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ORIGINAL PAPER

Evaluation on exposures to particulate matter at a junior secondary school: a comprehensive study on health risks and effective inflammatory responses in Northwestern China

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Abstract Air pollutant measurement and respiratory inflammatory tests were conducted at a junior secondary school in Xi'an, Northwestern China. Hazardous substances including particulate matters (PMs), black carbon (BC) and particle-bounded polycyclic aromatic hydrocarbons (PAHs) were quantified both indoors and outdoors of the school. Source characterization with organic tracers and particle-size distribution demonstrated that the school's air was mostly polluted by combustion emissions from the surrounding environment. The evaluation of health assessment related to air quality was conducted by two methods, including potential risk estimation of air pollutants and direct respiratory inflammatory test. The incremental lifetime cancer risks associated with PAHs were estimated and were 1.62×10^{-6} and 2.34×10^{-6}, respectively, for indoor and outdoor fine PMs. Both the values exceeded the threshold value of 1×10^{-6}, demonstrating that the carcinogenic PAHs are a health threat to the students. Respiratory inflammatory responses of 50 students who studied in the sample classroom were examined with a fractional exhaled nitric oxide (FeNO) test, with the

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aid of health questionnaires. The average FeNO concentration was 17.4 ± 8.5 ppb, which was slightly lower than the recommended level of 20 ppb established by the American Thoracic Society for children. However, a wide distribution and 6% of the values were > 35 ppb, suggesting that the potentials were still high for eosinophilic inflammation and responsiveness to corticosteroids. A preliminary interpretation of the relationship between air toxins and respiratory inflammatory response demonstrated the high exposure cancer risks and inflammatory responses of the students to PMs in the city.

Keywords PM · BC and PAHs · Cancer risks · FeNO · Health assessment · Northwestern China

Introduction

BC and PAHs are two important organic components in airborne PM. BC is produced from incomplete combustions of wood, biomass and fossil fuel (Koelmans et al. 2006; Cao et al. 2009), potentially stimulating productions of reactive oxygen species (ROS) and cytokine-mediated inflammation after inhaled by human (Alessandrini et al. 2009). BC also links with cardiopulmonary diseases and other health impacts (Lin et al. 2011). PAHs are known as air toxins and carcinogenic substances which are generated naturally or by a variety of anthropogenic emissions (Schauer et al. 1996; Xu et al. 2013). Both BC and PAHs are tracers for source apportionment and act as indices of potential health risks (Turpin and Andrews 2000; Mauderly and Chow 2008; Yuan et al. 2015).

Rather than the molecular characterization of air pollutants, direct respiratory inflammatory response test can examine health impact related to air quality. Nitric oxide (NO) is produced in human lung after respiratory inflammation and is widely used as an indicator for respiratory diseases (Lin et al. 2011; van der Valk et al. 2012), particularly of lower airways inflammation (Alving et al. 1993; Annesi-Maesano and Dinh-Xuan 2016). A FeNO test is a standard method on measurement of NO level in exhaled airs of participants (ATS 2005). A questionnaire is often conducted synchronously to collect a wide range of medical condition and history of the participants (Zhao et al. 2008). The relationships between the results of FeNO test and the information from questionnaires can provide insights of any factors that contributed to the respiratory inflammatory and subsequently implicated on the health assessment (Xu et al. 2011).

Many health-related assessments on the PM have been conducted in workspace or residential units. The evaluations of air toxin on age groups of children and youths are particularly critical, because their juvenile respiratory systems are being developed (Kulkarni and Grigg 2008). According to the statistics from the International Review of Curriculum and Assessment (INCA), the numbers of school days was 175–220 days per year and the numbers of school hours was 5–8 hour per school day (INCA 2009). In China, because of rapid economy growth and capitalization, the PM levels have been continuously increasing in most mega- and developing cities which was approximately 1–2 orders of magnitude higher than those in other developed countries (van Donkelaar et al. 2010). The PM and its components can cause adverse effects on the health and overall performance of children (Mendell and Heath 2005).

In this work, we quantified the BC in total resuspended particulate (TSP) and PAHs in five-stage size-distributed PMs collected at a junior secondary school in Xi'an, which is the largest and the most representative urban city in Northwestern China. The potential cancer risks were estimated with the levels of the organic components. The direct respiratory inflammatory tests and questionnaires were conducted for a class of 50 students. The relationships between the potential cancer risks and the respiratory inflammatory responses were interpreted. The study provides integrity of the health assessment related to the air quality at the school.

Materials and methods

Site description

The study was conducted at a junior secondary school, where located in the southwestern region of urban Xi'an city from May 16, 2012, to May 30, 2012. A ground-floor classroom of 50 grade one students, who aged from 12 to 14 years, was selected for the health assessment. The site was approximately 100 m away

from a medium-traffic flow road. The school was surrounded by residential communities. The major local pollution sources including vehicle emission and residential cooking activity were observed by the on-site survey. Regional pollutant transportation might also affect the airs as the site was at downwind position. Synchronous outdoor samples were collected on the rooftop of the same teaching building, where was 10 m above the ground level.

Particulate matter collection

Indoor instrumental package was set up on a desk at 1.2 m above the ground in center of the classroom. The height is consistent with usual breathing zone for a youth student. A cascade impactor, which was equipped with a cyclone to separate PM into five different particle-size fractions, was operated at a flow rate of 9.0 L min^{-1} (SKC Inc., Eighty-Four, PA, USA). Four pieces of pre-fired (780 °C, 4 h) 25-mm-diameter quartz-fiber filters (QM/A®, Whatman Inc., Buckinghamshire, UK) were collected the $PM_{>2.5}$, $PM_{2.5–1.0}$, $PM_{1.0–0.5}$, $PM_{0.5–0.25}$ μm (referred as: A, B, C and D, respectively), in line with one piece of pre-fired (780 °C, 4 h) 47-mm-diameter quartz-fiber filter (QM/A®, Whatman Inc.) for the last fraction of $PM_{<0.25\ \mu m}$ (E). The sampling operation was controlled by an electrical timer, which ran for 12 hour per day, while the flow rate was continuously regulated to avoid any critical drift. All filter samples were exchanged in each day at 08:00 a.m. local time.

Online observations and ventilation rate in classroom

BC in TSP, consistent with the particle cutoff size of the total five fractions from impactor, was measured with aethalometers (AE-16, Magee Scientific, Berkeley, CA, USA) at a flow rate of 4 L min^{-1}. The PM was collected on a quartz-fiber filter tape, and the BC was quantified by the attenuation of transmitted light at a wavelength of 880 nm. The accuracy of factory-calibrated instruments was ± 2%. The transmittance in inverse megameters (M m^{-1}) was converted to the unit of nanogram per cubic meter (ng m^{-3}) according the recommended constant of 16.6 m^2 g^{-1} (Zhu et al. 2010).

Indoor air quality meters (Q-TRAK 7575, TSI Inc., Shoreview, MN, USA) were used to collect for real-time data of relative humidity (RH), temperature (T) and concentrations of carbon dioxide (CO_2) at a 5-min time interval. The instruments were calibrated regularly. The resolutions, precisions and measurement ranges for each component were: 0.1, ± 3.0, 5–5% for RH; 0.1, ± 0.6, 0–60 °C for T; and 1, ± 50, 0–5000 ppm for CO_2, respectively (Xu et al. 2015). The average RH, T and CO_2 were 49.6 ± 7.0 and 54.3 ± 20.4%, 25.7 ± 1.4 and 24.2 ± 4.9 °C and 805 ± 97 and 412 ± 58 ppm in classroom indoors and outdoors, respectively, during this study.

The ventilation rate, as expressed in terms of air changes per hour (ACH), in the classroom was evaluated with the changes in CO_2 concentration in classroom (Clements-Croome et al. 2008). With the regulation standard of GB/T 17226-1998 (GB/T 17226-1998 1998), the ACH should not be < 3.0 and < 4.0 unit per hour for an indoor area in primary and secondary school, respectively. In this study, the ACH was 6.0 unit per hour, demonstrating a satisfy infiltration and exchange of airs through the five large windows (each of 1.9 m wide × 1.8 m high) and two doors (each of 0.9 m wide × 2.0 m high) in the target classroom, which were frequently opened during school time (Clements-Croome et al. 2008; Xu et al. 2015).

Gravimetric analysis

The PM-loaded filters were analyzed gravimetrically for mass concentrations with a Sartorius ME 5-F electronic microbalance (sensitivity ± 1 μg, Sartorius, Gottingen, Germany) (Xu et al. 2015, 2016). The filters were weighed before and after sampling after equilibration at T of 20–23 °C and RH of 35–45% for 24 h. All sampled filter replicate weights were satisfied the error limit (0.020 mg) in this study. The exposed samples and field blanks were air-tightly sealed and stored in a refrigerator at < −4 °C until chemical analysis.

PAH analysis

In-injection port thermal desorption (TD) coupled with gas chromatography/mass spectrometry (GC/MS) used in this study is a convenient method to determine PAHs from an aerosol filter sample. The approach involves a short sample preparation time (< 1 min), avoids contamination from solvent

impurities and is highly sensitive (Ho and Yu 2004; Chow et al. 2007; Ho et al. 2008, 2011; Wang et al. 2015; Xu et al. 2013, 2015, 2016). Details of the analytical method have been described in our previous publications (Ho and Yu 2004; Ho et al. 2008, 2011). Briefly, filters were split in aliquots of different sizes (one-third of the filter for stages A to D and a punch of 0.5–1.0 cm^2 for stage E) subjected to the aerosol loading. The injector temperature was lowered to 50 °C before loading of the TD tube. Once the sample was exchanged into the injector, the temperature was raised to 275 °C for the analytes desorption in a splitless mode, while the GC oven temperature was kept at 30 °C. After the injector temperature reached 275 °C, the GC temperature oven program started, and the analytes were then separated through a capillary column and detected by the MS. Table S1 in supplementary information (SI) lists sixteen target PAHs and their minimum detection limits (MDLs). The quality assurance and quality control (QA/QC) procedures for the TD-GC/MS method have been shown in elsewhere (Ho and Yu 2004; Ho et al. 2008, 2011).

Health assessment methods

PAH cancer risk model

It is well documented that many PAHs are mutagenic and carcinogenic (Yuan et al. 2015). Their health risks can be expressed by measurement of BaP-equivalent ($[BaP]_{eq}$) concentrations (Petry et al. 1996). $[BaP]_{eq}$ was calculated with the concentration of that compound of multiplied by its toxicity equivalency factor (TEF) as shown in Eq. (1) (Yu et al. 2008):

$$\Sigma[BaP]_{eq} = \Sigma(C_i \times \ TEF_i) \quad (1)$$

where C_i and TEF_i are the concentration (ng m^{-3}) and TEF (Nisbet and Lagoy 1992; Yu et al. 2008) of the single target PAH of i, respectively. According to the US EPA cancer risk model (US EPA 2004, 2011), the ILCRs of PAHs via inhalation way was calculated as follows:

$$\mathrm{ILCR} = [(\mathrm{C} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED} \times cf)/(\mathrm{BW} \times \mathrm{AT})] \times \mathrm{SF} \times \mathrm{ADAF} \quad (2)$$

where C represents $\Sigma[BaP]_{eq}$ concentration from Eq. (1) in PM (ng m^{-3}). The exposure parameters are chosen for 11- to < 16-year-old children, equivalent to the range of ages for the students involved in this study (US EPA 2004, 2011; Yu et al. 2008; Ma et al. 2014). IR represents the amount of air that a child inhaled each day (15.2 m^3 day^{-1}). EF refers to the exposure frequency (180 days $year^{-1}$). ED is the exposure duration (5 years) and *cf* represents the conversion factor (10^{-6} mg ng^{-1}). BW stands for the body weight (31.8 kg) (actual measurement in this study). AT represents the average lifetime (70 × 365 days). SF represents cancer slope factor of BaP [7.3 (mg kg^{-1} $day^{-1})^{-1}$] (Letter Health Consultation 2008; US EPA http://www.epa.gov/iris/subst/0136.htm). Early life susceptibility had to be assumed for the children in this study, while an age-dependent adjustment factor (ADAF) of three was applied to the cancer slope factor, meaning that a threefold of exposures occurring between the ages of 2 and 16 years of age (US EPA 2011). For the cancer risk, the value of 1×10^{-6} is an internationally accepted precautionary or threshold value, above which the risk is unacceptable (US EPA 2006).

FeNO test and questionnaires

The FeNO test was conducted using a portable NIOX MINO® device (Aerocrine AB, Solna, Sweden). Before the test, a face-to-face interview was conducted with a small-scale questionnaire surveyed their recent conditions on respiratory health, medication and diet. Students who eat, drink and smoke within 1 h before the test will not meet the FeNO test requirement. All 50 students were eligible to participate in the test. In the test, the student sat down on a chair, rinsed the mouth with pure water and emptied the lungs by thorough expiration. Then, the student was invited to deeply inhale air through a disposable filter, which removes any NO content from ambient air. The air was then breathed out through the device at an exhalation rate of 50 ± 5 ml s^{-1} (Xu et al. 2011).

The full questionnaires were handed out to the 50 students who studied in the sample classroom and were fully responded (100% response rate). The questions were specifically designed for the junior secondary students and focused on their health conditions and general habits, including: (1) any respiratory noise or wheezing sound (i.e., sharp whistle); (2) dermatological symptoms; (3) nasal symptoms; (4) asthma symptoms; (5) family status; (6) childhood living environment; (7) current living environment;

(8) dining habits; and (9) current health status. The questionnaires were answered by their parents or legal guardians.

Results and discussion

PM levels

Table 1 lists the size-distributed PM masses indoors and outdoors. The mean TSP (sum of the five size-distributed fractions) was 211.5 ± 48.8 and 138.2 ± 30.0 μg m^{-3} indoors and outdoors, respectively. Both the average and daily values outdoors were well below the air quality standard of 300 μg m^{-3} for TSP in China on a 24-h average (GB 3095-2012 2012). For $PM_{2.5}$ (PM with aerodynamic equivalent diameters < 2.5 μm), its mass is equal to the sum of four size fractions of $D_{p2.5–1.0\ \mu m}$, $D_{p1.0–0.5\ \mu m}$, $D_{p0.5–0.25\ \mu m}$ and $D_{p<0.25\ \mu m}$ (collected at the stages of B-E from the cascade impactors). The average $PM_{2.5}$ accounted for over half of TSP indoors (50.7%) and outdoors (52.4%). The average and daily outdoor $PM_{2.5}$, except three sampling days, was below the air quality standard of 75 μg m^{-3} in China on a 24-h average (GB 3095-2012 2012). The masses with different particle sizes showed V-shaped distributions, with higher values at $D_{p>2.5\ \mu m}$ and $D_{p<0.25\ \mu m}$ and a minimum at $D_{p0.5–0.25\ \mu m}$. On average, coarse particles ($D_{p>2.5\ \mu m}$) contributed 49.3 and 47.6% of the TSP indoors and outdoors, respectively. Very fine particles ($D_{p<0.25\ \mu m}$) accounted for 16.9% of the TSP indoors and 24.9% of outdoors.

Table 1 Summary of PM mass concentrations and their indoor-to-outdoor ratios (I/O)

PM mass (μg m^{-3})	Indoors (I)	Outdoors (O)	I/O
A: $D_{p>2.5\ \mu m}$	104.2 ± 77.2	65.8 ± 45.8	1.6
B: $D_{p2.5–1.0\ \mu m}$	38.7 ± 23.0	13.6 ± 5.0	2.8
C: $D_{p1.0–0.5\ \mu m}$	19.1 ± 12.8	14.1 ± 7.3	1.4
D: $D_{p0.5–0.25\ \mu m}$	13.6 ± 8.2	10.3 ± 4.2	1.3
E: $D_{p<0.25\ \mu m}$	35.8 ± 18.9	34.4 ± 16.3	1.0
$\sum$PM (TSP)	211.5 ± 48.8	138.2 ± 30.0	1.5
$PM_{2.5}$	107.3 ± 19.4	72.4 ± 13.3	1.5

The indoor-to-outdoor ratio (I/O) of masses with different particle sizes is also listed in Table 1. Higher I/O with $D_{p>2.5\ \mu m}$ and $D_{p2.5–1.0\ \mu m}$ (1.6 and 2.8, respectively) was resulted by coagulation and/or gas to particle condensation on the surface of preexisting particles and re-suspension of urban dust and soils, shedding of skin cells, spray of coughing and sneezing in the classroom (Mejía et al. 2011). Almeida et al. (2011) observed that re-suspension of deposited particles could strongly elevate the indoor PM levels. Less impact on finer particles (range of I/O 1.0–1.4) was found because the deposition velocities are often varied with a function of PM size.

Figure 1 presents the time series of daily TSP mass concentrations and their I/O. The TSP concentrations ranged from 90.0 to 504.4 μg m^{-3} and 59.8 to 299.7 μg m^{-3} indoors and outdoors, respectively. The average TSP in the occupied classroom on the school days (weekdays) was 247.9 μg m^{-3}, twice of that in the vacant classroom on the non-school days (weekends, 110.9 μg m^{-3}). Besides, the I/O values varied day-to-day and were equal to or greater than unity (1.0–2.3). In general, the I/O was generally higher than on the school days than the non-school days. The "weekend effect" can be ascribed to no human activities such as physical movement, cleaning and teaching activities (use of chalk) in the classroom (Tran et al. 2012). Routine cleaning (i.e., light but without use of detergent) is conducted daily after the class on the school days.

The correlations between indoor and outdoor TSP masses with different particle sizes were examined (Fig. 2). Statistically significant correlations ($p < 0.001$) were shown with linear regression (R^2) ranged from 0.414 to 0.912. The highest R^2 was seen on the coarse particles ($D_{p>2.5\ \mu m}$), in agreement with the best homology. For the fine PMs, they had higher potentials of chemical conversions that lead generation of new or secondary particles.

BC and CO_2

Figure 1 illustrates the daily averaged BC concentrations in TSP during the sampling period, with mean values of 3273.5 ± 1861.8 and 3639.4 ± 1768.5 ng m^{-3} indoors and outdoors, respectively. The daily trend of BC was consistent with the TSP, showing the lower values on the non-school days (Fig. 1). However, the average I/O of BC

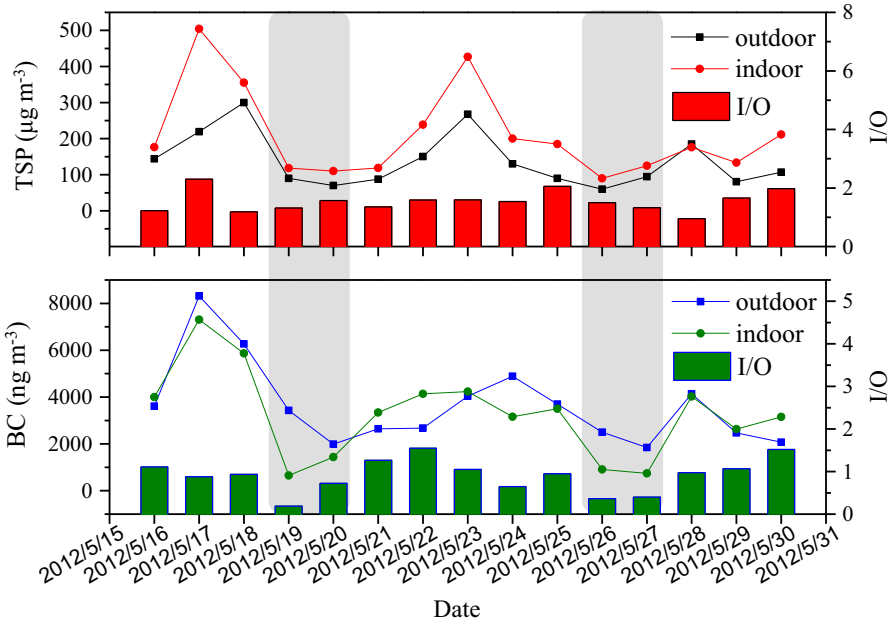

Fig. 1 Time series of indoor and outdoor TSP and BC concentrations, as well as their indoor-to-outdoor ratio (I/O) at the school (shadow zones: non-school days)

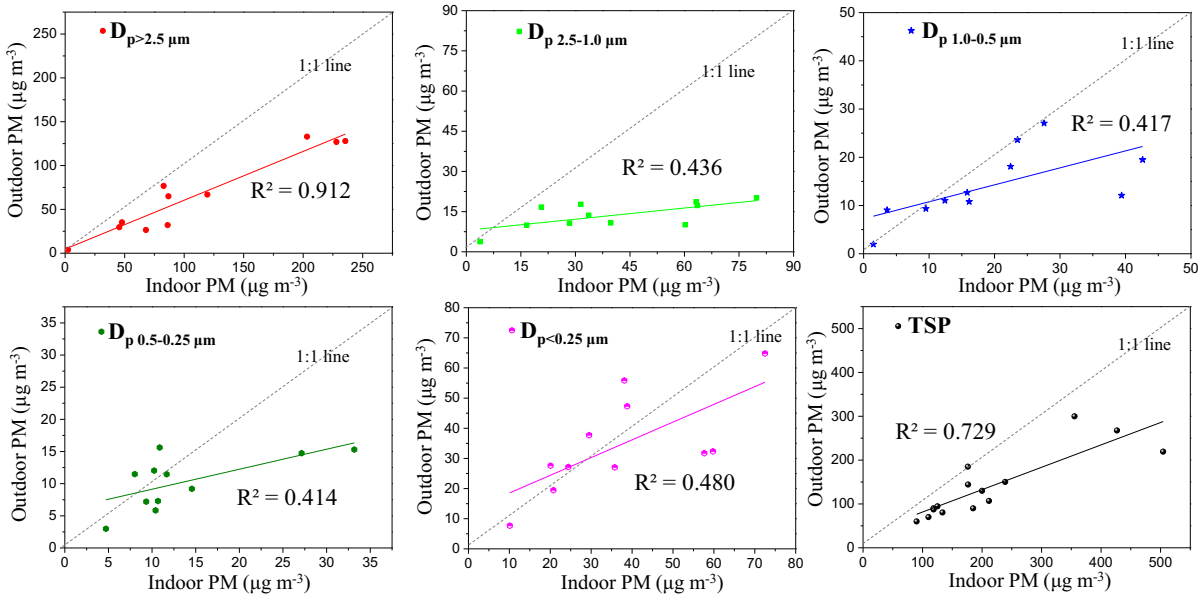

Fig. 2 Indoor–outdoor correlations of PM concentrations with different particle sizes

was below unity, with an average of 0.9 ± 0.4, suggesting the dominance of outdoor sources for BC. As we know, BC is an indicator of traffic-related emissions in urban environments (Koelmans et al. 2006; Cao et al. 2009).

Figure 3a demonstrates pronounced temporal variations on hourly averaged BC. Both indoor and outdoor trends were generally consistent, with four peaks observed in 06:00–07:00, 09:00–11:00, 16:00–19:00 and 22:00–23:00 local time. The daytime peaks were reliably impacted by the increase numbers of vehicle emission nearby during the rush hours. A decline of BC concentration was seen between 13:00 and 15:00, subjected to higher boundary layer in the atmosphere. The elevation of BC at the nighttime can be ascribed to more diesel-fueled vehicles for goods

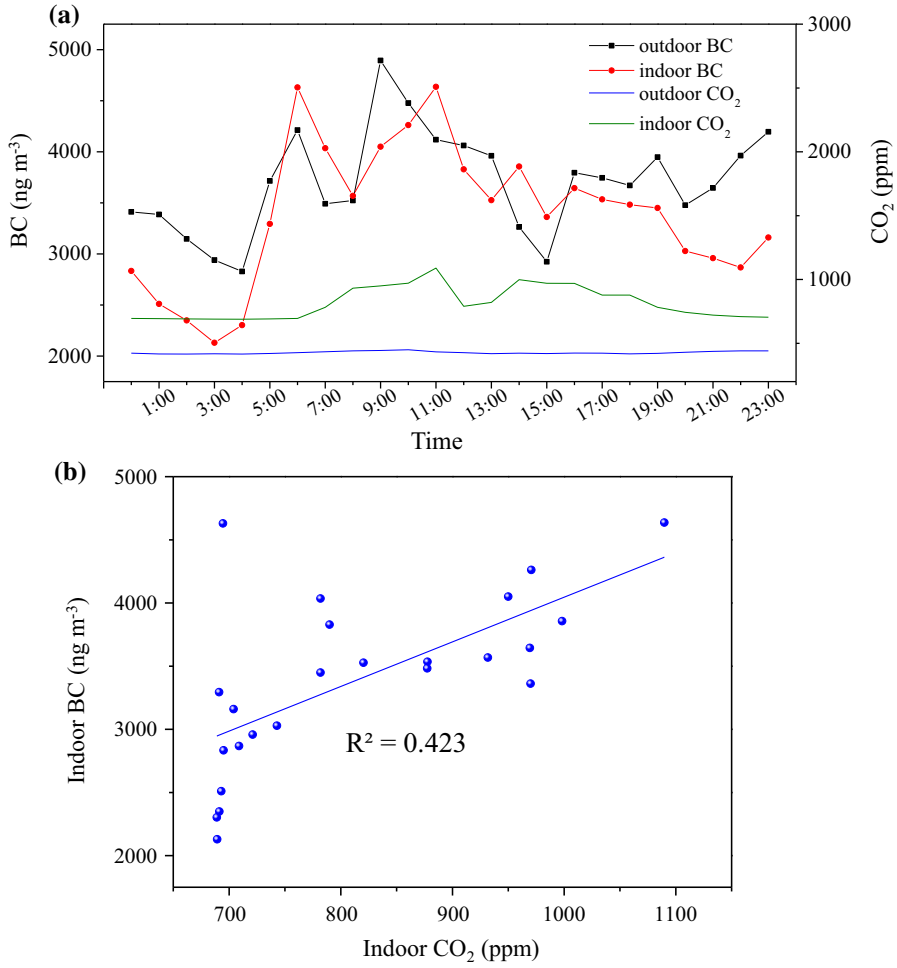

Fig. 3 **a** Temporal variations of the hourly averaged BC and CO_2 concentrations and **b** correlation between indoor concentrations of BC and CO_2

transportation and construction trucks. Indoor CO_2 is a tracer of human activity (Heudorf et al. 2009). High indoor CO_2 concentrations were observed from 07:00 to 18:00, ascribed to breathing of occupants (i.e., teachers or students) (Fig. 3a). In comparison, the outdoor CO_2 concentrations were much steady due to large air circulation. Figure 3b shows the correlations between the concentrations of indoor BC and CO_2, with a very fair R^2 of 0.423. This result supported that the indoor BC should not be produced by the activities inside the classroom, while no any combustion sources were presented.

PAHs

The concentrations, particle-size distributions and I/O of PAHs were discussed in our previous publication (Xu et al. 2015). Here, we characterized the potential pollution sources to the school airs with the diagnostic ratios of PAHs. Figure 4 illustrates the diagnostic ratios of indeno[1,2,3-cd]pyrene/(indeno[1,2,3-cd]pyrene + benzo[ghi]perylene) [IcdP/(IcdP + BghiP)] vs. benzo[a]anthracene/(benzo[a]anthracene + chrysene) [BaA/(BaA + CHR)] and anthracene/(anthracene + phenanthrene) [ANT/(ANT + PHE)] versus benzo[e]pyrene/(benzo[e]pyrene + benzo[a]pyrene) [BeP/(BeP + BaP)] with different particle sizes (Tobiszewski and Namiesnik 2012).

Yunker et al. (2002) documented that the IcdP/(BghiP + IcdP) of < 0.2, 0.2–0.5 and > 0.5 represents petrogenic, petroleum combustion and a mix of grass, wood and coal combustions, respectively. And BaA/(BaA + CHR) of < 0.2, 0.2–0.35 and > 0.35 refers to petrogenic, coal combustion and vehicular emissions, respectively (Akyüz and Çabuk 2010). As

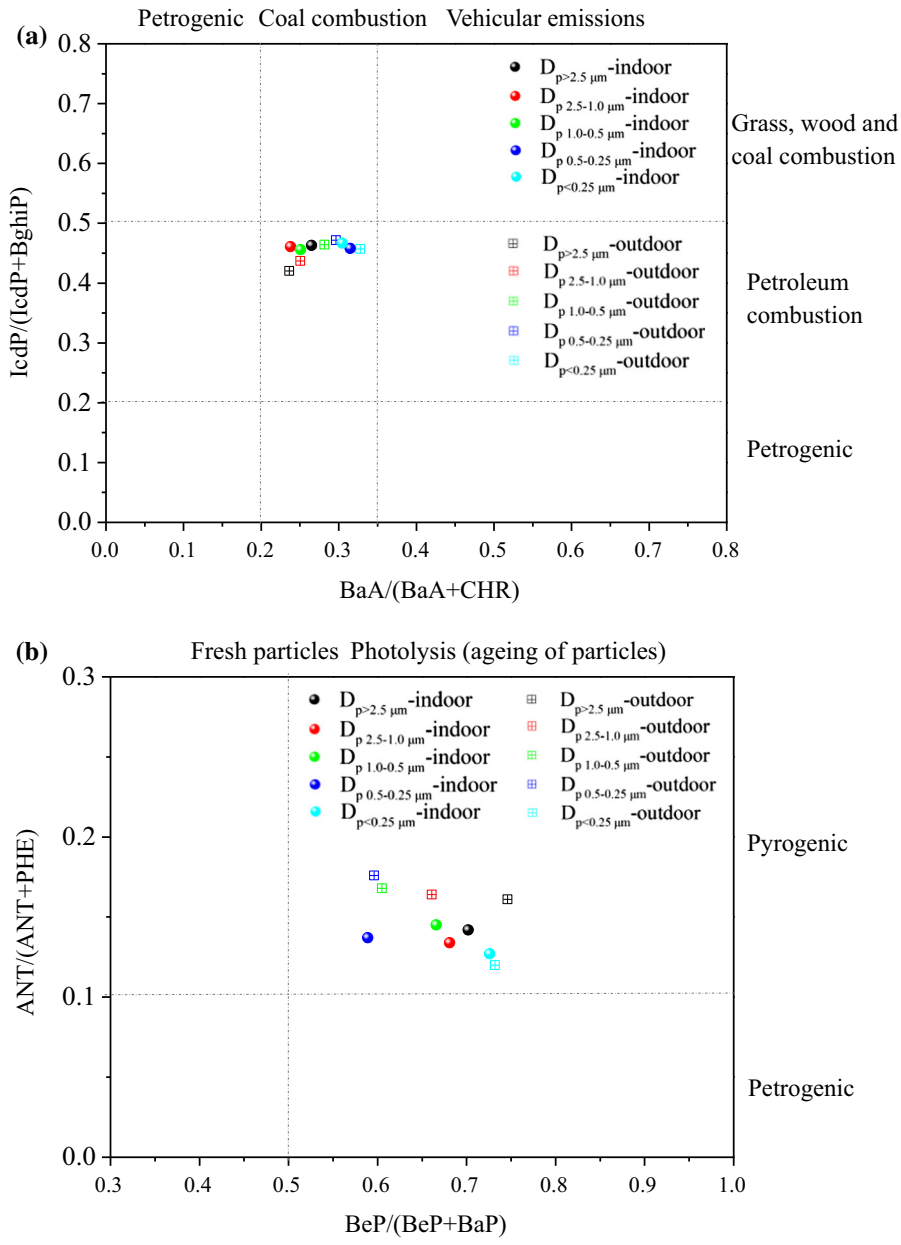

Fig. 4 Correlations between PAH diagnostic ratios with different size-distributed PAHs [**a**: IcdP/(IcdP + BghiP) vs. BaA/(BaA + CHR); **b** ANT/(ANT + PHE) vs. BeP/(BeP + BaP)]

shown in Fig. 4, the indoor and outdoor IcdP/(BghiP + IcdP) was 0.46–0.47 and 0.42–0.47, and BaA/(BaA + CHR) was 0.24–0.31 and 0.24–0.33, respectively, appointing to a mixed contribution of petroleum and coal combustions. Without any combustion sources, the indoor PAHs should be infiltrated and transported from the outdoors through regular air exchange. The values of IcdP/(BghiP + IcdP) increased with a decreasing of particle sizes, ascribed to stronger influences on the formation of fine fractions from grass, wood and coal combustions. Higher BaA/(BaA + CHR) was observed at $D_{p0.5–0.25\ \mu m}$ and $D_{p<0.25\ \mu m}$, with an average of 0.30 and 0.33, respectively, suggesting that the fine particles were predominantly affected by the traffic emissions.

The ANT/(PHE + ANT) was 0.13–0.15 and 0.12–0.18 indoors and outdoors, respectively (Fig. 4). ANT is less thermodynamically stable than PHE, and hence, the increase in ANT suggested a raise

in weights of combustion sources. The ANT/(PHE + ANT) of < 0.1 is usually associated with petrogenic sources, while the ratio of > 0.1 indicates the dominance of pyrogenic combustion (Pies et al. 2008). In this study, higher ANT/(PHE + ANT) with $D_{p<2.5\ \mu m}$ to $D_{p0.5–0.25\ \mu m}$ was found outdoors than indoors, suggesting that larger significance of pyrogenic sources in outdoors. Meanwhile, the BeP/(BeP + BaP) ranged 0.59–0.73 and 0.60–0.75 indoors and outdoors, respectively. The values were comparable with those reported in Guangzhou (0.41–0.72) and Shanghai (0.49–0.78) (Li et al. 2005; Gu et al. 2010) and agreed with that the photochemically degradation was minor. In between, the coarsest ($D_{p>2.5\ \mu m}$) and finest ($D_{p<0.25\ \mu m}$) particles were more prone to aging because of the potential re-suspension and the longer residence lifetime, respectively.

Mutual relationships of PM, BC and PAHs

PM and BC

The daily BC concentrations accounted for 1.6 ± 0.6% (0.6–2.8%) and 2.8 ± 0.9% (1.5–4.2%) in TSP indoors and outdoors, respectively. The higher compositions of BC outdoors were consistent with the fact that the incomplete combustion sources were more pronounced outdoors.

PM and PAHs

The mean compositions of total quantified PAHs in TSP with different particle sizes are presented in Table 2. The PAH compositions (0.07–1.07‰ and 0.07–1.72‰ indoors and outdoors, respectively) increased with a decrease in particle sizes, suggesting that the PAHs were enriched in finer particles, especially with $D_{p<0.25\ \mu m}$. In addition, the PAH proportions in TSP were higher outdoors than indoors, agreed with the apparent outdoor combustion sources.

BC and PAHs

The abundances of BghiP, a tracer of motor vehicle exhaust (Xu et al. 2013), was significantly correlated with the daily averaged BC levels, with high R^2 of 0.838 and 0.722 outdoors and indoors, respectively. (Figures were omitted.) Figure 5 further illustrates the variations of the correlation coefficients between BC and PAHs with different particle sizes and the number of aromatic rings on PAHs. BC can be divided into soot and char (charcoal) aerosols (Masiello 2004; Han et al. 2010), while PAHs are precursors of aromatic structure during the soot formation. A fraction of PAHs could be firmly entrapped with char matrix (Stanmore et al. 2001; Liu et al. 2011; Han et al. 2015). Hence, BC and PAHs are often supposed to be transported together in the atmosphere (Liu et al. 2011; Han et al. 2015). For outdoors, BC had fair to distinct correlations with PAHs, and the R^2 increased with the decreases in particle sizes. However, a reverse trend was observed indoors as the R^2 declined with the particle size decreased (Fig. 5a). The results supported that both BC and PAHs outdoors could be originated from the same combustion sources (i.e., traffic emission) and were more contributed in the formation of finer particles. On the other hand, stronger correlations between BC and PAHs were observed with more aromatic numbers in PAHs (i.e., 4–6 rings) outdoors (Fig. 5b). The lighter PAHs (i.e., 3 rings PAHs) can be volatized and partitioned in gaseous phase, while the heavier are mainly produced from the traffic emissions (Baek et al. 1991; Wang et al. 2007). However, no

Table 2 Compositions of total quantified PAHs in PM indoors and outdoors

	Indoor PAHs/PM (‰)	Outdoor PAHs/PM (‰)
A: $D_{p>2.5\ \mu m}$	0.07	0.07
B: $D_{p2.5–1.0\ \mu m}$	0.12	0.21
C: $D_{p1.0–0.5\ \mu m}$	0.28	0.31
D: $D_{p0.5–0.25\ \mu m}$	0.37	0.63
E: $D_{p<0.25\ \mu m}$	1.07	1.72
PAHs/$\sum$PM	0.28	0.56
PAHs/$PM_{2.5}$	0.50	1.01

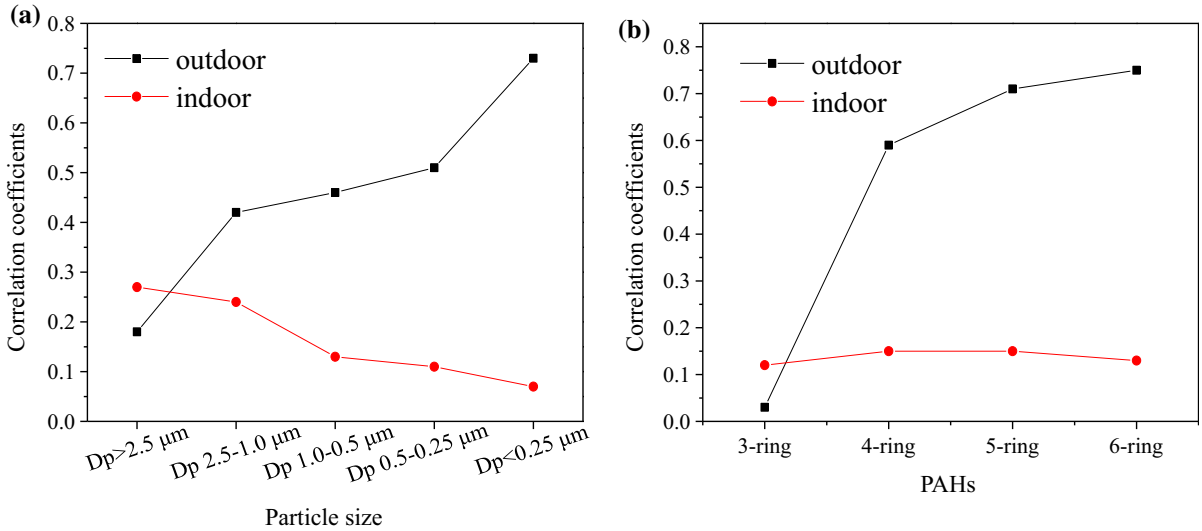

Fig. 5 Variations of correlation coefficients (R^2) of BC and PAHs with different **a** particle sizes and **b** number of aromatic rings at PAHs

such trend was observed indoors due to less direct influences from the combustion activities.

Potential risk estimation and direct health implication

PAH carcinogenic risks

Table 3 lists the ILCRs with different particle-size fractions. The ILCRs with $D_{p>2.5\ \mu m}$ and $D_{p2.5–1.0\ \mu m}$ were below 0.1×10^{-6} indoors and outdoors, suggesting negligible cancer risks of PAHs with the coarse mode particles. The ILCRs with $D_{p<0.25\ \mu m}$ were 5–25 times of those in larger-sized PMs. The finest PM has a long residence time in the atmosphere and thus can be transported over a long distance and eventually penetrated into the pulmonary system of human (Yu and Yu 2011). The PAHs associated with the finest PM posed the highest toxicity risk to the students. Besides, the average ILCRs were ~ 50% higher outdoors than indoors, implying the high toxicity originated from the outdoor pollution sources. $PM_{2.5}$ can directly enter to alveoli of lungs and is one of the major hazardous materials in the atmosphere (Gurman et al. 1984; Cao et al. 2013). The ILCRs were 1.62×10^{-6} and 2.34×10^{-6} indoors and outdoors, respectively. Both exceeded the threshold value of 1×10^{-6}, indicating that the carcinogenic PAHs are a threat to the students' health and subsequently alerting a need of effective control of PAH emission in this city in Northwestern China.

Table 3 Incremental lifetime cancer risks (ILCRs) for PAHs

Size (μm)	PAH ILCRs ($\times 10^{-6}$)	
	Indoors	Outdoors
$D_{p>2.5}$	0.096	0.076
$D_{p2.5–1.0}$	0.093	0.066
$D_{p1.0–0.5}$	0.13	0.19
$D_{p0.5–0.25}$	0.22	0.30
$D_{p<0.25}$	1.18	1.79
$PM_{2.5}$	1.62	2.34
$PM_{1.0}$	1.53	2.28
$PM_{0.5}$	1.40	2.09

According to our ILCR calculations, there were two cancer cases in one million of students induced by inhalation of $PM_{2.5}$-bound PAHs. The total number of secondary school students (11–16 years old) was approximately 453,000 in Xi'an at the end of 2012 (the year of this study) (Xi'an Statistical Yearbook 2013). It should be noted that the cancer risks could be potentially underestimated since the sampling was conducted in spring to summer, when both PM and PAH concentrations were lower than those quantified in winter (Wang et al. 2006).

FeNO test and questionnaire

In this study, the average age of the students was 13.0 ± 0.6 years, and the boy-to-girl ratio was 6:4. No student had backgrounds of neither asthma nor allergic diseases. The average FeNO level was 17.4 ± 8.5 ppb with a range of 6–44 ppb. Our FeNO values were much higher than those surveyed in other Chinese megacities, such as 13.3 ± 1.7 ppb (9–11-year-old healthy children) in Shanghai (Xu et al. 2011) and 10.7–13.8 ppb (11–18-year-old healthy youths) in Beijing (Li et al. 2010). Compared with the values measured with moderate atopic asthmatic children (21 ppb on average and ranged from 14 to 32 ppb for age of 10–12 years) in Europe (van der Valk et al. 2012), the FeNO level in Xi'an was slightly lower. The official clinical practice recommendations published by the ATS addresses that the FeNO value of > 35 ppb for children represents a high potential of eosinophilic inflammation and responsiveness to corticosteroids, while the value of < 20 ppb for children suggests those would unlikely occur. The FeNO levels between 20 and 35 ppb for children should be cautiously interpreted with reference to the clinical context (Dweik et al. 2011). Even though our average was below the threshold recommended for children, there was a wide distribution of the values. There was 6% in total exceeded the upper limit of 35 ppb, with a maximum value of 44 ppb. The distribution still implied a relatively high rate of respiratory inflammatory responses among the surveyed students in this study.

According to the guideline of ATS, we have divided the 50 students into three groups with the information summarized in the health and environmental questionnaires. The FeNO levels, anthropometric data and conditions related to their living environments are shown in Table S2 in SI (only shown the relatively influential factors in Table S2). Indoor and outdoor air pollutions are associated with a variety of health effects on either short-term or long-term effects (Wolkoff 2013). The top six influencing factors and the results were thus concluded (Fig. 6), including factor a: the FeNO levels for students who never suffered from the upper respiratory tract infections, colds or ear infections in the past three months (average: 14.2 ± 5.6 ppb) were much lower or less scattered than who suffered from more than once in the past three months (average: 19.2 ± 9.4 ppb) (Fig. 6a); factor b: students who grew up in the city had higher FeNO levels (average: 17.6 ± 9.1 ppb) than who grew up in the countryside or rural (average: 15.7 ± 6.2 ppb) (Fig. 6b); factor c: families with pets exhibited higher FeNO values (average: 19.1 ± 7.2 ppb) than no pet (average: 17.2 ± 9.0 ppb) (Fig. 6c); factor d: the student who lived in old houses (constructed before 2000) had lower FeNO (average: 15.1 ± 5.4 ppb) than who lived in house constructed after 2000 (average: 19.9 ± 10.3 ppb) (Fig. 6d); factor e and f: the student who lived in house had issues including leakage and damp on wall, floor or ceiling, and water condensation on window or glass, in the past year had higher FeNO levels of 24.0 ± 13.8 and 21.9 ± 11.8 ppb, respectively, in comparison with 15.8 ± 6.1 and 15.8 ± 6.2 ppb, respectively, for who lived in the good condition houses (Fig. 6e, f).

Our results were consistent with those findings from other studies. The health condition of participants is a key factor varying the FeNO levels. Any occurrence of upper respiratory tract infections, colds or ear infections in near history could elevate the respiratory inflammatory responses in the tests. In addition, the results of high FeNO values can be caused by the participants who lived in the apartment with new home decoration, furniture or flooring, leakage, damp or water condensation on window or glass. Environmental tobacco smoking (ETS) is a contributor to respiratory inflammatory responses as well (Zhao et al. 2008). For the students who exposed to ETS from birth to 1-year-old, the impact was much stronger than who exposed ETS with the same length at higher ages. It is thus worth emphasizing that the household ETS has an extremely high potential to affect the health of newborns and persistently harm to growth of children.

We have conducted a review on many relevant studies of chemical compositions in PM and respiratory inflammatory responses. Lin et al. (2011) reported that the BC and $PM_{2.5}$ concentrations were strongly associated with FeNO values for the students, which increased by 16.6% [95% confidence interval (CI), 14.1–19.2%] and 18.7% (95% CI, 15.0–22.5%) per interquartile range change (IQR) of BC (4.0 μg m^{-3}) and $PM_{2.5}$ (149 μg m^{-3}), respectively, in Beijing, China, during the Olympics Game in 2008. Chen et al. (2015) also reported that a 11.3% (95% CI, 0.6–23.0%) raise of FeNO value was observed on IQR of indoor $PM_{2.5}$ (64 μg m^{-3}) in Shanghai, China.

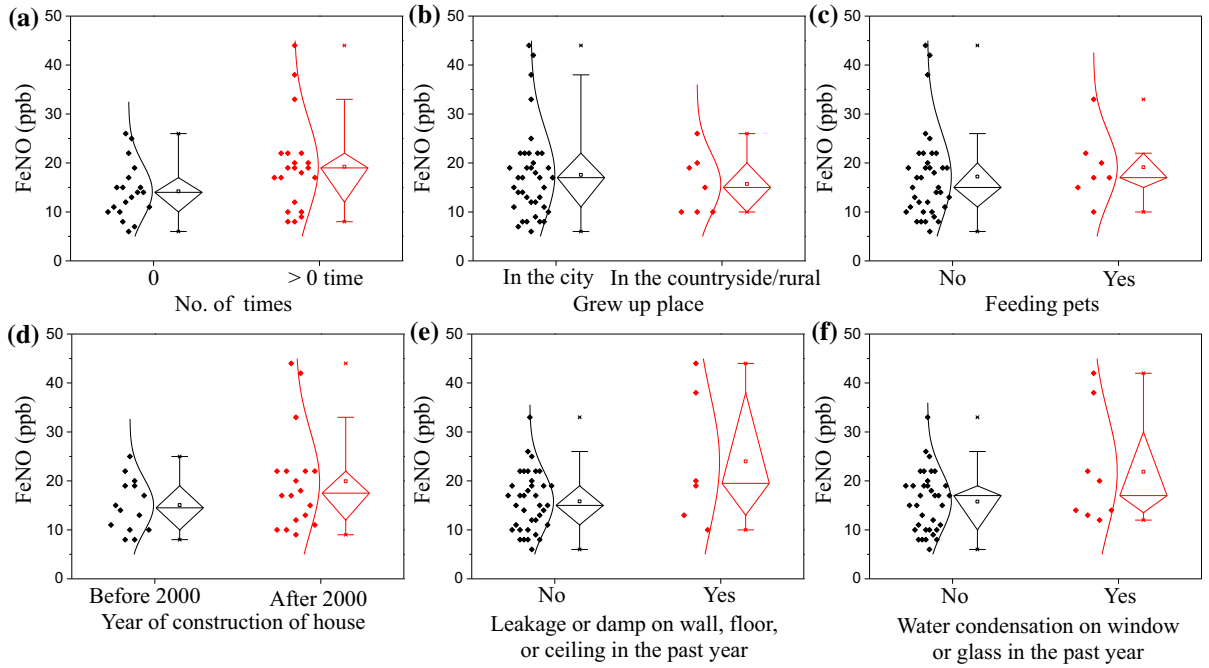

Fig. 6 Distributions of FeNO values with **a** number of times with the upper respiratory tract infections, colds or ear infections in the past three months; **b** grew up place; **c** feeding pets; **d** year of house construction; **e** leakage or damp on wall, floor or ceiling in the past year; **f** water condensation on window or glass in the house in the past year

The respiratory inflammatory responses for the children without seroatopy were correlated with domestic BC ($p = 0.012$) in New York, USA (Cornell et al. 2012). In addition, different metals in ambient airs highly varied the FeNO production (Rosa et al. 2014). Those studies supported the airborne PM and the toxic components had positive relationships with the respiratory inflammatory responses. PM is associated with acute respiratory inflammation, and more public health intervention on emission standard and execution is warranted.

Study limitations and health implication

Due to the political limits, the study involved 50 participants in one school and the samplings were conducted at a rather short period. The tactic might have limited the generalities of the results and increased uncertainties on the health assessments, potentially underestimating the scores because of no sample collection in heating period in winter (i.e., frequent haze days). Besides, there was no longtime series on the FeNO data for the students. It is thus difficult to interpret statistical correlations between the toxic components in PMs and the respiratory inflammatory responses. However, the approach is a major research direction in the future.

Conclusions

The outdoor pollution sources contributed a dominant fraction of PM indoors at the school in Xi'an, China. The elevations of BC and PAH levels were mainly caused by the combustion emissions from the surrounding environments. The toxic substances in fine PM being inhaled can pose high health risks to children and youths. The indoor and outdoor ILCRs exceeded the threshold value, indicating that the carcinogenic PAHs are a threat to the students' health. The high respiratory inflammatory responses demonstrated a potential link with the poor air quality in this heavily polluted city in Northwestern China. Efficient environmental protection controls and pollution reduction policies should be executed to suppress the health risks.

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