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COMPARATIVE STUDIES ON POLYAROMATIC HYDROCARBONS (PAHS) IN SOME EDIBLE OIL (SHEA BUTTER, COCONUT OIL AND PALM KERNEL OIL) SOLD IN NIGERIA

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ABSTRACT

Polyaromatic hydrocarbons (PAHs) are ever-present lipophilic substances, having varying levels of concentration in edible oils. Shea butter, coconut oil and palm kernel oil are used in Africa as component of traditional ointment. The study evaluated the concentration of polyaromatic hydrocarbons in Shea butter, coconut oil and palm kernel oil using gas chromatography with flame ionization detector. The polyaromatic hydrocarbons identified and quantified are: napthalene, acenaphthene, fluorene, phenapthrene, fluoranthene, pyrene, chrysene for Shea butter samples; napthalene, acenaphthene, phenanthrene, anthracene, pyrene for coconut oil samples while palm kernel oil samples have napthalene, acenaphthene, acenaphthylene,fluorene, phenanthrene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene and benzo(k)fluoranthene, The concentration of the sum of PAHs of Shea butter ranged from 7.63 - 44.71 ppm, coconut oil samples 7.81 - 19.24 ppm and palm kernel oil samples 25.09 - 71.55 ppm. Shea butter, coconut oil and palm kernel oil samples have concentration of benzo(a)pyrene above the set maximum permissible limit as revealed in the study. It is important that further research on the reduction and/or elimination of PAHs in Shea butter, coconut oil and palm kernel oil be developed.

Keywords: Edible oil, Polyaromatic hydrocarbons, Benzo(a)pyrene, Carcinogenic, Medicinal

INTRODUCTION

Polyaromatic hydrocarbons (PAHs) are ever-present chemical substances found in the environment. They are generated from incomplete combustion of organic substances (Lee *et al*., 1981, Iwegbue *et al.*, 2019; Hussein *et al*., 2016; Purcaro *et al*., 2006). Most polyaromatic hydrocarbons have been classified as mutagens or carcinogens (IARC, 1997). The United State Environmental Protection Agency classified 16 polyaromatic hydrocarbons as priority pollutants: napthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a) anthracene, chrysene, benzo (b) fluoranthene, benzo (k) fluoranthene, benzo(a) pyrene indeno (1,2,3-cd)pyrene, dibenz(a,h)anthracene, benzo(g,h,i)perylene (USEPA, 1985). Benzo(a)pyrene, dibenz(a,h)anthracene, naphthylene, benzo(a)anthracene, benzo (b)fluoranthene, benzo(k)fluoranthene, chrysene and indeno(1,2,3-cd)pyrene are considered probably or possibly carcinogenic to humans and animals (IARC, 1997). PAHs combine more easily with fat and oil than water, since they are lipophilic substances (Zhang *et al*., 2013)

therefore, the possibility of being found in edible oil is inevitable.

Edible oil serve as nutrient and favor in diet, important raw material for chemical, pharmaceutical and medical industries and a source of income both local and foreign (Mercy and Johannes, 2017, Macaire, 2010. Ekpa and Ekpe, 1996). Shea butter derived from the Shea tree (*Vitellaria paradoxa*), Coconut oil from Coconut tree (*Cocos nucifera*) and Palm kernel oil from Palm tree *(Elaeis guineensis*) are classified as edible and essential oils. A substance is edible when it can be eaten without harm and is non toxic to humans. it is essential if it is of high importance and necessary. Shea butter, coconut oil and palm kernel oil had been use as ingredients in medicinal ointments traditionally. Butyric acid in coconut oil is used to treat cancer, while lauric acid is effective in treating viral infections (Padmini *et al*., 201). In west Africa, Nigeria no exception, women use Shea butter, coconut oil and palm kernel oil as rubbing oil on their babies and children. Some women administer palm kernel oil to their babies and children when they are infected by pathogenic microorganisms, and have symptoms such as coughing and sneezing.

The body mass is directly proportional to the dosage administered (Walter et al., 2008). Shea butter, coconut oil and palm kernel oil are edible oil administered to children and babies as well. Edible oils are harmless and non-toxic. There are studies on the evaluation of PAHs on edible vegetable oil, frying oils, snacks and roasted food delicacies. The research work by Purcaro *et al* 2006) on frying oils had benzo(a)pyrene concentration in ranged from trace to 0.7 ppb while oil from fried snacks, had benzo(a)pyrene concerntration below 2 ppb. Princewill-Ogbonna and Adikwu, 2015 worked on refined and unrefined vegetable oil, recorded benzo(a) anthracene concentration of 19.2 ug/kg in refined oil while 4824 µg/kg and 1584 µg/kg was recorded for anthracene and flourene in unrefined oil. In the research work by Onojake *et al* 2017 on roasted food delicacies, acenaphthene, flourene, anthracene, pyrene, chrysene, benzo(k) fluoranthrene, benzo(b)fluoranthrene were detected in roasted plantain, and a sum concentration that ranged from 4.212 - 6.310 mg/kg. Roasted fish have flourene, pyrene, chrysene, benzo(b)flouranthrene, anthracene, having a sum concentration of 5.236 - 7.165 mg/kg. There is lack of records on the PAHs concentration in Shea butter, coconut oil and Palm kernel oil.

The aim of this study was to evaluate and compare the level of polyaromatic hydrocarbons (PAHs) in Shea butter, coconut oil and Palm kernel oil; and investigate which have more mutagenic and carcinogenic PAHs.

MATERIALS AND METHODS

Collection of Samples

The samples of edible oil, (Shea butter, coconut oil and palm kernel oil) were obtained from different markets; Oba market and mile one market (South-south region), Ekitiafor and New markets (South-East region). The samples were identified by the local vendors. All samples were transported aseptically to the laboratory for analysis. The samples were designated as follows; Shea butter (SSO1, SSO2, SSM1,SSM2, SEE1, SEE2, SEN1,SEN2, SSB, SEB), Coconut oil (CSO1,CSO2, CSM1,CSM2, CEE1, CEE2, CEN1, CEN2,

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CSB, CEB) Palm kernel (PSO1, PSO2, PSM1, PSM2, PEE1, PEE2, PEN1, PEN2, PSB, PEB).

Extraction of PAHS from Edible Oil Samples

The extraction of PAHs was carried out using a 1 liter separating funnel. Exactly 5g of the sample was weighed in a sample bottle. Methylene chloride (50 ml) was added to the sample bottle, and shaken for 30 seconds to rinse the inner surface. The mixture was transferred to a 1 liter separating funnel and then shaken for two minutes with periodic venting to release excess pressure. The organic layer was allowed to separate from the water phase for a minimum of ten minutes, and the extract was collected in a 250 ml Erlenmeyer flask.

Methylene chloride (60 ml) was added to the sample bottle and the extraction process was repeated for the second time, the extracts were combined in the 250 ml Erlenmeyer flask. A third extraction was performed in the same manner with 20 ml Methylene chloride.

The combined extracts were poured through a drying column containing packed cotton wool anhydrous sodium sulphate and silica gel. The extract was concentrated to 1.0 ml by passing nitrogen gas and then collected into a GC-vial bottle for GC analysis (AOAC, 2006).

Gas Chromatographic Analysis

The detection of PAHs in edible oil sample was conducted using Model Agilent 6890 Gas Chromatograph with flame ionization detector (GC-FID) fused to a capillary column Restex 30 meter at 0.53 mm ID. The following conditions were maintained during the GC analysis. Detector temperature, 330°C Injector temperature, 250°C , initial oven temperature 100°C, holding temperature/time 4°C , Ramp at 320°C , Standard Used, 610 PAH calibration mix A (Methylene choride), volume injected, 0.1 µl (AOAC, 2006).

RESULTS AND DISCUSSION

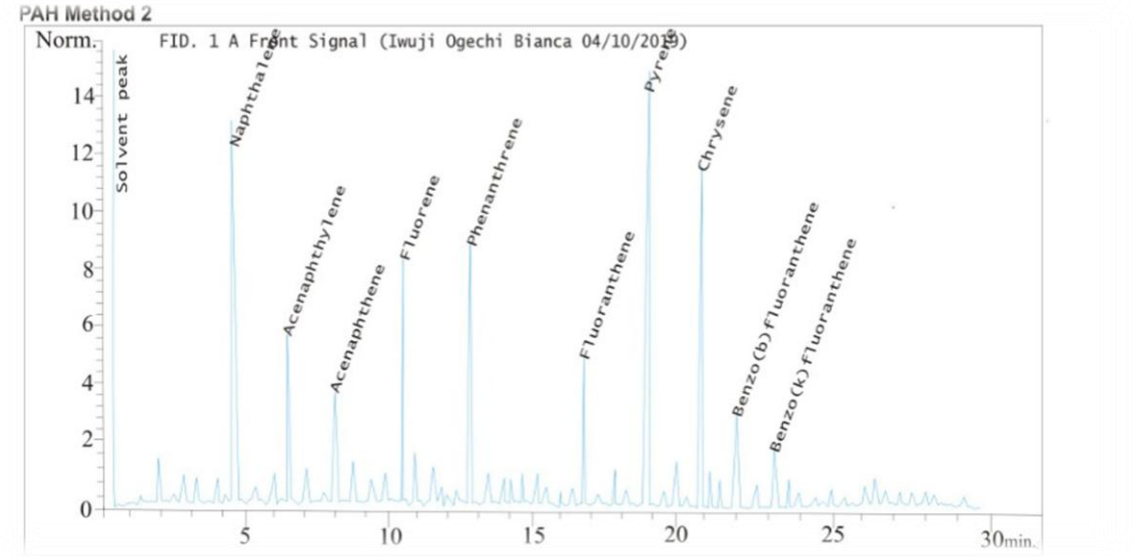

Fig. 1: Chromatogram profile of Polyaromatic hydrocarbons (PAHs) in Palm kernel oil samples

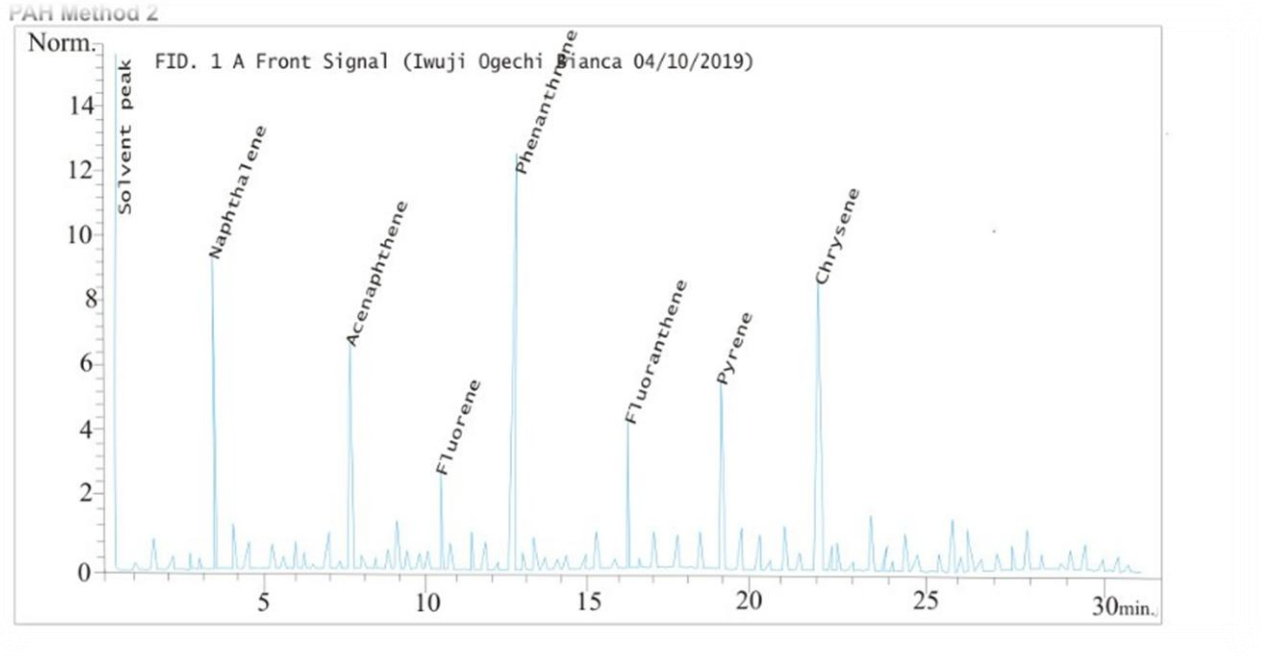

Fig. 2: Chromatogram profile of Polyaromatic hydrocarbons (PAHs) in Shea butter samples

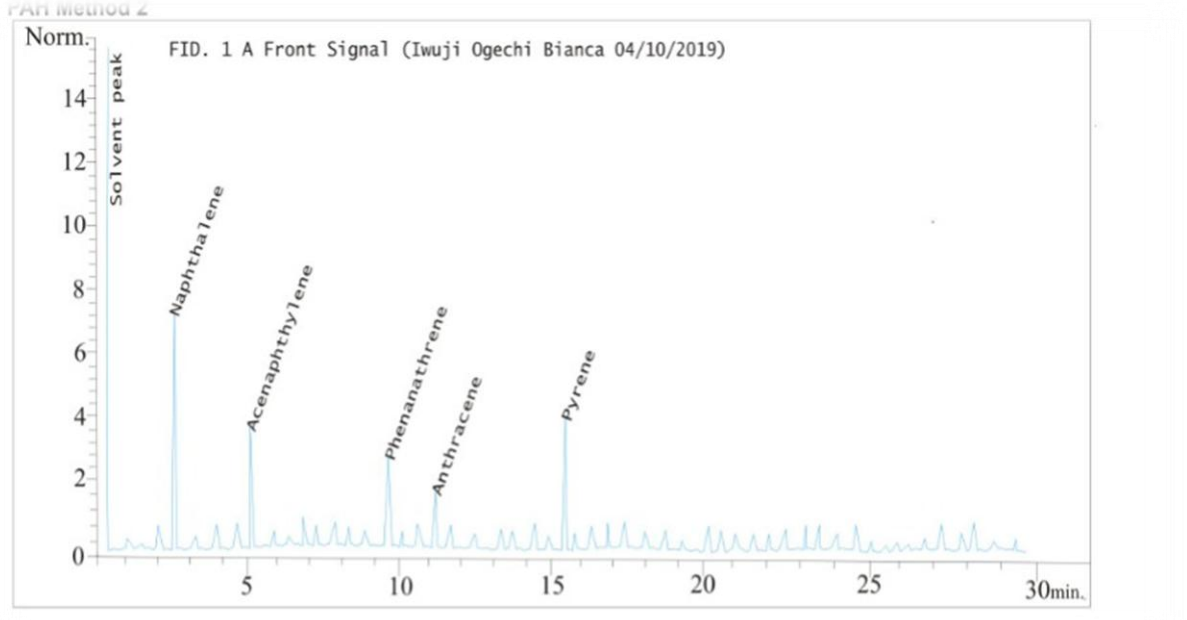

Fig.3: Chromatogram profile of Polyaromatic hydrocarbons (PAHs) in Coconut oil samples

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Table 1: PAH concentrations in Edible oil samples

PAHs (ppm)	Shea butter				Coconut oil				Palm kernel oil			
	SSO1	SSM1	SEE1	SEN1	CSO1	CSM1	CEE1	CEN1	PSO1	PSM1	PEE1	PEN1
Nap	0.57	-	9.79	5.56	0.87	1.54	7.86	3.43	2.64	2.16	7.57	13.32
Acy	-	-	-	-	-	-	3.29	0.22	-	1.28	3.34	5.21
Ace	-	-	6.07	3.32	-	-	-	-	-	-	10.44	3.07
Flu	1.54	1.04	2.15	-	1.26	2.84	-	4.31	3.59	3.20	2.21	7.31
Phe	1.79	1.25	11.77	12.73	1.52	2.16	2.98	3.83	4.81	4.54	9.71	8.74
Ant	-	0.38	-	-	-	-	1.94	-	-	-	6.36	-
Flt	1.33	0.87	3.22	4.01	0.57	-	-	-	2.59	2.31	-	4.89
Pyr	1.02	-	4.09	2.32	1.20	1.67	3.17	2.72	3.67	3.43	5.17	14.32
L-PAH	**6.25**	**3.54**	**37.09**	**27.94**	**5.42**	**8.21**	**19.24**	**14.51**	**17.3**	**16.92**	**44.80**	**56.86**
BaA	-	-	-	-	1.16	1.87	-	0.55	2.89	2.49	-	-
Chy	2.16	2.73	7.62	6.81	0.74	1.25	-	2.32	2.61	4.21	8.61	10.71
BbF	0.82	-	-	1.69	-	0.36	-	0.29	-	-	-	2.05
BkF	1.29	-	-	-	-	1.07	-	0.26	-	-	-	1.71
BaP	1.06	1.36	-	-	0.49	-	-	-	2.29	3.05	3.71	-
IndP	-	-	-	-	-	-	-	-	-	-	-	-
DBahA	-	-	-	-	-	-	-	-	-	-	-	-
BghiP	-	-	-	-	-	-	-	-	-	-	-	-
H-PAH	**5.33**	**4.09**	**7.62**	**8.50**	**2.39**	**4.45**	**-**	**3.42**	**7.79**	**9.75**	**12.32**	**14,47**
T-PAH	**11.58**	**7.63**	**44.71**	**36.44**	**7.81**	**12.66**	**19.24**	**17.93**	**25.09**	**26.67**	**57.12**	**71.33**

Key: Nap = Napthalene, Acy = Acenaphthylene, Ace = Acenaphthene, Flu = Fluorene, Phe = Phenanthrene, Ant = Anthracene, Fit = Fluoranthene, Pyr = Pyrene, BaA = Benz(a) anthracene, Chy = Chrysene, BbF = benzo (b) fluoranthene, BkF = benzo (K) fluoranthene, BaP = Benzo (a) pyrene, IndP = Indeno (1,2,3 -cd) pyrene, DBahA = Dibenz (a,h) anthracene, BghiP = Benzo (g,h,i) perylene.

The chromatogram of the edible oil samples is shown in Figs 1,2,3 having peaks of some priority polyaromatic hydrocarbons. The Palm kernel oil samples chromatogram has naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k) fluoranthene, 10 PAHs of the 16 USEPA PAHs (62.5 %). Naphthalene, benzo(a) anthracene, chrysene, benzo(b) fluoranthene, benzo(k)fluoranthene are five PAHs(31.25%) in palm kernel oil chromatogram that are classified as possible carcinogenic to humans (IARC, 1997; Iwegbue et al., 2019). Shea butter samples chromatogram show the presence of naphthalene, acenapthene, flourene, phenanthrene, fluoranthene, pyrene, chrysene, 7 PAHs of the 16 USEPA PAHs (43.25%). Naphthalene and chrysene are PAHs that are possible carcinogenic in Shea butter chromatogram, 12.5% of the 16 USEPA PAHs. Coconut oil samples chromatogram shows naphthalene, acenathene, phenanthene, anthracene, pyrene, 5 PAHs of the USEPA PAHs (31.25%). Naphthalene is currently classified as possible carcinogenic to humans (Princewill-Ogbonna and Adikwu, 2015). This is 6.25 % of 16 USEPA PAHs. This result is similar to those of other researchers (Purcaro *et al* 2006., Princewill-Ogbonna and Adikwu, 2015). The Chromatogram results (Figs 1,2,3) revealed that the levels of PAHs classified as propably/possible carinogenic to animals and humans have palm kernel oil > Shea butter oil > coconut oil. Benzo(a)anthracene is not present in Shea butter samples.

Table 1 lists the concentration of each PAH and the sum of light PAHs (naphthalene -

pyrene) and heavy PAHs (benzo(a) anthracene - benzo(g,h,i)perylene) in the edible oil samples. Benzo(a)pyrene a classified carcinogen to humans has concentration of 1.06 - 1.36 ppm (25.7 %) in Shea butter, 0.49 ppm (10.4 %) coconut oil and 2.29 - 3.71 ppm (63.8 %) in palm kernel oil samples All samples have benzo(a)pyrene concentration higher than 2ppb (0.49 to 3.71 ppm which is 490 to 3710 ppb), the set maximum level in oil and fat for direct consumption or use as ingredient in food (European Union Commission, Regulation No. 208/2005) . The level of benzo(a)pyrene has Palm kernel oil > Shea butter > coconut oil. This could be attributed to the production process of the edible oils (Purcaro *et al* 2006., Princewill-Ogbonna and Adikwu, 2015). The local production of Shea butter and palm kernel oils involve roasting of the kernel which exposes them to direct heat and combustion gases through which PAHs are introduced. This could be possibly why palm kernel oil and Shea butter samples have more PAHs than coconut oil samples which do not involve roasting in its production procedure (Akinola, et al., 2010). Palm kernel oil having higher levels of benzo(a)pyrene is more carcinogenic as described by Kenam and Ideli (2012).

The concentration of light PAHs in Shea butter samples range between 3.54 - 37.09 ppm (38.4%), 5.42 - 19.24 ppm (13.82%) in coconut oil and 16.92 - 56.86 ppm (45.7%) in palm kernel oil (Table 1). The concentration of heavy PAHs in Shea butter samples range between 4.09 - 8.50 ppm (33.5%), 2.39 - 4.45 ppm (15.7%) and 7.79 - 14.47 ppm (50.8%) The total concentration of light PAHs is greater than the total concentration of heavy PAHs (Table 1) Light PAHs although most are non-carcinogenic, play important role in enhancing the carcinogenic and toxic potential of other PAHs (Iwegbue et al., 2019; Hussein et al., 2016). The percentage of occurrences of the various PAHs in the edible oil samples as indicated in Table 1 has naphthalene, 99.67 %, acenaphthylene, 41.67 %, acenaphthene 33,33 %, fluorene 83.33 %., phenanthrene 100 %, anthracene 16.67 %, fluoranthene 66.67 %, pyrene 99.67 %, benz(a) anthracene 41.67%, chrysene 99.67%, benzo(b)fluoranthene 41.67 %, benzo(k)fluoranthene 33.33 %, benzo(a)pyrene 50 %. Indeno(1,2,3-cd)pyrene, dibenz(a,h)anthracene and benzo(g,h,i)perylene are not presence in all edible oil samples analyzed in this research. Although, the ANOVA on the mean concentration of Polyaromatic hydrocarbons of Shea butter, coconut oil and palm kernel oil shows no significant difference between means at $p \geq 0.05$, the percentage difference of the variances for benzo(a)pyrene concentration has palm kernel oil as 63.8%, Shea butter recorded 25.7% while coconut oil recorded 10.4%. Palm kernel oil samples has more than 50% of PAHs classified as propable/possible carcinogenic. Coconut oil and palm kernel oil both has similar chemical components and uses (Berger et al., 1991). Although coconut oil is more expensive than palm kernel oil; from the research work it has less PAHs that are carcinogenic so in the long run it is safer; taking the human health into consideration.

CONCLUSION

All results obtained indicate that Palm kernel oil possess more carcinogenic PAHs as such, its usage should be limited. Procedures to reduce the PAHs concentration in the edible oil such as

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deodorization and treatment with activated charcoal should be employed. NAFDAC and other regulatory agencies should put more effort in regulating the quality of the edible oil to ensure healthy consumption of edible oils.

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