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ORIGINAL PAPER

# Characteristics and health risk assessment of indoor and outdoor $PM_{2.5}$ in a rural village, in Northeast of China: impact of coal and biomass burning

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**Abstract** Fine particulate matter ($PM_{2.5}$) has health effects that may depend on its sources and chemical composition. In this study, characteristics of $PM_{2.5}$ chemical composition and health risk assessment from Songyuan, China, were investigated during day and night in indoor and outdoor from February 4 to 19, 2021. Relative high concentrations of $PM_{2.5}$ were obtained in indoor environment than outdoor, with $503.95 \pm 209.62$ μg/m$^3$ during the day and $357.52 \pm 232.81$ μg/m$^3$ at night for the indoor environment. Relatively high total carbon, organic carbons, elemental carbons, polycyclic aromatic hydrocarbons (PAHs), and oxygenated polycyclic aromatic hydrocarbons (OPAHs) were obtained in indoor environment. However, the average concentrations of PAHs were higher during night ($73.57 \pm 43.09$ ng/m$^3$) in indoor and OPAHs during day ($6.027 \pm 2.960$ ng/m$^3$) in outdoor. They had different I/O distributions of these compounds during day and night. Indeno(1,2,3-cd) pyrene was the dominant PAHs, and benzanthrone was the dominant OPAHs; this is different with the previous studies. The high indoor/outdoor ratios showed the indoor coal and biomass burning greatly affect the indoor pollutants. Average ILCR health risk assessment for PAHs was all higher than $10^{-6}$ for different age gender, suggesting there has potential cancer risk existed for populations living in the rural coal and biomass burning area Songyuan, China.

**Keywords** Characterization · $PM_{2.5}$ · PAHs · OPAH · Health risk · Coal and biomass burning

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## Introduction

Particulate matters (PMs) are small liquids or solids in the air, and it has severely affected atmospheric quality in China in recent decades (He et al., 2022). PMs are of different types, among which fine particulate matter ($PM_{2.5}$) is more toxic and can easily penetrate to lungs. $PM_{2.5}$ has major chemicals components such as organic carbons (OC), elemental carbons (EC), secondary organic

chemicals, polycyclic aromatic hydrocarbons (PAHs), and oxygenated polycyclic aromatic hydrocarbons (OPAHs) In rural areas of China, the major source of air pollution is residential use of solid fuels for combustion (e.g., crop residues, branches, wood, coal and straw), especially during the heating season (Gupta et al., 2023; Mukherjee & Agrawal, 2017; Zhang et al., 2013, 2016). Furthermore, during winter season people spend the majority of their time indoors. These factors trigger the severe PM indoor air pollution in China (Li et al., 2023; Zhang et al., 2023). Reports suggested that in 2015, the annual biomass energy utilization of Chinese residents was approximately 35 million tonnes of standard coal and approximately 95 million tonnes of burning coal (PRC National Bureau of Statistics, 2017; PRC National Energy Administration, 2016).

PAHs are largely diffused atmospheric and persistent organic pollutants, usually generate from fuel combustion (coal, petroleum, biomass, gasoline, and diesel). Some congeners of PAHs exhibited reproductive, immune, developmental, neurological, and hematological-toxicities (Chen et al., 2021). PM-bound PAHs were primarily from the adsorption of gas PAHs into particles. Some mutagenic and carcinogenic PAHs in $PM_{2.5}$ could easily penetrate into the lungs and damage respiratory system. Exposure to PAHs in mixture for long periods of time by skin contact or breathing could develop cancer risk in humans (ATSDR, 1995). Furthermore, PAHs could be converted into more hazardous derivatives such as nitrated, brominated, oxygenated, chlorinated, and hydroxylated PAHs after reactions with other chemicals, such as nitrogen oxides and ozone (Alves et al., 2017). OPAHs are produced through biomass burning or incomplete combustions of fossil fuels and formed through atmospheric processes (Wei et al., 2015). These oxidized derivatives are frequently found at similar levels as their parent PAHs (pPAHs) in the environments.

PM and associated chemicals studies focused on selected sources. For example, previously health risks caused by $PM_{2.5}$ and associated chemicals emitted from biomass fueled cooking were measured from rural Chinese home (Ding et al., 2012); incense burning emission at shrine Chiang Mai, Thailand (Bootdee et al., 2016); emitted by gasoline vehicles China (Zhao et al., 2020); typical Chinese residential cooking (Li et al., 2022); domestic solid fuel combustion in rural Guanzhong Basin, China (Li et al., 2019; Zhang et al., 2022). Meantime, the indoor and outdoor air-bounded PAHs in different microenvironments were also concerned (Mohammed et al., 2016; Li et al., 2017; Chen et al., 2017; Wang et al., 2017; Zhang et al., 2019b; Song et al., 2019; Mo et al., 2019; Kumar et al., 2020; Bai et al., 2020; Zhu et al., 2022). Several studies reported the health risk assessment of ambient $PM_{2.5}$ and associated chemicals which provides valuable information on the major $PM_{2.5}$ components (He et al., 2022; Zhou et al., 2018).

However, there is lack of studies of indoor and outdoor $PM_{2.5}$ regarding typical mixture rural coal and biomass burning, the so called Kang in Northern east in China. In this paper, our objectives are to measure the concentrations of $PM_{2.5}$ and the major chemical components and to calculate the health risk assessment from rural coal and biomass burning village located in Songyuan, Jilin Province, China.

## Material and methods

### Sites description and sampling

In this study, the sampling site was a rural village located in Songyuan, Jilin Province, where coal and firewood are the two most commonly used domestic fuels for daily cooking and winter heating. During the sampling period, the household mainly burned coal for heating and firewood cooking indoors, and the indoor sampler was placed next to the coal-fired stove, close to the wood-burning stove. The outdoor sampling point is ten meters away from the interior, surrounded by empty areas, without tall buildings, and far from factories and roads. The average minimum and maximum temperature during the sampling period were −16.6 °C and −6.4 °C, respectively. The sampling instrument settings are shown in Figure S1.

Mini-Volume $PM_{2.5}$ samplers (Airmetrics, Springfield, OR, USA) with a flow rate of 5 L/min and a quartz fiber filter with a diameter of 47 mm (QM/A®, Whatman Inc. U.K.) were used in this study. The sampling period was from February 4 to 19, 2021, and each sample was continuously collected for 12 h, with the sampling time from 09.00

to 21.00 during the day and from 21.00 to 09.00 the next day for the night. A total of 64 valid samples were collected in this study. The filters were placed in Teflon boxes and kept in a freezer at − 20 °C until analysis.

Mass and chemical analysis

The quartz membrane before and after sample collection was equilibrated for more than 24 h in a constant temperature and humidity chamber with a temperature of 20–23 °C and a relative humidity of 35–45% (MC108f, MeRyOu, Wuppertal, Germany), and then weighed using an ME5-F electronic microbalance (Sartorius, Germany).

The analysis of OC and EC analysis was performed by a thermo-optical carbon analyzer (DRI Model 2001) in an oxygen-free pure He environment at temperatures of 120 °C (OC1), 250 °C (OC2), 450 °C (OC3) and 550 °C (OC4), respectively; the 0.526 $cm^2$ filter was heated to convert the granular state on the filter paper into $CO_2$, which is catalyzed by $MnO_2$. The carbides formed during the carbonization of organic carbon are called cracked carbon (OPC). The final IMPROVE protocol specifies OC1 + OC2 + OC3 + OC4 + OPC as OC content and EC1 + EC2 + EC3 − OPC as EC content (Cao et al., 2007).

Half of the filter was used for the PAHs and OPAHs analysis. It was ultrasonic three times, with 5 mL dichloromethane mixed with methanol (V:V was 3:1), and then 5 mL dichloromethane for twice. Internal standards were added to control the extraction effective. Then, it was concentrated and for instrument analysis. An Agilent gas chromatography coupled with mass spectrometry (7890A-5975C) was used to qualitatively and quantitatively analyze PAHs, and a HP-5MS (30 m × 0.25 mm × 0.25 μm. Agilent) column was used. The temperature procedure for PAHs analysis was: the initial temperature was 60 °C, held for 1 min; then, it was heated to 150 °C at rate of 4 °C/min and maintained for 5 min, then raised to 300 °C at rate of 4 °C/min and maintained for 15 min. The ion source temperature of electron collision ion source (EI) was 230 °C, with an ionization energy of 70 eV, and a solvent delay time of 6 min. A total of 16 PAHs were analyzed in this experiment. For OPAHs analysis, the initial temperature was 80 °C, held for 3 min; then, it was heated to 200 °C at rate of 6.5 °C/min and maintained for 5 min and then raised to 300 °C at rate of 6.5 °C/min and kept for 5 min. The ion source temperature was 300 °C, and the ionization energy was 70 eV, with solvent delay of 6 min. A total of 9 OPAHs were analyzed in this study.

Quality assurance and control (QA/QC)

Before sampling, the filter membrane was roasted in a muffle furnace at a temperature of 780 °C for 4 h to prevent the adsorption and contamination of organic matter. During sampling, the samplers were calibrated routinely, and the variance for flow was approximately ± 2% (Wang et al., 2022a). The collected samples are kept in a freezer at < − 20 °C until analysis. The filed blanks were collected. For filters weighting, the absolute error is less than 0.015 mg (blank filter) and 0.020 mg (sample) for repeat weighing. The QA/QC procedures for carbon analysis in this study were same with our previous study (Wang et al., 2022b). The detection limits of EC and OC were both below 1.0 μg/$m^3$, and the relative deviation for replicate samples was smaller than 5% for TC (total carbon, TC), and 10% for OC and EC. Two internal standards phenanthrene-$d_{10}$ ($C_{14}D_{10}$) (98%, Aldrich, Milwaukee, WI, USA), chrysene-$d_{12}$ ($C_{18}D_{12}$) (98%, Sigma-Aldrich, Bellefonte, PA, USA) were added each sample for the PAHs and OPAHs analysis throughout the treatment process. Laboratory blank and blank with internal standards were treated same as the samples for each 12 samples. The recoveries of phenanthrene-$d_{10}$ and chrysene-$d_{12}$ were from 77.8 to 98.5% and from 89.3 to 106.5%, respectively. Repeat samples were done for each 20 samples and with a repeated error lower than 10%. Repeat instruments analysis was conducted randomly to check the instruments.

## Results and discussion

*Concentration of $PM_{2.5}$ mass*

The average concentration of $PM_{2.5}$ in indoor and outdoor environments in rural areas during the sampling period is shown in Table 1. The average concentrations of $PM_{2.5}$ during the day and night in the indoor were 503.95 ± 209.62 μg/$m^3$ and 357.52 ± 232.81 μg/

**Table 1** Summary of $PM_{2.5}$ and major chemical compositions (mean ± standard deviation) during the sampling period in this study

| Concentrations | Indoor | | Outdoor | |
|---|---|---|---|---|
| | Day | Night | Day | Night |
| $PM_{2.5}$ | 503.95 ± 209.62 | 357.52 ± 232.81 | 83.57 ± 41.38 | 66.10 ± 50.09 |
| OC | 241.66 ± 100.41 | 169.33 ± 120.77 | 20.86 ± 8.33 | 19.31 ± 11.31 |
| EC | 36.21 ± 20.98 | 30.19 ± 17.62 | 3.71 ± 2.31 | 3.96 ± 2.45 |
| OC1 | 21.27 ± 26.66 | 18.56 ± 30.98 | 0.54 ± 0.39 | 0.19 ± 0.43 |
| OC2 | 60.42 ± 30.34 | 40.76 ± 34.86 | 3.02 ± 1.23 | 2.17 ± 1.88 |
| OC3 | 92.88 ± 28.26 | 60.39 ± 31.28 | 6.07 ± 2.15 | 5.06 ± 2.53 |
| OC4 | 21.20 ± 3.34 | 17.92 ± 3.53 | 5.88 ± 1.75 | 5.77 ± 2.52 |
| OPC | 45.89 ± 24.05 | 31.69 ± 27.21 | 5.35 ± 3.40 | 6.11 ± 4.24 |
| EC1 | 74.00 ± 41.17 | 55.50 ± 40.13 | 8.13 ± 5.26 | 9.28 ± 2.12 |
| EC2 | 5.08 ± 1.58 | 4.03 ± 1.78 | 0.63 ± 0.30 | 0.53 ± 0.31 |
| EC3 | 3.01 ± 0.85 | 2.36 ± 1.05 | 0.31 ± 0.25 | 0.26 ± 0.36 |
| *Total carbon (TC)* | 277.87 ± 116.12 | 199.52 ± 134.71 | 24.58 ± 10.55 | 23.27 ± 13.62 |
| Naphthalene (NaP) | 0.013 ± 0.008 | 0.011 ± 0.009 | 1.26 ± 0.87 | 0.01 ± 0.01 |
| Acenaphthylene (ACY) | 0.47 ± 0.17 | 1.29 ± 1.88 | 0.32 ± 0.20 | 0.43 ± 0.65 |
| Acenaphthene (ACE) | 0.22 ± 0.22 | 0.31 ± 0.30 | 0.18 ± 0.13 | 0.19 ± 0.15 |
| Fluorene (FLO) | 0.14 ± 0.006 | 0.27 ± 0.16 | 0.30 ± 0.18 | 0.21 ± 0.28 |
| Phenanthrene (PHE) | 2.59 ± 1.07 | 3.59 ± 2.43 | 2.35 ± 0.84 | 2.00 ± 1.16 |
| Anthracene (ANT) | 0.26 ± 0.13 | 0.35 ± 0.28 | 0.24 ± 0.16 | 0.15 ± 0.06 |
| Fluoranthene (FLU) | 3.95 ± 1.93 | 6.15 ± 4.74 | 5.32 ± 5.29 | 3.84 ± 1.89 |
| Pyrene (PYR) | 3.34 ± 2.05 | 4.89 ± 3.17 | 4.29 ± 4.50 | 2.22 ± 1.56 |
| Benz(a)anthracene (BaA) | 3.63 ± 2.59 | 4.94 ± 3.42 | 1.81 ± 1.96 | 1.08 ± 0.64 |
| Chrysene (CHR) | 6.06 ± 4.45 | 7.72 ± 5.53 | 2.33 ± 2.22 | 1.08 ± 0.49 |
| Benzo(b)fluoranthene (BbF) | 3.94 ± 3.18 | 6.78 ± 4.90 | 2.04 ± 2.52 | 1.23 ± 1.25 |
| Benzo(k)fluoranthene (BkF) | 1.52 ± 1.46 | 3.20 ± 2.35 | 1.05 ± 1.08 | 0.37 ± 0.16 |
| Benzo(a)pyrene (BaP) | 4.11 ± 3.41 | 6.56 ± 4.87 | 1.54 ± 1.93 | 0.90 ± 0.68 |
| Indeno(1,2,3-cd)pyrene (IcdP) | 9.04 ± 2.68 | 16.03 ± 10.02 | 17.87 ± 18.19 | 11.00 ± 9.63 |
| Dibenz(a,h)anthracene (DahA) | 1.35 ± 1.16 | 1.55 ± 1.29 | 0.39 ± 0.35 | 0.22 ± 0.13 |
| Benzo(g,h,i)perylene (BghiP) | 4.52 ± 3.50 | 9.93 ± 7.21 | 2.57 ± 3.15 | 1.25 ± 0.90 |
| *Sum PAHs* | 45.14 ± 22.08 | 73.57 ± 43.09 | 43.85 ± 40.94 | 26.18 ± 15.92 |
| 1,4-Naphthoquinone (1,4-NaP) | 0.063 ± 0.037 | 0.125 ± 0.08 | 0.137 ± 0.096 | 0.099 ± 0.045 |
| 1,Naphthaldehyde (1-NaP) | 0.704 ± 0.284 | 0.961 ± 0.37 | 0.047 ± 0.038 | 0.085 ± 0.130 |
| 1-Acenaphthenone (1-Ace) | 0.625 ± 0.178 | 0.738 ± 0.30 | 0.932 ± 0.515 | 1.351 ± 0.509 |
| 9-Fluorene (9-FLO) | 0.381 ± 0.125 | 0.649 ± 0.38 | 1.606 ± 0.678 | 1.232 ± 0.423 |
| Anthraquinone (ANTQ) | 0.545 ± 0.316 | 0.718 ± 0.62 | 0.771 ± 0.533 | 0.627 ± 0.255 |
| 1,8-Naphthalic anhydride (1,8-NaAH) | 1.025 ± 0.583 | 1.247 ± 0.75 | 0.860 ± 0.662 | 0.512 ± 0.348 |
| Benzanthrone (BZA) | 5.419 ± 3.791 | 7.256 ± 5.78 | 1.480 ± 1.430 | 1.059 ± 0.542 |
| Benzo(a)anthracene-7,12-quinone (BaAQ) | 0.674 ± 0.511 | 1.009 ± 1.01 | 0.065 ± 0.0620 | 0.093 ± 0.081 |
| 5,12-Naphthacenequinone (5,12-NapQ) | 0.100 ± 0.122 | 0.152 ± 0.10 | 0.067 ± 0.039 | 0.018 ± 0.012 |
| *Sum OPAHs* | 9.535 ± 5.325 | 12.855 ± 8.42 | 6.027 ± 2.960 | 5.075 ± 1.653 |

μg/m$^3$ for $PM_{2.5}$ and carbon fractions, ng/m$^3$ for PAHs and OPAHs

m$^3$, respectively, which were pretty higher than the outdoors (83.57 ± 41.38 μg/m$^3$ in day and 66.10 ± 50.09 μg/m$^3$ in night). The high level of $PM_{2.5}$ could be due to households' indoor coal combustion and firewood burning for heating and cooking activities. The results suggested that high level of $PM_{2.5}$ in indoor could be health risk such as lungs and skin diseases to people living inside the room, and

during daytime, it is more toxic to public health and environment.

The average indoor concentration of $PM_{2.5}$ in this study was significantly higher than many researches developed in rural villages in China (see supporting Table S1), like rural Guizhou of firewood-burning households (Zhang et al. 2012), Qian'an rural area of Tangshan, Hebei Province (Zhang et al., 2020a) and rural Jiangsu (households of burning rice, and wood-burning) (Zhang et al., 2019a), which was higher than that of the two rural areas of Qinghai (278.37 μg/m$^3$ in Huzhu Village and 275.83 μg/m$^3$ in Datong Village) (Sun et al., 2021) and comparable to the average concentration of $PM_{2.5}$ in coal-fired indoor environment in Heshun Prefecture of Shanxi Province of 375.9 ± 572.8 μg/m$^3$ (Wang et al., 2022c). While the average outdoor daily $PM_{2.5}$ was lower than that of firewood-burning households in rural Guizhou (Zhang et al. 2012) and the two rural areas of Qinghai (Sun et al., 2021), compared with the $PM_{2.5}$ content of 66.1 ± 15.5 μg/m$^3$ of coal-fired households in rural Guizhou (Zhang et al. 2012). It is speculated that due to the low winter temperature in Songyuan, Jilin, the amount of coal and firewood burned and the burning time were much greater than those in the above-mentioned rural areas in indoor, and in the case of low outdoor temperature, households usually closed doors and windows to maintain a comfortable indoor temperature, but very little indoor and outdoor ventilation was not conducive to the diffusion of pollutants.

Meantime, the daily average concentration of indoor $PM_{2.5}$ during the sampling period is significantly higher than second-level national standard of China (75 μg/m$^3$) (Ambient Air Quality Standards, GB3095-2012), exceeding about 4.8 times of it, and significantly higher than the latest daily average concentration of ambient air $PM_{2.5}$ specified by the World Health Organization and WHO (15 μg/m$^3$), which could seriously endanger the health of local rural households.

## *Characteristics of $PM_{2.5}$*

### *Characteristics of carbon fraction*

The TC content of $PM_{2.5}$ collected during the sampling period was pretty higher indoor, of which indoor accounted for 55.14% (day) and 55.81% of $PM_{2.5}$ mass (night), than the outdoor 29.41% (day) and 35.20% (night), suggesting TC could be dangerous in indoor. The average concentrations of OC and EC during the outdoor day were 20.86 ± 8.33 μg/m$^3$ and 3.71 ± 2.31 μg/m$^3$, accounting for 24.96% and 4.44% of the $PM_{2.5}$ mass. The average concentrations of OC and EC at night were 19.31 ± 11.31 μg/m$^3$ and 3.96 ± 2.45 μg/m$^3$, accounting for 29.21% and 5.99% (both higher than the day). The average OC concentrations observed indoors were significantly higher than those observed outdoors, with the average concentrations of OC of 241.66 ± 100.41 μg/m$^3$ and 169.33 ± 120.77 μg/m$^3$ of day and night, with the contribution of OC to $PM_{2.5}$ observed during day and night was 47.95 and 47.36%, respectively, suggesting that the indoor burning activities emitted more carbon compounds and OC could pose toxic effects in indoor environment. The average indoor concentrations of EC were 36.21 ± 20.98 μg/m$^3$ and 30.19 ± 17.62 μg/m$^3$, and the EC to $PM_{2.5}$ during day and night was 7.18% and 8.44%, respectively. These results were similar with the previous studies from China (Liao et al., 2023), USA (Fine et al., 2008), and southern European cities (Alves et al., 2015).

The correlation between OC and EC can be used to identify the sources of them, where the high correlation coefficients ($R^2$) of EC and OC suggested the similar originations. The correlation between OC and EC from indoor and outdoor is shown in Figure S2. High correlation coefficients of outdoor EC were found between OC in $PM_{2.5}$ during day ($R^2 = 0.900$) and night ($R^2 = 0.827$), suggesting OC and EC have the similar sources (Liao et al., 2023). However, there was non-significant trends/relationship for indoor EC and OC. EC showed better correlations between indoors and outdoors than for OC, which was consistent with results reported by Xu et al. (2016). That OC and EC might come from different sources (Alves et al., 2015).

### *POC and SOC*

The sources of OC are divided into primary organic carbon (POC), emitted directly and secondary organic carbon (SOC), generated by complex reactions such as photochemical reactions and gas-particle conversion in the atmosphere. EC is chemically stable and comes directly from the incomplete combustion of fossil fuels and biomass

(Wang et al., 2022c). If the OC/EC value was more than 2.0, it indicates the presence of SOC (Li et al., 2020). The calculation of SOC and POC is shown in formulas (1) and (2).

$$SOC = OC - EC \times \left(\frac{OC}{EC}\right)_{min} \quad (1)$$

$$POC = OC - SOC \quad (2)$$

where SOC is the estimated secondary organic carbon content (μg/m$^3$); OC is the concentration of total organic carbon (μg/m$^3$); $(OC/EC)_{min}$ is usually the minimum observed OC/EC value. The concentrations and proportions of POC and SOC and ratio of OC/EC in $PM_{2.5}$ from indoor and outdoor are given in Table 2.

OC/EC ratio was higher (7.14) during the daytime in indoor as compared to night and outdoor, indicating that there was serious secondary organic carbon (SOC) pollution in the daytime. OC/EC ratio measured in our study was higher than reported for Seoul, which was 2.28 in summer and 1.89 in fall (Yoo et al., 2022).

Both concentrations of POC and SOC were higher in indoor than in outdoor, which was solid associated with the coal and biomass burning activities in indoor. This also could be concluded that the concentration of SOC in indoor daytime was higher than at night, and the contribution rates of indoor SOC to OC concentration were 48.60% in daytime and 34.36% at night. The SOC in outdoor day and night was estimated to be 6.35 μg/m$^3$ and 7.83 μg/m$^3$, respectively, accounting for 34.50% and 40.45% of the OC mass. The night SOC value is slightly higher than during the day, indicating that the secondary organic carbon pollution at night outdoors is more serious than during the day. However, the POC content in daytime was similar with that at night.

**Table 2** Concentrations and proportions of SOC and OC/EC ratios in $PM_{2.5}$ from indoor and outdoor in this study

| | Indoor | | Outdoor | |
|---|---|---|---|---|
| | Day | Night | Day | Night |
| OC/EC | 7.14 | 5.54 | 6.19 | 5.52 |
| $(OC/EC)_{min}$ | 3.59 | 3.32 | 4.03 | 3.16 |
| SOC (μg/m$^3$) | 120.15 | 73.97 | 6.35 | 7.83 |
| POC (μg/m$^3$) | 114.67 | 102.22 | 12.75 | 12.55 |
| SOC/OC (%) | 48.60 | 34.36 | 34.50 | 40.45 |

The SOC/OC estimated in this study was comparable with other regions, for example, rural Guangzhou (47%) (Lai et al., 2016), the suburbs of Guangzhou (41%) (Yuan et al., 2018), and Shanghai (48%) (Zhou et al., 2016). The large indoor value may be due to the increase of indoor coal consumption in northern winter, which leads to an increase in pollutant emissions; on the other hand, the cold weather and less ventilation time increase the concentration of indoor gaseous organic precursors, which is conducive to the secondary transformation of pollutants (Li et al., 2020). The above comparison results showed that the formation of secondary organic carbonaceous aerosols was significant in rural coal and biomass burning area Songyuan, China.

*Characteristics of PAHs and OPAHs*

PAHs and OPAHs are major chemical components, associated with particulate matter. They could posed carcinogenic and/or mutagenic affects even at low concentration. Comparison of the concentrations of PAHs and OPAHs revealed that indoor and outdoor environments during day and night exhibited dynamic trends (Fig. 1). The concentrations of PAHs and OPAHs were higher in indoor than in the outdoor, while they were higher during night in indoor and day in outdoor. The average PAHs in indoor were 45.14 (in day) and 73.57 ng/m$^3$ (at night). The PAHs concentrations were recorded higher as compared with our previous study in Xi'an China, where total PAHs concentrations were in the range 22.0–79.3 ng/m$^3$ and 32.9–95.4 ng/m$^3$ in indoors and 26.0–87.9 ng/m$^3$ in outdoors (Wang et al., 2017). The average PAHs in outdoor during sampling time were 43.85 daytime and 26.18 ng/m$^3$ night time. The PAHs were dominated by IcdP and BghiP, followed by CHR, BaP, and BbF in indoor air, and the concentration of IcdP (14.44 ng/m$^3$) was much higher than that of other substances (0.18–3.26 ng/m$^3$) in outdoor.

The indoor and outdoor concentrations of 16 PAHs in this study were lower to those in autumn season of Jinan, China (140.34 and 111.07 ng/m$^3$ in outdoor and indoor, respectively) (Zhu et al., 2015), and Beijing, which were 387.0 ng/m$^3$ and 187.3 ng/m$^3$ in outdoor and indoor environments, respectively

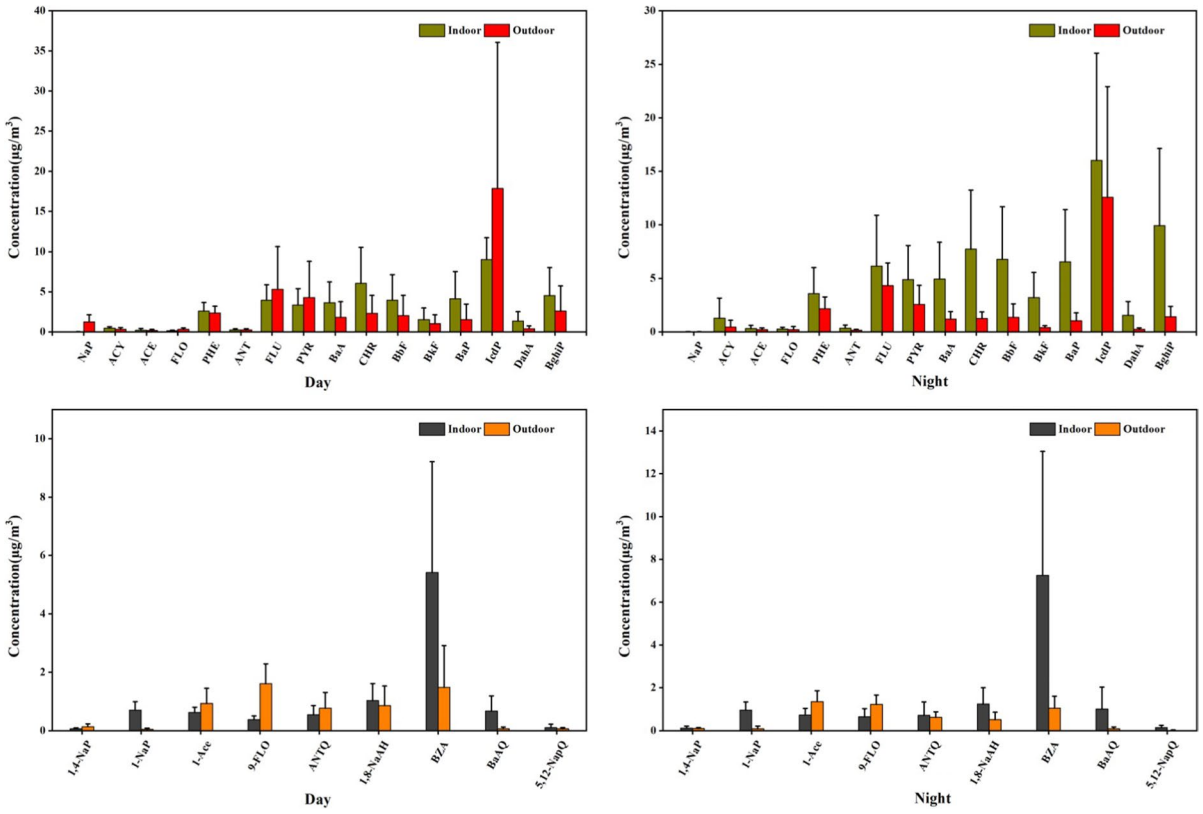


**Fig. 1** Concentrations of PAHs and OPAHs in day and night in indoor and outdoor in this study

(Han et al., 2016). Moreover, the concentrations were lower as reported for 16 $PM_{2.5}$-bound PAHs from Beijing (407.6 ng/m$^3$) (Shen et al., 2009) and Taiyuan (420.8 ng/m$^3$), China (Zhang et al., 2020b). However, the PAHs concentrations in the present study were comparable with the southern region of China. For example, the total 18 PAHs concentration in fine particles in Nanjing (50.6 ng/m$^3$ during pre-Spring Festival) (Kong et al., 2015); the concentration of $PM_{2.5}$-associated 15 ∑PAHs was 22.54 ng/m$^3$ during the autumn and winter in Nanchang (Liu et al., 2016); and 16 PAHs concentration in $PM_{2.5}$ was 23.7 ng/m$^3$ at Guangzhou with a heating source (Liu et al., 2015). In addition, the PAHs concentrations in this study were also lower than those in many areas of developed countries (Chang et al., 2006).

The most dominant profile of PAHs was IcdP in outdoor and indoor (41.22% and 21.12%, respectively). It was followed by FLU, PYR, PHE, and BghiP in outdoor (Fig. 2) and BghiP, CHR, BbF, and BaP, FLU in indoor. Meantime, ΣHMW-PAHs (5-, 6-rings) and LMW-PAHs (2-, 3-rings) also showed dynamic trends. ΣHMW-PAHs were dominant compounds in both indoor and outdoor environments. Similar trends were also observed in study conducted in Xian for outdoor and indoor (Wang et al., 2017) (Figure S3). This might be due to that LMW-PAHs have relatively higher vapor pressure and thus prefer to distribute in the gaseous phase, while the high molecular weight PAHs are prone to bound to particles.

The average OPAHs in indoor were 11.20 ng/m$^3$, showing that the daytime concentration was lower than the nighttime concentration. It was higher than in outdoor (5.82 ng/m$^3$), where daytime concentration was slightly higher than the nighttime concentration. For indoor OPAHs, the dominated profiles was BZA with average of 6.34 ng/m$^3$ and significantly higher than others, which was from 0.09 to 1.14 ng/m$^3$. For outdoor OPAHs, 9-FLO (1.48 ng/m$^3$), BZA (1.35 ng/m$^3$), and 1-Ace (1.20 ng/m$^3$) were also dominated, followed by ANTQ (0.74 ng/m$^3$)

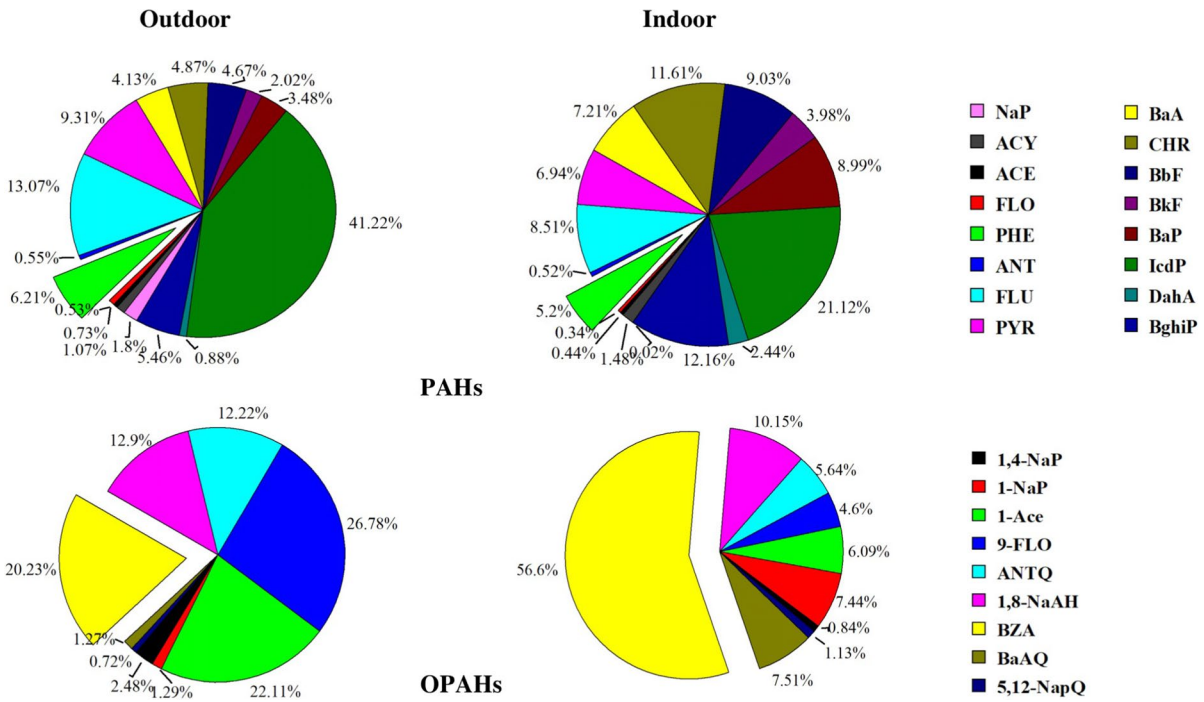


**Fig. 2** Profiles of PAHs and OPAHs in indoor and outdoor in this study

and 1,8-NaAH (0.73 ng/m$^3$), and other substances with lower levels. This was different with the study reported in Xian (Wang et al., 2017), which reported that among OPAHs, ANTQ dominates with concentrations of about 10 ng/m$^3$ in all three studied environments. Previous studies reported the higher level of OPAHs than our study. For example, the average total concentration of quantified OPAHs was 9.0 ± 9.0, 7.5 ± 4.6 and 7.5 ± 4.6 ng/m$^3$ for the personal exposure, indoor and outdoor samples (Li et al., 2019). The indoor and outdoor OPAHs in rural area in the middle-school in urban Xi'an was 16 ± 4.2 and 19 ± 3.9 ng/m$^3$, respectively (Wang et al., 2017). Higher air permeability and mobility are often observed in rural than urban area, resulting in a shorter suspending time of fine particles and thus less oxidation of pPAHs.

The ratios of 9-FLO/ FLO, ANTQ/ ANT, and BaAQ/BaA provide an indication for oxidation rates for FLO, ANT, and BaA. In this study, the average of 9FLO/ (FLO), ANTQ/ (ANT), and BaAQ/ (BaA) was 2.6 and 5.6, 2.1 and 3.8, 0.19 and 0.06, respectively, for indoor and outdoor, with no obvious difference for day and night. This was found to be lower than that in Xi'an ambient air, but higher than the oxygenation rate reported by Shen (2012). This suggested the important role of secondary formation processes for OPAHs (Hao et al., 2023; Kitanovski et al., 2020). It might due to the heterogeneous or homogeneous reactions between atmospheric oxidants (e.g., hydroxyl radical (OH) and ozone (O3)) and parent PAHs, which could generate OPAHs in the atmosphere (Huang et al., 2014).

## I/O ratios

Indoor/outdoor (I/O) ratios can serve as a representative of the relative intensities of the outdoor or indoor source (Kovacevic et al., 2015). Heating combustion sources trigger the higher outdoor aerosol concentrations which usually yield I/O ratios below 1. The I/O ratios of the carbon fractions, 16 PAHs, and 9 OPAHs are presented in Fig. 3.

The I/O ratios for carbon fractions were all higher than 1, and it has decreased for OC1 to OC4, with 41.7 (day) and 55.6 (night) for OC1 and then decreased to 3.8 (day) and 4.0 (night) for OC4. The EC1 to EC3 has similar values about 10. This can be

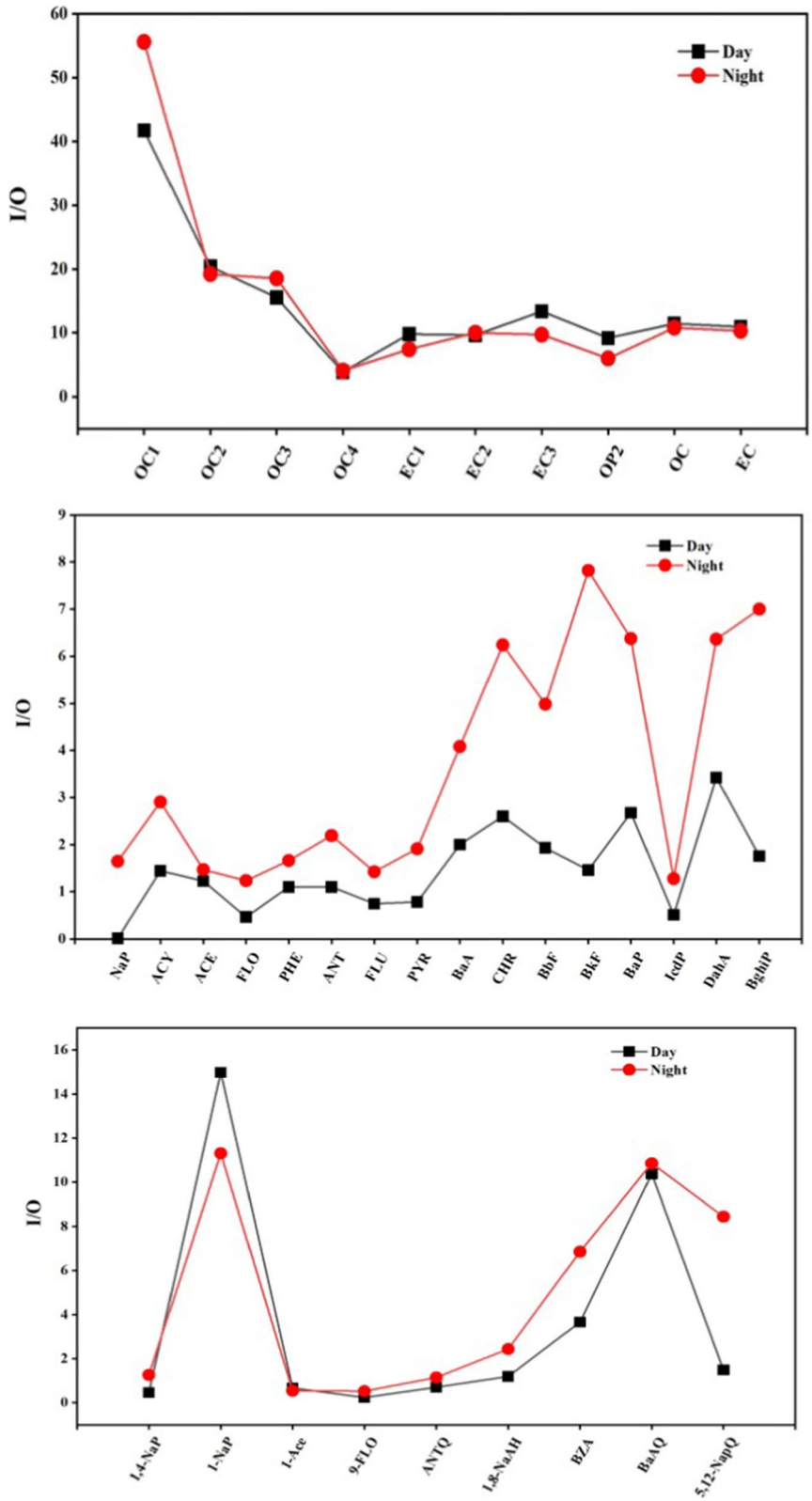


**Fig. 3** The I/O ratios for PAHs and OPAHs species during day and night

reflected that the indoor coal combustion and biomass burning really caused high pollution levels for indoor and then affected the outdoor air. There have no obvious differences between days and nights.

The I/O ratios of the 16 PAHs and 9 OPAHs indicated irregular trends. Except I/O ratios for NaP, FLO, FLU, PYR, and IcdP during daytime were less than 1, all the other I/O ratios of PAHs were greater than 1, showing that indoor source affects the outdoor pollution. All the I/O ratios at night were higher than in daytime, especially for the high molecular (5-ring and 6-ring) PAHs. The I/O ratios for 5-ring and 6-ring PAHs were from 0.51 to 3.4 in daytime and from 1.4 to 8.7 at night, respectively. This might due to the biomass burning and coal combustion in indoor for heating emitted more higher molecular PAHs (BkF, BbF, BghiP, BaP) at night.

The I/O ratios of the 9 OPAHs during days and at nights had the similar trends, and there are no obvious differences between days and night. The I/O ratios of 1-NaP were the highest (15.0 and 11.3 at day and night, respectively) followed by BaAQ (10.4 and 10.8 at day and night, respectively); these two were much higher than the other OPAHs congeners. The lower molecular OPAHs (1-NaP and 1-Ace) displayed higher I/O ratios in day, and the most of OPAHs I/O ratios were higher during night suggesting that OPAHs could pose toxic effects during night time. The higher ratio of OPAHs during night might be local mix of primary sources (Lammel et al., 2020); sampling regions due to many factors, such as differences in population density, life style, or meteorological conditions (Wang et al., 2022a) and the relative contributions of primary and secondary sources of OPAHs vary greatly between different sampling sites and periods (Li et al., 2015).

## Health risks assessment

The detailed calculation progress for incremental lifetime cancer risks (ILCRs) and relative parameters used in this study is displayed in Supplementary information. Previous study has discussed the effects of the parameters on the ILCR (Wang et al., 2022a). The results of the calculated ILCR via inhalation pathway for $PM_{2.5}$-PAHs are displayed in Fig. 4 and Figure S4. The previous studies have suggested that when the ILCR value is $\leq 10^{-6}$, the cancer risk is negligible; that between $10^{-6}$ and $10^{-4}$ designates a potential risk; when it is $\geq 10^{-4}$, there is a serious health risk (Xia et al., 2013).

The inhalational ILCR values of PAHs in indoor and outdoor for different groups showed different trends. For different age groups, the ILCR has decreasing trends by adults > children > seniors > adolescents. ILCR value was similar for boys, male

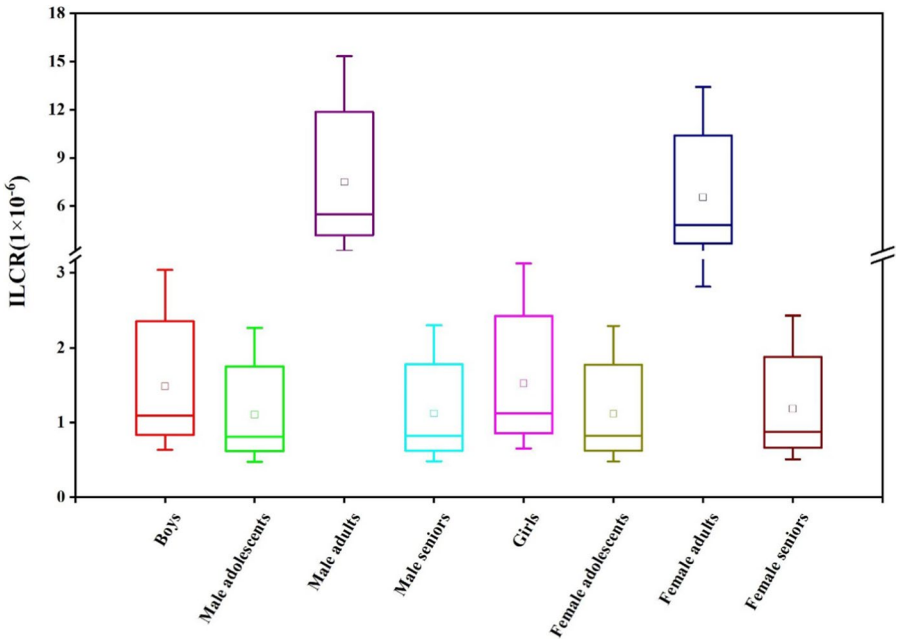


**Fig. 4** The ILCR assessment of PAHs for different groups

adolescents, male senior, girls, female adolescents, and female seniors (Fig. 4). Except for adults, the other groups of males were lower than the females. This is because of the differences of the body weight, inhalation rate and exposure duration between male and female. This result was in accordance with a study conducted in Shenzhen (Sun et al., 2015) and five urban cities of Zhejiang Province, China (Mo et al., 2019). The cancer risk reported in this study was lower as compared for the carcinogenic risk of housewives in northwest China (He et al., 2022). It needs to be added that according to the study by Buonanno et al. (2015), most toxicity (> 90%) in $PM_{2.5}$ contributed from the ultrafine particles (Dp < 0.1 μm), which have very large surface area to volume ratio (Pacitto et al., 2018). ILCRs for all groups in indoor were higher than $10^{-6}$ in this study, though we did not investigate other exposure pathways, such as ingestion and dermal exposure. The actual cancer risk may be higher than the levels reported in the present study. Therefore, changes to the domestic solid fuels and heating methods adopted in winter in this area are urgently required.

## Conclusion

In the present study, we were interested to see the impact of coal and biomass burning on $PM_{2.5}$ pollution and associated health risk in indoor and outdoor environment from Songyuan, China. The results concluded that $PM_{2.5}$ and major chemical components such as OC, EC, PAHs, and OPAHs showed much higher concentrations in indoor than in outdoor environment, suggesting the biomass burning and coal combustion for cooking and heating in indoors in this area could emit large amount of pollutants that could pose great health threat to people. The I/O ratios of these components showed different values during day and night, which might due to the emission and the secondary formation. The ILCRs for all groups during the sampling periods were higher than $10^{-6}$, revealing that there was potential risk to different age gender in Songyuan, China.

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**Authors' contributions** GM analyzed the data and prepared the paper; XL implemented the research; JW designed the experiment and reviewed and edited the manuscript; ZD and ML revised the manuscript; XL and LW provided the sampling equipment; YH and JC supported sources. All authors contributed to measurements, discussed results, and approved the manuscript.

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**Declarations**

**Competing interests** The authors declare that they have no conflict of interest.

**Ethics approval** Not applicable.

**Consent to participate** Not applicable.

**Consent to publish** Not applicable.

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