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## Levels of persistent organic pollutants (POPs) in foods from the first regional Sub-Saharan Africa Total Diet Study

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### ABSTRACT

For the first time, a multi-centre Total Diet Study was carried out in Benin, Cameroon, Mali and Nigeria. We collected and prepared as consumed 528 typical fatty foods from those areas and pooled these subsamples into 44 composites samples. These core foods were tested for a wide spectrum of POPs, including polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), brominated flame-retardants (BFRs), organochlorine compounds (OCs), perfluoro alkyl substances (PFAS) and chlorinated flame retardants (CFRs).

The POPs contamination levels were similar or lower than those reported in total diet studies previously conducted worldwide. In most cases, core foods belonging to fish food group presented higher POPs concentrations than the other food groups. Interestingly, we observed a difference in both contamination profile and concentration for smoked fish compared to non-smoked fish. Such finding suggests that the smoking process itself might account for a large proportion of the contamination. Further investigation would require the assessment of combustion materials used to smoke fish as a potential vehicle, which may contribute to the dietary exposure of the studied populations to POPs.

### 1. Introduction

Food is not only a source of nutrients as it also contains various other classes of chemicals, including persistent organic pollutants (POPs) (Camel et al., 2018). The environmental monitoring related to this class of substances, listed in the Stockholm Convention, is relevant

considering their widespread dissemination, long-term transport, long half-life, bioaccumulation and related toxicological impact in living organisms (UNEP, 2001; Amiard et al., 2016). The dietary exposure to POPs such as polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), brominated flame-retardants (BFRs), organochlorine compounds (OCs),

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perfluoro alkyl substances (PFAS) and chlorinated flame retardants (CFRs) may result in adverse toxicological effects to human health (WHO, 2003; Van den Berg et al., 2006; EFSA et al., 2018a, 2018b; Bruce-Vanderpuije et al., 2019).

Assessing the dietary exposure of a given population to chemicals such as POPs is based on various strategies. (FAO/WHO, 1985; WHO, 2009). An alternative to relying on data from food control systems is the use of the Total Diet Study (TDS) approach. These studies are based on a standardized method as recently recommended by WHO, FAO and EFSA: steps characterising a TDS include the selection of foods based on food consumption data to represent as best as possible a typical diet, their preparation to food as consumed and the subsequent pooling of related foods before analysis. (WHO, 2011; EFSA, 2011a, 2011b). Such cross-sectional surveys therefore enable to estimate the chronic exposure to chemical compounds through food consumption (Lee et al., 2015). Besides assessing dietary exposure to beneficial substances, TDS are also considered relevant public health tool to determine population dietary exposure to harmful chemicals across the entire diet.

Numerous TDS have already been conducted worldwide since the 1960s, including recent and major ones in Europe (Chekri et al., 2019), the United States of America (Hoffman-Pennesi et al., 2015), Canada (Juric et al., 2018), China (Gao et al., 2016) or South Korea (Shin et al., 2015). With regard to the African continent, a TDS was carried out in Yaounde, Cameroon between 2006 and 2010. This study scope included some pesticides (Gimou et al., 2008), metals and trace elements (Gimou et al., 2014), while POPs were not considered in this case. Some studies report POPs levels in African countries mainly in environmental matrices such as air, water, soil and sediments (Bogdal et al., 2013; Gioia et al., 2014) with specific focus available for Ghana (Bruce-Vanderpuije et al., 2019) and South Africa (Nieuwoudt et al., 2009; Verhaert et al., 2017; Govaerts et al., 2018). Most of these studies of environmental samples indicated that the levels of many POPs were increasing in Africa, instead of declining or remaining stable as could be expected after the ban of many of these chemicals worldwide and observed in other parts of the world (Adu-Kumi et al., 2012; Bogdal et al., 2013; Gioia et al., 2014). Consequently, several studies have been conducted in human to objectify and characterize internal exposure. Thus human biofluids such as serum (Linderholm et al., 2010; Luzardo et al., 2014; Bruce-Vanderpuije et al., 2019), urine (Bruce-Vanderpuije et al., 2019) or human milk (Kinyamu et al., 1998; Darnerud et al., 2011) have been analysed with regard to POPs contamination in a range of African populations (sub-Saharan and South Africa mainly). Associated results clearly demonstrate increasing Human exposure to POPs and the next question refers to the associated determinants. For the majority of populations that are not occupationally exposed to POPs, the main contamination route is known to be through dietary intake of food from animal origin. With regard to available literature, the lack of data in food is striking, as very few contamination data are reported. A recent systematic review focusing on Ghana compiled PCDD/Fs, PCBs, OCPs, PBDEs and HBCDs levels in fish, meat, dairy products, cereals, fruits and honey, and concluded at the same time to both large data gaps and high risks for the populations (Bruce-Vanderpuije et al., 2019). The present study is positioned in a context of urgent need to generate occurrence data in food in a public health perspective. The Sub-Saharan Africa Total Diet Study (SSA-TDS) is a multi-centre project aiming at investigating an extended list of food chemicals, within a large study population (Ingenbleek et al., 2019a, 2019b, 2019c; Jitaru et al., 2019). The SSA-TDS design and main methodological choices implemented by FAO in Benin, Cameroon, Mali and Nigeria between 2014 and 2018 have previously been described elsewhere (Ingenbleek et al., 2017). In this framework, we conducted the analysis of PCDD/Fs, PCBs, BFRs, CFRs, OCs, and PFAs from representative samples collected in eight African study centres and prepared as consumed. This article presents POPs occurrence data generated by the SSA-TDS.

## 2. Materials and methods

### 2.1. Sample collection and preparation

Two study centres within each country were selected, namely the Littoral of Benin and Borgou, Duala and North Cameroon, Bamako and Sikasso for Mali, Kano and Lagos in the case of Nigeria (Ingenbleek et al., 2017). In total, 528 subsamples of fatty foods (eggs, fish, meat, milk/dairy products, miscellaneous, nuts/seeds, oil/fat) were collected from Benin, Cameroon, Mali and Nigeria during the rainy season in October 2017. Briefly, the subsamples were collected, prepared and cooked as per the typical consumer behaviour. They were prepared individually, according to local recipe books (Vinakpon-Gbaguidi, 2003; Nya-Njike, 1998; Gautier and Mallet, 2006; Madubike, 2013) chosen for their representativeness of the study populations' diets. Recipes allowed for the identification of the processes used in the preparation of the foods, especially in terms of cooking time and temperature. Even if POPs are present at low level in tap water and condiments, as the exception of PFAS (Noorlander et al., 2011), distilled water was used instead of tap water and no condiments were added to avoid, as much as possible, external or cross contamination, considered a limitation of the present SSA-TDS. Subsamples from different food subgroups were not mixed to allow for the identification of the contamination source. Inedible parts were removed at the preparation stage, as a typical consumer would do. Then the 528 sampled foods were pooled in 12 subsamples of equal weight, of the same core food and from the same centre, to form 44 composite samples.

### 2.2. Analytical methods executive summaries

The choice of analyte groups resulted from a consultation among national stakeholders from Benin, Cameroon, Mali and Nigeria, and was discussed and agreed within a dedicated scientific committee, without applying the methodology proposed by the EU TDS Exposure project (Papadopoulos et al., 2015).

All analyses including PCDD/Fs, PCBs, BFRs, OCs, PFAs and PAHs analyses with the exception of CFRs, were carried out according to validated and accredited methods (ISO/IEC 17025:2005 standard). The full description of the analytical methods is available in the [supplementary information \(Table A1, SI\)](#).

#### 2.2.1. PCDD/Fs, PCBs and BFRs methods

17 PCDD/F congeners, 12 DL-PCB congeners, 6 NDL-PCB congeners, 8 PBDE congeners, 1 PBB and three HBCD isomers have been monitored in this SSA-TDS. To minimise environmental contamination during the extraction and purification steps, analyses were carried out in an overpressurized room and all clean laboratory glassware was rinsed with dichloromethane prior to use (Bichon et al., 2014). The extraction procedure implemented in the present study has been fully described elsewhere (Marchand et al., 2015) (Table A1, SI).

Briefly, food samples were freeze-dried before grinding. Appropriate labelled internal standards were added before automated solvent extraction under high temperature and high pressure. For PCDD/Fs, DL and NDL-PCBs, PBDEs and PBB, the obtained fat residual was then purified through three successive purification columns. The final purified extracts were then analysed by gas chromatography (7890A; Agilent Technologies, USA) coupled to a high-resolution mass spectrometer, double sector (JMS-700D and 800D; Jeol, Japan) operating at a resolution of 10,000 (10% valley) (Antignac et al., 2006; Cariou et al., 2010; Rivière et al., 2014). For HBCDs, only one purification column was needed and further liquid/liquid purification step was applied. The analysis of HBCDs was performed by liquid chromatography coupled to tandem mass spectrometry with negative electrospray as ionisation technique (LC-ESI(-)-MS/MS) (6410, Agilent Technologies, Santa Clara, CA, USA).

### 2.2.2. OCs method

The concentration of 9 organochlorine pesticides was characterized in the 44 food composite samples (for full protocol description see [Table A1, SI](#)). First, a 10 g of sample was mixed with 25 g of sodium sulphate and 25 g of Fontainebleau sand in a mortar in order to obtain dry and brittle product. This mixture was then poured in a glass column and eluted with an adapted solvent mixture in order to selectively isolate the fatty fraction. After adding the appropriate internal standards an aliquot of the obtained fat was then purified by two successive cryogenic centrifugations at  $-20^{\circ}\text{C}$  with solvent mixture. Both extracts were combined and evaporated to dryness. Two successive Solid Phase Extraction (SPE) purification steps were then carried out on the cryogenic extract. Organochlorines were separated by capillary gas chromatography (Trace GC Ultra Thermo Scientific) equipped with a programmed temperature vaporizer (PTV) injector and coupled to a Quantum XLS Triple Quadrupole (GC-MS/MS). The mass spectrometer was operated in electron ionization (EI) mode.

### 2.2.3. PFAS method

The analytical method was developed to determine the concentration of 5 perfluoroalkyl sulfonates and 9 perfluorocarboxylic acids ([Rivière et al., 2014](#)) ([Table A1, SI](#)). Solid food samples were freeze-dried, supplemented by twelve  $^{13}\text{C}$ -labelled quantification standards and extracted with an adapted solvent. After evaporation, food extracts were purified onto two consecutive SPE columns. Final purified extracts were analysed by LC-ESI(-)-MS/MS. Two transitions at least were monitored per analyte (except for PFBA and PFPA). Quantification was performed according to isotope dilution principles ([Table A1, SI](#)).

### 2.2.4. CFRs method

Trace analysis of 6 Dechlorane Related Compounds (DRCs), a class of CFRs, was performed according to previously published work ([Abdel Malak et al., 2018, 2019](#)). Briefly, lyophilized samples were extracted by Pressurized Liquid Extraction, except for oil samples. Purification of the extracts involved a multilayer silica gel column (acidic, neutral and basic layers) followed by Gel Permeation Chromatography. Purified extracts were analysed by gas chromatography (6890, HP, Palo Alto, CA, USA) coupled to high resolution mass spectrometry (JMS 700D, Jeol, Tokyo, Japan), operating at a resolution of 10,000 (10% valley), in the electron ionisation mode and in a single sequence. Identification relied on two diagnostic ions and quantification was performed through isotopic dilution using appropriate labelled internal standards ([Table A1, SI](#)).

### 2.2.5. QA/QC and reporting of results

To ensure the quality of the analysis, besides the use of appropriate internal standards in each sample, labelled external standards were systematically added at the end of each analytical process in order to determine recoveries.

Further, a continuous monitoring of the analytical procedure was implemented through procedural blanks. For BFRs and CFRs which are ubiquitous contaminants, blank concentration was systematically subtracted from the individual sample result to ensure that the reported contamination values arise from the sample itself. For PCDD/Fs, PCBs, PFAS and OCs, as analytical contamination is fully under control, i.e. lower than the concentration levels observed in the samples and regularly monitored through control chart, blank concentration was not deducted for these class of POPs.

Reproducibility was assessed using quality control samples (QC) regularly characterised over several years. QCs were as follows: a fish oil sample naturally contaminated with PCDD/Fs, PCBs, PBDEs and HCBDDs and possibly fortified with CFRs, and a fish sample naturally contaminated with PFAS.

The accuracy of most analytical methods is further ensured by regular participation of the laboratory to proficiency tests, such as those organized by the European Reference Laboratory (EURL) for POPs.

The measured concentrations of PCDD/Fs, DL-PCBs, NDL-PCBs, BFRs, CFRs, OCs and PFAS congeners in collected samples are expressed on a wet weight (ww) basis. The concept of “Upper-Bound (UB)” and “Lower-Bound (LB)” was used to report the results. ([EC, 2017](#)). UB and LB values have been calculated for all quantified parameters. A careful examination of the data with regard to guidelines provided by WHO and EFSA for the evaluation of low-level contaminations in food, conducted to the selection of UB values for further data analysis. The limit of quantification (LOQ) was set as the concentration corresponding to a signal to noise exceeding 3 and was calculated for each molecule, in each tested food sample. By using external standard, recoveries were determined for each class of POPs.

## 3. Results and discussion

Comparing mean contamination levels from one study to another requires caution, as food groups do not necessarily contain the same food items ([Sirot et al., 2012](#)). Moreover, in a TDS, samples are analyzed as consumed, i.e. cooked, whereas this is not the case in mere occurrence surveys, which are often based on the sampling of raw food commodities ([Windal et al., 2010](#); [Marin et al., 2011](#); [Mezzetta et al., 2011](#)). Besides, although composite samples have been considered in the present study, their limited number would require additional investigations in order to refine some hypothesis and conclusions.

### 3.1. PCDD/Fs, PCBs

First, results associated to PCC/Fs and PCBs are presented and discussed in a global and descriptive way. The 44 composite samples were tested for the 17 PCDD/Fs, the 12 DL-PCBs and the 6 NDL-PCBs. The UB concentrations of individual congeners, as WHO-TEQ values for PCDD/Fs, DL-PCBs and sum of PCDD/Fs & DL-PCBs, as well as the sum of the mass concentrations of NDL-PCBs in each food item are presented in [Table A2](#) (see [SI](#)). Concentrations are reported in pg/g ww for all the congeners and pg TEQ/g ww for the TEQ values. The maximum and minimum concentrations and the percentage of samples exceeding the LOQ is reported for each analyte. Recoveries associated to each compound were all between 50% and 120%. Overall, the concentration levels quantified in all samples were very low and the UB levels remained below the European MRLs ([EC, 2011](#)). Minimum values detected attest for the sensitivity of the applied analytical strategies while providing confident occurrence data related to chronic exposure. The results showed for each congener different proportions of left-censored data (percentage of samples in which the congener is not detected). For instance, while some contaminants have rarely been detected (e.g. 2,3,4,8-TCDD, 1,2,3,4,7,8,9-HpCDF, PCB-81, PCB-189 which presented a quantification rate under 40% (36%, 18%, 39% and 27% respectively) some others such as 5 NDL-PCBs (PCB-28, PCB-52, PCB-101, PCB-138 and PCB-153), PCB-77, PCB-118 and OCDD could be quantified in all investigated samples.

Then, in order to compare concentration levels between the different food groups, [Table 1](#) summarizes the mean concentrations expressed as WHO-TEQ values for PCDD/Fs, DL-PCBs and total PCDD/Fs DL-PCBs, and as the sum of NDL-PCBs concentrations by food group as discussed hereafter. Further, mean congeners detection rate (17 PCDD/Fs + 12 DL-PCBs + 6 NDL-PCBs) is presented by food group. This values which was observed higher for food groups fish, eggs, dairy products and meat (92%, 81%, 78%, 77% respectively) in comparison to nuts/seeds and oil/fat (54% for both), highlights most contaminated food groups of interest.

Fish samples ( $n = 9$  composite samples) presented higher concentration levels compared to the other food group matrices. Indeed, the mean UB concentrations values for WHO-TEQ-PCDD/F-PCB and the sum of the mass concentrations of NDL-PCBs were 0.278 pg/g ww, and 852 pg/g ww respectively, far above those determined for the eggs food group ( $n = 4$  composites), 0.046 pg/g ww and 64 pg/g ww, resp.,

**Table 1**

PCDD/Fs-WHO-TEQ (2005), DL-PCBs-WHO-TEQ (2005), PCDD/Fs + DL-PCBs-WHO-TEQ (2005) sum of NDL-PCBs mean concentrations and the associated mean quantification congeners rate calculated by food group. Results are expressed on a wet weight basis.

| Food group    | N  | Lipids (%) | Upperbound concentrations (pg/g ww) |                      |                       |       |                  |                      |  |                      | Mean of detection rate (%) <sup>*</sup> |
|---------------|----|------------|-------------------------------------|----------------------|-----------------------|-------|------------------|----------------------|--|----------------------|---|
|               |    |            | WHO-TEQ (2005) PCDD/F               |                      | WHO-TEQ (2005) PCB DL |       | TOTAL-TEQ (2005) |                      | Sum of 6 NDL-PCBs                          |                      |   |
|               |    |            | SSA-TDS                             | EU-MRL <sup>**</sup> | SSA-TDS               |       | SSA-TDS          | EU-MRL <sup>**</sup> | SSA-TDS                                    | EU-MRL <sup>**</sup> |   |
| EGGS          | 4  | 8.8        | 0.034                               | 2.5                  | 0.012                 | 0.046 | 5                | 64                   | 40.10 <sup>3</sup>                         | 81                   |   |
| FISH          | 9  | 6.9        | 0.161                               | 3.5                  | 0.117                 | 0.278 | 6.5              | 852                  | 75.10 <sup>3</sup> and 125.10 <sup>3</sup> | 92                   |   |
| MEAT          | 7  | 5.7        | 0.021                               | 2.5                  | 0.010                 | 0.030 | 4.0              | 44                   | 40.10 <sup>3</sup>                         | 77                   |   |
| MILK/DAIRY    | 7  | 11.2       | 0.032                               | 2.5                  | 0.017                 | 0.048 | 5.5              | 83                   | 40.10 <sup>3</sup>                         | 78                   |   |
| MISCELLANEOUS | 2  | 52.9       | 0.043                               | 0.75                 | 0.006                 | 0.050 | 1.25             | 29                   | 40.10 <sup>3</sup>                         | 63                   |   |
| NUTS/SEEDS    | 4  | 35.7       | 0.050                               | –                    | 0.012                 | 0.062 | –                | 39                   | –  | 54                   |   |
| OIL/FAT       | 11 | 98.2       | 0.102                               | 0.75                 | 0.021                 | 0.123 | 1.25             | 23                   | 40.10 <sup>3</sup>                         | 54                   |   |

\* Based on 17 PCDD/Fs + 12 DL-PCBs + 6 NDL-PCBs.

\*\* (EC, 2011).

and the meat food group (n = 7 composites), 0.030 pg/g ww, and 44 pg/g ww, resp. Similar observations were previously described in other European TDS conducted in Finland (Kiviranta et al., 2004), in Spain (Bocio and Domingo, 2005) and more recently in France (Sirot et al., 2012). For instance, in the second French Total Diet Study (TDS2) (Sirot et al., 2012), the highest levels reported for WHO-TEQ-PCDD/F-PCB and the sum of NDL-PCBs were also observed in the fish group (n = 46) (0.54 pg/g ww and 5263 pg/g ww respectively), whereas lower values for egg group (n = 30; 0.027 pg/g ww and 88 pg/g ww, resp.) and meat group (n = 80; 0.047 pg/g ww and 235 pg/g ww, resp.) were determined.

Moreover, in comparison to the second French TDS (Sirot et al., 2012), mean contamination levels for the fish food group in the present SSA-TDS were observed as lower for PCBs (factor of 2 for WHO-TEQ-PCBs and factor of 5 for NDL-PCBs) and higher for WHO-TEQ-PCDD/Fs. (SSA-TDS: n = 6; 0.161 pg/g ww vs TDS2: n = 66; 0.088 pg/g ww).

Deeper investigating the fish food group enabled assessing any potential relationship between their contamination profiles and, on the one hand, their origin (sea fish vs fresh water fish) or, on the second hand, their applied process (i.e. non-smoked vs smoked). The concentrations measured in the SSA-TDS fish composite samples (3 non-smoked and 6 smoked samples (from both sea and fresh water)) are reported in Table 2.

When comparing sea fish and fresh water fish contaminations in non-smoked samples, similar very low DL-PCB levels could be observed. Although obtained on limited sample set, such results allow drawing the hypothesis that no particular PCB contamination exist in those area to the contrary of what has already been described in France for instance in sea (Abarnou, 2008) or in river (Santiago et al., 1994). The

fact that the countries involved in the SSA-TDS are less industrialized could explain a lower environmental PCB contamination in Sub-Saharan rivers, which would be reflected by fish contamination levels.

When focusing on the smoking process, it appeared that the WHO-TEQ-PCDD/Fs mean values in smoked fish samples were three times higher than in non-smoked fish. Such phenomenon was not observed in the TDS2 (Sirot et al., 2012). Neither the difference of lipid content between the two groups (0.6 factor), nor the concentration factor resulting from the drying process undergone by smoked-fish could explain this observation. Possibly, the combustion material used during the smoking process could explain the observed contamination levels. Nonetheless, the concentration levels in the maximalist UB hypothesis that we determined remained below the current European Maximum Limits (EC, 2011).

With regard to the influence of smoking process on the contamination profile, PCDD/Fs (17 congeners), DL-PCBs (12 congeners) and NDL-PCBs (6 congeners) SSA-TDS mean patterns for non-smoked and smoked sample fish were established and compared to those from the TDS2 (Sirot et al., 2012) (Figure A1 a, b, c, SI). For DL-PCBs and NDL-PCBs, sample profiles were observed as comparable whether between smoked and non-smoked fish and also between the SSA and the TDS2. They highlighted the predominance in the contamination profiles of PCB-153, 138 and 180 for NDL-PCBs and PCB-118, 105, 156 and 167 for DL-PCBs. The establishment of PCDD/Fs profiles illustrated no difference between smoked and non-smoked fish samples which was also the case between the 2 TDS. 2,3,7,8-TCDF and OCDD were more predominant in SSA-TDS samples whereas 1,2,3,6,7,8-HCDD, 1,2,3,4,6,7,8-HpCDD and OCDD significantly contributed in the TDS2 samples. A previous study in Ghana (Bruce-Vanderpuije et al., 2019)

**Table 2**

PCDD/Fs-WHO-TEQ (2005), DL-PCBs-WHO-TEQ (2005), PCDD/Fs + DL-PCBs-WHO-TEQ (2005) sum of NDL-PCBs mean concentrations and their associated means for sea fish group and smoked fish group. Results are expressed on a wet weight basis.

| Core food        | Country  | Centre                | Lipids (%) | Upperbound concentrations (pg/g ww) |                       |                  |                   |
|------------------|----------|-----------------------|------------|-------------------------------------|-----------------------|------------------|-------------------|
|                  |          |                       |            | WHO-TEQ (2005) PCDD/F               | WHO-TEQ (2005) PCB DL | TOTAL-TEQ (2005) | Sum of 6 NDL-PCBs |
| Sea fish         | Cameroon | Douala                | 6.4        | 0.03                                | 0.08                  | 0.12             | 1116              |
| Sea fish         | Nigeria  | Lagos                 | 5.8        | 0.15                                | 0.27                  | 0.42             | 1571              |
| Fresh water fish | Cameroon | North                 | 2.5        | 0.01                                | 0.01                  | 0.02             | 51                |
|                  |          | Non smoked fish means | 4.9        | 0.07                                | 0.12                  | 0.19             | 912               |
| Smoked fish      | Cameroon | Douala                | 2.2        | 0.13                                | 0.06                  | 0.19             | 718               |
| Smoked fish      | Cameroon | North                 | 7.5        | 0.04                                | 0.02                  | 0.07             | 151               |
| Smoked fish      | Mali     | Bamako                | 8.9        | 0.48                                | 0.14                  | 0.63             | 1001              |
| Smoked fish      | Mali     | Sikasso               | 7.2        | 0.31                                | 0.08                  | 0.39             | 571               |
| Smoked fish      | Benin    | Borgou                | 14.0       | 0.26                                | 0.30                  | 0.56             | 1852              |
| Smoked Fish      | Benin    | Littoral              | 7.5        | 0.02                                | 0.08                  | 0.11             | 633               |
|                  |          | Smoked fish means     | 7.9        | 0.21                                | 0.12                  | 0.32             | 821               |

also determined PCB levels in different aquatic organisms in agreement with of the present SSA-TDS fish sample contamination levels and patterns.

With regard to the other food groups such as egg and meat, which presented lower contamination levels compared to fish, mean contamination levels were in the range of previously reported levels (Sirost et al., 2012).

Further, considering the geographical parameter, no significantly different congener profiles could be observed in any of the four countries and the determined concentrations within the same food group were in the same order of magnitude.

### 3.2. BFRs

Eight PBDES, three HBCDDs and one PBB congeners have been quantified in the 44 composite samples. The UB concentrations of individual congeners, of the sum of the markers PBDEs with (n = 8) or without (n = 7) BDE-209, and the sum of the 3 HBCDDs stereoisomers are presented in Table A4 (SI). Concentrations are reported in pg/g ww for all congeners. The maximum and minimum concentrations and the percentage of samples exceeding the LOQ were calculated for each analyte. Recoveries of all monitored BFRs were considered as acceptable (ranging from 50% to 120%).

- HBCDD. Low quantification rates, i.e. 25%, 0% and 16%, were obtained for  $\alpha$ ,  $\beta$ ,  $\gamma$ -HBCDD, respectively. Overall, very low concentration levels for HBCDDs (close to LOQs) were observed whatever the food groups and countries considered. Quantification could be performed in few samples only and the highest mean concentrations of the sum of the three HBCDD congeners ( $\alpha$ ,  $\beta$  and  $\gamma$ ) were observed in 2 palm oil samples from Benin (170 and 161 pg/g ww). The mean UB concentrations of all other food groups were lower than 25 pg/g ww. These results are comparable or slightly lower than those observed in previous similar studies in Europe (Kiviranta et al., 2004; Rivière et al., 2014). For instance, in the French study (Rivière et al., 2014), mean concentration levels for the sum of 3 HBCDDs associated to the milk, meat and fish food groups were 3 pg/g ww, 126 pg/g ww and 141 pg/g ww respectively.
- With regard to PBB 153 it could only be quantified in one composite sample of milk from Bamako, in which the concentration level (0.26 pg/g ww) was very close to the LOQs.
- The results associated to PBDEs showed high quantification rates for BDE-99, 100, 153 and 47 (95%, 89%, 84% and 82% respectively). BDE-209 could indeed be quantified in all composite samples involved in the study. As sampling plans differ from one country to another, the comparison of average contamination levels by country (Benin, Cameroon, Mali and Nigeria) does not allow however relevant conclusions to be drawn. Mean sum of the 7 PBDEs by country were determined in the range 0.91 to 57 pg/g ww. Probably due to a specific sampling (oil/fat and miscellaneous samples), Nigeria presented the lowest contamination mean value.

Table 3 summarizes the average concentrations of the individual congeners, the average associated blank in percentage, the average of sum of the seven and the sum of the eight PBDE congeners by food group and the mean congeners detection rate (8 PBDEs) per food group. While procedural blank contamination represents a significant part of contamination for BDE-28, 47, 99, 100 and 209, it could be considered as negligible for BDE-153, 154 and 183. Such observation was fully expected regarding the very low observed concentration levels, close to LOQs. The higher the concentration, the more the blank contribution is small or insignificant. As observed above for PCDD/Fs and PCBs, the mean value of the congeners detection rate was higher for food groups fish, eggs, meat and dairy products (94%, 91%, 80%, 70% respectively) than it was in the case of nuts/seeds and miscellaneous (50% and 38%).

**Table 3** PBDE congeners individual mean concentrations and their associated blank contribution (%), sum of marker's PBDEs with or without BDE-209 and associated detection congeners rate interval per food group. Results are expressed on a wet weight basis.

| Food Group    | N  | Upperbound concentrations (pg/g ww) |           |        |           |        |           |         |           |         |           |         |           |         |           |         |           | Sum of 7 indicator PBDEs | Sum of 8 indicator PBDEs | Mean of quantification rate (%) * |
|---------------|----|-------------------------------------|-----------|--------|-----------|--------|-----------|---------|-----------|---------|-----------|---------|-----------|---------|-----------|---------|-----------|--------------------------|--------------------------|-----------------------------------|
|               |    | BDE-28                              |           | BDE-47 |           | BDE-99 |           | BDE-100 |           | BDE-153 |           | BDE-154 |           | BDE-183 |           | BDE-209 |           |                          |                          |                                   |
|               |    | Sample                              | Blank (%) | Sample | Blank (%) | Sample | Blank (%) | Sample  | Blank (%) | Sample  | Blank (%) | Sample  | Blank (%) | Sample  | Blank (%) | Sample  | Blank (%) |                          |                          |                                   |
| EGGS          | 4  | 0.01                                | 30        | 1.13   | 29        | 1.21   | 10        | 0.47    | 2         | 1.17    | 0         | 0.38    | 0         | 2.53    | 0         | 1.47    | 8         | 6.9                      | 153                      | 91                                |
| FISH          | 9  | 1.99                                | 14        | 47.4   | 5         | 16.0   | 5         | 12.6    | 0         | 5.10    | 0         | 8.02    | 0         | 1.19    | 0         | 114     | 28        | 92                       | 206                      | 94                                |
| MEAT          | 7  | 0.02                                | 40        | 1.18   | 29        | 0.52   | 20        | 0.14    | 14        | 0.24    | 0         | 0.15    | 0         | 0.14    | 0         | 29      | 33        | 2.4                      | 31                       | 80                                |
| MILK/DAIRY    | 7  | 0.08                                | 28        | 3.93   | 38        | 1.84   | 26        | 0.51    | 13        | 0.62    | 0         | 0.24    | 0         | 0.19    | 0         | 26      | 34        | 7.4                      | 34                       | 70                                |
| MISCELLANEOUS | 2  | 0.003                               | 68        | 0.03   | 82        | 0.04   | 59        | 0.01    | 51        | 0.02    | 0         | 0.02    | 0         | 0.03    | 0         | 20      | 30        | 0.16                     | 21                       | 38                                |
| NUTS/SEEDS    | 4  | 0.05                                | 38        | 1.91   | 70        | 1.74   | 42        | 0.37    | 33        | 1.02    | 0         | 0.45    | 0         | 0.23    | 0         | 93      | 39        | 5.8                      | 99                       | 50                                |
| OIL/FAT       | 11 | 0.05                                | 31        | 0.38   | 80        | 0.57   | 45        | 0.26    | 15        | 0.83    | 0         | 0.33    | 0         | 0.37    | 0         | 72      | 43        | 2.8                      | 75                       | 60                                |

\* Based on 8 PBDEs.

The mean concentration levels for the sum of 7 PBDEs were comparable between eggs, meat, nuts/seeds and oil/fat groups (6.9 pg/g ww, 2.4 pg/g ww, 5.8 pg/g ww and 3.2 pg/g ww respectively). Besides, the sum of the 7 PBDE congeners in food groups eggs, meat and milk/dairy food groups, sampled in Cameroon presented the highest concentrations (16.8 pg/g ww, 6.1 pg/g ww and 31.5 pg/g ww respectively). This might be explained by a more extensive utilization of BFRs in Cameroon or by more important recycling activities leading to the emission of this kind of compounds, compared with the other countries, although we failed to gather additional information to support this hypothesis. As described in literature (Rivière et al., 2014), fish food group presented here also the highest mean sum of 7 PBDEs (92 pg/g ww), and the maximum concentration values were determined in 3 smoked fish samples, 2 from Mali (247 pg/g ww and 215 pg/g ww) and one from Benin (215 pg/g ww). These particular 3 samples also contained the highest PCDD/Fs concentration levels among all tested samples.

Overall, the PBDEs contamination levels, expressed as the sum of 7 PBDEs (excluding BDE-209), were observed as lower than those reported in European studies (Kiviranta et al., 2004; Rivière et al., 2014). For instance, eggs, fish, meat, milk/dairy products and oil/fat from the present SSA-TDS food groups exhibit lower mean concentration values (6.9 pg/g ww, 92 pg/g ww, 2.4 pg/g ww, 7.4 pg/g ww and 3.2 pg/g ww, resp.) compared to their counterparts in the French TDS2 (18 pg/g ww, 496 pg/g ww, 26 pg/g ww, 11 pg/g ww and 30 pg/g ww, resp.).

As observed for PCDD/Fs, the mean concentration of PBDEs in smoked fish composite samples was 5 times higher than it was in non-smoked sea fish samples. The smoking process could also be a source of PBDEs. Combustion material might be inappropriate and may certainly contain halogenated compounds such as BFRs. The specific combustion processes used in the areas we investigated could explain the differences of PBDEs contamination patterns observed between this study and others studies (Rivière et al., 2014).

As carried out for PCDD/Fs and PCBs, SSA-TDS mean PBDEs pattern for non-smoked and smoked sample fish were established and compared to those from the second French TDS (Rivière et al., 2014) (Figure A1-d, SI). PBDE profiles were observed as comparable between smoked and non-smoked fish from the SSA-TDS. The comparison of contamination patterns of the fish food group SSA-TDS with the French TDS2 presented differences on BDE 209 and BDE 47 respective contributions, 57%-21% and 7%-60%, resp. (Rivière et al., 2014). The occurrence of BDE 47 was also previously reported in other European studies of smoked products (Cruz et al., 2018).

### 3.3. OCs

Unlike the other POPs, organochlorine pesticides were not only screened in fatty matrices, but were also tested in other food composites as described elsewhere (Ingenbleek et al., 2019c). dealing with an additional set of pesticides monitored in the SSA-TDS Eight OCs listed in the Stockholm Convention (aldrin, hexachlorocyclohexane, chlordane, dieldrin, endrin, heptachlor, lindane, and DDT) were measured with an analytical method characterized by a LOD of 3 µg/kg ww and a LOQ of 10 µg/kg ww. None of these organochlorine pesticides was detected in any tested composite sample. Endosulfan was separately screened (LOD of 0.3 µg/kg ww and LOQ of 1.0 µg/kg ww). This OC was detected at four occasions; traces were found between LOD and LOQ in citrus and in cottonseed oil from Duala and in smoked fish from Bamako. One sample (cassava from the Littoral of Benin) was measured above the LOQ (1.5 µg/kg ww).

### 3.4. PFAS

The individual PFAS (n = 14) concentrations of the 44 composite samples are presented in Table 4.

Recoveries associated to each sample were determined and ranged from 30 to 80% (depending on food items). PFOS was the most

frequently detected PFAS, with values above LOQs in 25% of the samples. Long-chain perfluorocarboxylic acids were detected at rates of 18% (PFNA and PFUnA) and 14% (PFDA and PFDoA). The maximum concentration of PFOS has been observed in a smoked fish composite sample from Mali with a level of 10.4 µg/kg ww. The other levels measured were 10-times lower with concentrations of PFOS ranging from 0.02 µg/kg ww to 0.92 µg/kg ww. Long-chain perfluorocarboxylic acids concentration levels ranged from 0.01 to 0.89 µg/kg ww.

Fish samples again were observed as the most contaminated food group, with detection rate of 89% for PFOS and PFUnA, and 67% for PFNA, PFDA and PFDoA, as detailed in Table 5. PFPA was quantified in one composite sample from Cotonou, Benin, at a level of 2.60 µg/kg. Further, the comparison of levels of contamination between locations highlighted higher PFAS concentrations in the two study centers of Mali (Bamako and Sikasso). In smoked fish from Mali indeed, PFOS and long-chain perfluorocarboxylic acids were thus quantified at more than 10 times higher concentrations compared with the 3 other countries. It is unclear if the contamination of the fish collected in both Bamako and Sikasso results from a similar environmental contamination level, or if the fish of both sites had the same origin. An hypothesis to such contamination level could be emitted in relation with high pesticides levels quantified in those smoked fish samples as it has recently been acknowledged pesticides as a major source of PFOS contamination (Liu et al., 2017). Sulfluramid is the predominant PFOS/PFOA related pesticide and its main active ingredient is N-ethyl perfluorooctane sulfonamide (Et-FOSA) (Löfstedt Gilljam et al., 2016), which would ultimately transform to PFOS and PFOA through photolysis, oxidation, and biotransformation (Tomy et al., 2004; Martin et al., 2006; Plumlee et al., 2009).

Detection rate in beef samples was close to 0% (Table 6). Nevertheless, PFOS has been detected in two beef composite samples from the same country (Nigeria). Presence of traces of PFNA at 0.03 µg/kg ww reinforces the hypothesis of a specific contamination depending on the sampling site. Observed concentrations were low and consistently below 0.11 µg/kg. The detection of perfluoroalkylated substances in others food items was very rare. Nevertheless, traces of PFHpA (0.48 µg/kg ww) and PFOA (0.13 µg/kg ww) could be determined in food group nuts/seeds, in composite samples from Douala and from Sikasso respectively.

The mean UB PFAS concentrations were comparable to those reported in European TDSs. As an example, the mean concentration in foods from the French market were slightly lower in the case of fish samples (UB concentrations of 0.05 µg/kg fw vs. 0.08 µg/kg ww in this study for PFDoA, and 0.03 µg/kg ww vs. 0.22 µg/kg fw in this study for PFDA). In Spain (Domingo et al., 2012), concentrations in fish were higher for PFOS (UB concentrations of 2.7 µg/kg fw vs. 1.36 µg/kg ww in this study) and PFUnA (0.43 µg/kg fw vs. 0.15 µg/kg ww in this study) but lower for PFDoA (0.06 µg/kg fw vs. 0.08 µg/kg ww in this study).

### 3.5. CFRs

The concentration values for the 6 individual DRCs and their sums, expressed as lowerbound and upperbound values, are presented in Tables A8 and A9 (SI). No differences were observed between countries.

In terms of quantification rate (Table A8), one DRC or more was quantified in 43 out of the 44 samples, confirming DRCs are ubiquitous compounds. Dec-602 was the most frequently quantified DRC (95%), followed by *anti*- and *syn*-DP (91%). Yet, DP stereoisomers suffered procedural contamination involving higher limits of quantification. Other DRCs were less frequently detected (< 27%). Dec-601 was not identified in any of the investigated composite samples.

Total DRC concentrations, ranged up to 52 pg/g ww (UB, broth/bouillon cube from Lagos, Nigeria), with a mean value of about 17 pg/g ww (UB), which remains relatively low. DP isomers contributed in average to 61% of these sum in both scenarios (UB and LB), followed by

**Table 4**  
Occurrence and concentration of perfluoroalkylated substances ( $\mu\text{g}/\text{kg}$  wet weight) in the composite samples.

|                   | PFBS | PFHxS | PFHpS | PFOS  | PFDS | PFBA | PFPA | PFHxA | PFHpA | PFOA | PFNA | PFDA | PFUnA | PFDaA |
|-------------------|------|-------|-------|-------|------|------|------|-------|-------|------|------|------|-------|-------|
| N                 | 44   | 44    | 44    | 44    | 44   | 44   | 44   | 44    | 44    | 44   | 44   | 44   | 44    | 44    |
| n > LOQ           | 0    | 0     | 0     | 11    | 0    | 0    | 2    | 0     | 1     | 1    | 8    | 6    | 8     | 6     |
| % > LOQ           | 0%   | 0%    | 0%    | 25%   | 0%   | 0%   | 5%   | 0%    | 2%    | 2%   | 18%  | 14%  | 18%   | 14%   |
| Max concentration | 0    | 0     | 0     | 10.44 | 0    | 0    | 2.6  | 0     | 0.48  | 0.13 | 0.09 | 0.89 | 0.54  | 0.34  |
| Mean conc LB *    | 0    | 0     | 0     | 0.28  | 0    | 0    | 0.06 | 0     | 0.01  | 0    | 0.01 | 0.04 | 0.03  | 0.02  |
| Mean conc UB *    | 0.01 | 0.02  | 0.04  | 0.3   | 0.05 | 1.09 | 0.38 | 0.12  | 0.08  | 0.03 | 0.02 | 0.05 | 0.04  | 0.03  |

**Table 5**  
Concentrations of perfluoroalkylated substances (expressed in  $\mu\text{g}/\text{kg}$  of wet weight) in fish samples from different sites.

| Country  | Town    | Food Subgroup    | PFBS   | PFHxS  | PFHpS  | PFOS   | PFDS   | PFBA   | PFPA   | PFHxA  | PFHpA  | PFOA   | PFNA   | PFDA   | PFUnA  | PFDaA  |
|----------|---------|------------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| Cameroon | Douala  | Sea fish         | < 0.01 | < 0.01 | < 0.01 | 0.13   | < 0.01 | < 0.65 | 0.31   | < 0.07 | < 0.07 | < 0.01 | < 0.01 | < 0.01 | 0.02   | < 0.01 |
| Cameroon | Douala  | Smoked fish      | < 0.01 | < 0.01 | < 0.03 | < 0.02 | < 0.01 | < 0.61 | < 0.01 | < 0.07 | < 0.06 | < 0.01 | 0.03   | < 0.01 | 0.02   | < 0.01 |
| Cameroon | Garoua  | Fresh water fish | < 0.01 | < 0.01 | < 0.03 | 0.07   | < 0.02 | < 0.52 | < 0.01 | < 0.06 | < 0.05 | < 0.01 | 0.02   | 0.04   | 0.05   | 0.02   |
| Cameroon | Garoua  | Smoked fish      | < 0.01 | < 0.01 | < 0.01 | 0.12   | < 0.02 | < 0.82 | < 0.02 | < 0.09 | < 0.08 | < 0.01 | 0.03   | 0.06   | 0.09   | 0.03   |
| Mali     | Bamako  | Smoked fish      | < 0.01 | < 0.01 | < 0.02 | 0.92   | < 0.07 | < 0.53 | < 0.60 | < 0.40 | < 0.06 | < 0.08 | 0.09   | 0.89   | 0.54   | 0.34   |
| Mali     | Sikasso | Smoked fish      | < 0.01 | < 0.07 | < 0.01 | 10.44  | < 0.04 | < 0.22 | < 0.09 | < 0.34 | < 0.04 | < 0.04 | 0.08   | 0.83   | 0.47   | 0.23   |
| Benin    | Borgou  | Smoked fish      | < 0.01 | < 0.01 | < 0.01 | 0.39   | < 0.11 | < 0.51 | < 0.85 | < 0.14 | < 0.04 | < 0.05 | < 0.04 | 0.09   | 0.11   | 0.07   |
| Benin    | Cotonou | Smoked fish      | < 0.01 | < 0.01 | < 0.01 | 0.04   | < 0.04 | < 0.12 | 2.6    | < 0.08 | < 0.01 | < 0.02 | < 0.01 | < 0.01 | < 0.03 | < 0.01 |
| Nigeria  | Lagos   | Sea fish         | < 0.01 | < 0.01 | < 0.01 | 0.12   | < 0.01 | < 0.75 | < 0.20 | < 0.04 | < 0.01 | < 0.01 | 0.03   | 0.02   | 0.04   | 0.01   |

Dec-602 (19–34%, LB-UB). The blank contribution associated to the sum of 6 DRCs was between 19% and 71% among the food groups. As expected and previously mentioned for PBDEs, when determined concentrations were low or close to LOQs, the blank contribution was logically more important. *Anti*- and *syn*-DP represented more than 50% of the blank contribution whereas CP, Dec-601 and Dec-603 could not be most of the time quantified.

Two broth/bouillon cubes from Nigeria, were the most contaminated samples (Table 7, Table A8). These miscellaneous food samples contained about 7–11% of fat (99% dry matter). DP stereoisomers accounted for most of the contamination profiles. Unfortunately, broth/bouillon cubes were not sampled in the other countries so that it was not possible to investigate whether the geographical and/or the food nature/process could explain such levels.

The second most contaminated food group was fish, with an average total DRC concentrations of  $22 \text{ pg g}^{-1} \text{ ww}$  (UB). Interestingly, Dec-602 was predominant in DRC contamination profiles of fish (53–63% in average, depending on the considered scenario). Kim et al. (2014) reported values for  $N = 36$  fish samples within their study related to foodstuffs from South Korea. Interestingly, in the LB scenario the predominant congeners were DP isomers ( $36.34 \text{ pg g}^{-1} \text{ ww}$ , LB) followed by Dec-602 ( $3.99 \text{ pg g}^{-1} \text{ ww}$ , LB), highlighting regional differences. In our study, Dec-602 and CP were about 1.5 more concentrated in smoked ( $N = 6$ ) compared to non-smoked ( $N = 3$ ) fishes. This difference could only partly be explained by the lipid content, which varied according to the same proportion. Nonetheless, because DP stereoisomers were 4 to 7 times more concentrated in smoked fish than in non-smoked fish, depending on the lowerbound and upperbound scenario, our result suggest that the smoking process could be a DRC contamination source.

**Table 6**  
Concentrations of perfluoroalkylated substances (expressed in  $\mu\text{g}/\text{kg}$  of wet weight) in beef samples from different sites.

| Country  | Town    | Food Subgroup | PFBS   | PFHxS  | PFHpS  | PFOS   | PFDS   | PFBA   | PFPA   | PFHxA  | PFHpA  | PFOA   | PFNA   | PFDA   | PFUnA  | PFDaA  |
|----------|---------|---------------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|--------|
| Cameroon | Douala  | Beef          | < 0.01 | < 0.01 | < 0.05 | < 0.01 | < 0.02 | < 0.69 | < 0.01 | < 0.08 | < 0.07 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 |
| Cameroon | Garoua  | Beef          | < 0.01 | < 0.01 | < 0.04 | < 0.02 | < 0.02 | < 0.57 | < 0.01 | < 0.06 | < 0.06 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 |
| Mali     | Bamako  | Beef          | < 0.01 | < 0.01 | < 0.02 | < 0.05 | < 0.05 | < 0.06 | < 0.06 | < 0.03 | < 0.02 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 |
| Benin    | Borgou  | Beef          | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.03 | < 0.01 | < 0.03 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 |
| Benin    | Cotonou | Beef          | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.03 | < 0.06 | < 0.05 | < 0.06 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 | < 0.01 |
| Nigeria  | Kano    | Beef          | < 0.01 | < 0.01 | < 0.01 | 0.1    | < 0.04 | < 1.00 | < 0.07 | < 0.02 | < 0.01 | < 0.02 | < 0.02 | < 0.02 | < 0.01 | < 0.02 |
| Nigeria  | Lagos   | Beef          | < 0.01 | < 0.02 | < 0.01 | 0.11   | < 0.03 | < 0.91 | < 0.20 | < 0.07 | < 0.01 | < 0.02 | 0.03   | < 0.02 | < 0.01 | < 0.01 |

**Table 7**  
Sum of 6 DRC concentrations by food group and associated blank contribution (%). Results are expressed on a wet weight basis.

| Food Group    | N  | Sum of 6 DRCs ( $\text{pg}/\text{g}$ ww) |                 |                        |
|---------------|----|--|-----------------|------------------------|
|               |    | Lowerbound (LB)                          | Upperbound (UB) | Blank contribution (%) |
| EGGS          | 4  | 6.3                                      | 7.5             | 21                     |
| FISH          | 9  | 19.3                                     | 22.0            | 19                     |
| MEAT          | 7  | 10.2                                     | 12.0            | 21                     |
| MILK/DAIRY    | 7  | 1.0                                      | 3.3             | 71                     |
| MISCELLANEOUS | 2  | 29.9                                     | 37.5            | 28                     |
| NUTS/SEEDS    | 4  | 9.6                                      | 13.9            | 29                     |
| OIL/FAT       | 11 | 10.5                                     | 23.9            | 49                     |

Vegetable oils ( $N = 11$ ), containing almost 100% lipids, contained DRCs at levels similar to those previously reported in South Korea ( $N = 5$ , Kim et al., 2014), Japan ( $N = 5$ , Yasutake et al., 2018), Lebanon ( $N = 7$ , Abdel Malak et al., 2019) and Belgium ( $N = 2$ , L'Homme et al., 2015).

#### 4. Conclusion

While several studies have demonstrated over the last past 5 years an increased POPs level in African environmental samples conversely to the worldwide observed trends, the scientific community hypothesised such phenomenon to be linked with the recent and rapid transformation undergone by this part of the world with regard to the development of information communication technology (ICT) and the bulk import of



second-hand electrical and electrical devices from developed countries to support associated demands (Luzardo et al., 2014). In parallel, an illegal trade of associated wastes, known as e-waste, consisting in disassembling and burning such equipments (e.g. transformers) is reported to significantly contribute to PCBs, dioxins and PAHs environmental contamination. In this context, characterising the exposure of humans through food produced in such areas is imperative. However, studies that characterized the occurrence of POPs in African foods are seldom (Govaerts et al., 2018; Bruce-Vanderpuije et al., 2019) and concluded to gaps in occurrence data. The purpose of our article was to provide the first occurrence data concerning foods commonly eaten by some African populations in Benin, Cameroon, Mali, and Nigeria to a number of food chemicals, including some POPs, namely PCDD/Fs, PCBs, OCs, BFRs, PFAS and CFRs.

The POPs contamination levels quantified in the present SSA-TDS are equivalent or lower than those reported in previous international TDS (Kiviranta et al., 2004; Sirot et al., 2012; Rivière et al., 2014; Shin et al., 2015).

As expected, the highest POPs concentrations were determined in fish samples.

Moreover, we highlighted the smoking process as a possible contamination source of fish by some POPs, PCDD/Fs, PBDEs and PAHs (Ingenbleek et al., 2019b). The drying process, usually associated with hot fish smoking processes, that concentrates the dry matter including the bi-accumulated contaminants, does not suffice to explain the increase in contamination levels encountered in smoked fish samples compared with non-smoked fish. Our hypothesis is that the combustion material used in fish smoking processes may account for a large part of the POPs levels quantified in smoked fish.

If subsequent studies confirm our observation, two obvious consequences arise from this finding:

- Smoked fish, within the typical diet may be a significant contributor, in terms of proportion, to the dietary exposure to the POPs of our study populations.
- Improving the fish smoking process is a potential risk management option, to lower the dietary exposure to POPs of the study populations.

We would like to put in perspective the results of this SSA-TDS component dedicated to the study of POPs in typical African foods to mention that we also highlighted the contamination of smoked fish with:

1. Toxic fungi and bacterial secondary metabolites, namely aflatoxins and cereulide (Ingenbleek et al., 2019a)
2. 13 genotoxic and carcinogenic polycyclic aromatic hydrocarbons (Ingenbleek et al., 2019b)
3. Various concentrations of pesticides, including neurotoxic chlorpyrifos (Ingenbleek et al., 2019c). Such results also enabled drawing an hypothesis about pesticides as a source of PFOS contamination.

Therefore, studying the processes and practices, and in particular the combustion material used to produce smoked fish, the best practices that allow for the reduction of POPs in smoked fish could be included in the next updates of current Codes of Practice that are apply to fish (Codex Alimentarius, 2003, 2009).

#### CRedit authorship contribution statement

**Vincent Vaccher:** Formal analysis, Writing - original draft, Writing - review & editing. **Luc Ingenbleek:** Project administration, Conceptualization, Investigation, Methodology, Writing - original draft, Writing - review & editing. **Abimobola Adegboye:** Investigation. **Sétondji Epiphane Hossou:** Investigation. **Abdoulaye Zié Koné:** Investigation. **Awoyinka Dada Oyedele:** Investigation. **Chabi Sika**

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#### 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 and of the World Health Organization.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### Appendix A. Supplementary material

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