.35	1.64	0.55	0.72
pyrene	Pyr	4	0.001	0.21	1.10	0.39	0.46	0.22	1.16	0.40	0.48
benzo[a]anthracene	BaA	4	0.1	0.05	0.41	0.13	0.15	0.06	0.38	0.12	0.14
chrysene	Chr	4	0.01	0.27	1.56	0.40	0.55	0.26	1.32	0.42	0.54
benzo[b]fluoranthene	BbF	5	0.1	0.03	1.91	0.47	0.61	0.003	1.33	0.46	0.58
benzo[k]fluoranthene	BkF	5	0.1	0.03	1.16	0.16	0.30	0.10	1.35	0.20	0.36
benzo[a]pyrene	BaP	5	1	0.003	0.93	0.26	0.31	0.15	0.75	0.30	0.35
dibenzo[a,h]anthracene	DaA	5	1	0.16	1.25	0.26	0.39	0.007	1.54	0.27	0.38
indeno[1,2,3-cd]pyrene	IcdP	6	0.1	0.01	0.67	0.05	0.11	0.009	1.37	0.06	0.14
benzo[g,h,i]perylene	BghiP	6	0.01	0.18	1.39	0.31	0.46	0.17	1.14	0.33	0.45
sumof 16 PAHs	Σ16PAHs [b]		—	2.45	13.40	4.16	5.06	2.61	10.57	3.75	5.19
7 carcinogenic PAHs	Σ7CPAHs [c]		—	1.07	6.84	1.87	2.41	1.10	5.76	1.94	2.50

Abbreviations: [a] toxic equivalency factor. [b] The total concentration of 16 PAHs. [c] The total concentration of 7 carcinogenic PAHs.

3. Results and Discussion

3.1. Concentration of PAHs in Dustfall

Descriptive statistics of the observed PAHs concentrations in dustfall from libraries and dormitories are summarized in Table 1. There was no difference in the concentration of Σ16PAHs in atmospheric dustfall between the two functional areas. The levels of Σ16PAHs in the library areas was 2.45–13.4 μg·g^{-1}, with an average value of 5.06 μg·g^{-1};

the levels of Σ16PAHs in the dormitory varied from 2.61 to 10.6 μg·g^{-1}, and the average was 5.19 μg·g^{-1}. In libraries and dormitories, concentrations of Fla were in the range of 0.350–1.94 μg·g^{-1} and 0.350–1.64 μg·g^{-1}, which appeared to be the most abundant PAH individuals in dustfall. The order of the average value of Σ16PAHs was dormitory > library, which may be attributed to the different function and ventilation conditions. In addition, some research has indicated that the living convention, such as smoking in the bedroom and mothballs used as clothes antiseptics, resulted in the highest PAHs concentration in the dormitory [33].

The average concentrations of predominant individual PAHs decreased in the following order: Fla > Phe > BbF > Chr > Pyr > DaA. The dominant PAH individuals in this study were consistent with previous studies, which pointed out that Phe, Fla, BbF, and Chr accounted for approximately 50–70% of the total PAHs in atmospheric particulate matter [17,34,35]. The concentration of seven carcinogenic PAHs (Σ7CPAHs, including BaA, Chr, BbF, BkF, BaP, DaA, and IcdP) ranged from 1.07 to 6.84 μg·g^{-1} and from 1.10 to 5.76 μg·g^{-1} for libraries and dormitories, respectively. In this investigation, the concentrations of BaP in the dustfall of libraries and dormitories were between 0.003 and 0.930 μg·g^{-1}, with a mean of 0.330 μg·g^{-1}, which was lower than that in Tianjin [36], where the mean dustfall concentration of BaP varied from 0.110 to 5.66 μg·g^{-1} (mean 0.880 μg·g^{-1}). In contrast to Xian, an industrial city of China, the mean level of BaP was measured to be 0.52 μg·g^{-1}, and the range was 0.080 to 1.32 μg·g^{-1} [37]. Chen and Zhu [38] found that the PAH levels in atmospheric dustfall in Hangzhou were between 0.080 and 0.780 μg·g^{-1}, with an average value of 0.300 μg·g^{-1}. According to these comparison results, the BaP levels in the atmospheric dustfall of Wuhan colleges were at a moderate level.

The concentration of PAHs in this study was lower than those in Henan (6.00–79.0 μg·g^{-1}) [39], Xi'an (10.2–63.9 μg·g^{-1}, mean 30.1 μg·g^{-1}) [37], and Macao (2.26–22.9 μg·g^{-1}, mean 10.6 μg·g^{-1}) [40]. In addition, the PAHs in this study were compared to street/road-bound PAHs in other studies [17,41–47] (Table S3). Overall, the total concentration of PAHs in the dustfall of Wuhan universities were at a relative high level, which may be related to the seasons. Sample collections were conducted in winter, with low temperature, when energy consumption for residential heating increased [48,49]. In addition, the difference in PAHs concentration levels between cities may be attributed to the city's industrial structure, industrial layout, geographical environment, and meteorological conditions.

3.2. Composition of PAHs in Dustfall

The composition characteristics of PAHs in the two functional areas of 14 universities are shown in Figure 2. The 16 priority PAHs can be divided into low molecular weight (two–three rings, LMW) PAHs and high molecular weight (four–six rings, HMW) PAHs [50]. In the library, four-ring PAHs contributed the most and accounted for 38.5% of total PAHs, followed by five-ring (25.0%), three-ring (16.6%), six-ring (15.9%), and two-ring (3.91%). A similar contribution order was observed in the dormitory, with HMW-PAHs accounting for 79.2% of Σ16PAHs, while the LMW-PAHs accounted for only about 20.8%. Fla, Phe, and BbF were the three most abundant compounds in two functional areas, contributing to 38.4% of the total PAHs. Based on previous studies, Phe could be indicative of coke oven, coal combustion, and diesel engine origins [51]. BbF is a characteristic component of fossil fuel combustion [52]. Although the dormitory and library belong to two different types of areas, the proportion of 16 PAHs in the dustfall was similar, indicating that PAHs in the two functional areas may come from the same sources. A similar observation was reported by [17] for HMW PAHs, which had greater contributions to the total PAH concentrations than the LMW PAHs in dustfall. A possible explanation for these results is the physicochemical properties of PAHs. LMW PAHs have a higher vapor pressure and stronger volatility, so they mainly exist in the gas phase form and can be transported further, while HMW PAHs are more stable and less volatile, which can typically be associated with airborne particles, so with 90% of them existing in the form of particles [2,53]. Anyway,

much more attention should be paid to particulate PAHs, for the fact that HMW PAHs in the particulate phase were more carcinogenic than those of low molecular weights [33].

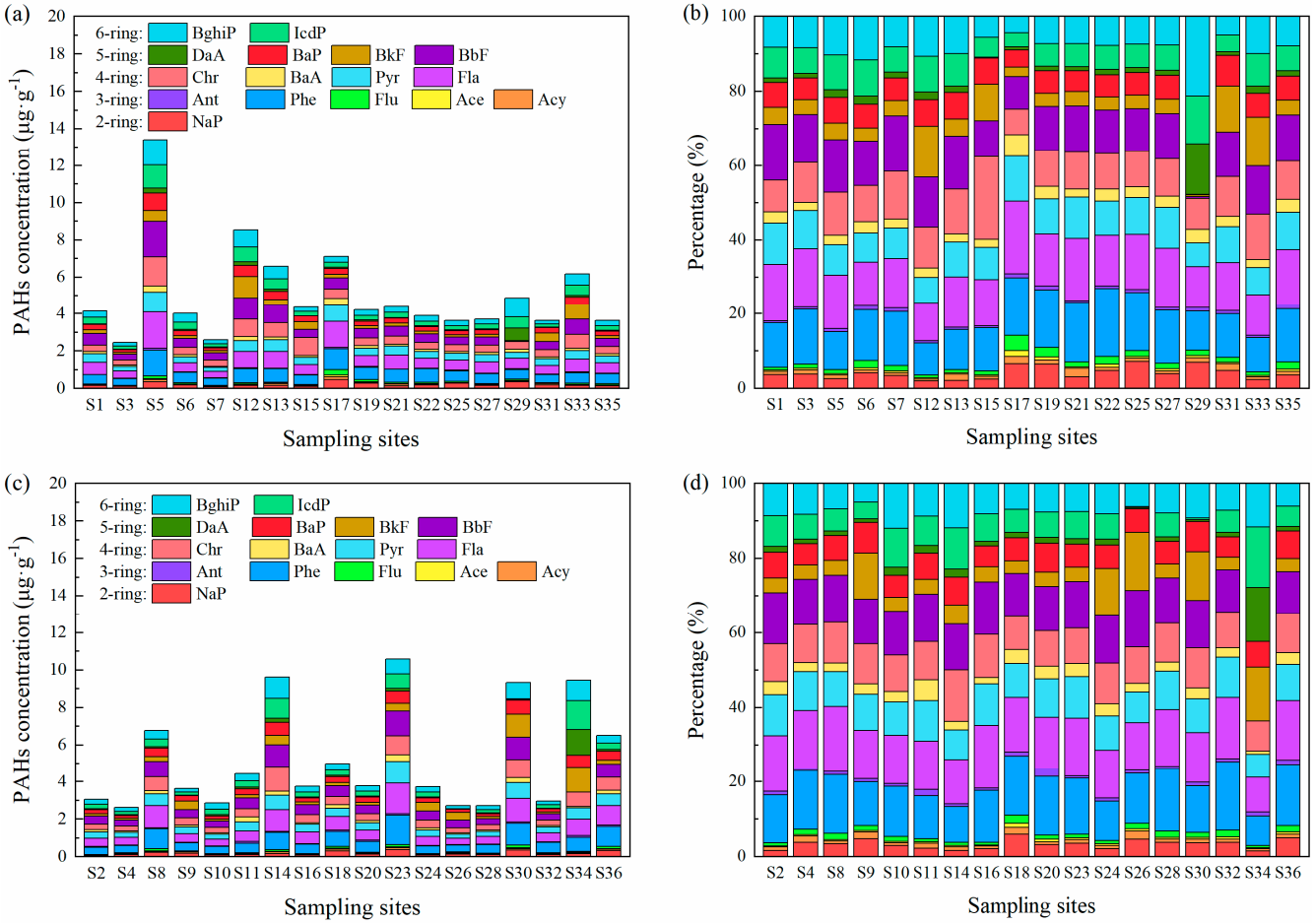

Figure 2. Concentration and percentage of individual PAH for each site: (**a**) concentration of PAH in libraries; (**b**) percentage of individual PAH in libraries; (**c**) concen tration of PAH in dormitories; (**d**) percentage of individual PAH in dormitories.

3.3. Spatial Distribution of PAHs

The spatial distribution of Σ16PAHs concentrations in dustfall from the libraries and dormitories are depicted in Figure 1. As shown in Figure 1, the concentrations of Σ16PAHs in the present study varied greatly, depending on the sampling location, and ranged from 2.45 to 13.4 μg·g^{-1}. In general, the content of PAHs gradually increased from the southwest to northeast, and the sampling sites located in the second ring road and third ring road, with frequent anthropogenic activities and heavy traffic, have higher PAHs content. It is well-known that vehicles and construction equipment emit large amounts of PAHs [2]. For the libraries, the highest concentrations of Σ16PAHs were observed at site WHU, followed by sites HBUT and WHUT, with average values of 13.4 μg·g^{-1}, 10.6 μg·g^{-1}, and 8.52 μg·g^{-1}, respectively. Among these, site WHU was located in the Chuhe Han Street commercial area, near East Lake tourist attractions; sites HBUT and WHUT were located on a main traffic road with long-term ground construction. The reason for this may be frequent and complex human activities and transportation, which were reported to be positively correlated with PAH concentrations [54]. Gunawardena [55] and Suman [56] had reported that motor vehicle emissions were the main source of PAHs, and heavy traffic loads could increase PAH emissions and accelerate their migration and transformation. In addition, there were many instances of heavy industrial machinery and equipment, which may be another reason for the high PAH contents of these sites. For dormitories,

the average Σ16PAHs concentrations observed at the 14 sites ranked as HUST > WHUT > HBUCM > ZUEL(N) > WHU > WUT > WHCC > CUG > ZUEL(S) > WUST > HBUT > WTU > HUE > HBU, different from the distribution of PAHs in library dustfall. The highest 16 PAHs concentration at the HUST site (10.6 $\mu g \cdot g^{-1}$) was 4.0 times higher than the lowest Σ16PAHs concentration at the HBU site (2.61 $\mu g \cdot g^{-1}$). The HUST site was located on the main road of the campus' transportation, with frequent vehicle transportation. By contrast, the HBU site was located in the living area, far away from the commercial area. This observation suggested that sample location appeared to be the important factor affecting dustfall PAHs concentration. Qi [40] demonstrated that the PAHs concentrations varied to a great extent at different sampling points and may be influenced by land use differences, and the sample location with frequent human activities would result in more PAHs emissions [57]. Sofia Augusto used the spatial modeling of PAHs to investigate the relations between the PAHs profile and different land uses, and the results showed that the distribution of PAHs in different land use areas (urban, industrial, and forest) was different. For example, four-ring PAHs were the most common PAHs in urban areas, while in industrial areas, five-ring and six-ring PAHs were the most common PAHs [58]. Environmental factors, such as temperatures and winds, are important in PAHs deposition and distribution [59], and the spatial distribution of PAHs is affected by many factors, which still need to be further explored.

3.4. Source Identification

The ratio of Ant/(Ant + Phe) < 0.1 indicates petrogenic sources (such as associated with oil spills and application of fuels in vehicles), and over 0.1 characterizes the combustion sources [26]. A value of Fla/(Fla + Pyr) less than 0.4 is assigned as petroleum combustion, from 0.4 to 0.5 is assigned as fuel oil source, and above 0.5 is assigned as biomass and coal burning. A ratio of BaA/(BaA+Chr) varying between 0.2 and 0.35 is identified as coal combustion, less than 0.2 is identified as mean oil pollution, and more than 0.35 is identified as vehicular emissions. The IcdP/(IcdP + BghiP) value < 0.2 is assigned as petrogenic source, from 0.2 to 0.5 is assigned as fossil fuel combustion, and above 0.5 is assigned as grass, coal, and wood combustion [26,60]. As shown in the cross-plot of Figure 3a, most samples had Ant/(Ant + Phe) ratios between 0.02 and 0.1 (except that there were three sampling sites located in a dormitory within the range of 0.1–0.14), indicating petrogenic sources, such as oil spills and the application of fuels in vehicles, in most of the sampling sites. Fla/(Fla + Pyr) ratios higher than 0.5 in this study implied that PAHs mainly came from coal and wood combustion. The ratio of BaA/(BaA + Chr) was recommended to distinguish motor vehicle emissions from wood burning emissions [27]. In Figure 3b, the ratio of BaA/(BaA + Chr) in the two functional areas is mostly less than 0.35, and IcdP/(IcdP + BghiP) almost falls within the range of 0.2–0.5, indicating that PAHs mainly came from mixed sources of fossil fuel, coal combustion, and vehicle emissions. According to the distribution of various points in the comprehensive map, we can obtain that the PAHs in the dustfall in libraries and dormitories are likely to have the same sources. It is speculated that the PAHs in the dust mainly come from the incomplete combustion of motor vehicle fuel, and the combustion products are discharged into the atmosphere and precipitated under the influence of natural factors, such as wind.

PCA was conducted on all samples, and the results are shown in Table 2. Three factors were extracted for the PAHs in the dustfall from Wuhan university campus, when eigenvalues were larger than 1 and the cumulative variance contribution rate reached 87.2%. The first principal component (PC1) explained 35.9% of the total variance, with high loading of 2–3 ring PAHs, such as Nap, Ace, and Flu, and moderate loading of Acy, Phe, and BaA. Nap, Acy, Ace, and Flu were considered to be from a coke oven [61,62]. PC2 contributed 29.6% to the total variance, with high loading of Chr, BbF, and BaP and moderated loading of Fla and Pyr. Chr and BaP were typical markers of coal combustion [63]. Fla and Pyr were often related to wood combustion [64]. In addition, a high emission ratio of BbF was also found in residential coal combustion [65]. Furthermore, it is hard to distinguish

coal and biomass combustion. Thus, PC2 was considered to be a source of coal and biomass combustion. PC3 explained 21.7% of the total variance, with high loading of DaA, IcdP, and BghiP and moderate loading of BkF. DaA and BghiP were widely used as markers of petroleum combustion [5]. IcdP was often related to diesel and gasoline engine emissions [66]. Additionally, BkF was a tracer of diesel exhaust [67]. Therefore, PC3 represented traffic-related source. Overall, the PCA and diagnostic ratios analysis showed that the source of PAHs in atmospheric dust was not controlled by a single factor, and the main sources were petroleum, coal and biomass combustion, and traffic-related sources.

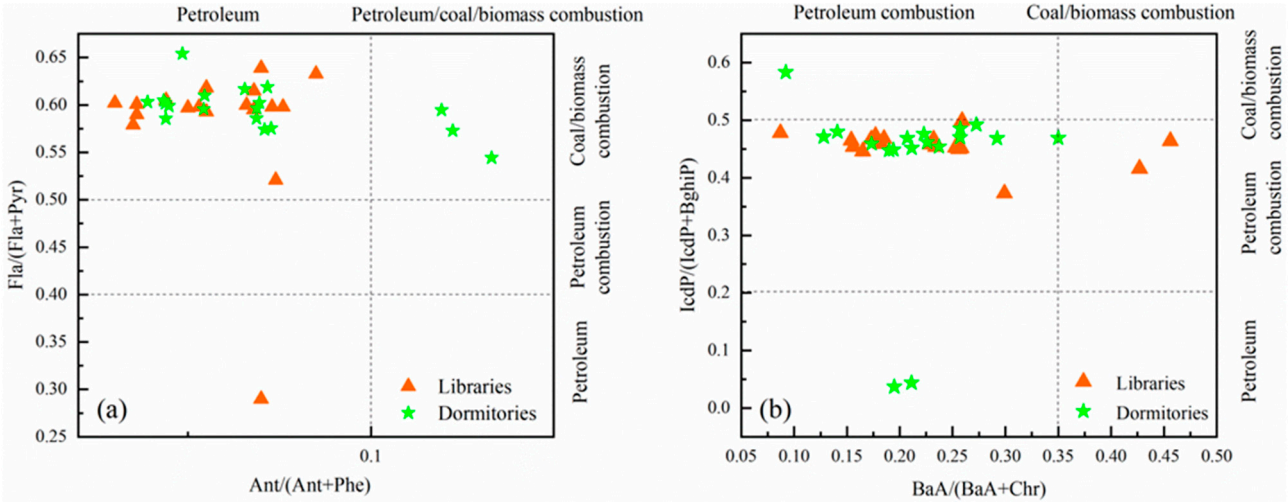

Figure 3. Scatter plot of diagnostic ratios: (**a**) Ant/(Ant+Phe) vs. Fla/(Fla+Pyr); (**b**) BaA/(BaA+Chr) vs. IcdP/(IcdP+BghiP).

Table 2. Source identification of PAHs in dustfall by PCA.

	PC1	PC2	PC3
NaP	**0.909**	0.144	0.113
Acy	**0.694**	0.413	0.145
Ace	**0.95**	0.122	0.015
Flu	**0.948**	0.129	0.03
Phe	**0.703**	0.596	0.204
Ant	0.583	0.348	0.557
Fla	0.652	**0.676**	0.263
Pyr	0.627	**0.694**	0.267
BaA	**0.811**	0.466	0.095
Chr	0.249	**0.83**	0.403
BbF	0.319	**0.928**	-0.016
BkF	-0.033	0.461	**0.624**
BaP	0.295	**0.81**	0.452
DaA	0.044	-0.173	**0.973**
IcdP	0.118	0.384	**0.837**
BghiP	0.237	0.53	**0.727**
% of variance	35.851	29.630	21.687
cumulative %	35.851	65.481	87.168

* bold: The dominant component with high loading.

3.5. Human Risk Assessments

In the library, the BaPeq concentrations of Σ16PAHs ranged from 363 μg·kg^{-1} to 2510 μg·kg^{-1}, with an average value of 823 μg·kg^{-1}. The total BaP_{eq} of seven carcinogenic PAHs were 820 μg·kg^{-1}, accounting for 99.7% of TEQs, indicating that seven carcinogenic PAHs were major carcinogenic contributors of Σ16PAHs. Among PAHs, DaA had the highest BaP_{eq} concentration of 1246 μg·kg^{-1}, accounting for 47.2% of total BaPeq. BaP,

considered to be the most hazardous among 16 PAHs, accounted for about 37.7% of TEQs. The total BaPeq of 16 PAHs in the dormitories (ranging from 271 to 2484 $\mu g \cdot kg^{-1}$, with an average value of 864 $\mu g \cdot kg^{-1}$) were 1.05 times higher than that in libraries. The mean BaPeq of Σ7CPAHs accounted for 99.7% of the Σ16PAHs, which was consistent with those in libraries. The toxic equivalent concentrations (TEQs) of dustfall in this study were higher than those in road dust of Ulsan, Korea (0.93l–68.8 $\mu g \cdot g^{-1}$) [68], Kabul (75.0 $\mu g \cdot kg^{-1}$), and Jalalabad (36.0 $\mu g \cdot kg^{-1}$) [69]. China has not stipulated the quality standards of PAHs in atmospheric dust; thus, the reference value of the TEQs of 10 PAHs in the Dutch soil management regulations was adopted (32.5 $ng \cdot g^{-1}$), which was calculated by the criterion concentration in soil and their corresponding TEF [70]. TEQs of PAHs in the dust from libraries and dormitories were higher than this value, which indicated the potential health risks.

Considering that there are mainly college students in universities, ILCRs were only performed on adults. The value of ILCRs $\leq 10^{-6}$ indicates a negligible risk, values between 10^{-6} and 10^{-4} indicate a potential health risk, and values > 10^{-4} indicates a high potential health risk [14]. Table 3 shows the ILCR levels of PAHs in the dustfall, calculated from Equations (2)–(5). The risk values for adults from libraries were ranged from 1.65×10^{-6} to 1.14×10^{-5}, with an average of 3.74×10^{-6}. The cancer risks for PAHs for the different routes of exposure decreased in the following order: dermal contact > ingestion > inhalation. ILCRsder was 2.39×10^{-6} and accounted for 64.0% of the total lifetime cancer risk, indicating that dermal contact was the predominant exposure pathway. In the dormitory, the values of ILCRs varied from 1.23×10^{-6} to 1.13×10^{-5} and were relatively higher than the priority level of risk (10^{-6}), which reflected that exposure to the dormitory dustfall PAHs poses a potential cancer risk for college students. Exposure to indoor dust-bound PAHs in Kuwait has a total cancer risk for adults of 2.15×10^{-3} [71]. Eom indicated that these health risks would be underestimated, since the cancer risk value was based on the BaP-equivalent concentration, rather than the actual PAH concentration [72], and suggested that we pay more attention to the health risks and importance of PAH exposure. Dermal contact (2.51×10^{-6}) was the predominant exposure pathway, followed by ingestion (1.414×10^{-6}) and inhalation (1.10×10^{-10}). This observation was consistent with previous studies [28,32], which indicated that PAH exposure through dermal contact presented a much greater health risk than either ingestion or inhalation of the PAHs. To make a conclusion, the students of Wuhan universities were frequently exposed to dustfall PAHs, and ILCR levels fell within the range of 10^{-4}–10^{-6} in libraries and dormitories, suggesting that there were potentially serious carcinogenic risks for exposed populations in the universities.

Table 3. Health risk assessment of PAHs in campus dustfall.

Exposure Pathways		$ILCRs_{ing}$ [a]	$ILCRs_{der}$ [b]	$ILCRs_{inh}$ [c]	ILCRs [d]
Libraries	mean	1.346×10^{-6}	2.391×10^{-6}	1.044×10^{-10}	3.738×10^{-6}
	min	5.937×10^{-7}	1.055×10^{-6}	4.605×10^{-11}	1.648×10^{-6}
	max	4.107×10^{-6}	7.296×10^{-6}	3.185×10^{-10}	1.140×10^{-5}
	median	1.017×10^{-6}	1.806×10^{-6}	7.885×10^{-11}	2.823×10^{-6}
Dormitories	mean	1.414×10^{-6}	2.511×10^{-6}	1.096×10^{-10}	3.925×10^{-6}
	min	4.429×10^{-7}	7.867×10^{-7}	3.435×10^{-11}	1.230×10^{-6}
	max	4.064×10^{-6}	7.220×10^{-6}	3.152×10^{-10}	1.128×10^{-5}
	median	1.016×10^{-6}	1.804×10^{-6}	7.876×10^{-11}	2.820×10^{-6}

Abbreviations: [a] the values of cancer risks derived from PAHs via ingestion. [b] the values of cancer risks derived from PAHs via dermal contact. [c] the values of cancer risks derived from PAHs via inhalation. [d] the sum of the risks of the above three exposure routes.

4. Conclusions

PAHs concentrations were analyzed in dustfall samples collected from the libraries and dormitories of 14 universities in Wuhan. PAHs were widely detected in the dustfall samples, and the average concentration of PAHs in dormitories was higher than in libraries.

The dominant species found in the dustfall were Fla, Phe, and BbF in both areas. The composition characteristics of PAHs in atmospheric dustfall in the two functional areas were similar, and both were dominated by HMW PAHs. However, PAHs content varied greatly at different sampling sites. The pollution levels of PAHs in colleges were higher near the second and third ring roads, with dense populations and heavy traffic, than other sites. Source identification studies indicated that petroleum, coal and biomass combustion, and motor vehicle emissions were the major sources for PAHs in dustfall in university environments. The college students were exposed to dustfall-bound PAHs, mainly via dermal contact and ingestion pathways. The ILCRs fluctuated within the range of 10^{-6}–10^{-4}, indicating that there were potentially serious carcinogenic risks for exposed students in Wuhan universities.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/atmos13101707/s1, Table S1. List of sampling sites; Table S2. Cancer risk assessment parameters; Table S3. Comparison of PAHs concentration in dust from some reported studies.

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