

Concentrations, particle-size distributions, and indoor/outdoor differences of polycyclic aromatic hydrocarbons (PAHs) in a middle school classroom in Xi'an, China

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Abstract Polycyclic aromatic hydrocarbons (PAHs) attached to particulate matter can affect respiratory health, especially the health of children, but information on the air quality in schools is generally lacking. This study investigated the PAH concentrations in a naturally ventilated classroom in Xi'an, China, from 16 to 31 May 2012. Particulate PAH concentrations were measured for samples collected on five-stage cascade impactors deployed inside the classroom and outside. PM_{2.5}-bound PAH concentrations were 53.2 ng m⁻³ indoors and 72.9 ng m⁻³ outdoors. PAHs attached to very fine particles (VFPs) accounted for ~70 % of the total PAHs. The PAH concentrations indoors were affected by the students' activities,

cleaning, and smoking, while outdoors, the main sources were motor vehicle emissions and contaminated road dust. Particle-bound PAHs infiltrated the classroom through open windows, but the activities of the students and staff were also associated with an increase of PAHs attached to particles larger than 1.0 μm, most likely through resuspension. Cycles in the sources led to PAH concentrations 2–3 times higher on weekdays compared to weekends, both indoors and outdoors. PAH toxicity risks inside the classroom were substantially lower than those outdoors, and the highest risks were associated with VFPs.

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Introduction

In urbanized regions, people spend roughly 80–90 % of their time indoors (Castro et al. 2011); therefore, indoor air quality (IAQ) is a major health concern (Pope et al. 2002). Children are especially at risk from air pollutants because their respiratory systems are not yet fully developed, making them more susceptible to respiratory pathologies than adults (Bennett and Zeman 1998; Kulkarni and Grigg 2008). Days at school amount to 175–220 days per year across the world, and the duration of a school day typically ranges from 5 to 8 h (INCA 2009; Almeida et al. 2011). Particulate matter (PM) concentrations in schools are influenced by numerous factors, including the numbers of students and staff; their types of activities (e.g., physical, cleaning, and teaching activities); flaking of building paint and other materials; the design and operation of ventilation and air filtration systems; and the sources and types of emissions (Dockery and Spengler 1981; Yu and Crump 1998; Lee and Chang 2000; Lai 2002; Daisey et al. 2003; Wolkoff et al. 2006; Heudorf et al. 2009; Guo et al. 2010; Alshitawi and Awbi 2011; Zhang and Zhu 2012). Numerous studies have demonstrated that elevated PM concentrations in classrooms can have significant adverse effects on children's health and overall performance (Mendell and Heath 2005; Tran et al. 2012).

The size of PM is important for human health because the particles' aerodynamic properties largely determine whether they will enter and be deposited in the respiratory system; this in turn has a major influence on the particles' potential for harmful effects (Gemenetzi et al. 2006). Coarse particles (PM with aerodynamic equivalent diameters of $D_p > 2.5 \mu\text{m}$) are typically primarily from low-temperature combustion sources, mechanical grinding processes, and physical transportation. Fine particles or $\text{PM}_{2.5}$ (PM with $D_p \leq 2.5 \mu\text{m}$) primarily originate from high-temperature combustion sources, such as motor vehicle emissions, coal and biomass burning, industrial processes, and complex chemical reactions involving gas-phase precursors (Seinfeld and Pandis 2006; Remer et al. 2009). $\text{PM}_{2.5}$ can be inhaled more deeply into the lungs than coarser

particles, and $\text{PM}_{2.5}$ also can penetrate through the lung lining into the circulatory system. Fine PM also remains suspended for longer periods of time than coarse PM; therefore, fine PM can be transported over longer distances compared with coarser particles. The implication of these differences is that fine particles are likely to play a more important role in affecting human health than coarse ones (Pope and Dockery 2006; Shen et al. 2009a, 2011).

In addition to physical factors, the chemical composition of PM, too, can have a strong influence on the health effects of aerosol populations. However, only limited data are available concerning the chemical properties of PM in schools; for example, data have been reported for water-soluble ions in Greek, German, and American schools (John et al. 2007; Diapouli et al. 2008; Fromme et al. 2008); for organic carbon (OC) and elemental carbon (EC) in German, Portuguese, and American classrooms (Ward et al. 2007; Fromme et al. 2008; Pegas et al. 2012); and for trace elements and metals in French, Dutch, and American schools (Janssen et al. 2001; Ward et al. 2007; John et al. 2007; Tran et al. 2012).

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic compounds of special interest to health scientists because they are ubiquitous in the environment and some can be cytotoxic, mutagenic, teratogenic, or carcinogenic for humans (WHO 1998). PAHs originate from a myriad of natural and anthropogenic sources, especially incomplete combustion of organic matter including coal and oil (Shibamoto 1998; Schauer et al. 2003; Okuda et al. 2010; Tobiszewski and Namiesnik 2012; Liu et al. 2013). To the best of our knowledge, no information on PAH concentrations in Chinese schoolrooms has been published in the open scientific literature. The purpose of this study was to determine the PAH concentrations for size-separated particles collected inside and outside a classroom and to assess their sources and dynamics, with the ultimate goal of assessing how these factors relate to the toxicity risks for school children.

Materials and methods

Site description

The middle school investigated for the study is located in the southwestern part of urban Xi'an, China. The

school environment is not directly influenced by the industrial emissions known to impact the atmosphere of Xi'an (Cao et al. 2005, 2007; Shen et al. 2008, 2009b, 2014). The closest main road is more than 200 m away, but vehicular traffic is a source of potential concern, as are the emissions from neighboring residential communities that are emitted every day, including weekends.

Samples were collected inside a school classroom and outdoors from 16 to 31 May 2012. The indoor sampling equipment was set up in a first-floor classroom that was occupied by 50 students aged 12–14 years during school hours: approximately from 8:00 to 11:30 a.m. and again from 1:30 to 5:00 p.m. local time during weekdays. Indoor sensors were located on a desk 1.2 m above the floor in the back of the classroom. An identical sampling apparatus was located outdoors, on the rooftop of the same teaching building, approximately 10 m above the ground and 10 m away from the indoor sampling site, horizontally. The classroom occupancy records, reported daily by the class adviser in a written log, showed a 78–90 % classroom occupancy rate. Cleaning was achieved daily by students during class hours. By contrast, little human activity took place in the school when it was not in session. The classroom was well ventilated as five large windows (1.9 m × 1.8 m) were open during school hours but closed after school ended (~5:00 pm). The classroom was a non-smoking area; however, smoking by older students or school staff could not be completely prevented.

PM sample collection

The PM samples were collected using a Sioutas Personal Cascade Impactor (SKC Inc., CA, USA), which operated at a flow rate of 9 l min⁻¹ and separated the aerosol particles into five size fractions. Pre-fired (780 °C, 3 h) 25-mm quartz fiber filters (QM/A[®], Whatman Inc., UK) were used for the first four stages: (stage A) PM_{>2.5}, also defined as coarse particles; (stage B) PM_{2.5–1.0} (D_p 1.0–2.5 μm); (stage C) PM_{1.0–0.5} (D_p 0.5–1.0 μm); (stage D) PM_{0.5–0.25} (D_p 0.25–0.5 μm). The fifth stage, denoted (E), collected particles with $D_p < 0.25$ μm, defined hereafter as very fine particles (VFPs). Stage E was adapted downstream from the cascade impactor itself using an inline 47-mm filter holder (NILU Innovation, Norway) and captured particles on a pre-fired (780 °C, 3 h) 47-mm quartz fiber filter (QM/A[®], Whatman Inc., UK). The

sum of all stages of the impactor is taken to represent the total suspended particle load or TSP.

The two sampling units used indoors and outdoors were compared, and the mass results did not display any bias. Sampling intervals were controlled by electrical timers, which allowed both the indoor and outdoor samplers to run for 30 min every hour, that is, approximately 12 out of 24 h. All PM-loaded filters were changed daily at ~8:00 a.m. local time before the beginning of the school day. Field blanks for each size fraction were collected separately for weekdays and weekends, and all reported results were corrected for backgrounds. 60 indoor and 60 outdoor samples were collected in total, excluding field blanks.

Indoor and outdoor meteorological data

TSI Q-Trak instruments (TSI Inc., USA) were operated inside and outside the classroom for real-time monitoring of temperature (T), relative humidity (RH), and CO₂; measurements for these variables were made simultaneously every 5 min. All instruments were calibrated and inter-compared for 48 h before and after the sampling period. Resolutions, precisions, and measurement ranges of the analyses were as follows: 0.1, ±0.6, 0–60 °C for T; 0.1, ±3.0, 5–95 % for RH; 1, ±50, 0–5,000 ppm for CO₂.

PM gravimetric analysis

PM samples were analyzed gravimetrically for mass concentrations with a Sartorius ME 5-F electronic microbalance (sensitivity ±1 μg, Sartorius, Germany). Filters were weighed before and after sampling after equilibration for 24 h at 20–23 °C and RH of 35–45 %. The absolute errors between duplicate weights were <0.015 mg for 47-mm blank filters, 0.020 mg for 47-mm sample filters, <0.010 mg for 25-mm blank filters, and 0.015 mg for 25-mm sample filters. The exposed samples and field blanks were tightly air sealed and stored in a refrigerator at less than –20 °C until chemical analysis.

Analysis of PAHs in PM

The in-injection port thermal desorption (TD)-gas chromatography/mass spectrometry (GC/MS) procedure used in our study is characterized by the high

sensitivity and short sample preparation time (<1 min), and it effectively avoids contamination from solvent impurities (Ho and Yu 2004; Chow et al. 2007; Van Drooge et al. 2009; Ho et al. 2008, 2011). Briefly, one-third of the 25-mm aerosol-loaded filters from stages A through D and 0.1–1.0 cm² aliquots from the 47-mm sample filters from stage E were used for PAH analyses. The filter strips were cut into small pieces and inserted into a TD tube, which had the same dimensions as the GC/MS injector liner. The temperature of the injector port was lowered to 50 °C before analysis; it was then raised to 275 °C for desorption in a splitless mode while the GC oven temperature was kept at 30 °C. After the injector temperature reached 275 °C, the GC separation and collection of mass spectra began. Limits of detection (LODs) for the 16 PAHs measured in this study can be found in Table 1. For discussion purposes, the sum of the concentrations of all measured PAHs is represented as \sum PAH, and \sum PAH-TSP is used to denote the sum of all quantified PAHs summed over all stages of the cascade impactors, \sum PAH-PM_{2.5} represents the sum of the PAHs in PM_{2.5}, and so forth.

In a previous study (Ho et al. 2008), replicate analyses of the same suite of PAHs as in the present study showed relative standard deviations (SD)

<10 %, with the majority <5 %. The accuracies for the PAH measurements, determined with NIST standard reference material SRM 1649a (urban dust), were within ± 5 % of the certified values. Comparisons of the TD-GC/MS and traditional solvent extraction (SE)-GC/MS method for ambient air samples have shown agreement within 11 % for nonpolar organic compounds (Ho et al. 2008). More information about the TD-GC/MS method used here can be found in Ho et al. (2008, 2011), Xu et al. (2013), and Cao et al. (2013).

Results and discussion

PAH concentrations and indoor/outdoor differences

The average PAH concentrations for each of the five stages of the impactors deployed inside the classroom and outside, as well as data on the temperature, relative humidity, and CO₂, are summarized in Table 2. Indoors, \sum PAH-TSP ranged from 4.9 to 162.4 ng m⁻³ and averaged (arithmetic mean) 60.0 ng m⁻³. In comparison, the range outdoors was 14.0–173.3 ng m⁻³ with an average of 77.7 ng m⁻³ (Fig. 1). Similarly, the mean concentrations of PAHs on particles with $D_p < 2.5$ μ m, that is, \sum PAH-PM_{2.5}, and the associated SDs were 53.2 ± 21.8 ng m⁻³ indoors and 72.9 ± 35.7 ng m⁻³ outdoors. Therefore, most of the PAHs were associated with the fine particle fraction, and, on average, PM_{2.5}-PAHs accounted for 88.7 % of the \sum PAH-TSP indoors and 93.7 % outdoors. The PAHs on VFPs (stage E) made up the largest proportions of the PAHs both indoors and outdoors by far: \sum PAH-VFP accounted for 63.7 % of the \sum PAH-TSP indoors and 76.1 % outdoors (Table 3).

The indoor/outdoor (I/O) concentration ratio as used here is a convenient indicator of the difference between the indoor and corresponding outdoor PAH levels. Hence, the ratios can be viewed as a measure of the relative intensities of the indoor versus outdoor sources (Huang et al. 2007; Crist et al. 2008). An I/O ratio greater than 1 indicates that the indoor sources were stronger than those outdoors, but on the contrary, if the indoor sources were weaker, the ratios would be less than unity (Crist et al. 2008). As shown in Table 2, the I/O ratios for \sum PAH-TSP and \sum PAH-PM_{2.5} were

Table 1 Limits of detections (LOD) for the TD-GC/MS for the analysis of PAH species in this study

PAHs (abbreviation)	Rings	LODs (ng sample ⁻¹)
Fluorene (FLO)	3	0.560
Phenanthrene (PHE)	3	0.165
Anthracene (ANT)	3	0.115
Fluoranthene (FLU)	4	0.061
Pyrene (PYR)	4	0.073
Benzo[a]anthracene (BaA)	4	0.025
Chrysene (CHRY)	4	0.041
Benzo[b]fluoranthene (BbF)	5	0.063
Benzo[k]fluoranthene (BkF)	5	0.055
Benzo[a]fluoranthene (BaF)	5	0.059
Benzo[e]pyrene (BeP)	5	0.026
Benzo[a]pyrene (BaP)	5	0.026
Perylene (PERY)	5	0.026
Indeno[1,2,3-cd]pyrene (IcdP)	6	0.036
Dibenzo[a,h]anthracene (DahA)	5	0.034
Benzo[ghi] perylene (BghiP)	6	0.066

Table 2 Summary of \sum PAH concentrations by cascade impactor stage and ancillary data during the sampling period

	Indoor (I)	Outdoor (O)	I/O ratio
Impactor stage	\sum PAH (ng m ⁻³) ^a		
Stage A: $D_p^b > 2.5 \mu\text{m}$	6.8 (±8.1)	4.9 (±4.5)	1.4 (±0.2)
Stage B: $D_p: 2.5\text{--}1.0 \mu\text{m}$	4.6 (±4.2)	2.9 (±2.1)	1.6 (±0.2)
Stage C: $D_p: 1.0\text{--}0.5 \mu\text{m}$	5.3 (±4.0)	4.4 (±5.7)	1.2 (±0.3)
Stage D: $D_p: 0.5\text{--}0.25 \mu\text{m}$	5.1 (±4.2)	6.5 (±5.8)	0.8 (±0.1)
Stage E: $D_p < 0.25 \mu\text{m}$	38.2 (±32.9)	59.1 (±54.2)	0.6 (±0.1)
PM _{2.5} (stages B + C + D + E)	53.2 (±21.8)	72.9 (±35.7)	0.7 (±0.3)
\sum Stages (stages A + B + C + D + E)	60.0 (±20.0)	77.7 (±32.3)	0.8 (±0.0)
Number of samples	60	60	60
Ancillary data			
Temperature (°C)	25.7 (±1.4)	24.2 (±4.9)	1.1 (±0.2)
Relative humidity (%)	49.6 (±7.0)	54.3 (±20.4)	0.9 (±0.2)
CO ₂ (ppm)	805 (±97)	412 (±58)	2.0 (±0.3)

^a Arithmetic mean (±SD) concentration

^b D_p mean is the aerodynamic equivalent diameter

0.7 and 0.8, respectively, suggesting greater impacts from outdoor PAH sources; these sources include automobile exhaust emissions, incomplete combustion of fossil fuels from heat or power generation, and fugitive emissions from industrial processes, motor

vehicles, and construction sites (Shibamoto 1998; Schauer et al. 2003).

Further examination of the data shows that the I/O ratios tended to increase as particle size increased. Indeed, I/O ratios >1 were observed for stages A, B, and C, suggesting a possible transfer of PAHs bound to PM larger than 0.5 μm from outdoors into the classroom. On the other hand, the I/O ratios for the smaller particles collected on stages D and E were <1, especially for the VFPs on stage E (0.6 ± 0.1), which suggests limited transfer of submicron particles from outdoors to indoors. Similar patterns were observed for PM mass concentrations; that is, the I/O ratios for PM mass were 1.7 for stage A, 3.0 for stage B, 1.5 for stage C, and then decreased to 1.3 for stage D and 1.0 for stage E. These I/O comparisons show that the PM mass loadings indoors were higher for all size fractions than those outdoors, but especially for particles larger than those collected on stage C (0.5–1.0 μm). This raises the possibility that fine particles and VFP entered the classroom with different transfer efficiencies or by different pathways than those for larger particles (Thatcher and Layton 1995; Tippayawong et al. 2009; Tiwari et al. 2013).

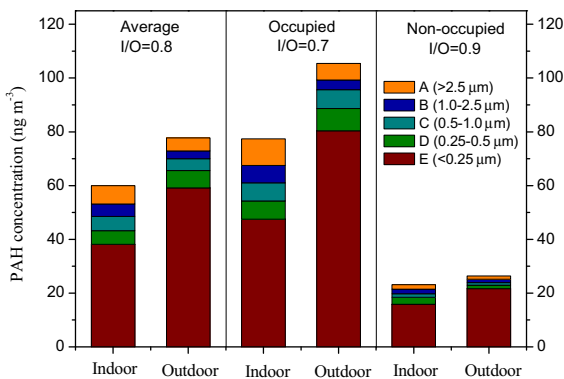


Fig. 1 Total measured PAH (\sum PAH) concentrations for cascade impactor stages A to E deployed in the school classroom and outdoors. Results are shown as the average for the entire sampling period, occupied period (weekdays), and unoccupied period (weekends)

Table 3 Percentages of total PAHs on total suspended particles on individual cascade impactor stages for indoor and outdoor samples

	Stage A (PM _{>2.5})	Stage B (PM _{2.5-1.0})	Stage C (PM _{1.0-0.5})	Stage D (PM _{0.5-0.25})	Stage E (PM _{<0.25})
Indoor (%)	11.3 ^a	7.7	8.9	8.4	63.7
Outdoor (%)	6.3	3.6	5.6	8.4	76.1

^a \sum PAH on stage A/ \sum PAH on total suspended particles (stages A + B + C + D + E)

As PAH levels in classroom environments have not been reported, here we compare the PAH concentrations from our study with those from selected reports for other indoor environments. The classroom \sum PAH concentrations found in our study were much lower than the levels reported from ten homes in north-central India sampled from 2006 to 2007 (Masih et al. 2010). For instance, the \sum PAH-TSP average for the classroom in our study (60 ng m^{-3}) is far less than the average PAH concentrations of $1,946.8 \text{ ng m}^{-3}$ in a kitchen, $1,666.8 \text{ ng m}^{-3}$ in a living room, and $1,212.6 \text{ ng m}^{-3}$ outdoors at an urban site in New Delhi. On the other hand, the \sum PAH-PM_{2.5} levels in the classroom in our study (53.2 ng m^{-3}) were slightly higher than those measured in Guangzhou, China, in 2002 (Li et al. 2005). There the PAH levels in PM_{2.5} ranged from 14.2 to 77.9 ng m^{-3} (mean 43.5 ng m^{-3}) in residential buildings and $15.8\text{--}84.8 \text{ ng m}^{-3}$ (mean 47.3 ng m^{-3}) outdoors; the PAH I/O ratios in the Guangzhou study were close to unity (0.9–1.1).

Student occupancy effect

This section describes the influence of the students' activities on the different PM size fractions in the classroom. The concentrations of PAHs in the occupied classroom were higher than in the unoccupied classroom, and particle-size distributions of the PAHs also were different (Fig. 1). Further inspection of the data shows that the indoor \sum PAH-TSP levels varied from 19.0 to 162.3 ng m^{-3} (77.4 ng m^{-3} on average) on weekdays, but were considerably lower, between 4.9 and 41.4 ng m^{-3} (23.1 ng m^{-3} on average), on weekends.

The average indoor CO₂ concentration during the sampling study was 805 ppm, and this was nearly twice the average concentration outdoors (Table 2). Most of the indoor CO₂ measurements were found to be within the 1,000 ppm limit (8-h average) recommended by the World Health Organization (WHO 2000). The air renewal rate was evaluated by comparing the indoor CO₂ values measured when the classroom was occupied with the concentrations when the room was not occupied but under the same ventilation conditions. The CO₂ concentration from 11:00 to 11:30 a.m., that is, when students were in the room, was 1,238 ppm, but the concentration dropped to 774 ppm from 11:30 to 12:00 a.m. when the

students left the room (lunch time). This is equivalent to a decrease of more than one-third of the starting concentration in 1 h. The results also show the average CO₂ I/O ratio was 2.2 during school hours (windows open) compared with 1.8 after school (windows closed) on weekdays and 1.6 (24-h average, windows closed) on weekends, respectively. The latter value suggests that some residual CO₂ remained in the classroom probably because of the poor ventilation when windows were closed, especially on weekends.

The PAH levels on weekdays, both inside the classroom and outdoors, were at least twice those on weekends. Indoors, this could be explained by two factors. First, PAHs attached to road dust and other particles may have infiltrated into the room through the doors and windows, or they may have been brought inside by the children. These particle-bound contaminants also may have been kept in suspension or resuspended by activities of the students and staff. More PM brought into the classroom by students would result in higher aerosol mass loadings on weekdays, and indeed, the indoor TSP mass concentrations on weekdays ($260.2 \text{ } \mu\text{g m}^{-3}$) were roughly twice those on weekends ($121.8 \text{ } \mu\text{g m}^{-3}$). The second and probably more important factor that likely influenced the weekday/weekend difference in PAH concentrations was the heavier motor vehicle traffic during weekdays. In fact, the concentrations of BghiP, which is a PAH marker for motor vehicle exhaust emissions (Shen et al. 2010), decreased dramatically from weekdays to weekends. The BghiP concentrations outdoors during the weekdays were nearly 20 times those on weekends, while the difference indoors was close to tenfold. Elevated BghiP concentrations on weekdays are consistent with the heavier traffic, which would lead to the infiltration of more contaminated outdoor air during the week, as the windows were open during class hours. The higher outdoor \sum PAH concentrations on weekdays versus weekends also can be explained by the greater vehicular emissions during the work week.

The students' physical activities during free periods and physical education classes and the cleaning of the school and playground also may have influenced the indoor/outdoor differences in PAH concentrations. On weekdays, the outdoor \sum PAH-TSP loadings were higher than those indoors: the average I/O \sum PAH-TSP ratio during the school week was 0.7, but on the weekends the I/O ratio was higher, 0.9. The latter is

consistent with the incomplete exchange of air on weekends that was deduced from the CO₂ data above and also with perturbations caused by actions of the students and school staff inside the school. Indeed, PAHs on the large particle stages (stages A and B) of the indoor sampler were significantly higher than those outdoors during the week (I/O for stages A and B were both ~1.7), suggesting that activities in the classroom or elsewhere in the school may have caused the resuspension of previously deposited contaminated particles.

In contrast, the PAHs on the smaller particle stages of the cascade impactor deployed outdoors (stages C to E) during the weekdays were higher than on the indoor sampler (I/O ratios for these stages were ~0.8). The elevated PAH concentrations on stages C, D, and E, especially outdoors, can be attributed to traffic emissions, even though the closest main road was more than 200 m from the sampling site. Other combustion sources, including residential emissions, especially those from cooking, also may have contributed to the higher outdoor PAH concentrations. As mentioned above, the I/O ratios for PM were all ≥1, and the I/O comparison for PM mass loadings showed higher ratios for coarse particles than for the fine and VFP fractions. One possible explanation for this is that the finer particles are less likely to infiltrate the classroom than coarser PM, but there are other possible explanations. Indeed, it is likely that activities in the school led to increases in both PM and PAHs due to the resuspension

of contaminated coarse particles, but unidentified coarse particle sources indoors may also have contributed to the I/O differences observed.

PAH ring-dependent characteristics

The size distributions of 3-, 4-, 5-, and 6-ring PAHs in the classroom and outdoors display similar trends (Fig. 2). The average concentrations indoors for the 3-, 4-, 5-, and 6-ring PAHs in TSP were 35.6 ± 8.0, 6.8 ± 1.5, 14.5 ± 4.4, and 4.7 ± 1.4 ng m⁻³, respectively, while the corresponding outdoor concentrations were 45.1 ± 13.8, 9.2 ± 2.8, 20.7 ± 6.9, and 6.5 ± 2.1 ng m⁻³. The 3- and 5-ring PAHs were the most abundant, accounting for 55–60 % and 20–25 % of ∑PAH, respectively. As Fig. 2 shows, the size-separated masses of 3-, 4-, and 5-ring PAHs displayed two modes both inside and outside the classroom: the primary one was below 0.25 μm (stage E), while the secondary one was above 2.5 μm (stage A). The 6-ring PAHs were concentrated in the very fine mode, and only small quantities of 6-ring PAHs were found in PM > 1 μm. Allen et al. (1996) have shown that most 3-ring PAHs are associated with fine particles when they are initially emitted, but the size distribution can shift upward because of volatilization, which is a function of the surface area-to-volume ratios of the particles. In addition, as the molecular weight of the PAHs increases, their volatility decreases, and this limits their transfer to coarser particles by this

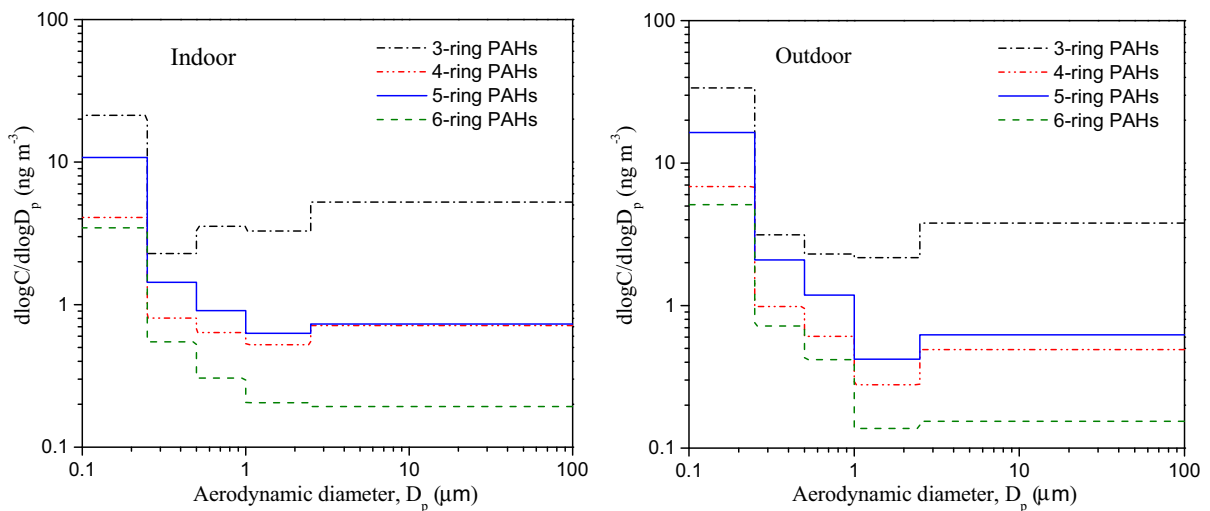


Fig. 2 Size distributions of 3-, 4-, 5-, and 6-ring PAHs inside the classroom and outdoors

mechanism. This appears evident in our data for the 4-ring PAHs and is even more pronounced for those made of 5- and 6-rings.

Figure 3 plots the I/O ratios for 3-, 4-, 5-, and 6-ring PAHs as a function of the cascade impactor particle size cuts. The average I/O ratios for 3-ring PAHs were 0.6, 0.7, 1.5, 1.5, and 1.4 for the impactor stages E, D, C, B, and A (increasing order of particle sizes), and the average was 1.2. For 4-ring PAHs, the ratios were 0.6, 0.8, 1.1, 1.9, and 1.5 (average 1.2). The I/O ratios for 5-ring PAHs on the same impactor stages were 0.7, 0.7, 0.8, 1.5, and 1.2 (average 1.0), and for 6-ring PAHs they were 0.7, 0.8, 0.7, 1.5, and 1.3 (average 1.0). The point here is that the I/O ratios for the 3- to 6-ring PAHs all displayed similar trends with respect to size. This also supports the discussion above that PAHs associated with coarser PM were more abundant in the classroom than outdoors. Indeed, the I/O ratios for 3- and 4-ring PAHs on stage C (D_p : 0.5–1.0 μm) were >1.0, while I/O ratios of 5- and 6-ring PAHs were <1.0.

The I/O patterns just discussed imply that 3- and 4-ring PAHs may have had some indoor sources for the PM captured on stage C; these include the shedding of skin cells, coughing, sneezing, and smoking by persons in the building (Slezakova et al. 2009). In addition, the I/O differences can be explained by the fact that 3- and 4-ring PAHs (molecular weight, MW < 252) exist in both the vapor and particulate phases, while the 5- and 6-ring PAHs (MW \geq 252) are mainly bound to particles (Bi et al. 2003; Ho et al. 2009; Tan et al. 2011). According to the gas/particle distribution theory, one

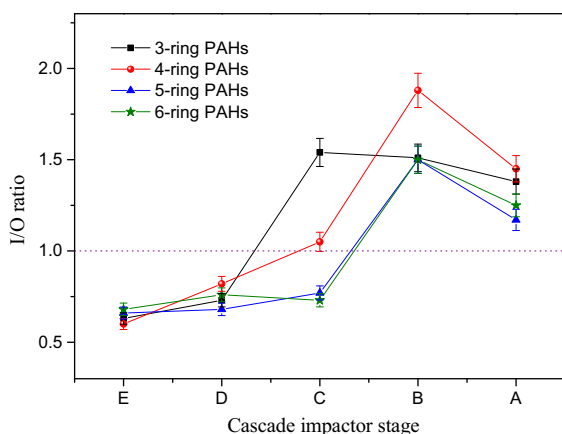


Fig. 3 Indoor/outdoor (I/O) ratios of 3-, 4-, 5-, and 6-ring PAHs for different particle size fractions in the school

would expect the 5- and 6-ring PAHs to be more stably bound to PM than 3-ring PAHs and for practical purposes irreversibly bound to the PM. On the other hand, 3-ring PAHs would be more likely to volatilize and then infiltrate into the classroom through the doors and windows. The average outdoor temperature during the study was 24.2 °C, and this was approximately 1.5 °C lower than the average temperature in the classroom (25.7 °C). Hence, aside from the possible effects of indoor PAH sources, the warmer temperatures indoors would vaporize more of the 3- and 4-ring PAHs compared with the outdoor environment.

Characteristics of specific individual PAHs

The relative contributions of the individual PAHs to \sum PAH for each of the five size fractions obtained with the cascade impactor are shown as pie charts in Fig. 4. FLO and PHE were the most abundant PAHs on all stages, and the next most abundant were BeP and ANT on stages A and B, BeP and BaP on stages C and D, and BeP and BbF for the VFP (stage E). The concentrations of BaP are of particular concern because that compound has been used as a general indicator of PAH carcinogenicity (Bi et al. 2003; Wang et al. 2006). The arithmetic mean BaP concentrations were 2.3 and 3.3 ng m^{-3} in the classroom and outdoors, respectively, and these values are 2–3 times the limits established in the air quality guidelines of the World Health Organization (1.0 ng m^{-3} , WHO 1998). With reference to the China National Ambient Air Quality Standards (CNAAQs, GB3095-2012), the BaP concentrations outdoors were comparable to the BaP standard (2.5 ng m^{-3} for the 24-h average of PM_{10}); however, the BaP levels indoors were twice the BaP limit in the China National Indoor Air Quality Standards (CNIAQS, GB/T18883-2002, 1.0 ng m^{-3} for the 24-h average in PM_{10}). The relative percentages of BaP to \sum PAH were inversely related to particle size, but the reasons for this remain unclear, and a break in the pattern was observed from stages D to E, both indoors and outdoors. The percentages of BaP to \sum PAH were 1.4, 2.4, 3.1, 6.1, and 4.0 % for stages A, B, C, D, and E indoors and 1.7, 2.6, 5.7, 6.6, and 3.8 % outdoors.

Figure 5 presents the average I/O ratios for specific PAHs, arranged in order of increasing molecular weight, for each of the five particle-size fractions obtained with the cascade impactors. For the coarser

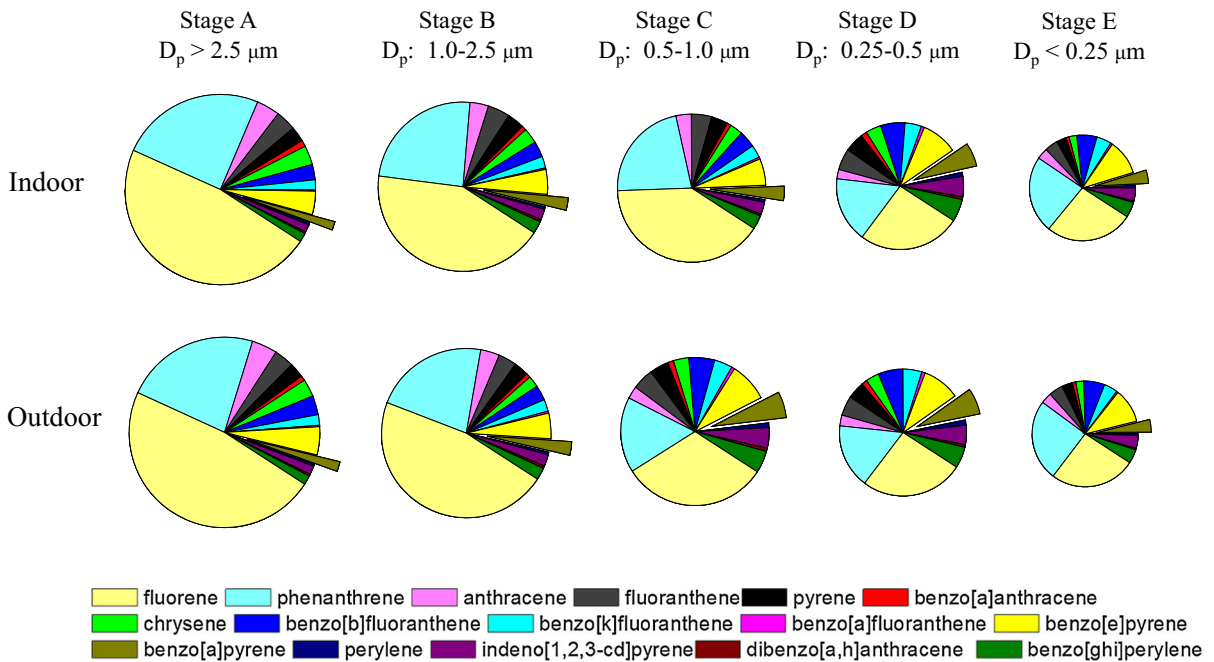


Fig. 4 Percentages of individual PAHs (%) for different particle size fractions collected in the classroom and outdoors (the sizes of the pie charts only roughly reflect the particles’ trend of decreasing sizes)

particles (stages A and B), the I/O ratios for all measured PAH species exceeded unity (1.1–1.7 for stage A and 1.1–2.1 for stage B). This is consistent with our earlier suggestion that the activities of students and staff may have led to the resuspension of coarse particles contaminated with PAHs. For stage C, the I/O ratios ranged from 0.7 to 1.6, and the values decreased as the molecular weight of the PAHs increased. In contrast, for stages D and E, the I/O ratios for the individual PAHs were all very similar and well below unity (~0.7 on average). One possible explanation for the lack of variability in stages D and E is that there were no significant indoor sources for particles captured on these impactor stages, which also supports the argument that the presence of students did not significantly change the PAH concentrations on stages D and E or the I/O ratios for the PAHs.

PAH toxicity risk

[BaP]_{eq} has been used as an indicator of the toxicity risks associated with exposure to PAHs (Petry et al. 1996; Bull and Collins 2013), and these risks can be estimated from the following Eq. (1):

$$\sum [\text{BaP}]_{\text{eq}} = \sum (C_i \times \text{TEF}_i) \tag{1}$$

In this formulation, $\sum [\text{BaP}]_{\text{eq}}$ aggregates the toxicity risk for all PAHs found in the different size fractions, C_i is the concentration of the compound of interest i (ng m^{-3}), and TEF_i is the toxicity equivalency factor (TEF) of compound i (Nisbet and Lagoy 1992; Petry et al. 1996).

The average $\sum [\text{BaP}]_{\text{eq}}$ values calculated in this way were $4.7 \pm 4.6 \text{ ng m}^{-3}$ inside the classroom and $6.6 \pm 7.0 \text{ ng m}^{-3}$ outdoors, suggesting considerably higher health risks for students when they are outdoors compared with indoors. Table 4 presents the [BaP]_{eq} calculations for the five particle size fractions, and it shows that the health risks from the particle-bound PAHs collected on stages A to D were low and probably negligible. Most notably, however, the [BaP]_{eq} values for stage E were 5–10 times higher than those of the larger particles, and 68 and 74 % of the [BaP]_{eq} could be ascribed to $\text{PM}_{<0.25}$ (VFP) both indoors and outdoors. Compared with larger particles, $\text{PM}_{<0.25}$ tends to remain suspended in the atmosphere longer, penetrate more deeply into the human body, and spread over longer distances (Yu and Yu 2011). Therefore, the PAHs associated with $\text{PM}_{<0.25}$ would

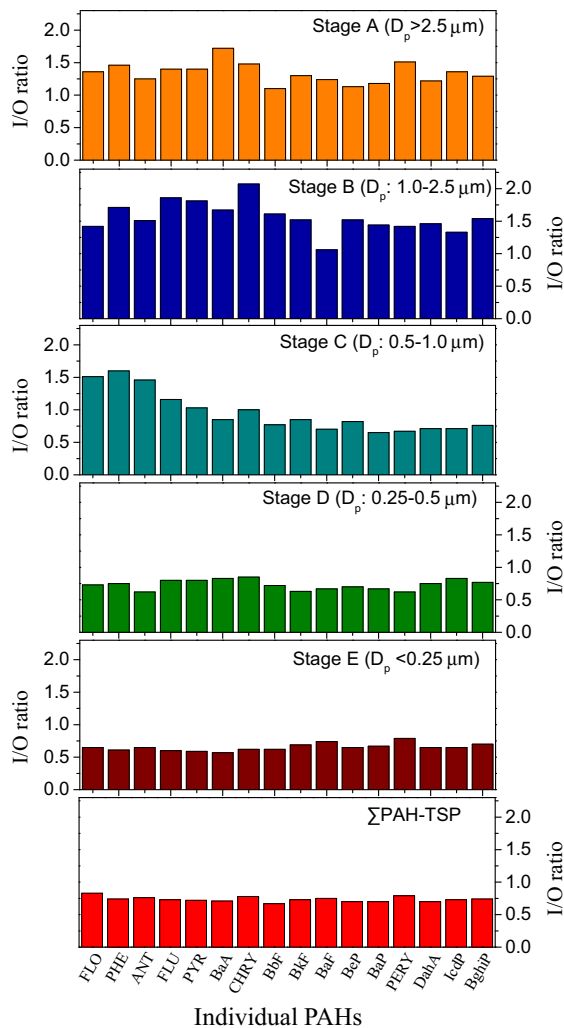


Fig. 5 Indoor/outdoor (I/O) ratios of individual PAHs collected on cascade impactor stages A to E and total suspended particles (Σ PAH-TSP, stages A + B + C + D + E)

likely pose the greatest toxicity risk and potentially do the greatest harm to the students' health.

Conclusions

This study provides detailed information on the concentrations and particle-size distributions of PAHs in a middle school in Xi'an, China. The PAH sources, dynamics, and toxicity risks were evaluated both inside a classroom and outside. The arithmetic mean concentrations of the total PAHs in TSP were 60.0 and 77.7 ng m^{-3} indoors and outdoors, respectively.

Table 4 PAHs equivalent toxicity ($[\text{BaP}]_{\text{eq}}$) in the school classroom and outdoors in relation to particle size fractions

Impactor stage	Size fraction (μm)	Indoor $[\text{BaP}]_{\text{eq}}$ (ng m^{-3})	Outdoor $[\text{BaP}]_{\text{eq}}$ (ng m^{-3})
A	$D_p > 2.5$	0.3 (± 0.3) ^a	0.2 (± 0.2)
B	$D_p: 2.5-1.0$	0.3 (± 0.3)	0.2 (± 0.1)
C	$D_p: 1.0-0.5$	0.4 (± 0.3)	0.5 (± 0.8)
D	$D_p: 0.5-0.25$	0.6 (± 0.6)	0.8 (± 0.7)
E	$D_p < 0.25$	3.2 (± 3.4)	4.9 (± 5.6)
Total	TSP ^b	4.7 (± 4.6)	6.6 (± 7.0)

^a Arithmetic mean (\pm SD)

^b TSP stands for total suspended particles

Approximately two-third of the total mass of PAHs was found at $\text{PM}_{<0.25}$, both indoors and outdoors, while the $\text{PM}_{2.5}$ fraction gathered 89 % of the Σ PAH mass indoors and 94 % outdoors.

Activities inside the school increased the PAH concentrations in particles larger than 1.0 μm . Ventilation was high when the windows were open, thus promoting air exchange. The I/O ratios for both Σ PAH-TSP and Σ PAH- $\text{PM}_{2.5}$ were less than unity, which indicates that PAHs were transported into the classroom from outside. The most likely sources of the contaminants were motor vehicle emissions and associated road dust. The PAH concentrations inside the classroom were clearly affected by activities from the students and staff as the I/O ratios were greater than unity for $\text{PM} > 1.0 \mu\text{m}$ in diameter. Other activities, including cleaning, as well as undetermined sources also may have affected the inside/outside differences and the particle-size distributions of the PAHs. Thermodynamic effects appeared to be a determinant of the PAH particle size distributions. Indeed, the volatilization of the light 3- to 5-ring PAHs led to bimodal distributions in both the classroom and outdoors. In contrast, the 6-ring PAHs, which are less volatile, displayed only one peak indoors. Moreover, ~ 70 % of the toxicity risks, calculated as $[\text{BaP}]_{\text{eq}}$, were attributable to the VFPs, and concentrations of PAHs indoors and their associated toxicity risks were apparently influenced by the ingress of contaminated outdoor air into the school. The indoor toxicity risks of PAHs on finer particles may have been reduced by the lower infiltration of contaminated fine particles, while

those for the coarser PM may have been affected by the activities of students themselves.

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