POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) CONCENTRATION IN SOME POPULAR COMMERCIAL BRANDS OF CIGARETTE IN NIGERIA

Supported by tetfund TERTIARY EDUCATION TRUST FUND

Ufuoma A. Igbuku[1,3], Bulouebibo Lari[2,3], Loretta C. Overah[3], Chukwujindu M. A. Iwegbue[3]*

[1]Department of Science Laboratory Technology, Delta State Polytechnic, Ozoro, Delta State, Nigeria
[2]Department of Science Laboratory Technology, Delta State University, Abraka, Nigeria
[3]Department of Chemistry, Delta State University, PMB 1 Abraka, Nigeria
*Correspondence author: maxipriestley@yahoo.com

Received: August 21, 2018 **Accepted:** September 30, 2018

Abstract: The concentrations of the USEPA 16 priority PAHs were investigated in 27 popular commercial brands of cigarette in the Nigerian market by using gas chromatography-mass spectrometry after extraction by ultra-sonication, using n-hexane/dichloromethane as solvent and for clean up. The risk of PAHs in these commercial brands of cigarette was assessed using the benzo[a]pyrene carcinogenic and mutagenic equivalency factors, and the sources of PAHs in these products were determined using the PAH isomeric ratios and principal component analysis. The concentrations of Σ16 PAHs in these commercial brands of cigarette ranged 425 to 10,300 µg kg^{-1} with an average of 2090 µg kg^{-1}. The compositional patterns of PAHs in these brands of cigarette follow the order 2-3 rings>4-rings>5-6 rings. The benzo[a]pyrene carcinogenic (BaPTEQ) and mutagenic (BaPMEQ) equivalency factor ranged from 0.1 to 731 with an average of 128 µg kg^{-1} and from 0.9 to 1090 µg kg^{-1} with an average of 150 µg kg^{-1}. The concentrations of the ΣPAHs in these samples are comparable to concentrations found in cigarette smoke and smokeless tobacco products in the literature. The PAHs isomer ratios indicate that the source of PAHs were mainly from petroleum, vehicular emissions, combustion of coal, wood and biomass.

Keywords: Cigarette, tobacco, PAHs, sources, toxic equivalency factor, Nigeria

Introduction

Cigarettes constitute the largest share of manufactured tobacco products in the world and about 96% of total sales. About 5.6 trillion cigarettes are produced annually which implies nearly 900 cigarettes per year for every person on the earth surface. Cigarettes are carefully engineered and designed products with different chemical additives added to the tobacco content, paper, and filter during the production process. Most cigarette manufacturing companies are increasingly using chemically modified, reconstituted tobacco made from discarded plant parts, stems and leaf ribs, tobacco dust, and reclaimed tobacco content to reduce the amount of tobacco in each cigarette and increase their profit margins (Ding *et al.*, 2008). Tobacco products are known to contain different carcinogenic compounds, or generate different carcinogenic compounds during its use which can cause cancer for the users. There are more than 60 known carcinogens in cigarette smoke and at least 16 in unburned tobacco. Among these, tobacco-specific nitrosamines (TSNA), polycyclic aromatic hydrocarbons (PAHs), and aromatic amines are well known to cause cancer. The concentrations of TSNAs and PAHs depend on the cigarette design and other chemical constituents. Tobacco usage is the main cause of cancers of lung, larynx, oral cavity, oesophagus, bladder and pancreas; and it is responsible for approximately 20% of all cancer deaths (Ding *et al.*, 2008).

PAHs are a group of over a hundred (100) pervasive organic compounds consisting of two or more fused aromatic rings/and pentacyclic rings in linear, angular or cluster formations. PAHs are primarily derived from the incomplete combustion or pyrolysis of organic matters including cigarette and natural combustion process such as forest fires and volcanic eruptions (Naccari *et al.*, 2011). They have been classified as hazardous compounds of environmental and health concerns because they are persistent and can undergo bioaccumulation and long-range transportation and deposition. Moreover, a number of them have well known carcinogenic, genotoxic and mutagenic properties. For these reasons, the United State Environmental Protection Agency (USEPA) has listed 16 PAHs as priority environmental pollutants based on their toxicity and occurrence frequencies that were further classified as follows; Benzo[a]pyrene (BaP) is carcinogenic (group 1), dibenz[a,h]anthracene is probably carcinogenic (group 2A) whereas naphthalene (Nap), benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF) and indeno [1,2,3-c,d] pyrene are classified as possible human carcinogen (group 2B) while acenaphthylene (Acy), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr) and benzo[g,h,i]perylene (BghiP) are not classified as their carcinogenicity to humans (International Agency for Research on Cancer, IARC, 2012).

A number of studies in the literature have documented the concentrations of PAHs in cigarettes, cigarette smoke and smokeless tobacco products [1, 4, 5, 6]. However, there are little or no published data on the concentrations of polycyclic aromatic hydrocarbons in popular commercial brands of cigarette in Nigeria. The objective of the present study was to determine the concentrations of the US EPA 16 priority PAHs in some popular commercial brands of cigarette in Nigeria with a view to providing information on the sources and risk of PAHs in these products.

Materials and Methods

Samples and sample collection

A total of 27 popular commercial brands of cigarettes were collected from tobacco shops in major towns in the Delta State of Nigeria. Information on the brand name and the origin of the cigarette are presented in Table 1.

Reagents

All chemicals and reagents used were of analytical grade. Acetone was obtained from Rieldel-de Haën (Seelze, Germany, and purity 99.8%) while dichloromethane (LC grade), anhydrous sodium sulfate (purity 99%), alumina was obtained from BDH (Poole, UK) and n-Hexane was obtained from Sigma-Aldrich (Steinheim, Germany). A PAH standard mixtures containing the US EPA 16 priority PAHs was obtained from Supelco (Bellefonte, PA, USA). Working mixed standard solutions containing all the PAHs were prepared by dilution of the stock solution with acetone and

stored at -10°C in darkness to avoid volatilization and photodegradation.

Sample preparation, extraction and clean up

The cigarette sticks were removed from its packs and the sticks in the each pack were crushed together, from which a subsample was obtained for PAHs analysis. A mass of 5.0 g of the cigarette sample was mixed with same amount of a drying agent, Na_2SO_4, until the mixture becomes free flowing. The resulting material was extracted by ultra-sonication with 30 mL of hexane/dichloromethane (DCM) (1:1 v/v) using ultra-sonic bath at 30°C for 30 min. The contents were filtered and the extraction process was repeated three times by sonication of the residue with a fresh mixture of hexane/dichloromethane each time as described above. The extracts were combined and rotary evaporated to 1 mL. The extract was cleaned up by solid phase extractions with 2 g of aluminium oxide (5% deactivated lower part). The PAHs were subsequently eluted with 15 mL of hexane, 15 mL hexane and dichloromethane (9:1) and 20 mL of hexane and dichloromethane (4:1). The eluted fractions were combined and evaporated to approximately 1.0 mL with a gentle stream of nitrogen.

Chromatographic analysis

The PAHs in the eluted fraction were measured with a gas chromatograph (HP 6890 Palo Alto, CA) equipped with a J&W Durabond 5 (cross-linked phenyl methyl siloxane) column (0.25 µm film thickness, 0.25 µm × 30 m) and a HP 5973 series mass-selective detector. The mass spectrometer was operated in the electron impact ionization mode (ionizing energy of 70 eV) and mass spectra were acquired by scanning from m/z 50 to 450 at 3.6 scans/s. The ion source and quadrupole temperature were 230 and 150°C, respectively. The operating conditions were as follows; the injection port and the GC/MS interface temperatures were 290 and 250°C, respectively. The column temperature was initially maintained at 80°C for 0.5 min and then ramped to 230 at 80°C/min and from 230 to 280°C at 5°C/min, and maintained at 280°C for 18 min; the solvent delay was 6 min. The injection volume was 2 µL in pulsed splitless mode and the carrier gas was helium with a linear velocity of 1 mL/min.

Quality control/quality assurance and statistical analysis

The quantification is by the use of external calibrations which were obtained with PAH solutions at five concentration levels. To evaluate the extraction efficiency for the targeted PAH compounds, known concentrations of standard PAH mixture were added to fresh portions of already analysed samples at three concentration levels and repeating all analytical steps from extraction to chromatographic analysis. The recoveries for the PAH compounds were in the range 79.6 to 108%. The relative standard deviations for replicate analyses (n = 3) were less than 6%. The r^2 values for the calibration lines were in the range of 0.9991 to 0.999 while limits of detection and quantification of the PAH compounds ranged from 0.3 to 2.0 µg kg^{-1} and from 1.0 to 6.0 µg kg^{-1}, respectively. The average inter-day and intra-day precisions of the method were 1.5 to 12.2% and 1.3 to 13.5%, respectively. Analysis of variance (ANOVA) was used to determine whether the differences observed in the concentrations of the PAHs among the brands are significant. Principal component analysis and PAH isomeric rations were used for source analysis of PAHs in these brands of cigarettes. The statistical calculations were performed with SPSS version 11.5.

Risk assessment

Toxic equivalency factor

The risk to human health from various PAH exposures can be established by evaluating the carcinogenic and mutagenic potency of the individual PAH compounds relative to that of BaP. The risks of PAHs in dust, foods, sediment and soils have been assessed using the BaP toxic carcinogenic (BaP_{TEQ}) and BaP mutagenic equivalency factors (BaP_{MEQ}) in the literature (Nisbet and LaGoy, 1992; USEPA, 1993; Durant *et al*., 1996; Larsen and Larsen, 1998).

The BaP carcinogenic equivalent (BaP_{TEQ}) for the individual PAHs can be estimated by using the equation:

$$BaP_{TEQ} = \Sigma\, Ci \times BaP_{TEF} \quad (1)$$

Where BaP_{TEF} is the cancer potency relative to BaP and Ci is the individual PAH concentration.

The BaP mutagenic equivalent (BaP_{MEQ}) for the individual PAHs can be estimated by the equation:

$$BaPMEQ = \Sigma\, Ci \times BaP_{MEF} \quad (2)$$

Where BaP_{MEQ} = the mutagenic potency relative to BaP and Ci = the individual PAH concentration. The BaP carcinogenic equivalency factors (BaP_{TEFs}) of the seven carcinogenic PAHs used were BaP (1), BaA (0.1), BbF (0.1), BkF (0.01), Chry (0.001), DahA (1) and IndP (0.1) US EPA, (1993). The BaP mutagenic potency factors (Ba_{PMEFs}) were BaP (1), BaA (0.082), BbF (0.25), BkF (0.11), Chry (0.017), DahA (0.29) and IndP (0.31) (Durant *et al*., 1996).

Results and Discussion

The concentrations of Σ16 PAHs in the 27 brands of cigarette investigated ranged between 425 and 10,300 µg kg^{-1} with a mean concentration of 2090 µg kg^{-1} (Table 1). Analysis of variance (ANOVA) ($p<0.05$) indicated the differences in the concentrations of Σ16 PAHs among the various brands are significant. The compositional patterns of PAHs varied from one to another. The difference in the concentration and composition pattern of PAHs in these brands of cigarette may be due to the origin and endogenous concentrations of PAHs in the tobacco, the numerous processes to which the tobacco is subjected to, and the additives used during the manufacturing of cigarette. The mechanisms for the formation of PAHs during smoking involve the degradation of complex molecules and formation of simpler molecules and free radical which recombines to form PAHs (Badger *et al*., 1965). Alternatively, PAHs can be form by the aromatisation of complex molecules e.g. phytosterols, to give different structures of PAHs. The main precursors include solanesol, phytosterols, terpenes, amino acids, nicotine, lipids, cellulose and several other tobacco components (Lam *et al*., 1985; Rodgman, 2001). In this cigarette samples, 3- and 4-ringed PAHs constituted significant proportion of the Σ16 PAHs (7.1 to 73.8 %), while 5 to 6-ringed constituted (7.2 to 86.2%) of the Σ16 PAHs. The dominance of 3- and 4-ringed PAHs in these cigarette samples may be due to the fact that the manufacturing processes involve low temperature operations not exceeding 400°C. In addition, high concentrations of 2-4-ringed PAHs were due to their high aqueous solubility. More than 4-ringed PAH compounds (BbF, BkF, BaP, DahA, IndP and BghiP) were detected in low concentrations due to their high water octanol coefficient and low aqueous solubility (Orecchio *et al.*, 2009). The concentrations of the ΣPAHs in these samples are comparable to concentrations found in cigarette smoke and smokeless tobacco products in the literature (Ding *et al*., 2008; Kalaitzoglou and Samara, 2005; McAdam *et al*., 2013). The concentrations of the 2-ringed PAHs ranged from 6.0 to 2416 µg kg^{-1} which constituted up to 71.3% of Σ16 PAHs in brand 6. The three ringed PAHs constituted 0.3 to 73.8% of the Σ16 PAHs in the brands of cigarettes. Phenanthrene was the dominant 3-ringed PAH compound in these brands of cigarette in terms of concentration and occurrence frequency. The dominance of Phenanthrene in tobacco products have been reported in the literature (Akpan *et al*., 2006). Phenanthrene constituted up to 34.4% of the Σ 16 PAHs. Anthracene was found in 17 out of the 27 samples investigated, with maximum concentration in brand 26 (1140 µg kg^{-1}). The contributions of anthracene to the Σ16 PAHs were less than 9% in all samples except for brand 26 (35.8%).

Acy, Ace and Flu were detected in not less than 20 out of the 27 samples examined, which constituted 0.1 to 9.6%, 0.1 to 70%, 0.1 to 18.6%, respectively of the Σ16 PAHs. The concentrations of Σ 4 ringed PAHs ranged from 16.0 to 1040 µg kg^{-1} with the highest concentration in brand 25. The 4-ringed PAH compounds constituted approximately 3.0 to 59.3% of the Σ16 PAHs in these samples. In this study, pyrene and chrysene were the dominant 4 ringed PAH compounds in these samples of cigarette. BaA was detected in 13 of the 27 brands analysed at concentrations in the range of 3.0 to 127 µg kg^{-1} which constituted less than 6% the Σ16 PAHs in these samples. Fluoranthene was detected in low concentrations relative to the other 4-ringed PAH compounds in these samples. Fluoranthene constituted less than 22% of the Σ16 PAHs in these samples.

The 5-ringed PAHs constituted 0.1 to 73.8% of the Σ16 PAHs in these brands of cigarette. The highest concentration of 5-ringed PAHs was observed in brand 18 (1600 µg kg^{-1}). BkF was the dominant 5-ringed PAH compound in these brands of cigarette. In these samples, brands 18 and 19 had higher concentrations of BkF relative to other samples. BkF constituted up to 59.6% of the Σ 16 PAHs in these brands of cigarette. BaP was found in 15 of the 27 brands investigated at concentrations between 1.0 and 251 µg kg^{-1} with maximum concentration in brand 19. The source of BaP in tobacco products may arise from environmental contamination of the leaf surface or inadvertent exposure to combustion fumes during processing (Akpan *et al.*, 2006). A wide concentration range of BaP has been documented for tobacco products in the literature. For instance, Akpan *et al.* (2006) found BaP concentrations of 0.971 to 117 µg kg^{-1} in smokeless tobacco products. Kalaitzoglou and Samara (2005) observed BaP concentrations of 0.8 to 10 ng/cigarette in particulate phase of mainstream cigarette smoke. Nair *et al.* (1987) reported BaP concentrations ranging from 27.0 to 119 µg kg^{-1} in masheri (pyrolysed tobacco products) in the India. BaP concentrations of 0.7 to 118 µg kg^{-1} have been reported in different kinds of smokeless tobacco products (Stepanov *et al*., 2008; 2010).

The 6-ringed PAHs constituted 0.1 to 39.3% of the Σ 16PAHs in these brands of cigarette. BghiP was the dominant 6-ringed PAH compound in these cigarette brands. IndP was detected in all the brands investigated except in brand 15. The highest concentration of IndP was observed in brand 19. IndP and BghiP make up 0.1 to 21.7% and 0.2 to 42.1% of the Σ16 PAHs in some of these samples. The concentrations of the seven carcinogenic PAHs (PAH_{7C}) in the cigarette samples ranged from 22 to 6828 µg kg^{-1} (see Table 3). The highest level of PAH_{7C} was observed in C19. Nevertheless, C8, C12, C13, C18, C19 and C24 contained PAH_{7C}. The PAH_{7C} concentrations in these samples accounted for 7.9 to 88.8% of the total PAHs. The CONTAM Panel of the EFSA in 2008 recommended that the concentration of BaP is not better indicator for the occurrence and toxicity effects of whole class of carcinogenic and genetoxic PAHs in foods. The EFSA recommended the use of PAH2 (BaP + Chry), PAH 4 (PAH2+BbF+BaA) or PAH8 (PAH4 + BkF + IndP+ DahA + BghiP) be as indicators of PAHs (EFSA, 2008), and concluded that a system of eight substances (PAH8) would not provide much added value as compared with that of the system of four substances (PAH4). The PAH2, PAH4 and PAH8 concentrations in these brands of cigarette ranged from nd to 831, nd to 1490 and 22 to 9448 µg kg^{-1} (Table 3). The sample, C19, contained the highest concentrations of PAH2, PAH4 and PAH8.

Table 1: Information on different brands of cigarette

Sample code	Brand	Country	Composition
C-1	St. Moritz	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-2	Rothmans	London/Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-3	London (menthol)	Nigeria with British Tobacco Company	Tar: 10 mg, Nicotine: 1 mg, carbon monoxide: 10 mg
C-4	Forum	Nigeria/USA	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-5	Green Spot	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-6	Pallmall	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-7	Oris	Germany	Tar: 7 mg, Nicotine: 0.6 mg, carbon monoxide: 7 mg
C-8	ACE	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-9	Aspen	Nigeria/UK	Tar: 10 mg, Nicotine: 0.8 mg, carbon monoxide: 10 mg
C-10	Consulate	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-11	YES	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-12	Benson & Hedges	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-13	Benson & Hedges (Switch)	London/Nigeria	Tar: 10 mg, Nicotine: 0.8 mg, carbon monoxide: 8 mg
C-14	Dunhill (master Blend	Nigeria with British Tobacco Company	Tar: 7 mg, Nicotine: 0.8 mg, carbon monoxide: 7 mg
C-15	Rocco	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-16	Titan (Philles)	USA	Pure Tobacco
C-17	Dunhill (Switch)	Nigeria with British Tobacco Company	Tar: 8 mg, Nicotine: 0.6 mg, carbon monoxide: 7 mg
C-18	Gold Bond (Filter)	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-19	Gold Bond (Menthol)	Nigeria	Tar: 10 mg, Nicotine: 0.8 mg, carbon monoxide: 10 mg
C-20	Blackstone	USA	Pure Tobacco
C-21	Peterfield Special	Nigeria	Tar: 10 mg, Nicotine: 1.0 mg, carbon monoxide: 10 mg
C-22	King Edward (The Seventh)	USA	Pure Tobacco
C-23	Dorchester	Nigeria with /UK	Tar: 10 mg, Nicotine: 0.8 mg, carbon monoxide: 10 mg
C-24	Captain Black	Nigeria	Nicotine (1%)
C-25	HAV-A-TAMPA (Jewels Vanilla)	USA	Pure Tobacco
C-26	Viceroy	USA	Tar: 10 mg, Nicotine: 0.7 mg, carbon monoxide: 10 mg
C-27	Sterling Blue	Nigeria/UK	Tar: 10 mg, Nicotine: 0.8 mg, carbon monoxide: 10 mg

Table 2: Concentrations of PAHs (µg kg^{-1}) in commercial brands of cigarette in Nigeria

	NaP	Acy	Ace	Flu	Phen	Ant	Flt	Pyr	BaA	Chry	BbF	BkF	BaP	IndP	DahA	BghiP	Σ16 PAI
C-1	717	2.0	3.0	19.0	283	2.0	79.0	704	127	185	71.0	101	11.0	74.0	52.0	46.0	2480
C-2	409	22.0	ND	18.0	196	30.0	80.0	46.0	ND	25.0	7.0	23.0	ND	38.0	8.0	175	1080
C-3	28.0	1.0	25	5.0	146	24.0	9.0	30.0	15.0	15.0	13.0	38.0	ND	15.0	52.0	11.0	425
C-4	418	3.0	254	10.0	188	9.0	17.0	514	24.0	165	ND	491	ND	23.0	6.0	45.0	2170
C-5	360	6.0	4.0	77.0	144	7.0	2.0	676	0.0	521	2.0	7.0	ND	346	16.0	5.0	2170
C-6	2416	ND	3.0	ND	109	7.0	ND	5.0	29.0	89.0	9.0	442	2.0	217	28.0	32.0	3390
C-7	300	1.0	7.0	6.0	139	67.0	9.0	404	18.0	85.0	22.0	1.0	6.0	4.0	6.0	42.0	1120
C-8	552	6.0	5.0	6.0	239	82.0	9.0	745	3.0	323	19.0	52.0	13.0	9.0	715	87.0	2870
C-9	286	ND	3475	8.0	105	43.0	5.0	38.0	5.0	100	16.0	735	6.0	7.0	88.0	ND	4920
C-10	175	27.0	376	3.0	75.0	38.0	2.0	273	9.0	19.0	12.0	41.0	9.0	18.0	31.0	ND	1110
C-11	446	16.0	2.0	3.0	158	250	4.0	666	ND	193	ND	ND	ND	7.0	45.0	1300	3090
C-12	79.0	91.0	10.0	18.0	143	0.0	6.0	205	ND	536	ND	276	ND	298	136	3.0	1800
C-13	86.0	14.0	2.0	6.0	11.0	51.0	10.0	63.0	ND	187	273	238	80.0	236	15.0	78.0	1350
C-14	121	ND	ND	ND	229	ND	ND	196	ND	113	57.0	213	ND	302	98.0	60.0	1390
C-15	ND	4.0	ND	ND	ND	ND	ND	ND	ND	16.0	ND	6.0	ND	ND	ND	ND	27.0
C-16	252	12.0	ND	66.0	28.0	ND	25.0	124	ND	248	49.0	517	2.0	262	100	22.0	1710
C-17	407	44.0	ND	113	18.0	ND	328	138	ND	173	ND	79.0	ND	19.0	123	174	1620
C-18	116	ND	33.0	ND	9.0	ND	ND	42.0	ND	291	181	1340	77.0	46.0	62.0	53.0	2250
C-19	317	98.0	66.0	ND	4.0	292	ND	57.0	ND	580	659	3680	251	620	235	3420	10300
C-20	1.0	1.0	39.0	2.0	ND	38.0	9.0	66.0	34.0	390	ND	67.0	2.0	63.0	40.0	37.0	788
C-21	4.0	1.0	5.0	ND	38.0	ND	88.0	82.0	ND	ND	ND	138	ND	18.0	29.0	1.0	403
C-22	2.0	ND	20.0	3.0	39.0	ND	1.0	148	13.0	49.0	ND	89.0	ND	43.0	47.0	33.0	487
C-23	6.0	ND	ND	17.0	11.0	ND	4.0	47.0	2.0	29.0	57.0	89.0	125	20.0	7.0	125	540
C-24	19.0	ND	2.0	ND	3.0	1.0	ND	2.0	ND	352	1050	49.0	42.0	2.0	68.0	228	1820
C-25	446	18.0	279	540	158	25.0	13.0	750	ND	277	ND	ND	ND	133	45.0	213	2900
C-26	245	16.0	35.0	83.0	294	1140	29.0	334	13.0	158	ND	318	ND	336	24.0	150	3170
C-27	101.0	4.0	ND	43.0	35.0	ND	25.0	215	ND	248	41.0	64.0	1.0	262	24.0	22.0	1090

Napthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phen), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Benzo[a]anthracene (BaA), Chrysene (Chry), Benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), Benzo[a]pyrene (BaP), Indeno[1,2,3-cd}perylene (IndP), Dibenz[a,h]anthracene (DahA) and Benzo[ghi]perylene (BghiP)

Table 3: The concentrations (µg kg^{-1}) of PAH homologues, seven carcinogenic PAHs and EFSA indicators in cigarette samples

	2 Rings	3 Rings	4 Rings	5 Rings	6 Rings	PAH_{7C}	PAH2	PAH4	PAH8
C-1	717	309	1100	235	120	621	196	394	667
C-2	409	266	151	38	213	101	25	32	276
C-3	28	201	69	103	26	148	15	43	159
C-4	418	464	720	497	68	709	165	189	754
C-5	360	238	1200	25	351	892	521	523	897
C-6	2420	119	123	481	249	816	91	129	848
C-7	300	220	516	35	46	142	91	131	184
C-8	552	338	1080	799	96	1134	336	358	1221
C-9	286	3630	148	845	7.0	957	106	127	957
C-10	175	519	303	93	18	139	28	49	139
C-11	446	429	863	45	1307	245	193	193	1545
C-12	79	262	747	412	301	1246	536	536	1249
C-13	86	84	260	606	314	1029	267	540	1107
C-14	121	229	309	368	362	783	113	170	843
C-15	ND	4.0	16	6.0	ND	22	16	16	22
C-16	252	106	397	668	284	1178	250	299	1200
C-17	407	175	639	202	193	394	173	173	568
C-18	116	42	333	1664	99	2001	368	549	2054
C-19	317	460	637	4828	4040	6028	831	1490	9448
C-20	1.0	80	499	109	100	596	392	426	633
C-21	4.0	44	170	167	19	185	ND	ND	186
C-22	2.0	62	211	136	76	241	49	62	274
C-23	6.0	28	82	278	145	329	154	213	454
C-24	19	6.0	354	1211	230	1565	394	1446	1793
C-25	446	1020	1040	45	346	455	277	277	668
C-26	245	1560	534	342	486	849	158	171	999
C-27	101	82	488	130	284	640	249	290	662

7C = Sum of the seven carcinogenic PAHs

Table 3: BaPTEQ and BaPMEQ (µg kg^{-1}) of PAHs in commercial brands of cigarette in Nigeria

	BaA	Chry	BbF	BkF	BaP	IndP	DahA	BaPTEQ	BaA	Chry	BbF	BkF	BaP	IndP	DahA	BaPMEQ
C-1	12.7	0.2	7.1	1.0	11.0	7.4	52.0	91.4	10.4	3.1	17.8	11.1	11.0	22.9	15.1	91.4
C-2	0.0	0.0	0.7	0.2	0.0	3.8	8.0	12.8	0.0	0.4	1.8	2.5	0.0	11.8	2.3	18.8
C-3	1.5	0.0	1.3	0.4	0.0	1.5	52.0	56.7	1.2	0.3	3.3	4.2	0.0	4.7	15.1	28.6
C-4	2.4	0.2	0.0	4.9	0.0	2.3	6.0	15.8	2.0	2.8	0.0	54.0	0.0	7.1	1.7	67.7
C-5	0.0	0.5	0.2	0.1	0.0	34.6	16.0	51.4	0.0	8.9	0.5	0.8	0.0	107	4.6	122
C-6	2.9	0.1	0.9	4.4	2.0	21.7	28.0	60.0	2.4	1.5	2.3	48.6	2.0	67.3	8.1	132
C-7	1.8	0.1	2.2	0.0	6.0	0.4	6.0	16.5	1.5	1.4	5.5	0.1	6.0	1.2	1.7	17.5
C-8	0.3	0.3	1.9	0.5	13.0	0.9	715	732	0.2	5.5	4.8	5.7	13.0	2.8	207	239
C-9	0.5	0.1	1.6	7.4	6.0	0.7	88.0	104	0.4	1.7	4.0	80.9	6.0	2.2	25.5	121
C-10	0.9	0.0	1.2	0.4	9.0	1.8	31.0	44.3	0.7	0.3	3.0	4.5	9.0	5.6	9.0	32.1
C-11	0.0	0.2	0.0	0.0	0.0	0.7	45.0	45.9	0.0	3.3	0.0	0.0	0.0	2.2	13.1	18.5
C-12	0.0	0.5	0.0	2.8	0.0	29.8	136	169	0.0	9.1	0.0	30.4	0.0	92.4	39.4	171
C-13	0.0	0.2	27.3	2.4	80.0	23.6	15.0	149	0.0	3.2	68.3	26.2	80.0	73.2	4.4	255
C-14	0.0	0.1	5.7	2.1	0.0	30.2	98.0	136	0.0	1.9	14.3	23.4	0.0	93.6	28.4	162
C-15	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.1	0.0	0.3	0.0	0.7	0.0	0.0	0.0	0.9
C-16	0.0	0.3	4.9	5.2	2.0	26.2	100	139	0.0	4.2	12.3	56.9	2.0	81.2	29.0	186
C-17	0.0	0.2	0.0	0.8	0.0	1.9	123	126	0.0	2.9	0.0	8.7	0.0	5.9	35.7	53.2
C-18	0.0	0.3	18.1	13.4	77.0	4.6	62.0	175	0.0	4.9	45.3	148	77.0	14.3	18.0	307
C-19	0.0	0.6	65.9	36.8	251	62.0	235	651	0.0	9.9	165	405	251	192	68.2	1090
C-20	3.4	0.4	0.0	0.7	2.0	6.3	40.0	52.8	2.8	6.6	0.0	7.4	2.0	19.5	11.6	49.9
C-21	0.0	0.0	0.0	1.4	0.0	1.8	29.0	32.2	0.0	0.0	0.0	15.2	0.0	5.6	8.4	29.2
C-22	1.3	0.1	0.0	0.9	0.0	4.3	47.0	53.5	1.1	0.8	0.0	9.8	0.0	13.3	13.6	38.6
C-23	0.2	0.0	5.7	0.9	125	2.0	7.0	141	0.2	0.5	14.3	9.8	125	6.2	2.0	158
C-24	0.0	0.4	105	0.5	42.0	0.2	68.0	216	0.0	6.0	263	5.4	42.0	0.6	19.7	337
C-25	0.0	0.3	0.0	0.0	0.0	13.3	45.0	58.6	0.0	4.7	0.0	0.0	0.0	41.2	13.1	59.0
C-26	1.3	0.2	0.0	3.2	0.0	33.6	24.0	62.2	1.1	2.7	0.0	35.0	0.0	104	7.0	150
C-27	0.0	0.3	4.1	0.6	1.0	26.2	24.0	56.2	0.0	4.2	10.3	7.0	1.0	81.2	7.0	111

Risk assessment

Toxicity equivalency factor

The estimated carcinogenic (BaP_{TEQ}) and mutagenic (BaP_{MEQ}) equivalency factors for these brands of cigarette are displayed in Table 3. The BaP_{MEQ} values ranged from 0.1 to 731 µg kg^{-1} with an average value of 128 µg kg^{-1}. In these brands of cigarette brand 15 had the lowest BaP_{TEQ} value, while brands 8 and 19 had higher BaP_{TEQ} values relative to other brands investigated. BaP_{TEQ} values of 11 out of the 26 brands studied were greater than 100 µg kg^{-1}. In these cigarette samples, the major contributors to the carcinogenic potency factor were DahA and IndP. However, in brands 13, 18, 19 and 23 BaP has significant impact on the BaP_{TEQ} values. The BaP_{MEQ} for these brands of cigarette ranged from 0.9 to 1090 µg kg^{-1} with an average of 150 µg kg^{-1}. Brands 19 and 15 had the highest and lowest BaP_{MEQ} values respectively. The BaP_{MEQ} values of 14 of these brands of cigarette were greater than 100 µg kg^{-1}. DahA, IndP and BbF have significant impacts on the mutagenic equivalency factors of these brands of cigarette, while BbF and BaP were the main factors in the mutagenic equivalency values of brands 24 and 23, respectively.

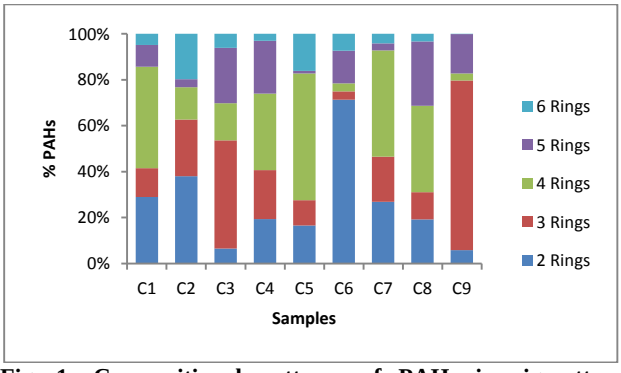

Fig. 1: Compositional patterns of PAHs in cigarette samples C1-C9

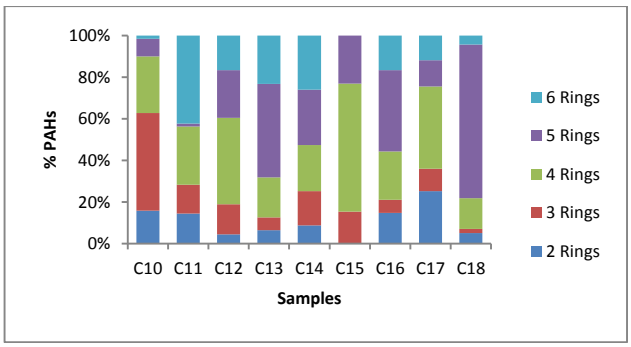

Fig. 2: Compositional patterns of PAHs in cigarette samples C10-C18

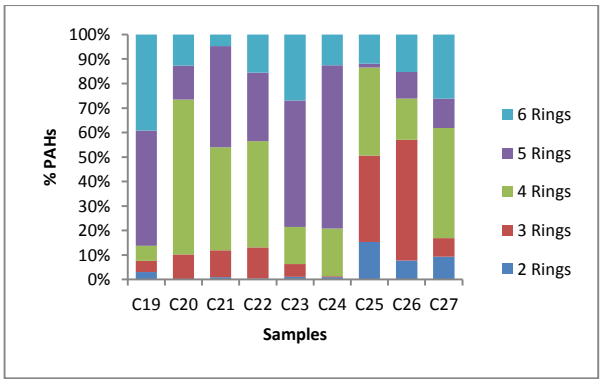

Fig. 3: Compositional patterns of PAHs in cigarette samples C19-C27

Sources of PAHs in the cigarette brands

Isomeric ratios: In environmental compartments, the molecular patterns generated by different sources are useful markers or fingerprints of the processes that generated the PAHs by studying their distribution in the samples (Yunker *et al*., 2002; Mannimo and Orecchio, 2008; Orecchio *et al*., 2009). The ratio of low molecular weight to high molecular weight (LMW/HMW), Ant/(Ant+Phe), BaA/(BaA+Chry), Flt/(Flt+Pyr), IndP/(IndP+BghiP) and BaP/BghiP etc. have

been used for the purposes of source diagnosis in the literature (Yunker *et al*., 2002; Essumang *et al*., 2009; Wang *et al*., 2011; Semlali *et al*., 2012). For example, the ratio of LMW/HMW is a reliable tool for discriminating petrogenic from pyrogenic origins of these contaminants. The lower values of the ratio indicate high prevalence of pyrolytic on petrogenic origin of the PAHs (Orecchio *et al*., 2009). Flt/(Flt+Pyr) is used to distinguish petrogenic and coal/wood, while BaP/BghiP ratio is used to distinguish traffic emissions from coal/biomass combustion sources. The major constraint to the use of isomeric ratios is that it does not provide quantitative information of the contribution of the PAHs source, especially for sample affected by mix sources (Kwon and Choi, 2013). The LMW/HMW ratio in our samples ranged from 0.1 to 3.92 with an average of 0.74. In our samples, the ratio of Ant/(Ant+Phe) ranged from 0.01 to 0.99 with an average value of 0.19. The ratio <0.1 usually depict low temperature sources while a ratio >0.10 depicts high temperature combustion process (Yunker *et al*., 2002; Giacolone *et al*., 2004; Mannimo and Orecchio, 2008; Orecchio *et al*., 2009). In our case, 12 of the 27 brands investigated have ratio values greater than 0.10, which indicated that higher temperature process were the dominant sources of PAHs in these 12 brands of cigarette. The BaA/(BaA+Chry) ranged between 0.01 to 0.5 in 12 of the 27 brands analysed. The BaA/(BaA+Chry) ratio <0.20 indicates petroleum, from 0.20 to 0.35 indicate either petroleum or combustion and >0.35 imply combustion (Orecchio *et al.*, 2009). The BaA/(BaA+Chry) ratio indicates that the source of PAHs in brands 2 and 3 was due to higher temperature process, and brands 6, 10 and 22 come from either petroleum or wood or coal combustion while petroleum combustion was responsible for PAH contamination of the other brands. The Flt/(Flt+Pyr)<0.4 is indicative of petroleum source while Flt/(Flt+Pyr) between 0.4 and 0.5 implies liquid fossil fuel combustion and Flt/(Flt+Pyr)>0.5 suggest biomass/coal combustion. The Flt/(Flt+Pyr) ratio of these samples ranged from 0.01 to 0.70 which indicate that the PAHs contamination of the majority of these samples was derived from petroleum while combustion of grass, wood or coal were the sources of PAHs in brand 2, 17 and 21 which may indicate environmental contamination from agricultural fires or from domestic or industrial heating sources.

Table 4: Diagnostic source ratios of PAHs in commercial brands of cigarette in Nigeria

	LMW/HMW	Ant/ (Ant+Phe)	BaA/ (BaA+Chry)	Flt/ (Flt+Pyr)	BaP/BghiP	CombPAH/ sum PAH
C-1	0.71	0.01	0.41	0.10	0.24	0.20
C-2	1.68	0.13	0.00	0.63	0.00	0.32
C-3	1.16	0.14	0.50	0.23	0.00	0.21
C-4	0.69	0.05	0.13	0.03	0.00	0.34
C-5	0.38	0.05	0.00	0.00	0.00	0.41
C-6	2.97	0.06	0.25	0.00	0.06	0.23
C-7	0.87	0.33	0.17	0.02	0.14	0.13
C-8	0.45	0.26	0.01	0.01	0.15	0.17
C-9	3.92	0.29	0.05	0.12	0.00	0.17
C-10	1.68	0.34	0.32	0.01	0.00	0.07
C-11	0.40	0.61	0.00	0.01	0.00	0.49
C-12	0.23	0.00	0.00	0.03	0.00	0.62
C-13	0.14	0.82	0.00	0.14	1.03	0.55
C-14	0.34	0.00	0.00	0.00	0.00	0.50
C-15	0.18	0.00	0.00	0.00	0.00	0.81
C-16	0.27	0.00	0.00	0.17	0.09	0.63
C-17	0.56	0.00	0.00	0.70	0.00	0.48
C-18	0.08	0.00	0.00	0.00	1.45	0.77
C-19	0.08	0.99	0.00	0.00	0.07	0.81
C-20	0.11	0.00	0.08	0.12	0.05	0.72
C-21	0.13	0.00	0.00	0.52	0.00	0.61
C-22	0.15	0.00	0.21	0.01	0.00	0.44
C-23	0.07	0.00	0.06	0.08	1.00	0.49
C-24	0.01	0.25	0.00	0.00	0.18	0.35
C-25	1.02	0.14	0.00	0.02	0.00	0.22
C-26	1.33	0.79	0.08	0.08	0.00	0.31
C-27	0.20	0.00	0.00	0.10	0.05	0.57

Fluoranthene (Flt), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chry), Phenanthrene (Phe), Lower molecular weight (LMW), High molecular weight (HMW), Combustion PAHs (CombPAH), Anthracene (Ant), Benzo(ghi)perylene (BghiP), Benzo(a)pyrene (BaP)

Principal component analysis: The principal component analysis is a useful statistical tool that represents the total variability of the data with minimum number of factors. Information on the chemical sources responsible for each component can be obtained by critically analysing the loadings of each component (Liu *et al.*, 2009). In this study, four components were extracted and collectively represented 79.77% of the data variance. Factor 1 accounted for 38.5% of the total variance and dominated with high positive loading in Acy, BbF, BkF, BaP, IndP and BghiP. This factor is consistence with the emission characteristics of PAHs from vehicular emissions (Li *et al*. 2007; Zhang *et al*., 2009).
BaP, BbF, BghiP, and IndP are source markers for gasoline emissions (Han *et al.*, 2009), while Acy is a typical product of low temperature pyrogenic processes such as wood combustion (Jerkins, 1996). Factor 2 accounted for 20.49% of the total variance and was heavily weighted by Flu, Phen and Pyr. This factor was consistent with emission characteristics of biomass, wood, and coal combustion (Wang *et al*., 2010). Flu and Phe are the source markers for coke oven (USEPA,1993). Factor 3 accounted for 13.63% and was heavily weighted by Nap and BaA. Nap is associated with incomplete combustion while BaA is usually a marker for diesel combustion (Khalili *et al*., 1995). Factor 4 accounted for 12.57% and has positive loading in Ant. Anthracene is tracer for wood/coal combustion.

Table 5: PCA Factor loadings after Varimax with Kaiser normalization rotation for PAHs in Cigarette sample

PAH	Factors			
Compound	Factor 1	Factor 2	Factor 3	Factor 4
Nap			.693	
Acy	.759			
Ace				.397
Flu		.628		
Phen		.684	.494	.305
Ant		.352		.540
Flt				-.726
Pyr		.822		
BaA			.812	
Chry	.705	.308		
BbF	.559	-.384		
BkF	.872			
BaP	.811	-.364		
IndP	.747			
DahA	.326	.301		
BghiP	.877			
% Variance	**33.05**	**20.49**	**13.63**	**12.57**

Conclusions

The results of the present study revealed that the compositional patterns of PAHs in these brands of cigarette follow the order 2-3 rings>4-rings>5-6 rings. The concentrations of the ΣPAHs in these samples are comparable to concentrations found in cigarette smoke and smokeless tobacco products in the literature. The BaP_{TEQ} and BaP_{MEQ} in the majority of these brands of cigarette samples were greater than the target value in soil. The PAHs isomer ratios indicate that the source of PAHs were mainly from petroleum and combustion of coal, wood and biomass.

Funding and Competing Interests

This research was self-funded and the authors have no competing interest to declare.

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