#### **ORIGINAL PAPER**



# Distribution, Sources and Health Risks of Polycyclic Aromatic Hydrocarbons (PAHs) in Household Dusts from Rural, Semi-urban and Urban Areas in the Niger Delta, Nigeria

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#### Abstract

Dusts from rural, semi-urban and urban areas of the Niger Delta, Nigeria were investigated for their polycyclic aromatic hydrocarbon (PAH) compositional patterns and sources, and risk of human exposure to PAHs in home dusts through non-dietary ingestion, inhalation and dermal contact pathways. The PAHs in the dust samples were extracted by ultra-sonication with hexane/dichloromethane and cleaned up on a silica gel/alumina column. The concentrations of the PAHs in the extracts were determined by gas chromatography–mass spectrometry. The  $\Sigma 16$  PAH concentrations in these household dusts varied from 60.0 to 1473, 124 to 2131 and 4531 to 111,914 µg kg<sup>-1</sup> for the rural, semi-urban and urban areas, respectively. The characteristic PAH distribution pattern in the household dusts from urban areas followed the order: 4 > 6 > 5 > 3 > 2 rings, while in the semi-urban and rural areas, the distribution patterns followed the order: 3 > 6 > 4 > 5 > 2 rings and 5 > 6 > 4 > 3 > 2 rings, respectively. The benzo[*a*]pyrene carcinogenic potency concentration of PAHs in dusts from homes in these areas varied from 161 to 3288 µg kg<sup>-1</sup>, while the mutagenic potency concentration values varied between 154 and 3466 µg kg<sup>-1</sup>. The estimated lifetime cancer risk values arising from exposure to PAHs in dust in homes from rural, semi-urban and urban areas were larger than the target value of  $10^{-6}$  (one chance in a million of equally exposed persons of the risk of suffering cancer or cancer-related diseases). Principal component analysis of the results suggested that the sources of PAHs in the dust from homes included cooking fuels and traffic emissions.

Keywords Home dusts · Non-dietary exposure · Health risk · Indoor environment quality · Niger Delta · Nigeria

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# Introduction

PAHs are widespread persistent organic pollutants found in virtually all environmental matrices including dust, soil, air and water. They are produced primarily from the combustion of biomass and fossil fuels as well as the pyrosynthesis of organic materials (Yan et al. 2015). In view of their widespread nature, mode of formation and toxicity profiles, PAHs are among the top priority pollutants in the human environment. In addition, a number of PAHs are known to be carcinogenic, mutagenic, genotoxic, immunotoxic and endocrine-disrupting chemicals (Iwegbue et al. 2018).

Indoor dust is a sensitive indicator of indoor environmental quality because its large surface area allows for the accumulation and conservation of contaminants for relatively longer periods than those adsorbed by outdoor dust that are more readily subjected to degradation, leaching and dilution, among other effects (Ong et al. 2007; USEPA 2011; Iwegbue et al. 2017). Therefore, household dust can provide valuable information on chronic exposure to indoor pollutants (Orecchio 2011; Iwegbue et al. 2017). The concentrations and distribution of PAHs in indoor dust are greatly influenced by the nature of the indoor activities, such as cooking habits, tobacco smoking, heating sources (coal, electricity or wood), lighting sources (kerosene lamp, candle, gas light or electricity), types of energy sources used for cooking (electricity, liquefied petroleum gas, wood, charcoal or kerosene stoves), burning of incense, the level of infiltration of outdoor dust which is governed by ventilation types, housekeeping habits and differences in residential settings (Gevao et al. 2007; Orecchio 2011; Peng et al. 2012; Shen et al. 2011; Lv and Zhu 2013; Derudi et al. 2014; Yang et al. 2015a). Thus, investigation of the concentrations of PAHs in indoor dust is a worthwhile exercise because humans are in frequent contact with dust (Wang et al. 2017) given the length of time that they spend indoors per day (>80%). Contaminants in household dusts constitute a major threat to more susceptible groups such as the elderly, and especially infants and toddlers because of their habits (Qi et al. 2014; Iwegbue et al. 2017). Unconscious ingestion of household dust, inhalation of contaminated air and dermal absorption are established non-dietary routes of human exposure to environmental contaminants including PAHs (Gevao et al. 2006, 2007; Harrad et al. 2006).

The presence of PAHs in household dusts has been documented (Iwegbue 2011; Essumang et al. 2016; Qi et al. 2014; Yan et al. 2015; Yang et al. 2015a, b; DellaValle et al. 2016; Oluseyi et al. 2016; Wang et al. 2017; Yassin et al. 2016). However, most of these studies were centred on urban environments of America, Asia and Europe. There is a paucity of data on PAH concentrations in indoor dust from rural, semi-urban and urban areas in most African countries and especially Nigeria. This study follows on from our earlier study in which the concentrations of metals in home dusts from these areas were determined (Iwegbue et al. 2017). To the best of our knowledge, this is the first investigation on PAHs in household dusts from rural, semi-urban and urban areas in Nigeria. The objective of the study was to determine the concentrations and risk of human exposure to PAHs in household dusts from rural, semi-urban and urban areas in the Niger Delta. This information will provide a useful guide for developing strategies for monitoring the quality of the indoor environment and risk management.

Warri (latitude 5°31"N and longitude 5°45"E), Abraka (lon-

gitude 6°06"E and latitude 5°48"N) and Emu-Uno (longitude

# **Materials and Methods**

### **Study Areas**

6°6″ and 6°42″E and latitude 6°31″ and 5°25″N) represent typical urban, semi-urban and rural settings, respectively, in the Niger Delta (Fig. 1). These areas are characterized by tropical climatic conditions with well-demarcated dry (November to April) and wet seasons (May to October) every year. There are occasional rainfalls during the dry season. The average annual rainfall in the study area is 2500 mm, while the average annual minimum and maximum air temperatures are 18 and 35 °C, respectively. The anthropogenic activities, demographic and edaphic characteristics, vegetation, geological and other relief features of the study area have been previously described (Iwegbue et al. 2009, 2012, 2016a, 2017; Iwegbue and Obi 2016).

# **Sample Collection**

Dust samples were collected from 60 homes in rural, semiurban and urban areas of Delta State, Nigeria during the months of November to December, 2016. The samples were collected by gentle sweeping of dust deposits on fans, chairs, floors, tables, window edges, shelves and other cabinetry with the aid of a brush into a dustpan. The dusts were collected in substantial quantities and transferred into clean amber glass bottles. The characteristic features of the homes sampled are given in Supplementary Material Table S1. The dust samples collected in the living room, bedrooms, kitchen and staircase within a building were pooled together to form a representative sample of that home. Acetone was used to clean the brush and dustpan after each sample collection in order to avoid carryover of dust particles from one sample to another. The samples were transported in a cooler chest containing ice to the laboratory, and subsequently air-dried in the dark at room temperature, filtered to pass through a 63-µm nylon sieve and stored in amber glass bottles at 4 °C.

### Reagents

The reagents for the analysis included dichloromethane and *n*-hexane (HPLC grade) (Rieldel-de Haën, Seelze, Germany), alumina, anhydrous sodium sulphate (purity 99%), silica gel (BDH Poole, UK) and a PAH standard mixture containing the US EPA 16 priority PAHs (Supelco, Bellefonte, PA, USA).

### Sample Extraction and Clean-up

A mass of 5.0 g of each household dust was homogenized with an equal amount of anhydrous sodium sulphate (activated at 550 °C for 3 h). The resulting homogenate was extracted with 30 mL of dichloromethane (DCM) and hexane (1:1 v/v) by ultra-sonication for 15 min at 35 °C. The extract was filtered through a 0.45- $\mu$ m filter and the extraction process was carried out three times with a fresh portion



of DCM/hexane on the residue. The extracts were combined and concentrated to approximately 2 mL with a rotary evaporator. The extract was cleaned up by passing it through a silica gel/alumina packed column which was loaded from bottom to top with 4.0 g of silica gel (5% deactivated) and 2.0 g of alumina (6% deactivated). The PAHs in the extract were subsequently eluted with hexane/DCM (1:1 v/v) and evaporated to approximately 1 mL with a gentle stream of high-purity nitrogen.

# **PAH Detection and Quantification**

A gas chromatograph (Agilent 6890 N, Agilent Technologies, Santa Clara, CA, USA) equipped with an Agilent 5975 mass selective detector (MSD) was used to effect the separation, detection and quantification of PAHs in the samples. The capillary column used for the separation was a J&W DB-5 cross-linked 5% phenylmethylsiloxane column with 0.25  $\mu$ m film thickness, 0.25 mm i.d. and 30 m length (J&W, USA). The sample injection volume was 1  $\mu$ L in pulsed splitless mode. The column temperature was initially set at 45 °C for 2 min, and then increased to 120 °C at a rate of 10 °C/min and finally to 300 °C at 5 °C/min and held there for 15 min. The ion source temperature was 200 °C, while the interface temperature was 280 °C. The abundance of quantification and confirmation ions alongside the retention times of the authentic PAH standards was used to confirm the identities of the PAHs in the samples.

### **Quality Control/Assurance and Statistical Analysis**

Quality assurance and quality control of the data were performed by analysing method blanks and spiked matrix samples alongside the samples. The extraction efficiency of the target PAHs was evaluated by means of a spike recovery method. In this case, known concentrations of individual PAHs were introduced into fresh portions of selected previously analysed samples at three concentration levels and all the analysis steps from extraction to chromatographic analysis were repeated. Average recoveries of 78-103% were achieved for the individual PAH compounds. The calibration curves of the PAHs had  $r^2$  values of 0.9995–0.9999. The limits of detection (LODs) and quantification (LOQs) refer to the concentrations that give a signal-to-noise ratio of 3 and 10, respectively, obtained by analysing blank samples (n=3). The LODs and LOQs for the PAHs varied from 0.03 to 0.2 and 0.1 to 0.6 µg kg<sup>-1</sup>, respectively. Inter-house differences in the PAH concentrations and compositions were established by means of analysis of variance (ANOVA). The statistical analyses were carried out with SPSS version 15.1 software. Source identification and apportionment of PAHs in the house dusts were determined from isomeric ratios and principal component analysis.

# **Health Risk Assessment**

Assessment of the human health risk derived from exposure to PAHs in dust from these homes was based on the  $C_{UCL95\%}$ concentration because the dataset showed an approximately non-normal distribution pattern. The  $C_{UCL95\%}$  (exposure-point upper confidence limit content, mg kg<sup>-1</sup>) refers to the upper limit of the 95% confidence interval for the mean which gives a measure of the "reasonable maximum exposure" (Hu et al. 2011; US EPA 1989; Zheng et al. 2010a, b). A description of the equation and terms used for the evaluation of the  $C_{UCL95\%}$  has been given elsewhere (Kurt-Karaku 2012; Iwegbue et al. 2017).

## **Carcinogenic and Mutagenic Potency**

The carcinogenic and mutagenic potency of PAHs in the homutagenic equivalency quotients (use dusts were estimated by comparing the toxicity or carcinogenic/mutagenic potency of the individual PAHs to that of benzo[a]pyrene (BaP). The BaP carcinogenic (BaP<sub>TEQ</sub>) and BaP<sub>MEQ</sub>) for the PAH compounds were estimated by means of the following equations:

$$BaP_{TEQ} = \sum C_i \times BaP_{TEF},$$
(1)

$$BaP_{MEQ} = \sum C_{i} \times BaP_{MEF},$$
(2)

where  $BaP_{TEF}$  is the carcinogenic potency relative to BaP,  $BaP_{MEF}$  is the mutagenic potency relative to BaP, and  $C_i$ is the concentration of the individual PAH compound. The values of the BaP carcinogenic (BaP<sub>TEF</sub>) and mutagenic (BaP<sub>MEF</sub>) equivalency factors for the seven carcinogenic PAHs are given in Table 1.

#### **Evaluation of Non-carcinogenic Risk**

The non-cancer risk expressed in terms of the hazard index (HI) is the sum of the hazard quotients (HQs) associated with human exposure to PAHs via non-dietary ingestion, dermal contact and inhalation pathways. The chronic daily intake (CDI) for the three exposure routes was based on the UCL95% concentrations of NaP, Acy, Ace, Flu, Phe, Ant, Flt and Pyr.

The CDI values for the three main exposure routes were calculated as follows:

$$CDI_{\text{ingestion}} = \frac{C_{\text{UCL}} \times \text{IngR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times 10^{-6},$$
 (3)

$$CDI_{inhalation} = \frac{C_{UCL} \times InhR \times EF \times ED}{PEF \times BW \times AT},$$
(4)

$$CDI_{dermal} = \frac{C_{UCL} \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6}.$$
(5)

Hazard index (HI) = 
$$\sum HQ = HQ_{ing} + HQ_{inh} + HQ_{derm}$$
,  
HQ =  $\frac{CDI_{nc}}{RfD}$ , (6)

where  $\text{CDI}_{\text{ingestion}}$ ,  $\text{CDI}_{\text{inhalation}}$  and  $\text{CDI}_{\text{dermal}}$  are the chronic daily intake for ingestion, inhalation and dermal contact, respectively, and  $C_{\text{UCL}}$  is the 95% UCL concentration. Under most programmes, if the HI value is less than 1, the exposed population is unlikely to experience considerable non-carcinogenic effects. If the HI values are greater than 1, the exposed population is likely to experience considerable non-carcinogenic effects.

#### **Evaluation of Incremental Lifetime Cancer Risk**

Residents of these homes are exposed to contaminants in indoor dust via three major pathways including non-dietary ingestion, dermal contact and inhalation of dust particles (IDI) (USEPA 2009). The incremental lifetime cancer risk (ILCR) of a resident's exposure to PAHs in house dust from these areas was evaluated as the sum of the individual risks from these three exposure routes. The ILCR in terms of IDI was calculated by following the model equations (Eqs. 6–9) and parameters with modifications (Table 1) of the United States Environmental Protection Agency (USEPA 1989, 2009).

$$ILCR_{ing} = \frac{C_{UCL} \times IngR \times EF \times ED \times CF \times SFO}{BW \times AT}$$
(7)

ILCR<sub>derm</sub>

$$=\frac{C_{UCL} \times SA \times AF_{soil} \times ABS \times EF \times ED \times CF \times SFO \times ABS_{GI}}{BW \times AT}, (8)$$

$$ILCR_{inh} = \frac{C_{UCL} \times EF \times ET \times ED \times IUR}{PEF \times AT^*}.$$
(9)

In this work, the dermal absorption factor (ABS) was taken as 0.13, the exposure frequency (EF) was 350 days/ yr, the exposure time (ET) was 24 h/day, the averaging time for non-carcinogenic risk ( $AT_{nc}$ ) is the exposure duration (ED) × 365, the averaging time for carcinogenic risk ( $AT_{ca}$ ) is the lifetime (LT) × 365, the particulate emission factor (PEF) is  $1.36 \times 10^9$  m<sup>3</sup>/kg, LT is 54.4 years, and the conversion factor (CF) is  $1.0 \times 10^{-6}$ . ILCR<sub>ing</sub>, ILCR<sub>derm</sub>, ILCR<sub>inh</sub> are the incremental lifetime cancer risk via ingestion, dermal contact and inhalation of dust particles, respectively. The qualitative ranking/significance of the lifetime cancer risks is given as follows: a

| Table 1 | Toxicological | parameters and | values of | variables | for estimation of | of human | health risk | assessment |
|---------|---------------|----------------|-----------|-----------|-------------------|----------|-------------|------------|
|---------|---------------|----------------|-----------|-----------|-------------------|----------|-------------|------------|

| PAHs | Toxicological param | eters of the investig | ated PAHs used fo | r health risk assessment |
|------|---------------------|-----------------------|-------------------|--------------------------|

|            | Oral ingestion reference dose (RfD <sub>o</sub> ) | Inhalation refer-<br>ence dose (RfD <sub>i</sub> ) | SFO <sub>ing</sub> (mg/kg/d) | IUR ( $\mu g/m^3$ )  | ABS <sub>GI</sub>  | BaP <sub>TEF</sub> | BaP <sub>MEF</sub> |
|------------|---|--|------------------------------|----------------------|--------------------|--------------------|--------------------|
| Nap        | $2 \times 10^{-2}$                                | $8.57 \times 10^{-4}$                              |                              |                      |                    |                    |                    |
| Acy        | $6 \times 10^{-2}$                                | $6 \times 10^{-2}$                                 |                              |                      |                    |                    |                    |
| Ace        | $6 \times 10^{-2}$                                | $6 \times 10^{-2}$                                 |                              |                      |                    |                    |                    |
| Flu        | $4 \times 10^{-2}$                                | $4 \times 10^{-2}$                                 |                              |                      |                    |                    |                    |
| Phen       | $3 \times 10^{-2}$                                | $3 \times 10^{-2}$                                 |                              |                      |                    |                    |                    |
| Ant        | $3 \times 10^{-1}$                                | $3 \times 10^{-1}$                                 |                              |                      |                    |                    |                    |
| Flt        | $4 \times 10^{-2}$                                | $4 \times 10^{-2}$                                 |                              |                      |                    |                    |                    |
| Pyr        | $3 \times 10^{-1}$                                | $3 \times 10^{-1}$                                 |                              |                      |                    |                    |                    |
| BaA        |   |  | $7.3 \times 10^{-1}$         | $1.1 \times 10^{-4}$ | 1                  | 0.1                | 0.082              |
| Chry       |   |  | $7.3 \times 10^{-3}$         | $1.1 \times 10^{-5}$ | 1                  | 0.001              | 0.017              |
| BbF        |   |  | $7.3 \times 10^{-1}$         | $1.1 \times 10^{-4}$ | 1                  | 0.1                | 0.25               |
| BkF        |   |  | $7.3 \times 10^{-2}$         | $1.1 \times 10^{-4}$ | 1                  | 0.01               | 0.11               |
| BaP        |   |  | 7.3                          | $1.1 \times 10^{-3}$ | 1                  | 1                  | 1                  |
| IndP       |   |  | $7.3 \times 10^{-1}$         | $1.1 \times 10^{-4}$ | 1                  | 0.1                | 0.31               |
| DahA       |   |  | 7.3                          | $1.2 \times 10^{-3}$ | 1                  | 1                  | 0.29               |
| References | USEPA (2012)                                      | USEPA (2012)                                       | USDOE (2011)                 | USEPA (2010)         | USEPA (2011)       | USEPA (2012)       | Durant (1996)      |
| PAHs       | Values of variables for                           | or estimation of hu                                | ıman health risk ass         | sessment             |                    |                    |                    |
|            | Variables   | Units  | Infant                       | Toddler              | Child              | Teen               | Adult              |
| Nap        | Age   | years  | 0–0.5                        | 0.6–5                | 6–12               | 13–20              | 21-65              |
| Acy        | Exposure duration (ED)                            | years  | 0.5                          | 4.5                  | 7.0                | 8.0                | 34.5               |
| Ace        | Body weight (BW)                                  | kg   | 8.2                          | 16.5                 | 32.9               | 59.7               | 65.0               |
| Flu        | Soil ingestion rate<br>(IngR)                     | mg/day   | 200                          | 200                  | 200                | 100                | 100                |
| Phen       | Soil to skin adherence factor                     | e mg/cm <sup>2</sup>                               | 0.2                          | 0.2                  | 0.2                | 0.07               | 0.07               |
| Ant        | Skin surface area                                 | $m^2$  | 203                          | 344                  | 586                | 908                | 1030               |
| Flt        | Inhalation rate                                   | m <sup>3</sup> /day                                | 2.0                          | 5.0                  | 12.0               | 21.0               | 50.0               |
| Pyr        |   |  |                              |                      |                    |                    |                    |
| BaA        |   |  |                              |                      |                    |                    |                    |
| Chry       |   |  |                              |                      |                    |                    |                    |
| BbF        |   |  |                              |                      |                    |                    |                    |
| BkF        |   |  |                              |                      |                    |                    |                    |
| BaP        |   |  |                              |                      |                    |                    |                    |
| IndP       |   |  |                              |                      |                    |                    |                    |
| DahA       |   |  |                              |                      |                    |                    |                    |
| References |   |  | Rout et al. (2013)           | Rout et al. (2013)   | Rout et al. (2013) | Rout et al. (2013) | Rout et al. (2013) |

value  $\leq 10^{-6}$  is considered a very low risk; greater than  $10^{-6}$  and less than or equal to  $10^{-4}$  a low risk; greater than  $10^{-4}$  and less than or equal to  $\leq 10^{-3}$  a moderate risk; greater than  $10^{-3}$  and less than  $10^{-1}$  a high risk and values greater than  $10^{-1}$  a considerable risk.

# **Results and Discussion**

# **Concentrations and Compositional Patterns of PAHs**

The results for the determination of the PAH concentrations in household dusts are displayed in Table 2. The  $\Sigma 16$ PAH concentrations in the dust samples from the urban,

Table 2PAH concentrations ( $\mu g k g^{-1}$ ) in indoor dust from three residential environments

|               | Rural (r | n = 20)  |        |        |      |        | Semi-urban $(n=20)$ |      |        |         |      |        |
|---------------|----------|----------|--------|--------|------|--------|---------------------|------|--------|---------|------|--------|
|               | Mean     | SD       | Median | Min    | Max  | UCL    | Mean                | SD   | Median | Min     | Max  | UCL    |
| Nap           | 41.0     | 45.7     | 13.0   | ND     | 110  | 47     | 17.2                | 12.3 | 14.0   | ND      | 39   | 19.5   |
| Acy           | 10.3     | 14.7     | 7.0    | ND     | 48.0 | 16     | 41.6                | 27.4 | 47.5   | 4.0     | 84   | 43.2   |
| Ace           | 8.6      | 11.1     | 4.0    | 1.0    | 30.0 | 12.3   | 60.3                | 47.7 | 70.0   | 5.0     | 136  | 62.5   |
| Flu           | 5.3      | 5.4      | 3.0    | 1.0    | 15.0 | 7.65   | 116                 | 62.0 | 125    | 5.0     | 210  | 115    |
| Phen          | 32.1     | 33.1     | 18.5   | 2.0    | 95.0 | 38.9   | 140                 | 78.1 | 144    | 2.0     | 238  | 137    |
| Ant           | 9.9      | 8.4      | 8.0    | 1.0    | 30.0 | 13     | 6.8                 | 4.21 | 8.0    | ND      | 11   | 8.17   |
| Flt           | 14.0     | 23.5     | 4.0    | ND     | 75.0 | 22.5   | 120                 | 78.3 | 126    | 2.0     | 221  | 119    |
| Pyr           | 8.4      | 9.4      | 4.0    | 1.0    | 31.0 | 11.9   | 14.3                | 12.4 | 10.5   | 1.0     | 38   | 17.2   |
| BaA           | 17.4     | 23.0     | 7.0    | 1.0    | 75.0 | 24.2   | 48.4                | 25.1 | 51.0   | ND      | 82   | 49.4   |
| Chry          | 38.8     | 47.2     | 25.5   | 2.0    | 158  | 50.9   | 55.2                | 43.2 | 51.0   | ND      | 121  | 59.2   |
| BbF           | 107      | 166      | 40.0   | 2.0    | 484  | 142    | 57.6                | 45.1 | 44.0   | ND      | 137  | 63.5   |
| BkF           | 76.6     | 87.1     | 44.5   | 1.0    | 235  | 89.7   | 60.2                | 62.5 | 36.0   | 4.0     | 203  | 71.5   |
| BaP           | 165      | 180      | 103    | 1.0    | 610  | 204    | 64.6                | 50.4 | 66.5   | 3.0     | 147  | 67.9   |
| DahA          | 93.0     | 45.2     | 102    | 11.0   | 144  | 90.8   | 111                 | 96.0 | 76.0   | 11.0    | 266  | 119    |
| IndP          | 104      | 96.6     | 60.0   | 8.0    | 269  | 117    | 60.9                | 49.0 | 35.0   | ND      | 149  | 68.9   |
| BghiP         | 103      | 95.3     | 71.5   | 2.0    | 298  | 822    | 247                 | 107  | 272    | ND      | 400  | 244    |
| Total         | 828      | 445      | 870    | 60.0   | 1473 |        | 1121                | 668  | 1263   | 124     | 2131 |        |
| Σ7C*          | 601      | 365      | 674    | 38.0   | 1117 |        | 413                 | 280  | 439.5  | 90.0    | 909  |        |
| Σ2-rings      | 41.0     | 45.7     | 13.0   | ND     | 110  |        | 15.5                | 12.8 | 13.0   | 0.0     | 39   |        |
| Σ3-rings      | 65.2     | 59.0     | 40.5   | 10.0   | 199  |        | 361                 | 200  | 394    | 20.0    | 591  |        |
| Σ4-rings      | 77.2     | 86.0     | 43.0   | 9.0    | 267  |        | 222                 | 148  | 255    | 3.0     | 426  |        |
| Σ5-rings      | 452      | 314      | 490.0  | 12.0   | 905  |        | 214                 | 175  | 176    | 7.0     | 522  |        |
| Σ6-rings      | 196      | 130      | 156.0  | 13.0   | 419  |        | 308                 | 211  | 295    | 60.0    | 636  |        |
|               |          | Urban (n | =20)   |        |      |        |                     |      |        |         |      |        |
|               |          | Mean     |        | SD     |      | Median |                     | Min  |        | Max     |      | UCL    |
| Nap           |          | 5025     |        | 4565   |      | 6335   |                     | ND   |        | 12,423  |      | 5103   |
| Acy           |          | 1411     |        | 804    |      | 1210   |                     | ND   |        | 2711    |      | 1465   |
| Ace           |          | ND       |        | ND     |      | ND     |                     | ND   |        | ND      |      | ND     |
| Flu           |          | 1354     |        | 1255   |      | 935.5  |                     | ND   |        | 4301    |      | 1451   |
| Phen          |          | 7705     |        | 3331   |      | 7477   |                     | ND   |        | 14852   |      | 7784   |
| Ant           |          | ND       |        | ND     |      | ND     |                     | ND   |        | ND      |      | ND     |
| Flt           |          | 688      |        | 911    |      | 424    |                     | ND   |        | 3760    |      | 810    |
| Pyr           |          | NA       |        | NA     |      | NA     |                     | NA   |        | NA      |      | NA     |
| BaA           |          | 876      |        | 736    |      | 553    |                     | ND   |        | 2604    |      | 921    |
| Chry          |          | 15141    |        | 11873  |      | 13,766 |                     | ND   |        | 49,219  |      | 15,965 |
| BbF           |          | 1813     |        | 2286   |      | 865    |                     | ND   |        | 9319    |      | 2082   |
| BkF           |          | 4088     |        | 4222   |      | 3414.5 |                     | ND   |        | 14,450  |      | 4468   |
| BaP           |          | 789      |        | 278    |      | 920    |                     | ND   |        | 1118    |      | 785    |
| DahA          |          | 2270     |        | 2339   |      | 1445   |                     | ND   |        | 8879    |      | 2493   |
| IndP          |          | 1812     |        | 1489   |      | 2227   |                     | ND   |        | 5061    |      | 1893   |
| BghiP         |          | 11,437   |        | 18,601 |      | 4843   |                     | ND   |        | 75,808  |      | 14,064 |
| Total         |          | 42,177   |        | 31,268 |      | 31,604 |                     | 4531 |        | 111,914 |      |        |
| $\Sigma 7C^*$ |          | 22,862   |        | 14,752 |      | 20,622 |                     | 911  |        | 54,271  |      |        |
| Σ2-rings      |          | 5025     |        | 4565   |      | 6335   |                     | ND   |        | 12,423  |      |        |
| Σ3-rings      |          | 5604     |        | 5474   |      | 4644   |                     | ND   |        | 15,795  |      |        |
| Σ4-rings      |          | 15,626   |        | 12,204 |      | 15,060 |                     | ND   |        | 49,643  |      |        |
| Σ5-rings      |          | 7005     |        | 6063   |      | 5428   |                     | 172  |        | 24,658  |      |        |

| Table 2 (continue) | ed)            |        |        |     |        |     |
|--------------------|----------------|--------|--------|-----|--------|-----|
|                    | Urban $(n=20)$ | )      |        |     |        |     |
|                    | Mean           | SD     | Median | Min | Max    | UCL |
| Σ6-rings           | 11,681         | 18,859 | 5768   | ND  | 78,035 |     |
|                    |                |        |        |     |        |     |

\*Σ7C refers to the sum of the seven carcinogenic PAHs

semi-urban and rural areas varied from 4531 to 111,914, 124 to 2131 and 60.0 to 1473  $\mu$ g kg<sup>-1</sup>, respectively. Analysis of variance (p < 0.05) indicates that the total PAH concentrations and compositions differed significantly among the households investigated in each area, while Tukey's test at p < 0.05 suggests that the differences in the mean concentrations of PAHs from the three areas are significant. The differences in the concentrations and compositions may have been influenced by variations in indoor activities, nature of the buildings and proximity to high-density traffic routes. In the urban area, dusts from buildings with electricity generators located in their corridors or nearer to the buildings showed larger PAH concentrations than those with electricity generators located far away from the buildings; likewise the influence of proximity to high-density traffic routes and kitchen location. In the homes that were sampled, a combination of gas- and kerosene-powered cooking systems were used. However, in the urban area few of the investigated buildings had the kitchen detached from the main building (room and parlour system) where a combination of wood, charcoal, kerosene or gas cooking systems are typically used. The levels of PAHs in the household dusts from the urban area were higher than those found in outdoor dust and soils from the same study area (Iwegbue and Obi 2016; Iwegbue et al. 2016). This may be due to outdoor dilution effects, as well as the influence of the indoor physical and chemical characteristics that protect PAHs from degradation processes such as photolysis and weathering that typically take place in the outdoor environment (Paustenbach et al. 1997; Wang et al. 2017). In addition, household dusts contain larger numbers of smaller particles alongside their high organic carbon content which could aid increased PAH adsorption than on their outdoor counterparts, soils and sediments (Molhave et al. 2000; Wu et al. 2005; Wang et al. 2017). Significantly higher concentrations of PAHs were found in home dusts from the urban area than those of the semi-urban and rural areas. This may be related to the fact that the kitchens in most of the homes sampled in the rural zone were detached from the living buildings and that there was relatively low traffic volume and industrial activity near the homes. Secondly, the majority of the houses in the rural zones had no fans, and their doors and windows were virtually open during most of the day, which could lead to a greater circulation of air within the indoor environment. This may lead to dilution and photodegradation of the PAHs since sunlight can easily penetrate into the house through the open windows and doors. In rural areas, buildings with their kitchens closer to the main house should show elevated concentrations compared with those with their kitchens far away from the living building.

The fuel for cooking in the rural area is exclusively wood. A comparison of the PAH concentrations in household dusts of this study with those previously reported for a wide variety of homes, but possibly determined following different extraction and chromatographic techniques, is shown in Table 3. The PAH concentrations in our samples fall into the global concentration range found in indoor environments (Table 3).

The PAH distribution in the home dusts from the urban area was in the order: 4 > 6 > 5 > 3 > 2 rings, while in the semi-urban and rural areas, the distribution patterns followed the order: 3 > 6 > 4 > 5 > 2 rings and 5 > 6 > 4 > 3 > 2 rings, respectively. The low molecular weight (2-3 ring) PAHs constituted 0.0-44.7%, while the high molecular weight (4-6 ring) PAHs constituted 55.3-100% of the total PAHs in the dust samples from these areas. The high molecular weight PAHs (HMW) showed dominance over the low molecular weight PAHs (LMW) in these household dusts, which may be related to the fact that LMW tend to be associated with gas-phase partitioning, whereas HMW tend to be associated with particulate phases as a result of their lipophilic characteristics (Ma et al. 2011; Orecchio 2011; Li et al. 2013; Wang et al. 2017). The 4-ring PAHs are the dominant PAH homologues in the household dust samples from the urban area with chrysene having the highest concentration and occurrence frequency. Of the total concentrations of 16 PAHs, 4-ring PAHs constituted up to 92.4%. Chrysene as an individual compound accounts for 0.0-91.7% of the concentrations of total PAHs in these home dusts. In the case of household dust from the semi-urban and rural areas, 3- and 5-ring PAHs are the respective dominant homologues. Phenanthrene was the dominant PAH in dust from homes in semiurban areas and contributed 1.6-20.5% of the total PAH concentrations. However, benzo(b)fluoranthene was the most prevalent compound in the home dusts from the rural areas, and constituted 1.8-32.9% of the total PAH concentrations. The 5- and 6-ring PAHs are responsible for 5.2–100% of the  $\Sigma 16$  PAH concentrations in dusts from urban homes, and benzo(k)fluoranthene and benzo(g, h, i) perylene were the respective dominant 5- and 6-ring homologues, while the

 Table 3
 Comparison of PAH concentrations in indoor dust with those reported in other parts of the world

| Country             | Location            | Sampling year                 | Sampler | No. of samples | particle size | No. of<br>PAHs | $\sum$ PAH (ng/g)  | BaP <sub>TEQ</sub><br>(ng/g) | References                            |
|---------------------|---------------------|-------------------------------|---------|----------------|---------------|----------------|--------------------|------------------------------|---------------------------------------|
| Nigeria             | Emu-Unor            | 2016                          | HB      | 20             | <63 µm        | 16             | 60.0–1473          | 347                          | This study                            |
| Nigeria             | Abraka              | 2016                          | HB      | 20             | <63 µm        | 16             | 124–2131           | 161                          | This study                            |
| Nigeria             | Warri               | 2016                          | HB      | 20             | <63 µm        | 15             | 4531–111,914       | 3288                         | This study                            |
| Australia           | Brisbane            | 2003                          | NA      | 11             | <1 mm         | 14             | 7440               | 106                          | Ayoko et al. (2005)                   |
| Brazil              |                     | 2008                          | HB      | 9              | NA            | 16             | 4091               | 288                          | Coronas et al. (2013)                 |
| Canada              | Ottawa              | 2002–2003                     | VC      | 51             | <150 µm       | 13             | 29,300             | 4724                         | Maertens et al. (2008)                |
| China               | Shanghai            | 2005                          | HB      | 27             | NA            | 16             | 20,674             | 4393                         | Ren et al. (2006)                     |
| China               | Hong Kong           | b                             | VC      | 55             | <100 µm       | 16             | 6070               | 635                          | Kang et al. (2010)                    |
| China               | Pearl Delta         | 2010                          | VC      | 55             | <100 µm       | 16             | 5910               | 345                          | Kang et al. (2011)                    |
| China               | Shanghai            | 2010                          | HB      | 22             | NA            | 16             | 11,575             | 829                          | Peng et al. (2012)                    |
| China               | Guangzhou           | 2010                          | VC      | 20             | <100 µm       | 16             | 5916               | 788                          | Wang et al. (2013a)                   |
| China               | Guangzhou           | 2011-2012                     | VC      | 70             | <100 µm       | 16             | 8130               | 843                          | Wang et al. (2013b)                   |
| China               | Qingyang            | 2011-2012                     | VC      | 70             | <100 µm       | 16             | 34,800             | 3446                         | Wang et al. (2013b)                   |
| China               | Changchun           | 2014–2015                     | HB      | 31             | NA            | 16             | 21,800–<br>329,600 | NA                           | Wang et al. (2017)                    |
| China               | 23 Cities           | 2010                          | HB      | 81             | NA            | 16             | 1000-470,000       | NA                           | Qi et al. (2014)                      |
| China               | Guizhou             | 2012                          | HB      | 88             | 80 µm         | 18             | 2180-14,200        | NA                           | Yang et al. (2015a)                   |
| China               | Xinxiang            | 2012                          | PC      | 20             | NA            | 16             | 1470-21,800        | NA                           | Yang et al. (2015b)                   |
| Germany             | Berlin              | 1997–98,<br>2000 <sup>c</sup> | VC      | 123            | Fine dust     | 18             | 6140 <sup>a</sup>  | 485                          | Fromme et al. (2004)                  |
| Ghana               | Cape coast/<br>KEEA | 2012                          | TS      | 10             | NA            | 15             | ND-3240            | 0.25–<br>218                 | Essumang et al. (2016)                |
| Greece              |                     | 2010                          | VC      | 11             | <62 µm        | 16             | 397                | 19                           | Christopoulou et al. (2012)           |
| Italy               | Palermo             | 2006                          | HB      | 45             | NA            | 16             | 5111               | 262                          | Mannino and Orec-<br>chio (2008)      |
| Kuwait              |                     | 2004                          | VC      | 24             | <63 µm        | 15             | 540                | 162                          | Gevao et al. (2007)                   |
| Kuwait              |                     | 2014                          | HB      | 13             | NA            | 16             | 1100               | NA                           | Yassin et al. (2016)                  |
| Nigeria             | Lagos               | 2016                          | HB      |                | NA            | 16             | 904–7677           | 41–719                       | Oluseyi et al. (2016)                 |
| Nigeria             | Delta State         | 2009                          | HB      | 30             | NA            | 17             | 127                | NR                           | Iwegbue (2011)                        |
| Poland              | Warsaw              | 2003-2004                     | VC      | 48             | <150 µm       | 16             | 35,030             | 2389                         | Tatur et al. (2009)                   |
| United King-<br>dom | Cambridge           | 2010                          | VC      | 1              | <63 µm        | 15             | 5095               | 345                          | Anders et al. (2012)                  |
| USA                 | Ohio                | 1992–1993                     | HVS3    | 24             | <150 µm       | 19             | 115,817            | 15,530                       | Chuang et al. (1993, 1995)            |
| USA                 | WA                  | 1992-1993                     | HVS3    | 9              | <150 µm       | 16             | 10,249             | 1235                         | Chuang et al. (1994)                  |
| USA                 | TX                  | 1993                          | HVS3    | 15             | < 53 µm       | 15             | 1715               | 198 <sup>c</sup>             | Mukerjee et al.<br>(1997)             |
| USA                 | NC                  | 1994                          | HVS3    | 24             | <150 µm       | 19             | 4200               | 439                          | Chuang et al. (1999)                  |
| USA                 | NC                  | 1995                          | HVS3    | 4              | <150 µm       | 19             | 3936               | 421                          | Chuang et al.<br>(1997a)              |
| USA                 | MD                  | 1995–1996                     | HVS3    | 126            | <150 µm       | 11             | 81,190             | 12,169                       | Egeghy et al. (2005);<br>USEPA (2011) |
| USA                 | KY                  | 1995–1996                     | HVS3    | 3              | <150 µm       | 19             | 3034               | 327                          | Chuang (1996)                         |
| USA                 | NC                  | 1996                          | HVS3    | 13             | <150 µm       | 19             | 3230               | 286                          | Chuang et al. (1997b)                 |
| USA                 | NC                  | 1996                          | HVS3    | 25             | <150 µm       | 10             | 20,100             | 3268                         | Lewis et al. (1999)                   |
| USA                 | AZ                  | 1996                          | HVS3    | 22             | <62 µm        | 19             | 1769               | 200                          | Chuang et al.<br>(1997b)              |
| USA                 | NC                  | 1997                          | HVS3    | 10             | <150 µm       | 19             | 2729               | 351                          | Wilson et al. (2001)                  |

| Country | Location        | Sampling<br>year                | Sampler | No. of samples | particle size | No. of<br>PAHs | $\sum$ PAH (ng/g) | BaP <sub>TEQ</sub><br>(ng/g) | References              |
|---------|-----------------|---------------------------------|---------|----------------|---------------|----------------|-------------------|------------------------------|-------------------------|
| USA     | NC              | 1997                            | HVS3    | 13             | <150 µm       | 19             | 2180              | 267                          | Wilson et al. (2003)    |
| USA     | MI/IA/CA/<br>WA | 1998–2000                       | VC      | 616            | <150 µm       | 7              | 8570              | 2103                         | Camann et al. (2002)    |
| USA     | MA              | 1999                            | VC      | 6              | <150 µm       | 2              | 5810              | 3191                         | Rudel et al. (2003)     |
| USA     | MA              | 1999–2001                       | VC      | 120            | <150 µm       | 4              | 5761              | 1680                         | Fromme et al. (2004)    |
| USA     | CA              | 2003-2005                       | HVS3    | 68             | <150 µm       | 9              | 383 <sup>a</sup>  | 63                           | Whitehead et al. (2012) |
| USA     | CA              | 2005-2007                       | HVS3    | 132            | NA            | 16             | 810 <sup>a</sup>  | 78                           | Hoh et al. (2012)       |
| USA     | CA              | 2001–2007                       | HVS3/VC | 583            | <150 µm       | 9              | 425 <sup>a</sup>  | 72                           | Whitehead et al. (2011) |
| USA     | CA              | 2001–2007,<br>2010 <sup>d</sup> | VC      | 494            | <150 µm       | 12             | 857 <sup>a</sup>  | 71                           | Whitehead et al. (2013) |
| USA     | TX              | 2008                            | HVS3    | 23             | <150 µm       | 16             | 73,521            | 7449                         | Mahler et al. (2010)    |
| USA     | CO              | 2010                            | VC      | 3              | <63 µm        | 15             | 3358              | 126                          | Anders et al. (2012)    |

Adopted from Ma and Harrad (2015) with some modifications

HVS3 high-volume small surface sampler, VC vacuum cleaner, HB hand brushing, PC pre-cleaned cotton, TS teflon sheet, NA not available, NR not reported

<sup>a</sup>Median value instead of average value

Table 3 (continued)

<sup>b</sup>The year of sampling was not provided

<sup>c</sup>Year 1999 was used for regression analysis

<sup>d</sup>Year 2005 was used for regression analysis

3-ring PAH homologues contributed less than 24% of the  $\Sigma 16$  PAHs in the dust from urban homes. Of the 3-ringed PAHs in the dust from the urban homes, phenanthrene was present in the highest concentration, while fluorene had the highest occurrence frequency. In this study, 3-ring PAHs, such as anthracene, acenaphthene and acenaphthylene, were not detected or found only in a limited number of dust samples from the urban area. This supports the fact that volatilization controls the fate of LMW, while PAHs with molecular weights above 200 are mostly associated with the particulate phase (Orecchio 2011). On the contrary, most of the 3-ringed PAHs were detected in dust samples from rural and semi-urban homes. The characteristic dominant compounds in the semi-urban and urban household dust samples were Phen, Chry, BkF and BghiP for 3-, 4-, 5- and 6-ring PAH homologues, respectively, while Phen, BaA, BbF and BghiP were the characteristic dominant 3-, 4-, 5- and 6-ring PAH compounds in dusts from homes in the rural area. This points to the fact that the fate and sources of PAHs in dusts from rural homes are entirely different from those of semiurban and urban areas. Nevertheless, the observed patterns in home dusts from semi-urban and urban areas correspond with the trend found in dusts from urban homes in Kuwait (Yassin et al. 2016).

# **Risk Assessment of PAHs in Household Dust**

### **Carcinogenic and Mutagenic Potency**

The carcinogenic and mutagenic potency of PAHs in the indoor samples expressed in terms of BaP<sub>TEO</sub> and  $BaP_{MEO}$  are shown in Table 4. The  $BaP_{TEO}$  and  $BaP_{MEO}$ were based on the UCL95% of the seven carcinogenic PAHs (PAH<sub>7C</sub>). The BaP<sub>TEO</sub> for dusts from homes from these areas varied from 161 to 3288  $\mu$ g kg<sup>-1</sup>, while the  $BaP_{MEO}$  values varied between 154 and 3466 µg kg<sup>-1</sup>. The  $BaP_{TEO}$  and  $BaP_{MEO}$  values for the dusts followed the order: urban > rural > semi-urban. The order of importance of the PAH<sub>7C</sub> to the BaP<sub>TEO</sub> values of home dusts from the urban area followed the order: DahA > BaP > IndP > BbF > BaA > BkF > Chry. This suggests that DahA, BaP and IndP are the main contributors to the carcinogenic potency of these household dusts, whereas the PAH compounds, BaP, IndP, DahA, BbF and BkF, are the main contributors to the mutagenic potency of the dusts from the investigated homes in the urban areas. The  $BaP_{TEO}$  and  $BaP_{MEO}$ values obtained for the household dusts from the urban area were far higher than those reported for outdoor dusts and soils from this area (Iwegbue and Obi 2016; Iwegbue et al. 2016). In the case of rural and semi-urban areas, BaP

and DahA were the main compounds responsible for the carcinogenic potency, while BaP was the main factor in the mutagenic potency of these dust samples. The  $BaP_{TEQ}$  values of the investigated household dusts from these areas correspond to the global temporal trends reported for indoor environments in the literature (Table 3).

### **Non-carcinogenic Risk**

The non-cancer risk was expressed in terms of the total hazard index (HI), which is the sum of the hazard quotient (HQ) values for the non-dietary ingestion (HQ<sub>ing</sub>), dermal contact (HQ<sub>derm</sub>) and inhalation (HQ<sub>inh</sub>) exposure pathways. The estimates of the HQ values for the three exposure pathways were based on the UCL95% concentration of the eight non-carcinogenic PAHs (NaP, Acy, Ace, Flu, Phen, Ant, Flt and Pyr). The HQ values for human exposure to PAHs in house dust from these areas followed the order:  $HQ_{ing} \gg HQ_{derm} > HQ_{inh}$  (Table 5). The non-dietary ingestion route contributes more than 85% to the total HI values. The HI values for the different age groups varied from  $1.47 \times 10^{-2}$  to 12.4 (Table 5). The HI values for human exposure to PAHs in dust in these homes followed the order: HI urban > HI rural > HI semi-urban. The HQ<sub>ing</sub> values for infants, toddlers and children were greater than those of teens and adults. This is related to the hand-to-mouth habits of these age groups. The HI values for PAHs in dust from urban homes were greater than 1, which suggests considerable non-carcinogenic risks associated with human exposure to these dust particles. However, HI values for PAHs in dusts from homes in the rural and semi-urban areas were less than 1. which implied that there was no considerable non-cancer risk associated with human exposure to PAHs in these house dusts. The PAH compounds NaP and Phen were the main contributors to the HI values of dusts from the rural and urban areas, whereas Flu, Phen and Flt were the major contributors to the HI values of dust in the semi-urban areas.

### **Incremental Lifetime Cancer Risk**

The ILCR reflects the age-specific potential cancer risk associated with human exposure to environmental PAH sources via non-dietary ingestion, inhalation of dust particles and dermal contact routes. The probabilistic cancer risks associated with exposure of adults and children residing in these homes to PAHs in dusts are shown in Table 5. The total cancer risk values for all human age groups in these areas ranged from  $2.3 \times 10^{-3}$  to  $5.76 \times 10^{-1}$ . The total cancer risk values for PAHs in household dust from these areas followed the order: urban  $\gg$  rural > semi-urban. The cancer risks associated with ingestion were of a higher magnitude than those of dermal and inhalation routes for all human age groups, which suggests that the ingestion pathway contributes significantly to the cancer risks. The cancer risks associated with children's exposure to PAHs in home dusts via ingestion and dermal contact routes were three times higher than the risk associated with the inhalation route. The cancer risk values associated with the ingestion and dermal contact pathways were greater than the accepted low risk value of  $10^{-6}$ . The cancer risk for children via the non-dietary ingestion route was higher than that of adults. This may be related to the hand-to-mouth habits of children. In addition, the PAH intake by children is greater than that of adults because of their smaller body weights. The total cancer risk values obtained for children and adult residents of the investigated homes were greater than the acceptable risk value of  $10^{-6}$ , which indicates a high potential carcinogenic risk in these areas from a single exposure source. BaP and DahA are the main contributors to the cancer risk values of dusts from homes in the study areas. The cancer risk values were higher than those obtained from exposure to PAHs in outdoor dust and soil in the same study area (Iwegbue and Obi 2016; Iwegbue et al. 2016a). The ILCR values suggested that there are 74-133,000 chances for children and 51-92,600 chances for adults in one million of equally exposed individuals of the risk of developing cancer or cancer-related illnesses in the study area. The result affirms the need for

|            | BaA  | Chry | BbF  | BkF  | BaP  | IndP | DahA | BaPTEQ |
|------------|------|------|------|------|------|------|------|--------|
| Rural      | 2.42 | 0.05 | 14.2 | 0.90 | 204  | 9.08 | 117  | 347    |
| Semi-urban | 4.94 | 0.06 | 6.35 | 0.71 | 67.9 | 11.9 | 68.9 | 161    |
| Urban      | 92.1 | 16.0 | 208  | 44.7 | 785  | 249  | 1893 | 3288   |
|            | BaA  | Chry | BbF  | BkF  | BaP  | IndP | DahA | BaPMEQ |
| Rural      | 1.99 | 0.87 | 35.5 | 9.86 | 204  | 28.2 | 33.8 | 314    |
| Semi-urban | 4.05 | 1.01 | 15.9 | 7.86 | 67.9 | 36.9 | 20.0 | 154    |
| Urban      | 75.5 | 271  | 520  | 492  | 785  | 773  | 549  | 3466   |

Table 4 $BaP_{TEQ}$  and  $BaP_{MEQ}$  of PAHs in indoor dust

remedial actions in these homes in order to minimize the risk of exposure to these contaminants.

# **PAH Source Apportionment**

### **Isomeric Ratios**

Source identification and apportionment provides useful information on the fate and transport of PAHs in indoor environments. In addition, source identification is useful for source control and risk minimization. A number of isomeric ratios, such as Ant/(Ant+Phen), BaA/(BaA+Chry), Flt/(Flt + Pyr) and IndP/(IndP + BghiP), and other indices including LMW/HMW, **SCOMB/TPAH** and PAH4/ PAH(5+6), have been used to differentiate between petrogenic and pyrogenic sources (Jamhari et al. 2014; Yang et al. 2015a, b; Iwegbue et al. 2016a, b, c). The BaA/ (BaA + Chry) and IndP/(IndP + Chry) ratios with values less than 0.2 indicate petroleum and petrogenic sources, BaA/(BaA+Chry) values between 0.2 and 0.35 and IndP/ (IndP + BghiP) between 0.2 and 0.5 suggest contributions from combustion of petroleum (such as liquid fossil fuels, vehicles and crude oil) and BaA/(BaA+Chry) and IndP/ (IndP + Chry) ratios > 0.5 indicate contributions from combustion of coal, grass and wood. The BaA/(BaA+Chry) and IndP/(IndP+Chry) ratios in these dust samples varied from 0.00 to 0.95 with the majority of the homes having values less than 0.2 (Table 6) which suggests that the PAHs in the house dusts arise from sources other than liquid fuel and wood combustion. In the rural and semi-urban areas, the ratio BaA/(BaA + Chry) ranged 0.29 to 0.93 and 0.00 to 0.75, respectively, which suggests that combustion of petroleum, coal and biomass are the sources of PAHs. The ratio of BaP/BghiP can be used to differentiate between coal combustion sources and traffic exhausts (Yang et al. 2015a). For example, BaP/BghiP values between 0.3 and 0.44 indicate that the PAHs come from automobile exhausts, whereas values between 0.9 and 6.6 suggest coal combustion sources (Sawicki 1962; Yang et al. 2015a). The BaP/BghiP values for dust from urban homes ranged from 0.00 to 6.98 with values less than 0.2 for the majority of these samples, while the values for the rural and semi-urban areas ranged from 0.29 to 5.07 and 0.00 to 0.63, respectively. The BaP/ BghiP ratios suggest that the PAHs in dusts from homes in the semi-urban and urban areas arise from traffic emission sources, while those of the rural areas come from biomass/ wood combustion sources. The LMW/HMW values less than 1 for the dust samples from homes in these areas indicate the prevalence of HMW-PAHs in these dusts. The  $\Sigma COMB/$ TPAHs ratio varied from 0.50 to 0.97, which suggests that pyrogenic sources are responsible for the PAH concentrations in the house dusts. The values of LMW/HMW and  $\Sigma$ COMB/TPAHs contrast those obtained for the BaA/

 Table 5
 Non-cancer and cancer risk of PAHs in dust of three residential zones

|            | Non-carcinog          | genic risk            |                       |                       | Carcinogenic risk     |                        |                       |                       |  |
|------------|-----------------------|-----------------------|-----------------------|-----------------------|-----------------------|------------------------|-----------------------|-----------------------|--|
|            | HQ <sub>ing</sub>     | HQ <sub>inh</sub>     | HQ <sub>derm</sub>    | HI                    | RISK <sub>ing</sub>   | RISK <sub>inh</sub>    | RISK <sub>derm</sub>  | Total cancer risk     |  |
| Infant     |                       |                       |                       |                       |                       |                        |                       |                       |  |
| Rural      | $1.17 \times 10^{-1}$ | $9.93 \times 10^{-6}$ | $2.52 \times 10^{-5}$ | $1.17 \times 10^{-1}$ | $5.93 \times 10^{-2}$ | $5.68 \times 10^{-10}$ | $1.56 \times 10^{-3}$ | $6.08 \times 10^{-2}$ |  |
| Semi-urban | $2.95 \times 10^{-1}$ | $6.12 \times 10^{-6}$ | $6.54 \times 10^{-5}$ | $2.96 \times 10^{-1}$ | $2.75 \times 10^{-2}$ | $2.70 \times 10^{-10}$ | $7.24 \times 10^{-4}$ | $2.82 \times 10^{-2}$ |  |
| Urban      | 12.4                  | $1.08 \times 10^{-3}$ | $2.83 \times 10^{-3}$ | 12.4                  | $5.61 \times 10^{-1}$ | $6.21 \times 10^{-9}$  | $1.48 \times 10^{-2}$ | $5.76 \times 10^{-1}$ |  |
| Toddler    |                       |                       |                       |                       |                       |                        |                       |                       |  |
| Rural      | $5.82 \times 10^{-2}$ | $1.23 \times 10^{-5}$ | $2.12 \times 10^{-5}$ | $5.82 \times 10^{-2}$ | $2.95 \times 10^{-2}$ | $1.42 \times 10^{-9}$  | $1.32 \times 10^{-3}$ | $3.08 \times 10^{-2}$ |  |
| Semi-urban | $1.48 \times 10^{-1}$ | $7.60 \times 10^{-6}$ | $5.51 \times 10^{-5}$ | $1.48 \times 10^{-1}$ | $1.36 \times 10^{-2}$ | $6.75 \times 10^{-10}$ | $6.10 \times 10^{-4}$ | $1.43 \times 10^{-2}$ |  |
| Urban      | 6.21                  | $1.35 \times 10^{-3}$ | $2.38 \times 10^{-3}$ | 6.21                  | $2.79 \times 10^{-1}$ | $1.55 \times 10^{-8}$  | $1.25 \times 10^{-2}$ | $2.91 \times 10^{-1}$ |  |
| Child      |                       |                       |                       |                       |                       |                        |                       |                       |  |
| Rural      | $2.91 \times 10^{-2}$ | $1.49 \times 10^{-5}$ | $1.81 \times 10^{-5}$ | $2.91 \times 10^{-2}$ | $1.48 \times 10^{-2}$ | $3.41 \times 10^{-9}$  | $1.13 \times 10^{-3}$ | $1.59 \times 10^{-2}$ |  |
| Semi-urban | $7.36 \times 10^{-2}$ | $9.15 \times 10^{-6}$ | $4.71 \times 10^{-5}$ | $7.37 \times 10^{-2}$ | $6.84 \times 10^{-3}$ | $1.62 \times 10^{-9}$  | $5.21 \times 10^{-4}$ | $7.36 \times 10^{-3}$ |  |
| Urban      | 3.09                  | $1.62 \times 10^{-3}$ | $2.03 \times 10^{-3}$ | 3.10                  | $1.40 \times 10^{-1}$ | $3.73 \times 10^{-8}$  | $1.07 \times 10^{-2}$ | $1.51 \times 10^{-1}$ |  |
| Teen       |                       |                       |                       |                       |                       |                        |                       |                       |  |
| Rural      | $1.60 \times 10^{-2}$ | $1.43 \times 10^{-5}$ | $5.42 \times 10^{-6}$ | $1.60 \times 10^{-2}$ | $8.14 \times 10^{-3}$ | $5.97 \times 10^{-9}$  | $3.36 \times 10^{-4}$ | $8.48 \times 10^{-3}$ |  |
| Semi-urban | $4.06 \times 10^{-2}$ | $8.83 \times 10^{-6}$ | $1.41 \times 10^{-5}$ | $4.06 \times 10^{-2}$ | $3.77 \times 10^{-3}$ | $2.83 \times 10^{-9}$  | $1.56 \times 10^{-4}$ | $3.93 \times 10^{-3}$ |  |
| Urban      | 1.70                  | $1.56 \times 10^{-3}$ | $6.08 \times 10^{-4}$ | 1.71                  | $7.71 \times 10^{-2}$ | $6.52 \times 10^{-8}$  | $3.19 \times 10^{-3}$ | $8.03 \times 10^{-2}$ |  |
| Adult      |                       |                       |                       |                       |                       |                        |                       |                       |  |
| Rural      | $1.47 \times 10^{-2}$ | $3.13 \times 10^{-5}$ | $5.65 \times 10^{-6}$ | $1.47 \times 10^{-2}$ | $4.74 \times 10^{-3}$ | $9.01 \times 10^{-9}$  | $2.22 \times 10^{-4}$ | $4.96 \times 10^{-3}$ |  |
| Semi-urban | $3.73 \times 10^{-2}$ | $1.93 \times 10^{-5}$ | $1.47 \times 10^{-5}$ | $3.73 \times 10^{-2}$ | $2.20 \times 10^{-3}$ | $4.28 \times 10^{-9}$  | $1.03 \times 10^{-4}$ | $2.30 \times 10^{-3}$ |  |
| Urban      | 1.57                  | $3.41 \times 10^{-3}$ | $6.34 \times 10^{-4}$ | 1.57                  | $4.49 \times 10^{-2}$ | $9.85 \times 10^{-8}$  | $2.10 \times 10^{-3}$ | $4.70 \times 10^{-2}$ |  |

(BaA + Chry) and IndP/(IndP + BghiP) ratios which support the fact that the sources of PAHs in a matrix may be different and occasional (Orecchio 2011). The total index is the sum of the single indices normalized with their respective limits between low- and high-temperature processes reported in the literature (Yunker et al. 2002; Barreca et al. 2014). A total index value greater than 4 suggests high-temperature combustion processes, while a total index value less than 4 relates to low-temperature combustion processes. As shown in Table 6, the total index values ranged from 5.74 to 10.7 with a mean of 8.23 for the rural area, and from 2.71 to 8.33 with a mean of 5.65 for the semi-urban area. This suggests that high-temperature combustion processes were responsible for the PAH concentrations in dusts from homes in the rural and semi-urban areas.

## **Principal Component Analysis**

Principal component analysis (PCA) is applied to simplify data that have a large number of correlated variables. This technique has been used for determining the distribution and source apportionment of PAHs in the environment. The principal components consist of the linear combination of the original variables that account for most of the variability in the dataset. The first component explains the greatest variability, while the successive components explain the smaller trends in a decreasing order of importance (Stout et al. 2001; DeMott et al. 2010). PCA can be used to visualize the

Table 6 Diagnostic source ratios for PAHs in indoor dust

relationships between multivariate samples in simple 2- or 3-dimensional PCA score plots (Johnson and Ehrlich 2002). In the PCA plot score, samples with similar characteristic features appear near one another, while those that exhibit different characteristic features are far away from each other. The PCA factor loadings after Varimax with Kaiser Normalization for the PAH compounds in the house dust are presented in Table 7.

In the rural area, the PCA of PAHs in household dust was resolved into four components explaining 85.5% of the total variance. Factor 1 accounts for 33.1% of the total variance and contained PAH compounds such as NaP, Phen, Ant, Pyr, Chry and BbF. NaP is characteristic of incomplete combustion-related sources (Jiang et al. 2009; Dong and Lee 2009; Wang et al. 2011). Factor 2 explained 26.5% of the total variance and was dominated by tricyclic PAHs such as Acy, Ace and Flu and tetracyclic PAH compounds including Flt and BaA. Factor 2 consists of compounds characteristic of lowtemperature combustion processes including wood combustion (Lv et al. 2010; Iwegbue et al. 2016b). Factor 3 accounts for 13.3% with BaP, IndP and BghiP as the dominating compounds. These are characteristic compounds for gasoline and gas engine emissions (Khalili et al. 1995; Larsen and Baker 2003; Jo and Lee 2009; Kwon and Choi 2014) and combustion of heavy oil (Harrison et al. 1996; Lee et al. 2004; Kwon and Choi 2014). BaP and BghiP are tracers for automobile emissions because these compounds have been found in high concentrations in traffic tunnels (Harrison et al. 1996;

|                     | Rural | Rural |        |      |        |      | Semi-urban |        |      |      |  |
|---------------------|-------|-------|--------|------|--------|------|------------|--------|------|------|--|
|                     | Mean  | SD    | Median | Min  | Max    | Mean | SD         | Median | Min  | Max  |  |
| BaA/(BaA+Chry)      | 0.65  | 0.24  | 0.61   | 0.29 | 0.93   | 0.44 | 0.27       | 0.54   | 0.00 | 0.75 |  |
| IndP/(IndP + BghiP) | 0.55  | 0.18  | 0.50   | 0.29 | 0.85   | 0.23 | 0.20       | 0.23   | 0.00 | 0.47 |  |
| Ant/(Ant+Phen)      | 0.27  | 0.09  | 0.27   | 0.11 | 0.45   | 0.09 | 0.21       | 0.01   | 0.00 | 0.67 |  |
| Flt/(Flt + Pyr)     | 0.46  | 0.26  | 0.50   | 0.00 | 0.82   | 0.86 | 0.11       | 0.89   | 0.67 | 0.97 |  |
| LMW/HMW             | 0.19  | 0.18  | 0.15   | 0.00 | 0.54   | 0.54 | 0.21       | 0.51   | 0.22 | 0.89 |  |
| COMB PAHs/TPAHs     | 0.72  | 0.11  | 0.68   | 0.59 | 0.88   | 0.62 | 0.10       | 0.62   | 0.50 | 0.82 |  |
| BaP/BghiP           | 2.10  | 1.82  | 1.79   | 0.29 | 5.07   | 0.26 | 0.19       | 0.29   | 0.00 | 0.63 |  |
| Total index         | 8.23  | 1.81  | 8.70   | 5.74 | 10.7   | 5.65 | 1.63       | 6.00   | 2.71 | 8.33 |  |
|                     |       | U     | rban   |      |        |      |            |        |      |      |  |
|                     |       | S     | D      |      | Median |      |            | Min    |      | Max  |  |
| BaA/(BaA+Chry)      |       | 0.    | 12     |      | 0.03   |      |            | 0.00   |      | 0.55 |  |
| IndP/(IndP+BghiP)   |       | 0.    | 21     |      | 0.00   |      |            | 0.00   |      | 0.95 |  |
| Ant/(Ant + Phen)    |       | 0.    | 00     |      | 0.00   |      |            | 0.00   |      | 0.00 |  |
| Flt/(Flt + Pyr)     |       | 0.    | 00     |      | 0.00   |      |            | 0.00   |      | 0.00 |  |
| LMW/HMW             |       | 0.    | 21     |      | 0.13   |      |            | 0.00   |      | 0.81 |  |
| COMB PAHs/TPAHs     |       | 0.    | 15     |      | 0.77   |      |            | 0.52   |      | 0.97 |  |
| BaP/BghiP           |       | 1.    | 52     |      | 0.00   |      |            | 0.00   |      | 6.98 |  |
| Total index         |       |       |        |      |        |      |            |        |      |      |  |

#### Table 7 PCA for PAHs in indoor dust after Varimax rotation

|                | Rural area |            |         |         | Semi-urban area |         |         |         |  |
|----------------|------------|------------|---------|---------|-----------------|---------|---------|---------|--|
|                | PCA1       | PCA2       | PCA3    | PCA4    | PCA1            | PCA2    | PCA3    | PCA4    |  |
| Nap            | 0.712      | 0.308      | - 0.237 | - 0.403 | - 0.147         | 0.882   | - 0.041 | 0.421   |  |
| Acy            | 0.223      | 0.922      | - 0.230 | - 0.119 | 0.298           | 0.397   | 0.264   | 0.802   |  |
| Ace            | - 0.040    | 0.862      | - 0.226 | - 0.156 | 0.233           | 0.122   | 0.911   | 0.260   |  |
| Flu            | - 0.157    | 0.871      | - 0.042 | - 0.065 | 0.658           | 0.043   | 0.645   | 0.327   |  |
| Phen           | 0.848      | 0.474      | - 0.101 | - 0.049 | 0.525           | 0.124   | 0.756   | 0.277   |  |
| Ant            | 0.882      | - 0.073    | - 0.193 | 0.227   | 0.169           | 0.916   | - 0.234 | - 0.049 |  |
| Flt            | 0.352      | 0.852      | -0.072  | - 0.129 | 0.636           | 0.122   | 0.690   | 0.251   |  |
| Pyr            | 0.945      | 0.188      | 0.133   | - 0.101 | 0.170           | 0.932   | 0.177   | 0.074   |  |
| Chry           | 0.927      | - 0.176    | 0.200   | - 0.016 | 0.402           | 0.748   | 0.324   | 0.102   |  |
| BaA            | 0.490      | 0.769      | 0.053   | - 0.068 | 0.688           | 0.043   | 0.667   | 0.148   |  |
| BbF            | 0.923      | 0.257      | - 0.102 | - 0.118 | 0.865           | 0.283   | 0.294   | 0.119   |  |
| BkF            | 0.006      | - 0.150    | 0.091   | 0.887   | 0.953           | 0.016   | 0.201   | 0.162   |  |
| BaP            | 0.430      | 0.024      | 0.743   | 0.057   | 0.729           | 0.258   | 0.276   | 0.525   |  |
| IndP           | - 0.193    | - 0.165    | 0.784   | 0.283   | 0.191           | - 0.454 | 0.402   | 0.702   |  |
| DahA           | - 0.079    | - 0.119    | 0.164   | 0.914   | 0.065           | 0.750   | 0.466   | - 0.115 |  |
| BhiP           | - 0.154    | - 0.220    | 0.802   | 0.035   | 0.592           | 0.143   | 0.212   | 0.668   |  |
| Eigen values   | 5.302      | 4.238      | 2.130   | 2.010   | 4.550           | 4.188   | 3.628   | 2.443   |  |
| % of variance  | 33.136     | 26.488     | 13.314  | 12.565  | 28.437          | 26.173  | 22.673  | 15.271  |  |
| Cumulative (%) | 33.136     | 59.624     | 72.938  | 85.504  | 28.437          | 54.611  | 77.284  | 92.555  |  |
|                |            | Urban area |         |         |                 |         |         |         |  |
|                |            | PCA1       |         | PCA2    |                 | PCA3    |         | PCA4    |  |
| Nap            |            | 0.787      |         | - 0.170 |                 | 0.159   |         | 0.233   |  |
| Acy            |            | 0.113      |         | 0.729   |                 | 0.191   |         | - 0.073 |  |
| Ace            |            |            |         |         |                 |         |         |         |  |
| Flu            |            | 0.323      |         | 0.577   |                 | 0.327   |         | 0.633   |  |
| Phen           |            | 0.504      |         | 0.025   |                 | 0.719   |         | 0.024   |  |
| Ant            |            |            |         |         |                 |         |         |         |  |
| Flt            |            | 0.138      |         | - 0.190 |                 | 0.078   |         | 0.863   |  |
| Pyr            |            |            |         |         |                 |         |         |         |  |
| Chry           |            | 0.348      |         | 0.691   |                 | 0.452   |         | 0.033   |  |
| BaA            |            | - 0.149    |         | 0.023   |                 | 0.862   |         | - 0.101 |  |
| BbF            |            | 0.813      |         | 0.409   |                 | - 0.023 |         | 0.294   |  |
| BkF            |            | - 0.226    |         | 0.834   |                 | - 0.132 |         | - 0.044 |  |
| BaP            |            | 0.179      |         | 0.055   |                 | - 0.139 |         | 0.844   |  |
| IndP           |            | 0.113      |         | 0.432   |                 | 0.763   |         | 0.142   |  |
| DahA           |            | 0.642      |         | - 0.171 |                 | 0.338   |         | 0.158   |  |
| BhiP           |            | 0.902      |         | 0.263   |                 | - 0.162 |         | 0.047   |  |
| Eigenvalues    |            | 3.135      |         | 2.560   |                 | 2.400   |         | 2.064   |  |
| % of variance  |            | 24.117     |         | 19.695  |                 | 18.463  |         | 15.878  |  |
| Cumulative (%) |            | 24.117     |         | 43.811  |                 | 62.274  |         | 78.153  |  |

Boldface numbers indicate variables with large positive correlations

Larsen and Baker 2003; Boonyatumanond et al. 2007; Iwegbue et al. 2016c). Factor 4 has BkF and DahA and explained 12.6% of the total variance. BkF and DahA are indicator compounds for diesel emissions (Lee and Dong 2011; Wang et al. 2013c; Iwegbue et al. 2016c). The PCA for PAHs in dusts from homes in the semi-urban area was resolved into four components, which accounted for 92.6% of the total variance. Factor 1 accounted for 28.4% of the total variance and was heavily weighted by Flu, Flt, BaA, BbF, BkF and BaP. These compounds are characteristic of

coal combustion and vehicular emissions (Hu et al. 2017). Factor 2 explained 26.2% with NaP, Pyr, Chry, DahA and NaP present and is associated with unburnt fossil fuel. Chry and DahA have been reported as characteristic compounds of diesel emissions (Marr et al. 1999; Simcik et al. 1999). Factor 3 explained 22.7% of the total variance and was heavily weighted by Ace, Flu, Phen, Ant, Flt and Chry. Factor 3 contains 3- and 4-ring PAH compounds that are characteristic of wood/biomass and fossil fuel combustion sources (Khalili et al. 1995; Simcik et al. 1999; Ravindra et al. 2008; Wang et al. 2013c; Kwon and Choi 2014). Factor 4 accounted for 15.3% of the total variance and has positive loadings in Acy, IndP and BghiP. IndP and BghiP are tracers for gasoline and gas engine emissions and combustion of heavy oil as mentioned earlier.

The PCA for PAHs in household dust from the urban area was resolved into four components accounting for 78.2% of the total variance. Factor 1 accounted for 24.1% of the total variance and was dominated by PAH compounds such as NaP, Phen, BbF, IndP and BghiP. NaP is associated with incomplete combustion-related processes (Simsick et al. 1999), while IndP and BghiP are tracers for gasoline and gas engine emissions (Khalili et al. 1995; Larsen and Baker 2003; Jo and Lee 2009; Kwon and Choi 2014) and combustion of heavy oil (Harrison et al. 1996; Lee et al. 2004; Kwon and Choi 2014). BbF and IndP were found as tracers of emissions from vehicular transportation (Esen et al. 2008; Kuang et al. 2011). Phen is related to emissions from fossil fuel combustion sources (Khalili et al. 1995; Kwon and Choi 2014). Factor 2 accounted for 19.7% of the total variance and was weighted by PAH compounds such as Acy, Flu, BaA and BkF. Factor 2 has a high weight on BkF and Acy. This could be related to diesel combustion because high levels of BkF and Acy are tracers of diesel vehicles (Randolph and Joel 2003; Kuang et al. 2011). Factor 3 has Phen, Chry and DahA, and was responsible for 18.5% of the variability in the data. The prevalence of Chry in the house dust is related to combustion of kerosene and cooking gas (Khalili et al. 1995). Factor 4 accounts for 15.9% of the dataset and was heavily weighted by Flu, Flt and BaP. Flu, Flt and Pyr are typical markers for wood and biomass combustion-related sources (Larsen and Baker 2003; Duval and Friedlander 2004). The PCA results demonstrated that the PAH compositional patterns in the house dust from the urban areas are influenced by outdoor dust infiltration, tracked-in dust and cooking fuel types.

# Conclusions

The 4-ring PAHs were the dominant PAH homologues in the household dust from the urban area with chrysene having the highest concentration and occurrence frequency. In the case of home dust from the semi-urban and rural areas, 3- and 5-ring PAHs are the respective dominant homologues. This points to the fact that the fate and sources of PAHs in dusts from rural homes are entirely different from those of semiurban and urban areas. The total cancer risk values obtained suggest that there is a potential carcinogenic risk for children and adult residents arising from exposure to PAHs in dusts from homes in the rural, semi-urban and urban areas. The PCA results demonstrated the influence of traffic emissions, biomass combustion and cooking fuel types on the sources and compositional patterns of PAHs in the house dusts. The results of this study affirm the need for remedial actions in the study area in order to minimize the risk of exposure to these contaminants. Therefore, further studies are warranted to determine the concentrations of halogenated hydrocarbons (such as PCBs, PBDEs, dioxins and organochlorine pesticides), bisphenols and phthalates in order to provide a detailed understanding of the pollution status of these indoor environments.

## **Compliance with Ethical Standards**

Conflict of interest The authors have no conflict of interest to declare.

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