RESEARCH ARTICLE



Particulate polycyclic aromatic hydrocarbons in rural households burning solid fuels in Xuanwei County, Southwest China: occurrence, size distribution, and health risks

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Abstract

The study is about the size distribution and health risks of polycyclic aromatic hydrocarbons (PAHs) in indoor environment of Xuanwei, Southwest China particle samples were collected by Anderson 8-stage impactor which was used to gather particle samples to nine size ranges. Size-segregated samples were collected in indoor from a rural village in Xuanwei during the non-heating and heating seasons. The results showed that the total concentrations of the indoor particulate matter (PM) were 757 ± 60 and $990 \pm 78 \ \mu g/m^3$ in non-heating and heating seasons, respectively. The total concentration of indoor PAHs reached to $8.42 \pm 0.53 \ \mu g/m^3$ in the heating season, which was considerably greater than the concentration in the non-heating season ($2.85 \pm 1.72 \ \mu g/m^3$). The size distribution of PAHs showed that PAHs were mainly enriched in PMs with the diameter <1.1 μ m. The diagnostic ratios (DR) and principal component analysis (PCA) showed that coal and wood for residential heating and cooking were the main sources of indoor PAHs. The results of the health risk showed that the total deposition concentration (DC) in the alveolar region (AR) was 0.25 and 0.68 $\mu g/m^3$ in the non-heating seasons respectively. Throughout the entire sampling periods, the lifetime cancer risk (*R*) based on DC of children and adults varied between 3.53×10^{-5} to 1.79×10^{-4} . During the heating season, the potential cancer risk of PAHs in adults was significant, exceeding 10^{-4} , with a rate of 96%.

Keywords Indoor pollution · Size distribution · Solid fuels · PAHs · Health risk

Highlights

- Nine size ranges of particle were collected to investigate indoor PAHs in Xuanwei.
- The highest concentration of PAHs appeared at <0.4 and 0.7~1.1 μ m.
- Indoor PAHs were mainly contributed by the combustions of coal and wood.
- Indoor PAHs in PM_{1.1} had higher inhalation exposure health risks.

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Introduction

PAHs is a collection of hydrocarbons containing two or more benzene rings and can bind with deoxyribonucleic acid to destroy their chemical structures, and have health hazards such as carcinogenicity as well as mutagenicity. Therefore, PAHs is one of the most important pollutants in environmental risk management and control (Li et al. 2014).

Xuanwei county in Yunnan Province is a high-risk area for lung cancer in China (Li et al. 2019a, 2019b). The incidence and mortality rates of lung cancer ranked first the Nation (Li et al. 2019a, 2019b). Some studies showed that the air pollution caused by household burning solid fuels was closely related to the incidence of lung cancer in Xuanwei (Li et al. 2019a, 2019b; Longyi et al. 2023; Zhang et al. 2022). The level of carcinogenic PAHs represented by benzo[a] pyrene (BaP) was closely related to the incidence of lung cancer. Coal and wood were the main household solid fuels, and coal combustion was the main source of indoor pollution in the local area. The combustion efficiency of these traditional solid fuels was relatively low in household stoves. Besides, PAHs and heavy metals emitted from these fuels had been considered as the factors for lung cancer in Xuanwei (Gastelum et al. 2020; He et al. 1991; Kim et al. 2014; Longyi et al. 2013).

China's research on PAHs in the ambient air mainly focuses on some large cities (Baihuan et al. 2019; Yu et al. 2018). There were few studies on the indoor PAHs in rural areas (Chen et al. 2016; Ding et al. 2012; Lv et al. 2009). Incomplete combustion of coal and wood in Xuanwei had caused high incidence of lung cancer and had been the subject of intense scrutiny in recent years (Zhang et al. 2016). The impact of PAHs in ambient air on human health was closely related to their size distribution (Zhang et al. 2020). The size distribution of PAHs in the ambient air had a significant correlation with their effect on human health. The distribution of PAHs' sizes could impact the transportation, deposition, and health risks of PAHs to the human body (Kwon and Kim 2013). Fine particles can enter the body more easily through respiration and circulation (Jiang et al. 2020; Liu et al. 2015). The particle size largely determines the deposition distribution of inhaled PM along the respiratory system. Larger size of inhaled PM is typically deposited in the head region (HA), while finer fractions tend to accumulate in the tracheobronchial region (TB) and alveolar region (AR), showing more efficient deposition and longer retention times. It was a critical factors that deposition of most of the target PAHs in AR with fine and ultrafine fractions (Clarke and Valentin 2005). Therefore, the size distribution of PAHs in ambient air was an important parameter for source and health risk (Zhang et al. 2020).

When calculating the health effect of PAHs in Chinese cities, few studies had taken into account the contribution of DC. Especially, the size distribution and inhalation exposure of PAHs had been found to have a significant impact on human health. The inclusion of the DC in the analysis is crucial for obtaining results that closely reflect the actual situation. The size distribution of PAHs showed obvious seasonal variation (Zhang et al. 2020). Therefore, this study investigated the concentrations, profile, size distribution, sources, and health risks of PAHs in nine size ranges in the non-heating and heating seasons in Xuanwei. In addition, the consideration was given to the PM's deposition effectiveness in the human organism, allowing us to evaluate the health effects on various parts of the respiratory system. This assessment aims to offer scientific proof for enhancing the air quality in comparable regions.

Materials and methods

Site and sampling

Xuanwei (25° 53′ 30″~26°44′ 50″ N, 103° 35′ 30″~104° 40′ 50″ E, altitude: 1948 m, area: 6052.96 km²) is located in the

northeast of Yunnan Province, with low oxygen concentration in atmosphere and significant air temperature variations. It is mainly based on agriculture, supplemented by industry. The region is rich in coal, iron, copper, and other mining resources, especially bituminous coal resources (Ren et al. 2016). Due to the abundant coal resources, most local residents use untreated coal as domestic fuel for cooking and heating. Xuanwei is a high incidence area of lung cancer caused by PAHs in indoor air from bituminous coal combustion. Mortality rate of the cancer in this region was as high as 91.1/100,000, especially the mortality rate of woman was six times high than the average value of China (Xiao et al. 2012). In recent years, there has been a development towards the increasing occurrence of lung cancer at a younger age in Xuanwei (Shao et al. 2023).

In Xuanwei, the wood stove was mainly used for cooking in the non-heating season, and the coal stove was mainly used for heating and cooking in the heating season. According to the survey, there was about 87% of using wood and coal for cooking and heating in the region. The building type is mainly bungalows which are mostly of brick construction. There were 81 households with 498 people in the village where the sampled houses were located. Wood and bituminous coal as domestic energy sources by the sampled households was chosen to be representative of this study. The natural conditions of the sample subjects were a residential population of four people, excluding smokers. The building area of is 92 m². The samples were collected for 16 days during 19~26, July and 7~14 November 2021. The indoor and outdoor samples were simultaneously collected for 8 h from 11:00 a.m. to 19:00 p.m. (non-heating season) and 10:00 a.m. to 18:00 p.m. (heating season). According to the requirements of Indoor Air Quality Standard (Bai and Li 2022), the monitoring points were set in bedroom and kitchen (non-heating season), bedroom and living room (heating season), and 1.5 m above the ground, which is basically the same height as the human breathing zone. The sampling locations are positioned with the bedroom located 1 m away from both the window and the wall, and the kitchen and living room situated at a distance of 1 m from the stove and the wall. The outdoor sampling point is located 1.5 m above the ground. The locating of the sample is beside the rural road, and the common motor vehicles in the villages are cars and diesel vehicles. The sampling sites, monitoring setup points, and fuel combustion locations are shown in Fig. 1.

Anderson 8-stage impactor particulate sampler (Model TE 20-800, Sample Flow: 28.3 L/min) can divide indoor PM into nine particle size segments (F1, F2, F3, F4, F5, F6, F7, F8, and F9 respectively represent 9.0~10.0, 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 μ m), which were gathered and placed into 9 quartz filters measuring 81 mm in diameter.





Outdoor sampling was conducted using $PM_{2.5}$ and $PM_{1.0}$ samplers (Model TH150, Wuhan Tianhong Intelligent Instrument) with a flow rate of 100 L/min. Impurities

were eliminated by annealing 90-mm-diameter quartz filters at 450°C for 3 h, after which airborne particles were gathered. Before and after sampling, the filters were kept in a desiccator at a temperature of 25° C and a relative humidity of 45% for 48 h. Subsequently, they were weighted, packed into sample cells, and stored at a temperature of -4° C until extraction and analysis.

PAHs analysis

Accelerated solvent extraction and nitrogen blow concentration were used for the pretreatment of PAHs. After extraction and concentration, 16 priority controlled PAHs were analyzed by gas chromatography-mass spectrometry (7808B~5977B, Agilent, USA) (Liu et al. 2016). The capillary column (HP-5MS: 30 m* 0.25 mm *0.25 µm) was used to separate the target compounds. The chromatographic conditions were as follows: high-purity helium was used as the carrier gas, with no split injection, an injection port temperature of 280°C, a sample volume of 1 µL, and a constant flow rate of 1.0 mL/min. The temperature difference in the GC oven was as follows: initially at 70°C held for 5 min, then gradually rising to 270°C at a rate of 10°C/min, and finally remaining constant at 300°C for 10 min, with a rate of 5°C/min. Quantification was performed using the scan mode in the electron ionization mass spectra (70 eV) covering a mass range (m/z) from 50 to 550 amu. The ion source, quadrupoles, and transfer line were heated to temperatures of 230°C, 150°C, and 280°C, correspondingly.

Strict quality control was carried out during the analysis of samples. The balance condition of the filters is ensured to be consistent before and after sampling. Then, same electronic balance was used for weighing and recording. Abnormal values caused by improper personal operation or equipment failure that shall be eliminated during data analysis. The data of filter samples were corrected by blank and recovery correction, ensuring that the recovery data fell between 67.5 and 103%.

Table S1 displayed the chemical compositions of the sixteen identified PAHs. The compounds were categorized into six groups based on the number of rings: naphthalene (Nap, 2-ring), acenaphthylene (Acy, 2-ring), acenaphthene (Ace, 3- ring), fluorene (Flu, 3-ring), phenanthrene (Phe, 3-ring), anthracene (Ant, 3-ring), fluoranthene (Fln, 4-ring), pyrene (Pyr, 4-ring), chrysene(Chr, 4-ring), benz[a] anthracene (BaA, 4-ring), benz[b]fluoranthene (BbF, 5-ring), benz[k] fluoranthene (BkF, 5-ring), benzo[a]pyrene (BaP, 5-ring), dibenzo[a, h]anthracene (DbA, 5-ring), indeno[1,2,3cd]pyrene (InD, 6-ring), benzo[g, h, i]-perylene (BghiP, 6-ring). PAHs can also be classified as low molecular weight (LMW-PAHs, 2 and 3-ring PAHs), medium molecular weight (MMW-PAHs, 5 and 6-ring PAHs) PAHs.

Diagnostic ratio (DR)

DR is a method used to identify primary pollution sources by assessing the variations of components in diverse source categories. It is extensively employed for distinguishing the specific source characteristics of PAHs (Li et al. 2023). Given the relatively consistent ratios between PAHs, certain groups of relatively stable substances can be utilized to ascertain the origin of PAHs.

Principal component analysis (PCA)

The U.S. Environmental Protection Agency (EPA) has endorsed and suggested PCA as a method for allocating resources. This is a crucial statistical technique that can decrease the complexity of extensive datasets and identify the necessary principal components to account for all the variations in these datasets, which is significantly fewer than the initial number of variables (Jain et al. 2018). The components are established with an orthogonal distribution using this method. Regardless of the number of variables considered in this study, the regression adjustment yielded consistent and straightforward results for each factor (Gupta et al. 2018). During the analysis, the factors were identified by choosing principal components that had eigenvalues higher than 1 based on prior research (Henry 1987; Henry et al. 1984). Their explanation and representation of the unique properties of individual PAHs in PM were successful in guiding the comprehension and management of PAHs pollution emission in the research location, ultimately contributing to the enhancement of the well-being of the nearby communities.

Deposition concentration assessment

Inhalable PM can be deposited in different parts of the human respiratory system, mainly in three parts: HA, TB, and AR (Jain et al. 2018; Lao et al. 2018). Therefore, the hierarchical sampling technique of atmospheric particulates is combined with the human respiratory tract model (A Lung Dose Evaluation Program, LUDEP model). It is possible to estimate the DC of particulate PAHs in different parts of the human respiratory system (Bair 1995; Drewnick et al. 2021). The calculation formula for the deposition fraction (DF) of PAHs in different parts of human respiratory system is as follows:

$$DF_{HA,i} = IF_{i} \left[\frac{1}{1 + \exp\left(6.84 + 1.83InD_{p,i}\right)} + \frac{1}{1 + \exp\left(0.924 - 1.885InD_{p,i}\right)} \right]$$
(1)
$$DF_{TB,i} = \frac{0.00352}{D_{p,i}} \left\{ \exp\left[-0.234(InD_{p,i} + 3.40)^{2}\right] + 63.9 \exp\left[-0.819(InD_{p,i} - 1.61)^{2}\right] \right\}$$
(2)

$$DF_{AR,i} = \frac{0.0155}{D_{p,i}} \left\{ \exp\left[-0.416 \left(\ln D_{p,i} + 2.84 \right)^2 \right] + 19.11 \exp\left[-0.482 \left(\ln D_{p,i} - 1.36 \right)^2 \right] \right\}$$
(3)

$$\text{IF} = 1 - 0.5 \left(1 - \frac{1}{1 + 0.00076 D_{p,i}^{2.8}} \right) \tag{4}$$

where $D_{p,i}$ is the geometric mean size of the *i*th particle size section of Anderson 8-stage impactor particulate sampler. The geometric mean size of F1 is 9.5 µm; IF is the inhalable fraction (IF) of environmental particulate. DF of PAHs in different parts of human respiratory system are calculated according to Equations (1)–(4), and then the deposition concentration (DC) of PAHs is calculated according to the following equation:

$$DC_{j,i} = \sum \left(DF_{j,i} \times \rho_i \right)$$
(5)

where ρ_i is the concentrations of PAHs in the *i*th particle size range, ng/m³; DF_{*j*,*i*} is the deposition fraction of PAHs in the *i*th particle size segment at different parts of the human respiratory system; DC_{*j*,*i*} is the deposition concentration, ng/m³.

Health risk assessment

The health risk assessment model was used to assess the health risk of PAHs pollution for children and adults. PAHs can enter human body through respiratory system, causing adverse effects on health. Therefore, the primary objective of this research is to evaluate the potential health hazards associated with inhaling substances.

PAHs are widely recognized as species that have mutagenic, carcinogenic, and teratogenic properties (Meng et al. 2019; Sun et al. 2021; Włóka et al. 2015). Compared to the LMW-PAHs, the HMW-PAHs exhibit greater toxicity. Due to its presence in the surrounding environment and significant health risks, BaP is widely acknowledged as a substitute for examining health impacts (Clergé et al. 2019). BaP_{eqi}, which stands for the concentration equivalent to BaP, serves as an indicator to enhance comprehension of toxicity. The carcinogenic potential of PAHs can be estimated using toxicity equivalency factors (TEF) (Mallah et al. 2022). BaP_{eqi} is calculated by multiplying the individual PAHs concentration by their TEF. The equations of TEQ is as follows:

$$TEQ = \sum BaP_{eqi} = \sum (DC_i \times TEF_i)$$
(6)

where DC_{*i*} is the DC of individual PAHs within the *i*th particle size range, ng/m³; TEF_{*i*} is the toxicity equivalency factors; BaP_{eqi} is the equivalent concentration of BaP, ng/m³; TEQ is the total carcinogenic equivalent concentration of PAHs, ng/m³. The average daily dose (ADD) for this specific pathway is determined using the subsequent equation.

$$ADD = \frac{TEQ \times IR_i \times ET \times EF \times ED \times CF}{BW \times AT}$$
(7)

where TEQ is the total carcinogenic equivalent concentration of PAHs, ng/m³; ADD is the average daily dose, mg/ (kg·d); IR_i is respiratory rate, m³/d; ET is exposure time, h/d; EF is exposure frequency, d/a; ED is exposure duration, a; CF is the conversion factor, kg/mg; BW is body weight, kg; AT is average lifetime, h. Among them, the human respiratory exposure parameters were selected from the exposure parameters manual for Chinese population (Table 1) (Protection 2013).

The calculation of the carcinogenic risk value *R* for PAHs is determined using the following formula (Tabatabai 1987), where *R* is the lifetime cancer risk of PAHs with the inhalation exposure, when *R* is between 10^{-6} and 10^{-4} , there is potential carcinogenic risk, and when $R \ge 10^{-4}$, there is a potentially high carcinogenic risk; *q* is the carcinogenic intensity coefficient of BaP, taking 3.14 (kg·d)/mg.

$$\mathbf{R} = q \times \text{ADD} \tag{8}$$

Results

Concentration of indoor PAHs

In the non-heating season, the total concentration of indoor PM was $757 \pm 60 \ \mu\text{g/m}^3$, in which the concentrations of F1, F2, F3, F4, F5, F6, F7, F8, and F9 were

Table 1	The exposure
paramet	er values

Parameter	Physical Significance	Unit	Children	Adult
EF	Exposure frequency (non-heating season)	d/a	245	245
	Exposure frequency (heating season)	d/a	120	120
ED	Exposure duration	а	6	30
ET	Exposure time	h/d	8	8
CF	Conversion coefficient	kg/mg	10-6	10-6
IR _i	Inhalation rate	m ³ /d	5	15
BW	Average body rate	kg	15	70
AT	Average lifetime (carcinogens)	h	25550	25550

52.5, 76.1, 73.9, 84.2, 83.1, 55.1, 82.6, 101, and 148 µg/m³ respectively. The particles <0.4 µm (F9) has the highest concentration. The mass median diameter (MMD) was 1.56 µm. In the heating season, the total concentration of indoor PM was 990 \pm 78 µg/m³. The average concentrations were 63.3, 73.8, 72.5, 73.2, 88.3, 121, 183, 203, and 111 µg/m³ respectively. The concentrations of particles in 0.4~1.1 µm (F7 and F8) size ranges were obviously higher than that in other ranges, and the 0.4~0.7 µm (F8) was the most abundant. The MMD was 1.44 µm.

The mean PAHs concentrations in this study were 2.85 \pm 1.72 µg/m³ (non-heating season) and 8.42 \pm 0.53 µg/m³ (heating season), which were 2.58~7.65 times higher than that in Linfen (Song et al. 2021), 15~30 times higher than that in Tangshan (Fang et al. 2020), and 55.5~90.5 times higher than that in Jilin (Bai and Li 2022). It can be shown that the concentrations of PAHs are significantly higher than those in many plain areas, which may be related to the incomplete combustion of fuel caused by the low oxygen concentration in the atmosphere of Xuanwei at high altitudes (Shi et al. 2023). As shown in Fig. 2, in the non-heating season, the cumulative concentrations of Fln and Pyr were 571 and 574 ng/m³, respectively, which were about two times higher than those of other monomer PAHs. The BaP was 169 ng/m^3 . In the heating season, the cumulative concentrations of Phe, Fln, Pyr, BaA, Chr, BbF, BkF, BaP, InD, and BghiP were relatively high, whose concentrations were 234, 533, 863, 1252, 1743, 942, 858, 1059, 331, and 378 ng/m³, respectively. The BaP concentrations far exceeds (169~1059 times) the maximum allowable risk level of indoor air (1 ng/m³) (Haigang et al. 2023). The BaP concentration is still much higher than that in Tangshan (3.64~61.6 ng/m³) (Fang et al. 2020), Hangzhou (average 3.31 ng/m³) (Lu et al. 2008), Shijiazhuang (40.1~44.1 ng/m³) (Tang et al. 2022), and Enshi (3.80~18.2 ng/m³) (Peng et al. 2018). However, the concentrations of BaP were significantly lower than those previously reports in Xuanwei (457 and 6269 ng/m³) (Lv et al. 2009). In recent years, more and more domestic stoves have been equipped with exhaust pipes when using fuel, which could reduce indoor PAHs concentrations and the risk of exposure to indoor air pollution and respiratory diseases for local people. It indicates that the residents of Xuanwei still suffer from relatively serious PAHs pollution (Table 2).

In the non-heating season, the peak values of size distribution of Acy, Ace, Phe, Ant, Fln, Pyr, BaA, Chr, BbF, BkF, BaP, InD, and BghiP were within the range of $0.4 \sim 0.7 \mu m$, DbA was within the range of $1.1 \sim 2.1 \mu m$, Nap and Flu were within the range of $< 0.4 \mu m$; in the heating season, the peak values of Acy, Ace, Flu, Phe, Ant, Fln, Pyr, BaA, Chr, BbF, BkF, BaP, DbA, InD, and BghiP were within the range of $< 0.4 \mu m$.

Indoor PAHs profiles

The profile of PAHs varies with different particle sizes ranges in non-heating and heating seasons (Figure S1). During the non-heating season, Phe, Fln, and Pyr exhibited higher proportions. The percentage composition of Phe was 14.2% (F2), 18.3% (F3), and 11.9% (F4), while Fln accounted for 27.5% (F6), 22.9% (F7), and 19.7% (F8). Pyr represented proportions of 22.1% (F6), 21.9% (F7), and 22.6% (F8). During the heating season, Nap, Fln, and Chr had higher contents within the particle size range of 3.3~10





Table 2 Comparison of BaPconcentration in indoor air inXuanwei with other cities. (ng/m³)

City	Sampling time/year	BaP concentration (non-heating season/ heating season)	PAHs species	Reference
Tangshan	2020	3.64/61.6	18	(Fang et al. 2020)
Hangzhou	2004	3.31	16	(Lu et al. 2008)
Shijiazhuang	2022	40.1/44.1	16	(Tang et al. 2022)
Xuanwei and Fuyuan	2009	457/6269	16	(Lv et al. 2009)
Enshi	2018	18.2/3.80	28	(Peng et al. 2018)
Xuanwei	2022	169/1059	16	This study

 μ m (F1~F4); higher contents of BaA, Chr, and BbF were found within the particle size range of <3.3 μ m (F5~F9), especially F8 and F9 (<0.7 μ m), the corresponding proportions were 15.1%, 21.1%, and 11.3%; 15.3%, 19.6%, and 11.2% respectively.

The percentage of PAHs with nine size ranges by ring number in the non-heating and heating seasons are shown in Fig. 3. The molecular structures of PAHs in the nine particle size ranges are also different. In the non-heating season, the PAHs emitted from wood combustion were mainly composed of MMW-PAHs. In the range of $<1.1 \mu m$ (F7~F9), the percentage of HMW-PAHs was significantly higher than that of LMW-PAHs. With the decrease of particle size, the relative content of 2- and 3-ring PAHs increased, while that of 4-ring PAHs decreased (F1~F5); 2- and 4-ring PAHs decreased (F6~F8). About 66.7%, 63.6%, 59.2%, and 54.3% of the 4-ring PAHs were concentrated on the <2.1 μ m (F6~F9). In the heating season, the relative content of 5- and 6-ring PAHs increased with the decrease of particle size, while 4-ring PAHs decreased. The higher proportion of 4 and 5-ring PAHs were concentrated on the $<2.1 \,\mu m$ (F6~F9). The 4-ring PAHs accounted for 53.1%, 52.1%,

54.8%, and 51% of the total PAHs, 32.8%, 37.3%, 35%, and 34.6% for 5-ring PAHs.

Size distribution of indoor PAHs

The particle sample is categorized into three modes based on size fractions: Aitken mode (< $0.4 \mu m$), accumulation mode ($0.4 \sim 2.1 \mu m$), and coarse mode ($2.1 \sim 10 \mu m$). In both non-heating and heating seasons, the PAHs exhibited a dualpeak mode (consisting of the Aitken mode and accumulation mode) in terms of their particle size distribution. In the non-heating period, the most prominent peak is observed $0.7 \sim 1.1 \mu m$, with another peak occurring $1.1 \sim 2.1 \mu m$ (Figure S2). During period of heating, the highest peak is observed at < $0.7 \mu m$ while another peak emerges within the range of $0.7 \sim 1.1 \mu m$ (Figure S3). The Aitken mode represents PM derived from combustion sources, while the accumulation mode signifies the presence of PAHs that have either condensed onto existing particles or grown through particle coagulation (Duan et al. 2005).

PAHs exhibited their highest concentrations in the size range of $0.7 \sim 1.1 \,\mu\text{m}$ during non-heating periods and $< 0.7 \,\mu\text{m}$ during heating periods, suggesting that particulate PAHs



Fig. 3 Percentage of PAHs with nine size ranges by ring numbers in non-heating and heating seasons

were predominantly found in submicron particles. During the non-heating season, the distribution of various PAHs with different numbers of rings in different size fractions was as follows: 7.4% in the Aitken mode, 66.5% in the accumulation mode, and 26.1% in the coarse mode for PAHs with 2 and 3-rings; 6.4%, 82%, and 11.6% for PAHs with 4-rings; and 7.1%, 71.7%, and 21.2% for PAHs with 5- and 6-rings. During the heating season, the percentages for 2- and 3-ring PAHs are 34.4%, 41.3%, and 24.3%; for 4-ring PAHs, the percentages are 45.6%, 50.9%, and 3.5%; and for 5 and 6-ring PAHs, the percentages are 49.4%, 48.5%, and 2.1%.

Concentration of outdoor PAHs

The concentrations of $PM_{2.5}$ and $PM_{1.0}$ outdoor were measured during both the non-heating and heating seasons (Fig. 4). The concentrations of $PM_{2.5}$ and $PM_{1.0}$ were 27.7~63.5 and 12.8~35.2 µg/m³ in the non-heating season, which were 33.2~63.2 and 28.5~52.7 µg/m³ in the heating season. The local outdoor PM concentration in the heating season is obviously higher than that in the non-heating season, which is lower than the class II concentration limit (75 µg/m³) specified in China's ambient air quality standard (GB 3095-2012). It was obviously lower than the household ambient PM concentrations level. Studies have shown that $PM_{2.5}$ emission factors (EFs) from household coal combustion may be about 100 times higher than that from coal combustion boilers in power plants (Zhang et al. 2018).

The concentrations of outdoor PAHs in $PM_{2.5}$ and $PM_{1.0}$ were 97.7 ± 51 and 67.9 ± 36 ng/m³ in the non-heating season, and 110 ± 67 and 86.1 ± 74 ng/m³ in the heating season (Table 3). The concentrations of BaP in $PM_{2.5}$ and $PM_{1.0}$ in non-heating and heating seasons exceeded the daily average standard value of BaP (2.5 ng/m³) (GB 3095-2012). Chr is a typical PAHs indicator in coal combustion process (Du



Fig. 4 The concentrations of outdoor $PM_{1.0}$ and $PM_{2.5}$ in non-heating and heating seasons

et al. 2017), which accounts for 5.80% and 10.5% of PAHs in $PM_{2.5}$, 7.02% and 11.6% of PAHs in $PM_{1.0}$ in non-heating and heating seasons, respectively.

The indoor PAHs concentration was significantly higher than the corresponding outdoor. However, the indoor PAHs concentration is positively correlated with the outdoor, indicating that the indoor and outdoor PAHs was affected by the same pollution source or the same environmental process (Du et al. 2017). Indoor-outdoor concentration ratios (I/O) of individual PAHs can provide some clues about pollutant's origin. An I/O > 1 indicates that an indoor source is contributing to the observed concentrations, while an I/O <1 suggest that the source of compounds is primarily outdoor (Dhabhai et al. 2023). In this study, the I/O ratio ranged between 1.3 (Nap) and 66.5 (InD) what clearly indicated that indoor combustion source was the dominant contributor to indoor levels during the non-heating and heating seasons (indoor $PM_{1,1}$ /outdoor $PM_{1,0}$). However, for Ace and DbA the ratios are 0.7 and 0.08 in the non-heating season; Acy, Ace, and Flu the ratios are 0.12, 0.46, and 0.74 in the heating season what could indicate contributions from outdoor sources. Many families use large quantities of bituminous coal and wood for heating and cooking, resulting in high indoor pollution. Therefore, the high concentrations of indoor PAHs are the primary causes of outdoor pollution.

Source of indoor and outdoor PAHs

Diagnostic ratios (DR)

PAH DRs are extensively employed for tracing the origins of PAHs or serving as a measure of the maturity of PM samples. Table S2 displayed the ratios of Ant/(Ant+Phe) versus BaA/(BaA+Chr) and Flu/(Flu+Pyr) versus InD/ (InD+BghiP). During the non-heating season, the indoor ratios of Ant/(Ant+Phe) and BaA/(BaA+Chr) ranged from 0.14 to 0.59 and 0.14 to 0.95, respectively, indicate that wood and coal combustion were the main sources. The ratio of Flu/(Flu+Pyr) higher than 0.2 indicates the oil burning. Wood and coal combustion are the primary sources, as indicates by ratios exceeding 0.5. If the value falls between 0.6 and 0.7, it is highly probable that the primary cause is the emission from diesel vehicles (Hu et al. 2018a, 2018b). The indoor ratio of Flu/(Flu+Pyr) ranged from 0.20 to 0.47 suggested that oil burning may be the primary source of PAHs. In this study, the range of the InD/(InD+BghiP) indoor ratio was between 0.37 and 0.88. Diesel vehicle emissions are considered the primary sources when the value falls between 0.35 and 0.7, whereas values exceeding 0.5 indicates that wood and coal combustion become the primary sources (Yunker et al. 2002). The outdoor ratios of Ant/(Ant+Phe), BaA/(BaA+Chr), Flu/(Flu+Pyr), and InD/(InD+BghiP)

 Table 3
 Concentrations of PAHs

 in PM_{2.5} and PM_{1.0} outdoor
 during sampling. (ng/m³)

PAHs Mean ± SD	PM _{2.5}		PM _{1.0}		
	Non-heating season	Heating season	Non-heating season	Heating season	
Nap	7.04 <u>+</u> 1.18	12.5±1.53	6.42±1.34	9.77 <u>±</u> 2.49	
Acy	1.71±1.45	4.58±3.72	1.49 <u>+</u> 1.44	4.34 <u>+</u> 3.99	
Ace	1.88±1.30	1.18 <u>+</u> 0.44	1.41±0.70	1.89 ± 1.05	
Flu	4.34 <u>+</u> 3.44	6.24 ± 2.70	3.78±0.85	3.25±2.34	
Phe	10.2 ± 1.72	9.27 ± 7.35	8.45±2.62	8.77 <u>±</u> 7.64	
Ant	1.77 ± 1.20	3.69 <u>±</u> 0.83	1.93 <u>+</u> 1.48	2.54 ± 2.22	
Fln	10.4 <u>+</u> 4.88	12.6±7.91	7.21±3.10	10.1±6.82	
Pyr	10.1 <u>+</u> 3.40	9.22 <u>+</u> 6.76	6.17±2.65	8.16 <u>+</u> 7.57	
BaA	8.35 <u>+</u> 0.83	7.72 <u>+</u> 3.70	5.29 <u>+</u> 2.40	5.92 <u>+</u> 5.87	
Chy	5.67±5.22	11.6 <u>+</u> 9.95	4.77±3.71	10.1±11.8	
BbF	10.6 <u>+</u> 8.72	13.19 <u>+</u> 6.77	7.23 <u>+</u> 6.45	7.97 <u>±</u> 8.11	
BkF	4.01 <u>+</u> 2.28	2.15 ± 1.25	2.50±0.83	6.06 ± 7.95	
BaP	7.69 <u>±</u> 3.88	8.11 <u>±</u> 6.15	2.93 <u>±</u> 2.43	3.85±2.87	
DbA	6.32 <u>+</u> 3.78	3.78±3.56	3.88 <u>+</u> 2.10	1.38±1.44	
InD	4.57 <u>+</u> 4.60	1.12 <u>+</u> 0.89	3.12±2.76	0.90 <u>±</u> 0.86	
BghiP	3.00 ± 3.45	3.49 ± 3.09	1.40±1.36	1.26 <u>±</u> 0.87	
PAH ₁₆	97.7±51.2	110 <u>+</u> 66.6	67.9±36.1	86.1±73.9	

were 0.24~0.44, 0.75~0.87, 0.48~0.57, and 0.68~0.75, respectively.

During the heating season, when the ratio of Ant/ (Ant+PA) is less than 0.1, diesel vehicle emission is regarded as the primary contributor. The ratio ranged from 0.1 to 0.28, which suggests that wood and coal combustion are the major sources (Ambade et al. 2022). When the ratio of BaA/(BaA+Chr) is less than 0.35, diesel vehicle emission is regarded as the primary contributor. On the other hand, when the ratio is greater than 0.35, wood and coal combustion are the main sources of PAHs (Hu et al. 2018a, 2018b; Martellini et al. 2012; Masiol et al. 2012). According to the indoor ratios of Ant/(Ant+Phe) and BaA/(BaA+Chr) in this study, ranging from 0.11 to 0.23 and 0.23 to 0.79, it can be inferred that wood and coal were the main sources. The indoor ratios of Flu/(Flu+Pyr) and InD/(InD+BghiP) ranged from 0.18 to 0.83 and 0.35 to 0.98, suggesting wood, coal combustion, and diesel vehicle emission might be the major sources of PAHs. The outdoor ratios of Ant/ (Ant+Phe), BaA/(BaA+Chr), Flu/(Flu+Pyr), and InD/ (InD+BghiP) were 0.44~0.57, 0.68~0.72, 0.65~0.85, and 0.59~0.72, respectively. Considering those ratios, the main source of indoor PAHs was coal and wood, while the main source of outdoor PAHs was coal used in power, chemical, and coking plants.

PCA analysis

To study the sources of finer particle sizes, DRs are inadequate for investigating the potential sources of PAHs (Tang et al. 2022). The PCA method is extensively employed for the identification of the main origins of PAHs. The results of PCA analysis in indoor environmental were displayed in Table S3 after performing PCA with varimax rotation. KMO-Bartlett spherical test was used to determine the significance of correlation variables. The KMO were 0.82 and 0.78 in non-heating and heating seasons, with P<0.001. The PAHs data were shown to be suitable for factorial analysis.

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According to PCA of indoor PAHs, three and two factors during the non-heating and heating season, respectively, were extracted from PAH variables. Three principal components in non-heating season, explained 84.4% of the sources. Factor 1 (71.9% of the total variance) was mainly defined by Acy, Ace, Phe, Ant, Fln, Pyr, BaA, Chr, BbF, BkF, BaP, DbA, and BghiP. Previous studies pointed that Acy and Ace were associated with wood combustion (Ambade et al. 2022). Phe, Ant, Fln, Pyr, BaA, Chr, BbF, BkF, BaP, DbA, and BghiP serve as indicators of the burning of coal, coke, and the emissions from vehicles, including the combustion of diesel and gasoline (Chen and Liang 2021). The high loading of LMW-PAHs (specifically Nap) accounted for 12.4% of the variance, which was factor 2. The variance was explained by factor 3 (InD) with 6.75%. Nap indicates the burning of coal while InD represented the release of pollutants from vehicles.

PCA extracted two principal components in the heating season, which explained 92.7% of the total variables. Factor 1 (47.6% of the total variance) consisted mainly of BaA, Chr, BbF, BkF, BaP, DbA, InD, and BghiP. Some researchers had reported that coal combustion can account for the presence of BaA, Chr, BbF, BkF, and BaP (Chen and Liang 2021); DbA, InD, and BghiP were considered as indicators of diesel

vehicle emission (Li et al. 2016; Marcia et al. 2012; Qi et al. 2021). Factor 2 (Acy, Ace, Flu, Phe, Ant, Fln) explained 45.1% of the variance.

PCA analyses of outdoor PAHs. From the sixteen PAHs variables, two factors were identified during both the non-heating and heating seasons (Table S4). The non-heating season, factor 1 (including Acy, Ace, Phe, Ant, Fln, Pyr, BaA, Chr, BbF, BkF, BaP, DbA, and BghiP) and factor 2 (including Nap and Ant) explained 72.0% and 12.8%, respectively. For the heating season, factor 1 (including BaA, Chr, BbF, BkF, BaP, DbA, and BghiP) and factor 2 (including Acy, Ace, Flu, Phe, Ant, and Fln) explained 45.4% and 42.0%, respectively.

Health risk assessment of indoor PAHs

Deposited in human respiratory tract

The deposition concentration (DC) of PAHs in the respiratory system was calculated by the formulas $(1)\sim(5)$ for HA, TB and AR during the non-heating and heating seasons (Table S5). During the heating season, the total DC was greater at 1909 ng/m³ compared to the non-heating season which recorded 1017 ng/m³. In the nonheating season, the concentrations deposited on HA, TB and AR were 708, 54.2, and 254 ng/m³; 1111, 113, and 683 ng/m³ in the heating season. The results showed that the particulate PAHs were mainly deposited in HA, less in AR and TB. This result was consistent with previous studies (Wang et al. 2019; Wang 2020). However, this study when solid fuel was used; the proportion of PAHs deposited in HA decreased, while that deposited in AR increased significantly. The experimental results showed that PAHs emitted from coal combustion are more likely to deposit in the alveoli of human body, causing serious harm to the health of residents.

Studies had shown that fine particles cannot be discharged after entering human alveoli (Luo et al. 2014). Chemical components in fine particles will activate oncogenes, inactivate tumor suppressor genes, cause mutation of genetic materials, and have greater harm to human health. Figure S4 showed DC of AR percentage for individual PAHs with nine size ranges in non-heating and heating seasons. The particle size range of $0.4 \sim 0.7 \mu m$ (F8) had the highest percentage contribution to the AR DC of PAHs, followed by the $0.7 \sim 1.1 \,\mu m$ (F7) in the non-heating season. The percentage contribution of particles greater than 3.3 µm (F1~F4) was smaller. During the heating season, the less than 0.4 µm (F9) significantly increased compared to the non-heating season. The percentage contribution of MMW-PAHs and HMW-PAHs to the AR DC was mainly composed of the less than 1.1 µm (F7~F9), accounting for over 90% of the total. Therefore, during the heating season when coal was used,

the percentage contribution of F7~F9 PAHs to the AR DC in indoor air was higher than during the non-heating season. When wood was used, the contribution rates varied across different particle size fractions. This indicated that the size distribution of PAHs emitted from solid fuels during the heating season was finer, which posed a greater health risk to humans.

Cancer risk assessment

The health risk of PAHs in non-heating and heating seasons is shown in Table 4. Compared with HA and TB areas, particles deposited in AR area were difficult to remove, so only DC AR was used to estimate *R*. The *R* values of children and adults based on PAHs DC in the non-heating season were 5.49×10^{-5} and 3.53×10^{-5} , respectively. The *R* values of children and adults were in the range of $10^{-6} \sim 10^{-4}$, indicating that there was potential carcinogenic risk of PAHs in indoor environment. The *R* values based on PAHs DC in the heating season were 1.79×10^{-4} for children and 1.15×10^{-4} for adults.

In the non-heating season, the maximum R values for children and adults based on BaP DC were 2.90×10^{-5} and 1.86×10^{-5} . The R values for children and adults of BaA, BbF, BkF, BaP, and DbA were in the range of $10^{-6} \sim 10^{-4}$, indicating potential cancer risk. In the heating season, the R values of BaA, Chr, BbF, BkF, and DbA ranged from 10^{-6} to 10^{-4} . The adult R values of BaA, Chr, BbF, BkF, BaP, and DbA were in the range of $10^{-6} \times 10^{-4}$.

 Table 4
 Lifetime cancer risk values of PAHs based on inhalation

 exposure route in non-heating and heating seasons

PAHs species	Non-heating season		Heating season	
	Children	Adult	Children	Adult
Nap	1.77×10^{-8}	1.14×10^{-8}	2.71×10^{-8}	1.74×10^{-8}
Acy	6.90×10^{-9}	4.43×10^{-9}	1.79×10^{-9}	1.15×10^{-9}
Ace	8.82×10^{-9}	5.67×10^{-9}	1.26×10^{-9}	8.11×10^{-10}
Flu	1.15×10^{-8}	7.38×10^{-9}	3.89×10^{-9}	2.50×10^{-9}
Phe	5.32×10^{-7}	3.42×10^{-7}	3.33×10^{-7}	2.14×10^{-7}
Ant	1.50×10^{-7}	9.61×10 ⁻⁸	5.93×10^{-8}	3.82×10^{-8}
Fln	9.57×10^{-8}	6.15×10^{-8}	6.35×10^{-8}	4.08×10^{-8}
Pyr	8.88×10^{-8}	5.71×10^{-8}	9.02×10^{-8}	5.80×10^{-8}
BaA	3.28×10^{-6}	2.11×10^{-6}	1.31×10^{-5}	8.40×10^{-6}
Chr	4.59×10^{-7}	2.95×10^{-7}	1.93×10^{-6}	1.24×10^{-6}
BbF	3.54×10^{-6}	2.28×10^{-6}	1.01×10^{-5}	6.47×10^{-6}
BkF	1.90×10^{-6}	1.22×10^{-6}	8.69×10^{-6}	5.59×10^{-6}
BaP	2.90×10^{-5}	1.86×10^{-5}	1.11×10^{-4}	7.14×10^{-5}
DbA	1.56×10^{-5}	1.00×10^{-5}	3.32×10^{-5}	2.13×10^{-5}
InD	1.61×10^{-7}	1.04×10^{-7}	5.20×10^{-7}	3.34×10^{-7}
BghiP	1.32×10^{-8}	8.49×10^{-9}	3.16×10^{-8}	2.03×10^{-8}
PAH ₁₆	5.49×10^{-5}	3.53×10^{-5}	1.79×10^{-4}	1.15×10^{-4}

carcinogenic risk. However, only the *R* value of children based on BaP DC was $>10^{-4}$ (1.11×10⁻⁴), indicating high carcinogenic risk for children.

Discussion

Households burning solid fuels was one of the major contributors to ambient PM concentration levels throughout the urban and rural areas (Du et al. 2017). In this study, the size distribution of indoor PM and PAHs concentrations tends to aggregate towards finer particle sizes. The results showed that the concentration level of indoor PM was high, and were easily enriched in <1.1 μ m in the non-heating and heating seasons. There was obvious difference in size distribution between different sampling periods. In the non-heating season, the PM from wood combustion mainly concentrates on <0.4 μ m (F9), while in the heating season, the PM from coal combustion mainly concentrates on 0.4~1.1 μ m (F7 and F8).

It was shown that the PAHs emitted from coal and wood combustion were easily enriched in $<1.1 \mu m$, which contrasts with previous finding suggesting that PAHs emitted from coal combustion tend to accumulated in $<2.1 \mu m$ (Shen et al. 2010). This could be attributed to the variations in stove types and coal characteristics. Earlier research indicated that PAHs released from burning coal had a tendency to form smaller particles, particularly fine particles measuring less than 1.0 µm in aerodynamic diameter (Cheng et al. 2019; Shen et al. 2014; GuoFeng et al. 2013; Romagnoli et al. 2017), suggesting that coal combustion for cooking and heating contributed significantly to the PAHs concentration in indoor environment. The extensive use of bituminous coal by residents for heating and cooking in the heating season results in an increase in the concentration of Chr in PAHs. Therefore, Xuanwei had existed PAHs contamination in the indoor air, which was related to the long-term use of solid fuels by residents in this area.

The profiles of PAHs indicated a predominant composition of MMW-PAHs. Compared to the study (Peng et al. 2018), where MMW-PAHs were mainly concentrated on <2.5 μ m, our study primarily focused on <2.1 μ m. In the non-heating season, particle PAHs were more enriched in 0.7~1.1 μ m, <0.4 μ m in the heating season. In terms of ring distribution, 4- and 5-ring PAHs exhibited a greater tendency to concentrate on <2.1 μ m.

The significant portion of PAHs was found to be concentrated on the accumulation mode, particularly MMW-PAHs during both the non-heating and heating seasons. HMW-PAHs are more easily distributed into fine particles than LMW-PAHs (GuoFeng et al. 2013). It is inconsistent with the particle size distribution of PAHs in coal-fired rural aerosol studied by Song et al. (Song et al. 2021). The size distribution of PAHs in indoor and outdoor air was similar, which indicated that there were similar contamination sources, indoor solid fuel combustion emissions (Peng et al. 2018). Therefore, some studies believe that the indoor use of solid fuel pollution may be the main reason for the high incidence of lung cancer in Xuanwei (Luo et al. 2014; Wang et al. 2019; Wang et al. 2021). Human activities were responsible for the contamination that led to the high occurrence of lung cancer in Xuanwei. It was closely related to the early diagnosis and treatment of cancer and the popularization of relevant anti-cancer knowledge (Shao et al. 2023).

Through the analysis of PAHs sources, it has been determined that reducing PAHs emissions at the source is an effective approach for pollution control. In this study, the sources of PAHs were resolved by using PCA and DR. The results of PCA showed that the PAHs had the same sources in the non-heating and heating season, which were coal and wood, followed by diesel vehicle emission. The PCA also quantitatively analyzed the partial source of coke combustion. Jungang Lv et al. believed that PAHs in Xuanwei came from the combustion of bituminous coal in the test area (Lv et al. 2009), so it was speculated that power, cooking, and chemical plants were the main sources of outdoor PAHs pollution. There are abundant mining operations of coal (bituminous coal), iron, and copper in Xuanwei, especially rich bituminous coal resources (Ren et al. 2016). Besides, most local residents used untreated coal as living fuel for cooking and heating. Another reason for serious air pollution is the terrain characteristics of the test area. Many villages are surrounded by mountains, and it is difficult for pollutants to effectively diffuse out of these areas, which leads to high PAH concentrations and favor lung cancer prevalence in Xuanwei.

The carcinogenic risk to children and adults from indoor PAHs in rural Xuanwei was further explored and evaluated by calculating from Equations (8) (Table 4). Indoor PAHs were found to be a carcinogenic risk for both children and adults. The R values of children and adults were all higher than 10^{-4} , which indicated high carcinogenic risk of PAHs. The *R* of PAHs in the heating season was higher than that in the non-heating season, which may be related to the use of untreated coal in the heating season. The high content of MMW-PAHs and HMW-PAHs indicated potential carcinogenic risk in both non-heating and heating seasons. BaP has a strong carcinogenic effect on humans and contributes most to R (Luo et al. 2014). Only the R value of children based on BaP DC was $>10^{-4}$ (1.11×10⁻⁴), indicating high carcinogenic risk for children. Children are sensitive to pollutants and expose to pollutants for a long time; therefore, the health risks to children are higher than those to other ages. This is inconsistent with the results of previous studies on the health risks of PAHs in other cities in China (Fang et al. 2020; Li et al. 2021), which may be closely related to the younger age trend of lung cancer in Xuanwei (Shao et al. 2023).

The results showed that the coal without purification process was used extensively during the heating season. The combustion of these coals emitted not only highly polluting PAHs, but also high levels of HMW-PAHs. The use of coal char emitted HMW-PAHs, which were generally considered to be more toxic (Du et al. 2018). This was related to the incidence of lung cancer in Xuanwei. Notable achievements, particularly in the Southwest, we were made as a result of the collective efforts of the public and government to enhance air quality. However, the rural regions in the southwest remain susceptible to severe air contamination. The burning of coal in residential areas can significantly contribute to the presence of PAHs, which is highly likely to be the cause of the low air quality and adverse health impacts. Hence, it is crucial to focus on enhancing clean energy technologies alongside implementing policy regulations.

Conclusions

The indoor PAHs concentrations in this study were 2.85 \pm 1.72 and 8.42 \pm 0.53 µg/m³ in non-heating and heating seasons. It can be shown that the concentration of PAHs was significantly higher than those in many plain areas, which may be related to the incomplete combustion of fuel caused by the low oxygen concentrations in the atmosphere of Xuanwei at high altitudes. The size distribution showed that PAHs were easily enriched in $<1.1 \mu m$. The highest concentration of PAHs appeared at 0.7~1.1 µm in the nonheating season and $< 0.4 \,\mu\text{m}$ in the heating season. The DR and PCA showed that the indoor PAHs were significantly influenced by human activities. During the non-heating season, the indoor ratios of Ant/(Ant+Phe), BaA/(BaA+Chr), Flu/(Flu+Pyr), and InD/(InD+BghiP) were 0.14~0.59, 0.14~0.95, 0.20~0.47, and 0.37~0.88; 0.11~0.23, 0.23~0.79, 0.18~0.83, and 0.35~0.98 during the heating season. The PCA results of indoor PAHs showed that three in the nonheating season and two factors in the heating season were extracted, which explained 84.4% and 92.7% of the sources. The coal and wood for residential heating and cooking were the main sources of indoor PAHs. Health-risk assessment showed that children and adults had potential cancer risk based on PAHs DC and BaP DC in the non-heating season. During the heating season, children and adults had high cancer risk based on PAHs DC, and children had high cancer risk based on BaP DC. The high concentrations of PAHs were one of the main contributing factors to the occurrence of lung cancer in the local population. That was still the main exposure risk for local residents.

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Author contributions All authors have contributed to the conceptualization and design of the study, as follows: Xinyu Han: formal analysis, writing—original draft, conceptualization. Dingshuang Li: formal analysis, investigation, writing—review editing. Jianwu Shi: conceptualization, supervision. Wei Du: investigation, date curation. Shuai Li: date curation, validation. Yuqi Xie: supervision, resource. Shihan Deng: date curation, methodology. Zhihao Wang: visualization, resource. Senlin Tian: project administration, validation. Ping Ning: validation, visualization. All authors commented on previous versions of the manuscript and all authors read and approved the final manuscript.

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Data availability All data generated or analyzed during this study are included in this manuscript and its supplementary information files. The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy.

Declarations

Ethical approval This work does not carry out research on humans and animals, and does not involve the use of human subjects.

Consent to participate Not applicable.

Consent to publish Not applicable.

Competing interests The authors declare no competing interests.

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