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Concentrations, sources and risk characterisation of polycyclic aromatic hydrocarbons (PAHs) in green, herbal and black tea products in Nigeria

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ABSTRACT

This study describes the analysis of 16 + 1 US-EPA Priority PAHs (polycyclic aromatic hydrocarbons) occurrence in twenty-three (23) imported and locally manufactured samples of green, black and herbal tea commercially marketed and widely consumed in Nigeria. A gas chromatograph (GC) coupled with an auto-sampler in tandem with a flame ionisation detector (FID) was used for the analysis. The percentage recoveries of each individual PAH varied between 90.24 and 108.92%. The degree of contamination expressed as sum of sixteen priority PAHs (Σ16PAHs) ranged between 1.63 ± 0.33–73.53 ± 6.07 μg/kg, 4.71 ± 0.23–79.61 ± 7.02 μg/kg, and 12.52 ± 0.15–26.89 ± 0.68 μg/kg, for green, herbal and black tea samples, respectively. Benzo(a)pyrene played a significant role in the carcinogenicity and mutagenicity potentials of the samples. The lifetime cancer incident risk assessments indicate higher cancer risk levels in herbal and black teas. Generally, children have higher lifetime probability of cancer risk than adults.

1. Introduction

In recent years, the rate of tea consumption in Nigeria has increased tremendously. Imported and locally produced tea products, including green, black, Oolong, fruit and herbal (moringa, chamomile, mint) teas, are widely consumed as household beverages, while trends also indicate increased consumption at offices, roadside and mobile kiosks, and bus terminals. On any given day, approximately 60% of the Nigerian population are likely to drink tea. More so, all over the world, tea is the second most consumed non-alcoholic drink, the first being water (Tea Fact sheet, 2014). In the last ten years, the world's tea production has increased, to about 5.35 million tonnes in 2013 according to Food and Agriculture Organization of the United Nations (FAO, 2015). Tea is a refreshing beverage that has several health benefits such as antioxidant effects (da Silva Pinto, 2013; Lee and Foo, 2013; Londoño et al., 2015; Wiseman and Rietveld, 2003), body weight control (Rains et al., 2011), cognitive performance (Jäger and Saaby, 2011), and decrease in cardiovascular disease (Wolfram, 2007).

Tea is widely produced from the plant of the *Theaceae* family known as *Camellia sinensis*. Over the years, several methods have been developed for processing different types of tea. These include the non-oxidised and non-fermented process to produce green tea, and the fully oxidised and fermented method, which results in the production of black tea. Herbal teas are widely produced from well-dried, ground (in some products), and processed roots, stem bark, seeds, or flowers of herbaceous plants, and may not necessarily contain *Camellia sinensis* leaves.

Several research studies have reported on the cumulative tea-drinking cancer reduction potential associated with several brands of tea products (Dora et al., 2003; Hakim et al., 2000; Su and Arab, 2002). Tea (*Camellia sinensis*) leaves contain naturally occurring anti-carcinogenic compounds such as flavonoids, theanine and epigallocatechin gallate (EGCG). However, several recent researches have reported that they also contain inorganic and organic carcinogenic contaminants, such as heavy metals, polycyclic aromatic hydrocarbons (PAHs), and pesticides, that are largely due to inputs from human-mediated activities (Fred-Ahmadu and Benson, 2017; Grover et al., 2013; Li et al., 2011; Lin and Zhu, 2004; Drabova et al., 2012; Pincemaille et al., 2014; Singh et al., 2011; EFSA, 2008).

Polycyclic aromatic hydrocarbons (PAHs) are a class of ubiquitous, persistent and toxic organic chemicals with two or more fused aromatic rings. Tea leaves have been shown to accumulate PAHs *via* aerial deposition from the environment and during processing stages like drying by wood or coal burning (Lin and Zhu, 2004). Due to the enormity of

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Table 1
General information about the samples used.

Product name	Code	Country of origin	Flavour	Manufacturer's nutrition facts
Ty-phoo Pure Green Tea	TWG	United Kingdom	–	100% green tea
Heladiv Green Tea	HGT	Sri-Lanka	anti-oxidant	total fat 0%, Na 0%, carbohydrate 0%, protein 0%
Gold blend Green Tea	GBG	Sri-Lanka	lemon & ginger	energy 0%, Na 0%, anti-oxidants 100–200 mg/200 mL
Super blend Green Tea	SBG	Sri Lanka	vanilla	Na 0%
Lipton Green Tea	LGB	USA	blackberry pomegranate	total fat 0 g, Na 0 mg, K 5 mg.
Lipton Green Tea	LGL	USA	lemon & ginseng	Na 0 mg, K 15 mg
Lipton Green Tea	LGR	USA	red goji raspberry	Na 0 mg, K 10 mg
Lipton Green Tea	LGJ	USA	jasmine passion with fruits	Na 0 mg, K 10 mg
Loyd Green Sense	LGS	Poland	aloe vera	Green tea 77%, white tea 20%
Bigelow Green Tea	BGT	USA	decaffeinated, aloe vera	1–8 mg caffeine
Twinings Pure Green Tea	TWG	United Kingdom	–	Green tea
Lipton Yellow Label Tea	LYL	Nigeria	–	energy 2 kJ/ < 1 kcal, protein 0.1 g, sugars 0 g, fat 0 g, fibre 0 g, Na 0 g.
Natural Liver Flush Tea	NLF	China	–	–
Top Tea	TTG	Nigeria	ginger	–
Tranquilizing & Brain nourishing Tea	TBN	China	–	–
Moringa Herbal Tea	MHT	Nigeria	–	–
Sahul Slim Herbal Tea	SSH	India	–	*Garcinia indica* 0.75 g, *Cyperus rotundus* 0.5 g, *Commiphora mukul* 0.6 g, *Garcinia pedunculata* 0.25 g, *Trigonella foenum gracecum* 0.2 g, *Clerodendrum phlomidis* 0.25 g, *Tinospora cordiofolia* 0.25 g, *Emblica officinalis* 0.25 g, *Terminalia chebula* 0.25 g, *Terminalia belerica* 0.25 g, *Zingiber officinale* 0.25 g, *Piper longum* 0.25 g, *Piper nigrum* 0.25 g, *Areca catechu* 0.25 g, *Terminalia arjuna* 0.25 g
Anti-hypertensive Tea	AHT	China	–	–
Joint Care Tea	JCT	China	–	–
Kidney Flush Tea	KFT	China	–	–
Anti-Cancer Tea	ACT	China	–	–
Top Tea	TTL	Nigeria	lime & lemon	–
Top Tea (Regular)	TTR	Nigeria	–	–

consumption of tea across the world populations, monitoring of PAHs concentrations in tea and the assessment of the associated human and environmental health risks is of high priority. More than one hundred PAHs congeners have been identified in environmental matrices, including air, soil, sediment, water, and food. They are known to possess carcinogenic, mutagenic and teratogenic potentials (McGrath et al., 2007) and, in recent years, there has been heightened health concerns regarding their occurrence food products. PAHs are classified as low molecular weight (LMW) PAHs when they possess a 2–3 fused ring structure and high molecular weight (HMW) when there are 4–6 fused aromatic rings. The LMW PAHs have higher vapour pressure and are found largely in air samples while the HMW PAHs are usually bound to particulates. The latter are more resistant to biodegradation, more toxic and persist longer in the environment (Qi et al., 2014).

PAHs are a product of natural processes, such as volcanic eruption, diagenesis, forest fires, crude oil, shale oil etc. (ATSDR, 1995), and anthropogenic activities, which include coal and wood burning, incomplete combustion of petrol and diesel (Mostert et al., 2010), liquid oil and fuel spills (da Silva and Bicego, 2010). US EPA identified 16 priority PAHs (acenaphthene, ACN, acenaphthylene, ACY, anthracene, ANT, benzo(a)anthracene, BaA, benzo(a)pyrene, BaP, benzo(b)fluoranthene, BbF, benzo(g,h,i)perylene, BghiP, dibenzo(a,h)anthracene, DahA, fluoranthene, FLA, benzo(k)fluoranthene, BkF, chrysene, CHR, indeno(1,2,3cd)pyrene, IP, phenanthrene, PHE, naphthalene, NAP, fluorene, FLR, and pyrene, PYR), which are representative of the hundreds of PAHs in the environment. From these 16 PAHs, EFSA (2008) selected PAH4 (BaP + CHR + BaA + BbF) and PAH8 (PAH4 + BkF + BghiP + DahA + IP) as biomarkers of PAHs occurrence in foods, based on their frequency of occurrence above detection limits in sample matrices. The occurrence of PAHs has been reported in many food items including coffee brew (Orecchio et al., 2009), fruits and vegetables (Camargo and Toledo, 2003), cereals (Orecchio and Papuzza, 2009), fish (Nwaichi and Ntorgbo, 2016), meat (Li et al., 2015), sugar cane (Silva et al., 2010), and edible oils (Hao et al., 2015). Some studies on the contamination of PAHs in *Camellia sinensis* have been conducted in China (Lin and Zhu, 2004), Germany (Zuin et al., 2005), Spain (Garcıa-Falcon et al., 2005), Czech Republic (Drabova et al., 2012), Luxembourg (Pincemaille et al., 2014), Argentina (Londoño et al., 2015) and Brazil (Milani et al., 2016). Several other reports on the contamination of PAHs exist (Adisa et al., 2015; Bishnoi et al., 2005; Ciemniak and Mocek, 2010; Chen et al., 2016; Duedahl-Olesen et al., 2015; Fiedler et al., 2002; Grover et al., 2013; Ishizaki et al., 2010; Iwegbue et al., 2015; Kayali-Sayadi et al., 1998; Khiadani et al., 2013; Li et al., 2011; Pincemaille et al., 2014; Schulz et al., 2014; Shi et al., 2016; Singh et al., 2011). However, this study presents a survey of US EPA 16 + 1 priority PAHs in branded green, black and herbal tea samples commercially sold in Nigeria.

Tea consumption has been identified as an important pathway of human exposure to many contaminants, including PAHs, and therefore, PAH contamination of *Camellia sinensis* and herbal tea may have serious health implications. The objectives of this study include (1) determination of the levels of polycyclic aromatic hydrocarbons (PAHs) in tea samples commercially sold and consumed in Nigeria; (2) identification of the sources of polycyclic aromatic hydrocarbons; (3) evaluation of the level of health risk associated with PAHs exposure through consumption of tea.

2. Experimental

2.1. Reagents

2.1.1. Analytical PAHs standard

The standard reference solution used was purchased from Accustandard (New Haven, CT) with components as follows: acenaphthene (ACN), acenaphthylene (ACY), anthracene (ANT), benzo(a)anthracene (BaA), benzo(a)pyrene (BaP), benzo(b)fluoranthene, (BbF),