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Polycyclic aromatic hydrocarbons in foods from the first regional total diet study in Sub-Saharan Africa: contamination profile and occurrence data

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ABSTRACT

As part of the first multi-centre Sub-Saharan Africa Total Diet Study, 660 typical foods from Benin, Cameroon, Mali, and Nigeria were purchased, prepared according to local consumption habits, and pooled into 55 composite samples. These core foods were tested for 15 + 1 EU priority polycyclic aromatic hydrocarbons, which were quantified by isotope dilution and gas chromatography tandem mass spectrometry. The sum of benzo[a]pyrene, benzo[a]anthracene, benzo[b]fluoranthene, and chrysene (PAH4) represented 77% of the 13 genotoxic and carcinogenic PAHs. The highest PAH4 concentration was quantified in sea and fresh water smoked fish (mean: 179.7 μg/kg; max: 560.4 μg/kg) and the PAH4 in all smoked fish composite samples exceeded the EU maximum limit of 12 μg/kg. Further, PAH4 in edible oils (including palm oil and peanut oil) exceeded the EU maximum limit of 10 μg/kg in 50% of the cases (mean 12.0 μg/kg; max: 60.6 μg/kg). These data can be used for assessing the contribution of core foods to dietary exposure and for risk characterization.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) may occur naturally in the environment, but they can also result from anthropic activity (Caballero, Finglas, & Toldra, 2015). Sources of PAHs exposure include diet (Bansal & Kim, 2015; Sun, Wu, & Gong, 2019); air (Kim, Jahan, Kabir, & Brown, 2013) especially for smokers; and contact via the skin (Champmartin, Jeandel, & Monnier, 2017). Food processes such as drying, grilling, and smoking are also likely to generate PAHs (Lee et al., 2016; Lu, Kuhnle, & Cheng, 2018; Rose et al., 2015; Singh, Varshney, & Agarwal, 2016; Zhu et al., 2018). Due to their lipophilic properties, the bioaccumulation of PAHs in adipose tissue is also likely to result in the contamination of fatty foods, such as animal products. The metabolites of PAHs have a propensity to form adducts with DNA (Ewa & Danuta, 2017). Among the PAHs, 15 are classified as genotoxic *in vitro* and *in vivo* (SCF, 2002). The Joint FAO/WHO Expert Committee on Food Additives concluded in its 64th session that 13 PAHs are carcinogenic in experimental animals (WHO, 2006). Based on the available occurrence data, the European Food Safety Authority published an opinion paper (EFSA, 2008) in which benzo[a]pyrene alone was reported to be an inadequate marker of PAH contamination in food. EFSA however concluded that the sum of benzo[a]pyrene, benz[a]anthracene, chrysene, and benzo[b]fluoranthene (PAH4) represented a suitable indicator of the total PAHs contamination in food stuffs. Following the release of this EFSA opinion, Commission Regulation (EC) No 1881/2006 was substituted by Commission Regulation (EU) No 835/2011 (European Commission, 2011), setting maximum limits for both benzo[a]pyrene and PAH4.

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One way of assessing the dietary exposure of populations to food chemicals such as PAHs is through the total diet study (TDS) approach (EFSA, 2011a). The characteristics of a TDS include the representativeness of the sampling and the preparation of the samples "as consumed", so that it represents a pertinent public health risk assessment tool. Most TDSs involve using the pooled sample approach to determine a mean and representative concentration at a low cost.

The World Health Organization (WHO) and the Food and Agriculture Organization of the United Nations (FAO) endorse the TDS methodology, which is both cost-effective and accurate in terms of human exposure to food chemicals. A joint publication by the European Food Safety Agency (EFSA), FAO, and WHO, entitled "Towards a harmonised total diet study approach," serves as a guidance document for research in this field (EFSA, 2011b).

In Sub-Saharan Africa (Cameroon), the first ever TDS targeted pesticide residues (Gimou, Charrondiere, Leblanc, & Pouillot, 2008) as well as metals and trace elements (Gimou et al., 2014). More recently, we carried out a regional TDS, implemented by FAO, in four African countries, between 2014 and 2018, together with four national food safety authorities, in close collaboration with the Centre Pasteur of Cameroon (CPC) and WHO (FAO, 2014; Ingenbleek et al., 2019). We denoted this project as the Sub-Saharan Africa Total Diet Study (SSA-TDS). The purpose of this project was to characterize the chemical contamination levels in typical foods collected in eight different African sites (two per country), located in Benin, Cameroon, Mali, and Nigeria, and to assess the dietary exposure of the populations in those areas. The health risk will subsequently be estimated by comparing the human dietary exposure to food chemicals, such as PAHs, to existing health-based guidance values or toxicological end-points. The methodology used in this study is described elsewhere (Ingenbleek et al., 2017).

In this paper, we present both the occurrence and the profiles of 16 PAHs in the composites of food samples collected from study centres in three coastal areas (Duala, the Littoral of Benin, and Lagos) and five non-coastal areas (Bamako, the Borgou region of Benin, Kano, the North of Cameroon, and Sikasso).

2. Experimental

2.1. Sample selection and preparation of foods as consumed

Food consumption data were obtained via household budget surveys validated by the National Statistics Authorities in Benin (2011), Cameroon (2007), Mali (2010), and Nigeria (2010), with data collected from a total of 72,979 households. The core foods of each study centre were selected on the basis of the relative importance of their mean consumption. This was achieved by selecting 27 to 40 different core foods per country from a list of 84 core foods in order to cover at least 90% of the mean total diet per adult male equivalent per day (Ingenbleek et al., 2017).

Each core food was sampled based on available representation criteria, such as market share or food origins, using 12 subsamples of equal size, prepared as consumed, and pooled into composites fit for laboratory tests. Subsamples were collected and prepared individually according to local recipe books (Gautier & Mallet, 2006; Madubike, 2013; Nya-Njike, 1998; Vinakpon-Gbaguidi, 2003). The books were selected by the competent national authorities for their representation of a typical diet of the populations studied. The expression "prepared individually" is used to denote that no salt, oil, or spices were added to the composite samples. Moreover, unlike in real situations, the core foods from different food subgroups were not mixed together. These recipes allow for the identification of the processes used in the preparation of the foods, especially in terms of cooking time and temperature. However, different ingredients were not mixed unless they belonged to the same core food. The inedible parts were removed at the preparation stage, as a typical consumer would do. Distilled water was used to prepare food instead of tap water to avoid contamination. Although avoiding tap water and condiments may lead to an underestimated concentration, the choice was justified to allow, as much as possible, for the identification of the contamination source.

A total of 660 purchases, or subsamples, were collected and selected for the 16 PAHs (PAH15 + 1) analysis in October 2017, mainly based on the assumption that purchased foods had undergone either drying or smoking processes. Once prepared and evenly pooled by 12 subsamples, 55 composite samples of 16 core foods (*including smoked fish, edible oils, tubers, broth cubes, dehydrated or concentrated milk, and sugar*) were formed (55*12 = 660) and used for laboratory testing.

Samples were frozen and shipped by plane in coolers with dry ice within a timeframe that never exceeded 24 h from kitchen laboratory (in Benin, Cameroon, Mali, and Nigeria) to the testing laboratory located in France.

2.2. Reagents and chemicals

All solvents used (e.g. dichloromethane, hexane, acetone, ethanol, cyclohexane, ethyl acetate, and toluene) were of picograde® quality and obtained from Promochem (Wesel, Germany). Florisil® (100–200 mesh) was obtained from Promochem (Wesel, Germany). SPE EnviChrom-P cartridges (80–160 μm spherical particles) were provided by Sigma-Aldrich (St. Quentin Fallavier, France). The isotopic-labelled internal standard compounds ^{13}C-PAHs and ^{12}C-PAHs were purchased from Promochem. Fluorinated PAHs were purchased from Chiron (Trondheim, Norway).

2.3. Laboratory sample preparation

The preparation of the samples was based on a previous method described by Veyrand et al. (2007). For solid materials, samples were freeze-dried. The dry residue was weighed in order to determine its water content. One gram of dry residue was taken and spiked with a mixture of ^{13}C-labelled internal standard (IS, n = 14). PAHs extraction was performed via pressurized liquid extraction using a Speed Extractor E-914 (Buchi). A cellulose filter was placed at the bottom of the cell and filled up with 15.0 g of Florisil®. The phase was pre-washed in the system with dichloromethane. One gram of the dry residue sample was introduced into the cell, and extraction was performed with a mixture hexane/acetone (50:50, v/v). For the oil matrices, 1.0 g was weighed and not further extracted. Food extracts and oil samples were then purified onto a SPE cartridge (EnviChrom-P) after stationary phase conditioning. After sample application, the SPE phase was washed with a mixture of cyclohexane/ethanol (70:30, v/v). Target compounds were eluted by 12 mL cyclohexane/ethyl acetate (40:60, v/v). Fluorinated PAHs were added at this stage as external standards. Two microliters of the final extract (in toluene) were analysed by GC–MS/MS.

2.4. Gas chromatography (GC-MS/MS) measurements

For GC–MS/MS analysis, a gas chromatograph (Agilent, 7890B Series) and a programmable oven with a temperature up to 350 °C were coupled to an Agilent 7010 triple quadrupole analyser (Agilent Technologies) operating in the electron ionization mode (70 eV). The sample extracts were injected in splitless mode (1 min). The injector temperature was set at 300 °C, whereas the transfer line was programmed at 350 °C. Helium (purity exceeding 99.99%) was used as the carrier gas at a flow rate of 1.0 mL/min. Separation was performed using an Agilent Select PAH column (30 m × 0.25 mm × 0.15 μm) (Les Ulis, France). The column temperature program was set as follows: 110 °C (1 min), 60 °C min^{-1} to 220 °C (0 min), 5 °C min^{-1} to 270 °C (0 min), 3 °C min^{-1} to 295 °C (0 min), 20 °C min^{-1} to 330 °C (10 min). The ion source was heated at 230 °C. Helium and nitrogen (flow set at 2.25 ml/min and 1.5 ml/min, respectively) were used as the collision gas. The PAHs were measured using two specific transitions (Table S1).

2.5. Performances

The method described has been validated and the performances were found fit-for-purpose: limits of quantification (LOQs) ranged from 0.026 to 0.055 μg kg^{-1} based on signal/noise. The linearity was assessed on seven calibration levels for each analyte over 0.1–50 μg kg^{-1} of dry matter. The determination coefficient (R^2) was higher than 0.99 for all analytes. Recoveries ranged from 50% to 70%.

2.6. Internal quality controls and statistical analysis

The accuracy of the method is verified on a yearly basis by way of a proficiency test organized by the European Union Reference Laboratory (JRC-IRMM, Geel, Belgium). In this study, a quality control sample and a blank sample were systematically incorporated in every batch; performances were checked via a quality control chart throughout the whole study.

2.7. Expression of concentration data

The mean and maximum PAH15 + 1 concentration of the 16 core foods (55 composites) is presented in Table 1. In addition, the PAH15 + 1 concentration of a selection of six core foods (25 composites), considering the dietary exposure contributions (manuscript in preparation), is presented in Table 2. Further, the PAH15 + 1 concentrations of all the 55 composites samples are shown in Table S2, together with the concentrations of pyrene, phenanthrene, anthracene, and fluoranthene.

Since these data will be used for the dietary exposure assessment, they are presented using the lower bound (LB: the concentration of non-quantified analytes set to zero) and upper bound (UB: the concentration of non-quantified analytes set to the limit of quantification) hypothesis. When an analyte is not detected, it does not mean that it is not present in the sample but that its concentration lies somewhere between zero and the analytical limit. Using the LB-UB hypothesis provides a range of concentrations around the actual analyte concentration in the sample, which cannot be more precisely defined. The uncertainty due to analytical limits is therefore taken into consideration.

To facilitate the presentation of our results, we only specify LB-UB in cases where some analytes were not detected, meaning that the LB and UB concentrations differ to a certain extent.

3. Results and discussion

3.1. Food contamination

3.1.1. Smoked fish

The multi-centre TDS sampling plan included 72 subsamples of smoked fish (*evenly pooled into 6 composite samples to obtain a mean concentration by study centre*). Smoked fish was not included in the list of Nigerian foods, but was included for Benin, Cameroon, and Mali, where smoked fish is more frequently consumed, according to our food consumption data. Whereas the mean daily consumption of smoked fish was 9.0 g/AME/day in Benin, 4.9 g/AME/day in Cameroon and 6.4 g/AME/day in Mali, it was only 1.9 g/AME/day in Nigeria (Ingenbleek et al., 2017).

Although the fish species could not be collected, it is likely that representative samples of marine species, in terms of their availability on the market, were collected in the Littoral of Benin, Duala, and Lagos. In Borgou, North Cameroon, Bamako, and Sikasso, fresh water fish species were available in the local markets. The fish were bought and washed, and any bones removed according to household practices. The fish were then boiled with distilled water in order to simulate the preparation of a stew.

Interestingly, the smoked fish samples collected in Benin accounted for both the highest (984.7 μg/kg in Borgou) and lowest (55.2 μg/kg in the Littoral) PAH15 + 1 concentrations, as shown in Table 2. PAH15 + 1 concentrations varied depending on the study centres from which the samples were collected (Table 2). The coefficients of variation (*SD/mean concentration*) of PAH15 + 1 congeners among 6 smoked fish composites were relatively high (111%).

Out of the seven SSA-TDS composites with the highest PAH15 + 1 concentration rank, six were smoked fish samples.

Mahugija and Njale (2018a) compared the occurrence of PAHs in three fish species caught in Tanzania, which were either sun-dried or smoked, and observed that: (i) PAHs levels were lower in sun-dried fish than in smoked fish, and (ii) the fish species did not significantly influence PAHs content. However, the same team recently showed that a reduction in PAHs concentrations by washing smoked-fish is species-specific (Mahugija & Njale, 2018b), ranging from 31.5 to 86.5%, depending on the species.

A high variance in PAH15 + 1 concentrations was found between the smoked fish composite samples from different study centres, suggesting that some local smoking practices generate more PAHs than others. PAHs levels may be significantly influenced by the lignin content of the type of wood used for the smoking process (García-Falcon & Simal-Gándara, 2005), as well as the fat content and the smoke-curing duration (Essumang, Dodoo, & Adjei, 2013). It would be useful to investigate this further and establish a typology of smoking practices, to determine which ones should be prohibited and which should be monitored. For example, it has been previously reported that some smoked-fish producers burn plastic bags or car tyres to generate the smoke. The Codex code of practice can provide a useful reference for the training of producers (Codex Alimentarius, 2009).

Whereas the use of innovative fish-smoking methods (Essumang, Dodoo, & Adjei, 2014) could be adopted to reduce the occurrence of high PAHs concentrations in fish, sun-drying may also be a safe alternative to fish smoking (Mahugija & Njale, 2018a). However, both the microbiological safety and the organoleptic perception of sun-dried fish compared to that of smoked fish in the context of African countries would need to be assessed. Alternatively, to preserve organoleptic preferences for the flavour of smoked fish, the use of approved smoke flavourings could be promoted and used, as prescribed by the Codex Code of Practice (CAC/RCP 52–2003), to preserve consumer preference while reducing exposure to PAHs. Specific marinades used for the reduction of PAHs concentrations in meat could also be explored for fish in the future (Viegas, Yebra-Pimentel, Martínez-Carballo, Simal-Gandara, & Ferreira, 2014). The antioxidant effect of spices may also contribute to prevent the formation of PAHs (Lu et al., 2018).

3.1.2. Edible vegetable oils

We collected 120 subsamples of the most common edible oils, including palm oil (48), peanut oil (24), and *other vegetable oils* (48): cottonseed oil (24) in Cameroon, cottonseed oil (9) and shea oil (3) in Mali. In Nigeria, the content of the *other vegetable oil* composite subsamples consisted of twelve samples: six branded samples made from soya or bleached palm oil, and another six whose sources could not be determined.

After being heated according to the local recipes, the 120 subsamples were aggregated into 10 composites by oil type and by study centre. Each was analysed as per the following sections.

3.2. Palm oil and palm nut

The 4 palm oil composites from the Littoral of Benin, Duala, Lagos, and Kano (Table 2) were tested for PAH15 + 1. Whereas the palm oil subsamples collected in Duala consisted of refined industrial oil, the subsamples collected in Benin and Nigeria were artisanal products (red palm oil samples). The refined bleached palm oil sample contained the lowest PAH15 + 1 concentration (LB: 1.9; UB: 2.0 μg/kg). The mean PAH15 + 1 concentration in the palm oil samples was LB: 22.6; UB: 22.7 μg/kg and the maximum was LB: 40.7; UB: 40.9 μg/kg (Littoral of

Table 1
Mean and maximum concentration (μg/kg) of PAH15+1 of 55 composite samples representing 16 core foods.

CORE FOOD	N			BaP	BaA	CHR	BbF	PAH4	BkF	IP	DbahA	BghiP	PAH8	MCH	BjF	DbalP	DbaeP	DbaiP	DbahP	PAH13	BcF	CPP	PAH15+1
Smoked fish	6	Mean	LB	26.34	51.60	70.58	31.15	**179.7**	11.93	13.59	2.21	10.31	**217.7**	3.59	17.21	1.58	1.04	0.26	0.17	**231.2**	12.53	56.05	**310.1**
			UB	26.34	51.60	70.58	31.15	**179.7**	11.93	13.59	2.21	10.31	**217.7**	3.63	17.21	1.61	1.04	0.26	0.17	**231.3**	12.53	56.05	**310.2**
		Max	UB	77.50	161.00	233.00	88.90	**560.4**	35.50	34.40	5.60	25.10	**661.0**	14.00	49.90	4.66	2.29	0.65	0.56	**708.0**	42.60	209.00	**984.7**
Peanut oil	2	Mean	LB	1.24	1.51	1.84	1.16	**5.7**	0.53	0.98	0.18	3.61	**11.0**	0.00	0.75	0.00	0.14	0.04	0.03	**8.4**	0.28	2.13	**14.4**
			UB	1.24	1.51	1.84	1.16	**5.7**	0.53	0.98	0.20	3.61	**11.0**	0.09	0.75	0.02	0.16	0.05	0.04	**8.5**	0.51	2.13	**14.8**
		Max	UB	2.09	2.10	2.30	1.89	**8.4**	0.86	1.67	0.35	5.21	**16.5**	0.09	1.24	0.02	0.28	0.08	0.06	**13.0**	0.55	2.88	**18.5**
Other vegetable oil	4	Mean	LB	2.04	4.61	6.73	2.44	**15.8**	0.99	1.06	0.20	1.12	**19.2**	0.00	1.27	0.09	0.09	0.03	0.01	**19.5**	0.97	3.42	**25.0**
			UB	2.04	4.61	6.73	2.44	**15.8**	0.99	1.06	0.22	1.12	**19.2**	0.13	1.27	0.10	0.11	0.04	0.02	**19.7**	1.00	3.42	**25.3**
		Max	UB	7.56	17.90	26.10	9.04	**60.6**	3.67	3.67	0.70	3.49	**72.1**	0.39	4.71	0.37	0.29	0.13	0.05	**74.6**	3.87	13.30	**95.2**
Palm oil	4	Mean	LB	2.00	3.25	3.82	2.23	**11.3**	1.11	1.78	0.26	1.79	**16.2**	0.00	1.50	0.00	0.18	0.05	0.01	**16.2**	1.17	3.46	**22.6**
			UB	2.00	3.25	3.82	2.23	**11.3**	1.11	1.78	0.26	1.79	**16.2**	0.05	1.50	0.03	0.18	0.05	0.03	**16.3**	1.18	3.46	**22.7**
		Max	UB	3.63	6.03	6.30	3.85	**19.8**	2.11	3.07	0.38	2.81	**28.2**	0.08	2.71	0.10	0.28	0.08	0.04	**28.7**	1.99	7.49	**40.9**
Other nuts/seeds	2	Mean	LB	1.86	3.59	4.04	1.86	**11.3**	0.90	1.10	0.18	0.97	**14.5**	0.00	1.01	0.15	0.10	0.04	0.02	**14.8**	0.88	4.18	**20.8**
			UB	1.88	3.59	4.04	1.86	**11.4**	0.90	1.10	0.19	0.99	**14.5**	0.05	1.01	0.15	0.11	0.04	0.03	**14.9**	0.89	4.18	**21.0**
		Max	UB	3.72	7.13	8.01	3.70	**22.6**	1.79	2.18	0.36	1.93	**28.8**	0.09	2.01	0.29	0.20	0.07	0.04	**29.6**	1.76	8.28	**41.6**
Other fat/oil	1	Mean	LB	0.22	0.32	0.41	0.28	**1.2**	0.13	0.21	0.05	0.34	**2.0**	0.00	0.13	0.00	0.05	0.00	0.00	**1.8**	0.00	0.16	**2.3**
			UB	0.22	0.32	0.41	0.28	**1.2**	0.13	0.21	0.05	0.34	**2.0**	0.03	0.13	0.03	0.05	0.05	0.03	**1.9**	0.12	0.16	**2.6**
		Max	UB	0.22	0.32	0.41	0.28	**1.2**	0.13	0.21	0.05	0.34	**2.0**	0.03	0.13	0.03	0.05	0.05	0.03	**1.9**	0.12	0.16	**2.6**
Palm nut	2	Mean	LB	0.56	0.71	0.84	0.61	**2.7**	0.25	0.61	0.08	0.67	**4.3**	0.00	0.41	0.08	0.07	0.00	0.00	**4.2**	0.25	0.55	**5.7**
			UB	0.57	0.71	0.84	0.61	**2.7**	0.25	0.61	0.08	0.68	**4.3**	0.03	0.41	0.08	0.08	0.02	0.01	**4.3**	0.27	0.55	**5.8**
		Max	UB	1.12	1.38	1.64	1.20	**5.3**	0.50	1.20	0.15	1.34	**8.5**	0.05	0.80	0.15	0.14	0.03	0.01	**8.4**	0.49	1.05	**11.3**
Chili/peper	3	Mean	LB	0.20	0.75	1.20	0.30	**2.4**	0.15	0.12	0.01	0.09	**2.8**	0.00	0.30	0.00	0.00	0.00	0.00	**3.0**	0.25	1.21	**4.6**
			UB	0.20	0.75	1.20	0.30	**2.4**	0.15	0.12	0.01	0.09	**2.8**	0.01	0.30	0.01	0.01	0.01	0.01	**3.1**	0.28	1.21	**4.7**
		Max	UB	0.32	1.19	1.64	0.42	**3.4**	0.24	0.17	0.02	0.15	**4.0**	0.01	0.47	0.01	0.01	0.01	0.01	**4.3**	0.44	1.88	**6.6**
Cassava dry	6	Mean	LB	0.07	0.13	0.16	0.08	**0.4**	0.04	0.07	0.00	0.06	**0.6**	0.00	0.06	0.00	0.00	0.00	0.00	**0.6**	0.00	0.13	**0.8**
			UB	0.08	0.13	0.16	0.08	**0.4**	0.04	0.07	0.03	0.08	**0.7**	0.02	0.06	0.01	0.02	0.02	0.02	**0.7**	0.03	0.13	**1.0**
		Max	UB	0.19	0.29	0.35	0.19	**1.0**	0.09	0.18	0.05	0.21	**1.6**	0.03	0.14	0.02	0.04	0.03	0.03	**1.6**	0.08	0.26	**2.2**
Broth/ bouillon cube	2	Mean	LB	0.00	0.10	0.15	0.10	**0.3**	0.04	0.06	0.00	0.00	**0.4**	0.00	0.04	0.00	0.00	0.00	0.00	**0.5**	0.00	0.08	**0.5**
			UB	0.10	0.10	0.15	0.10	**0.4**	0.04	0.06	0.04	0.17	**0.7**	0.02	0.04	0.01	0.04	0.01	0.01	**0.7**	0.03	0.08	**1.0**
		Max	UB	0.10	0.10	0.16	0.12	**0.5**	0.04	0.06	0.04	0.17	**0.8**	0.02	0.04	0.01	0.04	0.01	0.01	**0.8**	0.03	0.09	**1.0**
Peanuts	5	Mean	LB	0.00	0.09	0.09	0.03	**0.2**	0.01	0.02	0.00	0.00	**0.2**	0.00	0.02	0.00	0.00	0.00	0.00	**0.3**	0.00	0.11	**0.4**
			UB	0.07	0.09	0.10	0.05	**0.3**	0.03	0.03	0.04	0.08	**0.5**	0.02	0.03	0.02	0.03	0.03	0.03	**0.6**	0.04	0.11	**0.8**
		Max	UB	0.09	0.19	0.20	0.10	**0.6**	0.05	0.07	0.05	0.10	**0.9**	0.05	0.08	0.03	0.04	0.05	0.05	**1.1**	0.06	0.29	**1.5**
Sugar	6	Mean	LB	0.00	0.01	0.00	0.00	**0.0**	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	**0.0**
			UB	0.09	0.02	0.06	0.05	**0.2**	0.03	0.03	0.05	0.12	**0.4**	0.02	0.02	0.02	0.04	0.03	0.03	**0.5**	0.02	0.03	**0.6**
		Max	UB	0.11	0.03	0.08	0.05	**0.2**	0.03	0.04	0.05	0.18	**0.5**	0.04	0.02	0.03	0.04	0.06	0.07	**0.6**	0.03	0.04	**0.9**
Yam dry	1	Mean	LB	0.03	0.06	0.06	0.03	**0.2**	0.01	0.02	0.00	0.00	**0.2**	0.00	0.02	0.00	0.00	0.00	0.00	**0.2**	0.00	0.07	**0.3**
			UB	0.03	0.06	0.06	0.03	**0.2**	0.01	0.02	0.01	0.04	**0.3**	0.01	0.02	0.01	0.01	0.01	0.01	**0.3**	0.02	0.07	**0.4**
		Max	UB	0.03	0.06	0.06	0.03	**0.2**	0.01	0.02	0.01	0.04	**0.3**	0.01	0.02	0.01	0.01	0.01	0.01	**0.3**	0.02	0.07	**0.4**
Concentrated/ dehydrated milk	4	Mean	LB	0.00	0.03	0.00	0.00	**0.0**	0.00	0.01	0.00	0.00	**0.0**	0.00	0.01	0.00	0.00	0.00	0.00	**0.0**	0.00	0.03	**0.1**
			UB	0.07	0.03	0.05	0.04	**0.2**	0.02	0.03	0.04	0.10	**0.4**	0.02	0.02	0.02	0.03	0.02	0.02	**0.4**	0.03	0.03	**0.6**
		Max	UB	0.11	0.03	0.06	0.05	**0.2**	0.03	0.04	0.05	0.18	**0.5**	0.05	0.02	0.03	0.04	0.05	0.05	**0.6**	0.04	0.04	**0.9**
Yam fresh	5	Mean	LB	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	0.00	0.01	**0.0**	0.00	0.00	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	**0.0**
			UB	0.02	0.01	0.02	0.01	**0.1**	0.01	0.01	0.01	0.03	**0.1**	0.01	0.01	0.01	0.01	0.01	0.01	**0.2**	0.01	0.01	**0.2**
		Max	UB	0.04	0.01	0.02	0.02	**0.1**	0.01	0.01	0.02	0.06	**0.2**	0.01	0.01	0.01	0.01	0.01	0.01	**0.2**	0.01	0.01	**0.3**
Potato fresh	2	Mean	LB	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	0.00	0.00	0.00	0.00	**0.0**	0.00	0.00	**0.0**
			UB	0.02	0.01	0.01	0.01	**0.0**	0.01	0.01	0.01	0.02	**0.1**	0.01	0.01	0.01	0.01	0.01	0.01	**0.1**	0.01	0.01	**0.2**
		Max	UB	0.02	0.01	0.01	0.01	**0.1**	0.01	0.01	0.01	0.02	**0.1**	0.01	0.01	0.01	0.01	0.01	0.01	**0.1**	0.01	0.01	**0.2**
TOTAL	**55**																						

(*continued on next page*)

Table 1 (*continued*)

Legend:			PAH4	PAH8	PAH13	PAH15 + 1
	BaP	Benzo[a]pyrene	*	*	*	*
	BaA	Benz[a]anthracene	*	*	*	*
	CHR	Chrysene	*	*	*	*
	BbF	Benzo[b]fluoranthene	*	*	*	*
	BkF	Benzo[k]fluoranthene		*	*	*
	IP	Indeno[1,2,3,c-d]pyrene		*	*	*
	DbahA	Dibenz[a;h]anthracene		*	*	*
	BghiP	Benzo[g;h;i]perylene		*		*
	MCH	5-Methylchrysene			*	*
	BjF	Benzo[j]fluoranthene			*	*
	DbalP	Dibenzo[a;l]pyrene			*	*
	DbaeP	Dibenzo[a;e]pyrene			*	*
	DbaiP	Dibenzo[a;i]pyrene			*	*
	DbahP	Dibenzo[a;h]pyrene			*	*
	BcF	Benzo[c]fluorene				*
	CPP	Cyclopenta[c;d]pyrene				*

Benin). The palm oil used in food stuffs had been previously identified as a source of PAHs, with a mean level of 23 μg/kg (Fernandez-Gonzalez, Yebra-Pimentel, Martínez-Carballo, & Simal-Gándara, 2012).

Palm nut samples are equivalent to palm oil, which is the edible fraction of the palm nut, except that it is extracted at home, in a process involving crushing the nut, hot water, and phase separation. The mean PAH15 + 1 levels (LB: 5.7; UB: 5.8 μg/kg) in the palm nut extracts, or home-made palm oil, were lower than the ones observed in the palm oil available on the market.

3.3. Peanut oil and peanuts

Peanut oil composite samples were collected in Benin (Littoral) and Nigeria (Kano). The composite from Benin (Table 2) contained LB: 10.4; UB: 11.1 μg/kg PAH15 + 1. The peanut oil composite formed from subsamples collected in Kano, however, contained higher PAH15 + 1 concentrations (LB: 18.4; UB: 18.5 μg/kg).

In comparison with peanut oil, five peanut composite samples contained lower PAH15 + 1 concentrations (mean LB: 0.4; UB: 0.8; max UB: 1.5 μg/kg). The origin of the peanuts used for the peanut oil production was not identified, thus it was not possible to draw any conclusions regarding the impact of the oil extraction process on PAHs concentration.

3.4. Other vegetable oil

The composites of the food subgroup "*other vegetable oil*" exclusively consisted of cottonseed oil in Duala and North Cameroon. In Mali, however, it consisted of a mix of 75% cottonseed oil and 25% shea oil (also known as *karité*). The "other vegetable oil" composite from Nigeria was not known due to a lack of sufficient information collected from the market, but followed the sampling approach (Ingenbleek et al., 2017) and was therefore considered as being representative of food consumption habits.

It is unclear whether cottonseed oil or shea oil contributed most to the PAH15 + 1 content of the composite from Mali.

The coefficients of variation (SD/mean) of PAH15 + 1 concentrations among 10 edible oil composite samples were even higher than in the smoked fish composites (131%).

In 2016, Hao et al., 2016 studied the influence of deep-frying time, which increases PAHs content, especially the high ring (5-ring and above) content in edible oils, thus it is recommended to avoid repeated use of edible oils. In 2018, Zhu et al. concluded that the oil type influences the kinetics of PAH formation during the deep-frying process. In the case of edible oils collected for the SSA-TDS, it is unclear to which extent PAHs are mainly generated during the extraction process, prior to reaching consumers' home or at home (e.g. by frying, which was simulated in kitchen laboratories).

The comparison of PAH contents in typically-consumed oils in Africa before and after heating is needed in order to identify effective actions and reduce the occurrence of PAHs in typical African diets.

3.5. Chili/pepper

According to the study methodology (Ingenbleek et al., 2017), the core food selection is essentially based on food consumption data (by weight). Following this approach, chili pepper samples were only collected in three study centres (the Littoral of Benin, Lagos, and Kano (in Nigeria)).

The chili/pepper composite from Benin contained LB: 1.2; UB: 1.4 μg/kg PAH15 + 1, whereas chili pepper collected and prepared in Lagos contained LB: 5.9; UB: 6.0 μg/kg PAH15 + 1 (Table 2). Monago-Maraña, Pérez, Escandar, Muñoz de la Peña, and Galeano-Díaz (2016) and Fasano, Yebra-Pimentel, Martínez-Carballo, and Simal-Gándara (2016) detected higher concentrations in Spanish paprika samples, that were smoke-dried. We acknowledge that these products are not really

Table 2
Concentration (μg/kg) of PAH15 + 1 in a selection of 25 composite samples representing 6 core foods.

COMPOSITE SAMPLE	COUNTRY	CENTRE		BaP	BaA	CHR	BbF	PAH4	BkF	IP	DbahA	BghiP	PAH8	MCH	BjF	DbalP	DbaeP	DbaiP	DbahP	PAH13	BcF	CPP	PAH15 + 1
Smoked fish	BENIN	Littoral	LB	3.41	9.99	11.4	5.79	**30.6**	2.7	3.2	0.48	2.81	**39.8**	0.14	3.54	0	0.24	0	0	**40.9**	2.68	8.8	**55.2**
			UB	3.41	9.99	11.4	5.79	**30.6**	2.7	3.2	0.48	2.81	**39.8**	0.14	3.54	0.16	0.24	0.02	0.01	**41.1**	2.68	8.8	**55.4**
		Borgou	LB	77.5	161	233	88.9	**560.4**	35.5	34.4	5.6	25.1	**661.0**	14	49.9	4.7	2.29	0.65	0.56	**708.0**	42.6	209	**984.7**
			UB	77.5	161	233	88.9	**560.4**	35.5	34.4	5.6	25.1	**661.0**	14.00	49.9	4.7	2.29	0.65	0.56	**708.0**	42.6	209	**984.7**
	CAMEROON	Duala	LB	10.5	21.8	22.2	10.7	**65.2**	4.97	5.3	0.83	4.32	**80.6**	0	6.8	0.64	0.43	0.13	0.06	**84.4**	6.61	29.8	**125.1**
			UB	10.5	21.8	22.2	10.7	**65.2**	4.97	5.3	0.83	4.32	**80.6**	0.22	6.8	0.64	0.43	0.13	0.06	**84.6**	6.61	29.8	**125.3**
		North	LB	32.5	54.7	68.8	42.4	**198.4**	13.4	18.5	3.07	14.3	**247.7**	2.71	20.7	2.32	1.71	0.37	0.18	**261.4**	9.48	45.3	**330.4**
			UB	32.5	54.7	68.8	42.4	**198.4**	13.4	18.5	3.07	14.3	**247.7**	2.71	20.7	2.32	1.71	0.37	0.18	**261.4**	9.48	45.3	**330.4**
	MALI	Bamako	LB	17.4	30.0	46.3	20.3	**114.0**	7.35	10.6	1.72	8.00	**141.7**	2.52	11.5	1.1	0.87	0.2	0.1	**150.0**	6.33	22.4	**186.7**
			UB	17.4	30.0	46.3	20.3	**114.0**	7.35	10.6	1.72	8.00	**141.7**	2.52	11.5	1.1	0.87	0.2	0.1	**150.0**	6.33	22.4	**186.7**
		Sikasso	LB	16.7	32.1	41.8	18.8	**109.4**	7.68	9.51	1.55	7.30	**135.4**	2.16	10.8	0.77	0.71	0.2	0.09	**142.9**	7.49	21	**178.7**
			UB	16.7	32.1	41.8	18.8	**109.4**	7.68	9.51	1.55	7.3	**135.4**	2.16	10.8	0.77	0.71	0.2	0.09	**142.9**	7.49	21	**178.7**
Palm oil	BENIN	Littoral	LB	3.63	6.03	6.3	3.85	**19.8**	2.11	3.07	0.38	2.81	**28.2**	0	2.71	0	0.28	0.08	0	**28.4**	1.99	7.49	**40.7**
			UB	3.63	6.03	6.3	3.85	**19.8**	2.11	3.07	0.38	2.81	**28.2**	0.08	2.71	0.1	0.28	0.08	0.03	**28.7**	1.99	7.49	**40.9**
	CAMEROON	Duala	LB	0.2	0.15	0.21	0.25	**0.8**	0.09	0.23	0.05	0.39	**1.6**	0	0.11	0	0.05	0	0	**1.3**	0	0.17	**1.9**
			UB	0.2	0.15	0.21	0.25	**0.8**	0.09	0.23	0.05	0.39	**1.6**	0.03	0.11	0.01	0.05	0.01	0.02	**1.4**	0.03	0.17	**2.0**
	NIGERIA	Lagos	LB	1.79	3.1	3.8	2.13	**10.8**	1.01	1.6	0.26	1.67	**15.4**	0	1.36	0	0.17	0.04	0	**15.3**	1.1	2.54	**20.6**
			UB	1.79	3.1	3.8	2.13	**10.8**	1.01	1.6	0.26	1.67	**15.4**	0.06	1.36	0.01	0.17	0.04	0.02	**15.4**	1.1	2.54	**20.7**
		Kano	LB	2.36	3.73	4.96	2.67	**13.7**	1.21	2.21	0.34	2.29	**19.8**	0	1.83	0	0.23	0.06	0.04	**19.6**	1.58	3.63	**27.1**
			UB	2.36	3.73	4.96	2.67	**13.7**	1.21	2.21	0.34	2.29	**19.8**	0.03	1.83	0.01	0.23	0.06	0.04	**19.7**	1.58	3.63	**27.2**
Other vegetable oil	CAMEROON	Duala	LB	0.15	0.18	0.27	0.17	**0.8**	0.07	0.11	0	0.16	**1.1**	0	0.07	0	0	0	0	**1.0**	0	0.09	**1.3**
			UB	0.15	0.18	0.27	0.17	**0.8**	0.07	0.11	0.05	0.16	**1.2**	0.04	0.07	0.01	0.04	0.01	0.01	**1.2**	0.03	0.09	**1.5**
		North	LB	0.11	0.16	0.29	0.14	**0.7**	0.06	0.08	0	0.15	**1.0**	0	0.06	0	0	0	0	**0.9**	0	0.14	**1.2**
			UB	0.11	0.16	0.29	0.14	**0.7**	0.06	0.08	0.05	0.15	**1.0**	0.06	0.06	0.01	0.04	0.01	0.02	**1.1**	0.05	0.14	**1.4**
	MALI	Bamako	LB	7.56	17.9	26.1	9.04	**60.6**	3.67	3.67	0.7	3.49	**72.1**	0	4.71	0.37	0.29	0.13	0.05	**74.2**	3.87	13.3	**94.9**
			UB	7.56	17.9	26.1	9.04	**60.6**	3.67	3.67	0.7	3.49	**72.1**	0.39	4.71	0.37	0.29	0.13	0.05	**74.6**	3.87	13.3	**95.2**
	NIGERIA	Lagos	LB	0.32	0.19	0.27	0.39	**1.2**	0.14	0.36	0.09	0.67	**2.4**	0	0.22	0	0.08	0	0	**2.1**	0	0.14	**2.9**
			UB	0.32	0.19	0.27	0.39	**1.2**	0.14	0.36	0.09	0.67	**2.4**	0.03	0.22	0.01	0.08	0.02	0.01	**2.1**	0.04	0.14	**3.0**
Peanut oil	BENIN	Littoral	LB	0.39	0.91	1.38	0.43	**3.1**	0.19	0.28	0	5.21	**8.8**	0	0.26	0	0	0	0	**3.8**	0	1.37	**10.4**
			UB	0.39	0.91	1.38	0.43	**3.1**	0.19	0.28	0.04	5.21	**8.8**	0.08	0.26	0.01	0.04	0.01	0.01	**4.0**	0.46	1.37	**11.1**
	NIGERIA	Kano	LB	2.09	2.1	2.3	1.89	**8.4**	0.86	1.67	0.35	2.00	**13.3**	0	1.24	0	0.28	0.08	0.06	**12.9**	0.55	2.88	**18.4**
			UB	2.09	2.1	2.3	1.89	**8.4**	0.86	1.67	0.35	2.00	**13.3**	0.09	1.24	0.02	0.28	0.08	0.06	**13.0**	0.55	2.88	**18.5**
Chili/peper	BENIN	Littoral	LB	0.04	0.13	0.45	0.11	**0.7**	0.06	0.05	0	0.03	**0.9**	0	0.14	0	0	0	0	**1.0**	0	0.23	**1.2**
			UB	0.04	0.13	0.45	0.11	**0.7**	0.06	0.05	0.01	0.03	**0.9**	0.01	0.14	0.01	0.01	0.01	0.01	**1.0**	0.08	0.23	**1.4**
	NIGERIA	Lagos	LB	0.32	1.19	1.51	0.37	**3.4**	0.15	0.13	0.02	0.10	**3.8**	0	0.28	0	0	0	0	**4.0**	0.32	1.51	**5.9**
			UB	0.32	1.19	1.51	0.37	**3.4**	0.15	0.13	0.02	0.10	**3.8**	0.01	0.28	0.01	0.01	0.01	0.01	**4.0**	0.32	1.51	**6.0**
		Kano	LB	0.24	0.92	1.64	0.42	**3.2**	0.24	0.17	0.01	0.15	**3.8**	0	0.47	0	0	0	0	**4.1**	0.44	1.88	**6.6**
			UB	0.24	0.92	1.64	0.42	**3.2**	0.24	0.17	0.01	0.15	**3.8**	0.01	0.47	0.01	0.01	0.01	0.01	**4.2**	0.44	1.88	**6.6**
Cassava dry	BENIN	Littoral	LB	0.09	0.12	0.13	0.08	**0.4**	0.03	0.04	0.01	0.08	**0.6**	0	0.03	0	0	0	0	**0.5**	0	0.08	**0.7**
			UB	0.09	0.12	0.13	0.08	**0.4**	0.03	0.04	0.01	0.08	**0.6**	0.02	0.03	0.01	0.01	0.01	0.01	**0.6**	0.02	0.08	**0.8**
		Borgou	LB	0	0.02	0.03	0.01	**0.1**	0	0.01	0	0.04	**0.1**	0	0.007	0	0	0	0	**0.1**	0	0.03	**0.1**
			UB	0.01	0.02	0.03	0.01	**0.1**	0.01	0.01	0.01	0.04	**0.1**	0.01	0.007	0.01	0.01	0.01	0.01	**0.2**	0.01	0.03	**0.2**
	CAMEROON	Duala	LB	0.06	0.13	0.14	0.07	**0.4**	0.03	0.05	0	0.04	**0.5**	0	0.05	0	0	0	0	**0.5**	0	0.22	**0.8**
			UB	0.06	0.13	0.14	0.07	**0.4**	0.03	0.05	0.02	0.04	**0.5**	0.02	0.05	0.01	0.01	0.02	0.02	**0.6**	0.02	0.22	**0.9**
	MALI	Bamako	LB	0.19	0.29	0.35	0.19	**1.0**	0.09	0.18	0	0.21	**1.5**	0	0.14	0	0	0	0	**1.4**	0	0.26	**1.9**
			UB	0.19	0.29	0.35	0.19	**1.0**	0.09	0.18	0.05	0.21	**1.6**	0.03	0.14	0.02	0.04	0.03	0.03	**1.6**	0.08	0.26	**2.2**
		Sikasso	LB	0.09	0.2	0.26	0.13	**0.7**	0.06	0.12	0	0	**0.9**	0	0.1	0	0	0	0	**1.0**	0	0.19	**1.2**
			UB	0.09	0.2	0.26	0.13	**0.7**	0.06	0.12	0.05	0.1	**1.0**	0.01	0.1	0.02	0.04	0.03	0.03	**1.1**	0.06	0.19	**1.5**
	NIGERIA	Lagos	LB	0	0.02	0.02	0.01	**0.1**	0	0	0	0	**0.1**	0	0.007	0	0	0	0	**0.1**	0	0.02	**0.1**
			UB	0.01	0.02	0.02	0.01	**0.1**	0.01	0.01	0.01	0.02	**0.1**	0.01	0.007	0.01	0.01	0.01	0.01	**0.1**	0.01	0.02	**0.2**

(*continued on next page*)

Table 2 (*continued*)

			PAH4	PAH8	PAH13	PAH15 + 1
Legend:	BaP	Benzo[a]pyrene	*	*	*	*
	BaA	Benz[a]anthracene	*	*	*	*
	CHR	Chrysene	*	*	*	*
	BbF	Benzo[b]fluoranthene	*	*	*	*
	BkF	Benzo[k]fluoranthene		*	*	*
	IP	Indeno[1,2,3,c-d]pyrene		*	*	*
	DbahA	Dibenz[a;h]anthracene		*	*	*
	BghiP	Benzo[g;h;i]perylene		*		*
	MCH	5-Methylchrysene			*	*
	BjF	Benzo[j]fluoranthene			*	*
	DbaIP	Dibenzo[a;l]pyrene			*	*
	DbaeP	Dibenzo[a;e]pyrene			*	*
	DbaiP	Dibenzo[a;i]pyrene			*	*
	DbahP	Dibenzo[a;h]pyrene			*	*
	BcF	Benzo[c]fluorene				*
	CPP	Cyclopenta[c;d]pyrene				*

comparable because, unlike paprika, African pepper does not undergo a smoking process.

3.5.1. Tubers

All the samples discussed in this paper were prepared as consumed. However, in the study's food classification, a distinction was made between (1) tubers having undergone a drying process (e.g. "*cassava dry*"): before preparation, including rehydration, and (2) tubers prepared from fresh tubers (without resorting to drying at any stage (e.g. "*yam fresh*").

No PAH was detected in *yam fresh* and *potato fresh* (LB = 0) as displayed in Table 2, which means that the UB concentration is 100% censored data. The boiling and/or pounding of yam and potato did not generate any PAH15 + 1 congener above the analytical limit.

By contrast, the concentrations of PAH15 + 1 were quantified in 6 *cassava dry* composites, with LB: 0.1–1.9; UB: 0.2–2.2 μg/kg (Table 2), and 1 *yam dry* composite, with LB: 0.3; UB: 0.4 μg/kg (Table 1). The drying of tuber cossets (cassava and yam) in direct contact with roadside pavements was frequently observed in the field. The drying process, car emissions and direct contact with asphalt, could be sources of the PAHs detected in dried tubers.

3.5.2. Other core foods

Broth cubes, concentrated and dehydrated milk, and sugar were also tested for PAHs. Because these core foods were considered "ready to eat," they were not processed. The benzo[a]pyrene (BaP) concentrations in none of these composites exceeded the limit of detection (LB = 0), whereas it did for other congeners.

Broth cubes had a PAH15 + 1 concentration with a mean LB: 0.5; UB: 1.0; max: 1.0 μg/kg (Table 1).

Milk powder and concentrated milk composites contained low PAH15 + 1 content (mean LB: 0.1; UB: 0.6; max UB: 0.9 μg/kg). The difference between the LB and UB concentrations was relatively high due to frequent non-detection of analytes (UB/LB ratio superior to 5).

Similarly, in refined sugar samples, benzo[a]anthracene was the only detected PAH15 + 1 congener, in 1/6 composites (Duala), resulting in a mean PAH15 + 1 concentration of LB: 0.01; UB: 0.6 μg/kg (UB/LB ratio of 60).

3.6. Contamination profile

The proportion of each of the 15 + 1 EU priority PAHs was calculated from the mean UB values of 660 purchased foodstuffs, pooled evenly into 55 composite samples (Fig. 1). The main contributors were chrysene (22.4%), cyclopenta[c,d]pyrene (17.6%), benzo[a]anthracene (16.4%), benzo[b]fluoranthene (9.9%) and benzo[a]pyrene (8.5%), benzo[j]fluoranthene (5.5%), indeno[1,2,3-cd]pyrene (4.6%), benzo [g,h,i]perylene (4.0%), and benzo[k]fluoranthene (3.9%).

Taking all the samples into consideration, we noted that the PAH4 represented on average 77% of the 13 genotoxic and carcinogenic PAHs (PAH13 group listed in Table 1).

The cyclopenta[c,d]pyrene (CPP) was, surprisingly, the second congener in terms of proportion, which was three-fold the proportion (5.4%) of the second French TDS samples (Veyrand et al., 2013). Richter et al. (2000) enriched PAH-free sand with pyrene and observed that heating treatment generated CPP, most likely due to the interaction of pyrene with sand silica. This is a possible explanation of the relatively high CPP content in our food samples, given that pyrene was also present in our samples, as shown in Table S2. The presence of sand in the food could result from a lack of hygiene before or during the smoking or drying processes. In other words, it is possible that a large proportion of the CPP of our samples originates from the interaction between sand dust and pyrene.

In addition, the comparison between the proportions of PAH congeners in smoked fish (Fig. 2) versus edible oils (Fig. 3) suggests that these core foods share very similar PAH4 profiles. Tobiszewski and

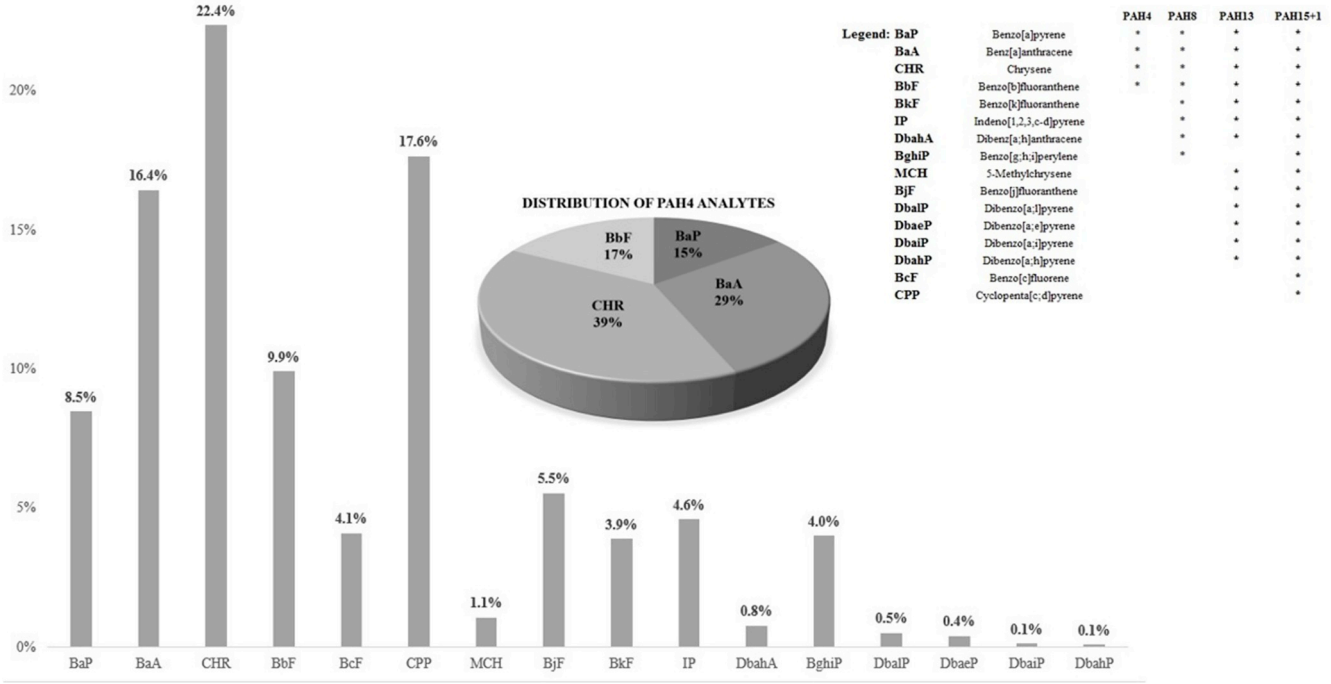

Fig. 1. Relative contribution of PAH congeners to PAH15 + 1.

Namieśnik (2012) discussed using the benzo[a]anthracene/(benzo[a]anthracene + chrysene) ratio as a means to identify the PAH emission source. Indeed, a ratio below 0.2 indicated that unburned petroleum was the main source of PAHs, whereas a ratio exceeding 0.2 indicated that the combustion of fuel or biomass (wood or grass) was the PAH source.

From the data of Veyrand et al. (2013), we determined that the benzo[a]anthracene/(benzo[a]anthracene + chrysene) ratio in composites from the French Total Diet Study was 0.30 in edible oils, and the presence of PAHs was interpreted as resulting from a heating process. In addition, a ratio of 0.20 was observed in molluscs, which indicates a petrogenic PAHs origin.

From the data of Mahugija and Njale, (2018a), we established that the average benzo[a]anthracene/(benzo[a]anthracene + chrysene) ratio of Tanzanian smoked fish samples was 0.38. In the SSA-TDS ratios were 0.44 (smoked fish), 0.42 (edible oils), LB: 0.45; UB: 0.63 (other core foods). These ratios confirm that a combustion process was the main source of PAHs in the SSA-TDS samples. The magnitude of carry-overs from feed to food of animal origin and from the soil to plants was previously assessed as being low to insignificant (Rey-Salgueiro, García-Falcón, Martínez-Carballo, González-Barreiro, & Simal-Gándara, 2008a, b), which is consistent with our finding.

Yebra-Pimental and colleagues (2012a, b) identified two feed groups, either contaminated via atmospheric or pyrolytic PAH sources, and reached similar conclusions using a cluster analysis.

Considering all the SSA-TDS samples, the determination coefficient obtained between the concentrations of BaP or PAH4 and PAH15 + 1 exceeded 0.99 (Fig. 4). The coefficient we obtained with data from 4

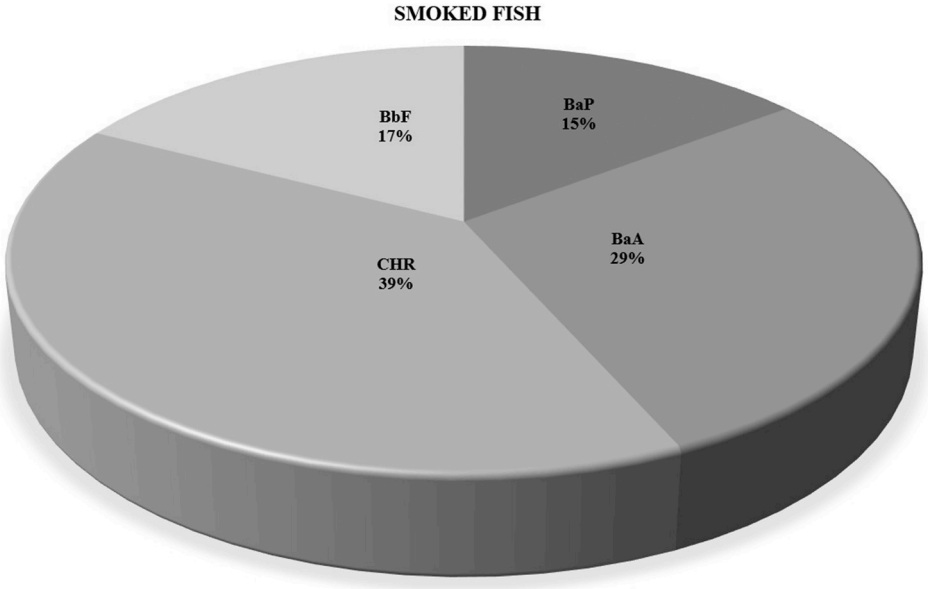

Fig. 2. Proportion of PAH4 congeners in smoked fish.

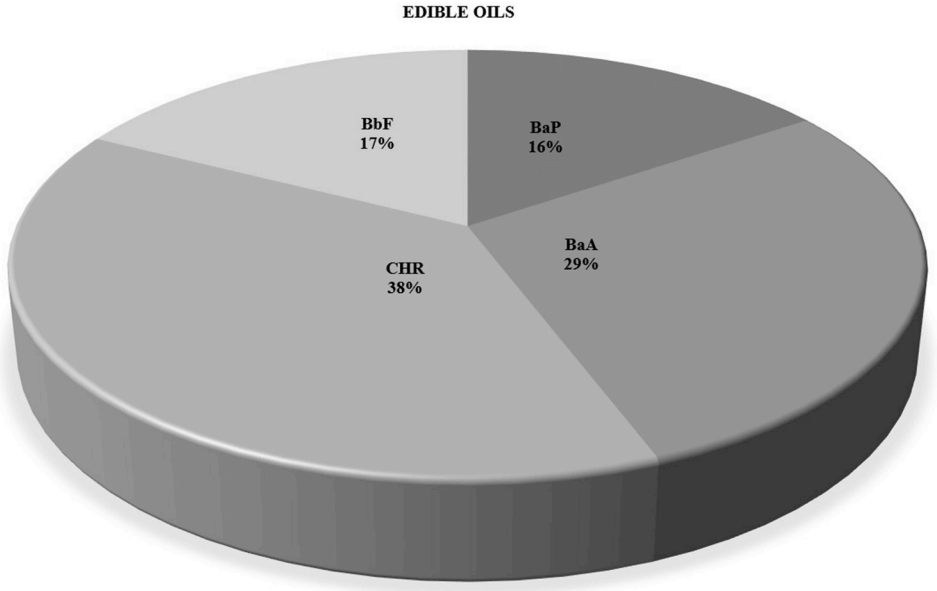

Fig. 3. Proportion of PAH4 congeners in edible oils.

countries was similar to the one reported by Veyrand et al. (2013) in the second French TDS (from 725 composite samples: $R^2 = 0.99$).

These results suggest that both BaP and PAH4 are pertinent markers of PAH15 + 1 contamination in the context of this TDS.

3.7. Concentrations versus regulation

The Codex Alimentarius has published a code of practice for the reduction of food contamination by PAHs resulting from smoking and direct drying processes (Codex Alimentarius, 2009). However, a limit for PAH concentrations in food stuffs has yet to be set. This is the reason why we compared previously observed PAH concentrations in this study with the standards set by Commission Regulation (EU) No 835/2011 (European Commission, 2011). This regulation was based on both the safety and the as-low-as-reasonably-achievable approach (ALARA). This regulation highlighted that PAH4 is a more suitable indicator of the total PAH contamination than BaP, and we therefore amended the limits accordingly.

Because we are dealing with pooled samples (composites systematically formed from 12 subsamples of equal weight) of foods prepared as consumed, in this study, we will not always be able to conclude with regard to the conformity of food commodities compared to the EU regulations, which applies to raw food commodities.

Table 1 shows that not only did all smoked fish composites exceed

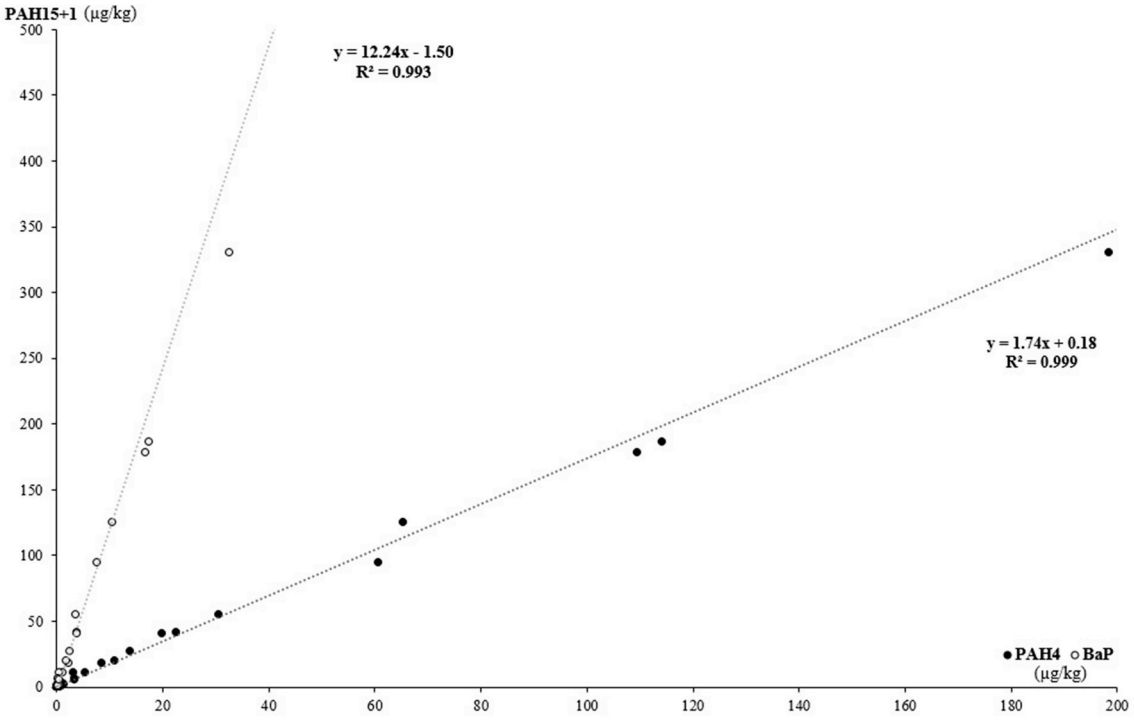

Fig. 4. Correlation of BaP and PAH4 with PAH15 + 1 concentration.

the EU maximum limit of 12.0 μg/kg fresh weight for PAH4 in muscle meat of smoked fish, but that the mean smoked fish concentration was 179.7 μg/kg PAH4, the highest mean concentration of all tested core foods.

The maximum PAH concentration of all our study samples occurred in the smoked fish composite collected in Borgou, which contained 39- and 47-fold the EU maximum limits for BaP and PAH4, respectively.

In spite of reducing PAH concentration by washing (Mahugija & Njale, 2018b), our TDS composites exceeded the EU regulation by 15-fold on average.

Other surveys were carried out in Africa. In Ghana, the EU regulation was exceeded 60-fold (Essumang, Dodoo, & Adjei, 2012), whereas in Tanzania, Mahugija and Njale (2018b) showed that washed smoked fish exceeded the EU regulation by an average of 158-fold.

Samples in Cambodia (Slámová, Fraňková, Hubáčková, & Banout, 2017) and Poland (Zachara, Gałkowska, & Juszczak, 2017) also exceeded the EU regulation for smoked fish by 2-50- and 6-fold, respectively. A survey in Iran showed that PAH4 in all smoked fish samples remained between 3 and 12 μg/kg (Mohammadi, Ghasemzadeh-Mohammadi, Haratian, Khaksar, & Chaichi, 2013), which conforms with EU regulations, as was the case in studies carried out in France (Veyrand et al., 2013) and Spain (Martorell et al., 2012).

The mean PAH4 contamination of the 4 palm oil samples in this study was 11.3 μg/kg, which exceeds the EU regulation (10.0 μg/kg).

Excluding the industrial bleached palm oil sample from Duala (0.2 μg/kg BaP; 0.8 μg/kg PAH4), the three red crude palm oil composites exceeded the EU regulation for PAH4 in oil (3/3). Three palm nut composite samples contained lower PAH4 concentrations (mean 2.7; max: 5.3 μg/kg) than the palm oil samples.

The peanut oil composite from Benin (Table 2) contained 0.4 μg/kg BaP and 3.1 of PAH4, and therefore complied with the EU regulations of 2.0 and 10.0 μg/kg, respectively. The peanut oil composite from Kano, however, exceeded the EU tolerance for BaP at 2.1 μg/kg, while remaining below the EU maximum limit in oils, with 8.4 μg/kg PAH4, meaning that it does not conform to regulations.

While the BaP and PAH4 concentrations in cottonseed oil composites were all tested and found to be 6–18-fold below the EU regulations (Table 2), with a range of 0.1–0.3 μg/kg BaP and 0.7–1.2 μg/kg for PAH4, the composites from subsamples collected in Mali exceeded 4-fold (BaP) and 6-fold (PAH4) the EU regulation applicable to oils, with concentrations of 7.6 and 60.6 μg/kg, respectively.

The occurrences of PAH4 in edible oil were lower in other studies carried out in Spain (Martorell et al., 2010) and in France (Veyrand et al., 2013), whereas the mean PAH4 concentrations were 1.9 and 1.96 μg/kg, respectively.

This difference may be explained by the different oil types (including olive oil, sunflower, and rapeseed oil, which were not included in the core food list of this TDS), different extraction processes (including heating versus cold press and refining processes), as well as different culinary practices (including heated versus non-heated oils). The complexity of factors influencing these levels currently limits our interpretation.

Activated carbon, as well as wood ash, may contribute to the reduction of PAH concentrations in oils (Yebra-Pimentel, Fernández-González, Martínez-Carballo, & Simal-Gándara, 2014), in addition to the refining process, including neutralization, bleaching, and deodorization (Rojo Camargo, Ramos Antoniolli, & Vicente, 2012).

4. Conclusion

There is currently no information regarding the dietary intake of PAHs by populations in any African country (Domingo & Nadal, 2015). The purpose of this component of the SSA-TDS was to begin to fill this gap in the field, beginning with the study of foods in Benin, Cameroon, Mali, and Nigeria.

4.1. Our main observations are as follows

- PAH concentrations exceeded EU regulations in smoked fish (100% of composite samples) and edible oils (50%).
- The profile of PAH4 congeners suggests that the PAH contamination mainly originates from food processing (*smoking, heating, drying, and possibly fuel combustion*).
- High variations in mean concentrations were not only observed between study centres of different countries but also between study centres from the same country (Benin and Cameroon).

Although the exposure data presented here will be of great help to risk managers from Benin, Cameroon, Mali, and Nigeria, by providing guidance for setting priorities, on the basis of comparisons with the EU regulation, we constructed the following specific recommendations for risk managers and their technical and financial partners:

1. Review local food processing practices (for *smoked fish, edible oils, and dried tubers*) and implement, where necessary, codes of practices (for *smoked fish and dried tubers*) as recommended by the Codex Alimentarius Commission.
2. Strengthen local analytical laboratory capacities in order to monitor the conformity with respect to PAH4 congeners.
3. Implement monitoring and surveillance plans with regard to PAH4 concentrations in smoked fish and edible oils as a priority.

Moreover, more TDS in other locations in Benin, Cameroon, Mali, and Nigeria, as well as in other countries in Sub-Saharan Africa, will need to be carried out in order to better document the risks resulting from dietary exposure to PAHs in this region.

Conflicts of interest

The authors declare that there is no conflict of interest.

Disclaimer

The views expressed in this publication are those of the authors and do not necessarily reflect the views and policies of the Food and Agriculture Organization of the United Nations.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.foodcont.2019.04.006.

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