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Characterization and Risk assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in Different Size Segments of Atmospheric Particles in Jinhua, China

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Abstract: Atmospheric particulate matter (PM) of different particle sizes was collected simultaneously from eight locations in Jinhua from December 2020 to January 2021, and the polycyclic aromatic hydrocarbons (PAHs) loaded on the collected PM were analyzed and evaluated. The mass concentrations of PAHs (Σ15PAHs) in the sampled areas ranged from 17.29 to 41.37 ng/m^3, with an average concentration of 29.78 ng/m^3. The distribution of Σ15PAHs concerning the PM_{10} particle size exhibited a three-peak pattern, with the highest concentration in the fine particle size segment. 4-ring PAHs were more likely to accumulate in the fine size segment, whereas 5-6-ring PAHs were more abundant in the coarse size segment, and 3-ring PAHs were more abundant in the medium size segment. The carcinogenic health risk of PAHs in different populations decreased in this order: adults > adolescents > children. Although the values of lifetime excess risk of carcinogenicity of each sampling particle size segment were less than 1×10^{-6}, the proportion of cancer risk in the fine particle segment was significantly higher than that in the medium or coarse particle size segment.

Keywords: Atmospheric particulate matter; Polycyclic aromatic hydrocarbons; Particle size segments; Pollution characteristics; Health

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds composed of two or more benzene rings linked together (Yan et al, 2018; Liu et al, 2018). PAH pollution has recently gained attention due to its typical carcinogenicity, mutagenicity, and teratogenicity (Kampa and Castanas, 2008; Poschl, 2005). To effectively control PAH pollution, many governments and companies have been monitoring the content of PAHs in the atmosphere and conducting detailed studies on their pollution characteristics since 1940 (Famiyeh et al., 2021; Valiere et al., 2022). Qu et al. (Qu et al., 2018) investigated the pollution characteristics of PAHs in Salerno (Italy). They found that the pollution characteristics at different locations varied significantly, with concentrations ranging from 9.58 to 15,818 ng/m^3. In addition, they found that 4-ring PAHs were the most abundant, accounting for 40.6% of the total PAHs, followed by 5-ring PAHs (29.2%), and 6-ring PAHs (17.6%), 3-ring PAHs (11.8%), and 2-ring PAHs (0.73%). Zheng et al. (Zheng et al., 2019) analyzed the characteristics of PAH pollution in Huainan (China) and found that the annual average value of 16 PAHs in PM_{10} was 188 ± 21.8 ng/m^3. Furthermore, they reported that 4-ring PAHs were the most abundant in all seasons, followed by 3-ring and 5-ring PAHs.

PAHs have typical carcinogenicity, teratogenicity, and mutagenicity. They can cause damage to most of the human body systems, such as the heart, respiratory, blood, immune, and endocrine systems (Chen et al,2005; Shiraiwa et al., 2017). Wang et al. (Wang et al., 2017) analyzed the potential carcinogenic health risks of PAHs in the atmosphere of Nanjing (China). They found that the values of lifetime excess risk of carcinogenicity (ILCR) caused by the total amount of PAHs in various populations were greater than 10^{-6}. The carcinogenic risk value decreases in this order: adults > adolescents > children and the elderly. Xia et al. (Xia et al., 2013) collected PAHs in the gas and particulate phases in Taiyuan's urban and rural areas (China). They found that the adult group had the highest carcinogenic risk value, and the carcinogenic risk of the female group was higher than that of the male group.

In this study, atmospheric particulate matter (PM) samples were collected from eight regions in Jinhua to investigate the concentration and composition characteristics of PAHs carried by atmospheric PM with different particle sizes. The United States Environmental Protection Agency (USEPA) model method was used to evaluate the health risks of atmospheric particulates of different particle sizes to different groups of people.

2. Materials and methods

2.1 Sampling

Jinhua is in the central part of Zhejiang Province, adjacent to Taizhou in the east, Lishui in the south, Quzhou in the west, and Shaoxing and Hangzhou in the north. It is an important transportation hub in China and one of the major cities in the YRD region. This study selected eight regions in Jinhua, including Yiwu, Yongkang, Lanxi, Wuyi, Dongyang, Pan'an, Wucheng, and Jindong. The sampling points are within 200 m of the monitoring points of the county-level municipal governments in Jinhua City, and there are no obvious pollution sources or sheltering buildings around. The sampling instrument is erected at approximately 1.5 m to avoid the impact of dust, and the sampling height is 25-30 m. The sampling period was from December 2020 to January 2021, with each sampling area sampled for 5-7 days and 23 h per day. The distribution of sampling points and the basic sample information are shown in Figure 1 and Table 1, respectively.

Table 1 Basic sampling information

Sampled sites	Longitude and latitude of sampling point	Sampling days	Number of samples	Meteorology
Wucheng	N29.13°, E119.64°	7 d (week 1)	N = 63	Sunny, gentle breeze
Jindong	N29.05°, E119.74°	7 d (week 2)	N = 63	partly cloudy, gentle breeze
Yongkang	N28.89°, E120.02°	6 d (week 4)	N = 54	Sunny, gentle breeze
Wuyi	N28.89°, E119.81°	7 d (week 5)	N = 63	partly cloudy, gentle breeze
Lanxi	N29.21°, E119.46°	5 d (week 7)	N = 45	Sunny, gentle breeze
Dongyang	N29.29°, E120.24°	7 d (week 8)	N = 63	Sunny, gentle breeze
Panan	N29.05°, E120.44°	7 d (week 9)	N = 63	Sunny, gentle breeze
Yiwu	N29.38°, E120.04°	6 d (week 10)	N = 54	partly cloudy, gentle breeze

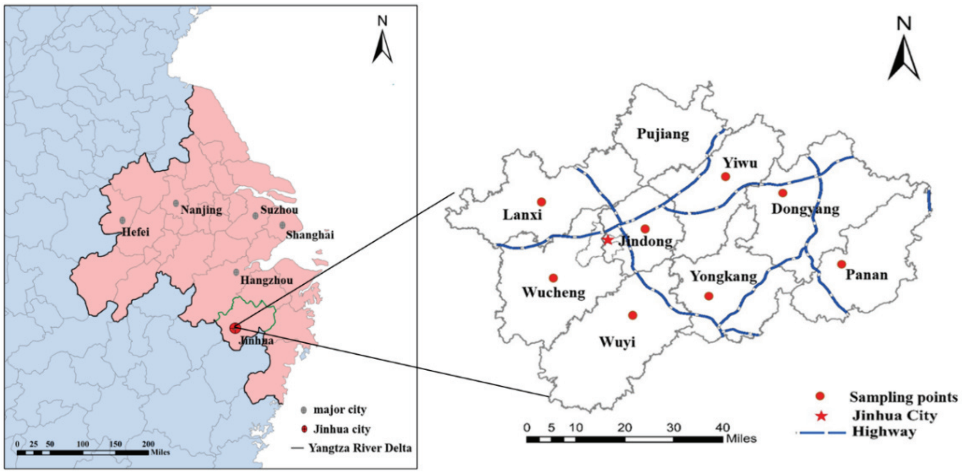

Figure 1 Geographical distribution of the sampling sites

Particulate samples were collected using an Anderson medium flow eight-stage sampler (TE-20-800, 28.3 L/min), where S_0 represents the 0th grade (0-0.4 μm) of the particle sampler, S_1 denotes the 1st grade (0.4-0.7 μm), S_2 the 2nd grade (0.7-1.1 μm), S_3 the 3rd grade (1.1-2.1 μm), S_4 the 4th grade (2.1-3.3 μm), S_5 the 5th grade (3.3-4.7 μm), S_6 the 6th grade (4.7-5.8 μm), S_7 the 7th grade (5.8-9 μm), and S_8 the 8th grade (9-10 μm). In this study, S_0–S_3 (0-2.1 μm) are collectively referred to as the fine size segment, S_4–S_6 (2.1-5.8 μm) the medium size segment, and S_7–S_8 (5.8-10 μm) the coarse size segment. All samples were collected on a glass fiber filter pre-baked in a muffle furnace at 450 °C for 4 h. Following sampling, the filter samples were sealed individually in an aluminum foil bag and stored in a freezer at -18 °C before analysis.

2.2 PAHs analysis

The determination method of PAHs in this study adopts the "Determination of Polycyclic Aromatic Hydrocarbons in Gas Phase and Particulate in Ambient Air and Waste Gas by Gas Chromatography-Mass Spectrometry Method" (HJ 646-2013) issued by the Ministry of Environmental Protection of the People's Republic of China. Sample pretreatment includes four steps: sample extraction, sample concentration, sample purification, and sample concentration and constant volume. This study used an Agilent 7890A-5975C gas chromatography-mass spectrometer to quantitatively and qualitatively analyze 15 PAHs. The composition spectra of PAHs were analyzed by scanning electron microscopy. The standard internal method established a standard curve to qualitatively and quantitatively analyze PAHs. The correlation coefficients of the calibration curves were all above 0.999, as shown in Table 2.

Table 2 15 optimal control PAHs standard curve and recovery rate

Number	PAHs	Standard curve line	R^2	Recovery rate
1	Acenaphthylene (ACY)	y = 0.0138x – 0.009	0.9999	80.25%
2	Acenaphthylene (ACE)	y = 0.0333x – 0.0125	0.9997	96.35%
3	Fluorene (FlO)	y = 0.0371x – 0.0135	0.9995	105.34%
4	Phenanthrene (PHE)	y = 0.0321x + 0.0261	0.9996	97.81%
6	Fluoranthene (FlA)	y = 0.0254x + 0.0201	0.9997	89.24%
7	Pyrene (PYR)	y = 0.0271x + 0.0363	0.9999	95.68%
8	Benzo[a]anthracene (BaA)	y = 0.0084x + 0.0101	0.9995	86.77%
9	Chrysene (CHR)	y = 0.0421x + 0.0332	0.9997	101.92%
10	Benzo[b]fluoranthene (BbF)	y = 0.0141x – 0.0078	0.9994	105.34%
11	Benzo[k]fluoranthene (BkF)	y = 0.0189x – 0.0491	0.9995	91.65%
12	Benzo[a]pyrene (BaP)	y = 0.0082x – 0.0061	0.9992	93.26%
13	Indenol[1,2,3-cd]pyren (ICDP)	y = 0.0118x – 0.0002	0.9995	87.94%
14	Dibenzo[a,h]anthracene (DaHA)	y = 0.0109x – 0.0003	0.9992	110.51%
15	Benzo[b]fluoranthene (BghiP)	y = 0.0245x – 0.0166	0.9993	98.67%

2.3 Quality assurance and quality control (QA/QC)

The samples in this study were sampled, analyzed, and quality controlled in strict accordance with the standards. All glass instruments are ultrasonically cleaned in an ultrasonic cleaner, pickled, and heated in a muffle furnace. Anhydrous sodium sulfate was baked in a muffle furnace at 550 °C, and alumina and silica gel were baked at 200 °C. The target compound is quantified using the calibration method, and the recovery rate corrects the final result after deducting the blank value.

2.4 Cancer risk assessment

The carcinogenic effect of each type of PAH monomer can be evaluated by the benzo[a]pyrene equivalent concentration (BaP_{eq}) and the toxic equivalency factors corresponding to the monomer.Based on analyzed PAHs in Jinhua atmospheric particles, the ILCR model was used to quantify and evaluate the health risks of fine atmospheric particles through respiratory exposure.

Referring to the model recommended by the USEPA and World Health Organization, the formula for calculating the ILCR of people inhaling PAHs through the respiratory route is as follows (Shen et al., 2019):

$$ILCR = \frac{TEQ \times CSFinh \times \sqrt[3]{\frac{BW}{70}} \times IRinh \times EF \times ED \times CF}{BW \times AT}$$

where ILCR denotes the lifetime carcinogenic risk of certain PAHs at a certain dose through respiratory exposure (dimensionless). TEQ denotes the concentration of ΣBaPeq of particulate PAHs (ng/m^3); CSF_{inh} denotes the carcinogenic intensity factor (mg/(kg·d)); BW denotes body weight (kg); IR_{inh} denotes the respiratory rate (m^3/d); EF denotes exposure frequency (d/a); ED denotes exposure years (a); AT denotes the average life span (d); CF denotes the conversion factor (10^{-6}). Table 3 shows the specific values.

Table 3 Values of PAHs toxicity exposure parameters

Expose parameters	children （1-11 years old）		adolescents （12-17 years old ）		adults （18-79 years old ）	
	man	woman	man	woman	man	woman
CSF_{inh}	3.14					
BW	17.2	16.5	47.1	44.8	60.2	53.1
IR_{inh}	8.4	8.4	13.1	13.1	17.7	14.5
IR_{ing}	200	200	200	200	100	100
EF	350	350	350	350	350	350
ED	6	6	14	14	30	30
AT	25,550					

3. Results and discussion

3.1 Particle size distribution characteristics of PAHs

The mass concentration of PAHs (Σ15PAHs) in PM_{10} of Jinhua ranged from 17.29 to 41.37 ng/m^3, and the average concentration was 29.78 ng/m^3. The PAH concentration in Yongkang was the highest, reaching 41.37 ng/m^3, and the concentration in Pan'an was the lowest, only 17.29 ng/m^3. The spatial distribution trend of the mass concentration of Σ15PAHs in the eight sampling areas is Yongkang (41.37 ng/m^3) > Yiwu (36.77 ng/m^3) > Lanxi (34.71 ng/m^3) > Wuyi (27.79 ng/m^3) > Jindong (27.22 ng/m^3) > Dongyang (26.69 ng/m^3) > Wucheng (26.41 ng/m^3) > Pan'an (17.29 ng/m^3), which is similar to the spatial distribution of PM_{10}. This result can indicate that the emission of atmospheric PAHs in Jinhua is closely related to the atmospheric PM emission. Yongkang and Lanxi are the industrial areas of Jinhua, while Yiwu has heavy traffic. The high atmospheric PM emissions in these places lead to high concentrations of PAHs carried on the atmospheric PM. On the contrary, Pan'an is an ecologically protected area in Jinhua, with less industry and less traffic, so the concentration of PAHs carried by atmospheric PM is the lowest.

Maximum, minimum, median, mean concentration, standard deviation and frequency density values for each size of the particulate matters and related Σ15PAHs mass concentrations during the winter of 2020-2021 in Jinhua are shown in Table 4. The mass frequency density curve of atmospheric PM_{10} exhibits a bimodal distribution pattern, with one peak in the S1 particle size and the other in the S_8 particle size. Unlike the mass frequency density curve of atmospheric PM_{10}, the mass frequency density curve of PAHs carried on PM_{10} exhibits a three-peak distribution pattern. The first and most important peak was in the S_1 particle size (0.4-0.7 μm), the second in the S_5 particle size (3.3-4.7 μm), and the third in the S_8 particle size (9-10 μm). Similar results were found in Beijing (China), where the highest concentration sum of PAHs was found in $PM_{0.5\text{-}1.0}$ (zhang et al 2021). Another consistent result came from the report by Shen et al. (Shen et al 2019a). They studied PAHs associated with size-segregated particulate matter at 10 cities in China and also found that particulate PAHs were dominant in fine size range of 0-1.1 μm. However, there was also an inconsistent finding. In the study of Shen et al. (Shen et al 2019a), the PAHs exhibited a unimodal pattern, while in our study, the PAHs presented a trimodal pattern, which should be related to the way of dividing the size of atmospheric particles.

Table 4 Maximum, minimum, median, mean concentration, standard deviation and frequency density values for each size of the particulate matters and related PAHs collected during the sampling periods

PM	S_0 0~0.4μm	S_1 0.4~0.7μm	S_2 0.7~1.1μm	S_3 1.1~2.1μm	S_4 2.1~3.3μm	S_5 3.3~.4.7μm	S_6 4.7~5.8μm	S_7 5.8~9μm	S_8 9~10μm	PM_{10} 0~10μm
Minimum (μg·m^{-3})	5.17	5.83	4.22	3.29	2.23	3.38	1.79	3.39	1.22	126.22
Maximum (μg·m^{-3})	15.33	25.20	16.96	12.92	10.50	13.06	10.92	16.26	17.67	33.98
Median (μg·m^{-3})	7.84	9.19	5.89	7.23	8.07	9.73	6.43	9.15	7.97	71.52
Mean (μg·m^{-3})	9.39	13.72	8.84	7.42	6.62	8.55	6.27	8.60	8.00	77.41
Standard deviation (μg·m^{-3})	3.51	7.26	4.88	3.33	2.96	3.49	3.14	4.23	5.69	32.55
Frequency density (μg·m^{-3}·μm^{-1})	23.48	45.73	22.10	7.42	5.52	6.11	5.70	2.69	8.00	7.74
Σ15PAHs	S_0	S_1	S_2	S_3	S_4	S_5	S_6	S_7	S_8	PM_{10}
Minimum (ng·m^{-3})	1.91	3.43	2.14	1.38	1.79	1.54	1.02	2.16	1.38	41.37
Maximum (ng·m^{-3})	4.89	12.52	5.53	3.28	2.78	3.37	3.30	4.08	3.02	17.29
Median (ng·m^{-3})	3.29	7.48	4.48	2.64	2.28	2.47	2.12	2.72	1.73	27.79
Mean (ng·m^{-3})	3.49	7.61	4.23	2.58	2.26	2.58	2.17	2.88	1.97	29.78
Standard deviation (ng·m^{-3})	0.98	2.64	1.07	0.60	0.35	0.62	0.77	0.59	0.63	9.81
Frequency density (ng·m^{-3}·μm^{-1})	8.73	25.37	10.58	2.58	1.88	6.11	1.97	0.90	1.97	2.98

According to the mass frequency density results of PAHs, in this study, we divided PM_{10} into three particle size segments: S_0–S_3 (0-2.1 μm, named fine size segment), S_4–S_6 (2.1-5.8 μm, medium size segment), and S_7–S_8 (5.8-10 μm, named coarse size segment). The PAH concentrations in the fine particle size segment were significantly higher than in the medium and coarse particle size segments. Therefore, PAHs in the atmosphere are more prevalent in fine particle size, which should be related to atmospheric PAHs' source and formation process.

Figure 2 shows the proportion of PAH monomers in the particles of different particle size segments in Jinhua. The top five PAH monomers in the coarse size segment were BbF, BkF, PHE, BaP, and ICDP, with proportions of 15.57%, 12.76%, 11.84%, 10.70%, and 9.87%, respectively, and a total proportion of 60.73%. The top five PAH monomers in the medium size segment were ANT, BkF, BbF, FLO, and BaP, with proportions of 16.14%, 12.42%, 10.89%, 9.87%, and 9.84%, respectively, and a total proportion of 59.15%. The top five PAH monomers in the fine size segment of the atmospheric particles in Jinhua City were CHR, BbF, PYR, BkF, and BaA, with proportions of 11.52%, 11.08%, 10.72%, 9.40%, and 8.75%, respectively, and a total proportion of 51.48%. Among the 15 different PAH monomers, BbF and BkF always ranked among the top five in each particle size segment and were the most abundant PAHs carried by atmospheric particles. Conversely, ACY and DaHA always ranked among the last five in each particle size segment and were the least PAHs carried by atmospheric particles. Furthermore, we find that the proportion of the top five PAHs decreases as the particle size segment becomes finer, indicating that the concentration distribution of each PAH in the fine particle size segment is more even. Figure 3 shows the distribution of PAHs with different ring numbers in different particle size segments. PAHs with 4 rings were more accumulated in the fine size segment than those with other rings, 5-6-ring PAHs were more accumulated in the coarse size segment, and PAHs with 3 rings are more likely to accumulate in the medium size segment.

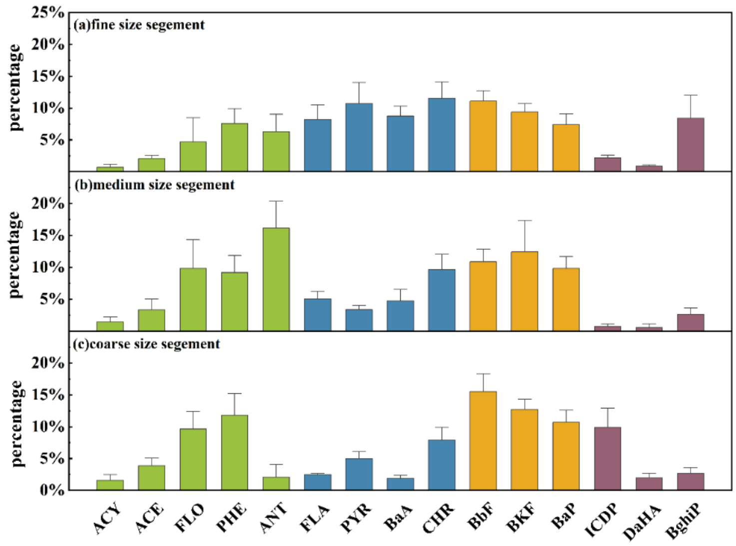

Figure2 Proportion of PAH monomer content in particles of different sizes in Jinhua

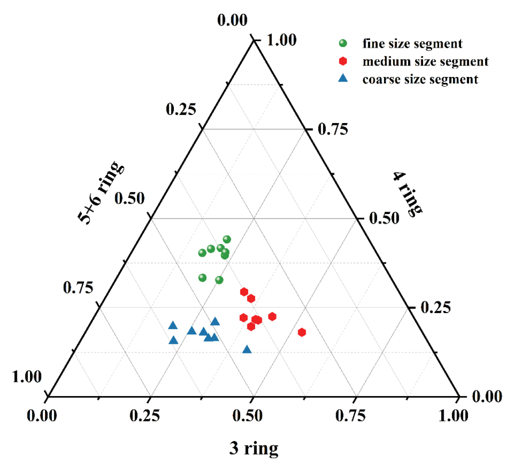

Figure 3 Distribution of PAHs with different ring numbers in different particle size segments

3.2 Cancer risk assessment

An estimation of the potential carcinogenic and mutagenic risks of exposure to PAHs was performed by calculating the inhalation lifetime cancer risk based on BaP-TEQ. The TEQ concentration on S_1 was much higher than that of other particle sizes, reaching 0.875 ng/m^3, while the concentration on S_4 was the smallest, only 0.209 ng/m^3, which meant the TEQ concentration in the fine size segment was significantly higher than that in the medium size segment or the coarse size segment. The TEQ concentration distribution was similar to the particle concentration distribution.

Figure 4 showed the ILCR values of each sample size of atmospheric particulate matter in Jinhua for various populations. The ILCR values of each particle size segment were all lower than 1×10^{-6}, indicating that the carcinogenic risk of PAHs of various particle sizes to humans was within an acceptable range. Though the cancer risk value of men was slightly higher than that of women, the difference was very small, which was related to the physiological characteristics of men and women.

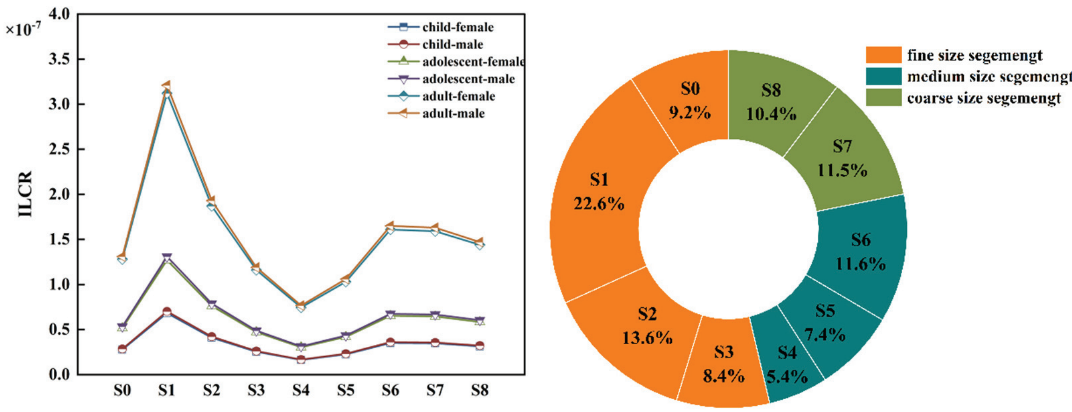

Figure 4 the ILCR and Proportion in different particle size

In this study, the carcinogenic risk of the 9 sampling particle sizes showed a trend of $S_1 > S_2 > S_6 > S_7 > S_8 > S_0 > S_3 > S_5 > S_4$, and the proportions of carcinogenic risk of the three particle segments (fine, medium, and coarse) were 53.7%, 24.4% and 21.9% respectively, indicating the carcinogenic risk in the fine size segment was much larger than that in the medium particle size and coarse particle size. Therefore, it is necessary to focus on the effective management of PAHs in the fine size segment of atmospheric particulates.

4. Conclusions

The total mass concentration of Σ15PAHs in PM_{10} of Jinhua ranged from 17.29 to 41.37 ng/m^3, with an average concentration of 29.78 ng/m^3. The distribution of PAHs on PM_{10} showed a trimodal pattern, with the highest concentration in the fine size segment. 4-ring PAHs were more likely to accumulate in the fine size segment, whereas 5-6-ring PAHs were more abundant in the coarse size segment, and 3-ring PAHs were more abundant in the medium size segment.The carcinogenic health risk values of PAHs for different populations differed, showing the characteristics of adults > adolescents > children. Although the ILCR value of each sampling particle size segment was less than 1×10^{-6}, the proportion of cancer risk in the fine segment was significantly higher than that in the medium or coarse size segments. The proportion of the carcinogenic risk of the three particle size segments was 53.7%, 24.4%, and 21.9%, respectively.

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Statements & Declarations

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