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Source identification and health risk assessments of polycyclic aromatic hydrocarbons in settled dusts from different population density areas of Ilorin, Nigeria

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Abstract Polycyclic aromatic hydrocarbons (PAHs) have attracted significant attention in recent times on account of their reasonably high environmental burden and extreme toxicity. Samples of indoor dusts were obtained daily over a period of 2 weeks from 10 residences located within low, medium, and high density residential areas of Ilorin City. The concentration levels, potential sources, and cancer health risks of sixteen polycyclic aromatic hydrocarbons (PAHs) were investigated using gas chromatography/mass spectrometry. PAHs total concentrations varied from 3.95±0.19 to 8.70±0.43 μg/g with arithmetic mean of 6.09±0.46 μg/g. Fluoranthene was the most dominant PAHs congener. High molecular weight (HMW) PAHs (4–6 rings) were the most prevalent PAHs and were responsible for 79.29% of total PAHs in sampled residences. Chrysene (Chry) was the most abundant compound among the 7 carcinogenic PAHs (CPAHs). Moreover, diagnostic ratios and positive matrix factorization (PMF) employed to apportion PAHs suggested that indoor dusts originated from indoor activities and infiltrating outdoor air pollutants. Diagnostic ratios revealed that PAHs are from mixed sources which include coal/wood combustion, non-traffic and traffic emissions, petroleum, petrogenic (gasoline), and petroleum combustion. Similarly, positive matrix factorization (PMF) model suggested five sources (factors) were responsible for PAHs in indoor dusts comprised of petroleum combustion and traffic emissions (60.05%), wood and biomass combustion emissions (20.84%), smoke from cooking, incense burning and tobacco (4.17%), gasoline combustion from non-traffic sources (13.89%), and emissions from coal burning and electronic devices (1.05%). The incremental lifetime carcinogenic risks (ILCR) of PAHs in adults and children estimated by applying benzo(a)pyrene (BaP) equivalent were within the satisfactory risk limits in Ilorin. Indoor PAHs emissions in Ilorin residences could be monitored and controlled by using data provided in this study.

Keywords Polycyclic aromatic hydrocarbons · Indoor dust · Residential area · Source · Cancer risk

Introduction

Human exposure to contaminants in urban indoor environments is a major concern considering that

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about 90% of peoples' daily activities in cities are carried out indoors (Adeniran et al., 2015, 2017, 2019; Stamatelopoulou et al., 2021). Depending on pollutants nature, indoor contaminants are mainly dispersed through indoor air and dust matrixes. Pollutants enter human body through the three contact routes of ingestion, inhalation, and dermal absorption (Qi et al., 2014). Owing to indoor dust's capacity to serve as depository, carrier, and sink of air pollutants, indoor dust sample collected over large surface area is more suitable for long-term assessment of human health risks in indoor environment compared to in situ sampling of indoor air for the same purpose (Iwegbue et al., 2019).

Polycyclic aromatic hydrocarbons (PAHs) are organic pollutants which are persistent, semi-volatile, lipophilic, and detected in practically every ecological matrix. PAHs are released predominantly from incomplete combustion of organic substances and fossil fuels, in addition to natural occurrences such as forest fires and volcanic eruptions (Wang et al., 2017). PAHs have dangerous impacts on living organisms far from their discharge sources due to their toxicity and ubiquitous nature (Iwegbue et al., 2019). Furthermore, some PAH compounds exhibit carcinogenic, immunotoxic, genotoxic, mutagenic, and endocrine-disrupting tendencies (Nováková et al., 2020). Intermediates of highly reactive epoxides are produced during metabolism of PAHs in the body of humans and animals. Adducts are formed when epoxides combine with DNA resulting in cancers and genetic mutations (Lundstedt, 2011; Pickering, 1999). Seven PAH compounds which include benz(a) anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(k)fluoranthene, chrysene, dibenz(ah)anthracene, and indeno(1,2,3-cd)pyrene are classified as possible human carcinogens (USEPA, 1993, 2021). Sources of PAHs in indoor environment encompass household cooking, smoking, incense burning, use of household products, heating activities, and infiltration of contaminated outdoor air emissions from burning of solid and fossil fuels, traffic, and industries during cross ventilation (Adeniran et al., 2017, 2019, 2020; Alamri et al., 2021; McDuffie et al., 2021; Odediran et al., 2021).

Low molecular weight (LMW) PAHs with 2–3 rings occur in gaseous phase and are more volatile, while on the other hand, high molecular weight (HMW) PAHs having 4–6 rings exist as particulates although they vaporize slightly (Alamri et al., 2021). Although PAHs have not been directly linked to asthma and allergies hitherto, outcomes of some studies indicated that exposure to PAHs might intensify the risks associated with symptoms of asthma and seroatopy in infants (Låg et al., 2020).

The nature of indoor settings (such as type of walls, floors, and ventilation), indoor activities (such as domestic heating, smoking, and cooking), and outdoor dusts penetration of enclosed environments allows indoor dusts to considerably incorporate higher PAHs concentrations than outdoor dusts (Iwegbue et al., 2019; Kang et al., 2010). To understand the exposure risks of indoor PAHs from residences, it is important to assess the concentrations, compositional pattern, and sources of PAHs in indoor dusts.

Several studies have related the patterns, sources, and health risks PAHs in indoor dusts in many cities of the world (Al-Harbi et al., 2020; Alamri et al., 2021; Ali, 2019; Pateraki et al., 2017; Qi et al., 2014; Stamatelopoulou et al., 2021; Wang et al., 2021a, b; Wang et al., 2017). Nevertheless, very limited studies have reported the episodes of PAHs sources and exposure risks in indoor dusts around Africa (Iwegbue et al., 2019; Munyeza et al., 2020). The deficiency of data on PAHs in indoor environment in Africa, especially, hampers the thorough grasp of the dangers associated with exposure to indoor PAHs, in addition to PAHs ultimate fate in the African-enclosed environments.

This study investigated the concentration distribution, sources, and carcinogenic health risks of PAHs in indoor dusts of different residential areas in Ilorin. Ilorin is one of the biggest metropolises with rapid urbanization and industrialization in Nigeria. Ilorin is a major commercial hub with mixed cultures of its residents migrating from the northern and southern Nigeria. The data of indoor PAHs from this research is crucial to the implementation of PAHs emission monitoring and control policies in Ilorin, a model African city.

Materials and method

Study area

The City of Ilorin is the state capital of Kwara in the north-central zone of Nigeria. Ilorin lies

between latitudes 8° 20′ N and 8.50° N and longitudes 4° 25′ E and 4.65° E with an area of about 100 km^2 (Zubair, 2006). It is a cultural and commercial hub situated between the forest and the savannah region of Nigeria, blending the art, music, and language of the northern and southern people. It has annual average temperature of 26.2 °C and rainfall of 1200 mm, the temperature ranged between 25 and 30 °C with March being the hottest month. Ilorin is the sixth-largest city in Nigeria. It is divided into three local government areas of Ilorin east, Ilorin south, and West with a total population of 777,667 according to the 2006 census (NPC, 2006). Its typical tropical climate is divided into dry weather (November to March) and wet weather (April–October). Ilorin has a fast growing population attributable to robust commercial and industrial activities in the city. Road dusts from paved and unpaved roads are the potential sources of outdoor dust around residential areas in Ilorin. The proximity of paved or unpaved roads from residences in Ilorin varied in radius from 25 m and above.

Indoor dusts were collected and analyzed for 16 United States Environmental Protection Agency (USEPA) priority PAHs (USEPA, 2002) at ten evenly and spatially distributed residences (sampling sites) in Ilorin as presented in Fig. 1. Sampling sites (residences) were selected from low, medium, and high density residential areas within Ilorin in August, 2020. The studied sampling sites were situated at Agbo Fulani (A1), Tanke (A2), Oloje area (A3), Oladosu Street (A4), Tanke, Pakata area (A5), Ubandawaki (A6), Sawmill/oko-erin (B), Unilorin GRA quarters (C1), Unilorin PS senior staff quarters (C2), and Offa garage (D). The geographic data of the designated ten (10) sampling indoor sites in Ilorin are shown in supplementary Table S1.

Indoor dust collection and analytical procedures

Collection of samples

Samples of indoor dust were collected pre-noon in August, 2020 (rainy season) from the surface of fans, windows, and floors at ten residences (one floor

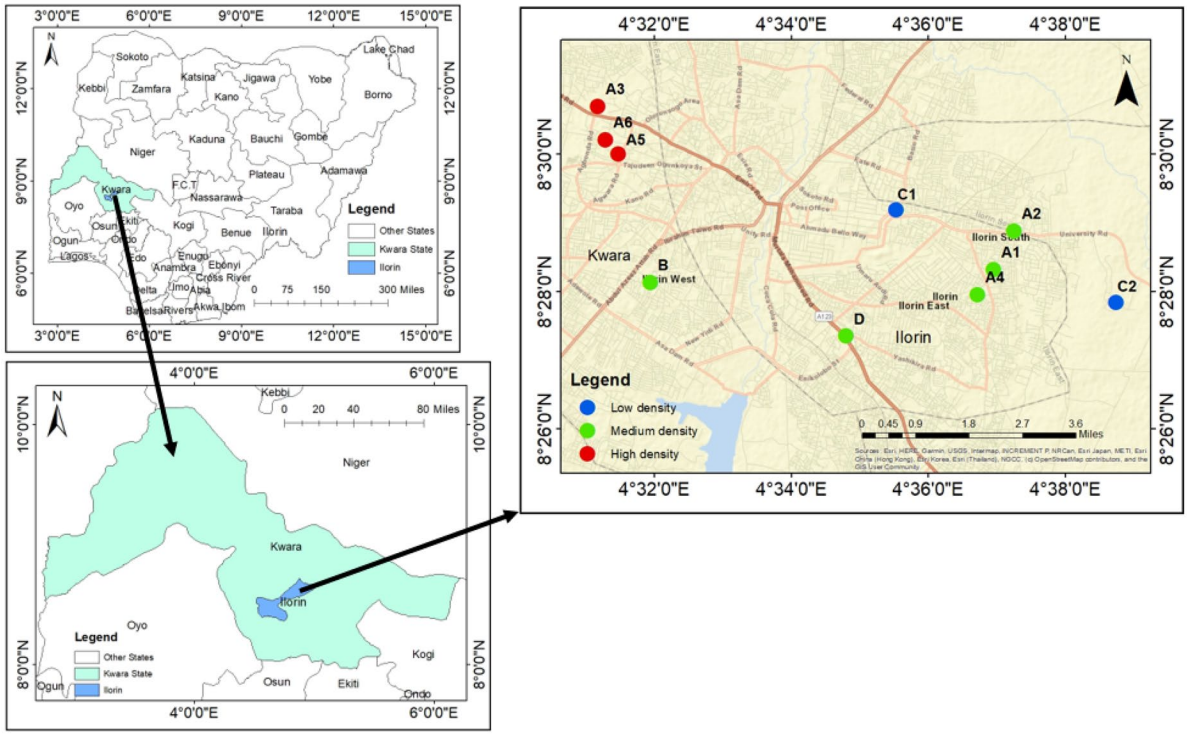

Fig. 1 Location of the study area

houses) by gentle sweeping action using new polyethylene brushes and plastic dustpans. Using a modified sampling protocol explained by Qi et al. (2014), three subsamples were collected and blended together to form one composite indoor dust sample of about 20 g at each of the ten indoor sites within Ilorin metropolis. The sampling sites comprised of two sites in the low density residential area (LDRA), four sites in the medium density residential area (MDRA), and three sites in high density residential area (HDRA). The criteria for the selection of the different population density areas are stated in the supplemental section. The new brushes were routinely pre-cleaned with deionized distilled water (DDW) to avoid cross-contamination of indoor dust samples. Extraneous substances such as paper, small pebbles, ash, cigarette butts, debris, and metal scraps were separated from the collected indoor dust samples. The dust samples were enfolded in aluminum foil before storing them in sealed polyethylene-zipper lock nylons to prevent dust re-suspension, leaching, and photo and thermal degradation of PAHs into other compounds and immediately moved to the laboratory in an ice chest at < 4 °C. Prior to extraction and analysis, all composite samples were dried in the dark and filtered into < 53 µm particle range which is of high risk to human (Soltani et al., 2015).

PAHs extraction and analysis

All indoor dust samples were extracted and analyzed for 16 priority PAHs, including naphthalene-Naph, acenaphthylene-Acy, phenanthrene-Phen, acenaphthene-Ace, fluorene-Flu, anthracene-Anth, fluoranthene-Flan, pyrene-Pyr, chrysene-Chry, benzo(a)anthracene-BaA, benzo(a)pyrene-BaP, indeno(1,2,3-cd)pyrene-IP, benzo(b)fluoranthene-BbF, benzo(k)fluoranthene-BkF, benzo(ghi)perylene-BghiP, and dibenzo(ah)anthracene-DBA. A previously described procedure by Du et al. (2021) was adopted for PAHs analyses.

Microwave accelerated extractions of samples was achieved by mixing 5 g of indoor dust sample with 30 ml of acetone:n-hexane mixture (v/v, 1:1) in a Teflon extraction vessel and then microwaved for 30 min at 120 °C (USEPA, 1999). Extracted samples were further cooled, sieved, and cleaned with silica gel (USEPA, 1996). One milliliter concentrate of extracts were obtained using calm flow of highly purified nitrogen gas and analyzed for 16 priority PAHs using GC–MS, after spiking with 200 ng of naphthalene-d8, acenaphthene-d10, and anthracene-d10 used as internal standards.

Gas chromatography-mass spectrometry (GC–MS, Agilent GC 6890, MS 5973, USA) in electron ionization mode (with ion source operating at 70 eV) was used for PAH analysis. The GC has a capillary column (Varian factor VF-4, VF–5 ms) with helium carrier gas (1 ml min^{-1} flow rate). The programmed oven temperature was kept at 80 °C for 2 min, heated to 260 °C for 6 min at 18 °C min^{-1} and then raised to 284 °C for 4 min at 4 °C min^{-1} (Verma et al., 2015). The sources and transfer line temperatures were functioning at 250 °C and 300 °C, respectively. For analytical accuracy and avoidance of overlapping molecular weights, MS functioned on selective ion scanning (SIS) mode. Samples were added by an auto sampler. Quantification of PAHs was done using internal standard method (Qi et al., 2014).

Quality control/quality assurance (QC/QA)

To attain a valid quantification with precision and accuracy, sample spikes, surrogate standards, and analytical blanks were sorted out after every 3 samples. Surrogate standards were spiked into each sample before extraction. For quality assurance, the repeated analyses' coefficient of variations (CVs) was around 3.9%. The spiked samples mean recovery efficiencies ranged from 85 to 118%. The relative standard deviations of the duplicate samples were < 14%. Targeted PAHs were not detected in method blank samples.

Benzo[a]pyrene equivalent (BaPeq) toxicity

The PAHs emissions' toxicity was calculated as benzo(a)pyrene total potency equivalence (BaPTPE) (Van den Berg et al., 2006) using Eq. 1:

$$\text{BaP TPE } (\mu g/g) = C_{PAH} \times (\text{PEF or TEF}) \quad (1)$$

where, C_{PAH} is the individual PAHs concentration, BaP TEF/PEF is the BaP toxicology or potency equivalency factor for specific PAHs (CCME, 2010). The PEF/TEF calculated in this study is presented in supplementary Table S3. B[a]P CPE is the summation of calculated BaP CPE, known as the BaP cancer potency equivalence for all

carcinogenic PAHs (Adesina et al., 2020; Manash Gope et al., 2018; Lee & Dong, 2011). In this study, the sum of concentrations of seven carcinogenic PAHs (Chry, BaA, BaP, IP, DBA, BbF, and BkF) recognized by International Agency for Research on Cancer (IARC, 2011) and denoted by CPAHs were expressed in percentage of other non-cancer PAHs (Hussain et al., 2015).

Source identification

Indicative ratios

Indicative ratios of PAHs are extensively applied to investigate PAH sources in the ecosystem (Agarwal, 2009). In this study, ratios of PAHs which include Flan/Pyr, BaA/Chry, BaP/BghiP, and Phen/Anth (Bucheli et al., 2004); Flu/(Flu + Pyr), Flan/(Flan + Pyr) IP/(IP + BghiP), and Anth/(Anth + Phen) were considered in determining PAHs sources (Manash Gope et al., 2018; Li et al., 2014; Yunker et al., 2002).

Positive matrix factorization (PMF)

Positive matrix factorization (PMF) is a technique used for identifying potential sources of various components in a mixture using multivariate receptor model (Odediran et al., 2021; Yu et al., 2015). PMF model breaks down the sample data matrix into matrices of source profile and source contribution. The type of component sources are determined using profile information acquired and components' inventories data (Niu et al., 2021; Yu et al., 2015). PMF is considered using Eqs. 2–4 (Manousakas et al., 2017). In this study, PMF 5.0 software was employed to quantitatively examine indoor PAHs sources and contributions in Ilorin residences. Additional explanation on PMF model had been reported in other studies (Odediran et al., 2021; Yuan et al., 2020).

$$x_{ij} = \sum_{k=1}^{p} g_{ik} f_{kf} + e_{ij} \tag{2}$$

x_{ij} is the measured mass concentration of jth PAH in sample i, p is sources of pollution in each sample, g_{ik} is the kth pollution source contribution rate in sample i, f_{kj} is the pollution source characteristic value k to the jth PAH concentration. The matrix of residual error, e_{ij}, is determined by minimizing the objective function Q (Tian et al., 2013; Yuan et al., 2020). Q which indicates the goodness of modeling is estimated using Eq. 2.

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{u_{ij}} \right)^2 \tag{3}$$

In this study, the amount of samples n is 10; Eq. 3 is used to calculate U_{ij} which represents uncertainty of jth PAH in sample i (Men et al., 2018).

$$U_{ij} = \begin{cases} \frac{5}{6} \times MDL, \ if \ x_{ij} \le MDL; or \ else, \\ \sqrt{(\sigma_j \times x_{ij})^2 + (MDL)^2}, \ if x_{ij} > MDL \end{cases} \tag{4}$$

x_{ij} is the concentration of jth PAH in sample i, and σ_j is the relative standard deviation of concentration of jth PAH.

Assessment of PAHs cancer risks

The PAHs incremental lifetime cancer risk (ILCR) were investigated using Eqs. 5–8 (USEPA, 1991, 2011). BaP TPE concentration corresponds to CS. Exposure parameters and factors information are shown in supplementary Table S2.

$$ILCR_{ingestion} = \frac{CS \times (CSF_{ing} \times \sqrt[3]{(\frac{BW}{70})} \times IR_{ing} \times EF \times ED)}{BW \times AT \times 10^6} \tag{5}$$

$$ILCR_{inhalation} = \frac{CS \times (CSF_{inh} \times \sqrt[3]{(\frac{BW}{70})} \times IR_{inh} \times EF \times ED)}{BW \times AT \times PEF} \tag{6}$$

$$ILCR_{dermal} = \frac{CS \times (CSF_{derm} \times \sqrt[3]{(\frac{BW}{70})} \times SA \times AF \times ABS \times EF \times ED)}{BW \times AT \times 10^6} \tag{7}$$

$$Carcinogenic\ Risk = ILCR_{ingestion} + ILCR_{inhalation} + ILCR_{dermal} \tag{8}$$

Results and discussion

PAHs distribution in indoor dust

In this study, all the 16 priority PAHs were identified, except for naphthalene (Table 1) with significant variation of PAHs concentrations and composition

observed in indoor dust samples ($p < 0.05$ was regarded as the statistical significance level). Total concentration of 16 PAHs in indoor dusts (Σ16 PAHs) varied from 3.95 ± 0.19 to 8.70 ± 0.43 μg/g with an arithmetic mean of 6.09 ± 0.28 μg/g. The most dominant PAHs congener was fluoranthene (with overall arithmetic mean 0.89 ± 0.29 μg/g) followed by Chrysene (0.74 ± 0.25 μg/g), benzo(b) fluoranthene (0.71 ± 0.31 μg/g) and phenathrene (0.70 ± 0.17 μg/g) while naphthalene (0.05 ± 0.01 μg/g) had the least PAHs concentration. The total indoor PAHs concentrations (Σ16PAHs) declined in the sequence C1, D, B, C2, A4, A5, A1, A2, A6, and A3 with overall percentages of 14.36%, 12.38%, 6.52%, 11.27%, 9.97%, 9.42%, 8.88%, 8.53%, 7.13%, and 6.52%, respectively.

Distribution of PAHs composition in this study showed that high molecular weight (HMW) PAHs (4–6 rings) were the predominant PAHs in indoor dust samples, accounting for 79.29% of overall PAHs in Ilorin-sampled residences. HMWPAHs have higher persistency and toxicity in the environment than low molecular weight (LMW) PAHs that are less toxic and highly prone to degradation. High abundance of HMWPAHs in indoor dusts analyzed in this study corroborate their characteristic ubiquitous contamination and high human exposure risk status in the environment. HMWPAHs have affinities to accumulate quickly near their sources whereas LMWPAHs have ability to be dispersed by air and deposited (multi-hop) far from their sources (Agarwal, 2009; Manash Gope et al., 2018). The total composition of PAHs rings at the ten sampling sites revealed the 4-ring PAHs as the most abundant PAHs in indoor samples (Supplementary Fig. S1). The abundance sequence of PAHs homolog in residential indoor dusts was 4 ring > 3 ring > 6 ring > 5 ring > 2 ring.

A comparison of total PAHs in indoor dusts of different cities across the world (Table 2) shows a higher mean concentration (6.09 μg/g) than the mean concentration levels in Palermo, Italy (5.11 μg/g), and Southern California, USA (0.91 μg/g), but lesser than those of Ottawa, Canada; Texas, USA; Shanghai, China; and 23 cities in China. The range of indoor PAHs concentration in this study was greater than that of Abraka and Warri, Nigeria, as well as Berlin, Germany.

Toxicity in respect of BaPeq

The benzo(a)pyrene toxicity equivalent (BaPeq) of PAHs obtained from Eq. 1 were used to calculate the PAHs carcinogenic toxicity strength in Ilorin indoor dust samples. The indoor dust in Ilorin may pose high exposure danger to residents over a long period. BaPTPE at the ten residences in this study varied from 0.70 ± 0.08 μg/g (A3) and 1.11 ± 0.04 μg/g (C1), with overall arithmetic mean BaPTPE of 1.00 ± 0.07 μg/g (supplementary Table S3). The highest total BaPTPE concentration was at C1 followed by D, C2, A2, A4, A1, A6, A5, A6, and A3, signifying that carcinogenic potency is ranked in Ilorin residences in the order LDRA > MDRA > HDRA which may be due to several factors such as nearness to road, cooking method, type of cooking fuel such as wood, charcoal, kerosene, liquefied petroleum gas (cooking gas), the type of indoor building materials, furniture, kitchenware, and paper products in residences, smoking habit, and incense burning for religious purposes and as fragrance.

Toxicity in respect of carcinogenic PAHs

The variation of carcinogenic PAHs (CPAHs) to other PAHs studied shows the CPAHs contributed 46.70% of the overall PAHs concentration in Ilorin indoor dust (Fig. 2). Chry was the predominant compound among the seven CPAHs. Chry denotes emissions from coal combustion and automobiles exhausts (Deka et al., 2016; Soltani et al., 2015).

The CPAHs abundance and contribution in decreasing order at the ten sampling sites were found to be 45.17%, 44.87%, 38.23%, 47.52%, 45.88%, 40.15%, 53.36%, 47.59%, 51.54%, and 46.53% at A1, A2, A3, A4, A5, A6, B, C1, C2, and D, respectively (Supplementary Fig. S2). The CPAHs abundance at the three residential types of LDRA, MDRA, and HDRA were 49.32%, 47.97%, and 42.06%, respectively. It was observed that the percentage of CPAHs in sampling sites followed the same trend of LDRA > MDRA > HDRA as in BaPTPE. Low density residential areas having higher PAHs concentrations than other residential type may be due to increased local anthropogenic activities such as bush burning; nearness to road traffic; burning of wastes near households; cooking method using wood,

Table 1 Sixteen USEPA priority PAHs in indoor dust of Ilorin

S/N	PAHs	Ring no	Composition (μg/g)—arithmetic mean and standard deviation										Ilorin overall
			A1	A2	A3	A4	A5	A6	B	C1	C2	D	
1	Naphthalene—Naph	2	BDL	ND	ND	ND	ND	ND	ND	0.06 ± 0.02	0.04 + 0.00	0.04 ± 0.02	0.05 ± 0.01
2	Acenaphthylene—Acy	3	0.10 ± 0.03	0.13 ± 0.01	0.10 ± 0.00	0.15 ± 0.00	0.15 ± 0.03	0.12 ± 0.03	0.18 ± 0.02	0.19 ± 0.00	0.14 ± 0.00	0.19 ± 0.00	0.15 ± 0.01
3	Acenaphthene—Ace	3	0.04 ± 0.01	0.05 ± 0.01	0.04 ± 0.00	0.05 ± 0.01	0.05 ± 0.01	0.04 ± 0.01	0.08 ± 0.01	0.06 ± 0.00	0.08 ± 0.01	0.09 ± 0.07	0.06 ± 0.01
4	Fluoren—Flu	3	0.21 ± 0.01	0.16 ± 0.02	0.23 ± 0.02	0.27 ± 0.03	0.26 ± 0.03	0.20 ± 0.02	0.36 ± 0.01	0.39 ± 0.03	0.32 ± 0.01	0.38 ± 0.06	0.28 ± 0.02
5	Phenanthrene—Phen	3	0.53 ± 0.03	0.80 ± 0.03	0.70 ± 0.00	0.77 ± 0.02	0.75 ± 0.03	0.79 ± 0.02	0.38 ± 0.01	0.98 ± 0.02	0.60 ± 0.06	0.66 ± 0.08	0.70 ± 0.03
6	Anthracene—Anth	3	0.06 ± 0.00	0.04 ± 0.00	0.07 ± 0.00	0.04 ± 0.00	0.05 ± 0.04	0.06 ± 0.00	0.13 ± 0.04	0.09 ± 0.02	0.04 ± 0.04	0.06 ± 0.02	0.06 ± 0.02
7	Fluoranthene—Flan	4	0.95 ± 0.07	0.68 ± 0.05	0.52 ± 0.03	0.79 ± 0.03	0.80 ± 0.02	0.51 ± 0.01	0.98 ± 0.02	1.42 ± 0.02	1.07 ± 0.01	1.15 ± 0.01	0.89 ± 0.02
8	Pyrene—Pyr	4	0.43 ± 0.02	0.36 ± 0.03	0.35 ± 0.03	0.57 ± 0.02	0.48 ± 0.02	0.32 ± 0.01	0.72 ± 0.02	0.51 ± 0.03	0.38 ± 0.01	0.83 ± 0.01	0.50 ± 0.02
9	Chrysene—Chry (carcinogenic)	4	0.72 ± 0.05	0.65 ± 0.03	0.46 ± 0.06	0.61 ± 0.04	0.63 ± 0.01	0.44 ± 0.09	0.84 ± 0.02	1.25 ± 0.02	1.00 ± 0.02	0.81 ± 0.01	0.74 ± 0.04
10	Benzo[a]anthracene—BaA (carcinogenic)	4	0.40 ± 0.02	0.40 ± 0.02	0.28 ± 0.02	0.46 ± 0.01	0.41 ± 0.02	0.24 ± 0.02	0.37 ± 0.02	0.63 ± 0.07	0.55 ± 0.04	0.52 ± 0.03	0.43 ± 0.03
11	Benzo[b]fluoranthene—BbF (carcinogenic)	5	0.52 ± 0.02	0.52 ± 0.03	0.15 ± 0.02	0.88 ± 0.08	0.80 ± 0.02	0.32 ± 0.06	1.03 ± 0.18	0.91 ± 0.02	0.95 ± 0.03	1.04 ± 0.04	0.71 ± 0.05
12	Benzo[k]fluoranthene—BkF (carcinogenic)	5	0.21 ± 0.03	0.16 ± 0.04	0.05 ± 0.00	0.10 ± 0.04	0.13 ± 0.01	0.08 ± 0.02	0.22 ± 0.02	0.13 ± 0.13	0.13 ± 0.05	0.12 ± 0.04	0.13 ± 0.03
13	Benzo[a]pyrene—BaP (carcinogenic)	5	0.17 ± 0.03	0.20 ± 0.00	0.19 ± 0.03	0.25 ± 0.06	0.17 ± 0.02	0.21 ± 0.03	0.32 ± 0.56	0.56 ± 0.08	0.14 ± 0.01	0.28 ± 0.01	0.25 ± 0.03
14	Indeno[1,2,3-cd] pyrene—IP (carcinogenic)	6	0.22 ± 0.02	0.27 ± 0.02	0.24 ± 0.07	0.30 ± 0.02	0.27 ± 0.02	0.30 ± 0.02	0.69 ± 0.02	0.48 ± 0.02	0.62 ± 0.02	0.40 ± 0.02	0.38 ± 0.02
15	Dibenz[a,h]anthracene—DBA (carcinogenic)	6	0.19 ± 0.04	0.12 ± 0.42	0.14 ± 0.05	0.27 ± 0.04	0.21 ± 0.04	0.16 ± 0.03	0.33 ± 0.07	0.18 ± 0.04	0.13 ± 0.05	0.32 ± 0.04	0.21 ± 0.08
16	Benzo[ghi]perylene—BghiP	6	0.63 ± 0.04	0.63 ± 0.04	0.43 ± 0.03	0.53 ± 0.05	0.55 ± 0.06	0.53 ± 0.02	0.36 ± 0.01	0.86 ± 0.01	0.64 ± 0.01	0.61 ± 0.01	0.58 ± 0.03
	Σ16 PAHs		5.38 ± 0.42	5.17 ± 0.76	3.95 ± 0.35	6.04 ± 0.46	5.71 ± 0.37	4.32 ± 0.40	6.99 ± 0.49	8.70 ± 0.44	6.83 ± 0.37	7.50 ± 0.47	6.09 ± 0.46
	Σ7CPAHs		2.43 ± 0.21	2.32 ± 0.56	1.51 ± 0.25	2.87 ± 0.30	2.62 ± 0.14	1.75 ± 0.28	3.80 ± 0.35	4.14 ± 0.28	3.52 ± 0.21	3.49 ± 0.2	2.85 ± 0.28
	BaPE		0.36 ± 0.06	0.35 ± 0.26	0.32 ± 0.06	0.53 ± 0.09	0.40 ± 0.05	0.37 ± 0.06	0.67 ± 0.08	0.81 ± 0.12	0.37 ± 0.04	0.61 ± 0.04	0.48 ± 0.09
	ΣBaP TPE		0.95 ± 0.08	0.98 ± 0.06	0.70 ± 0.08	0.97 ± 0.12	0.89 ± 0.08	0.84 ± 0.06	0.93 ± 0.06	1.65 ± 0.11	1.02 ± 0.04	1.11 ± 0.04	1.00 ± 0.07
	ΣLMW		0.94 ± 0.08	1.18 ± 0.07	1.14 ± 0.03	1.28 ± 0.06	1.26 ± 0.14	1.21 ± 0.08	1.13 ± 0.09	1.77 ± 0.10	1.22 ± 0.12	1.42 ± 0.25	1.29 ± 0.11
	ΣHMW		4.44 ± 0.34	3.99 ± 0.68	2.81 ± 0.32	4.76 ± 0.40	4.45 ± 0.23	3.11 ± 0.32	5.86 ± 0.41	6.93 ± 0.34	5.61 ± 0.25	6.08 ± 0.22	4.80 ± 0.35
	2 rings		BDL	ND	ND	ND	ND	ND	ND	0.06 ± 0.02	0.04 ± 0.00	0.04 ± 0.02	0.05 ± 0.01
	3 rings		0.94 ± 0.08	1.18 ± 0.07	1.14 ± 0.03	1.28 ± 0.06	1.26 ± 0.14	1.21 ± 0.08	1.13 ± 0.09	1.71 ± 0.07	1.18 ± 0.12	1.38 ± 0.23	1.24 ± 0.10
	4 rings		2.50 ± 0.16	2.09 ± 0.13	1.61 ± 0.12	2.43 ± 0.10	2.32 ± 0.07	1.51 ± 0.14	2.91 ± 0.07	3.81 ± 0.14	3.00 ± 0.08	3.31 ± 0.06	2.55 ± 0.11
	5 rings		0.90 ± 0.08	0.88 ± 0.08	0.39 ± 0.05	1.23 ± 0.18	1.10 ± 0.05	0.61 ± 0.12	1.57 ± 0.22	1.60 ± 0.13	1.22 ± 0.09	1.44 ± 0.09	1.09 ± 0.11
	6 rings		1.04 ± 0.10	1.02 ± 0.48	0.81 ± 0.15	1.10 ± 0.12	1.03 ± 0.11	0.99 ± 0.07	1.38 ± 0.11	1.52 ± 0.08	1.39 ± 0.08	1.33 ± 0.07	1.16 ± 0.14

biomass, or charcoal combustion; and in addition to the prevailing meteorological conditions which may promote PAHs dispersion. Most homes, cafeterias, and roadside food or snacks vendors in Ilorin employ wood or charcoal burning for cooking, frying, boiling, and roasting of plantain, bean cake, yam, maize, fish, and meat "suya and asun" and other nibbles.

PAHs source identification

PAHs specific ratios

Classification of PAHs sources in the environment are achieved using their indicative (diagnostic) ratios (Ferrara et al., 2020; Gope et al., 2020; Qi et al., 2020; Tobiszewski & Namiesnik, 2012). Plots of PAHs ratio proffer valuable and reliable data on PAHs sources in indoor dusts of Ilorin (Fig. 3). IP/IP + BghiP, Anth/Anth + Phen, and Flu/Flu + Pyr differentiate petrogenic sources from pyrolytic sources (Tobiszewski & Namiesnik, 2012). IP/BghiP is a pointer for vehicle exhausts. BaP/BghiP and IP/IP + BghiP are together evaluated as diagnostic indicator of traffic emissions and wood combustion (Yunker et al., 2002). In this study, ratios of Anth/Anth + Phen and Flu/Flu + Pyr as well as Anth/Anth + Phen and Flan/Flan + Pyr collectively indicate petroleum, petrogenic, and coal/wood combustion as sources of indoor PAHs in Ilorin. IP/IP + BghiP and BaP/BghiP ratios together classified indoor PAHs sources in Ilorin as petroleum combustion and non-traffic emissions (Fig. 3). Flan/Pyr and Phen/Anth jointly recognized coal combustion and petrogenic as PAHs sources of indoor PAHs in Ilorin.

Indoor emission in homes are predominantly characterized by emissions from traffic and non-traffic sources, cooking activities using charcoal, wood, kerosene, and cooking gas for roasting, boiling, frying of food and snacks, and other combustion sources such as tobacco smoking and incense smoke. Thus, PAHs in indoor dusts of Ilorin are from diverse sources.

The diagnostic PAHs ratios at several sampling sites suggested PAHs in indoor dusts of Ilorin are from different sources (supplementary Table S4) which include coal/wood combustion, non-traffic and traffic emissions, petroleum, petrogenic (gasoline), and petroleum combustion. PAHs could be emitted from combustion of lubricating oil (Khillare et al., 2005). Ratio of BaA/Chry is used as indicator to establish distances of PAHs sources (Gope et al., 2020; Manash Gope et al., 2018; Hussain et al., 2015); values above 1 denote that PAHs sources are local. All the sites in this study indicated BaA/Chry values were less than 1 indicating PAHs sources are not from local sources. (i.e., pollutants are from the outdoor sources).

PMF results

Source identification offers an informative guide to comprehend PAHs fate and transport in indoor environment. Data file of 16 priority PAHs concentrations and uncertainties were uploaded separately into the USEPA PMF 5.0 Software. The PAHs uncertainties in the indoor dust samples analyzed were estimated with Eq. 3 (USEPA, 2014). In this study, the number of PMF model runs was 20, fourteen of the PAHs studied were defined as "strong" having a signal to noise (S/N) ratio varying between 2.4 and 10.0 while Naph and BkF were defined as "weak" with small S/N ratio of 0.7 and 0.8, respectively. The PMF model was evaluated with factors between 2 and 9. A five factor simulation run gave a satisfactory absolute scaled residual of between − 3 and + 3 and least goodness of modeling (Q) values, demonstrating that the five factor solution perfectly matches and impacted the emission of the 16 PAHs. The PAHs factor profile in Ilorin indoor dusts are illustrated in Fig. 4. The method of detection limits (MDL) and error fractions employed for uncertainty assessment (using Eq. 3) are shown in supplementary Table S5.

Factor 1 has substantial emissions of Naph, Ace, Flu, Flan, Chry, BaA, BbF, BkF, IP, accounting for 60.05% of the total PAHs sources, signifying the impact of petroleum combustion and traffic emissions. Chry and BbF are specific markers of diesel combustion exhausts while Flu, Flan, and BkF are emissions from diesel engines (Boonyatumanond et al., 2006; Deka et al., 2016), BaA, Chry, BbF, IP are specific markers of gasoline emissions in vehicle exhausts (Kwon & Choi, 2014; Li et al., 2017). Naph, Ace, and BaA are emissions from different unburnt fossil fuels which include lubricating oil combustion, kerosene, natural gas combustion-LNG, aerosol sprays (petrochemicals), and unburnt diesel and gasoline (Gope et al., 2020; Khalili et al., 1995; Wang et al., 2000; Wong & Wang, 2001).

Table 2 Comparison of PAHs in indoor dust of different cities of the world (μg/g)

Indoor sampling site	Mean	Range	References
Ilorin, Nigeria	6.09	3.95–8.70	This study
Abraka and Warri, Nigeria	-	0.21–2.96	(Iwegbue et al., 2019)
Palermo, Italy	5.11	0.036–34.5	(Mannino & Orecchio, 2008)
Kuwait	-	1.6–16.7	(Saeed et al., 1998)
Berlin, Germany	-	0.1–1.4	(Fromme et al., 2004)
Ottawa, Canada	12.9	1.5–325	(Maertens et al., 2008)
Texas, USA	29.2	1.12–341	(Mahler et al., 2010)
Southern California, USA	0.91	0.163–439	(Hoh et al., 2012)
Shanghai, China	-	8.22–42.04	(Ren et al., 2006)
Shanghai, China	12.4	7.24–41.22	(Peng et al., 2012)
23 cities in China	30.9	1.0–466	(Qi et al., 2014)

Factor 2 contributed about 20.84% of total PAHs sources with high contamination of Phen and BghiP indicating wood and biomass combustion emissions. Phen and BghiP are released from wood and biomass burning (Deka et al., 2016). Factor 3 explained 4.17% of total measured PAH sources with high pollution of BaP which is an indicator of smoke from cooking, incense burning, and tobacco (Arfaeinia et al., 2017; Fromme et al., 2004). Factor 4 is responsible for 13.80% of total source of PAHs with greater pollution contributions of Acy, Pyr, BbF, and BDA which is recognized as gasoline emission (Y. Li et al., 2017) mostly from non-traffic sources such as gasoline used to power electric generators and electric motor for pepper- and grain-grinding machines. Generally, most households depend on

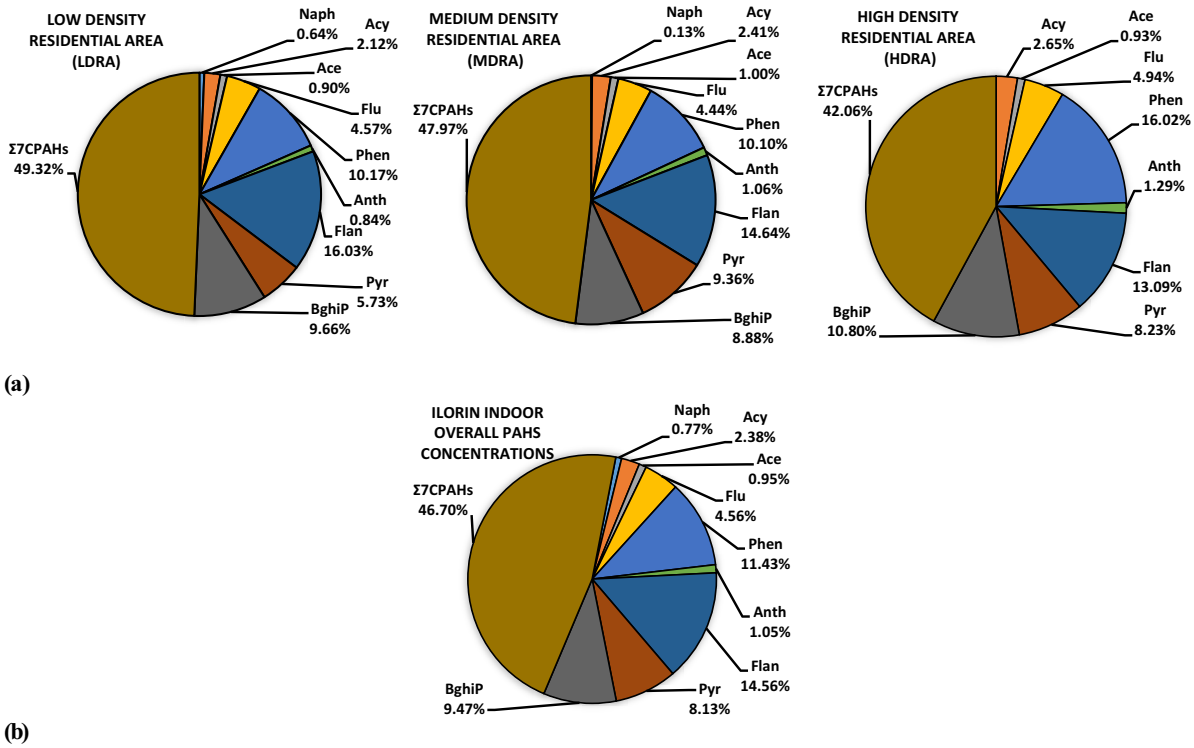

Fig. 2 Carcinogenic PAHs (CPAHs) abundance in indoor dusts of Ilorin: (**a**) Overall PAHs. (**b**) Residential type PAH concentrations

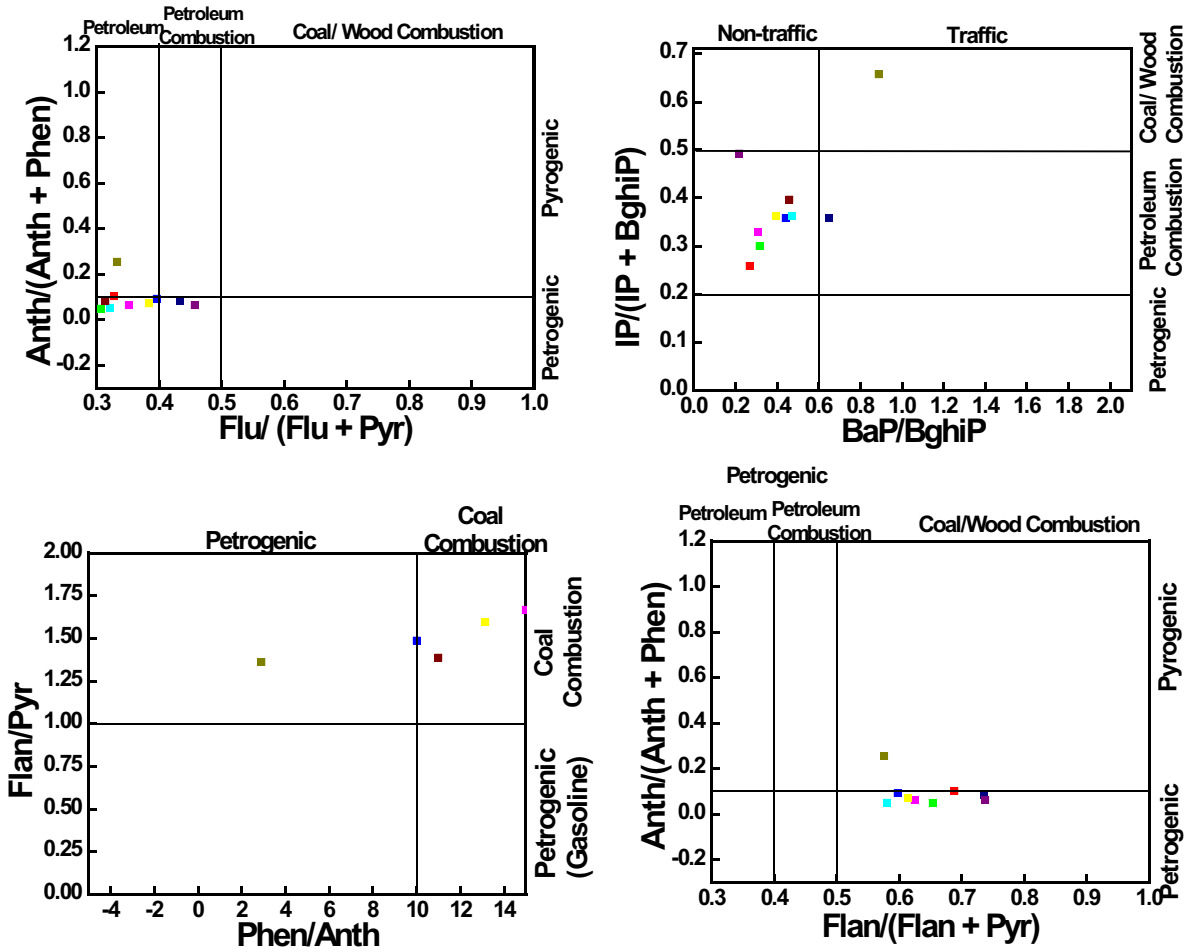

Fig. 3 PAHs diagnostic ratios in Ilorin indoor dusts

gasoline as alternative power supply in Nigeria (Y. Li et al., 2017). Factor 5 occupied 1.05% of the total PAHs sources with high contribution of Anth. Anth is associated with coal burning and electronic device emissions (Deka et al., 2016; Iwegbue et al., 2019; Lin & Lee, 1998; Mahgoub & Salih, 2017; Yang et al., 2017) from cooking with charcoal, incense burning, candle, local lamps (Atupa), electric stoves, television, and other indoor electric gadgets.

The five sources of PAHs accounting for PAHs in indoor dusts of residential areas include petroleum combustion and traffic emissions (60.05%), wood and biomass combustion emissions (20.84%), smoke from cooking, incense burning, and tobacco (4.17%), gasoline combustion from non-traffic sources (13.89%), and emissions from coal burning and electronic devices (1.05%).

Petroleum combustion and traffic emissions (60.05%) had the highest percentage factor contribution followed by wood and biomass combustion emissions (20.84%) while coal burning and electronic devices (1.05%) was responsible for the lowest factor contribution. In this study, emission from petroleum combustion (transportation), wood, and biomass combustion (cooking method) sources gave the highest PAHs contribution in residences. Results revealed that PAHs in indoor dust pose a big danger to human health and environment. Accumulation of PAHs in indoor dust may be due to day to day activities of residents which include incense burning for

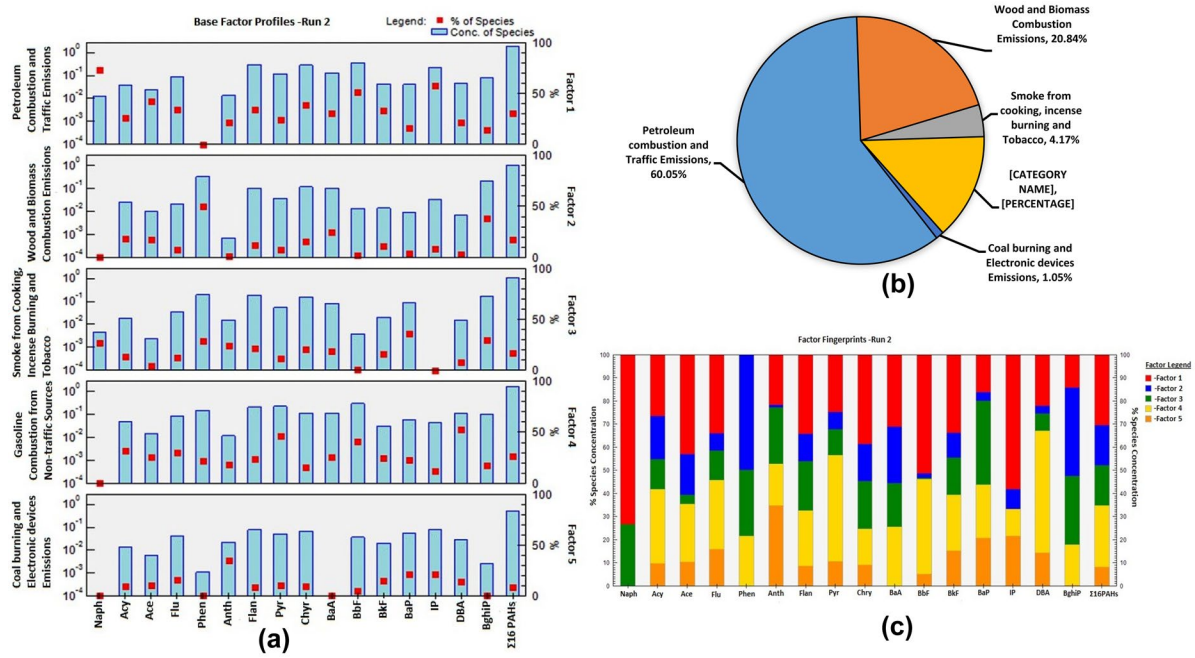

Fig. 4 (**a**) PMF model source profiles, (**b**) percentage factor contributions of PAHs, and (**c**) fingerprints of PAHs in Ilorin indoor dust

religious purposes and as perfume, tobacco smoking, cooking methods, and type of fuel used such as wood, charcoal, kerosene, liquefied petroleum gas (cooking gas). In addition to infiltration of outdoor emissions from petroleum combustion through electric generators, electric motors of grain- and pepper-grinding machines, and vehicular traffic emission into residences in the study area.

Evaluation of carcinogenic PAHs (CPAHs)

PAHs cancer risks through contact pathways of ingestion, inhalation, and dermal were evaluated using ILCR model using Eqs. 5–8 (Mon et al., 2020). Carcinogenic slope factor (CSF) and BaPTPE (carcinogenic toxicities relative to BaP) are calculated in Table 3 using BaPTPE data in supplementary Table S3. ILCR values between 10^{-4} and 10^{-6} signify likely health hazard; ILCR greater than 10^{-4} shows high potential health hazard while ILCR lesser or equal to 10^{-6} is acceptable (Hoseini et al., 2016).

The cancer risks for adults and children in Ilorin are presented in Fig. 5. PAHs cancer risks through inhalation with 10^{-9} magnitude were negligible compared to ingestion and dermal contact routes both with magnitudes of 10^{-6} in adults and children. In children, the range of cancer risks of PAHs in indoor dusts of Ilorin via ingestion and dermal contact varied from 3.10×10^{-6} (A3) to 7.31×10^{-6} (C1) and 3.86×10^{-6} (A3) to 9.11×10^{-6} (C1), respectively. For adults, the level of cancer risks via ingestion and dermal contact stretched from 2.78×10^{-6} (A3) to 6.55×10^{-6} (C1) and 4.93×10^{-6} (A3) to 1.16×10^{-5} (GAT), respectively, which were

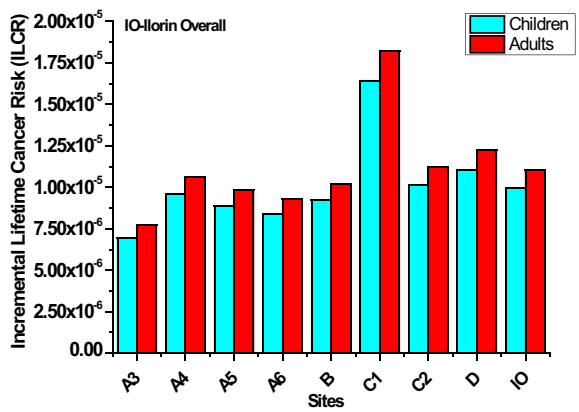

Fig. 5 Carcinogenic risk for adults and children in Ilorin

Table 3 BaPeq PAHs concentrations (CS) and cancer risks for Ilorin indoor dusts

Sites	ΣBaP TPE (CS)	$ILCR_{Child}$				$ILCR_{Adult}$			
		Ingestion	Inhalation	Dermal	Cancer risk	Ingestion	Inhalation	Dermal	Cancer risk
A1	0.95	4.19×10^{-6}	1.62×10^{-9}	5.22×10^{-6}	9.41×10^{-6}	3.75×10^{-6}	1.46×10^{-9}	6.67×10^{-6}	1.04×10^{-5}
A2	0.98	4.31×10^{-6}	1.67×10^{-9}	5.38×10^{-6}	9.69×10^{-6}	3.86×10^{-6}	1.50×10^{-9}	6.86×10^{-6}	1.07×10^{-5}
A3	0.70	3.10×10^{-6}	1.20×10^{-9}	3.86×10^{-6}	6.96×10^{-6}	2.78×10^{-6}	1.08×10^{-9}	4.93×10^{-6}	7.71×10^{-6}
A4	0.97	4.27×10^{-6}	1.66×10^{-9}	5.32×10^{-6}	9.59×10^{-6}	3.83×10^{-6}	1.48×10^{-9}	6.80×10^{-6}	1.06×10^{-5}
A5	0.89	3.95×10^{-6}	1.53×10^{-9}	4.92×10^{-6}	8.87×10^{-6}	3.54×10^{-6}	1.37×10^{-9}	6.28×10^{-6}	9.82×10^{-6}
A6	0.84	3.73×10^{-6}	1.44×10^{-9}	4.64×10^{-6}	8.37×10^{-6}	3.34×10^{-6}	1.29×10^{-9}	5.93×10^{-6}	9.27×10^{-6}
B	0.93	4.10×10^{-6}	1.59×10^{-9}	5.11×10^{-6}	9.21×10^{-6}	3.67×10^{-6}	1.42×10^{-9}	6.52×10^{-6}	1.02×10^{-5}
C1	1.65	7.31×10^{-6}	2.84×10^{-9}	9.11×10^{-6}	1.64×10^{-5}	6.55×10^{-6}	2.54×10^{-9}	1.16×10^{-5}	1.82×10^{-5}
C2	1.02	4.51×10^{-6}	1.75×10^{-9}	5.62×10^{-6}	1.01×10^{-5}	4.04×10^{-6}	1.57×10^{-9}	7.17×10^{-6}	1.12×10^{-5}
D	1.11	4.92×10^{-6}	1.91×10^{-9}	6.14×10^{-6}	1.11×10^{-5}	4.41×10^{-6}	1.71×10^{-9}	7.84×10^{-6}	1.22×10^{-5}
Ilorin overall	1.00	4.44×10^{-6}	1.72×10^{-9}	5.53×10^{-6}	9.97×10^{-6}	3.98×10^{-6}	1.54×10^{-9}	7.06×10^{-6}	1.10×10^{-5}

10^4 greater than risk through inhalation (10^{-9}). Generally, cancer risks in children via the three exposure pathways were higher than those in adults as shown in Table 3. Children are highly involved in hand to mouth actions through which they easily swallow polluted indoor dusts (Jiang et al., 2014). Thus, children absorb higher PAHs concentration than adults due to their lower body weight in relation to adults (Soltani et al., 2015). Dermal contact was the main exposure route with greater cancer risk in adults than children due to longer exposure duration (ED) and larger skin (dermal) contact surface area (SA) in adults. The total cancer risks (from ingestion, inhalation, and dermal contact) were higher in adults than in children as illustrated in Fig. 5.

In this study, C1 exhibited a higher indoor cancer risk in both children ($1.64-10^{-5}$) and adults (1.82×10^{-5}) which signifies potential indoor carcinogenic risk in Ilorin if trend continues. The cancer risks in both children and adults at the ten indoor sampling sites in Ilorin are ranked as C1 > D > C2 > A2 > A4 > A1 > B > A5 > A6 > A3. In comparison with the ILCR baseline, ILCR in both children and adults were within the satisfactory risk limits of $ILCR \leq 10^{-6}$ and between 10^{-4} and 10^{-6}, signifying the possibility of indoor carcinogenic risk in Ilorin. The results indicated that people (children and adults) of Ilorin metropolis were exposed to possible cancer risk principally through ingestion and dermal (skin) interaction with indoor dust.

Conclusion

This study probed into the concentration levels of 16 priority PAHs in indoor dusts by collecting indoor dust samples from the surface of fans, windows, and floors at ten residences (one floor houses) selected from low, medium, and high density residential areas inside Ilorin metropolis, Nigeria. The total concentrations of 16 priority PAHs (Σ16PAHs) analyzed in indoor dust samples of Ilorin residences ranged from 3.95 ± 0.19 to 8.70 ± 0.43 μg/g with an arithmetric mean of 6.09 ± 0.28 μg/g. In comparison with other PAHs studied, the most dominant PAHs in the indoor dust of Ilorin residences were fluoranthene. PAHs homolog abundance decreased in the order 4 ring > 3 ring > 6 ring > 5 ring > 2 ring. The carcinogenic potency of PAHs in Ilorin residences followed the sequence LDRA > MDRA > HDRA. Chrysene originating from combustion of coal and automobiles exhausts was the predominant compound among the seven CPAHs considered. Identification of PAHs sources using diagnostic ratios and positive matrix factorization (PMF) showed that the accumulation of PAHs in indoor dusts were influenced by emissions from indoor activities in addition to penetrating outdoor pollutants such as coal and wood combustion, electronic devices, smoke from cooking, incense burning and tobacco, traffic and petroleum products combustion, and gasoline combustion from non-traffic sources. Although the investigation of indoor PAHs cancer risks (ILCR) in adults and children that

is primarily via ingestion and dermal (skin) exposure pathways was satisfactory, the possibility of unacceptable PAHs cancer risk could arise if the current trend of PAHs emissions in indoor dusts within Ilorin residences are not controlled.

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Declarations

Ethics approval This is not applicable for this manuscript.

Consent to participate Yes

Consent for publication Yes

Competing interests The authors declare no competing interests.

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