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Par
Luc INGENBLEEK

Exposition par voie alimentaire de populations du Bénin, Cameroun, Mali et Nigéria à un large panel de substances chimiques

« Santé Publique »

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Rapporteurs avant soutenance :

Marie-Louise Scippo, Professeur Université de Liège
Jean-Pierre Cravedi, Directeur de Recherches INRA
Jean-Lou Dorne, Scientifique Senior EFSA

Composition du Jury :

Marie-Louise Scippo, Professeur Université de Liège
Président
Jean-Pierre Cravedi, Directeur de Recherches INRA,
Rapporteur
Jean-Lou Dorne, Scientifique Senior EFSA
Rapporteur
Bruno Le Bizec, Professeur Oniris, Directeur Laberca
Directeur de thèse
Sara Eyangoh, Directeur Scientifique Centre Pasteur
Co-encadrant de thèse
Jean-Charles Leblanc, Consultant Senior FAO
Co-encadrant de thèse

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INTRODUCTION

The Codex Alimentarius defines food safety as the assurance that food will not cause harm to the consumer when it is prepared and/or eaten according to its intended use (Codex Alimentarius, 2009a). The increasing awareness of consumers and public health institutions of the importance of eating safe food is compelling. Food chemicals can naturally occur or result from human activity. While residues may be present in foods because of an intentional use of food chemicals, contaminants are not intentionally added to foods. Although relatively recent worldwide industrial development may have accelerated or worsened adverse health consequences by increasing exposure to some residues and contaminants, the issue is far from new.

Due to the extensive use of lead pipes in the water distribution systems of declining Imperial Rome, the lead concentration in the drinking water is believed to have exceeded 100 times that of the local spring water (Delile et al., 2014) and is thought to have caused saturnism (Seaton, 2014), a severely debilitating brain affection. Besides lead, 200 chemicals may cause neurodevelopmental disorders such as autism, attention deficit disorder, mental retardation, and cerebral palsy (Granjean and Landrigan, 2006). These neurotoxins include arsenic, lead, methylmercury, and polychlorinated biphenyls, as well as chlorpyrifos (an organophosphate pesticide) and polybrominated diphenyl ethers (Granjean and Landrigan, 2015). Chemical food safety is therefore essential.

In order to better protect populations against adverse chemical effects, it is critical to assess the risks prior to addressing them. This is the purpose of risk analysis, a framework supported by international multilateral institutions, including the World Health Organization (WHO) and the Food and Agriculture Organization (FAO) of the United Nations. Risk assessment is the evidence-based component of risk analysis, which involves hazard identification, hazard characterization, the derivation of health-based guidance values (HBGVs), exposure assessment, and, finally, risk characterization.

The purpose of a total diet study (TDS) is to assess the dietary exposure of a population. It contributes to the risk assessment process and generates useful information for guiding risk mitigation measures. According to the WHO, FAO, and European Food Safety Authority (EFSA), a TDS consists of selecting, collecting, and analysing commonly consumed food purchased at a retail level, processing the food as for consumption, pooling the prepared food

items into representative food groups, homogenising the pooled samples, and analysing them for harmful and/or beneficial chemical substances. TDSs are designed to cover the whole diet and to measure the amount of each chemical substance of interest ingested by the population living in a country over their lifetime using low-level, average, and high-level consumption data, as appropriate for the substances being assessed (chronic dietary exposure). Exposure through drinking water and the water used in cooking should be included in the TDS assessment. Chronic dietary exposure calculations assist in determining whether specific food chemical substances pose a risk to health. The EFSA, FAO, and WHO document entitled 'Towards a harmonized Total Diet Study approach' was the first attempt to reach an agreement on the methodology (EFSA, 2014b).

National food safety authorities worldwide unanimously endorse the TDS approach. Many of them have carried out TDSs, including those in the USA, the UK, Canada, Australia, New Zealand, and France. Unfortunately, TDSs are challenging, both technically and financially, which explains why many countries have not yet been able to implement their own TDSs. In Africa, the lack of data relating to dietary exposure to food chemicals may be a consequence of national priorities focussing on food security, in terms of availability, to the detriment of food safety. Nonetheless, in 1996 the World Food Summit newly defined food security as '*access to sufficient, safe, and nutritious food which meets their dietary needs and food preferences for an active and healthy life*' (Berry et al., 2015). Food safety is therefore, by definition, a component of food security.

The first TDS ever carried out in Sub-Saharan Africa was conducted in Yaoundé, Cameroon, from 2006 and included pesticides (Gimou et al., 2008) as well as heavy metals and trace elements (Gimou et al., 2014). It received a preparation grant from the Standard and Trade Development Facility (STDF). The World Trade Organization hosts the STDF Secretariat, the governance of which also includes the World Bank, the WHO, the FAO of the United Nations, and the World Organization for Animal Health.

Lessons learned from the Yaoundé TDS experience emphasized the need for additional knowledge concerning the occurrence of food chemicals, dietary exposure, and food safety risks. In particular, the analytical limits of pesticide residues turned out to be a limiting factor, because the non-detects led to relatively high exposure uncertainty due to the large differences between the minimalist, lower-bound and maximalist, upper-bound hypotheses (Gimou et al., 2008).

The STDF agreed to fund a wider TDS involving four African countries: Benin, Cameroon, Mali, and Nigeria. I was involved in the project on behalf of the regional coordinator (of the Centre Pasteur of Cameroon) with technical supervision from an FAO Lead Technical Officer. This explains, to a certain extent, the convergence in methodology and objectives between that project and my doctoral training.

However, in the present document, which focuses on the public health component of the TDS, I do not substantially investigate any trade-related aspects, except where I briefly approach the question of conformity with current standards, which connects with trade issues. Risk managers need access to chemical food safety information in order to document the adequacy of the protection offered by the current Codex and/or national food standards in an African context. In light of typical exposure levels and the occurrence of residues and contaminants in core foods, which are the predominant contributors to exposure, it is possible to identify new standards for enhanced consumer protection.

In this thesis, I wanted to address the question

'*How safe is Africa's food*?',

as far as a selection of food chemicals and a limited number of African study centres are concerned. More specifically, in the context of the limited resources available to national food safety authorities and their partners, I wanted to contribute to answering the following question:

'*To what extent is the TDS approach a pertinent tool to identify and prioritize food chemical safety concerns in developing countries?*'

By pertinent, we mean (1) cost effective and (2) sufficiently accurate to identify specific exposure patterns and risks associated with the typical diet consumed by the studied population.

Objectives of the Sub-Saharan Africa Total Diet Study

In an attempt to answer the above-mentioned concerns, this thesis describes and discusses:

- Food consumption data
- Food contamination data

- Dietary exposure from the consumption and contamination data
- Risks from exposure to a selected range of food chemicals
- Communication of the risk assessment results to risk managers

In the first chapter of this thesis, I briefly describe the risk assessment process, as defined by international institutions, namely the WHO and FAO, as a component of the Codex Alimentarius risk analysis framework. I also present and discuss a selection of national TDSs. In the second chapter, dealing with the methodology, the key principles which governed the implementation of the Sub-Saharan Africa TDS (SSA-TDS) are explained. It includes the generation of food contamination data and the sampling approach.

In the third chapter, I present the results according to four lines of enquiry: The first subset of results is the food consumption data corresponding to each of the study populations, which we used for the completion of the dietary exposure assessment. We published the food consumption data in *Food and Chemical Toxicology*, along with the methodology. The second subset is the food chemical occurrence data. We generated five papers from the occurrence data during this thesis. We published the first occurrence data article in *Toxins*, dealing with the concentrations of mycotoxins, as well as other fungal, bacterial, and plant secondary metabolites. The second paper, which we submitted to *Food Control*, deals with the occurrence of polycyclic aromatic hydrocarbons in African foods. The third occurrence article, published in *Food Chemistry*, presents the occurrence of pesticides while the fourth paper, presenting the occurrence of Al, As, Cd, Hg, and Pb, was published in *Environment International*. The final occurrence paper deals with persistent organic pollutants (POPs) was published in *Environment International* too.

The third subset of results involves characterizing levels of risk through comparisons between the dietary exposure of study populations to a selection of food chemicals and the available HBGVs. Finally, to go beyond the simple assessment of risk and embrace the necessity of communicating our results to risk managers, we intend to submit an article to *The Lancet Global Health* with a focus on 24 food chemicals of safety concern, according to our data. The results of this study will guide risk managers, improving the monitoring, control, and surveillance of food chemical contamination to better protect consumers in Africa and beyond. We then discuss various limitations and perspectives, including the need for complementary studies.

‘*Science knows no country, because knowledge belongs to humanity, and is the torch which illuminates the world*’

Louis Pasteur (27 December 1822 - 28 September 1895)

1 CONTEXT

Introduction

The context in which we designed the SSA-TDS is explained in this chapter. First, the risk analysis framework, as understood by the WHO and FAO, is briefly described in this section, in order to explain to what extent TDSs may contribute to the risk assessment process. Once the exposure is known and the risks characterized, risk managers can utilize their resources efficiently to implement adequate mitigation measures. Risk analysis also offers the opportunity to rank the identified risks and to set evidence-based priorities to better protect consumer health. The risk analysis framework, as defined by the Codex Alimentarius, is shown in Figure 1. The risk analysis concept consists of three interrelated elements: the risk assessment, the science-based component; the risk management, the policy-based component; and the risk communication (WHO, 2009) that encompasses all aspects of the risk analysis. TDSs belong to the risk assessment process, although the data generated by TDSs are beneficial to guide risk management.

Figure 1: The risk analysis framework (FAO, 2006a)

Second, we describe some of the chemical hazards we assessed in this thesis. The emphasis is on several regulated mycotoxins, toxic trace elements, and polycyclic aromatic hydrocarbons and one pesticide. Finally, some of the TDSs which have been carried out worldwide over the last five decades are briefly described to inform discussion on the benefits of various approaches.

1.1. Risk Assessment

The risk assessment component of risk analysis comprises four subcomponents:

- Hazard identification
- Hazard characterization
- Exposure Assessment
- Risk characterization

1.1.1 Hazard identification and characterization

1.1.1.1 Toxicological profile of chemicals

An essential component of risk assessment is the gathering of available data concerning the chemical of interest. These data include the target organ, exposure route, exposure frequency, and exposure duration (Printemps and Rousselle, 2018). It is also useful to consider physicochemical properties such as molecular weight, solubility, lipophilicity, and degradation products, among others. For example, the half-lives of POPs are a very important aspect of their chronic toxicological profile.

1.1.1.2 Toxicological approaches

The forms of toxicity that are commonly investigated are as follows:

Acute toxicity

The unique ingestion of a single high dose of a chemical, acute toxicity mimics accidental exposure (Maes et al., 2016). Acute toxicity studies look at the exposure scenarios that happen in relatively rare events involving, for example, poisoning or suicide by ingestion of a chemical.

Non-acute toxicity

The effects of repeated dietary exposure can be studied over different durations:

- Sub-acute (2-4 weeks)
- Sub-chronic (3 months)
- Chronic (6-24 months)

Genotoxicity

Mutations can be induced by chemicals that form adducts with DNA and can alter its structure. This phenomenon can be studied *in vivo* or *in vitro* (Turkez et al., 2017). Genic and chromosomal alterations are likely to induce carcinogenesis and reproductive toxicity.

Carcinogenicity

Studying the carcinogenicity of genotoxic chemicals owing to their DNA interaction properties is particularly important as they pose a cancer risk even at very low doses. Non-genotoxic carcinogenic chemicals are thought to have a safe exposure threshold (Nohmi, 2018).

Reproductive toxicity

Reproductive toxicity includes adverse effects on fertility (Jeelani et al., 2016) and development, and teratogenicity (Zhang et al., 2016).

1.1.1.3 Hazard potency

Epidemiological studies (e.g., cohort or case-control studies) based on human data are highly preferable to assess food chemical potency, although these types of data are not available for many chemicals. The principle is to assess the dose-responses of chemicals in the human body. Several ethical concerns prevent the intentional administration of significant doses of certain chemicals to humans. A common substitute for human data which are more frequently used is observations obtained from animals. They can provide useful information on the toxicity of chemicals. Various doses of a given chemical are administered orally or injected into the animal and the potential toxicological effects are assessed.

Cell cultures can also be used to assess *in vitro* toxicity, as a substitute for *in vivo* human or animal toxicological data (Sambuy, 2005; Zeilinger et al., 2016). *In vitro* systems may be 2D or 3D (Godoy et al., 2013, Duval et al., 2017). The chemical structure of a substance can predict biological activity, to a certain extent, as shown in quantitative structure-activity relationship studies (Roy et al., 2015). Structural analogies between substances can be used (read-across) to

identify potential toxicity mechanisms, although (Alves et al., 2018) additional data are necessary to confirm or reject the predicted activity. Metabolomics can also contribute, when these data are available. Such information can help to assess the impact of chemicals using their metabolite profiles, in a broad sense (Chen et al., 2016; Zaitsu et al., 2016).

1.1.1.4 Health-based guidance values

HBGVs vary depending on the nature of the chemical. The Joint FAO/WHO Expert Committee on Food Additives (JECFA) developed the concept of a threshold of risk in 1956 (FAO, 2006b). The Joint FAO/WHO Meeting on Pesticide Residues (JMPR) uses the same principles (FAO, 2016). Based on a dose-response assessment, the concept consists of defining an exposure threshold under which the diet of consumers is either safe or of low safety concern.

In the case of non-carcinogenic residues (intentionally used in primary production), such as some pesticides and veterinary drugs, the JECFA and JMPR use an acceptable daily intake (ADI). It is calculated from the no-observed adverse effect level (NOAEL) divided by an uncertainty or safety factor. The NOAEL is typically 100-fold the ADI based on intraspecific variability (10) multiplied by interspecific variability (10). In the case of non-genotoxic-and-carcinogenic contaminants, such as heavy metals and some mycotoxins, the JECFA uses tolerable daily intake (TDI), provisional tolerable weekly intake (PTWI), or provisional tolerable monthly intake (PTMI). It is worth noting that the US Environmental Protection Agency (EPA) considers the word 'acceptable' to be somewhat non-scientific, which is why the EPA uses the terminology 'reference dose' (RfD) instead of ADI (Thompson et al., 2018). The benchmark dose (BMD) is a better estimate to derive than ADI or RfD, because it includes all the data necessary for a dose-response assessment (Guo and Mei, 2018).

1.1.1.5 Physiologically based pharmacokinetics

Physiologically based pharmacokinetic models are theoretical models, in which the body is represented as the sum of its compartments. This approach is useful to describe or predict the absorption, distribution, metabolism, and excretion of chemicals, and to extrapolate dose-response and intra- or inter-species variability.

1.1.1.6 Robustness of health-based guidance values

The confidence level of HBGVs depends on the following aspects:

- The pertinence of the health effect
- The relevance of the key study (human data; *in vivo* data or *in vitro* data)

- HBGV type (NOAEL or BMD)
- The quality of the data

Klimisch et al. (1997) proposed a systematic approach for evaluating the quality of experimental toxicological data.

1.1.2 Exposure assessment

Although knowing the toxicity of a substance is key, risk managers also need to know to what extent the chemical is likely to harm a population. It is necessary to identify and assess the various exposure routes. Some population groups are more exposed than others, due to occupational exposure for example. For farmers spraying pesticides, exposure through the air or through skin contact may be significantly higher than dietary exposure, especially when appropriate personal protective equipment is not available or not well understood (Bondori et al., 2018). Dietary exposure might also vary from one location to another. This can result from different food chemical concentration patterns, as well as from different food consumption habits.

Exposure can be assessed by forward dosimetry (the external exposure is obtained by assessing the dietary intake of food chemicals) or by reverse dosimetry (the internal exposure is reflected by biomarkers) (Bui et al., 2017). In general, external exposure is assessed using a combination of food consumption and food chemical concentration data.

1.1.2.1 Food consumption data

Food balance sheets

The most basic estimate of food consumption patterns relies on food balance sheets. This method consists of assessing the sum of locally grown and imported commodities, minus the exported ones. This approach assumes that an estimate of what is on the plate can be calculated from the available food divided by the population size. Data from the International Trade Center may be used (ITC, 2019) in the case of imported and exported food commodities, whereas the Food and Agriculture Organization Corporate Statistical Database provides official data on locally produced foods (FAO, 2019).

Food consumption data estimated through food balance sheets are highly aggregated at a national level. Hence, these data do not take into consideration any local differences or other specificities in dietary patterns, food preparation processes, or seasonal patterns. They also do not account for food losses or informal (non-reported) production, import, or export.

Based on food balance sheets, the WHO determined 17 distinct Global Environment Monitoring System (GEMS) food consumption profiles and named them 'food consumption cluster diets'. As shown in Figure 2, in the latest edition of these classifications (Kim et al., 2015; WHO, 2017), Benin and Cameroon were included in cluster n°3 along with Angola, Burundi, Congo, Côte d'Ivoire, the Democratic Republic of Congo, Ghana, Guinea, Liberia, Madagascar, Mozambique, Paraguay, Togo, and Zambia. Nigeria and Mali were included in cluster n°13 together with Kenya, Malawi, Namibia, Niger, Nigeria, Senegal, Somalia, Sudan, Swaziland, Tanzania, and Zimbabwe. Following the 5th international workshop on TDS held in Korea, the 8th recommendation is that, in the absence of food consumption data, countries are encouraged to use these food cluster diets.

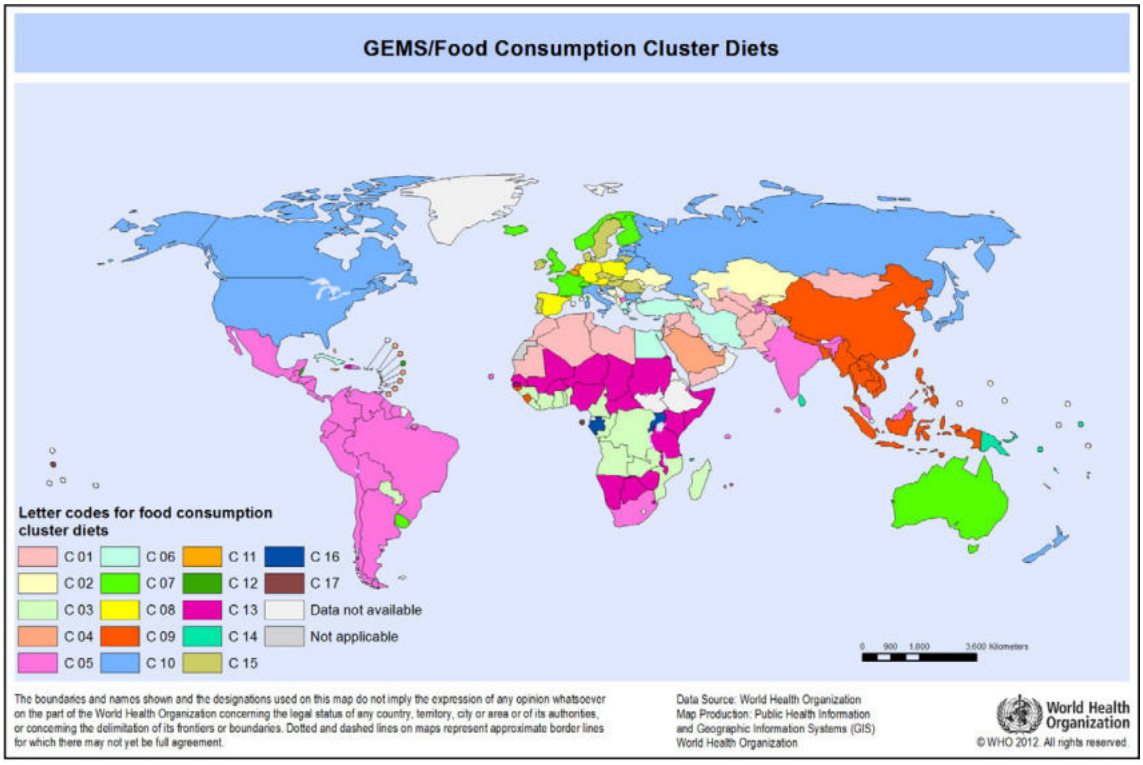


Figure 2: The 2012 WHO GEMS/Food Consumption Cluster Diets (Kim et al., 2015)

Household budget surveys

Household budget surveys (HBSs) are much more precise than food balance sheets. HBSs use declarations made by the head of a household and usually take into consideration more than 5 000 households (Rivière and Sirot, 2018) from different locations, so that local specificities in dietary patterns may be identified. Depending on the food classification system used in an HBS, several hundred food items can be recorded for each household, over a period ranging from 2 weeks to more than 36 weeks. However, data from HBSs do not take into consideration food preparation processes, seasonal patterns, or food losses. HBS data do not allow for the identification of age- or sex-specific food consumption patterns and do not include anthropometric parameters such as height and weight. This type of study, however, enables the identification of household-specific patterns.

In Africa, national stakeholders carry out HBSs about every five years. They are sometimes coupled with Health and Demographic Studies, and often benefit from financial and technical support from development partners (the United Nations Development Programme, UNICEF, or bilateral cooperation agencies). Most HBS executive reports are stored on the International Labour Organization website's central data catalogue (ILO, 2018). In Brazil, a comparison of HBS-derived food consumption data with individual consumption data showed that there is a good correlation between the two, especially in terms of the relative contributions of different foods to energy consumption, and even more so in the main food groups (de Oliveira et al., 2019).

Individual food consumption data

Individual food consumption data are collected based on 24-h recall questionnaires. By increasing the number of days that the exercise is conducted, the results become more reliable. It is common to recommend at least two non-consecutive days for each study subject. Individual food consumption data may be qualitative or quantitative. In the latter case, various approaches are used to estimate portion sizes, such as direct weighting of portions and estimates based on reference photographs, among others. Individual food consumption data usually includes anthropometric parameters such as the body weight, sex, and age of every study subject. The EFSA (2014) recommends recording at least 1 560 subjects, including six age groups with 260 subjects each.

However, recording seasonal variation in food consumption patterns is not usually possible when collecting individual food consumption data. St George et al. (2016), who performed such interviews with the African American youth, questioned the reliability of 24-h recall. With 1-3 different 24-h recalls, the reliability was 11-62%. To be able to achieve 80% reliability, the number of recalls varied depending on the type of intake parameter being considered. For energy intake, an 80% reliability would require 8 recalls, fat intake 13 recalls, fruit intake 21-32 recalls, and vegetable intake 21-25 recalls. There is currently a massive global effort for the production and dissemination of reliable and open access individual food consumption data, through the recently launched 'GIFT' platform (Leclercq et al., 2019).

1.1.2.2 Food chemical concentration data

Official control and surveillance plans may generate food contamination data when they exist and when the reports are available. Such data can also come from operators, through their auto-control activities. In Africa, academic researchers often provide these data. Depending on the data source and quality, a certain level of cautiousness is required when using these data for dietary exposure assessments.

Methods such as thin layer chromatography can detect aflatoxin spots as small as 0.5 ng (Hodges et al., 1964), quantified by fluorimetry. However, GC- and LC-MS/MS have become more common over the last two decades. An obvious advantage of MS/MS is the constant increase of the analytes within the same analysis; a constantly evolving sensitivity leading to high confidence in the interpretation of the signals due to the high specificity of the technique (Sulyok et al., 2007; Malachova et al., 2014). Trueness and precision characterize the accuracy of analytical methods (Menditto et al., 2017). Trueness is achieved by reducing the impact of systematic errors and bias, whereas precision results from the reduction of random errors, implying enhanced repeatability as shown in Figure 3. In other words, accuracy results from a combination of trueness and precision, as shown in Figure 4.

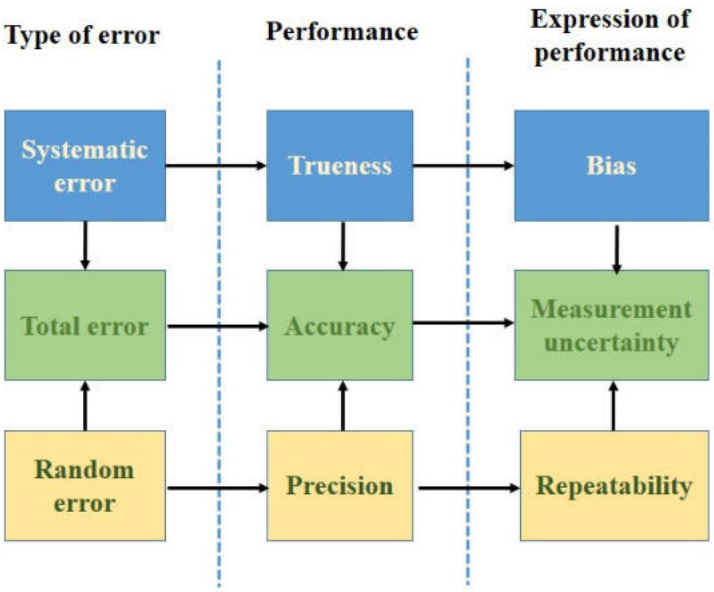


Figure 3: Relationship between type of error, performance, and their expression (Menditto et al., 2007)

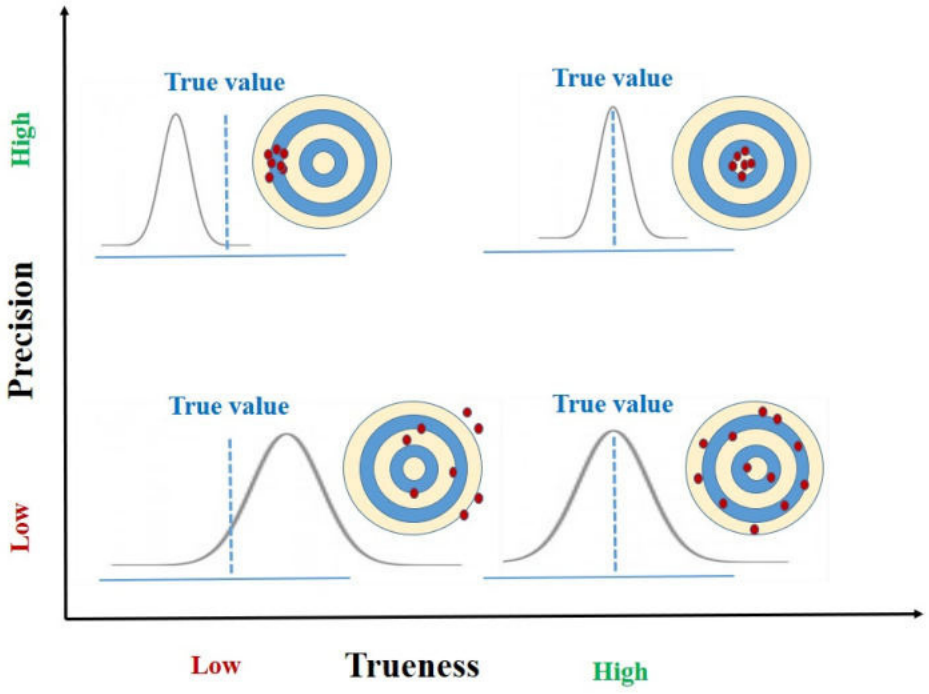


Figure 4: Effect of the association of accuracy and precision on the distribution of analytical data

Quality management systems are essential to guarantee confidence in the analytical data provided by laboratories. According to ISO 9000:2015, which defines the vocabulary of quality

management systems, the confidence provided by quality assurance is double: (1) internally to management and (2) externally to customers and third parties (ISO, 2015). While quality assurance focuses on the means to be able to achieve this, quality control refers to the outcome, for example by verifying processes through proficiency testing among several laboratories. The ISO/IEC 17025:2017 is the latest iteration of the voluntary international standard for general requirements for the competence of testing and calibration laboratories (ISO, 2017). Although accreditation to this standard is a definite asset for laboratories, many laboratories comply with this standard without undergoing the accreditation process, due to the extensive time and resources required.

Participation in proficiency testing is mandatory but is not sufficient for accreditation. It allows for the assessment and comparison of the ability of laboratories in a network (Vander Heyden and Smeyers-Verbeke, 2007). Spiking experiments allow the assessment of recovery by resorting to internal standards, which behave similarly to the tested analytes in a food matrix. Isotope dilution, which is a particular application of internal standards, involves the addition of an isotopically labelled compound. It is considered the most reliable test in terms of metrology (Lehmann, 2016). Deuterium (^{2}H) or carbon 13 (^{13}C) labelling is most often used.

The use of certified reference material (CRM) is necessary to standardize the calibration of analytical devices and to generate true data, thus making studies comparable with other, related TDSs. CRM consists of pure chemicals or pure food matrices of high-quality analytical standard (Krska and Welzig, 2003). Samples used in interlaboratory studies and associated with assigned values can be used to assure the comparability of the TDSs.

The TDS represents an adequate source for food contamination data. The combination of a representative sampling plan, and the preparation of foods as consumed, means that the confidence level in those food chemical concentration data is higher than for any other source. Resorting to a composite sample reduces the cost and enhances the representativeness of samples compared to the true mean concentration in the diet. In Figure 5, the 95% confidence interval of the mean observed concentration is shown, with two hypotheses in terms of the dispersion of concentrations among the sub-samples. In blue, the SD is 30% of the mean concentration, whereas in red, it is 100%. This corresponds to the formula (Sirot et al., 2009a):

$$CI = 1.96 \times \frac{SD}{\sqrt{n}}$$

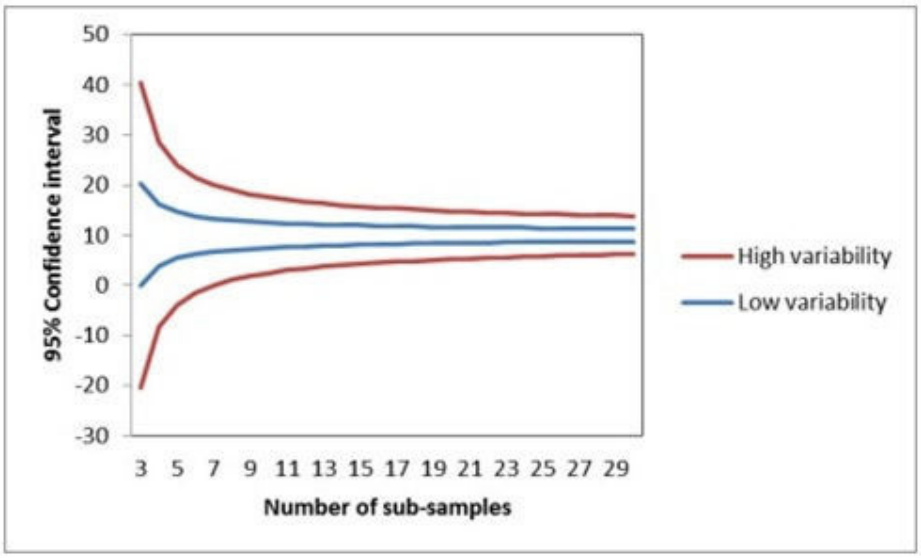


Figure 5: Variation of the confidence interval around the true mean concentration by subsample number (source: Oliver Lindtner)

In a hypothetical situation where the number of subsamples was infinite, the confidence interval and true mean concentration would be superposed.

The selection of chemicals/food matrix pairs to be included in a TDS may depend on scientific information related to the hazard's toxicity, to the known occurrence of the hazard in food, and to the consumption of the corresponding food item. The availability of corresponding analytical technology may be needed to be included in the planning, as well as the perceptions of the society, and, finally yet importantly, budget constraints. Decision making about the selection of analytes may result from an analytical hierarchy process (Papadopoulos et al., 2015). In this approach, experts' judgements are weighted and compared. The authors concluded that, in this analytical hierarchy process, the result depends heavily on the national situation and on available information. The list of analytes, therefore, varies highly through time and space and depends heavily on the TDS objectives.

Concerning the generated food contamination data, the IPCS and WHO (2014) recommend the use of two estimates of concentration, the lower-bound hypothesis, where the concentration of non-detects is set to zero, and the upper-bound hypothesis, where the concentration is set to the limit of detection. In cases where a concentration exceeds the limit of detection but remains

below the limit of quantification, the lower-bound value is the limit of detection and the upper-bound value is the limit of quantification.

1.1.2.3 External exposure calculation methods

Acute dietary exposure assessment

An acute assessment investigates the effect of a chemical ingested in a single dose, possibly from one food commodity (Petersen, 2013). TDS data are not adequate for acute dietary exposure assessment because they are based on food consumption patterns over several days (from two non-consecutive days to 36 weeks). According to the Environmental Health Criteria 240 (WHO, 2009), chronic dietary exposure may be calculated by different means:

Deterministic dietary exposure assessment

The point estimate approach is based on a certain level of food consumption (for example, the median, mean, or high percentile) multiplied by an individual observed mean concentration of food chemical which results from the analysis of pooled samples (Figure 6).

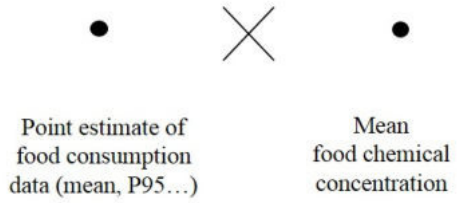


Figure 6: Deterministic dietary exposure assessment (source Dorne et al., 2009)

The limitation of the deterministic approach is that it takes into consideration neither the distribution of consumption level nor the distribution of the food chemical concentration. In other words, deterministic exposure assessments require caution when applying them to large populations (e.g., to a nation). For example, if the food consumption data are derived from food balance sheets, the mean and even the high percentile values are inconclusive concerning the whole population. Some population groups may present significantly different dietary patterns

from the typical national diet. It is therefore likely that these population groups also undergo different exposure patterns, compared to the rest of the population.

Probabilistic dietary exposure assessment

A probabilistic dietary assessment, in contrast to the deterministic approach, takes both the distribution of food consumption within the population group and the distribution of the food chemical concentration into account (Figure 7). The distribution of food consumption data means that individual food consumption data are available.

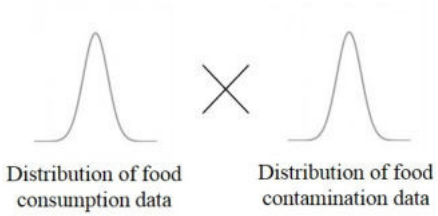


Figure 7: Probabilistic dietary exposure assessment (source Dorne et al., 2009)

A probabilistic dietary exposure assessment is the ideal scenario for a TDS, although it turns out to be much costlier than point estimation, due to the high costs of collecting individual food consumption data and characterizing the contamination of a large set of food samples.

Semi-distributional dietary exposure assessment

A semi-distributional dietary assessment is an intermediate approach between the deterministic and probabilistic methods, where the distribution of food consumption within the population group and the mean observed concentration of the food chemical concentration are taken into consideration (Figure 8).

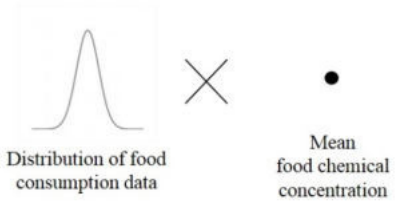


Figure 8: Semi-distributional dietary exposure assessment (source Dorne et al., 2009)

Monte-Carlo Simulations

Monte Carlo analysis generates variability from an existing data set and may be applied to dietary exposure assessments (Glorennec et al., 2016, Wu et al., 2019). Distributions of exposure following a Monte-Carlo calculation tend to reduce exposure at the extremes (high and low percentiles), and rare events may not be considered when a Monte Carlo analysis is used (Bertail et al., 2010).

1.1.3 Risk characterization

Risk characterization consists of comparing dietary exposure with HBGVs or other toxicological end points (e.g., benchmark dose lower confidence limits [BMDLs]). Toxicologists may propose using the HBGV with a threshold. However, in some case, for example with genotoxic and carcinogenic compounds, the threshold approach is not applicable to risk characterization.

1.1.3.1 Chemicals with toxicological thresholds

When exposure exceeds a chemical HBGV with a toxicological threshold (ADI, TDI, PMTDI, PTWI, or PTMI), it represents a safety concern for the population (WHO, 2009). Risk managers may consider various exposure levels for this comparison with the HBGV, including the mean exposure, the 95th percentile, and the 99th percentile, depending on the objectives they pursue.

1.1.3.2 Genotoxic carcinogens

For genotoxic and carcinogenic chemicals, it is impossible to state that a certain exposure level is safe. In contrast, the safety assessment involves a margin of exposure (MOE) for highly exposed individuals (in general the 95th percentile) with a BMDL. The MOE is the ratio between the BMDL and an estimated exposure level. When an MOE exceeds 10 000, the risk is considered of no safety concern (Benford et al., 2010).

Aflatoxin B1 is a genotoxic carcinogen and an exception to the MOE approach. The JECFA in its 83rd session proposed to assess a morbidity factor derived from the prevalence of the hepatitis B virus and the exposure in ng/kg body weight/day, by multiplying dietary exposure by 0.3 for carriers and by 0.01 for non-carriers. The morbidity factor expresses the number of additional cancer cases due to the aflatoxin B1 exposure, per 100 000 inhabitants per year (WHO, 2016).

1.1.3.3 Toxicological end points

For several chemicals, including lead, no threshold or MOE is applicable. An observed outcome is extrapolated from dietary exposure, such as increases in the blood pressure of adults or a loss of IQ in children (WHO, 2011).

1.2. Total Diet Studies

Although the scientific community benefits from 60 years of TDSs experience, interest in the topic seems to have expanded dramatically over the last twenty years. As shown in Figure 9, the number of articles located in a National Center for Biotechnology Information search by submitting the request 'Total diet study', from 1998 to 2018, has increased fourfold from 21 (1998-2002) to 86 (2014-2018) articles.

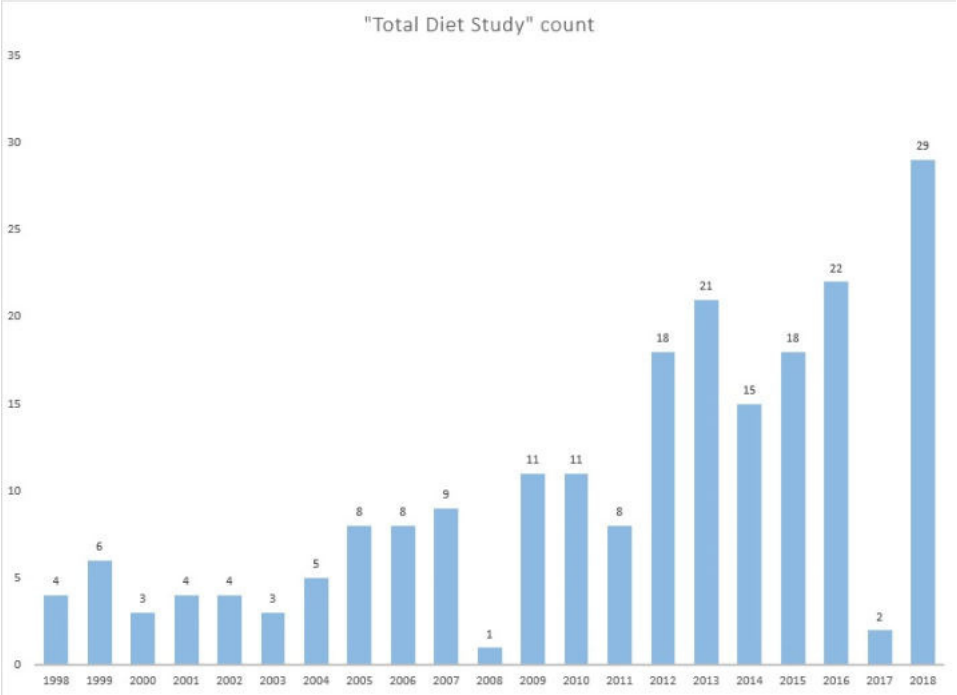


Figure 9: Number of articles recorded in the National Center for Biotechnology Information database corresponding to the request 'Total diet study' 1998-2018

1.2.1. US total diet studies

The TDS concept originally grew out of concerns about dietary exposure to pesticide residues and to radionuclide fallout, in the context of nuclear weapons testing (Egan, 2013). The launch of the first TDS dates to the early 1960s in the USA to monitor levels of selected substances in foods as consumed (Pennington and Gunderson, 1987; Egan et al., 2007). Previously, in 1957, the US Public Health Services had started measuring monthly Sr-90 levels in milk in five

geographic regions of the USA. At that time, public health officers assumed that milk was the main contributor to Sr-90 intake. In 1958, the independent Consumers Union carried out a wider study of milk samples from 48 US cities. In November 1959, a project led by the Consumers Union with the support of universities studied the diet of teenagers based on a typical 14-day menu, which is believed to be the first ever TDS. One important conclusion was that milk only contributed about half of the Sr-90 exposure in an average diet. In 1961, the Food and Drug Administration (USFDA) launched their first TDS in several cities with single analytical composites of 82 foods collected four times a year. In 1965, the FDA subdivided this quarterly market basket into 12 food groups.

Nowadays, the USFDA uses food consumption data from the National Health and Nutrition Survey (NHANES) conducted by the National Center for Health Statistics. The dietary component of the NHANES is known as What We Eat In America (WWEIA). WWEIA conducts 24-h recall interviews on two non-consecutive days (Ahluwalia, 2016). The first step is an in-person interview, followed by a phone interview held within the following three to ten days. Portion size is either estimated using 3D models of spoons, mugs, and so on, or by using a booklet containing drawings of various sizes of containers (Raper et al., 2004).

The USFDA TDSs analyse samples of 280 foods, collected as market baskets in 12 cities in four areas (west, north central, northeast, and south; three cities per area) four times a year (FDA, 2019). The number of analytes varies from year to year and included 350 food chemicals in 2014 (Lee et al., 2015) increasing to 800 in 2018 (FDA, 2019). The analytes considered by the USFDA TDSs include metals and trace elements, pesticides, radionuclides, POPs, and processing contaminants. The FDA pooled the three samples collected in the three different cities in each geographic area to form a sample. Other than that, the FDA did not pool the samples. The NHANES also collects anthropometric measures and biomarkers in addition to food samples.

1.2.2. UK total diet studies

The UK also has a long history of carrying out TDSs, starting in 1966. The food group approach was used in British TDSs and evolved from nine food groups (containing 115 foods) to 20 food groups (containing 68-119 foods) after 1981 (Peattie et al., 1983). More recently, food consumption data have been derived from the National Diet and Nutrition Survey, which is co-

funded by Public Health England and the Food Standard Agency. In this survey, about 1 000 subjects are investigated annually, including 500 children aged 1.5 to 17 years and 500 adults (Public Health England, 2019).

The UK TDS exposure assessment uses the Food Standards Agency distributional model called the Intake II Program. The 986 individual food samples were pooled into 20 food groups, which were then analysed. The analyte list in the UK included metals and trace elements, perfluorinated chemicals, dioxin and dioxin-like PCBs, acrylamide, PAHs, and uranium 238.

The Committee on Toxicity of Chemicals in Food (Shavila, 2013) noted that the uncertainty in these results was considerable, especially for perfluorinated compounds, which might be a consequence of the dilution effect of the food group approach. The UK TDS occurrence data which are available generally indicate low contamination (FERA, 2012), and to the best on my knowledge, the exposure assessments have not been published to date.

1.2.3. Australian total diet studies

In Australia, 24 TDSs were conducted from 1970-2014 by the Australian National Health and Medical Research Council and then by Food Standards Australia New Zealand (FSANZ, 2014). The food consumption data of the Australia TDS (ATDS) were derived from the National Nutrition Survey (Australian Bureau of Statistics, 2014) in which 13 858 respondents (aged 2 years or older) were presented with a 24-h recall questionnaire, including their height, weight, and blood pressure. On the second day, 1 489 of the respondents (11%) underwent a second 24-h recall.

The analytes covered by the 23rd ATDS were metals and trace elements, pesticide residues, veterinary drug residues, mycotoxins, PCBs, PAHs, and polybrominated diphenyl ethers (a total of 214 compounds) in 93 foods. In the 23rd ATDS report, the food-analyte combinations were provided (FSANZ, 2011) and are summarized in Table 1.

Table 1: Summary of food analyte combinations in the 23rd Australian TDS.

	Pesticides and veterinary drugs	Al, Cd, Pb	Hg, As	Antimicrobial residues	Aflatoxins B1, B2, G1 & G2	Aflatoxin M1	DON, ZEA	OTA	Fumonisin B1 & B2	Patulin
Cereals	●	●			●		●		●	
Tubers	●	●								
Baked beans	●	●			●				●	
Fruits	●	●								
Fruit juices	●	●								●
Peanuts & almonds	●	●			●					
Meat	●	●		●	●		●		●	
Eggs	●	●		●						
Seafood	●	●	●	●						
Dairy products	●	●		●		●				
Olive oil	●	●						●		
Hot beverages	●	●						●		
Infant foods	●	●			●	●				
Other foods	●	●			●					

In this table, it can be seen that mycotoxins (aflatoxins, DON, ZEA, and fumonisins) were tested in meat (in fact, only in meatpies). Mycotoxins were not detected in the ATDS; but as the limit of detection was not reported, it is tough to assess the robustness of these observations.

The latest ATDS, the 24th, included acrylamide, aluminium, and perchlorate, together with 30 packaging chemicals including bisphenol A and phthalates (FSANZ, 2014). The number of food items included in the ATDS varied between 60 and 96, depending on the year.

1.2.4. Canadian total diet studies

Canada also has a long history of implementing TDSs, starting in 1969 (Smith, 1971). Since 1999, Canadian TDSs have covered 10 cities and have screened pesticides, dioxins, PCBs, metals, and trace elements, as well as radionuclides. While pesticide exposure was concluded to be very low compared to the JMPR ADIs (Rawn et al., 2004), Canadian TDSs have shown a steep decrease in lead exposure from 0.8 µg/kg body weight/day in 1981 to 0.1 µg/kg body weight/day in 2000-2007. These results were consistent with two risk mitigation measures implemented in Canada:

- The conversion of lead soldered cans to lead free ones (1975-1982)
- The gradual phasing out of leaded gasoline (1975-1990)

This is one of the few documented successes of using TDS results to identify a risk and assess subsequent mitigations measures over time. The Canadian TDS analysed 159 composites collected in a different city every year. Each composite consists of four subsamples (Health Canada, 2013).

More recently, Juric et al. (2017, 2018) completed a risk assessment on a First Nations population and noted that the exposure to methylmercury and lead were 1.6 and 1.7 times higher than in the general Canadian population. The methylmercury P95 exposure remained well below the health-based guidance value, and as only 7.9% of participants met their recommended fish consumption, Health Canada recommended the consumption of more fish from lower trophic levels. The average lead exposure was associated with a 1.2 mmHg increase in systolic blood pressure (Juric et al., 2017). The major contributor was moose and deer meat, which indicated that lead-containing ammunition was putting the population at an elevated risk of lead toxicity (Juric et al., 2017).

1.2.5. Chinese total diet studies

The Chinese experience of conducting TDSs dates back to 1990 and includes five studies so far (Shi et al., 2017). The first Chinese TDS used food consumption data of 1 080 households from 12 provinces. The food consumption data were derived from a 3-day household survey plus three 24 h-recalls on non-consecutive days. Thirty households with mid-level economic status

were randomly selected in three survey sites (two rural and one urban) covering about 50% of the Chinese population aged 2 years and older. The subsamples were highly aggregated into 12 food composites from each of the four regions (each region included three provinces), so that the whole study covered 48 composites of cereals, legumes and nuts, meat, eggs, fish, dairy products, vegetables, fruits, sugar, beverages including water, condiments, and cooking oils. The number of tested analytes was 96 in the first Chinese TDS (Chen, 2013a). Out of 12 monitored organophosphate pesticides, five were detected in this TDS, methamidophos being the most prevalent. In all cases, pesticide exposures only accounted for a small percentage of the acceptable daily intake. No organophosphate pesticide exceeded the limit of detection in any of the four investigated regions.

In 1992, the second Chinese TDS included food consumption data corresponding to four age groups: 2-7, 8-12, 20-50 year-old male, and 20-50 year-old female. The number of samples increased to 120. From 2000, the Chinese TDS used a combination of the food group composite approach and an individual food approach. The decrease in dietary exposure to the organochlorine pesticides DDT and HCH, which were banned in 1985 in China, was consistently highlighted by the Chinese TDSs (Zhou et al., 2012).

Similarly, the comparison of previous results to those of the 5th Chinese TDS showed an obvious shift from the use of historical brominated flame retardants (such as PBDEs) to novel Brominated Flame Retardants (BFRs), in particular decabromodiphenylethane (Shi et al., 2016, 2017). Likewise, a steep decrease in dioxin and dioxin-like PCBs was observed thanks to the 5th Chinese TDS (Zhang et al., 2015). The acrylamide margin of exposure was as low as 565 in the same study (Gao et al., 2016). Sterigmatocystin and citrinin did not come up as health concerns in a typical Chinese diet and the aflatoxin B1 was only investigated in the first Chinese TDS. It was detected in only one food group composite and the authors concluded that the risk related to aflatoxins was acceptable (Qiu et al., 2017).

1.2.6. Hong Kong total diet study

The first TDS carried out in Hong Kong was based on a list of 150 foods established from two non-consecutive 24-h recalls of 5 008 individuals aged 20-84 years. The 1 800 collected foods

were acquired in three regions, namely Hong Kong Island, Kowloon, and the New Territories, and pooled to include three subsamples per composite (Chung et al., 2014).

The Hong Kong TDS showed that dietary exposure to inorganic arsenic, more than half of which was due to rice, was significant (Chung et al., 2014). The Hong Kong population was also at risk due to high exposure to aluminium, cadmium, and methylmercury. The exposure of women of childbearing age to methylmercury exceeded 150% of the provisional tolerable weekly intake (Chen et al., 2014). Stir-fried vegetables contributed to almost half of the acrylamide exposure and resulted in a non-protective MOE, below 10 000 (Wong et al., 2014a). The organochlorine pesticides exceeded the limit of detection in 55% of all composites, and mainly consisted of DDT (32%), HCB (30%), and endosulfan (22%) (Chen et al., 2015). The exposure to all pesticides, including organochlorine and organophosphates was, however, relatively low, the 95th percentile upper-bound exposure remaining below 24% of the acceptable daily intake (Wong et al., 2014b). Dietary exposure to dioxins and PCBs (Wong et al., 2013), as well as brominated flame-retardants (Chen et al., 2013), were not of safety concern according to the Hong Kong TDS.

1.2.7. French total diet studies

In France, the TDSs used the individual food approach. In 2000-2001, ANSES (formerly AFSSA) conducted the first French TDS (FTDS), including 18 trace elements and 14 mycotoxins and using five subsamples per composite. The total numbers of composites were 1 080 (trace elements) and 456 (mycotoxins). The study concluded that the risks for the French population were relatively low, but did not exclude that some specific population groups, such as children and vegetarians, could be at risk from ochratoxin A, deoxynivalenol, and zearalenone. Cereals contributed most to dietary exposure to the mycotoxins (Leblanc et al., 2005a, b).

In the meantime, the Calipso study investigated the contamination of fish with a TDS approach, targeting commonly consumed fish and analysing methylmercury, arsenic, and POPs. While contamination with trace elements was relatively homogenous in the four coastal areas investigated, the fish were more contaminated with POPs in Le Havre and least contaminated in Toulon. POP contamination seemed to follow a North-South gradient (AFSSA, 2006).

The second French TDS (FTDS2) included 445 analytes (2006-2010) and used 15 subsamples per composite. It used 212 different core foods and collected around 20 280 foods divided between 1 352 composites (Sirot, 2009). Interestingly, this study showed that, compared to the first FTDS, exposure to cadmium and aluminium was higher. In contrast, the lead, mercury, and arsenic exposure was lower but these risks could not be ruled out, due to high analytical limits (5 µg/kg for lead and 10 µg/kg for mercury) (Arnich et al., 2012). The FTDS2 also investigated exposure to food additives: annatto (E160b), nitrites (E249-250), sulphites (E220-228), and tartaric acid (E334). The results showed that, in 2.9% of the adult French population, exposure exceeded the ADI for sulphites, mainly due to wine consumption (Bemrah et al., 2012).

The study of acrylamide exposure revealed low margins of exposure, below 100 at the 95th percentile, due to the contribution of crisps and chips (Sirot et al., 2012a). Sirot and colleagues (2012b) highlighted that exposure to polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), and dioxin-like polychlorinated biphenyls (DL-PCBs) declined 3.2-fold since the previous evaluation, which followed a different methodology. This can be interpreted as displaying the efficiency of the risk management measures implemented after the previous evaluation (Sirot et al., 2012b).

The TDS2 showed that exposure to PAHs through food is not a major problem for French consumers (Veyrand et al., 2013). The estimated exposure to perfluoroalkyl acids and brominated flame-retardants was also unlikely to pose any risk, although gaps in the knowledge about some congeners were highlighted (Rivière et al., 2014). A specific case study of 188 French vegetarians used the FTDS2 data (Fleury et al., 2017). The result of this analysis showed that French vegetarians are less exposed to POPs and much more exposed to some mycotoxins and trace elements (Cd and Al) than the general population.

In addition, in 2010, ANSES conducted an infant TDS (iTDS) as a sub-component of the FTDS2, covering more than 500 analytes with 12 subsamples per composite (Hulin, 2014) representing the diet typically consumed by children under three years old. The sampling plan covered more than 80% of the typical infant diet with over 450 composites formed from the pooling of more than 5 500 food items. Occurrence data from the iTDS on lead, mercury (Guérin et al., 2017, 2018), acrylamide, and furan (Lambert et al., 2018a, b) were released. Exposure to cadmium (Jean et al., 2018) and to a selection of trace elements including Al, As, Pb, and Hg (Sirot et al., 2018) has been calculated. The iTDS showed that the risks with respect

to Al and methylmercury cannot be ruled out due to uncertainties, whereas Cd, Pb, and inorganic As pose problems for a significant proportion of children younger than 3 years in France.

1.2.8. Lebanese total diet studies

The Lebanese TDSs started with lead, cadmium, mercury, and radionuclides (Nasreddine et al., 2006). The authors reported the non-detection of Cs-134 and I-131, whereas Cs-137 was detected in only five samples. The mean lead exposure was only 7% of the PTWI, which was, at that time, 0.025 mg/kg body weight/week. The estimated cadmium and mercury exposures reached 17% and 5.6% of their respective PTWIs. In the following study, lead and cadmium exposure corresponded to 3.2% and 21.7% of their respective PTWIs, which was different from previous findings and could partly result from the fact that the sampling methodology was not the same (Nasreddine et al., 2010).

The Lebanese TDS also investigated some mycotoxins, in particular aflatoxin B1, ochratoxin A, and deoxynivalenol. Dietary exposure to OTA at the 95th percentile was 95.1% of the corresponding PTWI, and 355.8% in the case of DON (Raad et al., 2014). More recently, the same team looked at pesticides using the TDS approach. The number of detected pesticides was 18, the most prevalent of which were chlorpyrifos, procymidone, pirimiphos-methyl, dimethoate, and dieldrin (Nasreddine et al., 2016). In the minimalist hypothesis, the concentration of non-detected analytes was set to zero and the concentration of detected but unquantified analytes was set to the limit of quantification; all mean dietary exposure was below the acceptable daily intake. However, in the maximalist hypothesis which uses the analytical limits as the concentration, the mean dieldrin exposure was 100.7% and 128.7% of the ADI in urban and semi-rural areas, respectively. These exceedances in the upper-bound hypothesis is a source of uncertainty which results from inadequate analytical limits.

1.2.9. The first Cameroonian total diet study

Gimou et al. (2008) conducted a TDS in Yaounde targeting 46 pesticides in 63 composite food samples formed from composites of variable weight and prepared as consumed. The dietary

exposure of a selection of 557 households was calculated. The study noted the high uncertainty associated with the values (high upper-bound/lower-bound concentration ratios) but even in the upper-bound hypothesis, the dietary exposures were well below the JMPR ADIs.

Dietary exposure to lead in the Yaoundé population was noted as high (3.05 µg/kg body weight/day at the 95th percentile) due to the high contribution of cassava (Gimou et al., 2014b). The exposures to arsenic, cadmium, and inorganic mercury were not of concern according to the Yaoundé TDS.

1.2.10. Other total diet studies

Many other countries have reported their TDSs, including Cambodia (Cheng et al., 2013, 2016), Japan (Nakatani et al., 2011), India (Betsy et al., 2014), the Czech Republic, Fiji, Indonesia, Malaysia, The Netherlands, Spain, Sweden (Moy and Vannoort, 2013), and, more recently, Germany (Sarvan et al., 2017).

1.3. Critical analysis of various total diet studies

The purpose of this paragraph is not to criticize one methodology or another, as implementing a TDS is a learning curve for the scientific community, and the entities in charge do not all share the same resources, objectives, and limitations.

1.3.1. Target analytical limits

The most critical element of a TDS is probably the analytical limits (LOD/LOQ) of the testing method. When a large proportion of data consist of non-detects, it adds to the uncertainty of the exposure assessment. This is even more damaging if the non-detects include staple foods, because the quantity of food consumed augments dietary exposure in the upper-bound scenario. It is particularly important to carry out a safety assessment based on 100% of the censored data (meaning that no analyte exceeds the limit of detection), unless of course the upper-bound exposure is very low, much lower than the HBGV, in which case it is possible to rule out any

risk. In order to avoid spending large amounts of resources, energy, and time, without the assurance of obtaining tangible results, it is possible to define in advance the feasibility of a risk assessment exercise (Pité et al., 2018).

An upper-bound exposure, which reflects the theoretical exposure resulting from 100% of the non-detected data (pessimistic scenario), is calculated from a concentration equal to a target LOD or LOQ multiplied by food consumption data. In the case of 100% non-detects, the lower-bound exposure equals zero. It is pertinent to compare the upper-bound exposure, with a relevant HBGV. It may happen that this upper-bound exposure corresponding to 100% of the censored data exceeds the HBGV, which would be problematic. If, in a real situation, the lower-bound exposure is lower than the HBGV but the upper-bound exposure exceeds the HBGV, it is impossible to draw any conclusions concerning the risk. In such a situation, the analyte component of the TDS may require the analytical limits to be lowered, which is not always possible. If the analytical limits and food consumption data do not allow for the upper-bound exposure (corresponding to 100% of the censored data) to remain below the HBGV, it is necessary to evaluate the options.

If occurrence data from previous studies are available and show that a certain chemical is prevalent in the area of interest and that its concentration usually exceeds the analytical limit, it is worth testing the analyte in question. In that scenario, both the lower- and upper-bound exposures are likely to exceed the HBGV, which is important to report as it means that the population is at risk. However, if the analytical limits do not allow conclusions to be drawn concerning the risk, risk assessors may decide either to test the analyte anyway or not to include it in the analyte list. This decision is a strategic one and depends on available resources.

1.3.2. Number of subsamples per composite and the concentration confidence interval

One of the arguments for not pooling many subsamples together is that it dilutes the chemicals in concentrated samples with less concentrated ones (Churchill et al., 2013). This is why the Canada TDS used only four subsamples per composite. The advantage of this approach is that it makes it easier to identify the origin of the contamination. However, this approach means that the observed mean concentration will include a wide confidence interval around the true mean

concentration (Figure 5). There is, however, a costly solution to this issue, which consists of analysing several samples and using the mean concentration from those samples.

1.3.3. Aggregation level of composites

The TDS with the highest level of aggregation is certainly the UK TDS, with only 20 composites. Although this approach is highly feasible and cheap to run, the Committee on Toxicity of Chemicals in Food (2008) noted that the uncertainty was considerable, as a consequence of the dilution effect of the food group approach.

1.3.4. Geographic variability

Among the fundamental aspects driving TDS methodology, the question of whether to pool food samples from different geographic areas or not is critical. This question comes down to either forming nationally representative samples, or composites that are specific to certain smaller areas. In France, the FTDS data showed that there are no significant differences in contamination and exposure patterns between the French regions (Leblanc et al., 2005). The US TDS generates regionally representative samples, with three samples from three different cities (FDA, 2019), whereas the Canada TDS takes all the subsamples from the same city (Health Canada, 2013). Each method presents some advantages and drawbacks, which, at the end of the day, depend on the TDS objectives. Dofkova et al. (2016) presented a harmonized food list for five European countries, namely the Czech Republic, Finland, Germany, Iceland, and Portugal.

1.3.5. Seasonal variability

Through the seasons, the availability of foods such as fruits may vary, and so do their consumption patterns. Similarly, the contamination levels of foods may change, due to different environmental conditions, among other things. Pesticides are used at particular times of the year, which may vary from one crop to another, as well as from one place to another.

Mycotoxin-producing fungi thrive in certain climatic conditions (Gagiu et al., 2018). Because of these numerous parameters, capturing seasonal variability is challenging. For example, the design would have to include some reflection on the topic and the researchers involved in the project would need to keep the samples from different seasons separate if possible as it could help with understanding contamination determinants (Elegbede et al., 2017).

1.3.6. Population groups

Populations groups such as infants and young children or women of childbearing age have different susceptibility to the toxic effects of chemicals and can have different consumption habits. It is therefore valuable to be able to define, as precisely as possible, who eats what in a population. To achieve this goal, it is necessary to generate individual food consumption data, which relates who eats what within a household. The EFSA recommended the use of at least 1 560 individuals within six age groups (including 260 individuals of both sexes per age group) (EFSA, 2014c). To date, only one infant TDS has been reported (Hulin et al., 2014). Akhandaf et al. (2015) have proposed a cost-effective method of establishing food lists for specific population groups, based on the mean consumption of the general population.

'Tell me what you eat, and I will tell you who you are.'

Jean Anthelme Brillat Savarin (1 April 1755 - 2 February 1826)

2. METHODOLOGY

I had the opportunity to carry out this PhD study in parallel with a project lead by the FAO, WHO, and CPC with contributions by several national food safety authorities and submitted with the consent of the STDF. The inclusion of national teams from Benin, Cameroon, Mali, and Nigeria in the final SSA-TDS project proposal was possible thanks to their ability and motivation to contribute, in spite of the limited availability of funds.

2.1. Generation of food consumption data

2.1.1. Food classification and hierarchy

The fact that this study included four countries meant that each country had its own food classification system, including a specific number of food items (163-284) and specific terminology. Harmonizing and standardizing these food classification systems therefore became a challenge and a priority. In 2012, the FAO released the West Africa Food Composition Tables (FAO, 2012). We used the 13 food groups from this table equally in this study. These groups include cereals, tubers, legumes, vegetables, fruits, nuts and seeds, meat, eggs, fish, dairy products, oils and fats, beverages, and miscellaneous.

The aggregation level of these food groups was, however, too broad to highlight specific food safety issues. For example, it was expected that aflatoxins would be present in significant amounts in peanuts but not in beans, whereas both food commodities belong to the food group 'Legumes'. Similarly, we expected to find high amounts of PAH in smoked fish, and much less in other types of seafood.

Therefore, it seemed pertinent to include another stratum to the food classification system, which would:

1. Be harmonized among the four countries,
2. Characterize the pooling level of the sampling plan.

This stratum included the 84 subgroups considered the maximum pooling level for sampling. This means that subsamples from two different food subgroups should never be pooled in the same sample. The three strata of the food classification system are represented in Figure 10. The two higher strata, which are harmonized across the four countries, are shown in Table 2.

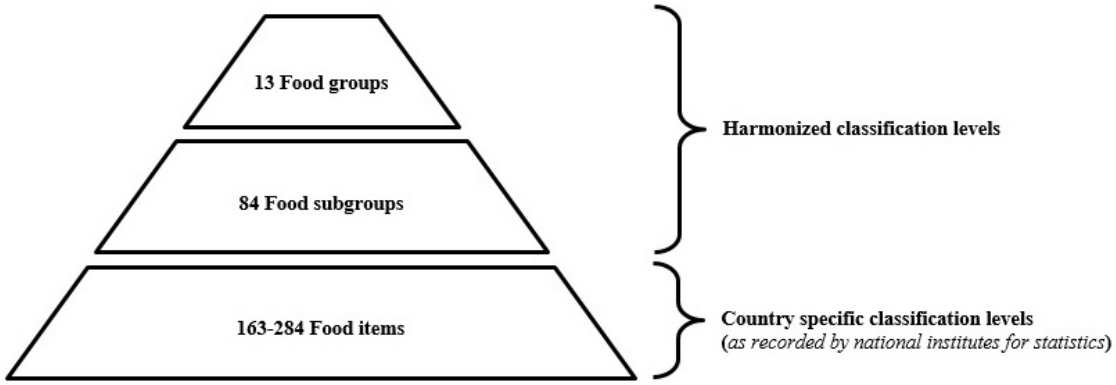


Figure 10: Food classification

Table 2: Food groups and subgroups of the SSA-TDS classification.

Food Group N°	Food Group	Food Subgroup N°	Food Subgroup
1	CEREALS	1.1	RICE
1	CEREALS	1.2	MAIZE
1	CEREALS	1.3	WHEAT/BREAD
1	CEREALS	1.4	PASTA
1	CEREALS	1.5	SORGHO
1	CEREALS	1.6	MIL
1	CEREALS	1.7	OTHER CEREALS
2	TUBERS	2.1	CASSAVA FRESH
2	TUBERS	2.2	CASSAVA DRY
2	TUBERS	2.3	YAM FRESH
2	TUBERS	2.4	YAM DRY
2	TUBERS	2.5	POTATO FRESH
2	TUBERS	2.6	POTATO DRY
2	TUBERS	2.7	SWEET POTATO
2	TUBERS	2.8	COCOYAM=TARO
2	TUBERS	2.9	MACABO
3	TUBERS	2.10	OTHER TUBERS
3	LEGUMES	3.1	BEANS
3	LEGUMES	3.2	PEANUTS
3	LEGUMES	3.3	PEAS
3	LEGUMES	3.4	SOJA
3	LEGUMES	3.5	OTHER LEGUMES
4	VEGETABLES	4.1	TOMATO
4	VEGETABLES	4.2	CARROT
4	VEGETABLES	4.3	GREEN LEAVES
4	VEGETABLES	4.4	COURGETTES, CUCUMBER & GROUND PEPER, EGGPLANT (GARDEN EGG)
4	VEGETABLES	4.5	CABBAGE
4	VEGETABLES	4.6	ONION & GARLIC
4	VEGETABLES	4.7	OKRO = GOMBO
4	VEGETABLES	4.8	PARSLEY, CELERY, BASIL & LEEK
4	VEGETABLES	4.9	OTHER VEGETABLES
5	FRUITS	5.1	BANANA
5	FRUITS	5.2	PLANTAIN
5	FRUITS	5.3	MANGO
5	FRUITS	5.4	PINEAPPLE
5	FRUITS	5.5	CITRUS (ORANGE, LEMON, LIME…)
5	FRUITS	5.6	AVOCADO
5	FRUITS	5.7	PAWPAW
5	FRUITS	5.8	MELON / WATERMELON
5	FRUITS	5.9	OTHER FRUITS
6	NUTS/SEEDS	6.1	COCONUT
6	NUTS/SEEDS	6.2	CASHEW NET
6	NUTS/SEEDS	6.3	PALM NUT
6	NUTS/SEEDS	6.4	OTHER NUTS/SEEDS
7	MEAT	7.1	BEEF
7	MEAT	7.2	POULTRY
7	MEAT	7.3	MUTTON/GOAT
7	MEAT	7.4	PORK
7	MEAT	7.5	PROCESSED MEAT
7	MEAT	7.6	GAME MEAT
7	MEAT	7.7	INSECTS
7	MEAT	7.8	OTHER MEAT
8	EGGS	8.1	POULTRY EGGS
9	FISH	9.1	SEA FISH
9	FISH	9.2	FRESH WATER FISH
9	FISH	9.3	SMOKED FISH
9	FISH	9.4	PROCESSED FISH
9	FISH	9.5	CRUSTACEANS/MOLLUSCS
9	FISH	9.6	OTHER SEAFOOD
10	MILK/DAIRY	10.1	FRESH/FERMENTED MILK
10	MILK/DAIRY	10.2	CONCENTRATED/DEHYDRATED MILK
10	MILK/DAIRY	10.3	OTHER MILK PRODUCTS
11	OIL/FAT	11.1	PALM OIL
11	OIL/FAT	11.2	GROUNDNUT OIL
11	OIL/FAT	11.3	OTHER VEGETAL OIL
11	OIL/FAT	11.4	OTHER FAT/OIL
12	BEVERAGES	12.1	WATER
12	BEVERAGES	12.2	FRUIT JUICE
12	BEVERAGES	12.3	TRADITIONAL SOFT DRINK
12	BEVERAGES	12.4	TRADITIONAL FERMENTED DRINK
12	BEVERAGES	12.5	INDUSTRIAL FERMENTED DRINK
12	BEVERAGES	12.6	INDUSTRIAL SOFT DRINK
12	BEVERAGES	12.7	SPIRITS
12	BEVERAGES	12.8	OTHER DRINKS
13	MISCELLANEOUS	13.1	SUGAR
13	MISCELLANEOUS	13.2	SALT
13	MISCELLANEOUS	13.3	BROTH/BOUILLON CUBE
13	MISCELLANEOUS	13.4	HONEY
13	MISCELLANEOUS	13.5	TEA
13	MISCELLANEOUS	13.6	COFFEE
13	MISCELLANEOUS	13.7	CHOCOLATE
13	MISCELLANEOUS	13.8	BABY MILK POWDER
13	MISCELLANEOUS	13.9	CHILI/PEPER
13	MISCELLANEOUS	13.10	OTHER MISCELLANEOUS

2.1.2. Food consumption data derivation process

We derived the food consumption data from HBSs available in Benin, Cameroon, Mali, and Nigeria and recorded them over a two-week period. The four HBSs gathered data from 72 979 households and included:

- the estimated value of food produced by the households for their own consumption
- the amount spent on each food commodity, which was recorded by the national institutes of statistics and expressed in local currency.

Corresponding tables were filled in for each country starting with the lowest ranking level (i.e. each national food item) and entering the edible fraction conversion factor, yield factor (reflecting weight change during the cooking process), and energy content obtained either from the West African Food Composition Table (FAO, 2012) or the French Food Composition Table (ANSES, 2013). In order to obtain a standardized unit to describe the energy intake of the study population, the values were converted into adult male energy intake equivalents (AMEs) using the equivalence scale from Nigeria (Table 3). This was possible because the sex and age of every household member was systematically recorded in each of the four HBSs.

Table 3: Adult equivalence factors provided by Nigeria.

Age Group	Male	Female
Less than 1 year	0.25	0.25
1 to less than 4 years	0.45	0.45
4 to less than 7 years	0.62	0.62
7 to less than 11 years	0.69	0.69
11 to less than 15 years	0.86	0.76
15 to less than 19 years	1.04	0.76
19 to less than 26 years	1.00	0.76
26 to less than 51 years	1.00	0.76
51 years and above	0.79	0.66

The relevance of using AMEs for estimating household energy requirements was summarized by (Weisell and Dop, 2012). The purpose of estimating the energy requirements of a household is to select households whose declared food consumption corresponds to a realistic range of energy intake.

We estimated food consumption data 'as consumed' in g/AME/day, derived using the following three-step process:

1. Food expenditure and food produced by households for their own consumption was reported by the national HBS in local currency over a two week period and converted into '*daily quantity of raw food commodity purchased*' with the help of a unit price database provided by each national institute of statistics.
2. The quantities of raw food commodities purchased and produced for household consumption were converted into '*daily quantity of edible raw food commodity*' using edible fraction conversion factors identified in the West African Food Composition Table (FAO, 2012).
3. The quantity of edible raw food was converted into '*daily amount of food as consumed*' using yield factors (FAO, 2012).

In order to eliminate biases due to under-reporting or over-reporting households, normal-reporting households were selected within the range of 1 200 kcal/AME/day to 5 100 kcal/AME/day. We removed all under-reporting and over-reporting households from the datasets. These extremes correspond to the mean energy requirement of an adult male of 60 kg (FAO, 2001) minus 45% for the lower limit and plus 45% for the higher limit. We chose these margins based on the hypothesis that 1 SD = 15%, in order to include households with energy requirements ± 3 SD. After applying these limits, we selected 61% of the 72 979 recorded households and formed a dataset of 44 431 normal reporting households.

2.1.3. Study populations

The study could have covered these 44 431 normal reporting households from Benin, Cameroon, Mali, and Nigeria. However, discussions with national stakeholders drew attention to the fact that, within a country, diets (and therefore dietary exposure) are likely to vary significantly. National stakeholders decided to select two study centres per country, with different diets. The breakdown of the number of households is represented in Table 4.

Table 4: Selected households from eight study centres.

COUNTRY	Centre	Selected households
BENIN	Littoral	1490
	Borgou	1004
CAMEROON	Duala	890
	North	508
MALI	Bamako	1318
	Sikasso	1015
NIGERIA	Lagos	301
	Kano	765
TOTAL		7291

2.1.4. Food lists

In order to keep the number of samples necessary to carry out the SSA-TDS as low as possible and to keep the cost of the study within the budgeted resources, the sampled national food lists did not cover 100% of the mean total diet. We reduced the number of required samples through inclusion criteria using a two-step approach. We first ordered the list of 84 food subgroups from the most to the least consumed, and selected the core foods which covered the first 90% of the national mean total diet by weight. Limiting the core food selection process to this first round would have limited the number of samples required to cover these 15-20 selected core foods, but would have excluded some important food groups such as meat and fish in some of the countries.

To avoid the almost exclusive coverage of staple foods, we included a second step in the core food selection process, to ensure significant coverage of each of the 13 food groups. Ideally, the coverage by weight of each food group (level 1) including the selected core foods (level 2) would also systematically be 90%. However, in order to allow for considerable reduction in the number of samples, we decided to only cover the core foods making up 50% of each food group if the food group represented less than 1% of the mean total diet, and 90% otherwise.

2.2. Sampling approach

2.2.1. Standard Operating Procedures

Standard Operating Procedures (SOPs) are necessary in order to harmonize sampling and allow comparison of results among study centres, as well as to avoid bias in contamination levels due

to the inadequate handling of samples. We derived the SSA-SOPs from the ones developed and used in the framework of the European TDS Exposure Project (Pité et al., 2018). We developed training materials and trained the staff in charge of collecting, preparing, and organizing the storage and transport of food samples. They were given mandatory training in four SOPs concerning:

1. The prerequisites prior to collecting samples,
2. The collection and shipment of samples from markets,
3. The reception and storage of samples in the kitchen-laboratory,
4. The preparation, pooling, and preservation of samples.

2.2.2. Pooling of samples

According to the EU TDS-Exposure Project SOPs, 12 subsamples of equal weight reflect, with a reasonable amount of uncertainty, the real mean concentration which would result from a theoretical situation where the number of subsamples is infinite. The 95% confidence interval of the concentration within a pooled composite sample containing n subsamples of equal weight around the true mean concentration is the following:

$$CI = 1.96 * \frac{SD}{\sqrt{n}}$$

The SSA-TDS methodology systematically used 12 subsamples of equal weight per composite sample.

2.2.3. Notion of national and local samples

To reduce the number of samples, without decreasing the coverage of the mean total diet by weight, we collected some samples in one study centre, but applied the concentration in both study centres in the same country. For example, for a strictly imported commodity, or when the production of a commodity mainly occurs in one of the study centres or in another location, we considered that contamination levels are likely to remain the same, regardless of the collection centre. In such cases, we considered the core food as a *national* core food. Otherwise, we

defined the core foods as *local* and collected them independently from both locations in the same country. The SSA-TDS used the principle of *local sample by default*, which means that, in the absence of justification as to why a core food is likely to be from the same origin, we collected it in both centres.

2.2.4. Breakdown of subsamples per composite

In paragraph 2.2.2., we mentioned the fact that composite samples are constructed from 12 subsamples of equal weight. One critical aspect to obtain representative samples is to use available data on the variability of samples and apply it in the sampling approach.

The data we exploited in the SSA-TDS were:

- The origins of imported food commodities
- The proportions of food item categories
- Main purchase locations

The origins of imported food commodities were available from the ITC website. The proportions of food item categories were obtained from the food consumption data of the food items at level 3 of the classification. We gathered information about main purchase places during preliminary market surveys.

2.2.5. Seasonal variation

We carried out the sampling plan with two collection campaigns. The first sampling wave took place simultaneously in the eight study centres during the rainy season (October 2017) and covered all the core foods in the 13 food groups. The second wave, during the dry season in February 2018, covered only the five first food groups, namely cereals, tubers, legumes, vegetables, and fruits, and excluded nuts and seeds, meat, eggs, seafood, dairy products, oil and fats, beverages, and miscellaneous.

2.3 Analytical plan

2.3.1. Analytical grid

The purpose of the analytical grid is to define the correspondence between the tested analyte groups and the sampled core foods. We tested the samples collected during the rainy season against all the analyte groups, as defined in the analytical grid, whereas we tested samples collected during the dry season for mycotoxins and pesticides only. We included core foods in the sampling of specific analyte groups partly based on assumptions that the chemicals were likely to be present in the food matrices. The inclusion criteria considered the test process and specific properties such as the lipophilicity of the core food. We also took resource limitations into consideration (Table 5).

Table 5: Analytical grid where 1 indicates which core foods were tested against an analyte group.

N°	Food Subgroup	Metals and trace elements	Pesticides	Mycotoxins	PAH	POPs
1.1	RICE	1	1	1		
1.2	MAIZE	1	1	1		
1.3	WHEAT/BREAD	1	1	1		
1.4	PASTA	1	1	1		
1.5	SORGHO	1	1	1		
1.6	MIL	1	1	1		
1.7	OTHER CEREALS	1	1			
2.1	CASSAVA FRESH	1	1			
2.2	CASSAVA DRY	1	1	1	1	
2.3	YAM FRESH	1	1			
2.4	YAM DRY	1	1	1	1	
2.5	POTATO FRESH	1	1			
2.6	POTATO DRY	1	1			
2.7	SWEET POTATO	1	1			
2.8	COCOYAM=TARO	1	1			
2.9	MACABO	1	1			
3.1	BEANS	1	1	1		
3.2	PEANUTS	1	1	1		
3.3	PEAS	1	1	1		
4.1	TOMATO	1	1			
4.3	GREEN LEAVES	1	1			
4.4	COURGETTES, CUCUMBER & GROUND PEPER, EGGPLANT (GARDEN EGG)	1	1			
4.5	CABBAGE	1	1			
4.6	ONION & GARLIC	1	1	1		
4.7	OKRO = GOMBO	1	1			
4.9	OTHER VEGETABLES	1	1			
5.1	BANANA	1	1			
5.2	PLANTAIN	1	1			
5.3	MANGO	1	1			
5.5	CITRUS (ORANGE, LEMON, LIME…)	1	1			
5.7	PAWPAW	1	1			
5.8	MELON / WATERMELON	1	1			
6.2	CASHEW NUT	1	1	1		
6.3	PALM NUT	1	1	1		
6.4	OTHER NUTS/SEEDS	1	1	1		
7.1	BEEF	1	1	1		1
7.2	POULTRY	1	1	1		1
7.3	MUTTON/GOAT	1	1	1		1
8.1	POULTRY EGGS	1		1		1
9.1	SEA FISH	1	1			1
9.2	FRESH WATER FISH	1	1			1
9.3	SMOKED FISH	1	1	1	1	1
9.5	CRUSTACEANS/MOLLUSCS	1	1			1
10.1	FRESH/FERMENTED MILK	1	1	1		1
10.2	CONCENTRATED/DEHYDRATED MILK	1	1	1	1	1
11.1	PALM OIL	1	1	1	1	1
11.2	GROUNDNUT OIL	1	1	1		1
11.3	OTHER VEGETAL OIL	1	1	1		1
11.4	OTHER FAT/OIL	1	1	1		1
12.1	WATER	1	1			
12.3	TRADITIONAL SOFT DRINK	1	1	1		
12.4	TRADITIONAL FERMENTED DRINK	1	1	1		
12.5	INDUSTRIAL FERMENTED DRINK	1	1	1		
12.6	INDUSTRIAL SOFT DRINK	1	1			
13.1	SUGAR	1	1			
13.2	SALT	1				
13.3	BROTH/BOUILLON CUBE	1	1	1	1	1
13.6	COFFEE	1	1	1	1	
13.7	CHOCOLATE	1	1	1	1	
13.9	CHILI/PEPER	1	1	1		
13.10	OTHER MISCELLANEOUS	1	1	1	1	

2.4. Generation of food chemical concentration data

No analytical developments or optimizations were attempted in this thesis, although the importance of using reliable and sensitive analyses (meaning with low limits of detection/quantification) was key to the success of the study. The analytical methods, including quality control, are described in detail in the occurrence articles presented in Chapter 3. They were mainly based on the latest technology involving mass spectrometry coupled with chromatography (either gas or liquid). The analytical methods were all validated and used in accredited laboratories that had been previously involved in TDSs.

Concerning data management, we expressed the concentrations so that any uncertainty due to censored data or non-detects could be taken into consideration. Table 6 describes the food chemical concentrations in the lower-bound and upper-bound hypotheses, depending on the analytical limits.

Table 6: Concentrations used for censored data in the minimalist and maximalist hypotheses.

Concentration level	Lower-bound hypothesis (LB)	Upper-bound hypothesis (UB)
Below LOD	0	LOD
Between LOD and LOQ	LOD	LOQ
Exceeding LOQ	Value	Value

Legend

LOD: limit of detection
LOQ: limit of quantificaion

2.5. Exposure assessment

The approach selected for this study is a form of semi-distributional exposure assessment. Although it is impossible to assess the individual consumption of household members from food consumption data derived from an HBS, the fact that the dietary exposure of 7 291 households was assessed means that an exposure distribution is possible. In contrast, the point estimate approach used for characterizing the food chemical concentration, which was based on the analysis of pooled samples, means that the contamination distribution is unknown.

The exposure was determined for each household at the second level of the food classification system (the food subgroups or core food groups), and divided by 60 kg, the assumed weight of an adult male.

$$E_i = \sum_{k=1}^{n} \frac{C_{i,k} \times L_k}{60}$$

Where:

E_i is the daily exposure of a household, normalized by consumption units or AMEs.

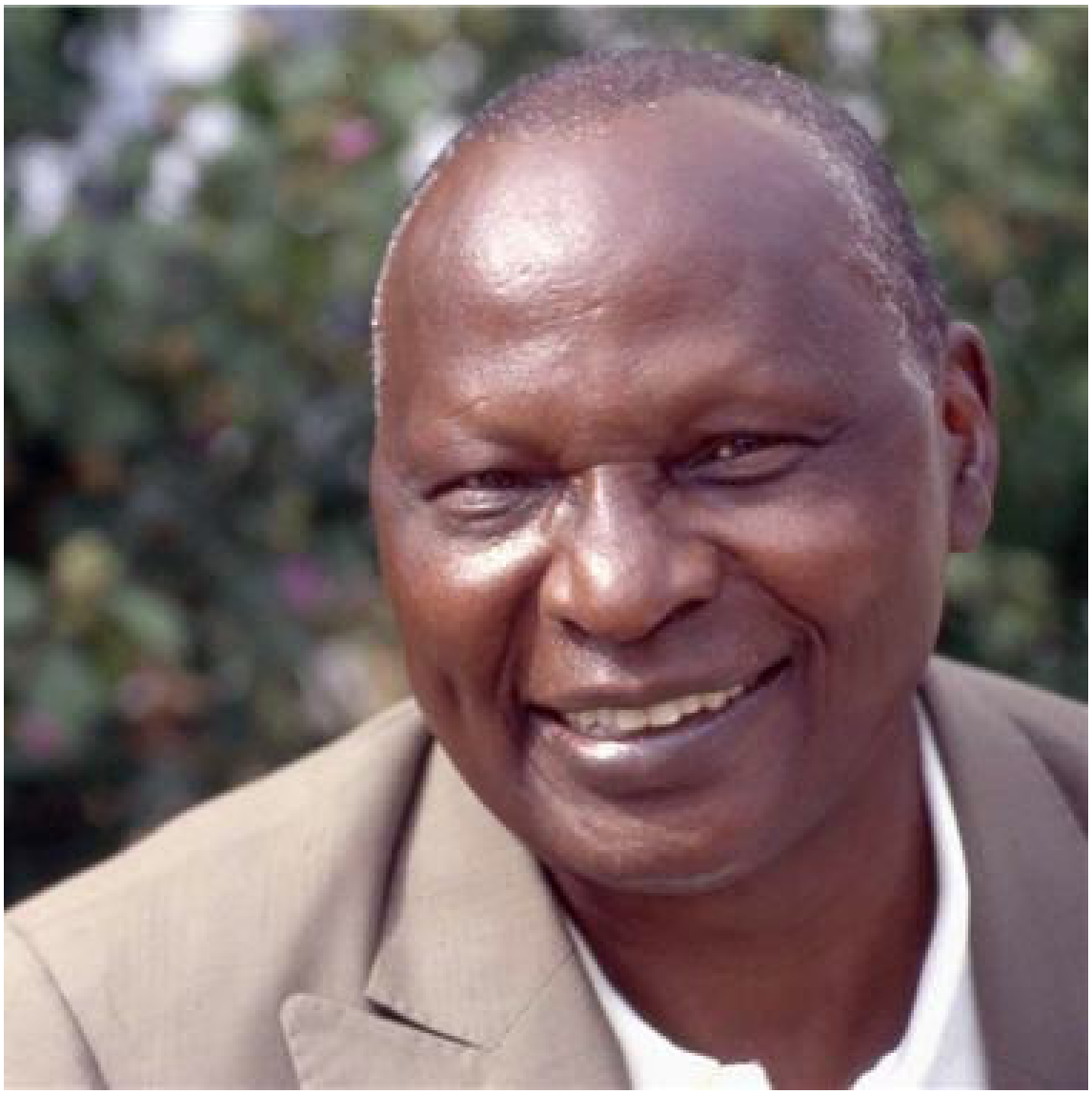
$C_{i,k}$ is the daily consumption of food subgroup k in normalized household i.

L_k is the level or mean concentration of the food chemical investigated in food subgroup k.

And:

- L_k is specific to each study centre if the core food is local, and specific to each country if the core food is national.
- L_k was obtained exclusively from samples collected during the rainy season, except in the case of pesticides and mycotoxins.
- In the case of pesticides and mycotoxins, L_k was the arithmetic mean of concentrations obtained from samples collected during the rainy and dry seasons for the first five food groups, namely cereals, tubers, legumes, vegetables, and fruits. For nuts and seeds, meat, eggs, seafood, dairy products, oil and fats, beverages, and miscellaneous, we analysed the pesticides and mycotoxin exclusively in samples collected during the rainy season.
- E_i was obtained with both the minimalist (lower-bound) and maximalist (upper-bound) hypothesis for each [household-chemical] couple.

The computation of the exposure values was performed using the software SAS.



'*The truth may redden your eyes, but it won't blind you*'

Ahmadou Kourouma (24 November 1927 - 11 December 2003)

3. RESULTS

The results of implementing the SSA-TDS are presented below.

They consist of seven articles at various stages of the peer review process.

- Food consumption data (published in *Food and Chemical Toxicology* 16.08.17)
- Mycotoxin occurrence data (published in *Toxins* 17.01.19)
- PAH occurrence data (published in *Food Control* 09.04.19)
- Pesticide occurrence data (published in *Food Chemistry* 30.06.19)
- Metal and trace element occurrence data (published in *Environment International*)
- POP occurrence data (published in *Environment International*)
- Exposure assessment (in preparation for submission to *The Lancet Global Health*)

3.1. Food Consumption Data

The food consumption data were not only assessed with respect to the normal-reporting households of the eight study centres (7 291 households), but for all the regions, districts, or states of Benin, Cameroon, Mali, and Nigeria. In total, we assessed the consumption of 44 431 households. We calculated food consumption at all levels of the food classification system: the national food items, food subgroups (also denoted as core foods), and food groups. We used each stratum of the food hierarchy for a different purpose in the SSA-TDS methodology:

- The food groups defined food consumption patterns.
- The food subgroups defined the core foods or composite samples tested in laboratories
- The national food items defined the breakdown of the subsamples in the composites

In the first article, entitled '*Methodology design of the regional Sub-Saharan Africa Total Diet Study in Benin, Cameroon, Mali and Nigeria*', the patterns based on the mean food consumption of cereals were compared between populations dwelling by the ocean (the Littoral area of Benin, Duala, and Lagos) with the other study centres (Borgou, North Cameroon, Bamako, Sikasso, and Kano). The diets of coastal populations contained significantly less cereal than the other population groups ($p = 0.001$). The typical diet of populations living close to the Sahel region is essentially based on cereals. Because these diets differ, we anticipated that dietary exposure patterns would differ accordingly.

In this first paper, food consumption data (the mean and 95th percentile) are presented at the level of food subgroups for the four countries (Benin, Cameroon, Mali, and Nigeria), as well as

for the eight study centres. Food and Chemical Toxicology accepted the publication of the related article well ahead of sample collection. The paper serves two purposes:

1. To offer an explanation and justification of the applied sampling approach,
2. To provide food consumption data compatible with a deterministic exposure assessment.

This paper lays the foundations for the SSA-TDS and describes the key methodological choices which apply to the occurrence and risk assessment articles. Citing this article avoids constantly repeating the methodology. The SSA-TDS methodology complies with three recommendations from a guidance document entitled '*Towards a harmonized Total Diet Study approach'* (EFSA, 2011): the sampling is representative of food consumption habits, the foods are prepared as consumed, and the samples are pooled.

The methodology was, by design, as cost-effective as possible. In the context of developing countries where budget is one of the most limiting factors, efficient use of the available resources is necessary. By reducing the number of samples needed to carry out a TDS, the cost of analysis is also decreased considerably. The idea is to demonstrate that a TDS may provide valuable food safety intelligence, without necessarily requesting a multimillion budget. Lowering the cost of implementing a TDS is key to the promotion of the approach and its replication in other developing countries, in Africa and beyond.

Another important aspect of the design is the multi-centre approach. The inclusion of eight study centres, where we assessed dietary exposure using both local food consumption data and local food contamination data, was a gamble. This approach is very costly, contradicting the objective mentioned in the previous paragraph. This compromise can only be justified and replicated if the risks characterized in each of the study centres vary to a significant degree. We could have opted for national TDSs, where nationally representative composites would result from collecting and pooling samples from various areas of the same country. Ultimately, we could have pooled samples from different countries before sending them to laboratories. Our approach was different, and we decided to study and compare the study centres separately.

Obtaining representative samples was another challenge. Pooling 12 sub-samples certainly contributed to that objective by considerably reducing the confidence interval around the true mean concentration, as shown in Figure 4. The individual food approach meant that we never mixed two different core foods. This aspect was essential in order to identify the origin of any

contamination, and more particularly, to determine which core foods were the main exposure contributors.

Which samples are representative of the core foods?

This key question also needed to be addressed and the next article provides some answers to it using a decision tree based on the analysis of the food balance sheet structure and food consumption at the most detailed level of food classification. We presented the food consumption data by country and by study centre, at the two higher levels of the food classification system, food groups and food subgroups. Food groups determine food consumption patterns. The consumption of cereals, in particular, is key when it comes to assessing the exposure profiles of population groups dwelling in various areas. Two aspects may explain this: specific food consumption (naturally) and different contamination patterns.

What are the determinants of specific exposure patterns?

Ultimately, we calibrated the methodology to answer these questions. If specific exposure patterns emerge from the study, it is necessary to be able to identify if this is due to high concentrations in a minority of food commodities, high consumption of specific foods, or if the contamination is ubiquitous. The answers will define the vulnerability of the population to these food safety issues. This aspect of identifying how easy an issue is to solve is already dallying on the side of risk management and goes beyond the sole responsibility of risk assessors.

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Methodology design of the regional Sub-Saharan Africa Total Diet Study in Benin, Cameroon, Mali and Nigeria

Luc Ingenbleek [a, b], Eric Jazet [c], Anaclet D. Dzossa [c], Samson B. Adebayo [d], Julius Ogungbangbe [e], Sylvestre Dansou [f], Zima J. Diallo [g], Christiant Kouebou [h], Abimbola Adegboye [d], Epiphane Hossou [i], Salimata Coulibaly [j], Sara Eyangoh [a], Bruno Le Bizec [b], Philippe Verger [k], Jean Kamanzi [l], Caroline Merten [l], Jean-Charles Leblanc [l, *]

[a] Centre Pasteur du Cameroun, (CPC), Yaounde, Cameroon
[b] LUNAM Université, Oniris, LABERCA, Nantes, France
[c] National Institute for Statistics, (NIS), Yaounde, Cameroon
[d] National Agency for Food and Drug Administration and Control, (NAFDAC), Abuja, Nigeria
[e] National Bureau of Statistics (NBS), Abuja, Nigeria
[f] Institut National de la Statistique et de l'Analyse Economique (INSAE), Cotonou, Benin
[g] Institut National de la Statistique (INSTAT), Bamako, Mali
[h] Institut de Recherche Agronomique pour le Développement (IRAD), Yaounde, Cameroon
[i] Agence Béninoise de Sécurité Sanitaire des Aliments (ABSSA), Cotonou, Benin
[j] Agence Nationale pour la Sécurité Sanitaire des Aliments (ANSSA), Bamako, Mali
[k] World Health Organization (WHO), Geneva, Switzerland
[l] Food and Agriculture Organization of the United Nations (FAO), Rome, Italy

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ABSTRACT

The core food model was described more than three decades ago, and has been used ever since to identify main food contributors to dietary intakes for both nutrients and other food chemicals. The Sub-Saharan Africa Total Diet Study (SSA-TDS) uses this model to describe the food consumption habits of some selected populations of Benin, Cameroon, Mali, and Nigeria, prior to use in the completion of quantitative risk assessments with regard to food chemicals. Food consumption data were derived from food expenditure data contained in national household budget surveys that were provided by the national institutes of statistics in each country. A classification of African foods was established for the purpose of the study and core foods were selected, so as to reflect 96 ± 1% of the average national total diet expressed in weight. Populations from eight study centers were selected by national stakeholders. This approach involves the purchase of 4020 individual foods, prepared as consumed and pooled into 335 food composite samples, for analysis of mycotoxins, PAHs, PCBs and dioxins, pesticides, metals and trace elements, PFAs, and BFRs. This sampling plan aims to provide a representative, cost effective, and replicable approach for deterministic dietary exposure assessments in developing countries.

1. Introduction

Evaluating the human exposure to potentially harmful substances is a key step in public health risk assessments. A better understanding of these exposures leads to evidence-based decision-making processes, providing for improved risk management at national and international levels.

The dietary exposure of a given population to food chemicals

* Corresponding author.
E-mail addresses: luc.ingenbleek@gmail.com (L. Ingenbleek), jazeteric@yahoo.fr (E. Jazet), adzossa@yahoo.fr (A.D. Dzossa), adebayosam@hotmail.com (S.B. Adebayo), juliusoyedeji@gmail.com (J. Ogungbangbe), dansousyl@yahoo.fr (S. Dansou), zimadiallo@gmail.com (Z.J. Diallo), kchristiant@yahoo.fr (C. Kouebou), adegboye.a@nafdac.gov.ng (A. Adegboye), hossepfr@yahoo.fr (E. Hossou), coulibalysalimat@yahoo.fr (S. Coulibaly), eyangoh@pasteur-yaounde.org (S. Eyangoh), bruno.lebizec@oniris-nantes.fr (B. Le Bizec), vergerp@who.int (P. Verger), kamanzijean@hotmail.com (J. Kamanzi), caroline.merten@efsa.europa.eu (C. Merten), JeanCharles.Leblanc@fao.org (J.-C. Leblanc).

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can be assessed by different approaches (FAO/WHO, 1985; WHO, 2009). The most refined approach for obtaining food chemical concentration data to be used in dietary exposure assessments involves purchasing the foods people eat and analysing these foods. Assessing the occurrence of chemicals of interest in foods as consumed in order to effectively estimate the dietary exposure for different population groups requires an efficient, cost-effective, and accurate method, such as Total Diet Studies (TDS). The TDS approach has been promoted and endorsed by the World Health Organization (WHO) along with the Food and Agriculture Organization of the United Nations (FAO) since the 1960s (WHO, 1968) and more recently in 2011 in a joint guidance document from European Food Safety Authority (EFSA), WHO and FAO (EFSA, 2011b).

TDS are designed to measure the average amount of a given substance ingested by a studied population. This public health oriented approach differs from classical chemical surveillance programs because: (1) it focuses on chemicals in the total diet rather than in specifically targeted food commodities and (2) it takes into consideration, to a certain extent, the impact of home cooking on the decomposition or formation of chemicals, as the foods are prepared as for consumption before analyses (WHO, 2007).

The core food approach was first described in 1982 for the US Total Diet Study (Pennington, 1983; Egan et al., 2007) and has since been used as a common tool by other food safety agencies around the World (EFSA, 2011a; WHO, 2009). A TDS enables identification of foods that are most highly consumed by a study population (in terms of quantity) and which foods contribute most to intakes of energy, nutrients, and other food chemicals. A core food list gathers the main foods representing at least 90% by weight of the average total diet. These foods are sampled and analyzed for the assessment of nutritional intakes or dietary exposure to other food chemicals of a given population.

Two specific aspects characterize a TDS: (1) the representativeness of the sampling and (2) the preparation of the samples "*as consumed*", so that it represents a pertinent public health risk assessment tool, as far as food safety and nutrition are concerned.

The four key steps of a TDS implementation within a specific population include (1) the identification of core foods (2) the derivation of both the average and the high-consumers daily food consumption (3) the sampling, preparation (i.e. prepared and cooked as per the typical consumer behavior), and laboratory analyses of the sampled core foods for nutrients and/or other food chemicals and (4) the exposure assessment and risk characterization obtained from consumption data multiplied by food chemicals concentration data.

Between 2006 and 2010, a TDS was implemented in the city of Yaoundé, Cameroon for the purpose of screening pesticides (Gimou et al., 2008) and metals and trace elements (Gimou et al., 2014). This first ever TDS implemented in Sub-Saharan Africa used a food list including 63 food items obtained from the pooling of national food items from the Cameroonian Household Budget Survey. The Sub-Saharan Africa Total Diet Study (SSA-TDS) is a wider project aiming to investigate a more extended number of food chemicals, within a larger study population.

The SSA-TDS was implemented by FAO in Benin, Cameroon, Mali and Nigeria between 2014 and 2017, together with the four national food safety authorities, in close collaboration with Center Pasteur of Cameroon (CPC) and WHO (FAO, 2014a).

Due to budget constraints, the national stakeholders of the four countries decided to select only two population groups per country. The basis for the selection of the two different population groups per country was distinct dietary behaviors, associated with distinct agro-ecological areas. These study centers include in each country (1) the most densely populated city (Bamako, Cotonou, Duala and Lagos), among which three are located by the Atlantic Ocean Coast, and (2) another study center located in a non-coastal area (the Sikasso Region of Mali, the Borgou Department of Benin, the North Region of Cameroon and the State of Kano of Nigeria).

The design and main methodological choices, forming the basis of the Sub-Saharan Africa Total Diet Study (SSA-TDS) in terms of selection of core foods, food sampling approach, food sample preparation and chemical substances looked for, which represent the main challenges for implementing and adapting the TDS approach for developing countries, are presented here below.

2. Materials and methods

2.1. Food classification and food consumption data

Food consumption data were derived from household budget surveys (HBS) available in Benin, Cameroon, Mali, and Nigeria. The four HBS gather data from a total of 72,979 households and include both the estimated value of food produced by households for their own consumption and the amount spent for each food commodity recorded by national institutes of statistics and expressed in local currency and recorded over a two-week period.

Data recorded by the four national institutes of statistics used heterogeneous food nomenclature, including the total number of distinct food items recorded ranging from 163 (Mali) to 284 (Cameroon). In order to generate comparable food consumption data among the 4 countries, two additional and harmonized levels were added to the food classification as shown in Fig. 1.

The adopted strategy consisted of setting up a corresponding table for each country between the national food items representing 100% of the average national diet and two additional levels that are of a higher ranking. The two additional levels are (1) 84 food subgroups, among which core foods are selected for the purpose of the study and considered to be the maximum pooling level for sampling and (2) 13 food groups taken from the food classification used in the West African Food Composition Table (FAO, 2012). These corresponding tables were filled for each country starting with the lowest ranking level (i.e. for each national food item table), entering edible fraction conversion factors, yield factor (reflecting weight change during the cooking process) and energy content either obtained from the West African Food Composition Table (FAO, 2012) or the French Food Composition Table (ANSES, 2013).

In order to obtain a standardized unit to describe the energy intake of the study population, the sex and age of every household member was systematically recorded in each of the four national household budget surveys (HBS), and converted into adult male equivalents (AME) using the equivalence scale from Nigeria (Table 1).

The relevance of using AME for estimating household energy requirement was summarized by the United Nations University (Weisell and Dop, 2012). Estimating the energy requirement of a household serves to select households whose declared food expenditure corresponds to a realistic range of energy intake.

Food consumption data were estimated as daily consumption of food "as consumed" in grams per adult male equivalent per day derived using the following three-step process: (1) food expenditure and food produced by households for their own consumption reported by national HBS in local currency recorded over a two-week period and converted into "daily quantity of raw food commodity purchased" with the help of a unit price database provided by each national institute of statistics (2) quantities of raw food commodity purchased or produced for household consumption converted into "daily quantity of edible raw food commodity" with edible fraction conversion factors identified in the West African

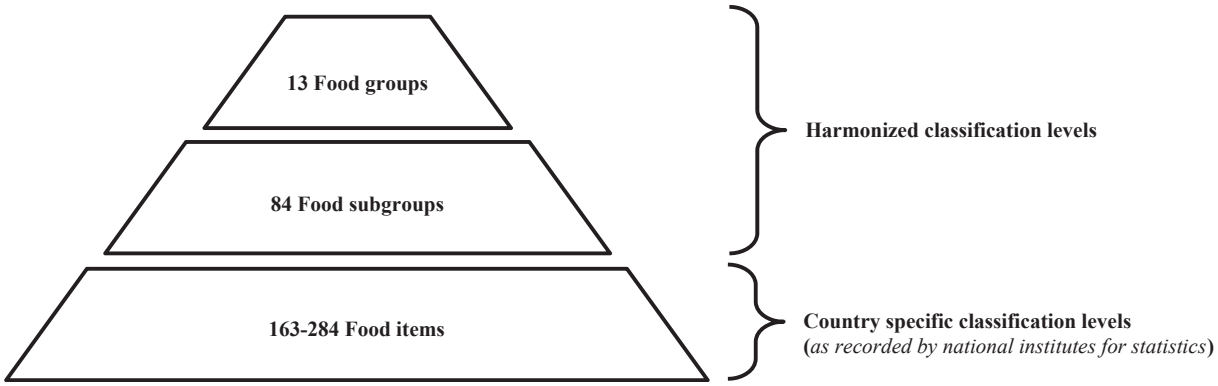

Fig. 1. Hierarchy of the food classification consisting of 3 strata representing each 100% of the average national total diet.

Table 1
Adult equivalence scale (source: National Bureau of Statistics, Nigeria).

Age Group	Male	Female
Less than 1 year	0.25	0.25
1 to less than 4 years	0.45	0.45
4 to less than 7 years	0.62	0.62
7 to less than 11 years	0.69	0.69
11 to less than 15 years	0.86	0.76
15 to less than 19 years	1.04	0.76
19 to less than 26 years	1.00	0.76
26 to less than 51 years	1.00	0.76
51 years and above	0.79	0.66

Food Composition Table (FAO, 2012) (3) quantity of edible raw food converted into "daily amount of food as consumed" with yield factors (FAO, 2012).

In order to eliminate biases due to under-reporting or over-reporting families, normal reporting households were selected within the range from 1200 kcal/AME/day to 5100 kcal/AME/day. Under-reporting and over-reporting households were discarded from datasets. These extremes correspond to the mean energy requirement of an adult male of 60 kg (FAO, 2001) minus 45% for the lower limit and plus 45% for the higher limit. These margins were selected on the basis of the hypothesis that 1SD = 15%, in order to include households energy requirements ± 3SD. After applying these limits, 61% of the 72,979 recorded households were selected to form a dataset of 44,431 normal reporting households.

The general description of the original food purchase datasets for the four countries before and after selection of normal reporting households is displayed in Table 2.

2.2. Selection of core foods

Four national core food lists were established (one per country) as the result of a selection process from a harmonized list of 84 food subgroups (Fig. 1). Representativeness criteria were set with two objectives (1) the coverage by core foods of the average total diet and (2) the coverage of each of the 13 food groups by the selected core foods. Each national list applies to the two study centers selected by each country, with a decision tree (Fig. 2) defining whether composite samples shall be collected locally, or nationally.

2.3. Core food representation versus total diet

In each of the 4 countries, the food subgroups (also named core foods in this study when selected for sampling) were ranked on the basis of their average consumption in grams per adult male equivalent per day and selected in descending order from the most consumed until reaching coverage of 90% of the total average national diet.

2.4. Core food representation versus main food groups

In order to ensure that the various food groups of the diet are adequately represented in the sampling approach, inclusion criteria for the establishment of the core food list were set as follows:

- If the national average consumption of the food group represents more than 1% of the total average national diet in weight, then the total average daily consumption of the food subgroups selected from this food group shall represent 90% of the food consumption of this group or more.

Table 2
Description of national household budget surveys used for setting up food consumption patterns.

Country	Benin	Cameroon	Mali	Nigeria	SSA Total diet study
Data source	National Institute for Statistics and Economic Analysis (INSAE)	National Institute for Statistics (NIS)	National Institute for Statistics (INSTAT)	National Bureau of Statistics (NBS)	TOTAL
National identification of survey	EMICoV	ECAM 3	MICS-ELIM	HNLSS	–
Year of survey implementation	2011	2007	2010	2010	–
Total number of households	17,667	11,347	8987	34,978	72,979
Number of selected households	13,967	8471	7834	14,159	44,431
Percentage of selected households	79%	75%	87%	40%	61%

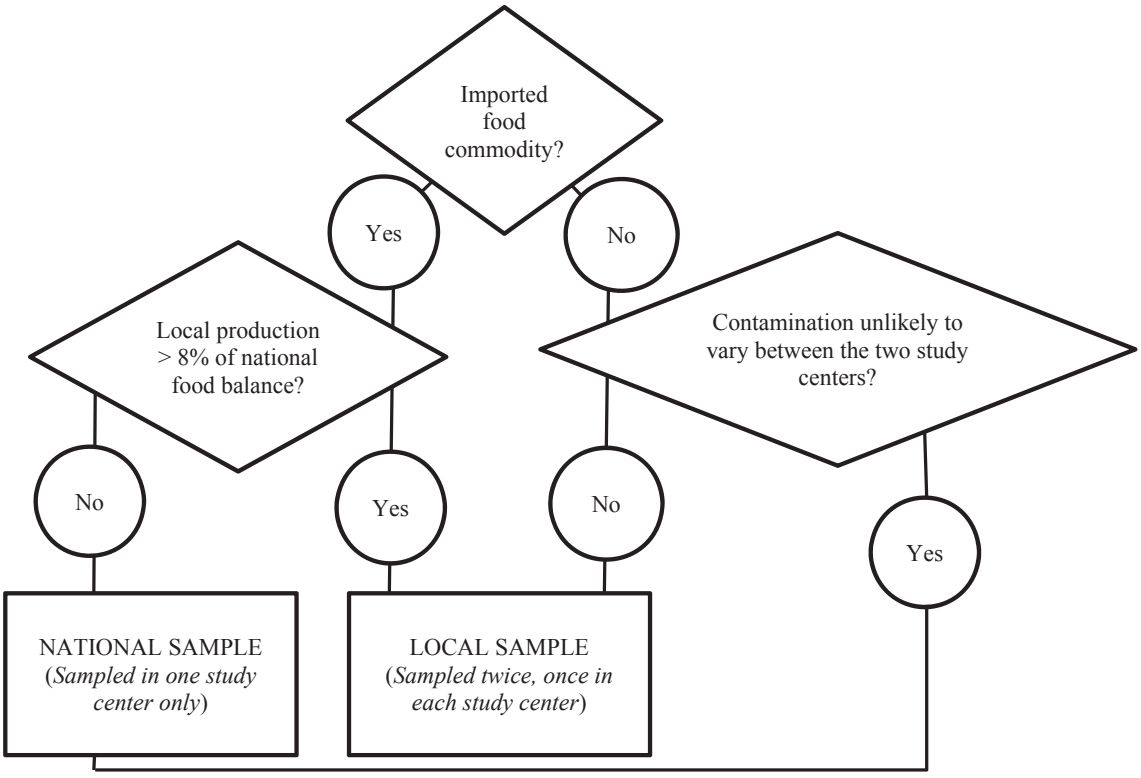

Fig. 2. Decision tree for choosing if a food commodity shall be nationally or locally sampled.

- If the food group average consumption in grams per adult male equivalent per day represents less than 1% of the total average national total diet in weight, then the total average daily consumption of the food subgroups selected from this food group shall represent at least 50% of the food consumption of this food group.

These criteria enable significant coverage of each food group with selected core foods, whilst making sure that the sampling approach focuses on most consumed food commodities. The logic is that, if core foods were also selected within food groups representing less than 1% in weight of the average national diet so as to cover 90% of the food group consumption (same criterion for all food groups), it was estimated that the number of samples needed would increase by 20% whereas the coverage in weight of the average total diet would only be 1% higher, which would not be cost effective.

2.5. Specifications of food composite samples

2.5.1. Inclusion of local specificity of samples

The SSA-TDS terminology slightly differs from the one used in the New Zealand and in the French TDS (Sirot et al., 2009). Instead of using the notion of "*regional food list*" versus "*national food list*", the SSA-TDS addresses the same concern by contrasting "*national sampling*" to "*local sampling*" from one single national food list. The SSA-TDS only includes two study centers per country, with supposedly distinct food consumption patterns and food supply. Because the variance in food chemical concentration pattern for some food commodities is prone to be higher than others, a decision was made as to its local sampling (food commodity sampled in both study centers) or if the food can reasonably be sampled nationally (collected in only one of the two study centers), as described in Fig. 2.

Local sampling is chosen by default in the absence of evidence that the food chemical concentration pattern is unlikely to vary between the two study centers or to impact significantly the resulting dietary exposure assessment. In contrast, a national sampling approach is chosen for food commodities which are mainly (1) imported, (2) industrially processed, or (3) mostly produced and/or consumed in the area where it is collected. In those later cases, although the food chemical concentration pattern is likely to vary from one location to the other, it is unlikely to significantly impact the dietary exposure pattern, which justifies the need of only one sample for both locations.

2.5.2. Food sampling methodology

2.5.2.1. Number of subsamples by composite sample. The SSA-TDS uses the individual food approach with twelve subsamples of the same food commodity per composite sample. The number of 12 subsamples of equal weight, representing each one 12th or slightly more than 8% of any specific composite sample was defined according the statistical basis published from the FP7 research program on TDS exposure (European Commission, 2016b), which was used as a benchmark and replicated in our study. The true standard deviation of concentration and the true mean concentration of a given substance in a food commodity in relation with the number of subsamples collected and pooled per composite sample were investigated. The width of the 95% confidence interval for the estimate of the true mean concentration was summarized, according to the number of pooled subsamples. In a situation where the true SD is unknown, which is the case for our study, hypotheses range from low variability (SD = 30% of true mean concentration) to high variability (SD = 100% of true mean concentration). The pooling of twelve subsamples turns out to be a cost effective approach, with limited impact on the confidence interval of food chemicals

concentration.

The adequate selection of representative subsamples to form individual composites is a major consideration of the sampling plan design (Tsukakoshi, 2011). In TDSs implemented in developed countries, it is common that the allocation of subsamples is proportional to market shares, which often refers to trademarks owned by known operators and clearly identifiable on the shelves of supermarket (Sirot et al., 2009). In developing countries however, this principle remains but needs to be adapted to the local food supply and distribution context. Most food commodities sold at the retail level in Africa, especially locally produced ones, do not bear any distinctive sign, brand, batch number, expiry date, or even a label. Food distribution in the Sub-Saharan Africa countries of this study mostly takes place in daily or weekly markets, involving a large number of ever-changing stakeholders.

2.5.2.2. Subsamples selection criteria. A specific sampling approach is used in the framework of the SSA-TDS that involves 3 major components: (1) the proportion of various origins, from which a given food commodity is imported, in the country where the sampling is taking place (2) the breakdown of the various food items recorder by the national institutes of statistics, at the most detailed level of ranking of the food classification and (3) information collected during field market surveys with regard to the flows of food commodities (Fig. 3).

As far as the sampling is concerned, preference is given to wholesale markets, which enabled the differentiation of the origin of the twelve collected subsamples, and thus to ensure that the intrinsic variability of subsamples is adequately taken into account in the pooled sample. Twelve distinct batches are collected randomly from wholesale markets located in the study center area.

The average national food balance sheet over five years was calculated from the International Trade Center (ITC) database for imported and exported food quantities (ITC, 2016), whereas local food production was extracted from the FAOSTAT database (FAO, 2014b). ITC data also include the origin(s) of food and the proportion of the each source in the food supply, which was reflected in the proportions of subsample of each origin of significant important (i.e. more than 8% or one twelfth of the food commodity supply).

The pooling level chosen for composite samples in this study is the second stratum of the classification pyramid (Fig. 1), consisting of 84 subgroups, including the selected core foods (Table 4). More detailed information available for some food commodities at the bottom stratum of the hierarchy can be exploited to define the proportions of each kind of subsample, which reflect the average behavior of the study populations. Because these data are available for each of the eight study centers enrolled in the project, as well as for the whole national population, these proportions are specific to each location, from where a sample is collected.

Based on the sampling plan, covering more than 90% of the average total diet, each study center was visited and questionnaires were submitted to local market leaders in order to identify (1) the main purchase areas (2) whether market places are wholesale or retail (3) the origins of food commodities (4) main cultivars or varieties (5) the seasonality, and (6) the average price for retail and wholesale. This information was used to define, for each applicable criterion, the breakdown of the twelve subsamples of equal size needed to obtain a representative food composite sample.

2.5.2.3. Seasonality. In order to capture the seasonal variance of food chemical concentration patterns, the SSA-TDS methodology takes into considerations two sampling waves, which are analyzed independently. Seasonal variability may reflect differences in the occurrence of food chemicals during the rainy season and the dry season, or due to different agricultural or post-harvest conditions and practices applied to the food supply throughout various times of the year.

The first sampling campaign includes all 13 food groups (cereals, tubers, legumes, vegetables, fruits, nuts and seeds, meat, eggs, seafood, dairy products, beverages and miscellaneous), and the seven analyte groups (metals and trace elements, mycotoxins, pesticides residues, polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCB and PCDD/F), perfluoroalkoxy alkanes (PFAs), and brominated flame retardants (BFRs)). Tap water is also collected as a representative composite sample in each study center.

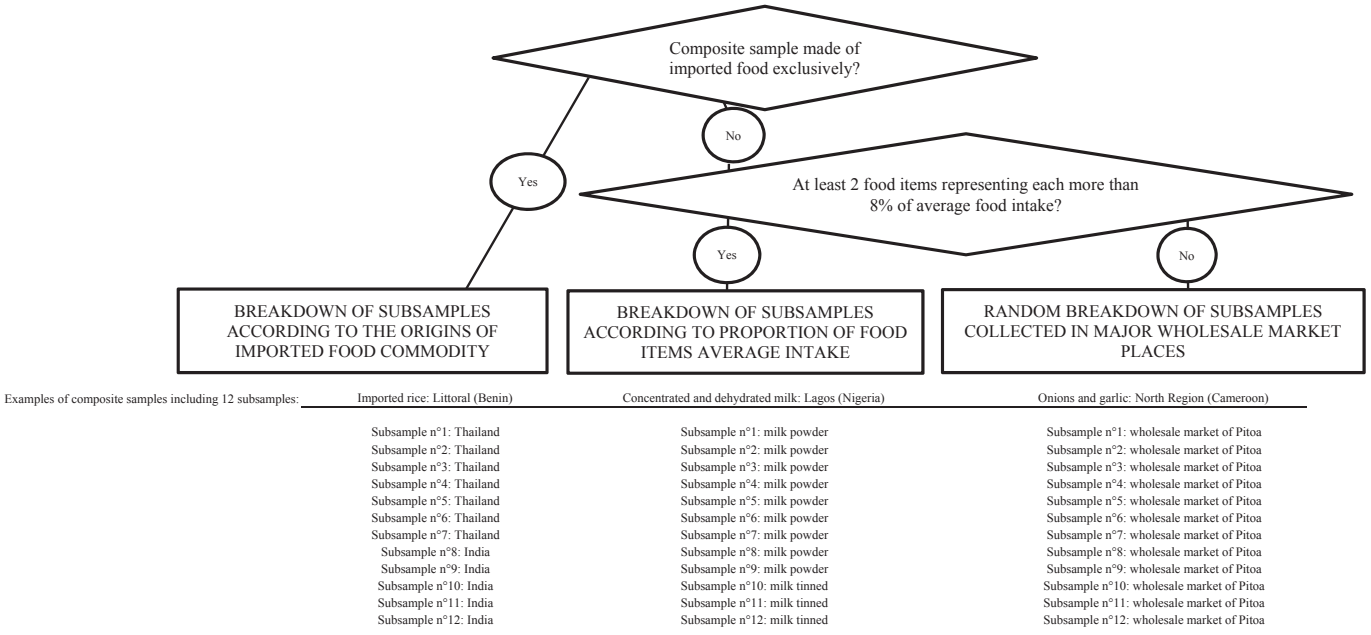

Fig. 3. Decision tree and breakdown of subsamples in composite samples.

Table 3
Analytical grid describing in each food group the selected core foods (matrices) tested against the seven analyte groups.

	Analyte groups	Minerals and Trace Elements	Mycotoxins	Pesticides residues	Polycyclic Aromatic Hydrocarbons	Polychlorinated Biphenyls and Dioxins, Perfluoroalkoxy alkanes, Brominated flame retardants
Food groups	Cereals	All matrices	All matrices	All matrices	Not tested	Not tested
	Tubers	All matrices	Dried tubers	All matrices	Dried tubers	Not tested
	Legumes	All matrices	All matrices	All matrices	Not tested	Not tested
	Vegetables	All matrices	Onion and Garlic	All matrices	Not tested	Not tested
	Fruits	All matrices	Not tested	All matrices	Not tested	Not tested
	Nuts & Seeds	All matrices	All matrices	All matrices	Not tested	Not tested
	Meat	All matrices	All matrices	All matrices	Not tested	All matrices
	Eggs	All matrices	All matrices	All matrices	Not tested	All matrices
	Seafood	All matrices	Smoked fish	All matrices	Smoked fish	All matrices
	Dairy	All matrices	All matrices	All matrices	Concentrated and dehydrated milk	All matrices
	Oil & Fat	All matrices	All matrices	All matrices	Oil and fat	All matrices
	Beverages	All matrices	Traditional & fermented drinks	All matrices	Not tested	Not tested
	Miscellaneous	All matrices	All but salt	All matrices	Peper, coffee, chocolate and broth	Coffee, chocolate and broth

Due to limited resources, the second sampling campaign focuses on core foods included in 5 main food groups, (cereals, tubers, legumes, vegetables and fruits). Tap water is also collected as a representative composite sample in each study center during the second wave. The second sampling campaign enables the screening of two analyte groups (mycotoxins and pesticides residues), the concentration of which is likely to vary due to agricultural practices, climatic and post-harvest conditions, which differ through the various times of the year.

2.5.3. Preparation of food as consumed

Subsamples are collected and prepared individually according to recipe books (Vinakpon-Gbaguidi, 2003; Nya-Njike, 1998; Gautier and Mallet, 2006; Madubike, 2013) , using inert kitchen utensils. These references are considered as representative of the diet of the study populations and were therefore selected by the national competent authorities. By the expression “prepared individually” it is meant that no salt, oil, nor spices are added to the composite samples, and that, unlike in real situations, core foods from different food subgroups are not mixed together. These recipe books allow the identification of the processes used in the preparation of the foods, especially cooking time and temperature. However, actual recipes are not prepared, as each composite sample only contains one core food or ingredient. The inedible parts are removed at the preparation stage, as a typical consumer would do. Distilled water is used to prepare food as consumed, instead of tap water, which is also part of the sampling. The quantity of water added during the cooking process of each of the 12 subsamples is measured by weighting the food at each stage of the process.

2.5.4. Core analyte list

The seven analyte groups included in this study are pesticides, metals and trace elements, mycotoxins, polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls, polychlorinated dibenzo-p-dioxins and dibenzofurans (PCB and PCDD/F) perfluoroalkoxy alkanes (PFAs) and brominated flame retardants (BFRs). These groups are described in the results section of this paper, as a core food chemicals list.

The choice of analyte groups was made by the national stakeholders (e.g. 30–50 food safety professionals per country), and was discussed with a scientific committee, without applying the methodology proposed by the EU TDS Exposure project (Papadopoulos et al., 2015).

2.5.5. Food chemical analysis

Analytical performance (LOD/LOQ) often becomes a limiting factor for risk assessors. In order to avoid as much as possible uncertainties in dietary exposure that could result from the inclusion of censored data with high analytical limits, special care was taken in the selection of testing laboratories so as to reach adequate limits of quantification for risk assessment purposes. In order to help laboratories to achieve this, calculations were made to assess satisfactory analytical limits. When a substance is not detected or cannot be quantified, two scenarios (upper bound and lower bound), based on the value of analytical limits, were used to estimate exposure. In the upper-bound scenario (UB), all results below analytical limit are considered to be equal to the analytical limit. In this TDS, as in the French infant TDS (Hulin et al., 2014), we estimated the maximum value of analytical limits of quantification (LOQ), so as to obtain a total UB exposure value of no more than 30% of the reference point or point of departure (e.g. acceptable daily intake (ADI) or tolerable daily intake (TDI)) in the case of non-detection or quantification of the substances looked for in all tested samples.

For margin of exposure (MOE) calculations (e.g for 13 PAHs, PCBs congeners and some mycotoxins), the harmonized approach for risk assessment of substances, which are both genotoxic and carcinogen established by WHO and EFSA applies (WHO, 2006; EFSA, 2005). The approach consists of verifying that analytical limits corresponds to a total UB exposure with a MOE above 10,000 compared to the applicable level of toxicological significance, and therefore associated with low levels of concern. For each selected substance, laboratories were required to reach targeted LOQs. All analytical limits, as well as which limit was used for the UB scenario (LOD or LOQ) targeted prior to analysis and actually reached by laboratories will be described in future articles dealing with the analytical results of this TDS.

The type of analysis performed on each composite sample primarily depends on the likelihood of finding food chemicals in the matrix. The analytical grid represented in Table 3 describes which matrices are tested against which analyte groups.

2.5.6. Data analysis

Households’ food purchase data, food prices, edible fraction conversion factors, yield factors, and energy content were processed with the SPSS 18.0 (IBM) software. The *t*-test for comparing mean consumption of cereals was completed with XLSTAT (AddinSoft).

Table 4
Selection of core foods from food subgroups on the basis of mean national daily consumption in grams per adult male equivalent.

Food group	Code	Food subgroup	BENIN				CAMEROON				MALI				NIGERIA			
			% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study	% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study	% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study	% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study
CEREALS	1.1	RICE	80	133.8	445.7	7.3	78	221.0	735.5	11	98	573.5	1419.2	33	76	211.7	860.9	13
	1.2	MAIZE	99	740.7	1709.3	41	69	384.9	1808.5	20	51	139.5	821.2	7.9	46	223.7	1452.7	13
	1.3	WHEAT/BREAD	35	14.1	152.2	NS	74	43.6	157.6	2.2	82	26.7	114.9	1.5	58	15.7	100.8	0.9
	1.4	PASTA	27	28.9	283.7	1.6	17	8.3	136.4	NS	31	7.7	89.2	NS	12	3.6	98.8	NS
	1.5	SORGHUM	13	53.3	896.1	2.9	12	122.8	2294.5	6.3	51	228.7	1270.4	13	44	250.4	1364.2	15
	1.6	MILLET	6	21.5	691.4	1.2	3	9.1	1291.5	NS	74	411.7	1525.6	23.5	34	181.2	1345.7	11
	1.7	OTHER CEREALS	14	12.6	253.3	NS	22	9.8	160.3	NS	20	15.6	433.6	NS	9	18.4	663.1	NS
TUBERS	2.1	CASSAVA FRESH	19	17.3	262.8	0.9	61	109.7	615.6	5.6	22	1.9	35.1	0.1	20	28.8	565.0	1.7
	2.2	CASSAVA DRY	69	206.8	799.3	11	37	83.3	927.3	4.3	23	8.7	121.7	0.5	50	268.0	1614.5	16
	2.3	YAM FRESH	46	78.8	490.8	4.3	32	27.8	264.7	1.4	34	2.8	25.3	0.2	50	84.4	531.9	5.0
	2.4	YAM DRY	6	8.2	394.8	NS	NR				0	0.03	.	NS	9	8.6	344.5	NS
	2.5	POTATO FRESH	2	0.5	78.3	NS	23	23.4	289.3	1.2	39	3.9	31.9	0.2	6	3.0	203.9	NS
	2.6	POTATO DRY	NR				1	2.2	2106.3	NS	NR				NR			
	2.7	SWEET POTATO	8	5.7	186.8	NS	41	47.7	332.0	2.4	68	12.7	54.4	0.7	14	6.2	179.8	NS
	2.8	COCOYAM	NR				12	21.5	484.5	1.1	2	0.2	37.7	NS	15	8.9	222.6	0.5
	2.9	MACABO	NR				51	67.8	378.0	3.5	NR				NR			
	2.10	OTHER TUBERS	NR				1	0.8	483.9	NS	16	3.03	57.30	NS	3	3.7	446.6	NS
LEGUMES	3.1	BEANS	67	88.0	338.8	4.8	71	81.4	349.9	4.2	72	27.2	104.9	1.5	68	71.9	384.1	4.3
	3.2	GROUNDNUTS	24	3.0	36.2	NS	85	36.4	123.4	1.9	89	18.8	57.5	1.1	33	7.0	101.7	0.4
	3.3	PEAS	NR				2	1.6	212.8	NS	9	1.5	87.6	NS	13	40.0	864.0	2.4
	3.4	SOJA	NR				2	1.0	137.2	NS	NR				4	3.4	331.3	NS
	3.5	OTHER LEGUMES	NR				5	0.3	17.8	NS	5	0.5	28.9	NS	8	5.0	306.7	NS
VEGETABLES	4.1	TOMATO	89	81.9	305.5	4.5	75	31.7	110.7	1.6	89	16.1	53.7	0.9	72	21.4	106.8	1.3
	4.2	CARROTS	2	0.1	12.7	NS	11	1.5	43.7	NS	17	0.2	3.7	NS	NR			
	4.3	GREEN LEAVES	37	3.6	32.5	NS	73	78.3	287.0	4.0	64	4.4	20.1	0.2	6	0.6	32.9	NS
	4.4	COURGETTES, CUCUMBER & GROUND PEPER	8	0.8	28.4	NS	23	1.3	18.4	NS	57	5.5	27.4	0.3	11	1.2	36.9	NS
	4.5	CABBAGE	1	0.1	20.1	NS	10	5.0	126.3	NS	52	2.0	10.8	0.1	3	0.3	41.1	NS
	4.6	ONION & GARLIC	80	12.8	45.5	0.7	73	8.0	32.6	0.4	91	9.6	33.2	0.5	74	8.0	33.9	0.5
	4.7	OKRO = GOMBO	39	4.7	35.3	0.3	62	9.7	46.7	0.5	70	3.3	12.6	0.2	59	19.8	132.8	1.2
	4.8	PARSLEY, CELERY, BASIL & LEEK	NR				49	3.6	22.3	NS	NR				NR			
	4.9	OTHER VEGETABLES	39	4.7	35.7	NS	21	1.4	23.0	NS	56	2.6	14.7	NS	57	15.1	122.6	0.9
FRUITS	5.1	BANANA	6	0.7	33.5	NS	48	69.0	461.6	3.5	47	3.1	25.0	0.2	14	1.9	65.1	0.1
	5.2	PLANTAIN	3	0.7	58.3	NS	52	73.8	416.8	3.8	27	2.4	30.8	0.1	18	14.3	284.0	0.9
	5.3	MANGO	0	0.0	19.9	NS	7	1.2	48.1	NS	79	11.7	46.9	0.7	3	0.9	162.0	NS
	5.4	PINEAPPLE	4	1.3	101.4	NS	8	1.6	65.4	NS	3	0.1	13.4	NS	4	0.6	56.3	NS
	5.5	CITRUS (ORANGE, LEMON, LIME…)	12	3.7	98.1	0.2	30	8.3	87.0	0.4	63	2.9	15.4	0.2	19	3.5	71.3	0.2
	5.6	AVOCADO	1	0.1	36.0	NS	17	2.5	45.0	NS	7	0.3	9.6	NS	2	0.5	125.8	NS
	5.7	PAWPAW	2	0.5	75.3	NS	8	3.1	120.5	NS	20	0.7	10.9	0.04	6	1.7	114.1	0.1
	5.8	MELON / WATERMELON	0	0.1	68.6	NS	5	1.9	107.2	NS	57	19.0	104.8	1.1	6	0.9	73.5	0.1
	5.9	OTHER FRUITS	1	0.1	37.9	NS	28	2.3	26.7	NS	77	4.1	15.1	NS	0	0.0	36.9	NS

(*continued on next page*)

Table 4 (*continued*)

Food group	Code	Food subgroup	BENIN				CAMEROON				MALI				NIGERIA			
			% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study	% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study	% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study	% Consumers	Mean daily consumption (g/AME/day)	P95 consumers only (g/AME/day)	% Total Diet covered by study
NUTS & SEEDS	6.1	COCONUT	2	0.2	34.3	NS	2	0.3	52.1	NS	4	0.1	5.2	NS	4	0.7	79.5	NS
	6.2	CASHEW NET	0.1	0.01	.	NS	0.05	0.002	.	NS	NR				0	0.1	143.7	NS
	6.3	PALM NUT	16	0.8	14.3	0.05	11	3.0	76.9	0.2	NR				3	0.3	49.6	0.02
	6.4	OTHER NUTS/ SEEDS	10	0.5	12.7	NS	39	2.7	23.2	NS	4	0.5	38.7	0.03	29	3.8	55.2	NS
MEAT	7.1	BEEF	16	3.3	59.1	0.2	43	8.3	55.0	0.4	82	10.4	45.7	0.6	55	7.8	48.1	0.5
	7.2	POULTRY	7	0.9	37.6	NS	19	1.6	34.1	NS	63	1.8	9.7	NS	5	1.0	58.7	NS
	7.3	MUTTON/GOAT	5	0.5	29.9	NS	8	1.3	65.1	NS	60	2.9	15.7	NS	15	1.5	35.1	NS
	7.4	PORK	2	0.2	23.9	NS	4	0.5	30.4	NS	0	0.004	.	NS	1	0.2	35.3	NS
	7.5	PROCESSED MEAT	3	1.5	144.3	NS	3	0.2	23.2	NS	14	0.7	18.9	NS	0	0.0	113.0	NS
	7.6	GAME MEAT	NR				9	1.3	51.5	NS	3	0.05	7.1	NS	3	0.4	61.2	NS
	7.7	INSECTS	NR				1	0.1	23.3	NS	NR				NR			
	7.8	OTHER MEAT	2	0.2	23.8	NS	1	0.1	28.1	NS	3	0.1	8.4	NS	2	0.1	23.0	NS
EGGS	8.1	POULTRY EGGS	14	1.4	28.8	0.1	25	3.3	42.9	0.2	24	16.9	263.2	1.0	10	1.6	69.0	0.1
SEAFOOD	9.1	SEA FISH	31	4.9	43.4	NS	67	15.2	63.6	0.8	72	3.1	14.3	NS	65	10.6	57.8	0.6
	9.2	FRESH WATER FISH	NR				12	1.8	48.6	0.1	20	0.4	5.8	NS	NR			
	9.3	SMOKED FISH	80	9.0	32.3	0.5	62	4.9	21.8	0.2	72	6.4	22.2	0.4	15	1.9	45.9	NS
	9.4	PROCESSED FISH	6	2.1	101.5	NS	11	0.7	19.8	NS	NR				1	0.1	34.6	NS
	9.5	CRUSTACEANS/ MOLLUSCS	8	0.2	9.2	NS	28	0.7	7.3	NS	0.1	0.0001	.	NS	23	0.9	15.0	NS
	9.6	OTHER SEAFOOD	NR				2	0.1	8.4	NS	6	0.3	15.4	NS	NR			
DAIRY	10.1	FRESH/ FERMENTED MILK	11	5.1	192.1	0.3	14	2.2	57.6	NS	70	7.2	37.0	0.4	11	2.5	89.0	NS
	10.2	CONCENTRATED/ DEHYDRATED MILK	8	4.0	184.0	0.2	20	5.3	97.3	0.3	63	11.6	64.5	0.7	22	8.0	123.2	0.5
	10.3	OTHER DAIRY PRODUCTS	14	2.4	47.4	NS	3	0.3	37.0	NS	5	0.3	11.7	NS	5	0.9	66.7	NS
OIL & FAT	11.1	PALM OIL	64	17.5	70.0	1.0	66	28.3	105.8	1.4	22	1.0	13.3	NS	74	16.9	73.7	1.0
	11.2	GROUNDNUT OIL	59	15.5	66.5	0.8	15	3.1	70.0	NS	56	8.6	41.6	0.5	32	4.7	52.1	0.3
	11.3	OTHER VEGETAL OIL	15	1.6	27.2	NS	34	8.8	70.8	0.5	71	6.3	28.4	0.4	16	3.1	67.1	0.2
	11.4	OTHER FAT/OIL	10	0.9	27.4	NS	22	0.8	15.3	NS	13	2.8	137.3	0.2	5	0.7	60.4	NS
BEVERAGES	12.1	WATER	15	67.2	1473.2	3.7	14	10.9	302.7	0.6	6	2.3	179.3	0.1	13	5.6	198.5	0.3
	12.2	FRUIT JUICE	NR				3	0.6	71.3	NS	34	1.2	13.6	NS	2	0.6	116.2	NS
	12.3	TRADITIONAL SOFT DRINK	7	5.3	295.2	0.3	10	3.7	128.2	NS	NR				15	6.1	179.7	0.4
	12.4	TRADITIONAL FERMENTED DRINK	2	0.4	111.9	NS	24	31.8	473.1	1.6	NR				5	2.4	182.3	0.1
	12.5	INDUSTRIAL FERMENTED DRINK	20	7.2	135.4	0.4	72	38.3	223.7	2.0	NR				6	2.0	127.6	0.1
	12.6	INDUSTRIAL SOFT DRINK	9	4.2	164.6	NS	29	6.7	92.4	0.3	21	2.6	42.4	0.2	19	6.5	137.5	0.4
	12.7	SPIRITS	8	1.7	79.1	NS	3	0.3	37.3	NS	NR				2	0.2	44.7	NS
	12.8	OTHER DRINKS	NR				4	0.6	51.2	NS	NR				5	0.8	88.7	NS

MISCELLANEOUS	13.1	SUGAR	36	5.8	52.0	NS	51	10.6	70.0	0.5	99	50.5	115.5	2.9	53	6.0	37.6	0.4		
	13.2	SALT	77	9.1	31.7	0.5	69	13.0	48.4	0.7	98	14.1	33.5	0.8	59	6.9	33.8	0.4		
	13.3	BROTH/ BOUILLON CUBE	87	5.7	18.3	NS	88	2.8	7.9	NS	90	3.0	7.4	NS	74	9.7	49.6	0.6		
	13.4	HONEY	0.4	0.02	21.2	NS	2	0.1	17.8	NS	0	0.0	8.5	NS	1	0.3	87.5	NS		
	13.5	TEA	2	0.7	107.6	NS	10	0.2	5.7	NS	72	1.4	5.3	NS	12	0.7	22.5	NS		
	13.6	COFFEE	3	0.6	72.0	NS	8	0.0	1.4	NS	60	0.8	4.2	NS	0	0.0	46.2	NS		
	13.7	CHOCOLATE	2	0.3	66.1	NS	22	0.7	11.7	NS	4	0.1	9.0	NS	10	1.2	47.5	NS		
	13.8	BABY MILK POWDER	1	0.2	69.2	NS	1	0.4	150.3	NS	1	0.2	55.6	NS	1	0.1	46.5	NS		
	13.9	CHILI PEPER	96	12.6	42.5	0.7	59	2.7	12.5	NS	97	15.8	56.0	NS	80	11.7	53.9	0.7		
	13.10	OTHER MISCELLANEOUS	98	106.0	417.0	5.8	98	127.9	628.8	6.5	81	8.8	39.2	0.5	65	13.1	90.3	0.8		
		TOTAL	100	1829	3373	94.9	100	1955	3350	95.6	100	1755	2910	96.9	100	1676	3146	95.5		

NS: non-selected food subgroup.
NR: non-recorded food subgroup (national statistics).

3. Results and discussion

3.1. Food consumption data

Estimates of daily consumption were calculated for each province or city of the four countries, for (1) the mean consumption based on the whole selected population in grams per adult male equivalent per day and for (2) consumption of high-level consumers, defined as those at the 95th percentile (P95) of consumers. The estimates are also expressed in grams per adult male equivalent per day and at the three strata of the food classification (food groups, food subgroups including the selected core foods and food items) described in Fig. 1.

Table 4 shows the mean national daily consumption data of populations for the 4 countries of the study presented at the second stratum of the SSA-TDS food classification pyramid (Fig. 1), consisting 84 food subgroups, among which core foods are selected.

By far, the most consumed food commodities (in mean weight per adult male equivalent per day) in each country are starchy products: maize (Benin: 740,7, Cameroon: 384,9, Mali: 139,5 and Nigeria: 223,7 g/AME/day), rice (Benin: 133,8, Cameroon: 221,0, Mali: 573,5 and Nigeria: 211,7 g/AME/day), sorghum (Benin: 53,3, Cameroon: 122,8, Mali: 228,7 and Nigeria: 250,4 g/AME/day), millet (Benin: 21,5, Cameroon: 9,1, Mali: 411,7 and Nigeria: 181,2 g/AME/day) and cassava dry (Benin: 206,8, Cameroon: 83,3, Mali: 8,7 and Nigeria: 268,0 g/AME/day).

The variety of core foods recorded (from 70 for Benin to 83 for Cameroon) is suitable for exposure assessment, as the sampled core foods enable the identification of the main contributors to the mean dietary exposure to food chemicals in each study center. The mean daily food consumption of core foods of the 8 study centers are displayed in Table 5.

3.2. Food consumption patterns

Dietary tendencies or patterns are better shown at the top stratum and the most aggregated level of the SSA-TDS food classification pyramid (Fig. 1), which consists of 13 food groups.

Staple foods (cereals and tubers) in particular represent the major part of the average total diet.

The dietary exposures for the various study populations may vary significantly within the same country, as a consequence of specific consumer behaviors, regardless of the concentration of substances of public health interest the diet For example, out of 8 study centers, 3 are located in coastal areas (Duala, the Littoral of Benin and Lagos) and 5 in non-coastal areas (Bamako, the Borgou region of Benin, Kano, the North of Cameroon and Sikasso), which can be associated with distinct food consumption patterns. In our study, populations located in densely populated coastal areas consume in average 597 g/AME/day of cereals, whereas the average daily consumption of populations located in non-coastal areas is 1247 g/AME/day (Table 6), which is significantly different (p-value: 0,001).

The next steps of the implementation of this study will enable the identification of specific exposure patterns effectively associated with the food consumption patterns described above.

3.3. Core food list

The core foods are identified and selected among food subgroups at the second stratum of the SSA-TDS food classification (Table 4). As it is the case for the 13 food groups defining the top stratum of the hierarchy, subgroups of the second level are harmonized among all study centers. In the adopted strategy, this stratum defines the aggregation level fit for producing food

Table 5
Study centers mean daily consumptions of food subgroups in grams per adult male equivalent per day.

Food group	Code	Food subgroup	BENIN				CAMEROON				MALI				NIGERIA			
			Littoral		Borgou		Duala		North		Bamako		Sikasso		Lagos		Kano	
			Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study
CEREALS	1.1	RICE	136	7	79	4	203	13	170	8	803	42	266	18	321	20	230	15
	1.2	MAIZE	576	29	766	41	124	8	922	44	121	6	419	29	80	5	242	16
	1.3	WHEAT/BREAD	42	NS	7.0	NS	77	5	42	2	50	3	9.2	1	55	3	21	1
	1.4	PASTA	79	4	26	1	18	NS	3.6	NS	10	NS	6.3	NS	23	NS	2.7	NS
	1.5	SORGHUM	0.8	0.04	186	10	0.3	0.02	243	12	127	7	213	15	1.7	0.1	448	29
	1.6	MILLET	1.5	0.1	26	1	0.4	NS	1.4	NS	191	10	253	17	3.6	0.2	255	17
	1.7	OTHER CEREALS	13.6	NS	4.2	NS	14	NS	27	NS	8.7	NS	4.9	NS	19	NS	51	NS
TUBERS	2.1	CASSAVA FRESH	3.5	0.2	22	1	66	4	15	1	0.7	0.04	1.4	0.1	7.0	0.4	6.8	0.4
	2.2	CASSAVA DRY	156	8	66	4	48	3	13	1	24	1	13	1	367	22	12	1
	2.3	YAM FRESH	51	3	222	12	28	2	7.5	0.4	5.1	0.3	3.1	0.2	157	10	14	1
	2.4	YAM DRY	12	NS	29	NS	NR				44.3%	NS	0.01	NS	24	1	0.03	0.002
	2.5	POTATO FRESH	2.7	NS	0.04	NS	29	2	1.1	0.1	9.6	1	3.3	0.2	6.3	NS	3.1	NS
	2.6	POTATO DRY	NR				0.8	NS	5.3	NS	NR				NR			
	2.7	SWEET POTATO	2.1	NS	0.4	NS	30	2	45	2	12	1	14	1	5.1	NS	7.3	NS
	2.8	COCOYAM	NR				16	1	2.6	0.1	0.1	NS	0.7	NS	3.5	0.2	0.6	0.04
	2.9	MACABO	NR				48	3	4.4	0.2	NR				NR			
	2.10	OTHER TUBERS	NR				0.000	NS	0.2	NS	1.3	NS	6.3	NS	0.1	NS	0.000	NS
LEGUMES	3.1	BEANS	48	2	41	2	58	4	71	3	25	1	29	2	141	9	53	3
	3.2	GROUNDNUTS	1.7	NS	2.2	NS	30	2	74	4	20	1	17	1	1.5	0.1	7.0	0.5
	3.3	PEAS	NR				0.8	NS	4.3	NS	4.0	NS	2.2	NS	3.8	0.2	0.6	0.04
	3.4	SOJA	NR				0.2	NS	0.2	NS	NR				0.5	NS	3.0	NS
	3.5	OTHER LEGUMES	NR				0.1	NS	0.8	NS	0.1	NS	2.8	NS	2.2	NS	1.0	NS
VEGETABLES	4.1	TOMATO	160	8	76	4	36	2	16	1	30	2	11	1	46	3	17	1
	4.2	CARROTS	0.6	NS	0.04	NS	3.0	NS	0.03	NS	0.6	NS	0.1	NS	NR			
	4.3	GREEN LEAVES	9.1	NS	0.4	NS	43	3	88	4	6.7	0.4	4.7	0.3	0.2	NS	0.1	NS
	4.4	COURGETTES, CUCUMBER & GROUND PEPER	1.4	NS	1.7	NS	1.5	NS	0.9	NS	12	1	5.8	0.4	0.4	NS	0.3	NS
	4.5	CABBAGE	0.3	NS	0.1	NS	4.6	NS	0.1	NS	3.9	0.2	1.3	0.1	0.3	NS	0.1	NS
	4.6	ONION & GARLIC	21	1	7.8	0.4	9.6	1	6.5	0.3	19	1	6.5	0.4	11	1	7.9	1
	4.7	OKRO = GOMBO	4.5	0.2	2.5	0.1	3.0	0.2	18	1	2.7	0.1	4.1	0.3	4.7	0.3	19	1
	4.8	PARSLEY, CELERY, BASIL & LEEK	NR				7.7	NS	0.3	NS	NR				NR			
	4.9	OTHER VEGETABLES	7.3	NS	0.6	NS	2.0	NS	3.1	NS	5.5	NS	1.8	NS	14	1	5.3	0.3
FRUITS	5.1	BANANA	1.6	NS	0.4	NS	93	6	3.0	0.1	11	1	2.2	0.2	0.9	0.1	1.4	0.1
	5.2	PLANTAIN	2.5	NS	0.1	NS	57	4	2.3	0.1	8.2	0.4	1.3	0.1	31	2	0.6	0.04
	5.3	MANGO	0.05	NS	0.01	NS	0.2	NS	0.7	NS	20	1	15	1	0.5	NS	0.4	NS
	5.4	PINEAPPLE	7.6	NS	0.01	NS	3.3	NS	0.0	NS	0.3	NS	0.2	NS	1.7	NS	0.04	NS
	5.5	CITRUS (ORANGE, LEMON, LIME…)	10	1	1.3	0.1	15	1	2.6	0.1	7.7	0.4	2.3	0.2	8.1	0.5	4.2	0.3
	5.6	AVOCADO	0.7	NS	0.04	NS	1.8	NS	0.5	NS	0.9	NS	0.4	NS	0.3	NS	0.1	NS
	5.7	PAWPAW	1.3	NS	0.20	NS	5.5	NS	0.2	NS	1.6	0.1	1.2	0.1	1.0	0.1	0.1	0.01
	5.8	MELON / WATERMELON	0.2	NS	0.000	NS	1.1	NS	7.0	NS	39	2	15	1	2.9	0.2	0.5	0.03
	5.9	OTHER FRUITS	0.5	NS	0.002	NS	1.0	NS	2.0	NS	4.5	NS	4.8	NS	0.0	NS	0.04	NS

NUTS & SEEDS	6.1	COCONUT	0.4		NS		0.1		NS		0.2		NS		0.000		NS		0.1		NS	
	6.2	CASHEW NET	0.01		NS		0.05		NS		0.000		NS		0.000		NS		NR			
	6.3	PALM NUT	1.7		0.1		0.1		0.003		0.8		0.05		0.01		0.0005		NR			
	6.4	OTHER NUTS/ SEEDS	0.4		NS		0.9		NS		2.1		NS		1.0		NS		0.1		0.01	
MEAT	7.1	BEEF	2.3		0.1		18		1		6.9		0.4		14		1		21		1	
	7.2	POULTRY	3.5		NS		0.3		NS		1.6		NS		1.7		NS		3.9		NS	
	7.3	MUTTON/GOAT	0.9		NS		0.7		NS		0.04		NS		4.7		NS		1.4		NS	
	7.4	PORK	0.2		NS		0.03		NS		0.6		NS		0.7		NS		0.01		NS	
	7.5	PROCESSED MEAT	5.5		NS		2.2		NS		0.7		NS		0.001		NS		2.6		NS	
	7.6	GAME MEAT	NR								0.2		NS		0.5		NS		0.1		NS	
	7.7	INSECTS	NR								0.000		NS		0.000		NS		NR			
	7.8	OTHER MEAT	0.4		NS		0.2		NS		0.02		NS		0.000		NS		0.05		NS	
EGGS	8.1	POULTRY EGGS	5.4		0.3		0.3		0.02		6.5		0.4		0.5		0.02		64.4		3.4	
SEAFOOD	9.1	SEA FISH	13.1		NS		3.0		NS		25		1.6		3.4		0.2		6.0		NS	
	9.2	FRESH WATER FISH			NR						0.4		0.0		6.8		0.3		0.1		NS	
	9.3	SMOKED FISH	9.0		0.4		3.2		0.2		5.0		0.3		2.8		0.1		7.1		0.4	
	9.4	PROCESSED FISH	9.4		NS		1.4		NS		1.6		NS		0.1		NS				NR	
	9.5	CRUSTACEANS/ MOLLUSCS	1.5		NS		0.02		NS		0.7		NS		0.02		NS		0.000		NS	
	9.6	OTHER SEAFOOD			NR						0.1		NS		0.02		NS		0.7		NS	
DAIRY	10.1	FRESH/ FERMENTED MILK	28		1.4		4.0		0.2		4.8		NS		1.9		NS		10		0.5	
	10.2	CONCENTRATED/ DEHYDRATED MILK	16		0.8		1.7		0.1		7.9		0.5		2.6		0.1		21		1.1	
	10.3	OTHER DAIRY PRODUCTS	1.8		NS		8.2		NS		0.4		NS		0.01		NS		0.3		NS	
OIL & FAT	11.1	PALM OIL	9.2		0.5		1.4		0.1		31		2.0		0.3		0.01		1.9		NS	
	11.2	GROUNDNUT OIL	26		1.3		10		0.6		3.0		NS		6.1		NS		11		0.6	
	11.3	OTHER VEGETAL OIL	1.5		NS		4.4		NS		7.8		0.5		13		0.6		5.1		0.3	
	11.4	OTHER FAT/OIL	1.7		NS		1.8		NS		2.3		NS		0.2		NS		2.6		0.1	
BEVERAGES	12.1	WATER	80		4.0		99		5.3		51		3.2		9.2		0.4		4.3		0.2	
	12.2	FRUIT JUICE			NR						2.2		NS		0.7		NS		3.7		NS	
	12.3	TRADITIONAL SOFT DRINK	39		1.9		6.1		0.3		8.4		NS		11		NS				NR	
	12.4	TRADITIONAL FERMENTED DRINK	0.2		NS		0.000		NS		7.4		0.5		47		2.2				NR	
	12.5	INDUSTRIAL FERMENTED DRINK	13		0.7		9.0		0.5		56		3.6		13		0.6				NR	
	12.6	INDUSTRIAL SOFT DRINK	16		NS		2.6		NS		13		0.8		2.3		0.1		6.7		0.4	
	12.7	SPIRITS	0.6		NS		2.6		NS		0.2		NS		0.01		NS				NR	
	12.8	OTHER DRINKS			NR						1.4		NS		0.5		NS				NR	

(*continued on next page*)

Table 5 (*continued*)

Food group	Code	Food subgroup	BENIN				CAMEROON				MALI				NIGERIA							
			Littoral		Borgou		Duala		North		Bamako		Sikasso		Lagos		Kano					
			Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study	Mean daily consumption (g/AME/day)	% Total Diet covered by study				
MISCELLANEOUS	13.1	SUGAR	8.5	NS	5.0	NS	7.8	0.5	27	1.3	53	2.8	34	2.3	5.7	0.3	8.7	0.6				
	13.2	SALT	2.5	0.1	7.1	0.4	12	0.8	10	0.5	9.6	0.5	11	0.7	2.3	0.1	8.7	0.6				
	13.3	BROTH/ BOUILLON CUBE	6.4	NS	4.0	NS	2.9	NS	2.7	NS	3.8	NS	2.3	NS	5.5	0.3	8.6	0.6				
	13.4	HONEY	0.1	NS	0.03	NS	0.1	NS	0.1	NS	0.001	NS	0.001	NS	0.04	NS	0.1	NS				
	13.5	TEA	1.7	NS	1.0	NS	0.1	NS	0.8	NS	1.5	NS	0.7	NS	2.4	NS	2.0	NS				
	13.6	COFFEE	2.2	NS	0.2	NS	0.03	NS	0.1	NS	1.1	NS	0.7	NS	0.02	NS	0.000	NS				
	13.7	CHOCOLATE	0.7	NS	0.05	NS	1.2	NS	0.04	NS	0.3	NS	0.1	NS	3.2	NS	0.3	NS				
	13.8	BABY MILK POWDER	1.1	NS	0.04	NS	0.8	NS	0.1	NS	0.5	NS	0.2	NS	0.5	NS	0.000	NS				
	13.9	CHILI PEPER	17	0.8	4.6	0.2	3.4	NS	0.8	NS	46	NS	8.7	NS	30	1.8	11	0.7				
	13.10	OTHER MISCELLANEOUS	335	16.6	104	5.6	204	13.0	111	5.3	16	0.8	6.6	0.5	20	1.2	7.8	0.5				
		TOTAL	**2017**	**90.8**	**1874**	**95.5**	**1563**	**93.3**	**2099**	**95.7**	**1893**	**94.2**	**1457**	**96.6**	**1645**	**94.8**	**1524**	**94.8**				

NS: non-selected food subgroup.
NR: non-recorded food subgroup (national statistics).

composite samples. Therefore, it is defined on the basis of the homogeneity of its subcomponents (ex: rice, maize …) and takes into consideration a number of processes, likely to impact contamination levels (ex: dried cassava, smoked fish …). In order to target core foods which contribute the most to the diet, each food group includes one specific subgroup, which gathers poorly defined or rare food commodities (ex: other cereals, other tubers …). The strategy adopted includes avoiding, when possible, these poorly defined food subgroups, the sampling of which would not be pertinent, because of the lack of information with regard to the actual nature of these food subgroups. Criteria defining the proportions of the food supply (e.g. type, variety, origin) are reflected in the breakdown of the twelve subsamples. Each criterion representing a proportion of 8% of the food supply or more is included in the pooled sample (Fig. 3).

The food balance sheet enabled us to identify the origin of imported food commodities. In the case of food commodities which are both imported and locally grown (ex: rice) the SSA-TDS strategy consists of producing two distinct composites, to be able to identify a source of contamination. This approach, including two composite samples, is compatible with a more precise subsequent exposure assessment based on the actual proportions of imported and locally produced food items.

The result of the core foods selection and the sampling plan are summarized in Table 7.

Some of the 84 foods subgroups defined in the food classification at level 2 of Fig. 1 were not recorded (from 1 non-recorded food subgroup for Cameroon to 14 in the case of Benin) by national institutes of statistics. However 100% of the average total diet is considered as recorded with a number of food subgroups: 70 for Benin, 83 for Cameroun, 71 for Mali and 77 for Nigeria. The selected core foods amount to 27 for Benin, 36 for Cameroun, 38 for Mali and 40 core foods in the case of Nigeria. In spite of the heterogeneous food classifications of the four original datasets, also reflected in the variability of the number of selected core foods (CV = 15%), the coverage of the average total diet by the selected core foods is very similar among the 8 study centers (CV = 2%).

3.4. Sampling strategy

The number of samples required to cover 100% of the core foods recorded in the household budget surveys once per study center is 602 (Table 7). Thanks to (1) the core foods selection methodology and (2) the introduction of national samples when applicable (sampled only once but applying to the two national study centers for exposure assessment), the number of selected core foods drops to 204 required samples (34%). This cost effective approach enables the coverage of 94,8 ± 2,1% of the average total diet by weight, including 92,3 ± 2,1% of the total diet covered by locally sampled core foods.

Taking into consideration the tap water composite samples compiled by each study center and two sampling campaigns, the subsamples collected in the eight city centers amounted to a total of 4020 purchases (63% for the first and 37% for the second campaign). As pooled samples include 12 subsamples each, the study gathers 204 composite samples of core foods plus 8 tap water composite samples for the first sampling campaign and 115 composite samples of core foods plus 8 tap water composite samples for the second campaign. Every composite sample undergoes up to 7 multi-analyte screening tests, which represents a total of 766 analyses (77% for the first and 23% for the second campaign).

3.5. Food chemicals list

Trace elements and metals selected include heavy metals

Table 6
Comparison of the mean consumption of cereals in grams per adult male equivalent per day in coastal and non coastal study centers.

Variable	Observations	Minimum (g/AME/day)	Maximum (g/AME/day)	Mean (g/AME/day)	SD (g/AME/day)
COASTAL AREAS	3	437	849	597	221
NON COASTAL AREAS	5	1094	1409	1247	121

Difference	−650
t (Observed value)	−5.5
|t| (Critical value)	2.4
DF	6
p-value (Two-tailed)	0.001
alpha	0.05

t-test for two independent samples/Two-tailed test:
95% confidence interval on the difference between the means: [-939;-362[
Test interpretation.
H0: The difference between the means is equal to 0.
Ha: The difference between the means is different from 0.
As the computed p-value is lower than the significance level alpha = 0,05, one should reject the null hypothesis H0, and accept the alternative hypothesis Ha. The risk to reject the null hypothesis H0 while it is true is lower than 0,15%.

Table 7
Coverage of total diet in weight by the sampling plan.

	Country	Benin		Cameroon		Mali		Nigeria		SSA Total Diet Study			
	Study center	Littoral	Borgou	Duala	North	Bamako	Sikasso	Lagos	Kano	Total	Mean	SD	CV
NUMBER OF CORE FOODS	Recorded core foods	70	70	83	83	71	71	77	77	602	75	6	7%
	Locally sampled core foods	24	18	33	19	35	19	33	23	204	26	8	28%
	National core food sampled elsewhere	3	9	3	17	3	19	7	17	78	10	7	71%
	Total sampled core foods	27	27	36	36	38	38	40	40	282	35	5	15%
COVERAGE OF AVERAGE DAILY INTAKE (g/AME/d)	Recorded core foods	2017	1874	1563	2099	1893	1457	1645	1524	–	1759	241	14%
	Locally sampled core foods	1826	1734	1455	1892	1824	1339	1541	1373	–	1623	213	14%
	National core food sampled elsewhere	7	55	4	115	4	77	19	72	353	44	43	95%
	Total sampled core foods	1833	1789	1459	2007	1828	1416	1560	1445	–	1667	222	13%
COVERAGE OF TOTAL DIET (%)	Recorded core foods	100	100	100	100	100	100	100	100	–	100	0	0%
	Locally sampled core foods	90.5	92.5	93.1	90.2	96.4	91.8	93.7	90.1	–	92.3	2.1	2%
	National core food sampled elsewhere	0.3	2.9	0.2	5.5	0.2	5.3	1.2	4.7	20.3	2.5	2.4	93%
	Total sampled core foods	90.8	95.5	93.3	95.7	96.6	97.1	94.9	94.8	–	94.8	2.1	2%

commonly monitored in food safety risk assessment as well as elements likely to migrate from cooking pots to food matrices. These are aluminium (Al), arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), mercury (Hg), nickel (Ni), lead (Pb) and tin (Sn) as a core list.

The core mycotoxins list includes a list of 23 substances screened in the FAO/WHO project on mycotoxins in sorghum (FAO, 2015) implemented in Burkina Faso, Ethiopia, Mali and Sudan (2012–2014). These are aflatoxins (B1, B2, G1, G2), altenuene, alternariol, alternariol monomethylether, deoxynivalenol (including 3-actetyldeoxynivalenol and 15-actelydeoxynivalenol), diacetoxyscirpenol, fumonisins (B1, B2, B3), fusarenon X, HT2 toxin, neosolaniol, nivalenol, ochratoxin A, roquefortine C, sterigmatocystin, T-2 toxin and zearalenone. The choice of those analytes is based on the expectation to find these substances (FAO, 2015) and will be extended, as recent advances in Africa have shown the occurrence of toxins never reported before in Cameroonian food (Abia et al., 2017).

An extraction from the European Commission Rapid Alert System for Food and Feed 2000–2016 showed that 99% of the alerts involving pesticides concentration above maximum residue limits in force involve 10 phytosanitary products (endosulfan, chlorpyrifos, profenefos, dichlorvos, dimethoate, ethephon, omethoate, trichlorfon, cypermethrin, lambda cyhalorthrin and permethrin) out of 109 substances recorded in Benin, Cameroun, Mali and Nigeria (European Commission, 2016a).

The availability of multi-analyte screening tests for organochlorine, organophasphorous and pyrethroids enables to cover all of these 10 actives substances, as well as the pesticides included in the Stockholm Convention, as a core pesticides residues list.

The PAHs selected for this study are the 13 genotoxic and carcinogenic substances evaluated by the Joint FAO/WHO Expert Committee on Food Additives, (WHO, 2006). These PAHs are benzo(*a*)pyrene, benzo(*a*)anthracene, benzo(*b*)fluoranthene, benzo(*j*)fluoranthene, benzo(k)fluoranthene, chrysene, dibenzo(a,h)pyrene, dibenzo(a,i)pyrene, dibenzo(a,e)pyrene, dibenzo(a,l)pyrene, dibenzo(a,h)anthracene, indeno(123-*cd*)pyrene and 5-methylchrysene.

The 12 coplanar or dioxin-like PCBs (PCB 77, PCB 81, PCB 105, PCB 114, PCB 118, PCB 123, PCB 126, PCB 156, PCB 157, PCB 167, PCB 169 and PCB 189) identified by the Joint FAO/WHO Expert Committee on Food Additives (WHO, 2002) as well as 6 non dioxin-like "indicator PCBs" (PCB 28, PCB 52, PCB 101, PCB 138, PCB 153 and PCB 180) plus PCB 128, for which adequate data were available to perform a risk characterization (WHO, 2015), are included in the core food chemicals list, as well as 10 polychlorinated dibenzo-*p*-dioxins and 7 polychlorinated dibenzofurans.

In addition to these food chemicals, 15 perfluoroalkoxy alkanes (PFAs) and 13 brominated flame retardants (BFRs) are going to be screened in the food samples.

3.6. Risks and limitations of this strategy

Although the percentage of selected households seemed rather inclusive for 3 countries as shown in Table 2 with 79% for Benin, 75% for Cameroon, and 87% in the case of Mali, we could not find a suitable explanation as to why the dataset from Nigeria only enabled to select 40% of the households, based on estimated energy requirements ± 45%. The significant number of households selected from the original datasets of household budget surveys (44.431 households) for the four countries needs to be balanced with the fact that the design of this study focuses on 8 study centers, covering in total 7.291 households or 16% of total normal reporting households. Although the whole dataset was used to establish nationally representative core food lists, the exposure assessment and sampling will reflect the exposure of two populations per country, but will hardly be considered as representative for the whole country. The European Food Safety Authority (EFSA, 2014) recommends the inclusion of 6 age groups containing each at least 260 subjects in dietary surveys and suggests not to discard over and under-reporting households. The SSA-TDS does not comply with these recommendations, and will not enable the estimation of the individual exposures for study subjects. Because the daily consumption data are derived from food expenditure data, based on questionnaires submitted to the head of household, they do not reflect the individual diet by gender or by age group either.

In particular, breast milk is not included in the core food list, because it is not recorded as a commercial food item. Weaning foods, although identified as a food subgroups, were not selected in the food list due to their low contribution to the total diet of the general population.

From this study, it is therefore impossible to target specific population groups, exposed to a particular risk, without additional data, thereby justifying further studies to cover the identified gaps.

The selection of core foods is solely based on mean daily consumption. Due to the lack of data describing the contamination patterns in Africa, the selection of core foods does not focus, in its design, on food commodities considered as high contributors to the dietary exposure or highly contaminated core foods. However, the fact that each food group is significantly represented (50% or 90% depending on the proportion of the total diet by weight covered by food group) means that a large variety of core food are included in the sampling, thus reducing the risk of skipping high contributors. However, the risk that a highly contaminated food item representing a low mean daily consumption is not taken into consideration exists, and cannot totally be ruled out.

The fact that we are preparing composite samples from one single core food with inert kitchen utensils presents a number of limits. This methodological choice enables the assessment of the contribution to the dietary exposure of each core food individually. However, it means that (1) the interaction between distinct core foods (for example due to osmosis or chemical phenomena such as Maillard reaction) and (2) the interaction between food contact materials and food matrices (Weidenhamer et al., 2017), both occurring during food preparation at household level and likely to impact the food chemical concentration of samples, are not taken into consideration. Therefore, this TDS methodology is more suited for screening environmental food chemicals, than for the purpose of detecting neo-formed substances such as acrylamide. This limit has impacted the selection of substances to look for. However, a few samples of the most consumed matrices will be prepared twice, with both traditional utensils and with inert kitchen utensils, in an attempt to capture a difference of concentration of elements in food samples.

The attempt to capture the seasonal variation of the concentration to food chemicals is another limitation of this study. In this methodology, we intend to focus on the most consumed food commodities (cereals, tubers, legumes, vegetables and fruits) food chemicals, which are highly likely to vary in terms of occurrence between dry and rainy seasons (mycotoxins and pesticides). This means that we will not be able to capture the variation of occurrence of the other food chemicals, nor the variation in occurrence of pesticides residues and mycotoxins in nuts and seeds, meat, eggs, fish, dairy products, oil and fats, beverages, and miscellaneous.

Finally we are aware that although cost effective, our approach tends to focus on main sources of the food supply (national sampling enabling to reduce the number of samples needed to cover the average total diet in weight). This means that the variation of the concentration of food chemicals will not be systematically be captured. It will however be the case for at least 90% of the average total diet.

4. Conclusions

We developed the core food model in this regional Sub-Saharan Africa Total Diet Study. The purpose of this TDS is to investigate a large number of food chemicals through a food sampling plan, which aims to be representative of the dietary habits of a large population of four African countries. This TDS has been adapted to the African context with limited resources, but nonetheless with a consistent and harmonized methodology. It complies with WHO and FAO recommendations and can, to a certain extent, be compared with other TDS implemented at international level by various national food safety authorities. Moreover this program is intended to provide concentration and dietary exposure data in Africa, as well as supporting international scientific advice for utilization by the Codex Alimentarius Commission. Therefore, it will contribute to consumer protection with regard to food safety issues, whilst providing evidences, likely to be used by the international community to tackle technical barriers to trade.

This study provides a baseline of concentrations and dietary exposure that can be used for comparison for future surveys in Africa. These types of baseline data are useful if risk management measures are implemented and if the impact of those can be evaluated.

Moreover, this TDS is likely to provide valuable information to the international risk assessment community concerning chemical concentrations and levels of dietary exposure for a specific area of the World, where this type of information may not be as common as in other areas of the World. This TDS can also be used to identify further work or research for either specific food chemicals or specific population groups, depending on the result of the study.

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The first paper, entitled '*Methodology Design of the Regional Sub-Saharan Africa Total Diet Study in Benin, Cameroon, Mali and Nigeria*', includes the first results of the SSA-TDS. The food consumption data contribute to the methodology in the sense that the core food list relies almost exclusively on food consumption data. We first displayed food consumption data by country, because we established core food lists at a national scale. We also presented the data assessed at the study centre level, which is key to be able to refine dietary exposure to the point of assessing distinct geographic areas.

The food consumption patterns were significantly different between the centres in coastal areas, including the Littoral area of Benin (Cotonou), Duala, and Lagos versus those in other areas, namely Borgou, the North of Cameroon, Bamako, Sikasso, and Kano, where cereals represented a larger part of the diet. The different food consumption patterns among different geographic areas (sometimes within the same country) justifies the multi-centre approach. Based on these data alone, one could foresee that we would obtain specific exposure patterns in the next steps of the study.

The core food list represents the framework of the sampling plan and covers more than 90% of the mean total diet by weight in each study centre. To avoid sampling only staple foods, such as cereals and tubers, the sampling method included core foods covering at least 50% of the food consumption of each food group, regardless of its relative importance in the diet. This rule was key to ensuring variety in the sampled core foods and to characterizing contamination patterns in a vast spectrum of foods.

Food consumption data at the most detailed level of classification, the structure of the food balance sheets, and the preliminary market surveys enabled us to identify the proportion of food items of the same core food that were pooled to form the composites sent to the laboratories. Individual food consumption data were not available in this study. A perspective in terms of research is to generate individual food consumption data in the countries involved in this study. The combination of individual food consumption data with the food contamination data generated by the SSA-TDS would be beneficial in terms of risk assessment. This would allow for the study of dietary exposure in the most vulnerable population groups, including infants and young children, as well as women of child bearing age.

3.2. Food Chemical Concentration Data

The food chemical concentration data (food contamination or food occurrence data) are presented hereafter with regards to **i)** mycotoxins and other fungal, bacterial, and plant secondary metabolites, **ii)** polycyclic aromatic hydrocarbons, **iii)** pesticide residues, **iv)** metals and trace elements, and **v)** POPs including dioxins and PCBs, brominated flame retardants, perfluorinated alkylated compounds, and dechloranes.

3.2.1. Mycotoxin occurrence data

Mycotoxins are secondary metabolites produced by moulds. Factors affecting mycotoxin production are the type of substrate, fungal species, moisture content of the substrate, humidity, temperature, and degree of physical damage to the kernels (Abrar et al., 2013). Although the introduction of maize in Africa began progressively during the sixteenth century, after its discovery by Christopher Colombus in America, maize only became a staple food in Sub-Saharan Africa during the twentieth century (Cherniwchan and Moreno-Cruz, 2019). Aflatoxin in maize was responsible for a major outbreak in 2004 in Kenya (Lewis et al., 2005). The acute toxicity of mycotoxins is probably just the tip of the iceberg, while chronic exposure to mycotoxins may be the submerged section. Establishing a relationship between chronic mycotoxin intake and health consequences from an epidemiological perspective is indeed complex. The long-term effects of mycotoxin exposure include liver cancer, retarded growth of children, and nephrotoxicity, and these affections may each have several causes.

We seized the opportunity of this TDS to study the occurrence of mycotoxins, not only in maize and peanuts, but also in a large variety of cereals, tubers, legumes, nuts and seeds, meat, fish, dairy products, oil and fats, and beverages. I took the following photograph (Figure 11) in North Benin, where schoolchildren stand close to maize, which dries in direct contact with the soil. On the rooftops, beans are also drying. This is the food that children eat daily. Frequently, children learn from their elders and participate in agricultural tasks, including the post-harvest handling of crops.

Figure 11: Maize used for school meals drying in direct contact with the soil in Banikoara, North Benin

Height-for-age is used as a chronic malnutrition indicator (de Onis et al., 2007). Stunting is a condition in which children are significantly too small compared to the growth standards corresponding to a certain age. Retarded growth is, among other causes such as inadequate nutrient intake and repeated infections, a consequence of mycotoxin exposure (Khlangwiset et al., 2011). Allowing direct contact between maize and the soil is a common but suboptimal practice in rural areas. This lack of hygiene is often the consequence of poor infrastructure and a lack of education concerning post-harvest issues (Omari et al., 2018). Fungi may thrive in this maize as contact with fungal spores from the environment is inevitable and the drying and storage conditions are suboptimal.

In the first SSA-TDS paper, we presented food consumption data. We noted that, on average, Lagos households had the lowest consumption rates of maize and peanuts. In contrast, Northern Cameroonians ate more maize and peanuts on average than households from other geographic areas.

But to what extent will mycotoxin concentrations vary among the various geographic areas?

Which core foods are most contaminated?

By which toxins?

toxins

MDPI

Article

Regional Sub-Saharan Africa Total Diet Study in Benin, Cameroon, Mali and Nigeria Reveals the Presence of 164 Mycotoxins and Other Secondary Metabolites in Foods

Luc Ingenbleek [1,2], Michael Sulyok [3], Abimbola Adegboye [4], Sètondji Epiphane Hossou [5], Abdoulaye Zié Koné [6], Awoyinka Dada Oyedele [4], Chabi Sika K. J. Kisito [7], Yara Koreissi Dembélé [8], Sara Eyangoh [1], Philippe Verger [9], Jean-Charles Leblanc [10,*], Bruno Le Bizec [2] and Rudolf Krska [3,11]

1 Centre Pasteur du Cameroun (CPC), Yaoundé BP1274, Cameroon; luc.ingenbleek@gmail.com (L.I.); eyangoh@pasteur-yaounde.org (S.E.)
2 LABERCA, Oniris, INRA, 44307 Nantes, France; Bruno.LeBizec@oniris-nantes.fr
3 Department IFA-Tulln, University of Natural Resources and Life Sciences, Vienna (BOKU), 3430 Tulln, Austria; michael.sulyok@boku.ac.at (M.S.); Rudolf.Krska@boku.ac.at (R.K.)
4 National Agency for Food and Drug Administration and Control (NAFDAC), Abuja 900288, Nigeria; adegboye.a@nafdac.gov.ng (A.A.); oyedele.dada@nafdac.gov.ng (A.D.O.)
5 Agence Béninoise de Sécurité Sanitaire des Aliments (ABSSA), Cotonou BP 362, Benin; hossepfr@yahoo.fr
6 Agence Nationale de la Sécurité Sanitaire des Aliments (ANSSA), Bamako BP 2362, Mali; ngolona@gmail.com
7 Laboratoire Central de Sécurité Sanitaire des Aliments (LCSSA), Cotonou BP 6874, Benin; kinnousika@yahoo.fr
8 Laboratoire de Technologie Alimentaire (LTA), Bamako BP 258, Mali; ykoreissidemb@gmail.com
9 World Health Organization (WHO), 1211 Geneva, Switzerland; vergerp@who.int
10 Food and Agriculture Organization of the United Nations (FAO), 00153 Rome, Italy
11 Institute for Global Food Security, School of Biological Sciences, Queens University Belfast, Belfast BT7 1NN, Northern Ireland, UK
* Correspondence: JeanCharles.Leblanc@fao.org

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Abstract: In the framework of the first multi-centre Sub-Saharan Africa Total Diet Study (SSA-TDS), 2328 commonly consumed foods were purchased, prepared as consumed and pooled into 194 composite samples of cereals, tubers, legumes, vegetables, nuts and seeds, dairy, oils, beverages and miscellaneous. Those core foods were tested for mycotoxins and other fungal, bacterial and plant secondary metabolites by liquid chromatography, coupled with tandem mass spectrometry. The highest aflatoxin concentrations were quantified in peanuts, peanut oil and maize. The mean concentration of the sum of aflatoxins AFB1, AFB2, AFG1 and AFG2 (AF_{tot}) in peanut samples (56.4 µg/kg) exceeded EU (4 µg/kg) and Codex (15 µg/kg) standards. The AF_{tot} concentration (max: 246.0 µg/kg) was associated with seasonal and geographic patterns and comprised, on average, 80% AFB1, the most potent aflatoxin. Although ochratoxin A concentrations rarely exceeded existing Codex standards, it was detected in unregulated foods. One palm oil composite sample contained 98 different metabolites, including 35.4 µg/kg of ochratoxin A. In total, 164 different metabolites were detected, with unspecific metabolites like asperglaucide, cyclo(L-pro-L-val), cyclo (L-pro-L-tyr), flavoglaucin, emodin and tryptophol occurring in more than 50% of composite samples. Aflatoxin B1 (AFB1), fumonisin B1 (FB1), sterigmatocystin (STC), ochratoxin A (OTA), citrinin (CIT) and many other secondary fungal metabolites are frequent co-contaminants in staple foods, such as maize and sorghum. Populations from North Cameroon and from Benin may, therefore, suffer chronic and simultaneous exposure to AFB1, FB1, STC, OTA and CIT, which are prevalent in their diet.

Keywords: Sub-Saharan Africa; aflatoxins; mycotoxins; total diet study; food contaminants; LC-MS/MS

1. Introduction

Mycotoxins are secondary metabolites produced by filamentous fungi in food commodities due to inadequate pre- or post-harvest conditions and practices. These fungal toxins are, therefore, naturally-occurring chemical hazards. Since they are structurally stable, mycotoxins are likely to persist in foods, even if toxin-producing moulds are eliminated during the food preparation process. Consumption of mycotoxin-contaminated food may result in acute or chronic affections, including non-communicable diseases. A particularly severe record of acute toxicity was reported after a major outbreak struck Kenya in 2004, resulting in 317 aflatoxicosis cases including 125 deaths [1]. This episode was the consequence of high exposure to aflatoxins due to the consumption of extensively-contaminated maize [2]. Long-term exposure to aflatoxin B1 or its precursors has been associated with genotoxicity and hepatocellular carcinoma [3,4]. Fumonisin B1 was associated with oesophageal cancer incidence in South Africa and some areas of China [5,6]. Growth impairment, the main indicator for child chronic malnutrition, is also associated with mycotoxin exposure [7–10]. Of the world's 161 million stunted children in 2013, about half live in Asia and over one-third live in Africa [11]. Although often overlooked as a possible cause of retarded growth, mycotoxins may contribute a significant public health burden in less developed countries [12].

An additive or synergistic effect of fumonisin and aflatoxin co-exposure in the development of preneoplastic lesions or hepatocellular carcinoma was suggested in laboratory animals [13–15].

Mycotoxins form the group of food chemicals which triggered the most cases of border rejection (489) recorded in the EU Rapid Alert System on Food and Feed [16]. According to the European Commission Regulation 1881/2006 [17], the maximum level for aflatoxins for peanuts and cereals intended for direct human consumption was set to 2 µg/kg of aflatoxin B1 (AFB1) and 4 µg/kg of the sum of AFB1, AFB2, AFG1 and AFG2. The maximum limit from the international standard [18] is 15 µg/kg of AFB1 or AF_{tot}, which only applies to a variety of nuts (including peanuts) intended for further processing (and 10 µg/kg for ready to eat dried figs, almonds, hazelnuts and pistachios).

In order to assess if the chronic intake of substances is likely to harm consumer health, it is pertinent to assess food safety risks by combining available toxicological studies, as well as food contamination and food consumption data.

One way of assessing the dietary exposure of populations to food chemicals, such as mycotoxins is the Total Diet Study (TDS) approach [19–24]. Two specific aspects characterize a TDS—(1) the representativeness of the sampling, and (2) the preparation of the samples "as consumed"—so that it represents a pertinent public health risk assessment tool as far as food safety and nutrition are concerned.

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 more accurately characterizes human exposure to food chemicals than mere occurrence studies [23].

Following a first experience in Sub-Saharan Africa [25,26], a regional TDS was implemented by FAO in Benin, Cameroon, Mali and Nigeria (2014 and 2018) by four national food safety authorities, in close collaboration with WHO and Centre Pasteur of Cameroon [27]. The purpose of this project is to assess the typical contamination levels of eight African population groups. The dietary exposure of those population groups will then be compared with existing health-based guidance values or end points.

The study methodology was described elsewhere [28].

In this paper, we are presenting the occurrence of mycotoxins and selected fungal, bacterial and plant toxins quantified in composite samples of foods prepared as consumed. The 194 composites

result from the pooling of 12 subsamples, representative of the food consumption habits of three study centres located in coastal areas (Duala, the Littoral of Benin and Lagos) and five study centres in non-coastal areas (Bamako, the Borgou region of Benin, Kano, North Cameroon and Sikasso).

2. Results

Since we are dealing with pooled samples (12 sub-samples per composite) of foods prepared as consumed in this study, we will not always be able to conclude with regard to the conformity of food commodities to selected standards [17,18], which, in most cases, apply to raw food commodities. This comparison is nonetheless useful, particularly when the mean concentration (quantified in a composite sample) exceeds or is close to the maximum legal limit of the substances of interest, because this means that at least one subsample out of 12 may have exceeded this limit.

Additionally, since these data will be used for a dietary exposure assessment, they are presented with (1) lower bound (i.e., LB: concentration of non-detected analytes set to zero and to the LOD for detected but non-quantified analytes) and (2) upper bound (i.e., UB: concentration of non-detected analytes set to LOD for non-detected analytes and to the limit of quantification (LOQ) for detected but non-quantified analytes) scenarios. This means that the uncertainty due to censored data will be taken into consideration. When LB–UB is not specified, it is meant that the difference between LB and UB concentrations in not perceptible or less than 0.1 µg/kg. Maximum concentration values are systematically UB concentrations.

Mycotoxins of public health and economic interest (including aflatoxins, fumonisins, ochratoxin A, zearalenone, deoxynivalenol and citrinin) represented 9% of the detected metabolites.

2.1. Aflatoxins

2.1.1. Aflatoxins in Maize

Composite samples were prepared with maize from each study centre (8) purchased during the rainy season (October 2017) and again during the dry season or harmattan (February 2018).

The AF_{tot} concentration in maize was significantly higher ($p < 0.05$) during the wet season (detected: 7/8; mean LB–UB: 22.2–22.5 µg/kg; max: 76.6 µg/kg) than during the dry season (detected: 4/8; mean LB–UB: 0.4–0.8 µg/kg; max: 2.7 µg/kg). Overall, we detected that AF_{tot} > LOD in 11 of 16 composites (69%) with a mean LB–UB concentration of 11.3–11.7 µg/kg (Table 1) in ready-to-eat maize samples, which exceeds the EU standard for both processed (4 µg/kg) and unprocessed maize to be subjected to sorting or physical treatment before human consumption or use as an ingredient (10 µg/kg). However, the fact that all maize composites collected during the dry season contained AF_{tot} concentrations that were below 4 µg/kg and, therefore, complied with EU standard needs to be emphasized.

Table 1. Occurrence and concentration of total aflatoxins (μg/kg wet weight) by core food and by study centre.

CORE FOOD		N	n > LOD	% > LOD	n > 4 μg/kg	% > 4 μg/kg	n > 15 μg/kg	% > 15 μg/kg	Mean Conc. *		Max Conc.	
									LB	UB	Season **	UB
Maize		16	11	69	5	31	3	19	11.3	11.7	Rainy	76.6
Peanut		10	8	80	5	50	5	50	56.4	56.7	Rainy	246.0
Peanut oil		2	2	100	2	100	2	100	60.2	60.4	Rainy	105.1
Beans		16	3	19	1	6	1	6	1.2	1.6	Dry	15.8
Sorghum		10	6	60	1	10	0	0	0.9	1.3	Rainy	4.9
Smoked fish		6	1	17	1	17	0	0	0.8	1.1	Rainy	4.9
Other core foods		134	11	8	0	0	0	0	0.1	0.5	Rainy	2.4
Total		194	42	22	15	8	11	6	4.7	5.1	Rainy	246.0
CENTRE		**N**	**n > LD**	**% > LD**	**n > 4 μg/kg**	**% > 4 μg/kg**	**n > 15 μg/kg**	**%> 15 μg/kg**	**Mean Conc. ***		**Max Conc.**	
									LB	**UB**	**Core food**	**UB**
BENIN	**Littoral**	26	5	19	3	12	3	12	7.6	8.0	Peanut oil	105.1
	Borgou	22	7	32	1	5	1	5	1.2	1.6	Maize	19.7
CAMEROON	**Duala**	29	3	10	0	0	0	0	0.2	0.6	Beans	3.0
	North	17	8	47	4	24	3	18	14.3	14.6	Peanuts	92.5
MALI	**Bamako**	27	4	15	2	7	1	4	9.4	9.8	Peanuts	246.0
	Sikasso	21	6	29	1	5	1	5	2.2	2.6	Peanuts	42.7
NIGERIA	**Lagos**	29	3	10	1	3	0	0	0.2	0.6	Maize	5.4
	Kano	23	6	26	3	13	3	13	5.6	6.0	Peanuts	96.6

* LB: lower-bound scenario where the concentration of non-detected analyte is zero and the concentration of detected but non-quantified analyte is the limit of detection. UB: upper-bound scenario where the concentration of non-detected analyte is the limit of detection and the concentration of detected but non-quantified analyte is the limit of quantification; ** Samples of the rainy season were collected in October 2017 and samples of the dry season were collected in February 2018.

There is currently no Codex standard applicable to aflatoxins in maize. AFB1, the most potent aflatoxin, represented 87.6% of the sum of AFB1, AFB2, AFG1 and AFG2 detected in maize samples (Table 2).

2.1.2. Aflatoxins in Peanut

As displayed in Table 1, the highest AF_{tot} concentration in this study was quantified in one peanut composite sample from Bamako (Mali): 246.0 µg/kg (mean LB–UB: 56.4–56.7 µg/kg). Aflatoxins were detected in 80% of peanut composites (rainy season: 100%, dry season: 60% detection exceeding LOD = 0.1 µg/kg). The mean AF_{tot} concentration in peanuts was 93.7–93.9 µg/kg (rainy season) and 19.1–19.4 µg/kg (dry season). A high variance of AF_{tot} levels in peanut was observed (CV > 100%). It was noted that while 50% of samples contained AF_{tot} concentrations below the EU standard (4 µg/kg) and 50% were above the Codex standard (15 µg/kg), the mean AF_{tot} concentrations exceeded both EU and Codex standards, regardless of the season. The proportion of AFB1 in peanut was 75.8% of the sum of AFB1, AFB2, AFG1 and AFG2 (Table 2).

Table 2. Proportions of aflatoxin B1, B2, G1 and G2 by core food and by weight.

CORE FOOD	AFB1 (%)	AFB2 (%)	AFG1 (%)	AFG2 (%)	Sum (%)
Maize	87.6	6.8	5.6	0.0	100
Peanut	75.8	14.3	9.4	0.5	100
Peanut oil	86.6	13.1	0.3	0.0	100
Other core foods	87.0	4.0	9.1	0.0	100
Total	80.1	12.1	7.5	0.3	100

2.1.3. Aflatoxins in Peanut Oil

Two composite samples of peanut oil were tested (Table 1) and both contained significant amounts of total aflatoxins: 15.8 µg/kg (Kano) and 105.1 µg/kg (Cotonou). There is currently no standard for aflatoxins in oil, and these concentrations exceed Codex standards available for processed and unprocessed peanuts. The proportion of AFB1 in peanut oil was 86.6% of the sum of AFB1, AFB2, AFG1 and AFG2 (Table 2).

2.1.4. Aflatoxins in Other Foods

Aflatoxins were detected in 60% of sorghum and 19% of bean composites. In Table 1, we reported that one bean sample contained 15.8 µg/kg AF_{tot}. One smoked fish composite contained 4.9 µg/kg AF_{tot}. The observed mean concentration of all tested core foods was below 1 µg/kg in Duala and Lagos (detection rate of 10%) but those recorded in North Cameroon exceeded 10 µg/kg (detection rate of 47%).

2.2. Fumonisins

FUM_{tot} (sum of fumonisins FB1, FB2, FB3 and FB4) were most concentrated in maize samples in all eight centres (Table 3). Although fumonisins were detected in 94% of ready-to-eat maize composites, all FUM_{tot} concentrations (mean LB–UB: 285.2–288.2 µg/kg; max: 855.9 µg/kg) remained below the Codex standard of 2 mg/kg applying to fumonisins in maize. Although there is no Codex standard for fumonisins in other foods than maize, other core food samples contained FUM_{tot} of up to 159.4 µg/kg. Apart from maize, composites containing fumonisins are sorghum (including a traditional fermented drink from North Cameroon processed from sorghum called bili-bili) and millet and tubers having undergone a drying process prior to being prepared as consumed (cassava and yam), as reported in Table 3. In food composites from Mali (Bamako and Sikasso), the mean FUM_{tot} concentration was three to ten-fold lower than in samples collected in the other study centres, with UB and LB scenario respectively. FUM_{tot} in our samples comprised 67.2% FB1, 18.9% FB2, 8.0% FB3 and 6.0% FB4. This is close to the proportions determined in maize samples (Table 4).

Table 3. Occurrence and concentration of total fumonisins (μg/kg wet weight) by core food and by study centre.

CORE FOOD		N	n > LOD	% > LOD	n > 10 μg/kg	% > 10 μg/kg	n > 400 μg/kg	% > 400 μg/kg	Mean Conc. *		Max Conc.	
									LB	UB	Season **	UB
Maize		16	15	94	15	94	4	25	285.2	288.2	Dry	855.9
Sorghum		10	5	50	5	50	0	0	20.0	36.1	Dry	159.4
Millet		8	1	13	1	13	0	0	5.0	13.6	Rainy	44.8
Traditional fermented drink		4	1	25	1	25	0	0	5.7	14.1	Rainy	29.3
Cassava dry		12	3	25	3	25	0	0	14.8	22.9	Dry	134.6
Yam dry		2	1	50	1	50	0	0	7.4	17.8	Rainy	21.7
Other core foods		142	2	1	0	0	0	0	0.04	9.2	Both	14.6
Total		194	28	14	26	13	4	2	26.4	34.8	Dry	855.9
CENTRE		**N**	**n > LOD**	**% > LOD**	**n > 10 μg/kg**	**% > 10 μg/kg**	**n > 400 μg/kg**	**% > 400 μg/kg**	**Mean Conc. ***		**Max Conc.**	
									LB	**UB**	**Core food**	**UB**
BENIN	**Littoral**	26	2	8	2	8	0	0	26.8	35.2	Maize	391.3
	Borgou	22	5	23	5	23	0	0	26.3	34.4	Maize	376.5
CAMEROON	**Duala**	29	5	17	4	14	0	0	19.0	27.1	Maize	241.7
	North	17	12	71	5	29	1	6	64.4	71.6	Maize	670.3
MALI	**Bamako**	27	3	11	2	7	0	0	2.0	11.0	Maize	40.6
	Sikasso	21	2	10	2	10	0	0	4.1	12.9	Maize	79.0
NIGERIA	**Lagos**	29	4	14	3	10	1	3	34.9	43.5	Maize	855.9
	Kano	23	3	13	3	13	2	9	45.9	54.1	Maize	589.9

* LB: lower-bound scenario where the concentration of non-detected analyte is zero and the concentration of detected but non-quantified analyte is the limit of detection. UB: upper-bound scenario where the concentration of non-detected analyte is the limit of detection and the concentration of detected but non-quantified analyte is the limit of quantification; ** Samples of the rainy season were collected in October 2017 and samples of the dry season were collected in February 2018.

Table 4. Proportions of fumonisins B1, B2, B3 and B4 by core food and by weight.

CORE FOOD	FB1 (%)	FB2 (%)	FB3 (%)	FB4 (%)	Sum (%)
Maize	65.9	19.3	8.4	6.4	100
Sorghum	76.7	15.8	4.6	2.8	100
Cassava dry	75.4	14.1	6.2	4.2	100
Other core foods	88.2	11.8	0.0	0.0	100
Total	67.2	18.9	8.0	6.0	100

The co-occurrence of FB1 and AFB1 was observed in 11 of 16 maize composites (69%) and four of 10 sorghum composites (40%), as well as one of eight millet composites (13%) and in one of 12 cassava dry samples (8%).

2.3. Sterigmatocystin (STC)

STC, which is a known aflatoxin precursor [29] was mostly prevalent in cooking oils (Table 5). STC was quantified in 50% of peanut composites (mean: 0.6 µg/kg; max: 2.9 µg/kg) and in all peanut oil samples (mean: 8.5 µg/kg; max: 8.7 µg/kg), which also contained aflatoxins (Table 1). Interestingly, STC was quantified in 100% of "other vegetable oil" samples (cottonseed oil in most cases), whereas aflatoxins were not detected in those composites (tested with the same limit of detection, LD = 0.1 µg/kg).

Contrarily, STC detection rate in maize was only 13%, whereas aflatoxins were detected in 69% of composite samples.

There is currently no Codex or EU standard for STC in any food commodity.

The co-occurrence of STC, AFB1 and FB1 was observed in four composites samples, all collected during the rainy season:

1. Maize (North Cameroon): 56.6 µg/kg AFB1; 458.5 µg/kg FB1; 1.0 µg/kg STC
2. Maize (Benin Littoral): 71.8 µg/kg AFB1; 179.0 µg/kg FB1; 0.075 (LB = limit of detection)–0.25 µg/kg (UB = limit of quantification) of STC, which was detected below the limit of quantification.
3. Sorghum (Borgou): 1.7 µg/kg AFB1; 33.5 µg/kg FB1; 0.5 µg/kg STC
4. Sorghum (Sikasso): 0.8 µg/kg AFB1; 12.5 µg/kg FB1; 2.4 µg/kg STC

Table 5. Occurrence and concentration of sterigmatocystin (µg/kg wet weight) by core food and study centre.

CORE FOOD		N	n > LOD	% > LOD	n > 1 µg/kg	% > 1 µg/kg	n > 4 µg/kg	% > 4 µg/kg	Mean Conc. * LB	Mean Conc. * UB	Max Conc. Season **	Max Conc. UB
Peanut oil		2	2	100	2	100	2	100	8.5	8.5	Rainy	8.7
Peanut		10	5	50	2	20	0	0	0.6	0.6	Rainy	2.9
Palm oil		4	3	75	3	75	1	25	2.0	2.0	Rainy	5.3
Other vegetable oil		4	4	100	3	75	1	25	3.9	3.9	Rainy	9.2
Sorghum		10	3	30	2	20	0	0	0.4	0.5	Rainy	2.4
Millet		8	2	25	1	13	1	13	0.6	0.7	Rainy	4.8
Other core foods		156	10	6	0	0	0	0	0.02	0.1	Rainy	1.0
Total		194	29	15	13	7	5	3	0.3	0.4	Rainy	9.2
CENTRE		**N**	**n > LOD**	**% > LOD**	**n > 1 µg/kg**	**% > 1 µg/kg**	**n > 4 µg/kg**	**% > 4 µg/kg**	**Mean Conc. * LB**	**Mean Conc. * UB**	**Max Conc. Core food**	**Max Conc. UB**
BENIN	**Littoral**	26	4	15	2	8	1	4	0.4	0.5	Peanut oil	8.3
	Borgou	22	2	9	0	0	0	0	0.03	0.1	Sorghum	0.5
CAMEROON	**Duala**	29	2	7	1	3	0	0	0.1	0.2	Other vegetable oil	3.0
	North	17	4	24	1	6	1	6	0.7	0.7	Other vegetable oil	9.2
MALI	**Bamako**	27	4	15	3	11	1	4	0.3	0.4	Millet	4.8
	Sikasso	21	6	29	3	14	0	0	0.4	0.4	Peanuts	2.9
NIGERIA	**Lagos**	29	3	10	1	3	1	3	0.2	0.3	Palm oil	5.3
	Kano	23	4	17	2	9	1	4	0.5	0.6	Peanut oil	8.7

* LB: lower-bound scenario where the concentration of non-detected analyte is zero and the concentration of detected but non-quantified analyte is the limit of detection. UB: upper-bound scenario where the concentration of non-detected analyte is the limit of detection and the concentration of detected but non-quantified analyte is the limit of quantification; ** Samples of the rainy season were collected in October 2017 and samples of the dry season were collected in February 2018.

2.4. Ochratoxin A (OTA)

OTA was detected in 10% of tested composite samples (Table 6). Six percent (6%) of all tested samples exceeded 1 μg/kg OTA, including maize (13%), wheat (pasta 50%) and peanut oil (50%). Only three samples contained OTA concentrations exceeding Codex standards applying to unprocessed wheat, barley or rye (5 μg/kg): sorghum (Sikasso: 5.6 μg/kg), rice (Borgou: 6.3 μg/kg) and palm oil (Benin Littoral: 35.4 μg/kg).

There is currently no standard regulating OTA in edible oils, rice and sorghum.

Table 6. Occurrence and concentration of ochratoxin A (µg/kg wet weight) by core food and by study centre.

CORE FOOD		N	n > LOD	% > LOD	n > 1µg/kg	% > 1µg/kg	n > 5µg/kg	% > 5µg/kg	Mean Conc. *		Max Conc.	
									LB	UB	Season **	UB
Palm oil		4	1	25	1	25	1	25	8.9	8.9	Rainy	35.4
Rice		16	5	31	4	25	1	6	0.9	0.9	Dry	6.3
Sorghum		10	2	20	2	20	1	10	0.8	0.9	Rainy	5.6
Maize		16	2	13	2	13	0	0	0.2	0.2	Rainy	1.4
Peanut oil		2	1	50	1	50	0	0	1.2	1.3	Rainy	2.5
Pasta		2	1	50	1	50	0	0	0.5	0.6	Rainy	1.1
Other core foods		144	7	5	0	0	0	0	0.03	0.1	Rainy	0.8
TOTAL		194	19	10	11	6	3	2	0.4	0.4	-	35.4
CENTRE		**N**	**n > LOD**	**% > LOD**	**n > 1 µg/kg**	**% > 1 µg/kg**	**n > 5 µg/kg**	**% > 5 µg/kg**	**Mean Conc. ***		**Max Conc.**	
									LB	**UB**	**Core food**	**UB**
BENIN	**Littoral**	26	5	19	5	19	1	4	1.6	1.6	Palm oil	35.4
	Borgou	22	3	14	3	14	1	5	0.4	0.5	Rice	6.3
CAMEROON	**Duala**	29	3	10	3	10	0	0	0.04	0.1	Cassava fresh	0.7
	North	17	3	18	3	18	0	0	0.2	0.3	Rice	2.0
MALI	**Bamako**	27	1	4	1	4	0	0	0.1	0.2	Maize	1.4
	Sikasso	21	1	5	1	5	1	5	0.3	0.4	Sorghum	5.6
NIGERIA	**Lagos**	29	0	0	0	0	0	0	0	0.1	ND	0.1
	Kano	23	2	9	3	13	0	0	0.2	0.3	Rice	2.6

* LB: lower-bound scenario where the concentration of non-detected analyte is zero and the concentration of detected but non-quantified analyte is the limit of detection. UB: upper-bound scenario where the concentration of non-detected analyte is the limit of detection and the concentration of detected but non-quantified analyte is the limit of quantification; ** Samples of the rainy season were collected in October 2017 and samples of the dry season were collected in February 2018.

2.5. Citrinin (CIT)

CIT was detected in 19% of all samples (Table 7), including maize (63%), sorghum (70%) and rice (38%). The only available citrinin standard (EU) applies to food supplements based on rice fermented by red yeast (2000 μg/kg). Ten percent (10%) of tested samples had CIT concentrations of 5 μg/kg or more and four maize composite samples exceeded 100 μg/kg (25% of maize samples and 2% of all samples).

Table 7. Occurrence and concentration of total citrinin (μg/kg wet weight) by core food and by study centre.

CORE FOOD		N	n > LOD	% > LOD	n > 5 μg/kg	% > 5 μg/kg	n > 100 μg/kg	% > 100 μg/kg	Mean Conc. * LB	Mean Conc. * UB	Max Conc. Season **	Max Conc. UB
Maize		16	10	63	9	56	4	25	76.4	76.8	Rainy	416.5
Sorghum		10	7	70	4	40	0	0	5.5	6.3	Rainy	18.2
Rice		16	6	38	3	19	0	0	2.8	3.2	Dry	18.0
Other core foods		152	14	9	4	3	0	0	0.3	1.0	Rainy	7.4
TOTAL		194	37	19	20	10	4	2	7.0	7.7	Rainy	416.5
CENTRE		**N**	**n > LOD**	**% > LOD**	**n > 5 μg/kg**	**% > 5 μg/kg**	**n > 100 μg/kg**	**% > 100 μg/kg**	**Mean Conc. * LB**	**Mean Conc. * UB**	**Max Conc. Core food**	**Max Conc. UB**
BENIN	**Littoral**	26	4	15	3	12	2	8	19.0	19.7	Maize	372.3
	Borgou	22	9	41	7	32	1	5	21.9	22.5	Maize	416.5
CAMEROON	**Duala**	29	4	14	2	7	1	3	5.7	6.5	Maize	123.6
	North	17	4	24	1	6	0	0	2.2	3.0	Maize	31.9
MALI	**Bamako**	27	2	7	0	0	0	0	0.1	0.9	Maize/Sorghum	2.5
	Sikasso	21	4	19	2	10	0	0	1.5	2.1	Sorghum	17.0
NIGERIA	**Lagos**	29	6	21	3	10	0	0	2.6	3.2	Maize	55.9
	Kano	23	4	17	2	9	0	0	3.4	4.0	Maize	61.4

* LB: lower-bound scenario where the concentration of non-detected analyte is zero and the concentration of detected but non-quantified analyte is the limit of detection. UB: upper-bound scenario where the concentration of non-detected analyte is the limit of detection and the concentration of detected but non-quantified analyte is the limit of quantification; ** Samples of the rainy season were collected in October 2017 and samples of the dry season were collected in February 2018.

2.6. Foods Contaminated by Other Regulated Mycotoxins

2.6.1. Zearalenone (ZEN)

ZEN was detected in 6% of samples and never exceeded EU standards of 100 μg/kg for maize intended for direct human consumption. However, the three composite samples containing the highest ZEN concentrations were collected in the same study centre (Duala): maize (wet season: 7.6 μg/kg; dry season: 97.0 μg/kg) and cassava having undergone a drying process prior to being prepared as consumed (dry season: 7.6 μg/kg).

There is currently no Codex standard for ZEN in foods.

2.6.2. Deoxynivalenol (DON)

DON was also detected in 6% of composite samples, including in (1) bread samples (detection rate: 100%) with a mean concentration of 68.8 μg/kg (min: 31.9 μg/kg; max: 134.6 μg/kg), (2) in 100% of pasta prepared as consumed (mean LB–UB: 9.8–14.3 μg/kg). This is inferior to Codex standards applying to DON cereal-based foods for children (200 μg/kg) and for wheat, maize and barley flour, meal, semolina and flakes (1000 μg/kg).

2.6.3. Ergot Alkaloids

Twelve ergot alkaloids were detected in foods processed from wheat (5 of 6 bread samples), with a mean concentration 62.4 of μg/kg, ranging from non-detected to 165.7 μg/kg, for the sum of ergocornine (1.4%), ergocorninine (0.9%), ergocristine (21.0%), ergocristinine (6.8%), ergocryptine (7.6%), ergocryptinine (2.2%), ergometrine (14.4%), ergometrinine (0.5%), ergosin (21.0%), ergosinine (1.4%), ergotamin (21.7%) and ergotaminine (1.0%). There is no Codex standard for ergot alkaloids, and, to the best of our knowledge, the only available standard (EU) is 0.5 g/kg for the sum of ergot alkaloids in unprocessed cereals, except for maize and rice.

2.7. Non-Detected Mycotoxins of Health and Economic Significance

T2 and HT2 toxins, patulin and diacetoxyscirpenol were never detected in this present study.

2.8. Remarks on a Selection of Other Secondary Fungal, Bacterial and Plant Metabolites

2.8.1. *Aspergillus fumigatus* Metabolites in Palm Oil

The presence of 11 *Aspergillus fumigatus* metabolites was observed in palm oil composites only. Bisdethiomethylgliotoxin was detected in three of four samples, with a mean (LB–UB) concentration of 117.7–118.0 μg/kg. Tryptoquivaline was detected in three of four samples (mean: 81.6–81.8 μg/kg). Gliotoxin was detected in three of four samples (mean: 36.6–36.2 μg/kg). Helvolvic acid was detected in two of four samples (mean 27.2–29.3 μg/kg). Fumigaclavin was detected in three of four samples (mean: 16.6–16.9 μg/kg). Fumagillin was detected in one of four samples (mean: 10.9–13.4 μg/kg). Methylsulochrin was detected in four of four samples (mean: 8.54 μg/kg). Pyripyropene A was detected in one of four samples (mean: 5.7–5.8 μg/kg). Fumitremorgin was detected in three of four samples (mean: 3.6–3.7 μg/kg). Pseurotin A was detected in two of four samples (mean: 2.1–2.8 μg/kg). Pyripyropene D was detected in one of four samples (mean 0.3–0.5 μg/kg). Little is known about these substances, which are not likely to represent a threat to consumer at these concentrations. Their presence, however, reveals that *Aspergillus fumigatus*, a human pathogen, may thrive in the palm oil production chain at some point between the palm tree and final production. Therefore, it represents a risk to value chain operators, if not to consumers [30].

2.8.2. Cereulide in Smoked Fish

The bacterial metabolite cereulide was only detected five times (2.6%) in 194 samples, but was quantified in three of six or 50% of smoked fish samples. Mean (LB–UB) cereulide concentration

in smoked fish was 0.8–0.9 µg/kg, and the maximum concentration was 2.5 µg/kg. The two other composites containing cereulide concentrations above the detection limit of 0.19 µg/kg were beef (2.0 µg/kg) and palm oil (0.7 µg/kg).

2.8.3. Cyanogenic Glucosides in Cassava

Following TDS methodology, all samples were prepared as consumed, but a distinction was made between cassava samples having undergone size reduction, fermentation and drying processes (e.g., cossets or gari using dehydration as preservation and toxins reduction means) before preparation, including rehydration (cassava dry) [31], and other cassava samples (cassava fresh).

Exposure to cyanogenic glucosides, such as linamarin and lotaustralin, may cause serious motor neuron diseases, called konzo [32–35].

A seasonal pattern was observed, with higher concentrations of both linamarin and lotaustralin in fresh cassava during the dry season ($p < 0.05$), which was already reported by previous studies on the matter [36].

While linamarin concentrations ranged from below LD (2.3 µg/kg) to 317 mg/kg wet weight (mean: 134 mg/kg) in cassava fresh samples, it was quantified between 0.15 mg/kg and 18 mg/kg (mean: 2.8 mg/kg) in cassava dry composite samples (1:47 ratio).

Similarly, lotaustralin ranged from 0.04 mg/kg to 0.66 mg/kg (mean: 0.16 mg/kg) in cassava dry and from below LD (1.3 µg/kg) to 18 mg/kg (mean: 6.1 mg/kg) in cassava fresh (1:26 ratio).

Overall, linamarin and lotaustralin were less concentrated in dry cassava samples ($p > 0.05$) than in fresh cassava. The wide range of cyanogenic glucoside concentrations in dry cassava composites (max/min ratio of 120:1 in the case of linamarin and 510:1 for lotaustralin) may be explained by different processing practices, such as the use of the wetting method in cassava flour [37,38], although we were not able to verify these aspects from information requested during the collection of samples.

Four composite samples of cassava fresh were collected in each country during the wet season and again during the dry season or harmattan. Surprisingly, neither linamarin, nor lotaustralin were detected above LD (2.3 and 1.3 µg/kg wet weight, respectively) in samples collected in Nigeria, whereas concentrations varied from 93 to 101 mg/kg (wet season) and from 198 to 317 mg/kg (dry season) in Benin, Cameroon and Mali. We have not figured out the reason of this Nigeria-specific pattern, which may include different cassava varieties or cultivars [39] as well as different cooking methods [40–42].

2.8.4. Low Contaminated Core Foods

We observed relatively low or no occurrence of mycotoxins and other toxins in foods prepared from fresh yam without dehydration processes, in rice and in traditional, soft and fermented drinks, as well as in sugar, onion, garlic, and eggs.

2.9. Secondary Metabolites Profile

Figure 1 shows 62 of the most frequently occurring metabolites out of 164 analytes, on the basis of detection in our samples.

More than a third (36%) of detected metabolites are unspecific to any fungi genera and might also be of plant origin.

Among most prevalent metabolites, six were detected in more than 50% of samples:

- asperglaucide (141 samples or 73% of 194 samples);
- cyclo(L-pro-L-val) (138 samples or 71%);
- cyclo(L-pro-L-tyr) (123 samples or 63%);
- flavoglaucin (105 samples or 54%);
- emodin (103 samples or 53%); and
- tryptophol (99 samples or 51%).

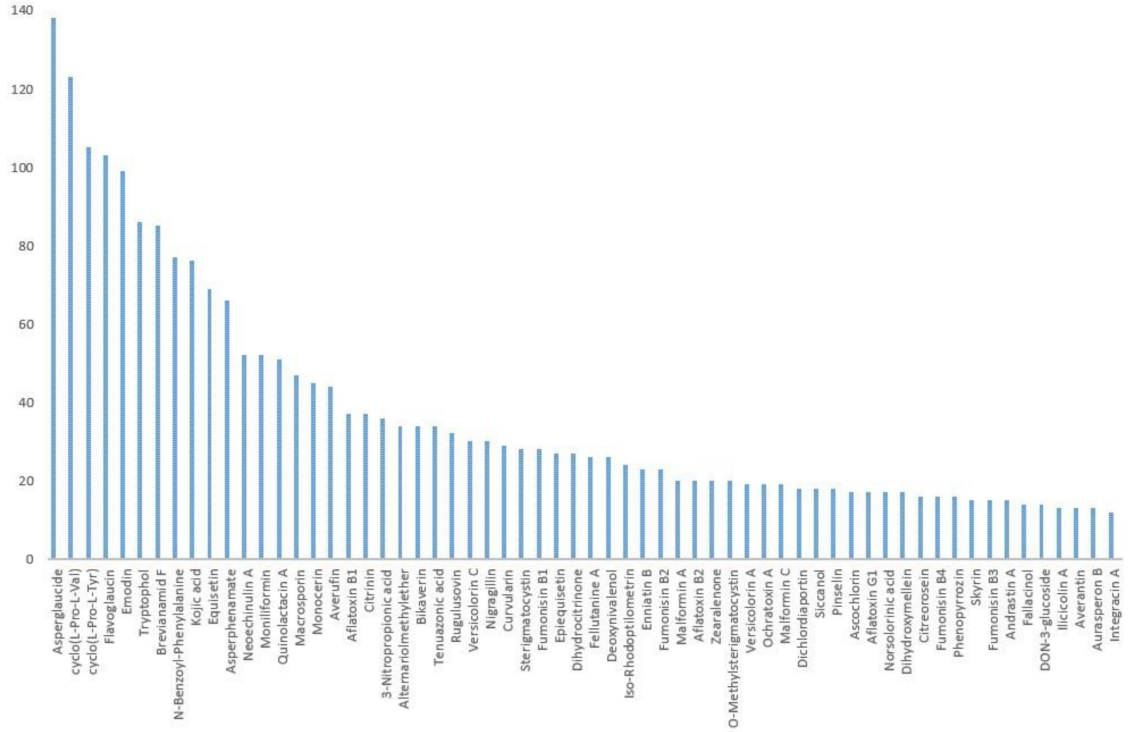

Figure 1. Most frequently detected secondary metabolites in Sub-Saharan Africa Total Diet Study (SSA-TDS) composite samples.

Crude red palm oil composite samples from Cotonou (Benin), Lagos and Kano (Nigeria) showed a higher number of metabolite concentrations above LOD than any other sample (n: 3; mean: 88; min–max: 83–98 metabolites), as noted in Table 8.

Table 8. Range of metabolites detected by core food composite samples.

Range Number of Analytes (min-max)	Composite Samples (n)	CORE FOODS
(51–98)	3	Palm oil
(21–50)	46	Maize, dried tubers, sorghum, peanuts, bread, various oils
(11–20)	62	Beans, dried cassava, rice, millet, smoked fish, onion and garlic, fermented drinks
(6–10)	45	Onion and garlic, meat, tubers, dairy products, rice, traditional soft drinks
(1–5)	36	Fresh tubers, sugar, onion and garlic, rice, eggs
0	2	Onion and garlic
(0–98)	194	TOTAL

All analytical data including quality control checks are enclosed in Table S1.

3. Discussion

First of all, the fact that 164 metabolites were detected in typical African foods does not mean that all of them represent a threat to human health. As of now, in the case of many analytes, the lack of knowledge on their toxicity and their combined (and potentially synergistic) effect with other substances limits our interpretation.

It does, however, represent a contribution to knowledge which may be used when new toxicological data with regards to some of these metabolites will be available. Therefore, the uniqueness of the multi-analyte LC–MS/MS approach used in this study, which enabled the occurrence characterization of a wide range of toxins and other fungal, plant and bacterial secondary metabolites, needs to be emphasized.

In the rest of this discussion, we found it relevant to focus on mycotoxins of public health and international trade significance.

The prevalence of mycotoxins in maize and peanut samples, though, has often been highlighted in previous surveys [43].

Worldwide, several total diet studies from various countries have included mycotoxins, including France [44–46], Canada [47], Lebanon [48], Vietnam [49], and China [50].

In Africa, studies of urinary biomarkers [51–53], surveys of food commodities [54,55], and the analysis foods prepared as consumed [56], have contributed to the rise in attention of the public health community to the threat that mycotoxins represent.

Unsurprisingly, the high concentration of the sum of AFB1, AFB2, AFG1 and AFG2 in African foods as consumed is probably the most significant public health and trade outcome of this multi-mycotoxin analysis compared with other regions of the world [57]. The fact that peanut oil may contain high AF_{tot} concentrations has only recently been described [58]. Peanut oil, peanut and maize are, therefore, likely to contribute significantly to AFB1 exposure, which will be used to characterize the risk of hepatocellular carcinoma.

The presence of fumonisins in staple foods such as maize, with concentrations below the Codex standard of 2 mg/kg, does not guarantee safety for our study populations. The Joint Expert Committee on Food Additives and Contaminants (JECFA) noted in the 83rd session [14], that the current worldwide exposure estimate was established with occurrence data belonging to countries of the WHO European region, and there was no available information on fumonisin levels in maize from the African, Eastern Mediterranean and South-East Asia regions. The JECFA also noted [14] that the interaction between AFB1, a compound with known genotoxic properties, and fumonisins, which have the potential to induce regenerative cell proliferation, is a concern. The completion of the dietary exposure assessment to the sum of FB1, FB2, FB3 and FB4 with the data presented in this paper will also result in conclusions with regards to the adequacy of protective levels of current Codex tolerances in the context of Africa (manuscript in preparation).

Surprisingly, the presence of a high concentration of AF precursor sterigmatocystin, not only in peanut oil but also in cottonseed oil and palm oil, was noted. In contrast, AF was never detected in cottonseed oil but detected in only one of four palm oil samples (0.5 µg/kg AFB1 in a red palm oil composite from sub-samples collected in the Littoral of Benin). This may be due to the production of STC by non-aflatoxigenic *Aspergilli*, such as *Aspergillus nidulans* [59], as well as other fungi genera [60]. The fact that we quantified STC in millet and sorghum composite samples is consistent with recent findings in sorghum [55]. Sorghum and millet, therefore, also qualify as potential STC dietary exposure contributors, noting that typical Sub-Saharan-Sahelian diets largely rely on these cereals [28].

The fact that citrinin was most concentrated in maize means that maize is likely to be a major contributor to CIT dietary exposures in centres where (1) maize CIT concentrations were high and (2) maize is consumed in large amounts.

We would like to bring forward the absence of Codex standards for mycotoxins in edible oils and, in light of occurrence data submitted in this paper, the need for surveillance of mycotoxin contamination levels in edible oils. The presence of (1) OTA in one palm oil with 97 other secondary metabolites, (2) high AF_{tot} concentrations in peanut oil, and (3) the presence of STC in cotton seed oil supports the need for an elaboration in the Codex code of practice for the production of safe edible oil.

Results of the risk characterization (manuscript in preparation) using this occurrence data and adequate food consumption data will clarify to what extent edible oils, as well as other core foods, contribute to the total dietary exposure to mycotoxins in Africa.

Mycotoxin exposure risk mitigation measures include growth prevention of toxin-producing fungi via biocontrol [61] in the field, good post-harvest practices [62] and mycotoxin degradation [63].

As human co-exposure to natural toxins through typical African foods is currently inevitable, national food safety authorities need to ensure that risk assessments are carried out properly to safeguard human health and to maintain international trade.

As demonstrated by the current study, AFB1, FB1, STC and many other secondary fungal metabolites are frequent co-contaminants in many foods (such as maize and sorghum) that threaten human health. Populations in North Cameroon and from Benin (where multiple toxins, including ABF1, FB1 and STC, have been detected within the TDS) may suffer repeated simultaneous exposure to natural toxins. In a recent study [15], the combined effects of various toxins at realistic concentrations were further investigated and revealed additive, antagonistic or synergistic effects. The results have confirmed that combinations of toxins may pose a considerable risk to human health. Clearly, further research is needed to understand the mechanics of toxicological interactions in order to effectively protect public health. Moreover, more TDSs in other locations of Benin, Cameroon, Mali and Nigeria, as well as in other countries belonging to Sub-Saharan Africa need to be carried out to better document actual dietary exposure levels to natural toxins in this region.

4. Conclusions

At this stage of the SSA-TDS project, the first ever multi-centric total diet study carried out in Africa, we have detected 164 secondary metabolites. However, our main results with regards to the occurrence of regulated mycotoxins in eight study centres are as follows:

- Mean AF_{tot} concentrations exceed EU and Codex tolerances applying to peanuts. Similar AF_{tot} levels were quantified in peanut oil (although no Codex or EU standards are currently available for edible oils), as well as in maize samples (aflatoxins in maize are not currently regulated by Codex).
- The TDS approach allowed for the capture of seasonal variations of the AF_{tot} contamination pattern in maize, which contains higher concentrations in samples collected during the rainy season.
- The geographic component of the AF_{tot} contamination pattern was suggested by variations in the mean AF_{tot} concentrations among study centres, which was also observed between two study centres from the same country (Duala versus North Cameroon).

Due to the systematic approach applied to this study, we consider these data fit for the completion of chronic dietary exposure assessment of mycotoxins, for which a health-based guidance value is available (e.g., Tolerable Daily Intake (TDI) or end point for genotoxic carcinogenic substances using the margin of exposure approach). We will then be able to take into consideration food consumption data, at the household level, for eight population groups. We expect maize, peanut and peanut oil to contribute to most of the dietary exposure to AFB1. Likewise, we expect maize to contribute highly to FUM_{tot} and CIT dietary exposure. However, other core foods, in which lower mycotoxin concentrations were estimated, especially highly-consumed staple foods, may also significantly contribute to households' total dietary exposure.

Although Codex maximum limits were not exceeded in the case of FUM_{tot} and OTA, a household dietary exposure assessment will enable risk characterization of the investigated population groups. From this exercise, we will be able to conclude whether currently available Codex mycotoxin standards are sufficiently protective to African consumers.

The dietary exposure assessment of our study populations (manuscript in preparation) will provide guidance to risk managers from Benin, Cameroon, Mali and Nigeria for the identification of national priorities to the consumer protection agenda. We can nonetheless readily address our recommendations to risk managers based on AF_{tot} occurrence data referencing Codex standards only. It will indeed be beneficial for health and trade if national food safety authorities, with the support of their technical and financial partners, draft and implement a road-map and mobilize adequate resources taking the following into consideration:

- Food commodity value chain structures and organization;
- Prevention of field contamination by toxin-producing fungi; and
- Post-harvest practices with emphasis on hygiene, drying and storage conditions.

This will reduce the occurrence and concentrations of mycotoxins in African foods.

To date, these observations about STC occurrence in maize and in oils are new findings which were not reported or highlighted by the last JECFA evaluation of mycotoxins (2016) due to a lack of data at the time of the assessment.

Mitigation measures from Codex Alimentarius may include the updating of current codes of practices and standards and the elaboration of new ones to contribute to the reduction of natural toxins occurrence. This is in an effort to effectively safeguard African consumers' health and food quality.

5. Experimental

5.1. Sample Selection and Preparation of Foods as Consumed

Food consumption data were derived from household budget surveys generated by National Statistics Authorities, from Benin, Cameroon, Mali and Nigeria and gathering a total of 72,979 households. Core foods of each study centre were selected based on the relative importance of their mean consumption [28], so as to cover at least 90% of the mean total diet in grams per adult male equivalent per day (g/AME/d).

Each core food was sampled through available representation criteria [64] (such as market share or the origins of the food) using 12 subsamples of equal size, prepared as consumed and pooled into composites, which underwent laboratory tests. The subsamples were prepared individually according to recipe books [65–68]. These references are considered as representative of the diet of the study populations and were, therefore, selected by the representatives of national competent authorities. These recipe books allow the identification of the processes used in the preparation of the foods, especially cooking time and temperature. The actual recipes were, however, not prepared as each composite sample only contained one core food or ingredient. The inedible parts were removed at the preparation stage, as a typical consumer would do. Distilled water instead off tap water was used to prepare food as consumed to avoid contamination. The quantity of water added during the cooking process of each of the 12 subsamples was measured by weighing the food at each stage of the preparation process.

Two seasons were captured [69] for five main food groups, which cover staple foods and most of the mean total diet by weight (i.e., cereals, tubers, legumes, vegetables and fruits):

- The rainy season in October 2017; and
- The dry season, or harmattan, in February 2018.

Other food groups were collected during the rainy season only (i.e., nuts and seeds, dairy, oils, beverages and miscellaneous).

Among 335 composite samples, 194 consisted of foods which may be stored in conditions allowing for the growth of moulds and, consequently, are likely to comprise mycotoxins. Those 194 composite samples were selected for mycotoxin analysis. Samples were frozen and shipped by air in coolers with dry ice, within a timeframe never exceeding 24 hours, from the kitchen laboratory (Benin, Cameroon, Mali and Nigeria) to the testing laboratory (Austria).

5.2. Reagents and Chemicals

LC gradient grade methanol and acetonitrile, as well as MS grade ammonium acetate and glacial acetic acid (p.a.), were purchased from Sigma Aldrich (Vienna, Austria). A Purelab Ultra system (ELGA LabWater, Celle, Germany) was used for further purification of reverse osmosis water.

Standards of fungal and bacterial metabolites were obtained either as gifts from various research groups or from the following commercial sources: Romer Labs®Inc. (Tulln, Austria), Sigma-Aldrich (Vienna, Austria), BioAustralis (Smithfiled, Australia), AnalytiCon Discovery (Potsdam, Germany), Fermentek (Jerusalem, Israel), Iris Biotech GmbH (Marktredwitz, Germany), Enzo Life Sciences Europe (Lausanne, Switzerland) and LGC Promochem GmbH (Wesel, Germany). Stock solutions of each analyte were prepared by dissolving the solid substance in acetonitrile, acetonitrile/water 1:1 (v/v),

methanol, methanol/water 1:1 (*v*/*v*) or water. Thirty-four combined working solutions were prepared by mixing the stock solutions of the corresponding analytes for easier handling and were stored at −20 °C. The final working solution was freshly prepared prior to spiking experiments through mixing of the combined working solutions.

5.3. Laboratory Sample Preparation

Twenty millilitres (20 mL) of extraction solvent (acetonitrile/water/acetic acid 79:20:1, *v*/*v*/*v*) were added to 5 g of sample. The samples were extracted for 90 minutes using a GFL 3017 rotary shaker (GFL, Burgwedel, Germany) and subsequently centrifuged for two minutes at 3000 rpm (radius 15 cm) on a GS-6 centrifuge (Beckman Coulter Inc., Fullerton, CA, USA). The extracts were diluted (1:1) with dilution solvent (acetonitrile/water/acetic acid 20:79:1, *v*/*v*/*v*). After appropriate mixing, 5 µL of the diluted extract was injected into the LC-MS/MS system without further pre-treatment.

5.4. LC-MS/MS Parameters

Metabolite analysis was carried out using a 1290 Series HPLC System (Agilent, Waldbronn, Germany) coupled to a QTrap 5500 LC-MS/MS System (Applied Biosystems SCIEX, Foster City, CA, USA) equipped with Turbo Ion Spray electrospray ionization source, as described earlier [70]. Chromatographic separation was performed at 25 °C on a Gemini® C_{18}-column (150 × 4.6 mm i.d., 5 µm particle size) equipped with a C_{18} 4 × 3 mm i.d. security guard cartridge (Phenomenex, Torrance, CA, USA). Confirmation of positive metabolite identification was carried out by two instances of scheduled multiple reaction monitoring (MRMs) which yielded 4.0 identification points according to the European Commission decision 2002/657 [71].

In order to further decrease the limits of detection (LODs) for aflatoxin B1 and ochratoxin A, larger aliquots of 20 µL of the diluted extracts (previously fortified with the related ^{13}C-labelled internal standards) were re-analysed using the QTrap 6500 LC-MS/MS system while keeping all other method parameters constant.

5.5. Quantification and Quality Control

Quantification was performed using external calibration based on serial dilution of a multi-analyte stock solution. Results were corrected using apparent recoveries that were determined for each of the investigated matrices by spiking experiments. The accuracy of the method is verified on a continuous basis by participation in a proficiency testing scheme organized by BIPEA (Gennevilliers, France) with a current success rate (i.e., a z-score between −2 and 2) of >94% of the >900 results submitted.

Supplementary Materials: The following are available online at http://www.mdpi.com/2072-6651/11/1/54/s1. Table S1: Raw Analytical Data.

Author Contributions: The first draft was produced by L.I. and M.S. and was then reviewed by R.K., A.A., and J.-C.L. Technical support was provided by J.-C.L., P.V., and B.L.B. National coordination, including sample collection, was assured by A.A., A.Z.K., S.E.H., and S.E. Food preparation was supervised by A.D.O., C.S.K.J.K., Y.K.D., and L.I. M.S. and R.K. supervised laboratory tests. 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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Conflicts of Interest: The authors declare that there is no conflict of interest.

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The second paper, entitled '*Regional Sub-Saharan Africa Total Diet Study reveals the presence of 164 mycotoxins and other secondary metabolites in foods'* includes the first published occurrence data of the SSA-TDS.

The mycotoxin occurrence data available before this study mainly focused on aflatoxins in Africa. Most frequently, these studies tested raw food commodities including maize and peanuts. 'Preparation as consumed' is a novelty in the four countries covered by the present study. Through this paper, we highlighted that:

- Aflatoxin detects were frequent in maize and peanuts, but also in other core foods such as peanut oil and beans, among many others. We observed frequent exceedances of the Codex standards applicable to peanuts. Codex standards for aflatoxins in maize and oils are not currently available.
- Fumonisins are mainly prevalent in maize but never exceeded the Codex standard.
- Sterigmatocystin was prevalent in peanuts, peanut oil, and cottonseed oil.
- We observed the co-occurrence of aflatoxins, fumonisins, and sterigmatocystin in maize and sorghum.
- Red crude palm oil samples contained the highest number of metabolites, up to 98, including a high ochratoxin A concentration in one composite.
- We described a wide spectrum of fungal, bacterial, and plant secondary metabolites in the typical foods of several African population groups, which was unprecedented.
- In the case of many of these metabolites, toxicological data are not available to date.
- The SSA-TDS methodology captured the seasonal variation of contamination patterns, with more frequent contamination during the rainy season than during the dry season.

Exposure assessment is necessary to be able to draw conclusions concerning the risks that mycotoxins represent in Benin, Cameroon, Mali, and Nigeria. Although available data on the combined effects of mycotoxin mixtures are currently scarce, this study characterized a variety of mycotoxin mixtures encountered in Africa. In addition, to date toxicologists have not determined HBGVs for many of the secondary metabolites detected in this study. Future research concerning some of these substances to determine the toxicological endpoints of single compounds and representative mixtures, would allow this study to make further contributions to risk assessment efforts in Sub-Saharan Africa.

3.2.2. Polycyclic aromatic hydrocarbon occurrence data

The most commonly studied polycyclic aromatic hydrocarbon is, without discussion benzo[a]pyrene (BaP). Most toxicological data applicable to PAHs were obtained from studies concentrating on BaP. Evidence suggests that, in some food samples, BaP may not be detected above the limit of determination, whereas other PAHs are.

The Codex Alimentarius did not propose a Codex maximum limit for PAH, although the institution has endorsed a code of practice for the reduction of the contamination of food with polycyclic aromatic hydrocarbons (PAH) during smoking and drying processes (CAC/RCP 68-2009). The European Commission, however, proposed Commission Regulation (EU) No 835/2011 on 19 August 2011, amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs, in which both BaP and PAH4 are regulated, with specific maximum limits applying to each. The occurrence of BaP, PAH4, and PAH13 (which was subsequently used to complete the risk assessment) are all presented and discussed in the upcoming paper submitted to *Food Control*.

The title we chose for the third paper is '*Polycyclic aromatic hydrocarbons (PAHs) in foods from the first regional Total Diet Study in Sub-Saharan Africa: contamination profile and occurrence data'*.

During an 18-h road trip from Yaounde, central Cameroon, to Garoua in the north, I took the picture shown in Figure 12. I observed similar situations travelling from the south to the north of Benin.

Figure 12: Tuber cossets drying on the side of the road near Ngaoundere, Cameroon

Do cossets that dry in direct contact with the asphalt contain PAHs?

What about non-dried tubers?

Of all the core foods, the selection of smoked fish was an obvious choice given the abundant literature concerning PAHs and smoked fish in Sub-Saharan Africa. Figure 13 shows a picture of smoked fish from a weekly market around Garoua, North Cameroon. The dark colour of the fish is reminiscent of the combustion process.

Figure 13: Typical smoked fish as sold on the market in the North of Cameroon

The PAHs facilitate the preservation of the fish at ambient temperatures (Sikorski and Kalodziejska, 2002). Chaber and Cunningham (2015) tested samples of illegally imported and seized fish from West and Central Africa and found that the concentration of 15 carcinogenic PAHs was in the range of 133 to 406 µg/kg and that of BaP was in the range of 15 to 45 µg/kg. In the meantime, EU regulation (EU) No 835/2011 of 19 August 2011, amending Regulation (EC) No 1881/2006 as regards maximum levels for polycyclic aromatic hydrocarbons in foodstuffs (European Commission, 2011), lowered the maximum limit from 5 µg/kg to 2 µg/kg BaP, as from 01.09.2014. PAHs represent an obvious market access problem as far as smoked fish is concerned.

What about the other food commodities?

But to what extent do PAHs also represent a food safety issue?

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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

Luc Ingenbleek[a,b], Bruno Veyrand[b], Abimbola Adegboye[c], Sètondji Epiphane Hossou[d], Abdoulaye Zié Koné[e], Awoyinka Dada Oyedele[c], Chabi Sika K.J. Kisito[f], Yara Koreissi Dembélé[g], Sara Eyangoh[a], Philippe Verger[h], Jean-Charles Leblanc[i,*], Sophie Durand[b], Anaïs Venisseau[b], Philippe Marchand[b], Bruno Le Bizec[b]

[a] Centre Pasteur du Cameroun, (CPC), Yaounde, Cameroon
[b] LABERCA, Oniris, INRA, Nantes, France
[c] National Agency for Food and Drug Administration and Control, (NAFDAC), Abuja, Nigeria
[d] Agence Béninoise de Sécurité Sanitaire des Aliments (ABSSA), Cotonou, Benin
[e] Agence Nationale pour la Sécurité Sanitaire des Aliments (ANSSA), Bamako, Mali
[f] Laboratoire Central de Sécurité Sanitaire des Aliments (LCSSA), Cotonou, Benin
[g] Laboratoire de Technologie Alimentaire (LTA), Bamako, Mali
[h] World Health Organization (WHO), Geneva, Switzerland
[i] Food and Agriculture Organization of the United Nations (FAO), Rome, Italy

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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.

* Corresponding author. Food and Agriculture Organization of the United Nations (FAO), Rome, Italy.
E-mail address: JeanCharles.Leblanc@fao.org (J.-C. Leblanc).

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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*)

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			*	*
	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

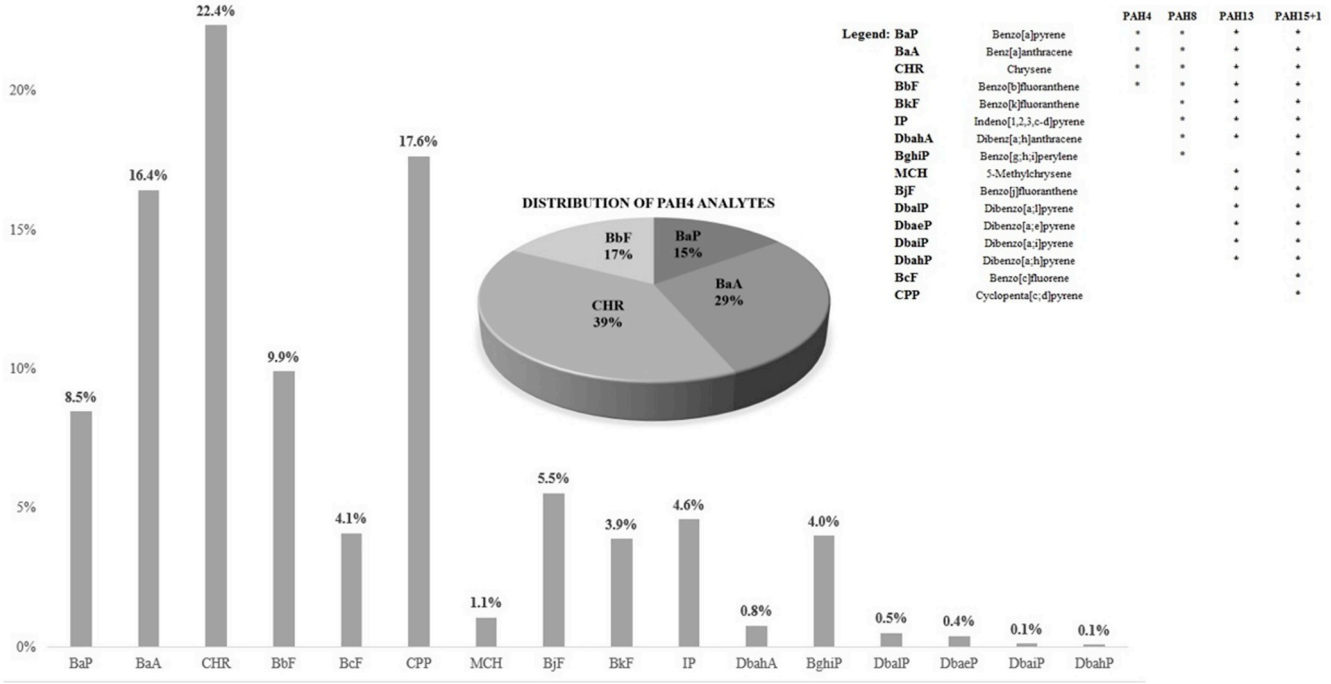

Fig. 1. Relative contribution of PAH congeners to PAH15 + 1.

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

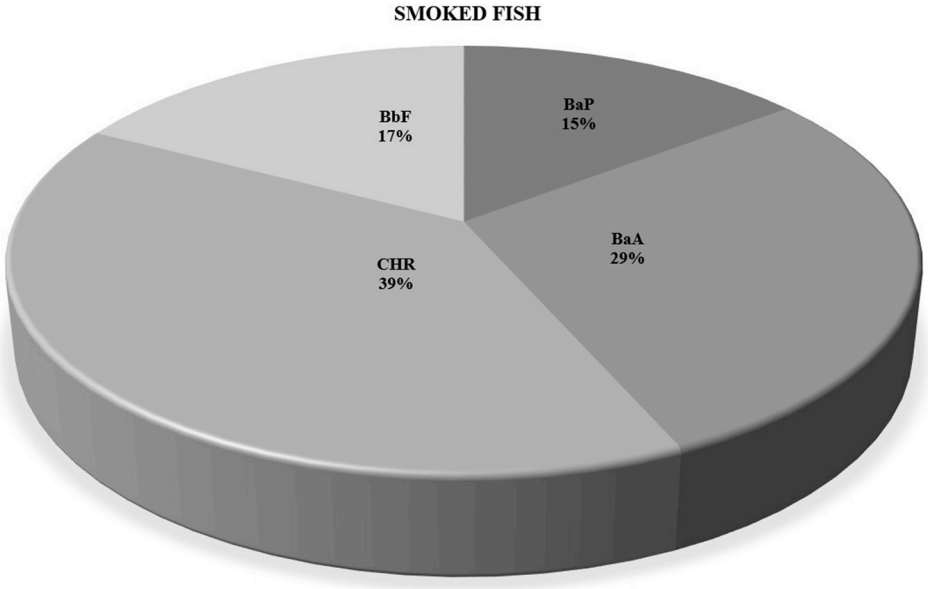

Fig. 2. Proportion of PAH4 congeners in smoked fish.

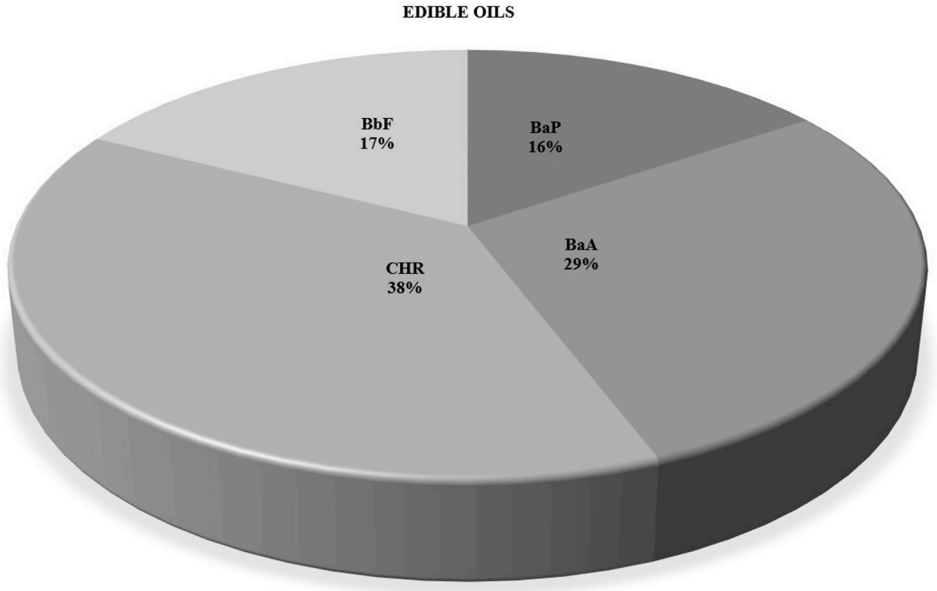

Fig. 3. Proportion of PAH4 congeners in edible oils.

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

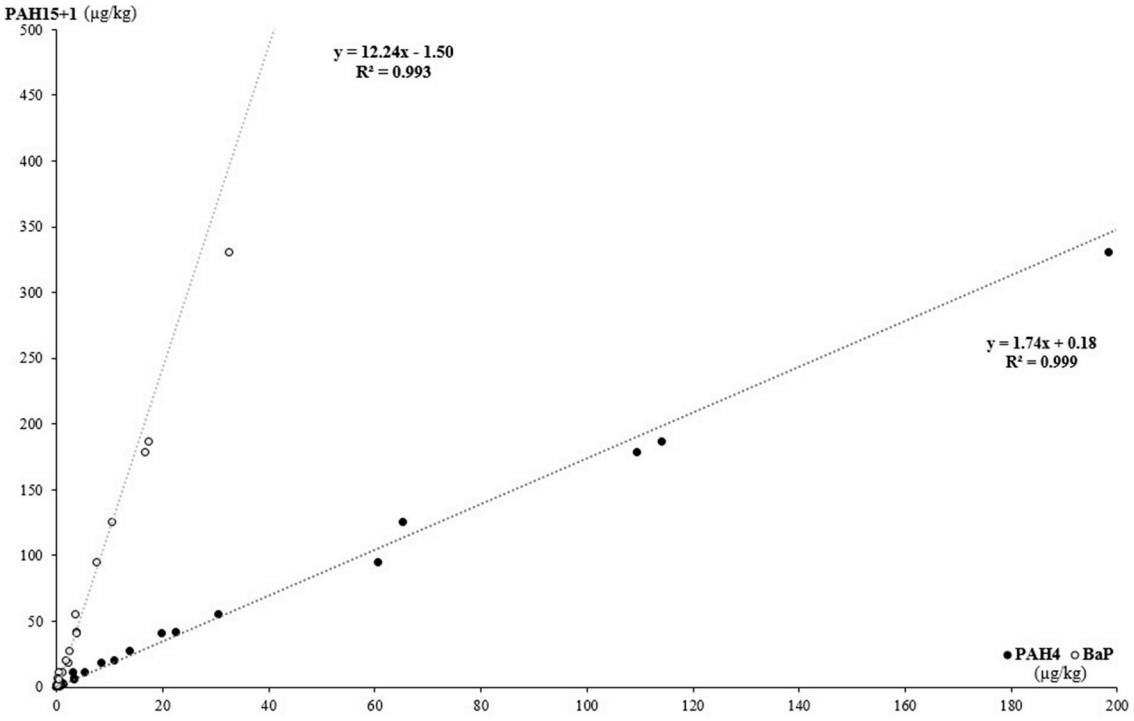

Fig. 4. Correlation of BaP and PAH4 with PAH15 + 1 concentration.

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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The third paper is entitled '*Polycyclic aromatic hydrocarbons (PAHs) in foods from the first Regional Total Diet Study in Sub-Saharan Africa: contamination profile and occurrence data'*.

Through this paper, we highlighted:

- The relatively high concentrations of PAHs in smoked fish and edible oils
- Both in the case of smoked fish and edible oils, the contamination profile has the typical signature of a combustion process.
- The presence of PAHs in foods prepared from dried tubers compared to foods prepared from fresh tubers
- The concentration of PAHs in smoked fish varied extensively from one centre to another, including within the same country.
- Core foods such as dehydrated and concentrated milk and sugar contained very low concentrations of PAHs.
- PAH levels exceeded the EU regulations in 100% of smoked fish and 50% of edible oils.

Moreover, an interesting feature of the contamination profile obtained in the SSA-TDS samples was the unusually high concentration of cyclopenta[c,d]pyrene (CPP) in the vicinity of threefold the proportions reported by previous studies. A tentative explanation of the high CPP content is that some of it originates from a reaction of heated pyrene in the presence of sand silica, which was previously simulated *in vitro*. If this hypothesis is correct and translates to actual food processes, ensuring sand-free or optimum hygiene conditions for smoking and heating processes could contribute to lowering the PAH concentrations in foods. Unfortunately, most PAH occurrence studies conducted in Africa before the SSA-TDS did not report the CPP concentration. Including CPP in multi-analyte tests in future research would contribute to a better understanding of this specific contamination profile encountered in Africa.

This paper, however, does not inform the extent to which PAHs represent a threat to human health. The exposure assessment is necessary to be able to draw conclusions concerning the risk that PAHs represent in Benin, Cameroon, Mali, and Nigeria.

3.2.3. Pesticide occurrence data

According to the EU Rapid Alert System for Food and Feed, 99% of alerts from Benin, Cameroon, Mali, and Nigeria (2000-2016) resulted from 10 pesticides, including endosulfan, chlorpyrifos, profenofos, dichlorvos, dimethoate, ethephon, omethoate, trichlorfon, cypermethrin, lambda-cyhalothrin, and permethrin (European Commission, 2016a).

Figure 14: Pesticide bottles sold in a food market of Pitoa, North Cameroon

Pesticide are easily accessible in Africa, and are sold in main food markets, as shown in Figure 14. Small packaging means that farmers can afford pesticides to increase crops production and yield. In Figure 15, children's food is contained in soda bottles and pesticides containers. In the village where I took this picture, running water was not available. Water is scarce in Banikoara, and villagers draw their necessary water from the community well. It is impossible to be certain whether the pesticide container was properly washed before it was converted into a food container.

Figure 15: Pesticide container transformed into a flask used by schoolchildren in Banikoara, North Benin

The purpose of a TDS is not to capture contamination resulting from such practices. The lack of awareness in this situation is, however, puzzling and needs addressing. The fact that some parents do not realize that recycling a pesticide container could expose their children to a chemical hazard also draws attention to the fact that contamination sources may be unexpected.

Omari and colleagues (2018) observed that the perception of food safety concerns such as pesticide residues in Africa was rather lower than that of other food safety issues relating to hygiene. Awareness about the risks is influenced by the level of knowledge and possibly by gender. Although most occurrence studies focus on pesticides in vegetables and fruits, we decided to look for pesticides in every food group. We also tried to capture the seasonal variation in contamination patterns.

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Sub-Saharan Africa total diet study in Benin, Cameroon, Mali and Nigeria: Pesticides occurrence in foods

Luc Ingenbleek[a,b], Renwei Hu[c], Lionel Lopes Pereira[c], Anaïs Paineau[c], Isabelle Colet[c], Abdoulaye Zié Koné[d], Abimbola Adegboye[e], Sètondji Epiphane Hossou[f], Yara Dembélé[g], Awoyinka Dada Oyedele[e], Chabi Sika K.J. Kisito[h], Sara Eyangoh[a], Philippe Verger[i], Jean-Charles Leblanc[j,*], Bruno Le Bizec[b]

[a] Centre Pasteur du Cameroun, (CPC), Yaoundé, Cameroon
[b] LUNAM Université, Oniris, LABERCA, Nantes, France
[c] Inovalys – Le Mans, Official Laboratory of Analysis, France
[d] Agence Nationale de la Sécurité Sanitaire des Aliments (ANSSA), Bamako, Mali
[e] National Agency for Food and Drug Administration and Control, (NAFDAC), Abuja, Nigeria
[f] Agence Béninoise de Sécurité Sanitaire des Aliments (ABSSA), Cotonou, Benin
[g] Laboratoire de Technologie Alimentaire, (LTA), Bamako, Mali
[h] Laboratoire Central de Sécurité Sanitaire des Aliments (LC-SSA), Cotonou, Benin
[i] World Health Organization (WHO), Geneva, Switzerland
[j] Food and Agriculture Organization of the United Nations (FAO), Rome, Italy

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ABSTRACT

In the framework of the first regional Total Diet Study in Sub-Saharan Africa, 3696 foodstuffs, commonly consumed in Benin, Cameroon, Mali and Nigeria were purchased, prepared as consumed and pooled into 308 composite samples. Those core foods were tested for up to 470 pesticides residues by liquid and gas chromatography coupled with tandem mass spectrometry.

39 pesticides were detected with 294 total occurrences, including 47.3% organophosphate pesticides and 35.7% pyrethroids. More specifically, 6 substances represented 75.5% of all 3 organophosphates and 3 pyrethroids: chlorpyrifos (22.4%) cypermethrin (18.0%) dichlorvos (13.6%), lambda cyhalothrin (8.2%), permethrin (7.5%) and profenofos (5.8%).

One pesticide or more was detected in 45.8% of samples.

Strikingly, several pesticides were quantified in 2 composite samples of smoked fish from Mali: chlorpyrifos (5236–18 084 µg/kg), profenofos (30–182 µg/kg), cypermethrin (22–250 µg/kg), cyfluthrin (16–117 µg/kg), lambda cyhalothrin (9–17 µg/kg) and permethrin (3–6 µg/kg).

1. Introduction

The “pesticide” terminology describes a variety of substances, including among others insecticides, herbicides, fungicides and growth regulators (WHO, 2010). Loss of agricultural commodities, curtailed by pesticide utilization, needs to be balanced with negative health effects. Acute toxicity, accidentally (Pouokam, Lemnyuy-Album, Ndikontar, & Sidatt, 2017) or through self-poisoning mainly, can lead to nausea, cough, skin and eye irritation and respiratory failure, tachycardia, ulceration of the lips, tongue, pharynx and larynx and an estimated 300,000 deaths per year (Eddleston, 2015; Pearson et al., 2017). In this study, we aimed to generate mean contamination data, adequately reflecting the chronic exposure to pesticides from a typical diet, in specific areas. Consequences of chronic exposure to pesticides may include metabolic disorders, such as diabetes (Montgomery, Kame, Saldana, Alavanja, & Sandler, 2008; Starling et al., 2014), reproductive disruption (Merviel et al., 2017; Ueker et al., 2016), genotoxicity (Eastmond & Balakrishnan, 2010) carcinogenesis (Engel et al., 2017; Uyemura, Stopper, Martin, & Kannen, 2017), neurological (Muñoz-Quezada et al., 2016; Schmidt et al., 2017; Shelton et al., 2014; Wagner-Schuman et al., 2015) and sensory disorders (Sturza et al., 2016).

One way of assessing the dietary exposure of populations to food

* Corresponding author.
E-mail address: JeanCharles.Leblanc@fao.org (J.-C. Leblanc).

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chemicals, such as pesticide residues, is the Total Diet Study (TDS) approach (EFSA, 2011). Two specific aspects characterize a TDS: (1) the representativeness of the sampling and (2) the preparation of the samples "*as consumed*".

WHO and FAO endorse the TDS methodology (EFSA, 2011) as a pertinent public health risk assessment tool.

A regional Sub-Saharan Africa TDS (SSA-TDS), which generated the data presented in this paper, was implemented by FAO in Benin, Cameroon, Mali and Nigeria between 2014 and 2018, together with the four national food safety authorities, in close collaboration with Centre Pasteur of Cameroon (CPC) and WHO (FAO, 2014; Ingenbleek, Sulok, et al., 2019; Ingenbleek, Veyrand, et al., 2019). The purpose of this project was to assess the contamination levels of the food of 8 African population groups. The dietary exposure of those population groups will then be compared with existing health-based guidance values or end points. The study methodology including the derivation of food consumption data from national household budget surveys was described elsewhere (Ingenbleek et al., 2017).

In this paper, we are presenting the occurrence of pesticides in composite samples, each resulting from the pooling of 12 subsamples. These composites are representative of the food consumption habits of 3 population groups living in coastal areas (Duala, the Littoral of Benin and Lagos) and 5 population groups in non-coastal areas (Bamako, the Borgou region of Benin, Kano, the North of Cameroon and Sikasso). Although our analytical plan included organochlorine pesticides, we will not discuss their occurrence in this paper, but in another article (in preparation) with other persistent organic pollutants (POPs).

2. Experimental

2.1. Sample selection and preparation of foods as consumed

Food consumption data were derived from household budget surveys validated by National Statistics Authorities, from Benin, Cameroon, Mali and Nigeria and gathering a total of 72,979 households. Core foods of each study centre were selected on the basis of the relative importance of mean food consumption by weight (Ingenbleek et al., 2017), so as to cover at least 90% of the mean total diet in grams per adult male equivalent per day (g/AME/d).

Each core food was sampled through available representation criteria, such as market share or the origins of the food, 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 recipe books (Gautier & Mallet, 2006; Madubike, 2013; Nya-Njike, 1998; Vinakpon-Gbaguidi, 2003), using inert kitchen utensils. These references are considered as representative of the diet of the study populations and were therefore selected by the national competent authorities. These recipe books allow the identification of the processes used in the preparation of the foods, especially cooking time and temperature. The actual recipes were however not prepared, as each composite sample only contained one food type or ingredient. The inedible parts were removed at the preparation stage, as a typical consumer would do. In order to avoid contamination, distilled water was used to prepare food as consumed, instead of tap water, which is also part of the sampling. The quantity of water added during the cooking process of each of the 12 subsamples was measured by weighing the food at each stage of the preparation process.

Two seasons were captured for 5 main food groups, which cover staple food and most of the mean total diet by weight (cereals, tubers, legumes, vegetables and fruits):

- the rainy season in October-November 2017;
- the dry season or harmattan in February 2018.

Other food groups were collected during the rainy season only (nuts and seeds, dairy, oils, beverages and miscellaneous).

Tap water was also tested for pesticide residues, with 16 composite samples of 12 representative subsamples by study centre for both the rainy and the dry season.

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

2.2. Chemicals and reagents

Acetone (pestinorm) and petroleum ether (40–60 °C) were obtained from VWR, acetonitrile (for LCMS grade), ethyl ether (pestipur), heptane (≥99%), hexane (pestipur), methanol (for HPLC grade), toluene (for analysis ISO ACS reag) and anhydrous sodium sulphate from Carlo Erba, dichloromethane (pestipur) from Biosolve, *n*-dodecane (For synthesis) from Merck and ammonium acetate from Fisher Chemical (France). Salts mixture 1 (4 g of anhydrous magnesium sulphate, 1 g of NaCl, 1 g ± 0.1 g of trisodium citrate dihydrate and 0.5 g of disodiumhydrogencitrates hydrate), mixture 2 (150 mg of Primary Secondary Amines (PSA) and 900 mg of anhydrous magnesium sulfate), mixture 3 (150 mg of PSA, 150 mg C18, 900 mg of anhydrous magnesium sulfate) and mixture 4 (150 mg PSA, 45 mg graphitized black carbon, 900 mg magnesium anhydrous sulfate) were purchased from Macherey Nagel (Chromabond Mix I, III, VI et V). Analytical standards (purity from 81.2% to 99.5%) of pesticides and metabolites were purchased from Dr. Ehrenstorfer GmbH (Germany) and Sigma-Aldrich. PTFE filter (Chromofil – 0.45 µm, for ethephon) was from Macherey Nagel, SPE C18 cartridge (12 ml/2 g; Sep-Pak®) from Waters (France) and Florisil cartridge (6 ml/1 g; Chromabond®) from Macherey Nagel.

Stock solutions of individual pesticide standards (1 mg/ml) were prepared by weighing in 20 ml of toluene, methanol, or acetonitrile according to their solubility and stability. Intermediate stock standard mixtures of pesticide were prepared in toluene (for GC mixture) and in methanol or acetonitrile (for LC mixture) and were stored at −20 °C. Working standard solutions were prepared by dilutions of the intermediate stock standard solutions in ethanol and stored at 4 °C.

2.3. Analytical methods

Three analytical methods were used in this study.

2.3.1 Multiresidue method QuEChERS for food of plant origin

The method used (Quick Easy Cheap Effective Rugged Safe, QuEChERS) was based on the standard method NF EN 15662 (AFNOR, 2009). 470 pesticides were analysed, including 122 compounds on GC and 348 on LC in this method.

The sample was blended to obtain a homogeneous sample, and 5 g (± 1%) for water-poor products or 10 g (± 1%) for water-rich products were weighed in a 50 ml screw-cap centrifuge tube and extracted with 10 ml of acetonitrile containing 100 µl of the internal standards (IS: HCH gamma D6, Chlorpyrifos-methyl D6, Diazinon D10 and PCB 170 for GC and Bentazone D6, Carbofuran D3, Isoproturon D6, Terbuthylazine D5, Propiconazole D5 for LC) at 2.5 µg/ml. After salts (mixture 1) addition, the mixture was shaken intensively and centrifuged for 20 min at 3000 rpm and kept at −15 °C for phase separation. 6 ml of the organic phase was taken for the clean-up with different bulk sorbents (the mixture 2, 3 or 4) depending on the type of matrix and $MgSO_4$ anhydrous to remove the residual water. Extracts were shaken by vortex and centrifuged for 15 min at 3000 rpm and kept at −15 °C. 1 ml of supernatant was transferred into a vial for LC-MS/MS analysis. To get better specificity and sensitivity for LC-MS/MS analysis, 348 pesticides were divided into two groups according to their properties. So, 2 injections were made on the LC-MS/MS: one for methanol mobile phases (331 compounds) and another for acetonitrile mobile phases (17 compounds shown in Section 2.4.3). Another 1 ml of supernatant was transferred into a vial in the presence of 25 µl of *n*-dodecane and evaporated to dryness at 35 °C under nitrogen steam. The residue was dissolved in 225 µl of a solvent mixture (heptane/acetone;

90/10; v/v). The final extract was then transferred into the conical vial for GC–MS/MS analysis after ultrasonic shock.

2.3.1. The ANSES POPs 10 and POPs 11 methods for food of animal origin

The POPs 10 method (ANSES, PBM Pest LSA-INS-0165, Version 04, 2015) for analysis of organochlorines and the POPs 11 method (ANSES, PBM Pest LSA-INS-0166, Version 05, 2015) for analysis of organophosphorus are official methods recognized by the Ministry of Agriculture for the official control of pesticides in animal commodities as part of surveillance plans. In this study, 70 of the pesticides were analysed with these methods.

2.3.1.1. Extraction for the ANSES POPs 10 and POPs 11 methods. First, the fat in sample was extracted, and its percentage (MG) calculated and then the pesticides were analysed in the extracted fat.

The sample was blended to obtain a homogeneous sample, and 10 g mixed with 25 g of sodium sulphate and 25 g of Fontainebleau sand in a mortar to obtain a dry and brittle product. The extraction column was sealed with glass wool and a 2-cm thick layer of anhydrous sodium sulphate, and the mixture obtained previously was then poured. The column was then eluted with 3 × 50 ml of solvent mixture (hexane/acetone; 2/1; v/v), and the mortar thoroughly rinsed. The eluate was collected in a zymark tube surmounted by a funnel containing anhydrous sodium sulphate, and the funnel rinsed with 10 ml of the solvent mixture, which was collected into the same tube. The solvent mixture was then evaporated under nitrogen to about 5 ml at 35 °C, the fat extract transferred to a tared tube, and the zymark tube rinsed with 3 × 1 ml of hexane, which was collected into the tared tube. The fat extract was evaporated under nitrogen at about 35 °C, and the amount of fat weighed at constant weight.

2.3.1.2. Cryogenic extraction of pesticides for the POPs 10 and POPs 11 methods. A 0.5 g aliquot of the fat obtained was taken in a tube of 12 ml. 100 µl of the IS-1 (HCH gamma D6, Chlorpyriphos methyl D6 and Cypermethrin D6 at 0.2 µg/ml) for POPs 10 or the IS-2 (Diazinon D10 and Propiconazole D5 at 2.5 µg/kg) for POPs 11 and then 3 ml of solvent mixture (acetonitrile/dichloromethane; 75/25; v/v) were added. The tube was vortexed, and centrifuged for 20 min at 3000 rpm and kept at −15 °C. The supernatant was then transferred to another tared tube. A second cryogenic centrifugation was made with the same solvent mixture. Both extracts were combined and evaporated to dryness under a stream of nitrogen (set at 35 °C).

2.3.1.3. Purification on SPE columns for the POPs 10 and POPs 11 methods. Two types of cartridges (C18 and Florisil) were used for POPs 10 and only one (C18) for POPs 11.

2.3.1.3.1. Purification on a cartridge C18 (POPs 10). The cartridge was conditioned with 5 ml of petroleum ether, 5 ml of acetone and then 2 × 5 ml of methanol. The cryogenic extract was dissolved into 1 ml of acetonitrile and deposited on the C18 cartridge, and the cartridge eluted with 10 ml of acetonitrile by rinsing the sample tube. The eluate was evaporated to dryness under a stream of nitrogen (set at 35 °C) in the presence of 100 µl of *n*-dodecane. 900 µl of hexane was added and shaken for 1 min, by vortex, then ultrasonically for 5 min, for the next purification step on the Florisil cartridge.

2.3.1.3.2. Purification on a cartridge of Florisil (POPs 10). The cartridge was conditioned with 10 ml of hexane. The extract obtained after C18 was deposited on the Florisil cartridge. The cartridge was eluted by rinsing the sample tube with 2 ml then 10 ml of solvent mixture (petroleum ether/ethyl ether; 98/2; v/v), then 12 ml of another solvent mixture (petroleum ether/ethyl ether; 85/15, v/v). The eluate was evaporated to dryness under a stream of nitrogen (set at 35 °C) in the presence of 100 µl of *n*-dodecane. 900 µl of hexane was added and shaken for 1 min, by vortex, then sonicated for 5 min, and transferred into an injection vial suitable for GC.

2.3.1.3.3. Purification on a cartridge C18 (POPs 11). The cartridge was conditioned with 5 ml of methanol, 5 ml of acetone and then 5 ml of solvent mixture (acetonitrile/dichloromethane; 95/5; v/v). The cryogenic extract was dissolved into 1 ml of the solvent mixture and deposited on the C18 cartridge, and the cartridge eluted with 10 ml of methanol by rinsing the sample tube. The eluate was evaporated to dryness under a stream of nitrogen (set at 35 °C), and the residue dissolved in 1 ml of ethanol. 500 µl of the solution was taken into a vial of 1 ml containing 50 µl of *n*-dodecane, and evaporated to dryness under a stream of nitrogen (set at 35 °C). The residue was then dissolved with 450 µl of hexane for GC-MS/MS analysis. Another half of the ethanol solution was evaporated to dryness under a stream of nitrogen (set at 35 °C), and the residue dissolved with 500 µl of solvent mixture (methanol/H_2O; 50/50; v/v) for LC-MS/MS analysis.

2.4. GC-MS/MS and LC-MS/MS measurements

2.4.1. GC-MS/MS for multiresidue QuEChERS

A trace GC Ultra (Thermo Scientific) equipped with a split/splitless injector and a Quantum XLS Triple Quadrupole was used. A CP SIL 8CB column (Agilent) of 60 m × 0.25 mm ID × 0.25 µm was used for chromatographic separation, with helium (99.999%) as carrier gas (1.5 ml/min.). The mass spectrometer was operated in electron impact (EI) positive ionization mode, and data were acquired using selected reaction monitoring (SRM). Argon (1.5 mTorr) was used as collision gas, and the source temperature was set at 250 °C. The injection volume was 1 µl in splitless mode.

2.4.2. GC-MS/MS for POPs 10 and POPs 11

A trace GC Ultra (Thermo Scientific) equipped with a programmed temperature vaporizer (PTV) injector and a Quantum XLS Triple Quadrupole was used. A CP SIL 8CB column (Agilent) of 60 m × 0.25 mm ID × 0.25 µm was used for chromatographic separation, with helium (99.999%) as carrier gas (1.5 ml/min). The mass spectrometer was operated in electron impact (EI) positive ionization mode, and data were acquired using selected reaction monitoring (SRM). Argon (1.5 mTorr) was used as the collision gas, and the source temperature was set at 280 °C. The injection volume was 2 µl in PTV mode.

2.4.3. LC-MS/MS for multiresidue QuEChERS

A Shimadzu LC (NEXERA X2) system coupled to a triple-quadrupole mass spectrometer was used (8060, Shimadzu, Kyoto, Japan). Chromatographic separation was carried out at 40 °C using an Accucore C18 150 mm × 2.1 mm × 2.6 µm column (Thermo Electron, France) with a pre-column (Accucore C18 2.1 mm × 2.6 µm). The methanol mobile phases were methanol + 2 mM ammonium acetate + 0.002% formic acid + 5 ml H_2O and H_2O + 2 mM ammonium acetate + 0.002% formic acid. The acetonitrile mobile phases were acetonitrile + 0.05% formic acid and H_2O + 0.05% formic acid. Injection volume was 5 µl, and the flow at 0.3 ml/min. ESI (electrospray ionization) was operated in a positive/negative polarity switching mode and data were acquired in multiple reaction monitoring mode (MRM). Interface voltage was at 1500 V, interface temperature, at 350 °C and CID Gas at 230 kPa.

2.4.4. LC-MS/MS for organophosphorus (POPs 11)

A LC (Dionex Ultimate 3000) system coupled to a triple-quadrupole mass spectrometer was used (TSQ Quantiva, Thermo Scientific, France). Chromatographic separation was carried out at 40 °C using an Accucore C18 150 mm × 2.1 mm × 2.6 µm column (Thermo Electron, France) with a pre-column (Accucore C18 2.1 mm × 2.6 µm). The mobile phases were methanol + 0.5 mM ammonium acetate and H_2O + 0.5 mM ammonium acetate. Injection volume was 5 µl, and the flow at 0.3 ml/min. ESI was operated in a positive/negative polarity switching mode and data were acquired in MRM. The mass spectrometer parameters were: spray voltage of positive ion of 4000 V and

negative of 2500 V; sheath gas of 40 (arbitrage); aux gas of 15 (arb); ion transfer tube temperature of 300 °C and vaporizer temperature of 300 °C.

2.5. Analytical performances

In this study, all applied methods of analysis have been accredited by COFRAC (**Co**mité **Fr**ançais d'**Ac**créditation, France). The QuEChERS method has been used since 2010, the POPs 10 method since 2011, the POPs 11 method since 2014.

The means recoveries of these methods were between 70% and 120% and their precision (RSD, Relative Standard Deviation %) was less than 20% for at least the accredited analytes. Therefore, it meets the requirements of the EU international standards (SANTE 31181, 2017).

The limits of quantification (LQ) for the QuEChERS method were different depending on the compounds, and were between 1 µg/kg (for 10 priority compounds: chlorpyrifos, cypermethrin, dichlorvos, dimethoate, endosulfan, lambda cyhalothrin, omethoate, permethrin, profenofos and trichlorfon) and 10 µg/kg (for others). The LQ for POPs 10 and 11, depending on the compounds and matrices considered, was between 1 and 8 µg/kg.

2.6. Internal quality controls

The quality of the analysis is ensured by analyzing internal reference material as well as blank control for each analytical series. The accuracy of the method is ensured by regular participation in inter laboratory tests such as EUPT (EU-Proficiency Tests), BIPEA (Interprofessional Bureau of Analytical Studies) and FAPAS (Proficiency testing from Fera).

In each batch of analyses, matrix-matched calibration with at least 4 levels was performed by using an internal standard method for quantification to reduce systematic errors and matrix effects; the deviations of the measured signal from their values estimated with linear regression (residuals) was controlled (± 20%); a reagent blank and a matrix blank were checked for contamination (< LD) and a checkpoint (QC) supplemented close to LQ was conducted for performance verification (individual recovery from 60% to 140% and means one from 70% to 120%).

3. Results

3.1. Occurrence of pesticides

No pesticide residue was detected > LD in any of the 16 tap water composite samples.

The list and proportion of detected pesticide residues with concentration > LD is presented in Table 1. Fig. 1 shows that 89.8% of pesticide occurrences concern insecticides and 4.8% are fungicides.

Organophoshates (47.3% of detected compounds) and pyrethroids (35.7%) represented the majority of occurrences, while neonicotinoids (acetamiprid and imidacloprid) represented 5.1% of all occurrences.

Other pesticides (21 analytes) represented 35 incidences or 11.9% of total occurrence.

3.2. Co-occurrence of pesticides

Fig. 2 shows the co-occurrence of pesticide in the SSA-TDS composite samples. Of 308 composite food samples, no pesticide residue was detected in 167 samples (54.2%). Whereas one pesticide only was detected in 72 samples (23.4%), 69 samples contained more than one and up to 8 pesticides (22.4%) and 36 samples contained 3 pesticides or more (11.7%).

Table 1
Occurrence of detected pesticides.

Pesticide	Chemical class	Main use	Occurrences (n)
Chlorpyrifos	OP	INS	66
Cypermethrin	PYR	INS	53
Dichlorvos	OP	INS	40
Lambda Cyhalothrin	PYR	INS	24
Permetrhin	PYR	INS	22
Profenofos	OP	INS	17
Acetamiprid	NN	INS	11
Piperonyl butoxide	Other class	SYN	7
Pirimiphos methyl	OP	INS	6
Chlormequat	Other class	GR	4
Imidacloprid	NN	INS	4
Deltamethrin	PYR	INS	3
Dimethoate	OP	INS	3
Indoxacarb	Other class	INS	3
Omethoate	OP	INS	3
Chlorantraniliprole	Other class	INS	2
Cyfluthrin	PYR	INS	2
Fenpropimorph	Other class	FUN	2
Imazalil	Other class	FUN	2
2,4-D	Other class	HER	1
3,5-Dichloroaniline	Other class	ME	1
Acrinathrin	PYR	INS	1
Atrazine	Other class	HER	1
Azoxystrobin	Other class	FUN	1
Boscalid	Other class	FUN	1
Carbendazim	Other class	FUN	1
Chlorpropham	Other class	HER	1
Chlorpyrifos methyl	OP	INS	1
Malathion	OP	INS	1
Metalaxyl	Other class	FUN	1
Orthophenylphenol	Other class	FUN	1
Phthalimide	Other class	ME	1
Propamocarb	Other class	FUN	1
Propiconazole	Other class	FUN	1
Pyrimethanil	Other class	FUN	1
Thiabendazole	Other class	FUN	1
Triazophos	OP	INS	1
Trichlorfon	OP	INS	1
Tricyclazole	Other class	FUN	1
TOTAL			**294**

Legend: FUN, Fungicide; GR, Growth regulator; INS, Insecticide; HER, Herbicide; ME, Metabolite; NN, Neonicotinoid; OP, Organophosphate; PYR, Pyrethroid; SYN, Synergist.

3.3. Contamination by most prevalent pesticides

The sum of 6 most frequently detected substances chlorpyrifos (22.4%), cypermethrin (18.0%), dichlorvos (13.6%), permethrin (7.5%), lambda cyhalothrin (7.5%) and profenofos (5.8%) covered 75.5% of 294 occurrences (Fig. 3). Like 88.1% of pesticide occurrences in SSA-TDS samples these 6 analytes are either organophosphates, or pyrethroids.

Table 2 and Table 3 present the occurrence of these 6 pesticides, by food group and by study centre respectively.

So as to take into account the measurement uncertainty due to censored data *(non-detected analytes)* the mean concentrations presented are lower bound and upper bound values (LB-UB), which means that we use as concentration:

- Zero (LB) and the limit of detection (UB) for non-occurrence;
- The limit of detection (LB) and limit of quantification (UB) for detected but non quantified data.

3.3.1. Chlorpyrifos

Out of 66 chlorpyrifos occurrences, 37 exceeded LQ (1 µg/kg). The highest chlorpyrifos concentrations (Table 3) were quantified in two smoked fish samples composite samples from Mali (Bamako: 18 084 mg/kg and Sikasso: 5236 µg/kg). Those high concentrations alone

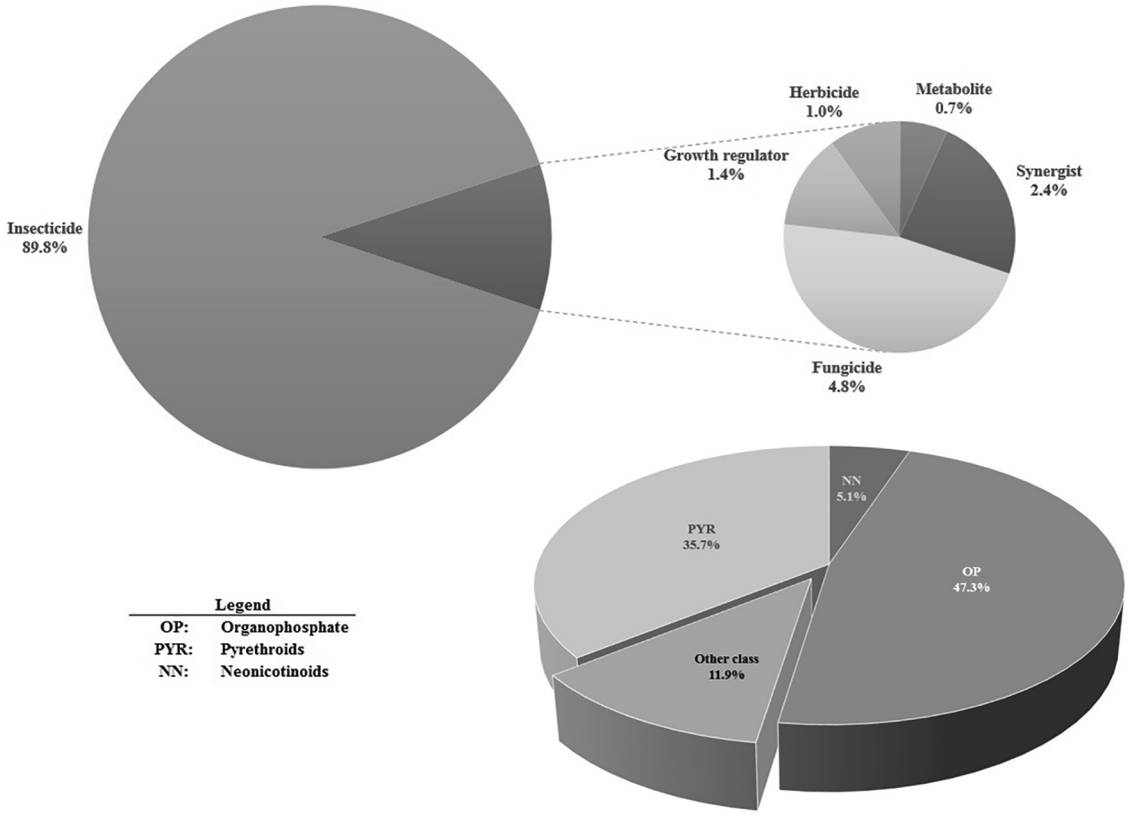

Fig. 1. Proportions of pesticide occurrences by main use and by chemical class.

considerably lift the mean chlorpyrifos concentration in fish up, which thereby becomes the highest concentration of all food groups (Table 2). Ten samples contained chlorpyrifos levels above 10 µg/kg, including one composite of peanut oil from Kano: 95 µg/kg, two composites of tomato from Duala: 26 and 91 µg/kg, rice from Cotonou (Benin): 56 µg/kg, leafy vegetables from Garoua: 33 µg/kg, vegetables from Lagos: 29 µg/kg, bread from Duala: with 14 µg/kg and peanuts from Bamako: with 11 µg/kg.

3.3.2. Dichlorvos

Of 40 dichlorvos occurrences, 38 exceeded LQ (1 µg/kg) and 11 composites comprised concentrations above 10 µg/kg. Dichlorvos was often detected in legumes (32% > LD), cereals (21% > LD), and tubers (20% > LD), as shown in Table 2. Dichlorvos was most frequently detected in Nigerian foods (Lagos 56% and Kano 15%), followed by Cameroon (Duala: 8% and Garoua 7%). Consistently, the maximum dichlorvos concentration (61 µg/kg) was quantified in a bean sample from Kano (Nigeria). Dichlorvos was only detected once in Benin

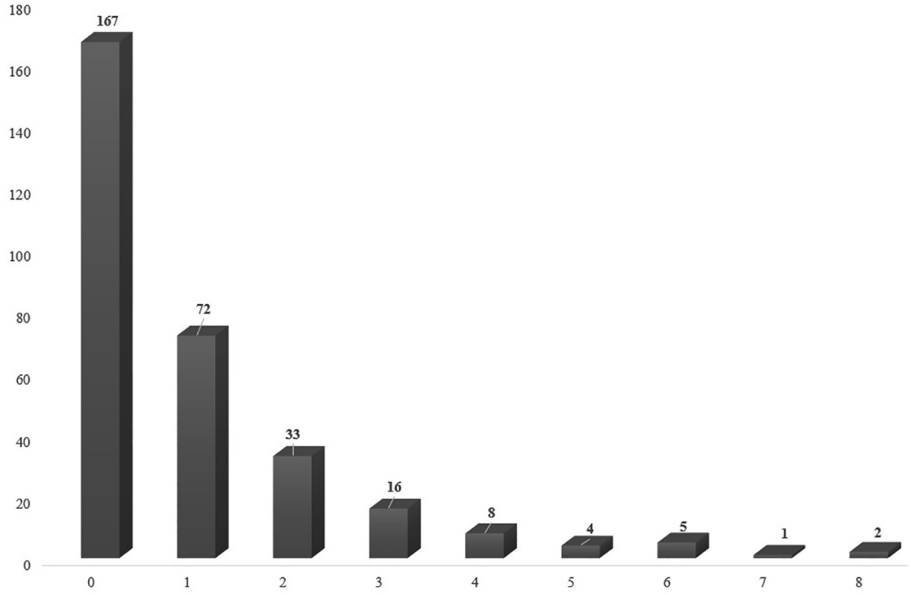

Fig. 2. Number of co-occurrences in SSA-TDS composite samples.

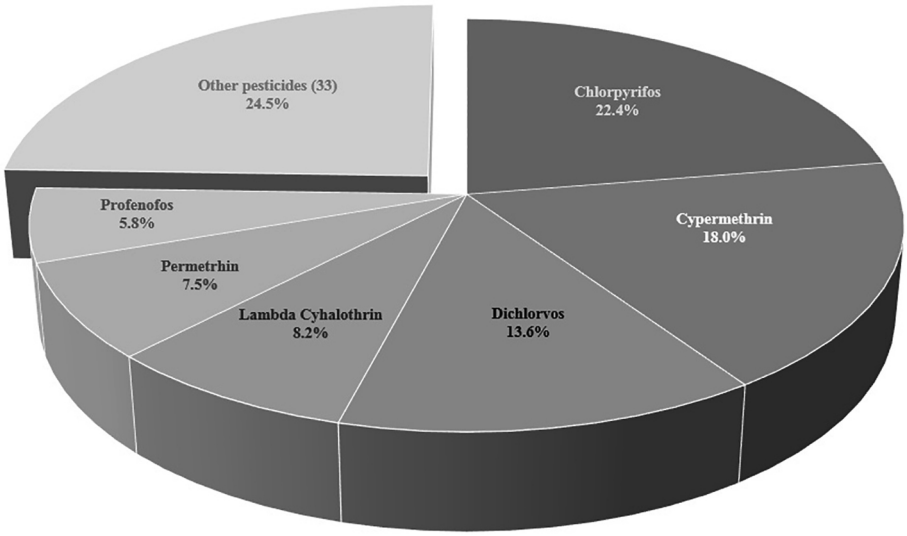

Fig. 3. Proportion of occurences of 6 most frequently detected pesticides in SSA-TDS composite samples.

(cassava: 1.0 µg/kg) and was not detected at all in Malian foods (Table 2).

3.3.3. Profenofos

Profenofos was more frequently detected in foods from Mali (Bamako: 9% and Sikasso 16%) than in other study centres. Concentrations in excess of 10 µg/kg were only quantified in Mali in two composites of leafy vegetables (16–555 µg/kg) and two composites smoked fish (30–182 µg/kg) collected in two distinct centres. Profenofos was also quantified (> LQ) in Bamako, in tomato (2 µg/kg) and plantain (3 µg/kg). Apart from samples collected in Mali, only one composite sample from Lagos exceeded the analytical limit of quantification (chili pepper: 7 µg/kg).

3.3.4. Cypermetrin

The highest cypermethrin concentrations were quantified in two tomato composite samples from Duala (240–258 µg/kg) and in smoked fish from Bamako (250 µg/kg). Cypermethrin was quantified in leafy vegetables collected during the dry season in Cameroon (Duala: 235 µg/kg; Garoua: 126 µg/kg) but was not detected (LD = 0.3 µg/kg) in leafy vegetables collected during the wet season. The same observation applies to leafy vegetables in Mali, which contained cypermethrin during the dry season (2–34 µg/kg) but was not detected during the rainy season. Similarly, okro collected contained cypermethrin in Kano (4 µg/kg) and Garoua (3 µg/kg) during the dry season, but concentrations did not exceed LD in samples collected during the wet season.

3.3.5. Lambda cyhalothrin

Lambda cyhalothrin was detected 24 times including in 16 vegetable samples (29% > LD) and in smoked fish from 3 different study centres (Garoua: 5 µg/kg; Bamako: 9 µg/kg; Sikasso: 17 µg/kg). 8/12 tomato composites also contained lambda cyhalothrin (ND-28 µg/kg), as well as 3/8 of leafy vegetables (ND-41 µg/kg), in which the maximum concentration was quantified.

We also quantified lambda cyhalothrin in beans from Borgou (8 µg/kg) and from Kano (12 µg/kg), bread from Bamako (1 µg/kg) and maize from the littoral of Benin (1 µg/kg).

3.3.6. Permethrin

Several bean composites samples enclosed permethrin concentrations (7/16 or 44%) unlike other legumes (peanuts and peas: 0%), which results in a permethrin detection rate of 25% in legumes (Table 2). Beans from Cameroon (Garoua: 475 µg/kg), and Benin (Borgou: 67 µg/kg) contained the highest permethrin content. Permethrin was also quantified in beans from Bamako: 2 µg/kg; Duala: 12 µg/kg; Lagos: 2 µg/kg; Sikasso: 2 µg/kg. Permethrin concentrations in excess of 10 µg/kg were quantified in cassava dry from Bamako (49 µg/kg), palm oil from Lagos (17 µg/kg), sesame seeds from Kano (11 µg/kg) and smoked fish from Garoua (15 µg/kg). Smoked fish from Mali also contained permethrin (Bamako: 6 µg/kg; Sikasso: 3 µg/kg). Vegetables from Duala also enclosed permethrin (leafy vegetables: 6 µg/kg; onion and garlic: LB: 0.3; UB: 1.0 µg/kg), as well as pepper from Lagos (3 µg/kg). Traces of permethrin (LB: 0.3; UB: 1.0 µg/kg) were detected in two beef composites from Bamako and Borgou, in broth from Kano and citrus (oranges) from Bamako.

3.3.7. Seasonal variation

In order to compare the occurrences of the 6 most prevalent pesticides in food groups, which were both collected (1) during the wet season and (2) during the dry season (cereals, tubers, legumes, vegetables and fruits) we completed a *t* test. Both the total number of occurrence ($p < 0.05$) and the number of occurrences below the limit of quantification (LQ = 0.001 mg/kg), which we arbitrarily qualify of "*traces*", enabled identification of a seasonal pattern ($p < 0.01$). Pesticides were more frequently detected in samples collected during the wet season. However, no particular detection or concentration pattern was determined with regard to quantified samples (> LQ).

3.4. Contamination by other pesticides

Acetamiprid was detected in 11 composites, with higher concentrations in leafy vegetables (94–241 µg/kg) from Garoua. Two bean composites (19–23 µg/kg) from Garoua, one composite of leafy vegetables from Sikasso (31 µg/kg) and two composites of tomato (Duala: 14 µg/kg; Lagos: 15 µg/kg) also exceeded 10 µg/kg.

Of 4 imidacloprid occurrences, only 1 composite (maize from Bamako: 35 µg/kg) contained concentration above 10 µg/kg.

Pirimiphos methyl was detected in wheat bread and pasta from Bamako, Duala and Cotonou (3–181 µg/kg).

Piperonyl butoxide, a synergist was quantified in the same bread samples collected in Bamako, Duala (35–45 µg/kg), and in pasta from Cotonou (17 µg/kg). One millet (Bamako composite) also contained LB: 3; UB: 10 µg/kg of piperonyl butoxide.

Interestingly, growth regulator chlormequat was also detected in bread from Bamako, Duala and Lagos (3–12 µg/kg).

Omethoate was quantified in Duala (tomato: 135 µg/kg), Lagos

Table 2
detection rate (> LD), mean and maximum concentration (μg/kg wet weight) of most frequently detected pesticides by food group.

		ORGANOPHOSPHATES											
		Chlorpyrifos				Dichlorvos				Profenofos			
	Samples (n)	> LD (%)	Concentration			> LD (%)	Concentration			> LD (%)	Concentration		
FOOD GROUP			Mean		Max		Mean		Max		Mean		Max
			LB	UB	Matrix		LB	UB	Matrix		LB	UB	Matrix
CEREALS	58	10	1.4	1.6	56.3 Rice	21	1.1	1.3	13.5 Bread	0	0.0	0.3	ND –
TUBERS	**50**	12	0.4	0.6	7.9 Cassava dry	20	2.0	2.2	18.6 Cassava dry	0	0.0	0.3	ND –
LEGUMES	**28**	32	1.1	1.3	10.8 Peanut	32	5.1	5.4	61.0 Beans	11	0.0	0.4	NQ Beans
VEGETABLES	**56**	27	3.6	3.8	91.2 Tomato	7	0.5	0.8	10.0 Tomato	13	10.2	10.6	555.0 Leafy vegetables
FRUITS	**36**	8	0.0	0.3	NQ Citrus	11	0.3	0.6	5.6 Citrus	6	0.1	0.4	3.2 Plantain
NUTS/SEEDS	**4**	75	2.5	2.6	5.2 Sesame	0	0.0	0.3	ND –	0	0.0	0.3	ND –
MEAT	**7**	71	0.4	0.6	1.1 Beef	0	0.0	0.3	ND –	14	0.0	0.3	NQ Beef
EGGS	**4**	75	0.2	0.3	NQ Hen eggs	0	0.0	0.3	ND –	25	0.1	0.3	NQ Hen egg
FISH	**9**	67	2,591	2,591	18,084 Smoked fish	0	0.0	0.3	ND –	22	23.5	23.8	181.5 Smoked fish
DAIRY	**7**	14	0.0	0.3	NQ Milk powder	0	0.0	0.3	ND –	0	0.0	0.3	ND –
OIL/FAT	**11**	45	8.9	9.2	94.7 Peanut oil	0	0.0	0.3	ND –	0	0.0	0.3	ND –
BEVERAGES	**20**	0	0.0	0.3	ND –	5	0.2	0.5	3.0 Beer	0	0.0	0.3	ND –
MISCELLANEOUS	**18**	22	0.2	0.5	2.1 Chili pepper	0	0.0	0.3	ND –	6	0.4	0.6	6.5 Chili pepper
TOTAL (n)	**308**	**66**				**40**				**17**			
(%)		**21.4**				**13.0**				**5.5**			

	PYRETHROIDS											
	Cypermethrin				Lambda cyhalothrin				Permethrin			
	> LD (%)	Concentration			> LD (%)	Concentration			> LD (%)	Concentration		
FOOD GROUP		Mean		Max		Mean		Max		Mean		Max
		LB	UB	Matrix		LB	UB	Matrix		LB	UB	Matrix
CEREALS	7	1.6	1.9	65.1 Pasta	3	0.0	0.3	1.378 Bread	0	0.0	0.3	ND –
TUBERS	8	0.2	0.5	5.0 Cassava dry	0	0.0	0.3	ND –	6	1.2	1.5	49.1 Cassava dry

(*continued on next page*)

Table 2 (*continued*)

	PYRETHROIDS										
	Cypermethrin				Lambda cyhalothrin				Permethrin		
	> LD (%)	Concentration			> LD (%)	Concentration			> LD (%)	Concentration	
FOOD GROUP		Mean		Max		Mean		Max		Mean	Max
		LB	UB	Matrix		LB	UB	Matrix		LB UB	Matrix
LEGUMES	11	0.9	1.1	13.8 Peas	7	0.7	1.0	12.1 Beans	25	20.2 20.5	475.4 Beans
VEGETABLES	34	16.8	17.0	257.8 Tomato	29	1.9	2.2	41.1 Leafy vegetables	4	0.1 0.4	6.4 Leafy vegetables
FRUITS	19	0.7	0.9	8.9 Citrus	3	0.0	0.3	NQ Watermelon	3	0.0 0.3	NQ Citrus
NUTS/SEEDS	25	0.4	0.6	1.6 Sesame	0	0.0	0.3	ND –	25	2.7 2.9	10.7 Sesame
MEAT	57	2.3	2.5	10.0 Beef	0	0.0	0.3	ND –	29	0.1 0.3	NQ Beef
EGGS	25	0.3	0.5	1.4 Hen egg	0	0.0	0.3	ND –	0	0.0 0.3	ND –
FISH	44	30.4	30.7	250.0 Smoked fish	33	3.4	3.6	16.8 Smoked fish	33	2.6 2.8	14.7 Smoked fish
DAIRY	43	0.5	0.7	2.8 Milk powder	0	0.0	0.3	ND –	0	0.0 0.3	ND –
OIL/FAT	0	0.0	0.3	ND –	0	0.0	0.3	ND –	9	1.6 1.8	17.3 Palm oil
BEVERAGES	0	0.0	0.3	ND –	0	0.0	0.3	ND –	0	0.0 0.3	ND –
MISCELLANEOUS	17	0.5	0.8	4.0 Chili pepper	0	0.0	0.3	ND –	11	0.2 0.4	2.5 Chili pepper
TOTAL	**53** **17.2**				**24** **7.8**				**22** **7.1**		

Legend: ND: Non detected; NQ: Non quantified.

(peas: 8 μg/kg; other vegetables: 5 μg/kg). Dimethoate concentrations were also quantified (Duala tomato: 31 μg/kg; Lagos peas: 15 μg/kg; and Lagos other vegetables 46 μg/kg).

4. Discussion

Neither the food preparation process (Huan, Xu, Jiang, Chen, & Luo, 2015; Mahugija, Kayombo, & Peter, 2017), nor the dilution factor due to the pooling of 12 subsamples decreased initial pesticide concentration of raw agricultural commodities to a level that could not be detected in 46% of our food samples. The occurrence of 294 incidences of 39 pesticides scattered in 141/308 samples was possible thanks to the combination of (1) a sensitive analytical method (LD: 0.3; LQ: 1.0 μg/kg) for 10 priority analytes and (2) the collection of 12 different subsamples of equal weight by composite. Low limit of detection also ensured low uncertainty of measurement due to limited contribution of censored data to the conservative upper bound scenario. Had the sensitivity of the method been lower, for example with LD = 5 μg/kg, like in a previous total diet study by Gimou, Charrondiere, Leblanc, and Pouillot (2008), and assuming comparable recoveries, we would have obtained 121 incidences, including 85 incidences of chlorpyrifos, dichlorvos, profenofos, cypermethrin, lambda cyhalothrin and permethrin (70%).

In a recent study, (Lehmann, Turrero, Kolia, Konaté, & de Alencastro, 2017) was looking for 31 residues, with higher analytical limits than in the SSA-TDS, through the collection of raw agricultural commodities in Burkina Faso. This team detected 16 pesticides in vegetables and drinking water and concluded that vegetables they collected were 36% above Codex MRLs.

In this present study, it is impossible to conclude with regard to the conformity of pooled subsamples of food prepared as consumed against MRLs, which apply to commodities. It is however obvious that at least one of the 12 pooled subsamples contained pesticide, up to 12 times the amount quantified in the composite.

In the absence of Codex MRL, a food sample is non-conforming, regardless of the concentration.

In order to illustrate this, although chlorpyrifos is approved by the international standard, Codex Alimentarius (2018), did not publish a chlorpyrifos maximum residue limit (MRL) for commodities such as fish, tomato, peanut or peanut oil, from which these foods were prepared. Therefore, 15 composites all contained at least one subsample, which did not conform to Codex standard applicable to chlorpyrifos in commodities, from which they were prepared.

We would also like to emphasize that, of 39 detected pesticides, 11 (including 3 of the 6 most frequently detected residues in our analytical plan) are not currently approved by the EU (atrazine, carbendazim, cyfluthrin, dichlorvos, omethoate, permethrin, profenofos, triazofos, trichlorfon, tricyclazole). The European Commission retains more stringent regulation with regard to MRLs compared with Codex (European Commission, 2018).

Chlorpyrifos, only detected in fruits in the second French TDS, and in the NZ TDS (FSANZ, 2002; Nougadère et al., 2012), was detected in all food groups in the SSA-TDS, with the exception of beverages. The absence of concentrations > LD in beverages phenomenon may be due to degradation of residues in drinks (Radford, Panuwet, Hunter, Barr, & Ryan, 2018). In a study on market foods in Shaanxi Province, China (Wang, Wang, Zhang, Wang, & Guo, 2013) the detection rate of chlorpyrifos on individual raw agricultural commodities (1.75%) was over 12 fold lower than in SSA-TDS pooled samples (21.4%). We noted that (Wang et al., 2013) only detected chlorpyrifos in 5 Shaanxi Province raw vegetable samples (mean: 35 μg/kg; max: 129 μg/kg).

Strikingly, both the maximum chlorpyrifos concentration of all SSA-TDS composites (18 084 μg/kg) and the highest mean concentration (2591 μg/kg) applied to food group fish, due to the contribution of smoked fish from Mali. The identification of the origin of the presence of chlorpyrifos at such high levels in smoked fish (environmental contamination or catch/post catch bad practice) requires further investigations with regard to handling (capture techniques, processing, storage), and to the environment. Food group "*oil and fat*" contained, to a lesser amount chlorpyrifos concentration (9 μg/kg), mainly due to the contribution of one peanut oil sample from Kano (95 μg/kg).

Dichlorvos was mainly detected in Nigeria (33 of 40 occurrences) and was notably prevalent in legumes with 32% samples with concentration > LD (mean: 5; max: 61 μg/kg). In the second French TDS, dichlorvos was only detected in one fruit sample and was below LQ (10 μg/kg). In Shaanxi Province (Wang et al., 2013), dichlorvos was detected in 2 samples (mean: 1.6 μg/kg; max: 2.3 μg/kg), which is considerably lower than in composite samples from the SSA-TDS. Codex standard did not publish maximum residue limit (MRL) for dichlorvos in legumes or vegetables but was detected 16 times in food composites prepared from those commodities.

Profenofos was not tested in the French and NZ TDS and was not detected in Shaanxi either. Of all profenofos occurrences obtained in this present study, 58% occurred in Mali (10/17), including 4 quantified concentrations in leafy vegetables (16–555 μg/kg) and smoked fish (30–182 μg/kg). By comparison, Lehmann et al. detected profenofos in raw sorrel (median: 619 μg/kg; max: 2999 μg/kg) in Burkina Faso. There is no Codex MRL for profenofos in leafy vegetables, legumes, onion, plantain or fish, and yet profenofos was detected in foods prepared from these commodities with 14 occurrences.

Cypermethrin was detected in 34% of SSA-TDS vegetable samples (with concentrations mean: 17 μg/kg; max: 258 μg/kg). The tomato composite from Duala exceeded Codex MRL of 0.2 mg/kg. By contrast, cypermethrin was not detected > LD, neither in the second French TDS nor in the NZ TDS (FSANZ, 2002) but was detected (mean: 124 μg/kg; max: 631 μg/kg) in raw individual samples of sorrel from Burkina Faso.

Lambda cyhalothrin was detected in 29% of SSA TDS vegetable samples (mean: 2 μg/kg; max: 41 μg/kg) and only 2% > LD in vegetables from the second French TDS (mean: 12; max: 200 μg/kg). The difference in detection rate may be explained by different analytical limits of detection, as well as by different contamination patterns.

Permethrin was not detected in a Chinese survey (Wang et al., 2013), it was only detected (< LQ) in one vegetable sample from the French TDS (Nougadère et al., 2012) and was quantified in several vegetable samples, prepared as consumed from the New Zealand TDS (mean: 10 μg/kg; max: 140 μg/kg). In the SSA-TDS, permethrin was detected in 25% of legume samples (44% of bean composites), up to 475 μg/kg in beans, two of which exceeded the Code MRL for soy bean of 50 μg/kg (as no specific Codex MRL applicable to beans is recorded).

From a more general perspective, our survey of pesticide concentrations in African foods prepared as consumed, included the characterization of seasonal patterns, although the fact that traces of pesticide not in excess of 1 μg/kg are more frequently detected during the wet season is unlikely to matter with regard to consumer safety.

The completion of dietary exposure assessment and comparison of households' actual intakes with acceptable daily intakes (manuscript in preparation) will be useful to include pesticide residues in the list of food safety priority concerns. We expect that the issue of chlorpyrifos found with exceptionally high levels in smoked fish in Mali foreshadows extensive health problems, which need to be tackled soonest.

5. Conclusion

To the best of our knowledge, a multi-centre total diet study is unprecedented in Africa. This systematic approach, covering 13 food groups and up to 470 analytes reveals the presence pesticides residues in foods as consumed, rather than in raw agricultural commodities. The concentrations we obtained therefore reflect a typical diet of 7291 households.

In order to conclude with regard to the occurrence of pesticides in 8 study centres:

Table 3
detection rate (> LD), mean and maximum concentration (μg/kg wet weight) of most frequently detected pesticides by study centre.

			ORGANOPHOSPHATES											
			Chlorpyrifos				Dichlorvos				Profenofos			
		Samples (n)	> LD (%)	Concentration			> LD (%)	Concentration			> LD (%)	Concentration		
Centre				Mean		Max		Mean		Max		Mean		Max
				LB	UB	Matrix		LB	UB	Matrix		LB	UB	Matrix
BENIN	Littoral	33	12	1.3	1.6	56.3 Rice	3	0.0	0.3	1.0 Cassava dry	0	0.0	0.3	0.3 –
	Borgou	28	7	0.0	0.3	1.0 Smoked fish	0	0.0	0.3	0.3 –	4	0.0	0.3	1.0 Beef
CAMER-OON	Duala	50	24	2.5	2.7	91.2 Tomato	8	0.6	0.8	18.3 Peanuts	4	0.0	0.3	1.0 Onion & garlic
	Garoua	28	25	1.1	1.5	33.0 Leafy vegetables	7	0.9	1.2	0.0 Peanuts	4	0.0	0.3	1.0 Tomato
MALI	Bamako	54	17	548.2	548.5	18,084 Smoked fish	0	0.0	0.3	0.3 –	9	14.0	14.3	555.0 Leafy vegetables
	Sikasso	32	16	276.1	276.4	5,236 Smoked fish	0	0.0	0.3	0.3 –	16	2.0	2.3	30.0 Smoked fish
NIGERIA	Lagos	50	36	1.0	1.2	28.7 Other vegetables	56	3.2	3.4	18.6 Cassava fresh	6	0.2	0.5	19.0 Chili pepper
	Kano	33	27	4.8	5.0	94.7 Peanut oil	15	1.7	2.0	61.0 Beans	0	0.0	0.3	0.3 –
TOTAL	(n)	308	66				40				17			
	(%)		21.4				13.0				5.5			

	PYRETHROIDS											
	Cypermethrin				Lambda cyhalothrin				Permethrin			
	> LD (%)	Concentration			> LD (%)	Concentration			> LD (%)	Concentration		
Centre		Mean		Max		Mean		Max		Mean		Max
		LB	UB	Matrix		LB	UB	Matrix		LB	UB	Matrix
BENIN	15	1.6	1.8	65.1 Pasta	3	0.0	0.3	1.1 Maize	0	0.0	0.3	0.3 –
	11	0.3	0.5	5.6 Beans	4	0.2	0.5	8.0 Beans	11	2.3	2.5	67.4 Beans
CAMER-OON	12	11.6	11.8	257.8 Tomato	6	0.7	1.0	28.4 Tomato	8	0.3	0.6	12.3 Beans
	21	4.1	4.4	126.3 Leafy vegetables	18	1.6	1.8	41.1 Leafy vegetables	7	14.0	14.3	474.4 Beans

(*continued on next page*)

Table 3 (*continued*)

Centre	PYRETHROIDS													
	Cypermethrin					Lambda cyhalothrin				Permethrin				
	> LD (%)	Concentration		Max		> LD (%)	Concentration		Max	> LD (%)	Concentration		Max	
		Mean					Mean				Mean			
		LB	UB	Matrix			LB	UB	Matrix		LB	UB	Matrix	
MALI	7	7.8	8.1	250.0	Smoked fish	13	0.4	0.6	8.6	9	1.0	1.3	49.1	Cassava dry
	9	2.1	2.4	34.2	Leafy vegetables	9	0.9	1.2	16.8	9	0.3	0.6	3.3	Smoked fish
NIGERIA	34	1.4	1.6	13.8	Peas	6	0.2	0.4	5.8	6	0.6	0.9	17.3	Palm oil
	27	1.2	1.4	10.0	Beef	3	0.3	0.6	12.1	6	0.5	0.8	10.8	Other nuts/ seeds
TOTAL	**53**					**24**				**22**				
	17.2					**7.8**				**7.1**				

- The preparation of foods as consumed (e.g. washing and cooking) and the pooling of 12 samples (dilution effect) enabled characterization of significant and realistic contamination levels of food.
- The TDS approach applied to this present study enabled capturing of seasonal variation of detection (frequent presence of pesticide traces detected in the rainy season).
- The TDS methodology enabled identification of geographic patterns (dichlorvos frequently detected in Nigeria, high concentrations of chlorpyrifos in smoked fish from Mali).
- 6 most prevalent pesticides generated 75.5% of occurrences, which justifies scrutiny and examination of these analytes as top priority in terms of pesticide risk assessment.

Our next step is the completion of dietary exposure assessment of study populations to 6 most prevalent pesticides, taking into consideration food consumption data, at household level, for the populations of our 8 study centres. Although the exposure will be of great help to provide guidance to risk managers from Benin, Cameroon, Mali and Nigeria in setting the priority agenda, we would like to address our preliminary recommendations to risk managers and their technical and financial partners:

1. Strengthen laboratory capacity to monitor most prevalent pesticides in commodities
2. Implement a monitoring and surveillance plan with regard to concentrations of at least 6 of the most frequently detected pesticides
3. Review practices in the smoked fish value chain in Mali
4. Sensitize smoked fish value chain operators to the health consequences of high exposure to pesticides and to good practices

Additional recommendations will be available, once dietary exposure and risk characterization completed.

6. 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 the World Health Organization.

Declaration of Competing Interest

The authors declare that there is no conflict of interest.

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Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.fochx.2019.100034.

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The third paper is entitled '*Polycyclic aromatic hydrocarbons (PAHs) in foods from the first Regional Total Diet Study in Sub-Saharan Africa: contamination profile and occurrence data'*.

Through this paper, we highlighted:

- The relatively high concentrations of PAHs in smoked fish and edible oils
- Both in the case of smoked fish and edible oils, the contamination profile has the typical signature of a combustion process.
- The presence of PAHs in foods prepared from dried tubers compared to foods prepared from fresh tubers
- The concentration of PAHs in smoked fish varied extensively from one centre to another, including within the same country.
- Core foods such as dehydrated and concentrated milk and sugar contained very low concentrations of PAHs.
- PAH levels exceeded the EU regulations in 100% of smoked fish and 50% of edible oils.

Moreover, an interesting feature of the contamination profile obtained in the SSA-TDS samples was the unusually high concentration of cyclopenta[c,d]pyrene (CPP) in the vicinity of threefold the proportions reported by previous studies. A tentative explanation of the high CPP content is that some of it originates from a reaction of heated pyrene in the presence of sand silica, which was previously simulated *in vitro*. If this hypothesis is correct and translates to actual food processes, ensuring sand-free or optimum hygiene conditions for smoking and heating processes could contribute to lowering the PAH concentrations in foods. Unfortunately, most PAH occurrence studies conducted in Africa before the SSA-TDS did not report the CPP concentration. Including CPP in multi-analyte tests in future research would contribute to a better understanding of this specific contamination profile encountered in Africa.

This paper, however, does not inform the extent to which PAHs represent a threat to human health. The exposure assessment is necessary to be able to draw conclusions concerning the risk that PAHs represent in Benin, Cameroon, Mali, and Nigeria.

3.2.3. Pesticide occurrence data

According to the EU Rapid Alert System for Food and Feed, 99% of alerts from Benin, Cameroon, Mali, and Nigeria (2000-2016) resulted from 10 pesticides, including endosulfan, chlorpyrifos, profenofos, dichlorvos, dimethoate, ethephon, omethoate, trichlorfon, cypermethrin, lambda-cyhalothrin, and permethrin (European Commission, 2016a).

Figure 14: Pesticide bottles sold in a food market of Pitoa, North Cameroon

Pesticide are easily accessible in Africa, and are sold in main food markets, as shown in Figure 14. Small packaging means that farmers can afford pesticides to increase crops production and yield. In Figure 15, children's food is contained in soda bottles and pesticides containers. In the village where I took this picture, running water was not available. Water is scarce in Banikoara, and villagers draw their necessary water from the community well. It is impossible to be certain whether the pesticide container was properly washed before it was converted into a food container.

Figure 15: Pesticide container transformed into a flask used by schoolchildren in Banikoara, North Benin

The purpose of a TDS is not to capture contamination resulting from such practices. The lack of awareness in this situation is, however, puzzling and needs addressing. The fact that some parents do not realize that recycling a pesticide container could expose their children to a chemical hazard also draws attention to the fact that contamination sources may be unexpected.

Omari and colleagues (2018) observed that the perception of food safety concerns such as pesticide residues in Africa was rather lower than that of other food safety issues relating to hygiene. Awareness about the risks is influenced by the level of knowledge and possibly by gender. Although most occurrence studies focus on pesticides in vegetables and fruits, we decided to look for pesticides in every food group. We also tried to capture the seasonal variation in contamination patterns.

The fourth paper is entitled '*Sub-Saharan Africa Total Diet Study in Benin, Cameroon, Mali and Nigeria: pesticide occurrence in foods'*.

These data exclude the organochlorine pesticides, which will be included in another occurrence paper about POPs. Through this paper, we highlighted:

- Of the TDS composite samples, 46% contained at least one pesticide.
- Up to eight pesticides were detected in the same core food composites.
- Organophosphate pesticides and pyrethroids represented 83.0% of detects.
- Six pesticides represented 75.5% of detects (chlorpyrifos, dichlorvos, profenofos, cypermethrin, lambda-cyhalothrin, and permethrin).
- The highest concentration was for chlorpyrifos quantified in smoked fish from the two areas of Mali: Bamako 18 mg/kg and Sikasso 5 mg/kg.
- The methodology allowed for the capture of seasonal variation in contamination patterns, with more frequent detects during the rainy season.

In terms of public health, it is now critical to identify the extent of the addition of pesticides in smoked fish, to sensitize the producers, and to propose viable alternatives to preserve fish. The fact that chlorpyrifos was detected in two different areas, namely Bamako and Sikasso, suggests that smoked fish producers from other locations in Mali, and possibly elsewhere in Africa, might resort to similar practices, probably at the detriment of public health.

The present article does not, however, present the health risks associated with the dietary exposures of the study populations to the detected pesticides. In other words, more effort is needed in terms of risk assessment, including in areas beyond the study centres involved in this study.

Depending on the outcomes of risk assessments, risk managers from Mali need to work on risk management options, possibly including recommendations concerning diet, investigations of current and alternative practices to reduce contamination levels, and the monitoring of contamination levels.

3.2.4. Metals and trace elements (Al, As, Cd, Hg, and Pb)

Because of the sum of evidence concerning the toxicity of aluminium, arsenic, cadmium, mercury, and lead we considered them a priority and selected these trace elements for inclusion in this occurrence data article. Trace elements may be found everywhere, which is why we included all the core foods in our sampling plan. Water is also a source of metals and trace elements, so tap water was also tested.

Aluminium is known for its contribution to renal damage and may cause hydronephrosis and urethral dilatation, as well as contribute to the presence and obstruction of calculi (WHO, 2011). No cognitive impairment has been reported following dietary exposure to aluminium, although reduced grip strength was observed (WHO, 2011). Recently, the unusually high aluminium content in the brain tissues of patients with familial Alzheimer disease was highlighted (Mirza et al., 2017), suggesting that a combination of genetic and environmental factors, including exposure to aluminium, may contribute to disease onset and progression. Elevated aluminium reduces the lifespan of erythrocytes and interferes with haemoglobin synthesis; these factors contribute to the anaemia that develops after prolonged aluminium exposure in patients with compromised kidney function (Willhite et al., 2014).

Inorganic arsenic may cause lung cancer, bladder cancer, and skin lesions (WHO, 2011). Inorganic mercury may be responsible for kidney weight change, proximal tubule damage, and progressive nephropathy (WHO, 2011). Methylmercury, which is mainly present in fish, is associated with neurotoxicity (JECFA, 2007). Unfortunately, the ICP-MS analytical method that we used did not allow for chemical form speciation.

Cadmium also produces renal damage, which is the most sensitive endpoint (WHO, 2011), as well as neurodevelopmental disorders (Rodriguez-Barranco, 2014), endometrial cancer (McElroy et al., 2017), and various other types of cancer (Richter et al., 2017). Lead exposure is associated with neurodevelopment (Liu et al., 2014) and systolic blood pressure disorders (Gambelunghe et al., 2016).

Figure 16: A traditional cooking pot used in Bamako

Some traditional cookware is made from scrap metal (mainly aluminium) and is commonly used to prepare food in Africa. The cooking pot shown in Figure 16 is from Bamako, Mali. The apparent porosity of the surface may enhance the leaching of metals and trace elements from the pot to the food.

In addition to testing the whole diet and tap water, some composites were duplicated to assess the contribution of artisanal cookware made from scrap metal to contamination, as the leaching of lead from aluminium cookware was previously reported by Weidenhamer et al. (2017). In a proper migration study, protocols avoid the intrinsic variability of food matrices and study variation based on fixed acetic acid concentration, standardized temperature, and time. Because it was not the primary purpose of the TDS, we did not reproduce these studies but duplicated some food samples to compare trace element content.

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Occurrence of 30 trace elements in foods from a multi-centre Sub-Saharan Africa Total Diet Study: Focus on Al, As, Cd, Hg, and Pb

Petru Jitaru[a], Luc Ingenbleek[b,c], Nathalie Marchond[a], Clémence Laurent[a], Abimbola Adegboye[d], Sètondji Epiphane Hossou[e], Abdoulaye Zié Koné[f], Awoyinka Dada Oyedele[g], Chabi Sika K.J. Kisito[h], Yara Koreissi Dembélé[h], Sara Eyangoh[b], Philippe Verger[i], Bruno Le Bizec[c], Jean-Charles Leblanc[j,*], Thierry Guérin[a]

[a] Université Paris-Est, Anses, Laboratory for Food Safety, F-94701 Maisons-Alfort, France
[b] Centre Pasteur du Cameroun (CPC), Yaoundé, Cameroon
[c] LABERCA, ONIRIS/INRA, Nantes, France
[d] National Agency for Food and Drug Administration and Control (NAFDAC), Abuja, Nigeria
[e] Agence Béninoise de Sécurité Sanitaire des Aliments (ABSSA), Cotonou, Benin
[f] Agence Nationale pour la Sécurité Sanitaire des Aliments (ANSSA), Bamako, Mali
[g] Laboratoire Central de Sécurité Sanitaire des Aliments (LCSSA), Cotonou, Benin
[h] Laboratoire de Technologie Alimentaire (LTA), Bamako, Mali
[i] World Health Organization (WHO), Geneva, Switzerland
[j] Food and Agriculture Organization of the United Nations (FAO), Rome, Italy

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ABSTRACT

This paper reports occurrence data related to 30 trace elements in food composite samples from a multi-regional Sub-Saharan Africa Total Diet Study. Herein, 2700 samples grouped in 225 food composite samples corresponding to 13 food groups: cereals, tubers, legumes, vegetables, fruits, nuts/seeds, meat, eggs, fish, milk/dairy, oil/fats, and beverages from eight locations in four countries, namely Benin (Littoral/Borgou), Cameroon (Duala/North), Mali (Bamako/Sikasso), and Nigeria (Lagos/Kano) were prepared as consumed, pooled, and analysed using a validated method based on inductively coupled plasma-mass spectrometry. The occurrence data for Al, As, Cd, Hg, and Pb as regulated by the *Codex Alimentarius* are discussed herein. Although the levels of As, Cd, Hg, and Pb were above the limit of quantification, they were below the maximum limits set by the Codex in most samples analysed. A distinct feature was observed for cereals and tubers, as they were mostly contaminated with Al and Pb. A pilot study regarding the impact of using artisanal cookware (made from recycled aluminium) on the contamination of food samples was performed. Relevant contamination with Al and Pb when cooking tomato samples from Cameroon and Nigeria using artisanal aluminium cookware was compared to that when cooked using stainless-steel.

1. Introduction

Food contaminants can originate from the environment or from specific practices and processes as the food is taken from field to fork. Thus, safety assessment of food is challenging given the diversity of available foods and the variety of agricultural, processing, and culinary practices worldwide. Among potential food hazards, inorganic chemical contaminants, such as trace elements, are of particular interest due to the chronic exposure of consumers to and potential long-term health effects of metal exposure (Rehman et al., 2018).

Trace As (inorganic), Pb, Cd, and Hg are routinely monitored in Europe and other countries (Council of the European Union, 2015a,b, 2014, 2011, 2008, 2006), but other inorganic contaminants are not regulated and monitoring data are limited. Recommendations can be generated within the framework of total diet studies (TDS) according to previously published protocols (WHO, 2006, EFSA, 2011a,b; Moy and Vannoort, 2013, Ingenbleek et al., 2017; Papadopoulos et al., 2015; Turrini et al., 2017).

TDSs are endorsed by the World Health Organisation (WHO) and the Food and Agriculture Organization of the United Nations (FAO) has been tasked with assessing the chemical contamination of food prepared as consumed (EFSA, 2011b; FAO/WHO, 2009). This allows for

* Corresponding author.
E-mail address: JeanCharles.Leblanc@fao.org (J.-C. Leblanc).

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the estimation of human dietary exposure after matching contamination data (occurrence) with consumption patterns based on representative samples. TDSs provide scientific information to national authorities to address the risks of food-related chemical hazards for public health protection.

TDSs have been performed in several countries such as the USA (U.S. Food and Drug Administration, 2017; Egan et al., 2007), the UK (UK Report on the Total Diet Study, 2014), Germany (BfR, 2015), Canada (Rawn et al., 2004; Dabeka and Cao, 2013), Italy (Carnovale et al., 2000), Spain (Marin et al., 2017), Australia (Abbey et al., 2013; Food Standards Australia New Zealand, 2014), and Cameroon (Gimou et al., 2013, 2014a, 2014b). In France, two TDSs targeting the general population were performed in 2004 and 2011 (Leblanc et al., 2005; Arnich et al., 2012).

Information concerning the exposure of African population to chemical hazards via food consumption is extremely scarce. To date, published TDS data concerning trace elements in Africa are available only for Cameroon (Gimou et al., 2013, 2014a, 2014b).

The data presented herein were generated in the framework of the Sub-Saharan Africa Total Diet Study (SSA-TDS), which involved two study centres (see Section 2.1) in each of the four countries: Benin, Cameron, Mali, and Nigeria under the leadership of the FAO jointly with the WHO and Centre Pasteur of Cameroon (Ingenbleek et al., 2019a,b,c). Herein, the occurrence data related to 30 inorganic contaminants including Al, As, Cd, Hg, and Pb in 225 composite samples of food are reported. The sampling plan included 194 food composite samples (representing 2338 subsamples prepared using stainless-steel cookware). Additionally, eight tap water composite samples (representing 96 subsamples) were collected and 23 migration study composite samples (representing 276 subsamples prepared using traditional cookware) were obtained within the framework of SSA-TDS. The total chemical content was determined using an accredited method based on inductively coupled plasma-mass spectrometry (ICP-MS) followed by acid digestion (Chevallier et al., 2015). Herein, the occurrence data relating to toxic trace elements (Al, As, Cd, Hg, and Pb) for which JECFA has proposed health-based guidance values or endpoints were examined in detail (WHO, 2011). The occurrence data related to 25 additional inorganic elements are provided in the Supplementary Material (Tables S1 and S2).

Supplementary data associated with this article can be found, in the online version, at https://doi.org/10.1016/j.envint.2019.105197.

2. Material and methods

2.1. Food samples, classification, and food consumption data methodology

The food samples analysed herein were obtained within the framework of SSA-TDS whose methodology and implementation design are described elsewhere (Ingenbleek et al., 2017). Briefly, food consumption data were derived from household budget surveys (HBS) in Benin, Cameroon, Mali, and Nigeria, starting from food expenditure data processed with a unit price database, edible fraction conversion factors, and cooking yield factors obtained from the West Africa Food Composition Table (FAO, 2012).

In October 2017, 2700 sub-samples were collected from eight Sub-Saharan regions as follows: Littoral and Borgou in Benin; Duala and North in Cameroon; Bamako and Sikasso in Mali; and Lagos and Kano in Nigeria. The samples were grouped into 225 food composite samples and subsequently into 13 food groups,: *(i)* cereals, *(ii)* tubers, *(iii)* legumes, *(iv)* vegetables, *(v)* fruits, *(vi)* nuts/seeds, *(vii)* meat, *(viii)* eggs, *(ix)* fish, *(x)* milk/dairy, (xi) (oil/fats), *(xii)* beverages, and *(xiii)* miscellaneous. The SSA-TDS sampling plan was performed based on food consumption data and some less consumed but highly contaminated items may have been omitted for sampling cost effectiveness. Eight tap water samples (not listed here as food) were also included.

The sampling plan was designed to obtain a representative coverage of the most consumed food groups by weight. Hence, the coverage of food groups representing ≥1% of total food consumption was set so as to include a variety of food samples that covered at least 90% of the food groups defined above. However, when food groups represented < 1% of the mean total food consumption, the sampling covered a minimum of 50% of the food groups (Ingenbleek et al., 2017). This approach was used to reduce the number of samples and decrease the cost of the sampling and analysis, while focusing on the most commonly consumed foods representative of the typical diet of the population.

Table 1 summarises the core foods and their proportion of the mean national total diet obtained using the sampling plan in Benin, Cameroon, Mali, and Nigeria.

It is difficult to compare TDS occurrence data due to differences in samples chosen to obtain a given food core, account for natural background presence of trace elements, contamination control, and culinary practices used to prepare the consumed food samples.

Table 1
Coverage of the mean national total diet (TD) by the SSA-TDS sampling plan.[a]

Food core	Benin		Cameroon		Mali		Nigeria	
	% mean TD	No. of samples	% mean TD	No. of samples	% mean TD	No. of samples	% mean TD	No. of samples
Cereals	53.5	7	39.5	6	78.6	9	52.7	7
Tubers	16.6	5	19.5	8	1.7	7	23.8	5
Legumes	4.8	2	6.0	4	2.6	4	7.1	4
Vegetables	5.4	6	6.5	7	2.3	9	3.8	6
Fruits	0.2	1	7.7	3	2.3	7	1.3	7
Nuts/Seeds	0.0	1	0.2	1	0.0	1	0.2	1
Meat	0.2	2	0.4	2	0.6	1	0.5	2
Eggs	0.1	1	0.2	1	1.0	1	0.1	1
Fish	0.5	2	1.1	4	0.4	2	0.6	1
Milk/Dairy	0.5	3	0.3	1	1.1	2	0.5	1
Oil/Fats	1.8	2	1.9	3	1.0	2	1.5	4
Beverages	4.4	5	4.5	6	0.3	2	1.4	7
Miscellaneous	7.0	3	7.8	4	4.9	3	2.8	8
Total	**94.9**	**40**	**95.6**	**50**	**96.8**	**50**	**96.3**	**54**

[a] Ingenbleek et al. (2017).

2.2. Preparation of the food samples

Herein, 225 food sub-samples (approximately 1 kg) were prepared as consumed. A schematic representation of the sampling methodology is provided in Fig. 1 including those prepared for the migration study with relatively inert cookware composed of stainless-steel.

Although this type of cookware is not representative of common cooking practices in the four countries of interest, the use of stainless-steel allowed for identification of the contamination source.

This study was performed to obtain occurrence data of trace elements from contamination of the food itself and not arising from cooking practices. However, a pilot study related to the impact of the traditional cookware made of recycled aluminium on food contamination during cooking is also addressed herein. Thus, five to six of the most consumed foods in each country including tap water, rice, maize, sorghum, millet, and cassava, as well as an acidic matrix (tomato) were split into two identical portions and each portion was cooked under the same conditions using the two types of cookware mentioned above (Fig. 2).

Distilled water was used for cooking to prevent sample contamination from tap water. Similarly, this type of water is not representative of the culinary practices in the African countries, but prevented food contamination from tap water. Tap water composite samples were also analysed and were collected at 12 sites considered to be representative of the cities of interest, as for the other core foods. Identical amounts of each of the samples were subsequently pooled and analysed as for the other composite samples.

Dry foodstuffs (cereals, tubers, and dried legumes/vegetables) were also prepared as consumed by rehydrating the respective matrices with distilled water according to national standard culinary practices (Gautier and Mallet, 2006; Madubike, 2013; Nya-Njike, 1998; Vinakpon-Gbaguidib, 2003). Generally, the food products were ground before preparation, with the exception of rice, in compliance with food consumption habits.

All samples were shipped frozen by air with dry ice from the kitchen laboratory (Benin, Cameroon, Mali and Nigeria) to the analysis laboratory (France). The transportation timeframe did not exceed 24 h. The samples were kept frozen for periods not exceeding three months (−20 °C) prior to analysis.

2.3. ICP-MS analysis

The samples were analysed using an in-house validated and accredited method (French Accreditation Body-COFRAC) based on ICP-MS using acidic microwave digestion as reported elsewhere (Chevallier et al., 2015). Briefly, 0.2–0.4 g of the sample was precisely weighed into a quartz digestion vessel and subsequently pre-digested with 3 mL of ultrapure nitric acid (HNO_3, 67% v/v, VWR chemicals, Prolabo). Then, 3 mL of ultrapure water (18.2 mΩ·cm, Millipore SA, Saint-Quentin-en-Yvelines, France) was added and the sample mineralised using a closed (high-pressure) microwave system (Multiwave 3000 and Multiwave PRO, Anton-Paar, Courtaboeuf, France). The digests were quantitatively transferred to 50 mL (certified volume) polypropylene tubes and filled with ultrapure water. The concentrations of 30 inorganic contaminants were determined by ICP-MS (Agilent 7700, Agilent Technologies, Les Ulis, France). A solution of mixed internal standards (IS), including scandium (Sc), indium (In), bismuth (Bi), rhenium (Re), yttrium (Y), and gold (Au) was added to all blanks, standards, and food

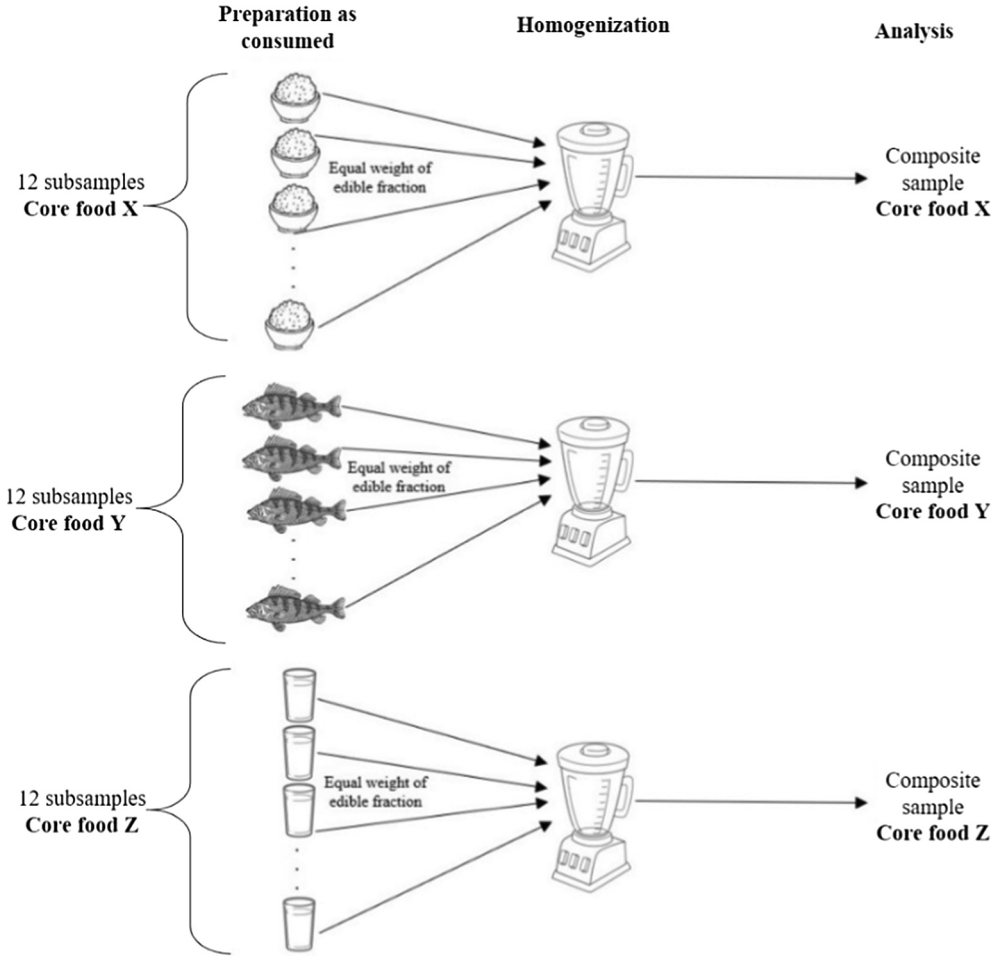

Fig. 1. Schematic representation of the core food composite sample formation from 12 subsamples of equal weight.

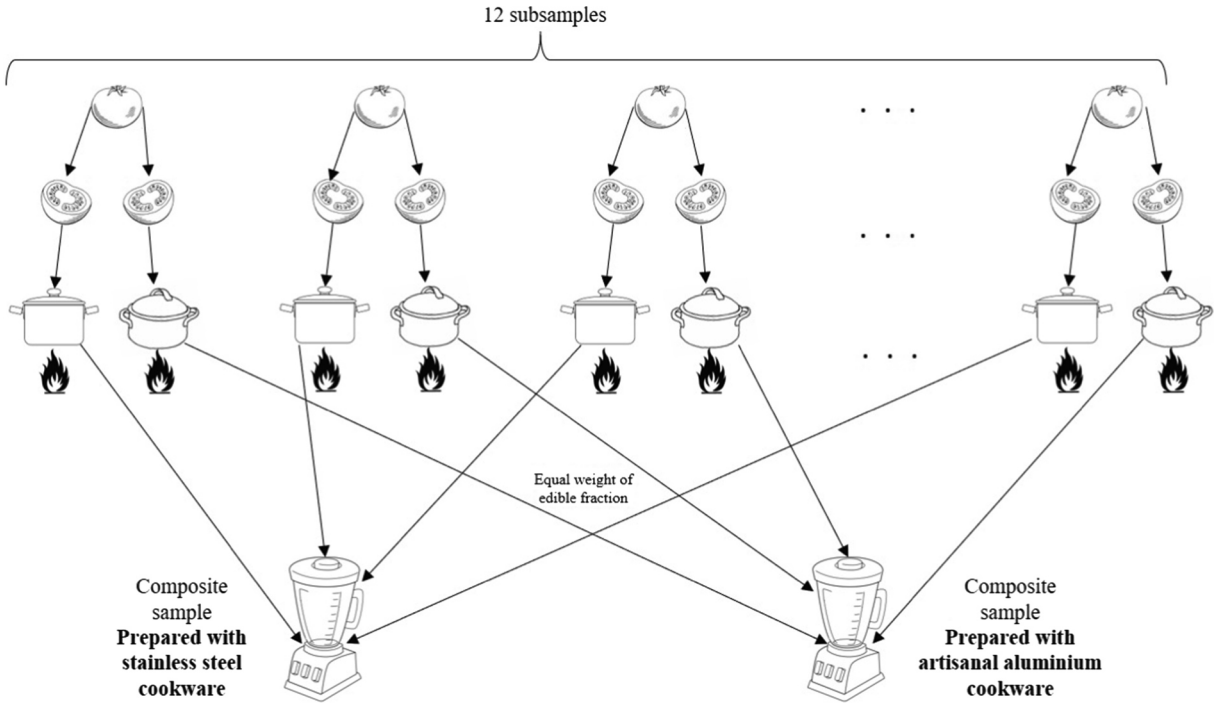

Fig. 2. Schematic representation of the processes of sample duplication prepared using stainless-steel or aluminium cookware.

samples to correct for non-spectral interferences and instrumental drift (each IS was spiked at 2 ng mL^{-1}). The ICP-MS washing solution (6%, v/v) contained gold (Au) at 10 mg L^{-1} to reduce memory effects related to Hg analysis. The limits of quantification (LOQ) and intermediate precision (CV_R, %) for two concentrations ($\leq 2 \times$ LOQ and $> 2 \times$ LOQ) of the accredited method are reported in Table 2.

2.4. Internal quality control

Internal quality controls were used to ensure data reliability and a measurement was considered valid only when all the acceptance criteria were globally satisfied (Chevallier et al., 2015).

Method accuracy and precision were assessed daily via analysis of three certified reference materials (CRM), namely ERM-278K (European reference material, mussel tissue, European Commission), TORT-2 (lobster hepatopancreas) from the National Institute of Standards and Technology (NIST, USA), and SRM 1548 - typical diet (NIST, USA). The analysis of a real sample batch was considered valid when the analyte concentration in each CRM fell within the confidence interval (CI) calculated based on the certified value ($X_{certified}$) (Eq. (1)).

$$CI = X_{certified} \pm \left[k \times \frac{CV_R \times X_{certified}}{100 \times \sqrt{N}} \right] \quad (1)$$

Table 2
Limits of quantification (LOQ in mg/kg fresh weight) and intermediate precision (CV_R, %) for As, Al, Pb, Cd, and Hg.[a]

Analyte	LOQ[b] (mg/kg)	CV_R (%)	
		Sample level $\leq 2 \times$ LOQ	Sample level $> 2 \times$ LOQ
Al	0.033–0.17	20	12
As	0.0008–0.004	20	12
Cd	0.0002–0.001	12	10
Hg	0.003–0.017	15	10
Pb	0.001–0.005	15	8

[a] LOQ and CV_R for the other trace elements measured were provided in Chevallier et al. (2015).
[b] Depending on the amount of analysed sample.

where

CI, confidence interval;
CV_R, intermediate precision;
k = 3 (p = 99%);
N, number of replicates.

For the analytes not present in the CRM or not certified, the method accuracy was assessed by determination of the corresponding recovery factors at two spiking levels depending on the analyte. The method accuracy was considered acceptable when the recovery factors ranged between 80 and 120% for all spiking levels.

A standard solution containing each analyte at a concentration of LOQ + 3 × CV_R was also analysed in parallel with each batch to assess the measurements reliability at concentrations close to the LOQ. This analysis was considered valid if the measured concentration fell within the CI obtained from the method validation using a similar equation as Eq. (1). Most data (> 90%) related to the analysis of CRM and control standard solution (LOQ + 3 × CV_R level) complied with the CI calculated for the inorganic contaminants investigated herein.

2.5. Calculations and statistical methods

The trends were assessed by Student' tests (Microsoft ® Excel® software, 2016) for simplicity due to the relatively low number of samples subjected to statistical analysis.

The data presented herein are the upper bound (UB) concentrations, meaning that the concentration of non-detected analytes was set to LOD for non-detected analytes and to the LOQ for detected but non-quantified analytes. The lower bound concentrations indicated that the concentration of non-detected analytes was zero for non-detected analytes and was assumed to be the LOD for detected but non-quantified analytes, as presented in the Supplementary Data. This indicates that the uncertainty due to censored data was considered, as recommended for TDSs (EFSA, 2011a). All concentrations presented herein are expressed in mg/kg fresh weight.

3. Results and discussion

The occurrence data in terms of Al, As, Cd, Hg, and Pb, which are commonly regulated inorganic contaminants in a large variety of food matrices either by the EC or Codex are reported for the composite samples of the SSA-TDS. In addition to the actual concentrations of these analytes in the various core foods discussed as a function of study centre, the proportions of samples for each of the measured levels of Al, As, Cd, Hg, and Pb exceeding the LOQ are shown in Table 3 (these samples are referred to as quantified samples, which denotes samples with analyte levels exceeding the LOQ).

The concentrations of Al, As, Cd, Hg, and Pb measured in each composite sample of the food groups analysed for the four countries including the eight tap water samples are also reported in Table 4. The low concentrations of Al, As, Cd, Hg, and Pb in the water samples, considering the measurement uncertainty, should conform to the Codex Standard (1981) and are unlikely to contribute significantly to the overall dietary exposure.

The overall mean for a given element as well as the minimum and maximum values were provided for each given analyte in each composite sample.

The concentrations of Al, As, Cd, Hg, and Pb (including the minimum and maximum) for each core food and each study centre are listed in Table 5.

The data obtained for the pilot study assessing contamination with Al, As, Cd, Hg, and Pb when preparing tomato and the other core foods in stainless-steel or traditional artisanal aluminium cookware are provided in Table 6. Contamination is expressed as a concentration factor (CF) that represents the ratio between the sample concentration prepared in aluminium cookware and that measured in the stainless-steel cookware prepared samples.

Supplemental Table S1 lists the mean and minimum/maximum (min-max) concentrations of the 25 inorganic contaminants in various core foods (LB and UB) for the four countries, whereas supplemental Table S2 reports the same type of data in various food groups depending on the study location.

3.1. Total arsenic (As_t)

The highest As_t quantification fractions were observed for the samples collected in Mali, which were mostly found in core foods (except for fruits, milk/dairy, and beverages) and with a 100% proportion in all samples of nuts/seeds, meat, eggs, and fish (Table 3). As_t was quantified in 100% of the meat samples, except in for those obtained from Cameroon, where the quantification fraction was 50%. As_t was quantified in cereals from the four countries, with rates ranging from 57% (Benin) to 83% (Cameroon). The lowest As_t quantification rate was obtained for the fruits, eggs, and milk/dairy food groups.

The highest mean concentration of As_t (Table 4) was measured in fish (0.71 mg/kg) and nuts/seeds (0.030 mg/kg). The most contaminated samples were the smoked and sea fish (≅1.06 mg/kg), whereas, As_t levels in freshwater fish were considerably lower (0.016 mg/kg). For the cereals, As_t levels were considerably higher in rice (0.024 mg/kg) compared to those of the other core foods such as maize, wheat/bread, pasta, sorghum, and millet (< 0.002–0.009 mg/kg; Table 4).

Current European regulations only specify a limit for inorganic As (As_i) of 0.10 mg/kg for rice destined for the production of infants and young children foods to 0.30 mg/kg in rice waffles, rice wafers, rice crackers, and rice cakes (Council of the European Union, 2015a). The mean As_t levels in the four countries showed that the rice was considerably lower (≅8 fold) than the European regulated limits and the Codex maximum (Codex Alimentarius, 1995) for inorganic arsenic in regular rice (0.20 mg/kg).

Concerning the fish samples, the highest As_t level (≅1.0 mg/kg) was observed in sea fish, indicating that sea fishery products are more contaminated with arsenic compared to freshwater fish. However, the dominant As species in marine organisms is arsenobetaine, which is the least toxic of all As species (Hong et al., 2014; EFSA, 2009).

Speciation data to determine the amount of inorganic As, which is the most toxic fraction, are needed (EFSA, 2014) to better characterise the contamination of fishery products with As and assess the impact of fishery product consumption on the general population in the Sub-

Table 3
Fraction (%) of quantified samples of the food groups in each country (Benin, Cameroon, Mali, and Nigeria) participating in the SAA-TD.

Analyte	Country	Cereals	Tubers	Legumes	Vegetables	Fruits	Nuts/Seeds	Meat	Eggs	Fish	Milk/Dairy	Oil	Beverages	Miscellaneous
n[a]	Benin	7	5	2	6	1	1	2	1	2	3	2	5	3
	Cameroon	6	8	4	7	3	1	2	1	4	1	3	6	4
	Mali	9	7	4	9	7	1	1	1	2	2	2	2	3
	Nigeria	7	5	4	6	7	1	2	1	1	1	4	7	8
Al	Benin	100	100	100	100	100	100	100	0	100	100	100	60	100
	Cameroon	100	100	100	100	100	100	100	0	100	100	0	83	100
	Mali	100	100	100	100	100	100	100	100	100	100	100	0	100
	Nigeria	100	100	100	100	100	100	100	100	100	100	75	57	100
As	Benin	57	20	0	33	0	0	100	0	0	33	0	60	67
	Cameroon	83	0	25	29	0	100	50	0	100	0	0	33	50
	Mali	44	29	75	22	0	100	100	100	100	0	50	0	33
	Nigeria	71	0	0	33	14	0	100	0	100	0	50	29	50
Cd	Benin	71	60	100	100	0	100	50	0	100	0	0	40	100
	Cameroon	67	88	100	100	33	0	0	0	75	0	0	17	50
	Mali	o	86	75	89	0	100	100	0	100	0	0	0	0
	Nigeria	71	60	100	100	14	100	100	0	100	0	0	57	75
Hg	Benin	0	20	0	0	100	0	50	0	100	0	0	0	0
	Cameroon	0	0	0	0	0	0	0	0	100	0	0	0	25
	Mali	0	0	0	0	0	0	0	0	100	0	0	0	0
	Nigeria	0	0	0	0	0	0	0	0	100	0	0	0	38
Pb	Benin	57	80	0	83	0	0	100	0	100	67	50	40	67
	Cameroon	67	50	50	100	67	100	100	0	50	0	0	50	50
	Mali	33	57	75	22	0	100	100	100	100	50	0	0	33
	Nigeria	100	100	100	100	86	100	100	100	100	0	50	57	88

[a] Number of samples analysed in each food group.

Table 4
Mean ($\bar{X}$), minimum (min), and maximum (max) upper bound concentrations of Al, As, Cd, Hg, and Pb (mg/kg) in the core foods of the four countries (Benin, Cameroon, Mali and Nigeria).

FOOD			Concentration (C, mg/kg)									
	Composite sample	n	Al		As		Cd		Hg		Pb	
			$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max
CEREALS	Rice	8	0.85	0.28–1.47	0.024	0.009–0.045	0.004	0.001–0.008	0.0040	[a]	0.004	0.001–0.009
	Maize	8	3.36	0.77–8.63	0.002	0.001–0.002	0.0004	0.0003–0.001	0.0040	[a]	0.007	0.001–0.022
	Wheat/bread	3	5.49	1.80–8.60	0.009	0.004–0.017	0.011	0.004–0.017	0.0080	[a]	0.017	0.005–0.036
	Pasta	1	1.26	[a]	0.004	[a]	0.004	[a]	0.0080	[a]	0.003	[a]
	Sorghum	5	25.0	4.42–61.5	0.009	0.001–0.017	0.0006	0.0003–0.001	0.0040	[a]	0.037	0.003–0.095
	Millet	4	20.7	1.84–60.1	0.006	0.001–0.019	0.002	0.001–0.004	0.0040	[a]	0.026	0.001–0.062
	Mean		**9.44**		**0.009**		**0.004**		**0.0053**		**0.016**	
TUBERS	Cassava fresh	4	6.81	0.64–19.0	0.002	0.001–0.002	0.001	0.0005–0.003	0.005	0.004–0.008	0.057	0.033–0.12
	Cassava dry	6	24.7	4.2–93.5	0.011	0.001–0.050	0.002	0.0003–0.004	0.006	0.004–0.008	0.22	0.035–0.53
	Yam fresh	5	1.63	0.21–6.11	0.001	0.001–0.002	0.002	0.001–0.004	0.012	0.004–0.041	0.006	0.001–0.019
	Yam dry	1	12.5	[a]	0.002	[a]	0.001	[a]	0.004	[a]	0.13	0.13
	Potato fresh	2	1.27	1.20–1.30	0.001	[a]	0.012	0.001–0.022	0.004	[a]	0.002	0.001–0.002
	Sweet potato	4	0.78	0.54–1.15	0.001	[a]	0.001	0.0003–0.002	0.004	[a]	0.003	0.003–0.004
	Cocoyam/taro	2	0.96	0.39–1.5	0.002	0.001–0.002	0.003	0.0003–0.005	0.004	[a]	0.004	0.003–0.005
	Macabo	1	0.55	[a]	0.001	[a]	0.001	[a]	0.004	[a]	0.003	[a]
	Mean		**6.16**		**0.003**		**0.003**		**0.0054**		**0.052**	
LEGUMES	Beans	8	2.40	1.7–3.7	0.0020	0.001–0.002	0.001	0.0003–0.002	0.004	0.004	0.009	0.001–0.030
	Peanuts	5	16.7	1.3–39.4	0.0060	0.004–0.10	0.010	0.002–0.014	0.008	0.008	0.019	0.005–0.031
	Peas	1	2.95	2.95	0.0010	[a]	0.001	[a]	0.004	0.004	0.018	[a]
	Mean		**7.34**		**0.0030**		**0.004**		**0.005**		**0.015**	
VEGETABLES	Tomato	8	1.92	0.33–4.63	0.002	0.001–0.005	0.004	0.001–0.008	0.004	[a]	0.007	0.001–0.013
	Green leaves	4	32.3	9.9–77.1	0.006	0.002–0.013	0.007	0.0005–0.020	0.005	0.004–0.010	0.037	0.011–0.10
	Cabbage	1	0.28	[a]	0.001	[a]	0.004	[a]	0.004	[a]	0.001	[a]
	Onion/garlic	8	1.31	0.18–2.74	0.001	0.001–0.003	0.008	0.002–0.018	0.004	[a]	0.010	0.001–0.045
	Okro/gombo	5	1.89	0.91–4.43	0.001	0.001	0.003	0.001–0.010	0.004	0.0040	0.011	0.001–0.021
	Other vegetables	1	24.2	[a]	0.005	[a]	0.008	[a]	0.004	0.0040	0.12	[a]
	vegetables	1	0.24	[a]	0.001	[a]	0.001	[a]	0.004	0.0040	0.001	[a]
	Mean		**8.87**		**0.003**		**0.005**		**0.004**		**0.027**	
FRUITS	Banana	4	0.26	0.19–0.45	0.001	[a]	0.0005	0.0003–0.001	0.004	[a]	0.005	0.001–0.012
	Plantain	3	1.45	0.13–3.92	0.001	[a]	0.0003	[a]	0.004	[a]	0.003	0.001–0.005
	Mango	1	0.22	[a]	0.001	[a]	0.0003	[a]	0.004	[a]	0.003	[a]
	Citrus	5	1.04	0.15–1.97	0.001	0.001–0.003	0.0003	[a]	0.010	0.004–0.035	0.014	0.001–0.047
	Pawpaw	2	0.17	0.12–0.22	0.001	[a]	0.0003	[a]	0.004	0.004	0.004	0.001–0.006
	Watermelon/melon	3	0.87	0.15–2.20	0.001	0.001–0.002	0.0009	0.0003–0.002	0.004	0.004	0.008	0.001–0.022
	Mean		**0.67**		**0.001**		**0.0004**		**0.005**		**0.006**	
NUTS/SEEDS	Palm nut	2	1.47	0.92–2.01	0.006	0.004–0.008	0.0008	0.0005–0.001	0.008	[a]	0.008	0.003–0.014
	Other nuts/seeds	2	333	0.92–662	0.054	0.004–0.103	0.011	0.008–0.014	0.008	[a]	0.18	0.023–0.33
	Mean		**167**		**0.030**		**0.006**		**0.008**		**0.092**	
MEAT	Beef	7	6.73	0.72–21.5	0.004	0.001–0.008	0.010	0.0003–0.64	0.010	0.004–0.045	0.069	0.007–0.26
	Mean		**6.73**		**0.004**		**0.010**		**0.010**		**0.069**	
EGGS	Poultry eggs	4	0.42	0.42–0.98	0.002	0.001–0.002	0.0003	0.0003	0.004	0.004	0.003	0.001–0.008
	Mean		**0.42**		**0.002**		**0.0003**		**0.004**		**0.003**	
FISH	Sea fish	2	0.31	0.23–0.39	1.05	1.00–1.10	0.015	[a]	0.043	0.040–0.046	0.008	0.003–0.013
	Fresh water fish	1	0.52	[a]	0.016	[a]	0.0003	[a]	0.011	0.011	0.003	0.003
	Smoked fish	6	101	0.60–373	1.06	0.02–3.08	0.040	0.002–0.18	0.058	0.034–0.101	0.11	0.007–0.25
	Mean				**0.71**		**0.018**		**0.024**		**0.040**	
MILK / DAIRY	Fresh/fermented milk	3	0.24	0.087–0.35	0.0007	0.0004–0.0008	0.0001	[a]	0.002	[a]	0.002	0.001–0.003
	Dehydrated milk	4	0.48	0.28–0.79	0.005	0.004–0.006	0.0006	0.0005–0.001	0.008	[a]	0.003	0.003–0.005
	Mean		**0.36**		**0.003**		**0.0004**		**0.005**		**0.003**	
OIL / FATS	Palm oil	4	3.24	0.08–6.71	0.005	0.004–0.005	0.0006	0.0005–0.001	0.008	[a]	0.022	0.003–0.053
	Groundnut oil	2	0.26	0.21–0.31	0.002	[a]	0.0005	[a]	0.008	[a]	0.005	0.005
	Other vegetables oil	4	0.38	0.083–1.26	0.005	0.002–0.008	0.0005	[a]	0.008	[a]	0.004	0.003–0.005
	Other fat/oil	1	0.17	[a]	0.004	[a]	0.0005	[a]	0.008	[a]	0.003	0.0030
	Mean		**1.01**		**0.004**		**0.0005**		**0.008**		**0.009**	
BEVERAGES	Water	7	0.022	0.017–0.055	0.0005	0.0002–0.0008	0.0001	[a]	0.002	[a]	0.001	[a]
	Traditional soft drink	3	19.0	0.57–50.2	0.0011	0.0004–0.002	0.002	0.001–0.002	0.002	[a]	0.010	0.002–0.023
	Traditional fermented drink	4	2.08	0.17–4.77	0.001	0.0004–0.002	0.002	0.0001–0.004	0.0020	[a]	0.006	0.001–0.011
	Industrial soft drink	3	0.04	0.017–0.087	0.0005	0.0004–0.0008	0.0001	[a]	0.002	[a]	0.001	[a]
	Industrial fermented drink	3	0.075	0.041–0.097	0.002	0.001–0.003	0.0002	0.0001–0.0003	0.002	[a]	0.003	0.001–0.008
	Mean		**4.24**		**0.001**		**0.0008**		**0.002**		**0.004**	

(*continued on next page*)

Table 4 (*continued*)

FOOD			Concentration (C, mg/kg)									
	Composite sample	n	Al		As		Cd		Hg		Pb	
			$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max
MISC.[b]	Sugar	6	0.38	0.20–0.79	0.002	[a]	0.001	0.0005–0.001	0.008	[a]	0.003	0.003–0.005
	Salt	7	11.3	1.07–38.9	0.017	0.007–0.029	0.001	0.001–0.002	0.048	0.008–0.16	0.054	0.024–0.079
	Broth	2	5.09	4.34–5.84	0.038	0.007–0.069	0.004	0.002–0.005	0.022	0.017–0.028	0.023	[a]
	Chili/pepper	3	6.66	3.59–7.43	0.002	0.001–0.002	0.004	0.002–0.005	0.004	[a]	0.009	0.003–0.013
	Mean		**4.69**		**0.012**		**0.002**		**0.016**		**0.018**	
WATER	Tap water	8	0.089	0.017–0.31	0.0005	0.0004–0.001	0.0001	0.0001–0.0003	0.002	[a]	0.001	0.001–0.002
	Mean		**0.089**		**0.0005**		0.0001	[a]	0.002	[a]	0.001	[a]

[a] Upper bound value (no min/max available).
[b] Miscellaneous.

Saharan region, but this task was beyond the scope of the TDS. Additionally, the ratio of organic and inorganic arsenic in fishery products varied greatly, complicating the assessment of risks related to As_i exposure from consumption of this food type.

Table 5 shows the occurrence data related to As_t in various food groups as a function of study locations. The highest As_t level (3.08 mg/kg) was observed in a smoked fish sample from Borgou, Benin (not shown here). This is twice the level measured in the freshwater fish sampled at the other Benin location. Nevertheless, the origin of As_t could not explain unambiguously attributed to the environment or due to the water loss during drying.

A similar trend was observed for the two Cameroon location, where different levels of As_t in fish were obtained (approximately 43-fold higher in Duala compared to in North Cameroon). Again, the two samples corresponded to different fish species and origins (sea and freshwater).

Apart from fish, levels of As_t exceeding the LOQ were measured in samples from the miscellaneous food group collected in Lagos, Nigeria, with a sample of broth/bouillon cube being the most contaminated (0.069 mg/kg, not shown here) along with rice composite samples from Duala, Cameroon (0.023 mg/kg).

3.2. Lead

From Table 3, Pb levels were above the LOQ in all meat samples in the four countries and in all fish samples collected in Benin, Mali, and Nigeria. Pb was also quantified in all vegetables and nuts from Cameroon, in all nuts and eggs from Mali, and in all cereals, tubers, legumes, vegetable, nuts/seeds, and eggs from Nigeria. The mean Pb levels amongst the core foods ranged from 0.001 mg/kg (LOQ; oil/fats group) to 0.052 mg/kg in tubers (Table 4).

Pb levels in samples from various locations ranged between 0.001 (LOQ) and 0.33 mg/kg for a nuts/seed sample from Sikasso, Mali and 0.23–0.25 mg/kg in fish from Mali (Sikasso) and Benin (Borgou; Table 5). The highest Pb levels observed in fish slightly exceeded the Codex maximum limit (0.30 mg/kg), likely due to the predominant use of leaded gasoline in these countries. This type of fuel may result in environmental Pb contamination via organolead species, which are more bioavailable compared to inorganic lead (Tiwari et al., 2013). Other sources of Pb contaminants in the aquatic environment and ultimately the fish include leaded paint, the materials in contact with the foods (including grinders), and specific food processing practices such as smoking. However, these sources are impossible to discriminate, and it cannot be determined whether environmental or a post capture process contaminated the fish. Speciation analysis of organolead compounds is necessary to discriminate the different origins of Pb, but this exceeds the scope of the study.

Pb concentrations measured in tubers from Mali and vegetables from Cameroon were relatively higher than the levels observed in the other samples. It would be interesting to analyse soil samples to examine soil contamination correlation with Pb concentrations in tubers and vegetables since cassava tubers can readily accumulate metal contaminants from polluted soils (Nworu et al., 2018).

3.3. Cadmium

Cadmium (Cd) was quantified in most food samples with a maximum quantification rate being observed in legumes and vegetable from Benin, Cameroon, and Nigeria; from fish in Benin, Mali, and Nigeria; and from nuts/seeds in Benin, Mali, and Nigeria (Table 3). The mean Cd concentration in various food cores ranged from 0.0003 mg/kg (LOQ) in eggs to 0.018 mg/kg in fish (Table 4). Cd was quantified in meat from Nigeria (Lagos, 0.064 mg/kg), fish from Borgou, Benin (0.18 mg/kg), nuts/seeds from Kano, Nigeria (0.014 mg/kg), and vegetables from Duala, Cameroon (0.011 mg/kg), whereas the lowest mean Cd level was observed for the fruit and milk/dairy food groups (Table 5). Regarding Cd distribution amongst the study centres, the concentrations ranged from 0.0002 mg/kg (LOQ) to 0.18 mg/kg in a fish sample from Benin (Borgou; Table 5).

The highest Cd concentration (0.036 mg/kg) in the fish group was lower than the maximum limit currently set by the EU (0.05 mg/kg; Council of the European Union, 2014). Only one sample exceeded the prescribed maximum Cd level (fish sample from Benin at 0.18 mg/kg), whereas the Codex Alimentarius does not specify a maximum Cd content for fish.

3.4. Mercury

Mercury (Hg) showed the smallest quantification rate in the samples analysed herein ranging from 0.002 mg/kg (LOQ) to 0.16 mg/kg in a salt sample collected in Duala, Cameroon (not shown here as mean values only are shown in Table 5). As expected, Hg was quantified in all seafood samples with means ranging from 0.008 mg/kg (crustacean/molluscs) to 0.10 mg/kg (smoked fish, Bamako, Mali). The maximum Hg concentration was considerably lower than the Codex maximum limits (0.5 and 1.0 mg/kg for non-predatory and predatory fish species, respectively). No Hg was detected in any sample from Mali except in the fish samples. Previous studies highlighted that the most prevalent Hg form in fish is methylmercury (MeHg), the most toxic form. MeHg frequently exceeds 70% of the total Hg in fish (Lescord et al., 2018). Nevertheless, determination of the actual amount of MeHg species requires speciation analysis, which is beyond the scope of this study.

Apart from the fish, wherein Hg was quantified in all samples from the four countries, Hg was more frequently quantified in Benin with a 20% quantification rate in tubers (n = 5), 100% in fruits (n = 1), and 50% in meat (n = 2; Table 3). Hg was quantified in the beef composite sample collected in Benin (Borgou, 0.045 mg/kg), fresh yam composite sample from Borgou (0.041 mg/kg), one tomato composite sample

Table 5
Mean ($\bar{X}$), minimum (min), and maximum (max) upper bound concentrations of Al, As, Cd, Hg, and Pb (mg/kg) in various food groups of the SAA-TDS reported by study location.

FOOD	Country	Centre	Concentration (mg/kg)									
			Al		As		Cd		Hg		Pb	
			$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max
CEREALS	Benin	Littoral	1.50	0.30–2.93	0.008	0.002–0.019	0.003	0.0003–0.004	0.006	0.004–0.008	0.003	0.002–0.003
		Borgou	31.6	0.34–61.5	0.012	0.002–0.019	0.002	0.0003–0.002	0.004	[a]	0.024	0.001–0.052
	Cameroon	Duala	3.3	0.29–6.04	0.017	0.002–0.045	0.006	0.0003–0.011	0.006	0.004–0.008	0.006	0.003–0.011
		Garoua	15.6	1.47–36.6	0.008	0.002–0.014	0.002	0.0005–0.003	0.004	[a]	0.012	0.003–0.027
	Mali	Bamako	6.97	0.89–16.7	0.010	0.001–0.037	0.002	0.0003–0.004	0.005	0.004–0.008	0.004	0.001–0.006
		Sikasso	2.08	0.77–4.42	0.007	0.001–0.025	0.0007	0.0003–0.001	0.004	[a]	0.002	0.001–0.003
	Nigeria	Lagos	3.68	0.83–8.60	0.012	0.001–0.017	0.007	0.004–0.017	0.006	0.004–0.008	0.017	0.004–0.036
		Kano	4.40	1.36–7.61	0.010	0.002–0.029	0.002	0.0003–0.006	0.004	[a]	0.047	0.009–0.095
TUBERS	Benin	Littoral	2.28	0.38–4.17	0.0010	[a]	0.0007	0.0003–0.001	0.004	[a]	0.037	0.003–0.071
		Borgou	8.62	4.02–15.7	0.003	0.001–0.005	0.002	0.0005–0.004	0.018	0.004–0.041	0.056	0.019–0.116
	Cameroon	Duala	2.40	0.39–8.91	0.001	0.001–0.002	0.002	0.001–0.005	0.004	[a]	0.024	0.003–0.123
		Garoua	1.15	[a]	0.001	[a]	0.0003	[a]	0.004	[a]	0.003	[a]
	Mali	Bamako	23.9	0.73–93.5	0.013	0.001–0.05	0.007	0.001–0.022	0.005	0.004–0.008	0.14	0.003–0.533
		Sikasso	6.96	0.65–19.5	0.003	0.001–0.007	0.002	0.0005–0.004	0.006	0.004–0.008	0.18	0.003–0.488
	Nigeria	Lagos	7.94	0.39–19.0	0.001	0.001–0.002	0.0007	0.0003–0.001	0.004	[a]	0.045	0.003–0.125
		Kano	na		na		Na		na		na	
LEGUMES	Benin	Littoral	1.66	[a]	0.001	[a]	0.001	[a]	0.004	[a]	0.003	[a]
		Borgou	1.72	[a]	0.001	[a]	0.001	[a]	0.004	[a]	0.003	[a]
	Cameroon	Duala	1.58	1.27–1.89	0.003	0.002–0.004	0.005	0.001–0.008	0.006	0.004–0.008	0.004	0.003–0.005
		Garoua	11.9	2.42–21.3	0.004	0.001–0.006	0.007	0.001–0.013	0.006	0.004–0.008	0.018	0.005–0.031
	Mali	Bamako	9.89	3.33–16.5	0.004	0.002–0.005	0.007	0.002–0.011	0.006	0.004–0.008	0.006	0.004–0.008
		Sikasso	20.7	2.05–39.4	0.006	0.001–0.010	0.007	0.0003–0.014	0.006	0.004–0.008	0.013	0.001–0.025
	Nigeria	Lagos	2.70	2.39–2.95	0.002	0.001–0.002	0.001	[a]	0.004	[a]	0.022	0.018–0.026
		Kano	4.29	3.66–4.92	0.003	0.002–0.004	0.002	0.001–0.002	0.006	0.004–0.008	0.029	0.028–0.030
VEGETABLES	Benin	Littoral	2.21	0.94–4.22	0.002	0.001–0.004	0.005	0.003–0.008	0.004	[a]	0.004	0.003–0.005
		Borgou	2.80	1.31–4.63	0.002	0.001–0.005	0.006	0.003–0.010	0.004	[a]	0.006	0.005–0.008
	Cameroon	Duala	26.3	0.52–77.1	0.006	0.002–0.013	0.013	0.001–0.02	0.006	0.004–0.008	0.053	0.013–0.102
		Garoua	5.80	0.18–19.8	0.002	0.001–0.002	0.004	0.002–0.006	0.004	[a]	0.015	0.005–0.023
	Mali	Bamako	2.48	0.24–9.87	0.002	0.001–0.005	0.005	0.001–0.015	0.004	[a]	0.004	0.001–0.012
		Sikasso	6.40	0.65–22.2	0.001	0.001–0.002	0.004	0.0005–0.011	0.004	[a]	0.004	0.001–0.011
	Nigeria	Lagos	9.14	0.47–24.2	0.002	0.001–0.005	0.006	0.004–0.008	0.004	[a]	0.049	0.006–0.123
		Kano	2.13	0.53–4.43	0.002	0.001–0.003	0.004	0.001–0.006	0.004	[a]	0.012	0.005–0.020
FRUITS	Benin	Littoral	0.15	[a]	0.001	[a]	0.0003	[a]	0.035	[a]	0.001	[a]
		Borgou	na		na		Na		na		na	
	Cameroon	Duala	2	0.48–3.92	0.001	[a]	0.0005	0.0003–0.001	0.004	[a]	0.005	0.003–0.009
		Garoua	na		na		Na		na		na	
	Mali	Bamako	0.31	0.12–1.03	0.001	[a]	0.0003	[a]	0.004	[a]	0.002	0.001–0.003
		Sikasso	0.26	[a]	0.001	[a]	0.0005	[a]	0.004	[a]	0.001	[a]
	Nigeria	Lagos	0.97	0.15–2.20	0.002	0.001–0.003	0.0006	0.0003–0.002	0.004	[a]	0.018	0.005–0.047
		Kano	0.91	0.24–1.58	0.001	[a]	0.0003	[a]	0.004	[a]	0.005	0.003–0.008
NUTS/SEEDS	Benin	Littoral	0.92	1	0.004	0.004	0.0010	[a]	0.008	[a]	0.003	[a]
		Borgou	na		na		Na		na		na	
	Cameroon	Duala	2.01	[a]	0.008	[a]	0.0005	[a]	0.008	[a]	0.014	[a]
		Garoua	na		na		Na		na		na	
	Mali	Bamako	na		na		Na		na		na	
		Sikasso	662	[a]	0.10	[a]	0.008	[a]			0.33	[a]
	Nigeria	Lagos	na		na		Na		na		na	
		Kano	3.53	[a]	0.04	[a]	0.014	[a]	0.008	[a]	0.023	[a]
MEAT	Benin	Littoral	1.40	[a]	0.002	[a]	0.0010	[a]	0.004	[a]	0.036	[a]
		Borgou	1.06	[a]	0.001	[a]	0.0003	[a]	0.045	[a]	0.019	[a]
	Cameroon	Duala	0.72	[a]	0.002	[a]	0.0005	[a]	0.004	[a]	0.007	[a]
		Garoua	1.30	[a]	0.003	[a]	0.0005	[a]	0.004	[a]	0.011	[a]
	Mali	Bamako	4.00	[a]	0.005	[a]	0.001	[a]	0.004	[a]	0.074	[a]
		Sikasso	na		na		Na				na	
	Nigeria	Lagos	17.2	[a]	0.008	[a]	0.064	[a]	0.004	[a]	0.082	[a]
		Kano	21.5	[a]	0.006	[a]	0.005	[a]	0.004	[a]	0.26	[a]
EGGS	Benin	Littoral	0.083	0.083	0.001	[a]	0.0003	[a]	0.004	[a]	0.001	[a]
		Borgou	na		na		Na		na		na	
	Cameroon	Duala	0.040	[a]	0.002	[a]	0.0003	[a]	0.004	[a]	0.001	[a]
		Garoua	na		na		Na		na		na	
	Mali	Bamako	0.59	[a]	0.0020	[a]	0.0003	[a]	0.0040	[a]	0.0030	[a]
		Sikasso	na		na		Na		na		na	
	Nigeria	Lagos	0.98	[a]	0.002	[a]	0.0003	[a]	0.004	[a]	0.008	[a]
		Kano	na		na		Na		na		na	

(*continued on next page*)

Table 5 (*continued*)

FOOD	Country	Centre	Concentration (mg/kg)									
			Al		As		Cd		Hg		Pb	
			$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max	$\bar{X}$	min–max
FISH	Benin	Littoral	0.60	[a]	1.44	[a]	0.027	[a]	0.101	[a]	0.070	[a]
		Borgou	71	[a]	3.08	[a]	0.18	[a]	0.045	[a]	0.25	[a]
	Cameroon	Duala	1.84	0.23–3.44	1.41	1.11–1.72	0.012	0.009–0.015	0.039	0.038–0.040	0.013	0.003–0.023
		Garoua	7.71	0.52–14.9	0.033	0.016–0.049	0.001	0.0003–0.002	0.023	0.011–0.034	0.013	0.003–0.0023
	Mali	Bamako	144	[a]	0.024	[a]	0.008	[a]	0.072	[a]	0.118	[a]
		Sikasso	373	[a]	0.046	[a]	0.011	[a]	0.058	[a]	0.23	[a]
	Nigeria	Lagos	0.39	[a]	1.00	[a]	0.015	[a]	0.046	[a]	0.013	[a]
		Kano	na		na		Na		na		na	
MILK/DAIRY	Benin	Littoral	0.37	0.27–0.47	0.003	0.0008–0.006	0.0006	[a]	0.005	0.002–0.008	0.002	0.001–0.003
		Borgou	0.09	[a]	0.0004	[a]	0.0001	[a]	0.002	[a]	0.002	[a]
	Cameroon	Duala	0.28	[a]	0.004	[a]	0.0005	[a]	0.008	[a]	0.003	[a]
		Garoua	na		na		Na		na			
	Mali	Bamako	0.36	0.35–0.37	0.002	0.001–0.004	0.0003	0.0001–0.0005	0.005	0.002–0.008	0.0025	[a]
		Sikasso	na		na		Na					
	Nigeria	Lagos	0.79	[a]	0.004	[a]	0.0005	[a]	0.008	[a]	0.0050	[a]
		Kano	na		na		Na		na		na	

Mean ($\bar{X}$) and minimum/maximum (min-max).
na: data not available due to lack of analysed samples.
[a] Upper bound value (no min/max available).

(Littoral, 0.010 mg/kg), and a citrus sample (Littoral 0.035 mg/kg; not shown here).

In Nigeria and Cameroon, Hg was quantified in salt samples (miscellaneous food group) at 0.098 mg/kg in a sample from Kano/Nigeria and 0.166 mg/kg from Duala, Cameroon (not shown here).

3.5. Aluminium (Al)

Al levels were above LOQ in all samples collected from the four countries except in the case of eggs collected from Benin and Cameroon, one edible oil sample collected from Cameroon, and one beverage sample from Mali (Table 3). The most extensive variability of Al concentrations was observed for cereals and tubers (Table 4), with a maximum of 662 mg/kg measured in sesame seeds from Sikasso, Mali (Table 5).

Relatively high Al levels were quantified in fish collected from Mali (144 and 373 mg/kg in Bamako and Sikasso, respectively), in a traditional soft drink (beverages group) from Borgou, Benin (50.2 mg/kg), and two salt samples (miscellaneous group) from Littoral (39.0 mg/kg) and Borgou (11.4 mg/kg). Unfortunately, the Al contamination sources of these food items was not clearly assessed and requires further investigation.

3.6. Impact of artisanal cookware on food contamination during cooking

The relatively large variety of artisanal cooking utensils made in Africa from recycled aluminium and the impact of this traditional cookware on food contamination with trace amounts of the five inorganic contaminants studied herein (As, Al, Cd, Hg, and Pb) during cooking were assessed.

The tomato core food, with a relatively acidic matrix, was considered individually as it was previously shown that acidity increases leaching of trace elements from metallic food contact materials (Weidenhamer et al., 2017, Street et al, 2019). The comparative contamination data of the traditional cookware for each country are shown in Table 6 for tomatoes and other mixed core foods. The ratios between the Al, As, Cd, Hg, and Pb concentration obtained using stainless-steel or artisanal cookware (referred to as contamination factors, CF) are reported. It should be noted that the pots were not identical in the four countries studied herein, which may affect the statistical significance of the obtained data.

Among the five trace toxic elements studied herein, relevant contamination of Al and Pb due to the artisanal cookware was observed for the tomatoes (Table 6).

Pb leaching indicates the presence of this contaminant in the alloy of the artisanal cookware considering that such artisanal cookware are generally manufactured from low quality metal waste. For Al, the maximum impact of artisanal cookware was observed in Cameroon and Nigeria with CF values of 17 and 21, respectively. Similar behaviour was observed for Pb, with the highest contamination observed in Cameroon (CF = 26) and Nigeria (CF = 13). The tomato contamination with Al and Pb was lower in Benin and Mali (CF $<$ 6 for both Al and Pb). These data are consistent with the data reported by Weidenhamer et al. (2017) that showed artisanal cookware composed of recycled aluminium may be a significant source of Al and Pb contamination. Therefore, exposure to toxic trace elements leached from inexpensive, artisanal aluminium cookware produced from recycled metallic waste may pose a public health concern in the developing world, including African countries. Apart from Al and Pb, the concentration factors were relatively low (CF $\cong$ 3) for As and Cd in tomatoes in Cameroon, while for the other core foods, contamination was negligible, with the exception of Pb in Mali (CF = 5).

Statistical tests (p value) were used to compare the artisanal cookware contamination with Al, As, Cd, Hg, and Pb of tomato composite samples and other core foods from all four countries. Significantly higher contamination ($p < 0.05$, not shown here) of tomatoes was observed compared to those of the other core foods only for Al. This confirms that meals prepared with cooked tomatoes may be prone to contamination with toxic trace metals from the traditional cookware due to their acidity, promoting leaching of Al particularly from artisanal cooking utensils composed of recycled Al.

4. Conclusions

Herein, the concentrations of 30 inorganic (elemental) contaminants in foods collected within the first multi-centre regional Sub-Saharan African TDS (Benin, Mali, Cameroon, and Nigeria) are presented. The discussion focuses on the occurrence data of Al, As, Cd, Hg, and Pb.

Heterogeneous levels of these contaminants were observed in different core foods, countries, and between two locations within the same country. In most of the samples, the levels of the four highly regulated

Table 6
Concentrations and concentration factors (CF) of Al, As, Cd, Hg, and Pb (mg/kg) in core foods prepared with stainless-steel and traditional aluminium cookware.

CORE FOOD	Country	n	Concentration (mg/kg)						
			Al			As			Cd
			Stainless steel	Aluminium	CF[a]	Stainless steel	Aluminium	CF[a]	Stainless steel
TOMATO	Benin	1	4.22	8.70	2.1	0.004	0.005	1.3	0.008
	Cameroun	1	1.33	22.8	17	0.002	0.008	4.0	0.001
	Mali	1	0.33	1.14	3.5	0.001	0.001	1.0	0.004
	Nigeria	1	0.47	9.70	21	0.001	0.002	2.0	0.004
	Mean ± SD[b]		1.59 ± 1.81	16 ± 9.0	**6.7**	0.002 ± 0.001	0.004 ± 0.003	**2.0**	0.004 ± 0.003
OTHER FOODS	Benin	5	1.51	1.74	1.1	0.004	0.0045	1.0	0.001
	Cameroun	5	9.19	8.15	0.9	0.012	0.0102	0.8	0.002
	Mali	4	1.79	3.73	2.1	0.010	0.010	1.0	0.001
	Nigeria	5	3.10	4.11	1.3	0.002	0.003	1.1	0.0004
	Mean ± SD[b]		3.90 ± 3.60	4.43 ± 2.69	**1.1**	0.007 ± 0.0048	0.0079 ± 0.004	**1.0**	0.001 ± 0.001

CORE FOOD	Concentration (mg/kg)							
	Cd		Hg			Pb		
	Aluminium	CF[a]	Stainless steel	Aluminium	CF[a]	Stainless steel	Aluminium	CF[a]
TOMATO	0.010	1.2	0.004	0.010	2.4	0.005	0.007	1.4
	0.003	3.0	0.004	0.004	1.0	0.013	0.34	26
	0.004	1.0	0.004	0.004	1.0	0.001	0.008	6.2
	0.004	1.0	0.004	0.004	1.0	0.006	0.078	13
	0.005 ± 0.003	**1.3**	0.004	0.00 ± 0.003	**1.5**	0.006 ± 0.005	0.11 ± 0.16	**18**
OTHER FOODS	0.002	2.2	0.003	0.003	1.0	0.016	0.016	1.0
	0.002	1.1	0.003	0.003	1.0	0.008	0.018	2.2
	0.002	1.5	0.004	0.004	1.0	0.002	0.011	5.2
	0.0005	1.2	0.003	0.003	1.0	0.036	0.025	0.7
	0.002 ± 0.001	**2.0**	0.003 ± 0.0002	0.003 ± 0.0002	**1.0**	0.015 ± 0.015	0.018 ± 0.006	**1.2**

In bold: mean concentration calculated per trace elements for all countries per core food.
[a] Concentration factor (ratio between the level measured in the same sample prepared with aluminium and stainless-steel cookware).
[b] Standard deviation.

inorganic contaminants (As, Cd, Hg, and Pb) were consistently lower compared to the maxima set by the current Codex or European regulations. It should be noted that the staple foods, cereals and tubers, were frequently contaminated with Al and Pb.

The magnitude of the migration of Al and Pb from artisanal aluminium cookware to the prepared food was particularly noticeable during the preparation of tomato samples. This may represent a significant contribution to the dietary exposure towards these toxic elements, which can be reduced by using stainless-steel kitchen utensils.

The next phase in this research project will be to use the occurrence data to characterise dietary exposure of the studied populations.

5. Disclaimer

The views expressed in this information product are those of the authors and do not necessarily reflect the views or policies of FAO and WHO.

Declaration of Competing Interest

The authors declared that there is no conflict of interest.

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The fifth paper is entitled '*Occurrence of 30 trace elements in foods from a multi-centre Sub-Saharan Africa Total Diet Study: focus on Al, As, Cd, Hg and Pb'.*

In this paper, we highlighted:

- Al was found above the limit of detection in all food groups and in 100% of the core foods belonging to the cereals, tubers, legumes, vegetables, fruits, nuts and seeds, meat, fish, dairy, and miscellaneous food groups.
- The highest Al concentration was found in a composite of sesame seeds (662 mg/kg), along with 0.329 mg/kg of Pb.
- Pb concentrations more frequently exceeded the LOQ in Nigeria, than in the other countries, where it was quantified in all the food groups except dairy products.
- The highest mean concentration of Pb was measured in tubers.
- The As_{tot} in rice was 6 to 8-fold lower than the Codex maximum limit applicable to As_{tot} As_i.
- The maximum Cd concentration was measured in fish, but the mean fish concentration did not exceed the EU maximum limit.
- Hg in fish was considerably lower than the Codex maximum limit.
- Cookware type influenced the Pb and Al content of foods when preparing acidic food such as tomato sauce.

With additional lead and aluminium coming from commonly used artisanal pots, compared to the stainless steel utensils used for the study, the TDS results may underestimate dietary exposure to Pb and Al. Risk managers might consider recommending stainless steel and other relatively inert cooking utensils to prepare foods, although it is impossible, from our data to comment on the magnitude of the problem.

This paper, however, does not inform to what extent Al, As, Cd, Hg, and Pb represent a threat to human health, and if they do, in which areas. Once the exposure assessment is completed, we will be able to draw conclusions concerning the risk that these trace elements represent in Benin, Cameroon, Mali, and Nigeria. In particular, it will be interesting to find out if the fact that Pb is more frequently detected in Nigeria is equally reflected in the dietary exposure of the populations of Lagos and Kano, compared to the exposure encountered in other areas.

3.2.5. Persistent Organic Pollutants and other environmental contaminants

POPs, unlike other chemicals, are not defined following their chemical nature, but according to four properties:

1. persistence,
2. bioaccumulation,
3. long-range transport,
4. and toxicity to humans.

The Stockholm Convention came into force in 2004. It presented a list of 12 chemicals from four screening criteria also known as the dirty dozen (WHO, 2008). The 12 substances included organochlorine insecticides such as aldrin, chlordane, DDT, dieldrin, endrin, heptachlor, hexachlorobenzene, mirex, and toxaphene, as well as industrial chemicals such as PCCDs, PCDFs, and PCBs. As of 2017, 16 additional POPs, including PFOS, brominated flame-retardants, and endosulfan, supplemented the initial list and 181 members have ratified the convention, as shown in Figure 17 (UNEP, 2017).

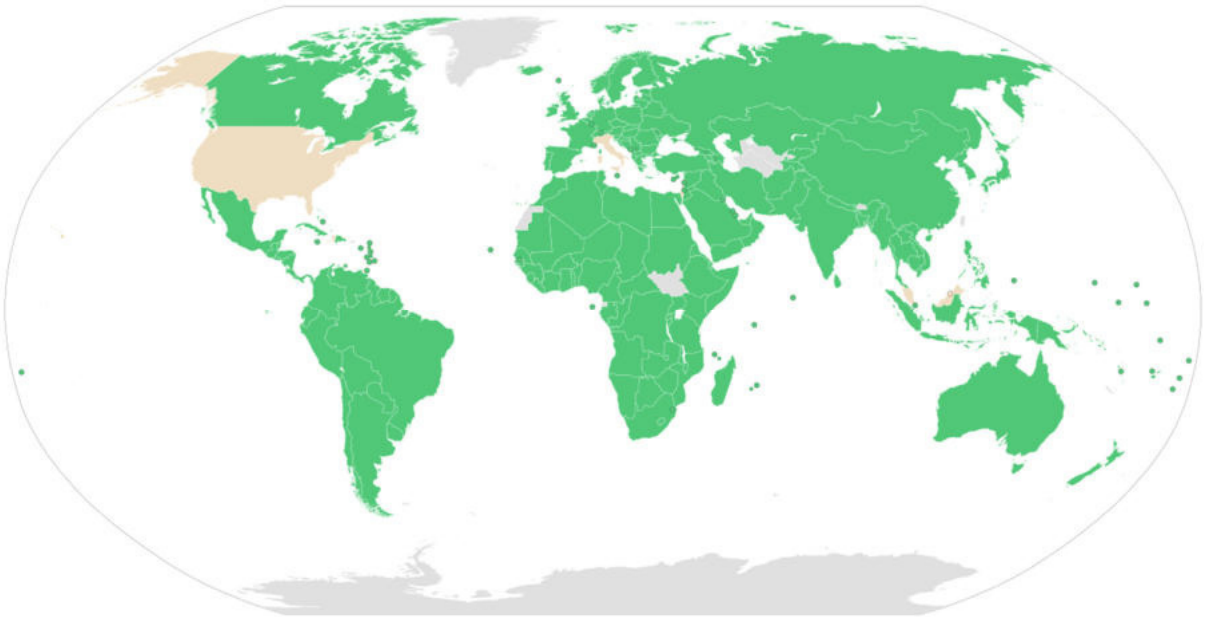

Figure 17: Parties who ratified the Stockholm Convention as of 2017, shown in green

Recently McLachlan (2018) advocated for a more prominent role for POP screening assessments. In other words, the author emphasized some limitations of the Stockholm Convention. For example, PFAAs were not included in the list of POPs, as they do not bioaccumulate with a concentration factor >5 000 in aquatic species, as stipulated in the Stockholm Convention. PFOS, however, was included in the B annex of the Stockholm convention aiming to restrict the use of those chemicals. PFOA is currently a POP candidate, without meeting the Stockholm Convention criteria for bioaccumulation concentration factor. In October 2017, the POPs Review Committee recommended to include PFOA on Annex A (elimination) or B (restriction) (UNEP, 2018).

Long-range transport of POPs cause contamination as far as the Antarctic environment (Mwangi et al., 2016). Fish are commonly studied subjects for monitoring POP contamination. In Tanzania, POP levels in wild and farmed fish were compared (Mwakalapa et al., 2018). The authors did not detect any PFAAS. The most prevalent POP was p,p'-DDE, a DDT metabolite, which was 572-fold higher in wild fish than in farmed fish from the same area of Mtwara.

A recent review by Bruce-Vanderpuije and colleagues (2019) on POP environmental and human exposure in Ghana, highlighted inconsistencies among study methods and analytes, which make comparisons and temporal trends challenging to assess. The authors highlighted the need for more consistent and widespread monitoring programs

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

Vincent Vaccher[a], Luc Ingenbleek[a,b], Abimobola Adegboye[c], Sétondji Epiphane Hossou[d], Abdoulaye Zié Koné[e], Awoyinka Dada Oyedele[c], Chabi Sika K.J. Kisito[f], Yara Koreissi Dembélé[g], Reinwei Hu[h], Inas Adbel Malak[a], Ronan Cariou[a], Anaïs Vénisseau[a], Bruno Veyrand[a], Philippe Marchand[a], Sara Eyangoh[b], Philippe Verger[i], Gaud Dervilly-Pinel[a], Jean-Charles Leblanc[j,*], Bruno Le Bizec[a]

[a] LABERCA, Oniris, INRA, F-44307 Nantes, France
[b] Centre Pasteur du Cameroun (CPC), Yaoundé BP1274, Cameroon
[c] National Agency for Food and Drug Administration and Control (NAFDAC), Abuja 900288, Nigeria
[d] Agence Béninoise de Sécurité Sanitaire des Aliments (ABSSA), BP 362 Cotonou, Benin
[e] Agence Nationale de la Sécurité Sanitaire des Aliments (ANSSA), Bamako BP 2362, Mali
[f] Laboratoire Central de Sécurité Sanitaire des Aliments (LCSSA), Cotonou BP 6874, Benin
[g] Laboratoire de Technologie Alimentaire (LTA), Bamako BP 258, Mali
[h] Inovalys, Official Laboratory of Analysis, Le Mans, France
[i] World Health Organization (WHO), 1211 Amman, Jordan
[j] Food and Agriculture Organization of the United Nations (FAO), 00153 Rome, Italy

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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),

* Corresponding author.
E-mail addresses: vincent.vaccher@oniris-nantes.fr (V. Vaccher), adegboye.a@nafdac.gov.ng (A. Adegboye), oyedele.dada@nafdac.gov.ng (A.D. Oyedele), reinwei.hu@inovalys.fr (R. Hu), ronan.cariou@oniris-nantes.fr (R. Cariou), anais.venisseau@oniris-nantes.fr (A. Vénisseau), bruno.veyrand@oniris-nantes.fr (B. Veyrand), philippe.marchand@oniris-nantes.fr (P. Marchand), eyangoh@pasteur-yaounde.org (S. Eyangoh), vergerp@who.int (P. Verger), gaud.dervilly@oniris-nantes.fr (G. Dervilly-Pinel), jeancharles.leblanc@fao.org (J.-C. Leblanc), bruno.lebizec@oniris-nantes.fr (B. Le Bizec).

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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 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 stricking, 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 °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}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 HBCDDs 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 massic 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 massic 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 (%)*
			WHO-TEQ (2005) PCDD/F		WHO-TEQ (2005) PCB DL	TOTAL-TEQ (2005)		Sum of 6 NDL-PCBs		
			SSA-TDS	EU-MRL**	SSA-TDS	SSA-TDS	EU-MRL**	SSA-TDS	EU-MRL**	
EGGS	4	8.8	0.034	2.5	0.012	0.046	5	64	40.10^3	81
FISH	9	6.9	0.161	3.5	0.117	0.278	6.5	852	75.10^3 and 125.10^3	92
MEAT	7	5.7	0.021	2.5	0.010	0.030	4.0	44	40.10^3	77
MILK/DAIRY	7	11.2	0.032	2.5	0.017	0.048	5.5	83	40.10^3	78
MISCELLANEOUS	2	52.9	0.043	0.75	0.006	0.050	1.25	29	40.10^3	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	40.10^3	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 (Sirot 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 α, β, γ-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 (α, β and γ) 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)																					
		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 (%)	Sum of 7 indicator PBDEs	Sum of 8 indicator PBDEs	Mean of quantification rate (%) *			
EGGS	4	0.01	30	1.13	29	1.21	10	0.47	2	1.17	0	0.38	0	2.53	0	147	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 perflurocarboxylic 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 sulphonamide (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 slighty 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 (μg/kg wet weight) in the composite samples.

	PFBS	PFHxS	PFHpS	PFOS	PFDS	PFBA	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFUnA	PFDoA
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 μg/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	PFDoA
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 pg g^{-1} 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 pg g^{-1} ww, LB) followed by Dec-602 (3.99 pg g^{-1} 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 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 (pg/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

Table 6
Concentrations of perfluoroalkylated substances (expressed in μg/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	PFDoA
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

second-hand electronical 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 K.J. Kisito**: Investigation. **Yara Koreissi Dembélé**: Investigation. **Inas Adbel Malak**: Formal analysis. **Ronan Cariou**: Writing - original draft. **Anaïs Vénisseau**: Formal analysis. **Bruno Veyrand**: Formal analysis, Writing - original draft. **Philippe Marchand**: Methodology, Validation. **Sara Eyangoh**: Supervision. **Philippe Verger**: Conceptualization. **Gaud Dervilly-Pinel**: Writing - review & editing. **Jean-Charles Leblanc**: Project administration, Conceptualization, Methodology, Supervision, Writing - review & editing. **Bruno Le Bizec**: Methodology, Supervision, Validation, Writing - review & editing.

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Disclaimer

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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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.105413.

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The sixth paper is entitled *'Levels of Persistent Organic Pollutants (POPs) in foods from the first Regional Sub-Saharan Africa Total Diet Study'*.

In this paper, we highlighted:

- The levels of POPs are relatively low compared with other TDS occurrence data and regulated levels.
- POPs are more frequently detected in fish than in any other food group.
- In smoked fish, the PCDD/F levels in the SSA-TDS exceeded those in the French TDS, whereas levels in non-smoked fish remained low.
- The PCB levels in the smoked and non-smoked fish in the SSA-TDS were lower than those in the French TDS.
- It can be assumed that the smoking process generates some POPs, which might be connected to the use of burning plastic as a smoke source.

These data also support the hypothesis that the higher concentration of PFOS in smoked fish from Mali may relate to the presence of pesticides (highlighted in the 5th paper) in those foods. If this hypothesis is confirmed, tackling the utilization of pesticides in smoked fish will also reduce PFOS levels.

3.3. Dietary Exposure Assessment and Risk Characterization

Dietary exposure is determined from a combination of food consumption data, presented in the first paper, and occurrence data, presented in the other articles. A box-and-whisker diagram can be used to represent the dispersion of households, within a given study centre. The middle of the box is the median (quartile 2), whereas the bottom and the top of the box represent quartiles 1 and 3, respectively. The whiskers represent 1.5 times the interquartile range, as shown in Figure 18 adapted from Burgard (2013). In a normal distribution, 99.3% of the population is included between the whiskers' extremities.

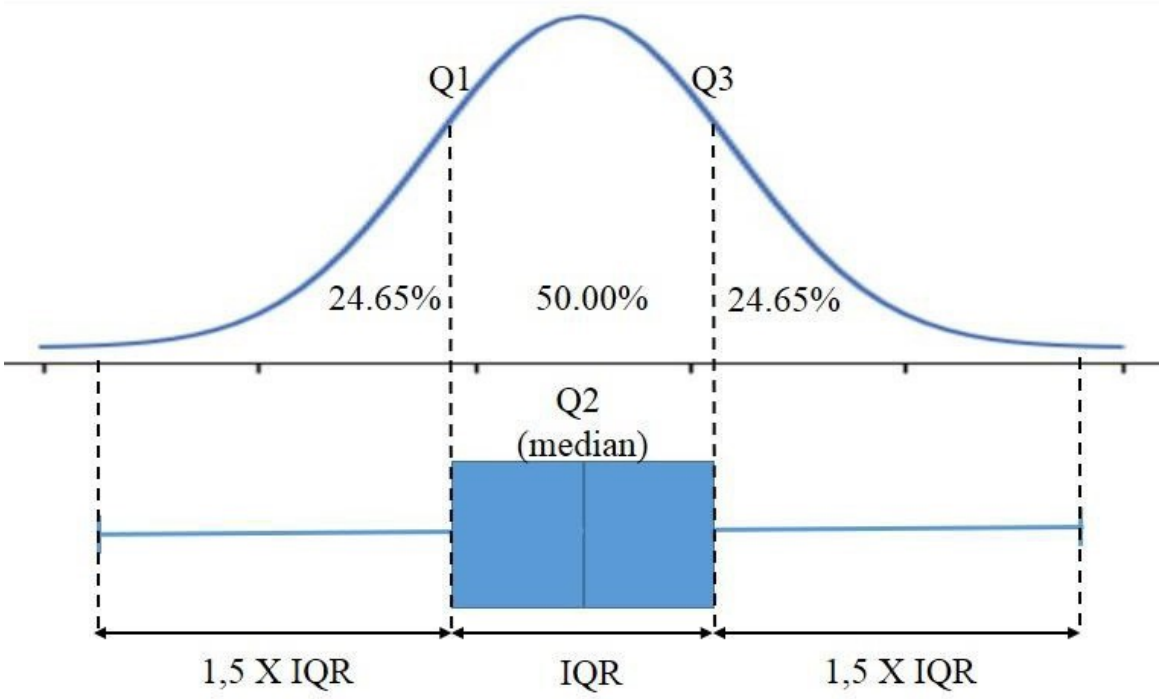


Figure 18: Box-and-whisker representation and probability density of a normal population (Burgard, 2013)

To make better use of the space in the graphs, which include various centres with exposure levels of different magnitude, we excluded the outliers (which represent <1% of the population probability density) from the box-and-whisker diagrams.

3.3.1. Dietary exposure to mycotoxins

3.3.1.1. Aflatoxin B1

The aflatoxin B1 (AFB1) is the most potent aflatoxin. Based on available epidemiological and toxicological data, the JECFA estimated that the morbidity factor could be derived from a combination of dietary exposure and the prevalence of the hepatitis B virus (WHO, 2016). Schweitzer et al. (2015) assessed the prevalence of [$HBAg^{+}$] in Benin (15.6%), Cameroon (12.2%), Mali (13.1%), and Nigeria (9.8%). In this model, the number of additional cases of liver cancer per 100 000 inhabitants per year were determined by multiplying the exposure (in ng/kg body weight/day) by 0.3 for carriers and 0.01 for non-carriers of the hepatitis B virus antigen.

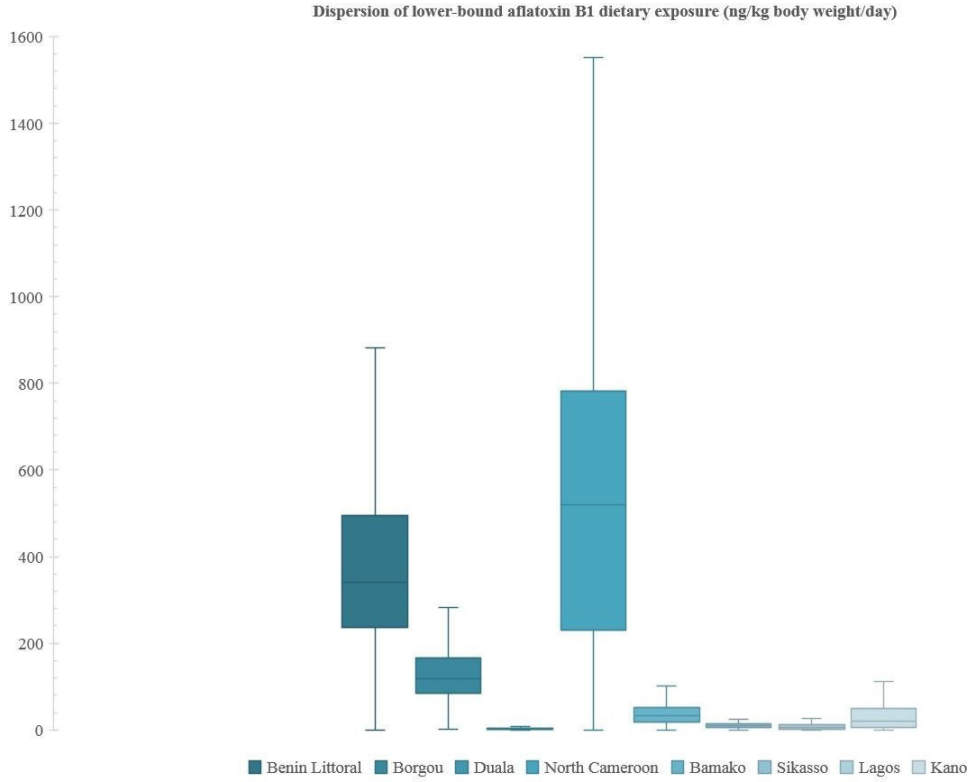


Figure 19: Box-and-whisker plot of ABF1 lower-bound exposures in eight study centres

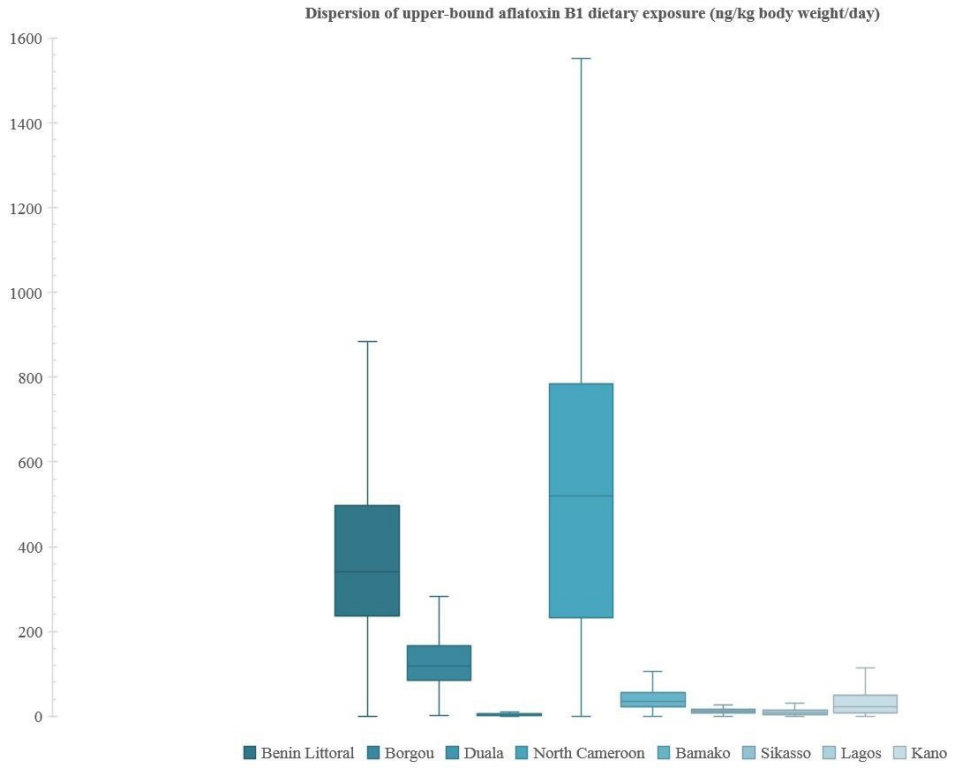


Figure 20: Box-and-whisker plot of ABF1 upper-bound exposures in eight study centres

The difference between the lower- and upper-bound diagrams (Figures 19 and 20) is hardly perceptible thanks to the low analytical limit combined with the relatively high concentrations.

Table 7: Exposures and additional cases of cancer due to AFB1 exposure.

Exposure (ng/kg bw/d)		Mean		P95		Population	Prevalence*	Additional cases of cancer per year**			Contributors		
		LB	UB	LB	UB	(inhabitants)	HBsAg+ (%)	Cases/10E5/year	Study centre	Total (country extrapolation)	Core food	%LB	%UB
BENIN	Littoral	393	394	845	847	678 874	15.6	21.8	148	1 668	Maize	89	88
	Borgou	132	134	258	261	1 202 095		7.4	89		Maize	84	83
											Peanut oil	12	12
CAMEROON	Duala	3	4	9	10	2 510 263	12.2	0.2	5	2 826	Beans	45	34
											Maize	38	30
	North	525	526	1 117	1 117	1 687 959		23.9	404		Peanut	17	13
											Maize	82	82
											Peanut	18	18
MALI	Bamako	38	41	84	89	3 337 122	13.1	2.0	65	228	Peanut	89	83
											Peanut	50	42
											Sorghum	37	31
	Sikasso	10	12	21	23	2 625 919		0.6	16		Maize	40	34
											Rice	38	33
NIGERIA	Lagos	9	10	30	32	12 009 000	9.8	0.4	48	1 729	Maize	75	72
											Peanut	14	13
	Kano	35	37	123	125	10 077 638		1.4	142				

*Schweitzer et al. (2015) The Lancet
** JECFA 2016, 83rd Report

The highest morbidity factor corresponds to North Cameroon (23.9 additional cases per 100 000 per year) and the Littoral area of Benin (21.8 additional cases per 100 000 per year). In both cases, maize is the main contributor to chronic AFB1 dietary exposure (Table 7).

1.1.1.1 Fumonisins

In the case of fumonisins, AFB1 synergists, the JECFA estimated that a group PMTDI of 2 µg/kg body weight/day is appropriate for the completion of chronic dietary exposure assessments (WHO, 2016). In the case of the SSA-TDS, the exposure group considered fumonisin B1, B2, B3, and B4.

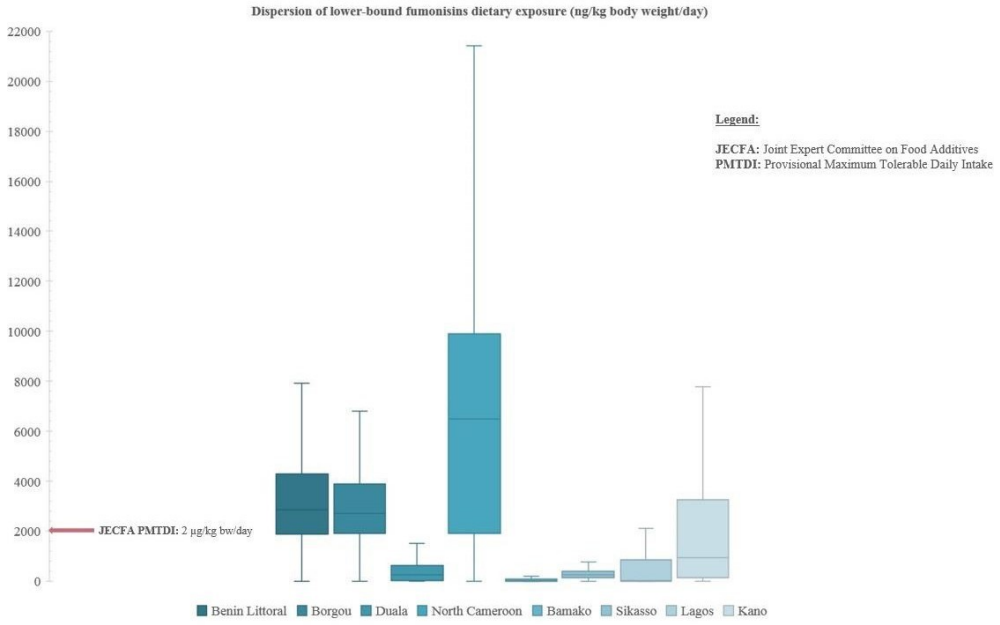


Figure 21: Box-and-whisker plot of fumonisin lower-bound exposure in eight study centres

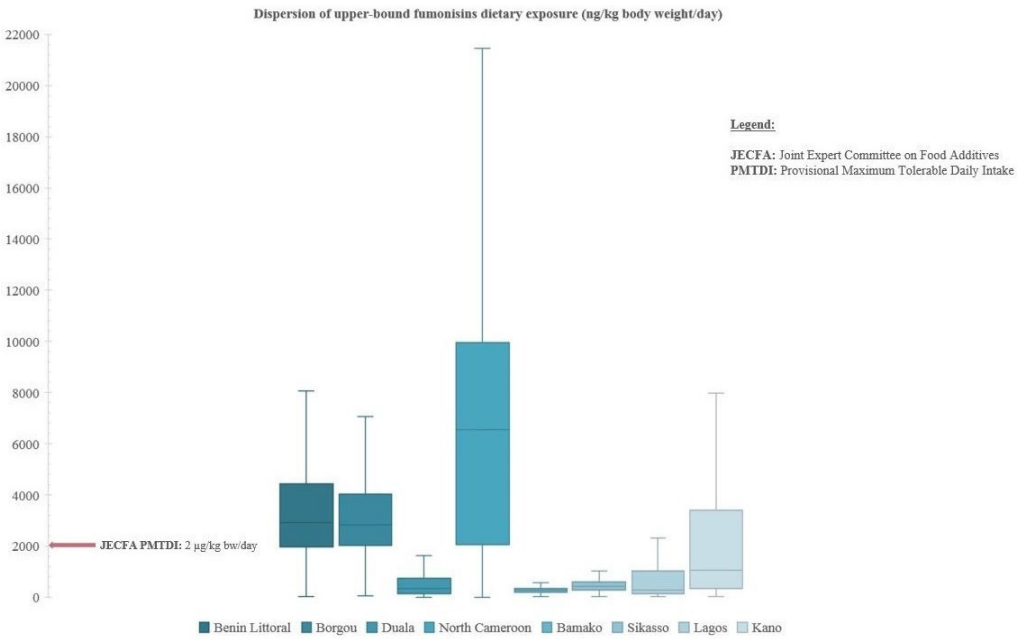


Figure 22: Box-and-whisker plot of fumonisin upper-bound exposure in eight study centres

The difference between the lower- and upper-bound diagrams (Figures 21 and 22) is hardly perceptible thanks to the low limits of detection combined with relatively high fumonisin concentrations.

Table 8: Exposure to the sum of fumonisin B1, B2, B3, and B4 and exceedance of the PMTDI.

Exposure (ng/kg bw/d)		Mean		P95		%> PMTDI (2µg/kg bw/d)		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	3 349	3 450	7 496	7 615	**72**	**74**	Maize	100	97
	Borgou	3 088	3 222	6 104	6 260	**72**	**75**	Maize	96	95
CAMEROON	Duala	472	566	1 646	1 764	3	4	Maize	86	72
								Cassava dry	13	12
MALI	North	6 754	6 826	16 093	16 169	**75**	**75**	Maize	99	98
	Bamako	52	286	221	522	0	0	Maize	100	23
	Sikasso	282	460	639	878	0	0	Maize	92	67
NIGERIA	Lagos	666	855	3 507	3 658	**10**	**12**	Maize	100	77
	Kano	2 188	2 352	8 504	8 656	**38**	**39**	Maize	94	88

* JECFA 2016, 83rd Report

In every study centre, the main exposure contributor was maize (Table 8).

3.3.1.3. Sterigmatocystin

Sterigmatocystin is both an AFB1 precursor and a genotoxic carcinogen. The JECFA proposed a BMDL10 of 0.16 mg/kg body weight/day. Figure 23 and 24 show the lower-bound and upper-bound dietary exposure boxplots, respectively. Table 9 presents the concentrations and the associated margins of exposures. The difference between the two scenarios is more perceptible where the exposure is low, as it is in Duala, for example.

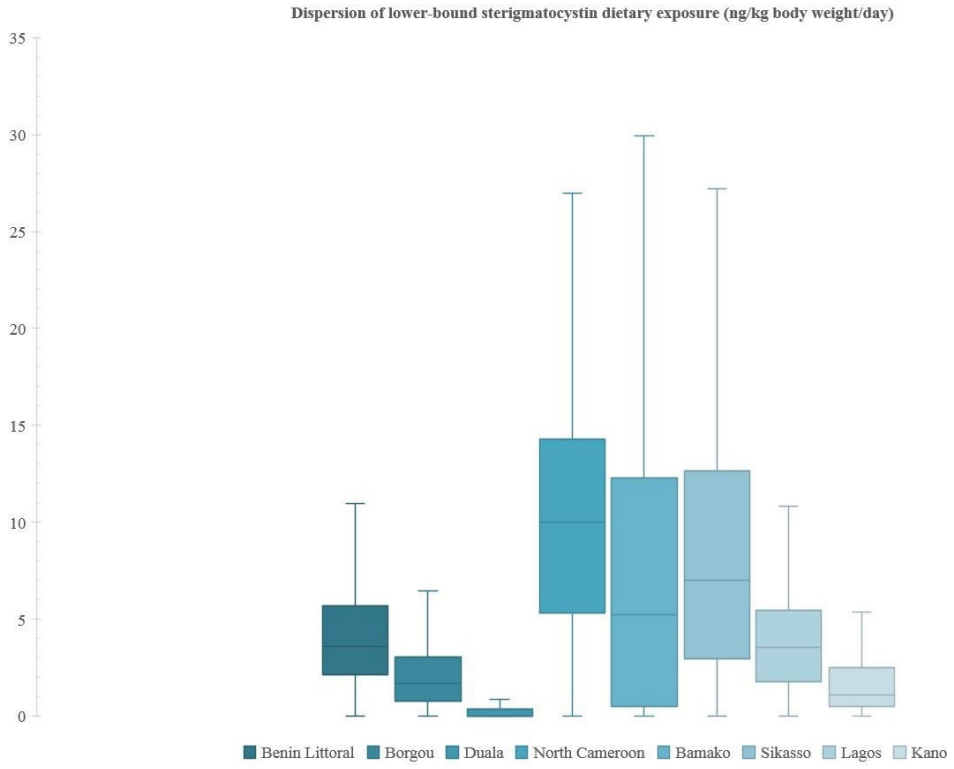


Figure 23: Box-and-whisker plot of sterigmatocystin lower-bound exposure in eight study centres

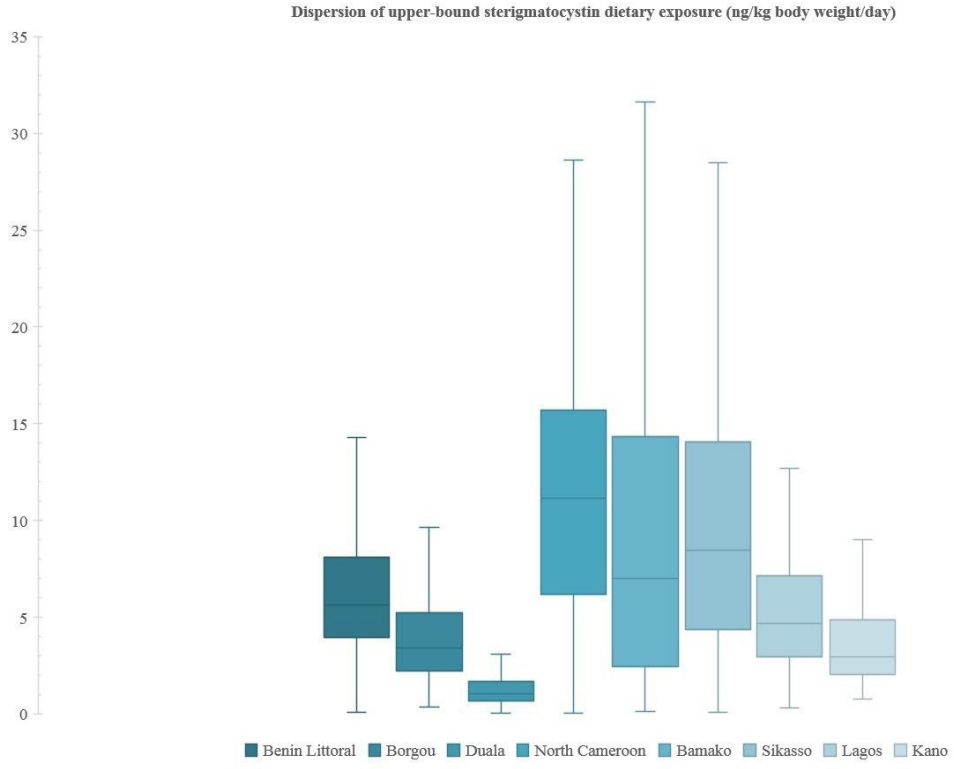


Figure 24: Box-and-whisker plot of sterigmatocystin upper-bound exposure in eight study centres

Table 9: Exposure to sterigmatocystin and margin of exposure (MOE).

Exposure (ng/kg bw/d)		Mean		P95		MOE / BMDL10 (0,16mg/kg bw/d)*				Contributors		
		LB	UB	LB	UB	Mean (LB)	Mean (UB)	P95 (LB)	P95 (UB)	Core food	%LB	%UB
BENIN	Littoral	4	6	10	13	36 566	24 896	15 543	11 899	Peanut oil	82	56
										Maize	8	24
	Borgou	2	4	7	10	68 992	38 556	22 384	16 611	Peanut oil	63	35
										Sorghum	32	21
CAMEROON	Duala	0.4	1	2	3	402 566	119 625	71 392	45 927	Other vegetable oil	98	29
	North	10	11	21	23	15 686	14 175	**7 484**	**6 896**	Maize	73	71
										Other vegetable oil	19	17
MALI	Bamako	8	10	27	30	19 823	16 193	**5 853**	**5 369**	Millet	95	79
	Sikasso	9	10	22	24	18 298	15 773	**7 189**	**6 596**	Sorghum	78	67
										Rice	13	13
NIGERIA	Lagos	4	6	10	12	38 954	28 977	15 471	13 058	Rice	47	28
										Palm oil	37	38
	Kano	2	4	8	10	75 909	40 193	20 649	16 050	Peanut oil	66	35
										Palm oil	26	14

* JECFA 2016, 83rd Report

The Sterigmatocystin 95th percentile margin of exposure was below 10 000 in both the lower-bound and upper-bound scenarios in North Cameroon, Bamako, and Sikasso, where the main contributors were various cereals (maize, millet, sorghum, and rice) and vegetable oils (cotton seed oil and shea butter oil). A safety concern therefore exists in these areas.

3.3.1.4. Ochratoxin A

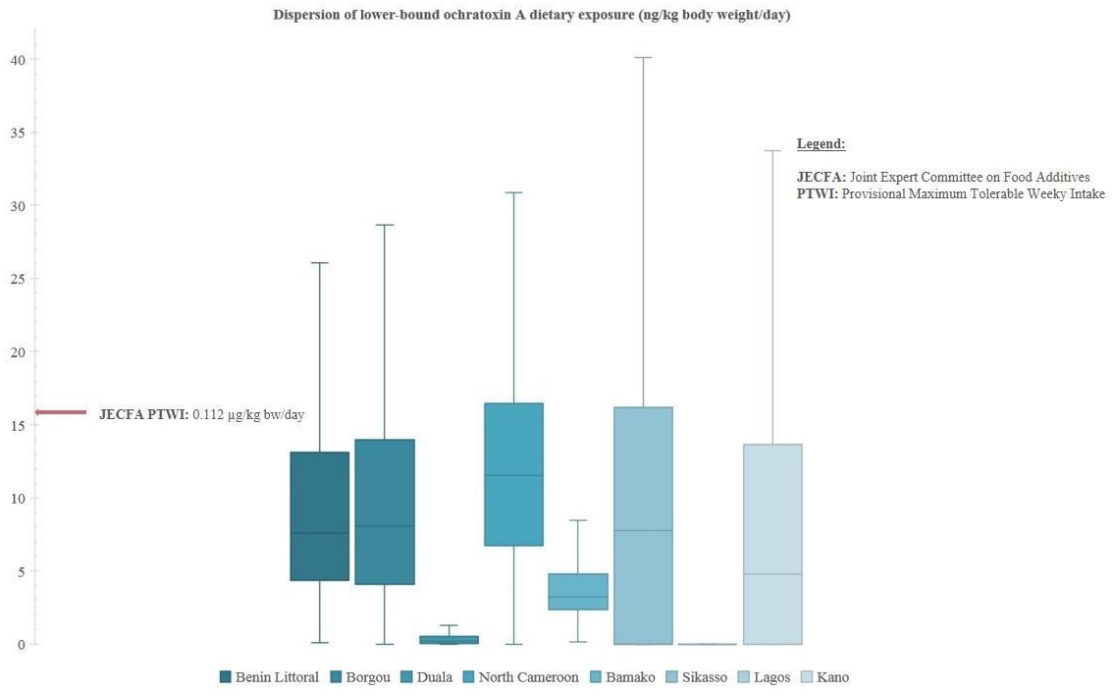


Figure 25: Box-and-whisker plot of ochratoxin A lower-bound exposure in eight study centres

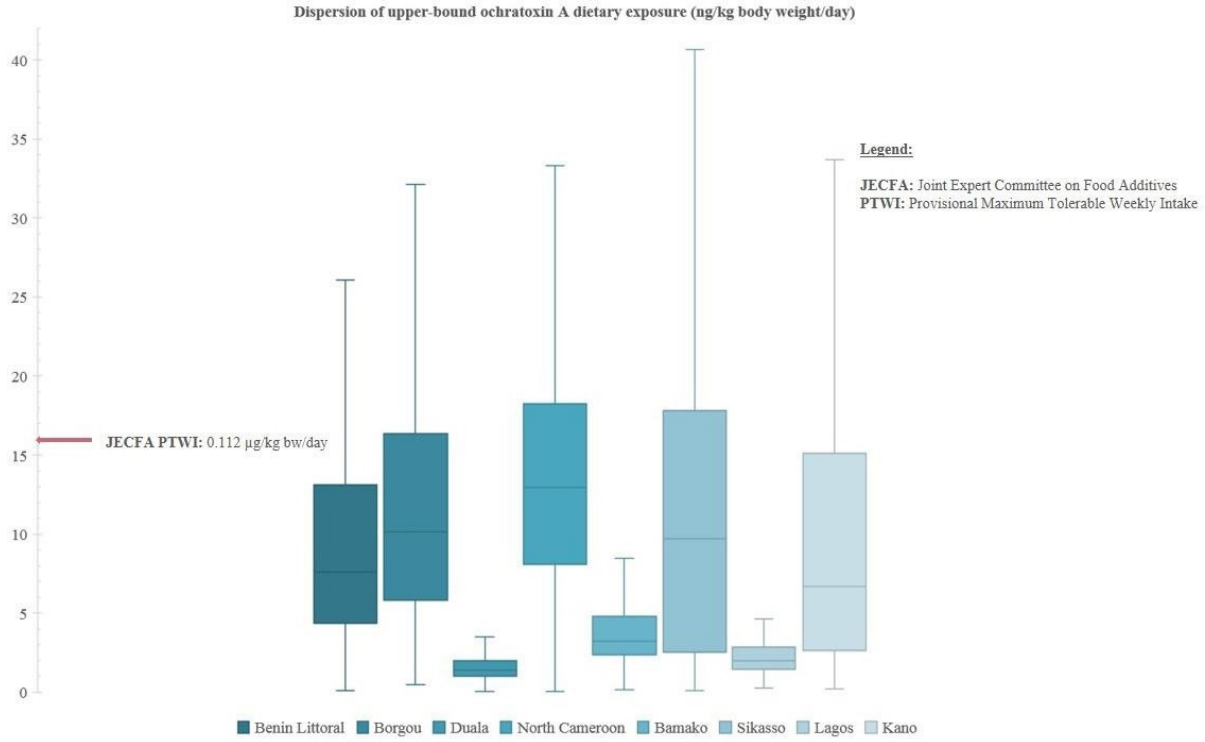


Figure 26: Box-and-whisker plot of ochratoxin A upper-bound exposure in eight study centres

Table 10: Exposure to ochratoxin A and exceedance of the JECFA PTWI.

Exposure (ng/kg bw/d)		Mean		P95		%> PTWI (0,112µg/kg bw/week)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	8	10	24	27	**14**	**17**	Palm oil	65	54
								Peanut oil	13	11
								Cassava dry	12	11
								Rice	44	37
	Borgou	10	13	30	33	**20**	**27**	Sorghum	41	36
								Palm oil	8	6
CAMEROON	Duala	0	2	1	3	0	0	Cassava fresh	90	26
	North	12	13	25	27	**27**	**34**	Maize	72	70
								Rice	24	22
MALI	Bamako	1	4	6	9	0	0	Maize	100	38
	Sikasso	10	12	30	32	**25**	**30**	Sorghum	100	84
NIGERIA	Lagos	0.001	2	0.007	4	0	0	Other nuts/seeds	100	0.1
	Kano	9	11	32	35	**20**	**23**	Rice	100	83

* JECFA 2007, 68th Report

The JECFA PTWI of 0.112 µg/kg body weight/week based on the kidney toxicity of ochratoxin A corresponds to a daily intake of 16 ng/kg body weight (WHO, 2007). In five study centres,

namely the Littoral area of Benin, Borgou, North Cameroon, Sikasso, and Kano, some households exceeded this exposure level. The difference between the lower- and upper-bound diagrams (Figures 25 and 26, Table 10) is only slightly perceptible thanks to low limits of detection combined with relatively high ochratoxin A concentrations. The contributors vary from palm oil in the Littoral area of Benin to rice, maize, or sorghum elsewhere.

3.3.1.5. Citrinin

The JECFA has not yet proposed a health-based guidance value applicable to citrinin.
The EFSA, however, proposed a PMTDI of 0.2 µg/kg body weight/day.

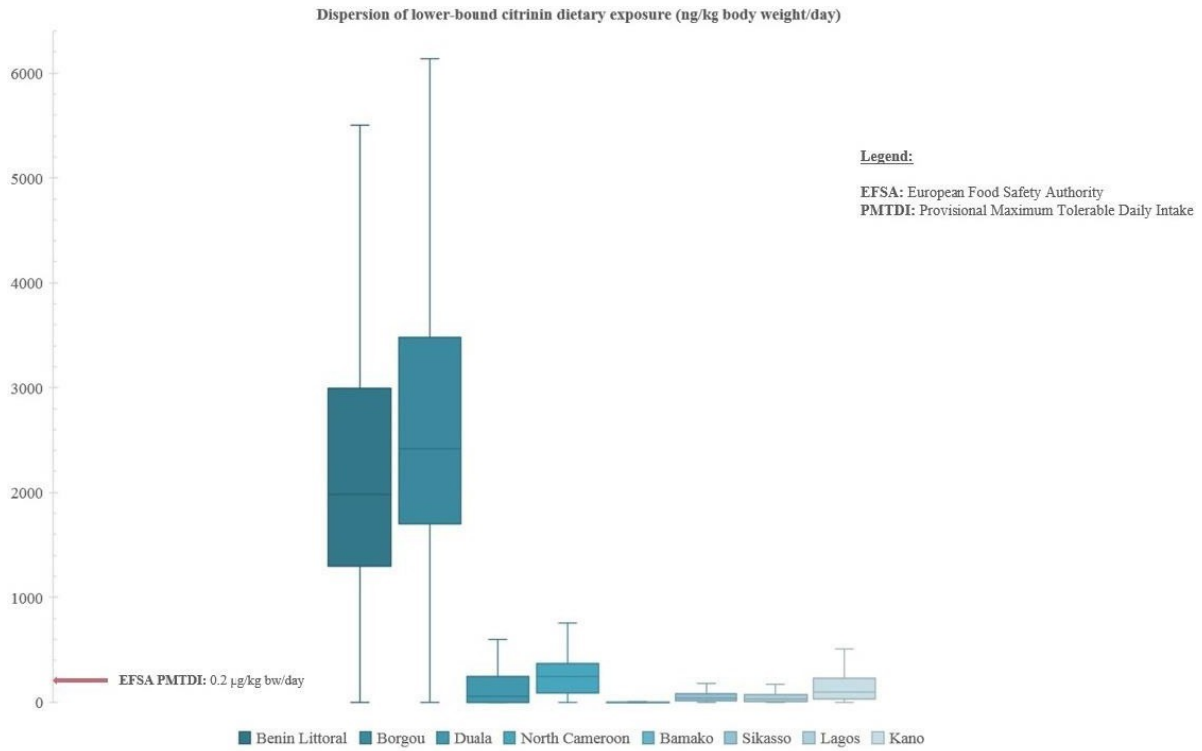


Figure 27: Box-and-whisker plot of citrinin lower-bound exposure in eight study centres

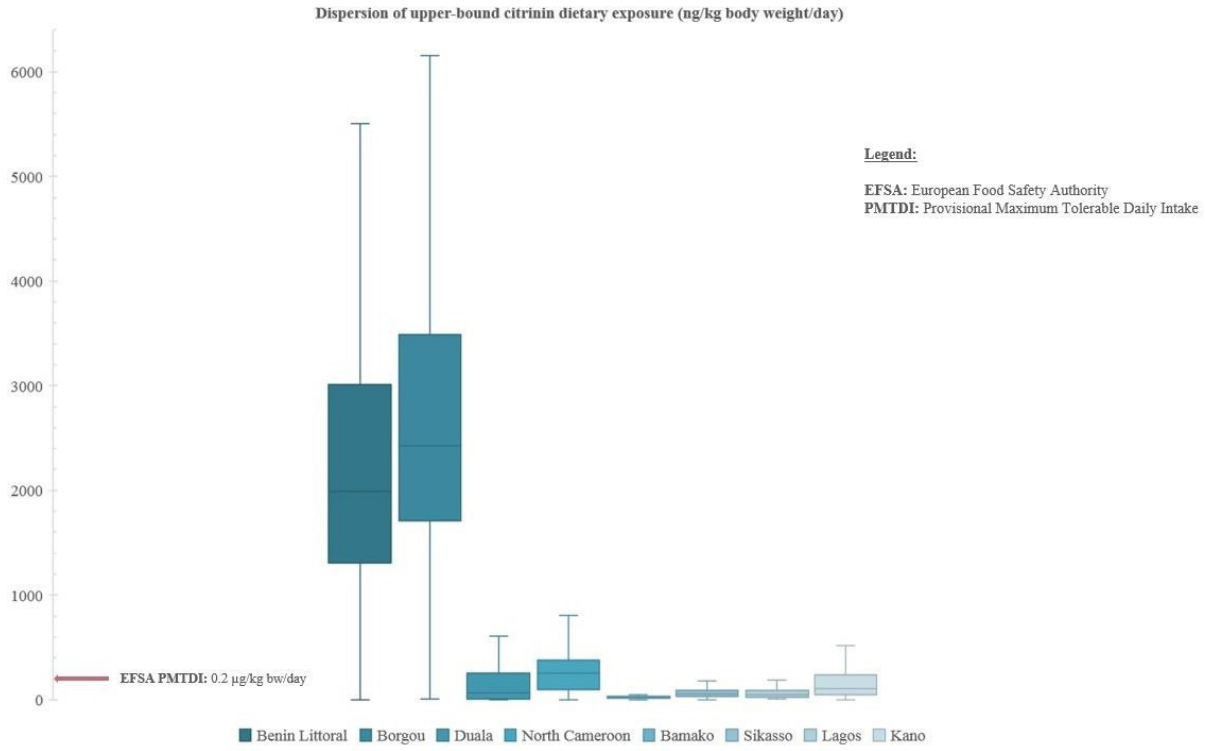


Figure 28: Box-and-whisker plot of citrinin upper-bound exposure in eight study centres

Table 11: Exposure to citrinin and exceedance of EFSA PMTDI.

Exposure (ng/kg bw/d)		Mean		P95		%> PMTDI nephrotoxicity (0,2µg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	**Littoral**	2 332	2 341	5 217	5 228	**98**	**98**	Maize	100	100
	Borgou	2 760	2 767	5 490	5 499	**100**	**100**	Maize	98	98
CAMEROON	**Duala**	171	180	641	655	**30**	**31**	Maize	100	95
								Maize	98	96
	North	252	263	597	615	**60**	**62**	Maize	49	14
MALI	**Bamako**	2	23	5	41	0	0	Sorghum	51	15
								Rice	16	16
	Sikasso	55	67	145	160	0	1	Sorghum	84	68
								Maize	64	53
NIGERIA	**Lagos**	58	71	214	225	**6**	**7**	Rice	29	27
								Maize	78	74
	Kano	158	169	525	544	**30**	**31**	Millet	15	14

*EFSA, 2012

The citrinin exposure in Benin, where maize is clearly the main staple food, was much higher than in the other study centres. The chronic dietary exposure of the populations in Mali was the lowest (100 to 1 000-fold lower than those of the Benin populations). In Mali, the major staple

foods are rice, sorghum, and millet. The difference between the lower- and upper-bound

diagrams (Figures 27 and 28, Table 11) is not perceptible thanks to low limits of detection combined with relatively high concentrations.

3.3.1.6. Zearalenone

Zearalenone chronic dietary exposure in the SSA-TDS, at the 95th percentile, never exceeded the JECFA PMTDI of 0.5 µg/kg body weight/day. It was, however, close, with 4.5-4.6% of the population of Duala exceeding that value (Table 12).

Table 12: Exposure to zearalenone and exceedance of JECFA PMTDI.

Exposure (ng/kg bw/d**)**		**Mean**		**P95**		**%> PMTDI (**0,5µg/kg bw/d**)**		**Contributors**		
		LB	**UB**	**LB**	**UB**	**LB**	**UB**	**Core food**	**%LB**	**%UB**
BENIN	**Littoral**	8	11	18	23	0	0	Maize	100	81
	Borgou	0.4	5	2	11	0	0	Cassava dry	90	9
								Millet	10	3
CAMEROON	**Duala**	144	146	481	484	4.5	4.6	Maize	75	74
								Cassava dry	25	24
	North	10	14	0	11	0.4	0.4	Cassava dry	100	68
MALI	**Bamako**	1	6	2	9	0	0	Wheat/bread	100	12
	Sikasso	0.1	4	0.4	7	0	0	Wheat/bread	100	3
NIGERIA	**Lagos**	11	15	30	36	0	0	Rice	86	68
								Weat/bread	13	10
	Kano	1	5	3	10	0	0	Wheat/bread	95	11

* JECFA 2000, 53rd Report

The main zearalenone contributors were maize and cassava (having undergone a drying process, before rehydration and preparation as consumed).

3.3.1.7. Deoxynivalenol

Deoxynivalenol (also known as vomitoxin) was mainly detected in bread made from imported wheat and, as shown in Table 13, never exceeded the JECFA PMTDI of 1 µg/kg body weight/day.

Table 13: Exposure to deoxynivalenol and exceedance of JECFA PMTDI.

Exposure (ng/kg bw/d)		Mean		P95		%> PMTDI (1µg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	17	104	52	204	0	0	Pasta	77	18
								Yam fresh	14	8
	Borgou	42	136	140	287	0	0	Sorghum	66	25
								Millet	22	8
								Pasta	10	5
CAMEROON	Duala	47	101	108	194	0	0	Wheat/bread	88	41
								Maize	9	17
	North	11	105	58	205	0	0	Wheat/bread	96	10
								Maize		
MALI	Bamako	49	151	134	258	0	0	Wheat/bread	98	32
	Sikasso	9	96	31	157	0	0	Wheat/bread	94	9
								Yam fresh	6	1
NIGERIA	Lagos	106	204	334	526	0	0	Wheat/bread	89	47
								Yam fresh	10	17
	Kano	45	146	171	314	0	0	Wheat/bread	80	24
								Maize	18	24

* JECFA 2011, 72nd Report

3.3.1.8. Nivalenol

Nivalenol dietary exposure was low in all study centres, as shown in Table 14. The difference between the lower-bound and upper-bound exposure was large where the exposure was particularly low, so in all study centres except in Duala. The contribution of maize and dry cassava in Duala, where the nivalenol exposure was highest, resulted in a 95th percentile exposure of 26-27% of the JECFA TDI of 1.2 µg/kg body weight/day.

Table 14: Exposure to nivalenol and exceedance of JECFA PMTDI.

Exposure (ng/kg bw/d)		Mean		P95		%> TDI (1,2µg/kg bw/d)		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	1	18	8	34	0	0	Concentrated/dehydrated milk	92	6
								Sorghum	7	1
	Borgou	27	51	85	125	0	0	Sorghum	80	44
								Maize	19	42
CAMEROON	Duala	93	101	317	327	0	0	Cassava dry	23	21
								Maize	77	71
	North	12	39	19	77	0	0	Cassava dry	49	15
								Maize	51	68
MALI	Bamako	0	19	0	31	0	0	-	-	-
	Sikasso	0	14	0	24	0	0	-	-	-
NIGERIA	Lagos	0	18	0	34	0	0	-	-	-
	Kano	0	18	0	35	0	0	-	-	-

* JECFA 2011, 72nd Report

3.3.2. Dietary exposure to polycyclic aromatic hydrocarbons

The JECFA recommended a surrogate approach, where the MOE is the BMDL of benzo[a]pyrene (BMDL10 of 100 µg BaP/kg body weight/day) divided by exposure to the sum of 13 genotoxic carcinogenic polycyclic aromatic hydrocarbons (PAH13) (WHO, 2006).

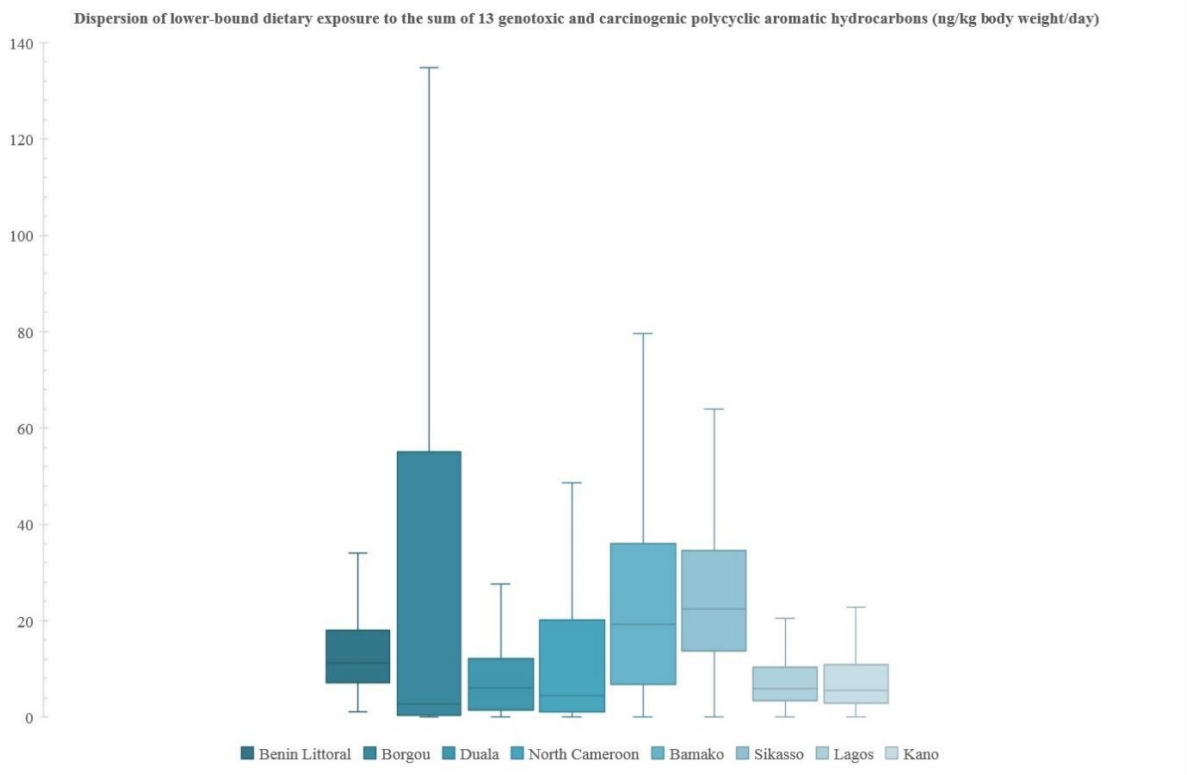


Figure 29: Box-and-whisker plot of the sum of PAH13 lower-bound exposure in eight study centres

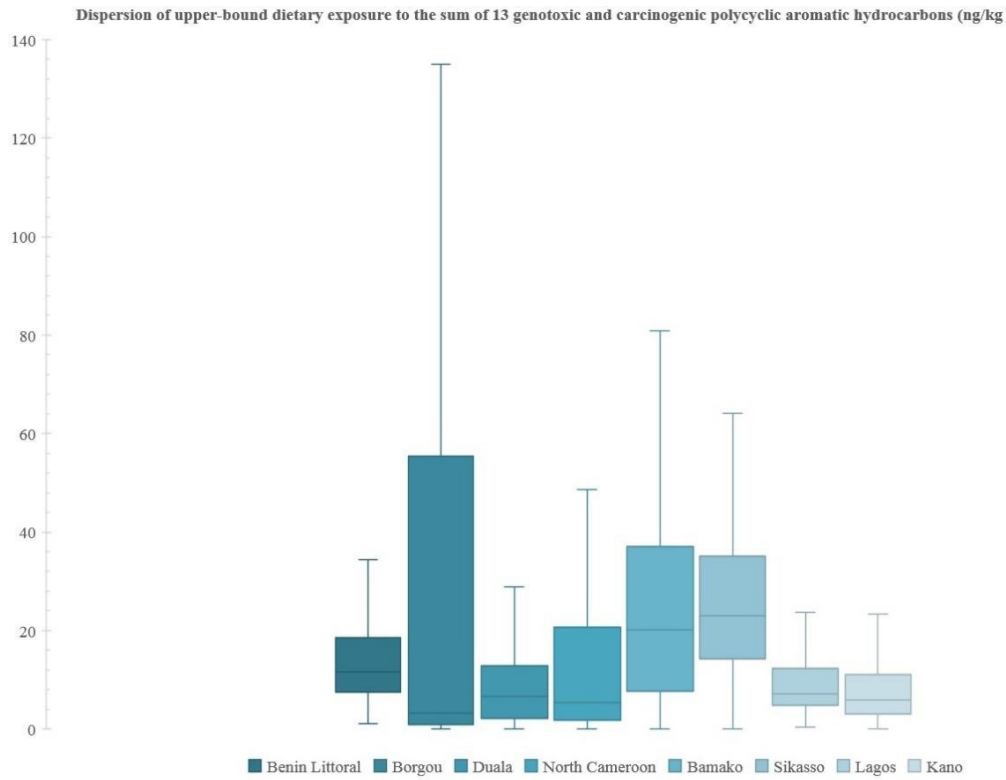


Figure 30: Box-and-whisker plot of the sum of PAH13 upper-bound exposure in eight study centres

Table 15: Exposure to the sum of PAH13 and margin of exposure compared to the BaP BMDL10.

Exposure (ng/kg bw/d)		Mean		P95		MOE (PAH13) / BMDL10 (100µg/kg bw/d) BaP*				Contributors		
		LB	UB	LB	UB	Mean (LB)	Mean (UB)	P95 (LB)	P95 (UB)	Core food	%LB	%UB
BENIN	Littoral	14	14	33	34	**7 238**	**6 995**	**2 998**	**2 924**	Smoked fish Palm oil Peanut oil Cassava dry	44 32 12 10	43 31 12 11
	Borgou	40	40	169	169	**2 531**	**2 504**	**591**	**590**	Smoked fish Peanut oil Palm oil	96 2 2	95 2 2
CAMEROON	Duala	8	9	26	27	11 791	11 046	**3 881**	**3 735**	Smoked fish Palm oil Casava dry	83 8 5	78 8 6
	North	13	14	48	50	**7 475**	**7 060**	**2 063**	**2 001**	Smoked fish Peanuts	91 6	86 9
MALI	Bamako	25	26	69	70	**4 007**	**3 870**	**1 444**	**1 425**	Smoked fish Other vegetable oil	71 25	69 25
	Sikasso	25	26	55	56	**3 938**	**3 863**	**1 808**	**1 796**	Smoked fish Other vegetable oil	67 32	66 31
NIGERIA	Lagos	8	10	21	24	12 286	10 454	**4 691**	**4 226**	Palm oil Chili Peanut oil	54 24 10	47 21 8
	Kano	9	9	27	27	11 467	11 145	**3 714**	**3 657**	Palm oil Peanut oil Chili	67 24 8	65 23 8

* JECFA 2006, 64th Report

The margin of exposure is lower than 10 000 for highly exposed households (95th percentile) in the eight study centres. It was as low as 590-591 in Borgou, where the concentration of smoked fish exceeded that of any other centre or sample of the study (Figures 29 and 30, Table 15). In Benin, Mali, and North Cameroon, even the mean chronic dietary exposures corresponded to an MOE below 10 000.

3.3.3. Dietary exposure to pesticides

As presented in the pesticide occurrence paper submitted to *Food Chemistry* (December 2018), six pesticides covered 75% of all pesticide occurrences in the SSA-TDS samples, three organophosphates (chlorpyrifos, dichlorvos, and profenofos) and three pyrethroids (cypermethrin, lambda-cyhalothrin, and permethrin).

3.3.3.1. Chlorpyrifos

Chlorpyrifos was detected in 21% of the SSA-TDS samples. Exposure is the only way to assess to what extent the frequency of occurrence matters from a public health perspective.

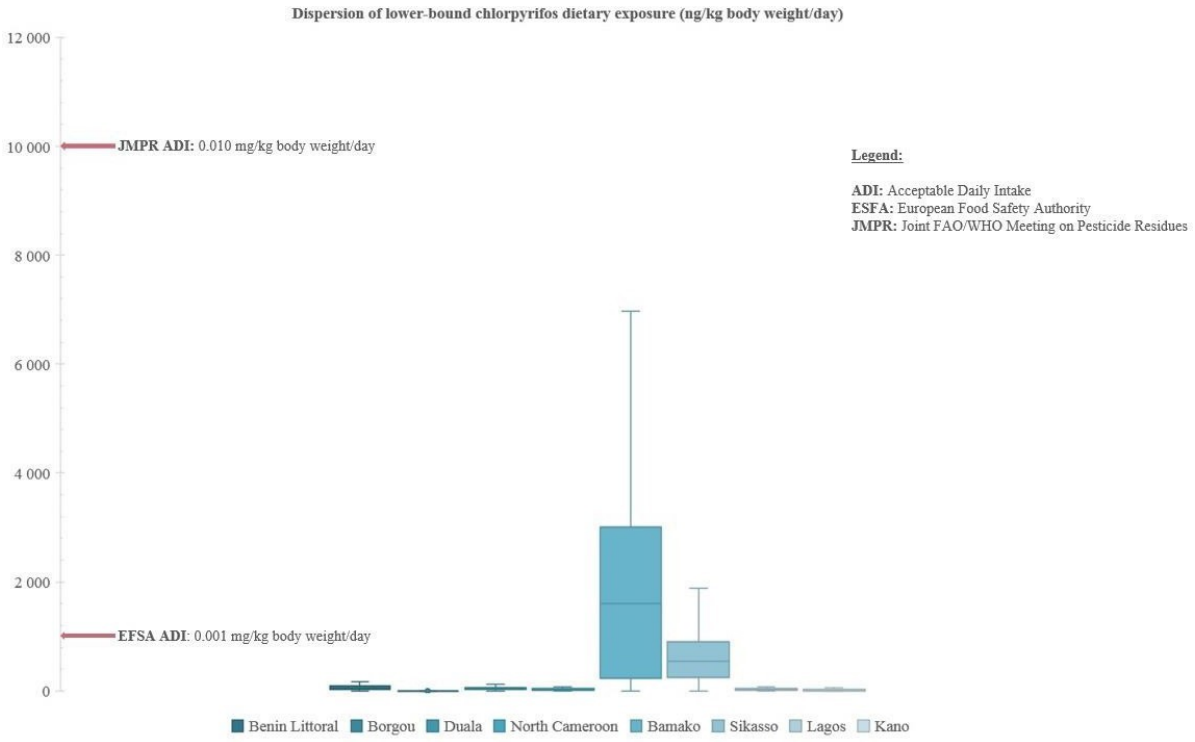


Figure 31: Box-and-whisker plot of chlorpyrifos lower-bound exposure in eight study centres

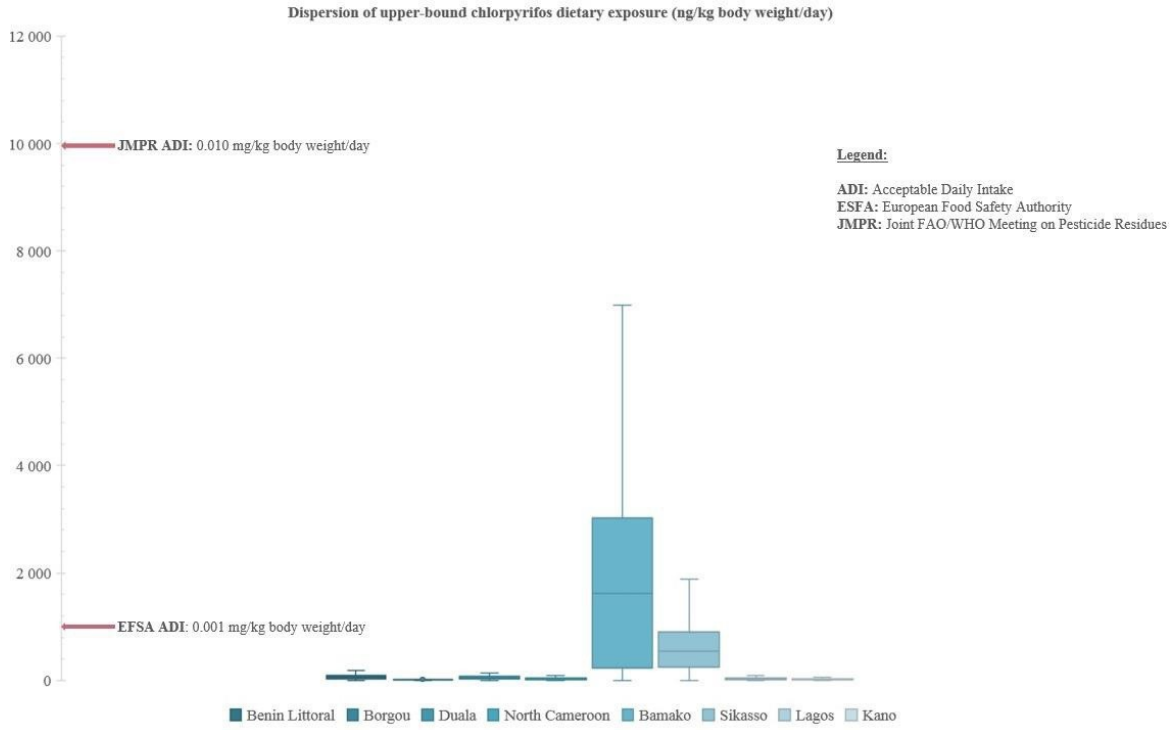


Figure 32: Box-and-whisker plot of chlorpyrifos upper-bound exposure in eight study centres

Table 16: Exposure to chlorpyrifos and exceedance of JMPR and EFSA ADIs.

Exposure (ng/kg bw/d)		Mean		P95		%> JMPR ADI (0,01mg/kg bw/d)*		%> EFSA ADI (0,001mg/kg bw/d)**		Contributors		
		LB	UB	LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	64	71	164	177	0%	0%	0%	0%	Rice	100	90
	Borgou	0.1	8	0.3	16	0%	0%	0%	0%	Beef	84	1
										Smoked fish	15	0,2
CAMEROON	Duala	51	56	114	123	0%	0%	0%	0%	Tomato	70	63
										Wheat/bread	18	17
	North	29	37	69	78	0%	0%	0%	0%	Green leaves	84	67
MALI	Bamako	2 153	2 161	6 962	6 970	0.3%	0.3%	**61%**	**62%**	Smoked fish	100	99
	Sikasso	624	631	1 586	1 595	0%	0%	**21%**	**22%**	Smoked fish	100	99
NIGERIA	Lagos	29	34	68	75	0%	0%	0%	0%	Cassava dry	45	41
										Peanut oil	20	17
	Kano	22	28	85	91	0%	0%	0%	0%	Peanut oil	69	55
										Beans	14	11

* JMPR 2004, 178th Report
**EFSA 2014

The chronic dietary exposure only exceeded the JMPR (2004) ADI of 0.01 mg/kg body weight/day in four of the 7 291 households from Bamako, due to the almost exclusive contribution of smoked fish to chlorpyrifos dietary exposure. However, considering the EFSA ADI of 0.001 mg/kg body weight/day, the percentage of households which exceeded the threshold, was considerably higher (61-62% in Bamako and 21-22% in Sikasso). The difference between the chlorpyrifos lower-bound and upper-bound

exposures in Figures 31 and 32, and in Table 16 is low in Mali, which means that the

uncertainty due to censored data is minimal. It is important to note that the smoked fish samples used for the dietary exposure calculations were not the same (local samples collected in both centres and prepared as consumed). No household outside of Mali was exposed in excess of the JMPR or EFSA chlorpyrifos ADIs.

3.3.3.2. Dichlorvos

Table 17: Exposure to dichlorvos and exceedance of JMPR ADI.

Exposure (ng/kg bw/d)		Mean		P95		%> JMPR ADI (0,004mg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	0	7	1	14	0	0	Cassava dry	100	11
	Borgou	0.0	8	0.0	16	0	0	ND	-	-
CAMEROON	Duala	13	18	26	33	0	0	Wheat/bread	46	34
								Peanuts	35	25
								Maize	14	11
	North	23	31	64	72	0	0	Peanuts	92	68
MALI	Bamako	0	8	0	13	0	0	ND	-	-
	Sikasso	0	7	0	11	0	0	ND	-	-
NIGERIA	Lagos	163	164	325	330	0	0	Cassava dry	34	34
								Yam fresh	18	18
								Rice	17	16
								Beans	14	14
	Kano	58	63	173	180	0	0	Beans	49	44
								Rice	28	27

* JMPR 2011, 211th Report

No household was exposed above the JMPR ADI of 0.004 mg/kg body weight/day. The dichlorvos dietary exposure of the populations in Nigeria was, however, 3-30-fold higher than that of the populations in Benin, Cameroon, and Mali. As shown in Table 17, the difference between the lower-bound and upper-bound dichlorvos dietary exposure is important, except in Nigeria, where the exposure exceeds that of the other countries. The exposure pattern is consistent with the dichlorvos detection rates for Nigeria presented in the occurrence data article.

3.3.3.3. Profenofos

Table 18: Exposure to profenofos and exceedance of JMPR ADI.

Exposure (ng/kg bw/d)		Mean		P95		%> JMPR ADI (0,03mg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	**Littoral**	0	7	0	14	0	0	-	-	-
CAMEROON	**Borgou**	0.1	8	0.3	16	0	0	Beef	100	1
	Duala	0.1	6	0.2	12	0	0	Peanuts	76	5
								Onion and Garlic	24	2
	North	0.04	8	0	15	0	0	Tomato	100	2
MALI	**Bamako**	54	62	128	138	0	0	Green leaves	58	50
								Smoked fish	40	35
	Sikasso	4	11	10	19	0	0	Smoked fish	82	32
								Green leaves	14	6
NIGERIA	**Lagos**	3	11	8	20	0	0	Chili	97	30
	Kano	0	7	0	14	0	0	Plantain	53	0,04
								Peans	47	0,04

* JMPR 2007

Dietary exposure to profenofos was low both in the lower-bound and upper-bound hypothesis, and no household was exposed above the ADI, as shown in Table 18. The uncertainty due to non-detects was high with the exception of Bamako, where exposure was higher than in other centres. The exposure pattern is consistent with the profenofos detection rates for Mali presented in the occurrence data article.

3.3.3.4. Cypermethrin

Table 19: Exposure to cypermethrin and exceedance of JMPR ADI.

Exposure (ng/kg bw/d)		Mean		P95		%> JMPR ADI (0,02mg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	50	56	147	155	0	0	Pasta	87	77
								Cassava dry	13	12
	Borgou	17	25	86	96	0	0	Pasta	85	58
								Beans	11	8
CAMEROON	Duala	239	245	614	622	0	0	Tomato	63	62
								Green leaves	35	35
	North	96	104	240	249	0	0	Green leaves	96	89
								Smoked fish	86	69
MALI	Bamako	35	43	103	113	0	0	Wheat/bread	14	11
								Smoked fish	53	22
	Sikasso	5	12	10	20	0	0	Wheat/bread	18	8
								Cassava dry	28	26
NIGERIA	Lagos	29	34	59	66	0	0	Beans	20	18
								Wheat/bread	16	14
	Kano	6	13	19	31	0	0	Wheat/bread	28	14
								Tomato	13	6
								Chili	12	6
								Gombo=okro	10	5

* JMPR 2006; 187th report

No household was exposed in excess of the JMPR cypermethrin ADI (WHO, 2006). The highest chronic dietary exposures were found in the populations of Cameroon, due to the contributions of tomatoes and green leaves, as shown in Table 19. In Duala and North Cameroon, the difference between the lower-bound and upper-bound exposure is contained below 10%. Interestingly, although the cypermethrin detection rates reported in the occurrence data article were higher in Nigeria, the exposure of the populations of Kano and Lagos were among the lowest, as shown in Table 19. This might be explained by a more extensive application of good agricultural practices, while more extensively resorting to cypermethrin in Nigeria.

3.3.3.5. Lambda-cyhalothrin

Table 20: Exposure to lambda-cyhalothrin and exceedance of JMPR ADI.

Exposure (ng/kg bw/d)		Mean		P95		%> JMPR ADI (0,02mg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	5	12	12	23	0	0	Maize	100	60
	Borgou	3	11	11	23	0	0	Beans	100	26
CAMEROON	Duala	13	19	34	44	0	0	Tomato	100	68
	North	33	40	82	91	0	0	Green leaves	98	79
MALI	Bamako	3	11	5	18	0	0	Smoked fish	40	9
								Tomato	25	6
								Wheat/bread	23	6
	Sikasso	2	9	5	15	0	0	Smoked fish	89	22
NIGERIA	Lagos	2	9	6	19	0	0	Other vegetables	51	10
								Tomato	49	11
	Kano	6	13	22	31	0	0	Beans	94	43

* JMPR 2007; 191st report

No household was exposed in excess of the JMPR lambda-cyhalothrin ADI (2007). The highest chronic dietary exposures were assessed for the populations of Cameroon, due to the contribution of tomatoes and green leaves. As shown in Table 20, with the exception of North Cameroon, the difference between the lower-bound and upper-bound lambda-cyhalothrin exposures exceeded 10%.

3.3.3.6. Permethrin

Table 21: Exposure to permethrin and exceedance of JMPR ADI.

Exposure (ng/kg bw/d)		Mean		P95		%> JMPR ADI (0,05mg/kg bw/d)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	0	7	0	14	0	0	-	-	-
	Borgou	28	36	110	121	0	0	Beans	83	64
								Cassava dry	17	14
CAMEROON	Duala	8	15	24	33	0	0	Beans	71	42
								Green leaves	27	16
	North	281	289	1 084	1 093	0	0	Beans	100	97
MALI	Bamako	12	20	50	59	0	0	Cassava dry	83	49
								Beans	10	6
	Sikasso	1	8	3	13	0	0	Beans	70	12
								Smoked fish	27	5
NIGERIA	Lagos	8	15	22	33	0	0	Palm oil	60	33
								Beans	23	15
								Chili	15	8
	Kano	1	8	2	14	0	0	Other nuts and seeds	93	7

* JMPR 1999; 153rd report

No household was exposed in excess of the JMPR permethrin ADI (1999). The highest dietary exposures were assessed for the populations of North Cameroon, due to the contribution of beans. Nonetheless, none of the 95th percentile permethrin dietary exposures exceeded 2% of the JMPR ADI. As shown in Table 21, at the maximum exposure, the uncertainty due to the difference between the lower-bound and upper-bound exposures remained below 1%.

3.3.3.7. Other detected pesticides

Chronic dietary exposure to other detected pesticides (half of which were only detected once in the whole study), were assessed. Because it does not matter much if the lower-bound exposure is very different when the upper-bound exposure is already very low compared to the available ADI, only the upper-bound values are displayed in Table 22. The dietary exposure assessment shown in this table reveals no particular health concerns due to the intake of pesticides, with a maximum coverage of the ADI of only 16% (North Cameroon) in the maximalist hypothesis where the concentration of all the non-detects is the limit of detection.

Table 22: Percentage of pesticide ADI covered by the upper-bound 95th percentile dietary exposure.

Chemical	Evaluation year (JMPR by default)	ADI (mg/kg)	CAMEROON		BENIN		MALI		NIGERIA	
			Duala	North	Littoral	Borgou	Bamako	Sikasso	Lagos	Kano
2,4-D	2001	0.01	1	1	1	2	1	1	1	1
Acetamiprid	2011	0.07	0	1	0	0	0	0	0	0
Acrinathrin	2013 (EFSA)	0.01	1	1	1	2	1	1	1	1
Atrazine	2007	0.02	1	1	1	1	1	1	1	1
Azoxystrobin	2008	0.2	0	0	0	0	0	0	0	0
Boscalid	2006	0.04	0	0	0	0	0	0	0	0
Carbendazim	2005	0.03	0	0	0	1	0	0	0	0
Chlorantraniliprole	2008	2	0	0	0	0	0	0	0	0
Chlormequat	1999	0.05	0	0	0	0	0	0	0	0
Chlorpropham	2005	0.05	0	0	0	0	0	0	0	0
Chlorpyriphos methyl	2009	0.01	1	1	1	2	1	1	1	1
Cyfluthrin	2006	0.04	0	0	0	0	0	0	0	0
Deltamethrine	2002	0.01	1	1	1	2	1	1	1	1
Dimethoate	2003	0.002	8	1	1	1	1	1	2	1
Endosulfan	1998	0.006	0	0	0	0	0	0	0	0
Fenpropimorphe	2004	0.003	4	5	5	5	4	4	4	5
Imazalil	2005	0.03	0	0	0	1	0	0	0	0
Imidacloprid	2001	0.06	0	0	0	0	0	0	0	0
Indoxacarb	2005	0.01	1	2	1	2	1	1	1	1
Malathion	2003	0.3	0	0	0	0	0	0	0	0
Metalaxyl	2002	0.08	0	0	0	0	0	0	0	0
Orthophenylphenol	2008 (EFSA)	0.4	0	0	0	0	0	0	0	0
Phtalimide=Folpet	2007	0.1	0	0	0	0	0	0	0	0
Piperonyl butoxide	2001	0.2	0	0	0	0	0	0	0	0
Pirimiphos methyl	2006	0.03	1	1	0	1	1	0	0	0
Prochloraze	2001	0.01	1	1	1	2	1	1	1	1
Propamocarb	2005	0.4	0	0	0	0	0	0	0	0
Propiconazole	2007	0.07	0	0	0	0	0	0	0	0
Pyrimethanil	2007	0.2	0	0	0	0	0	0	0	0
Thiabendazole	2006	0.1	0	0	0	0	0	0	0	0
Triazophos	2002	0.001	11	15	14	16	14	11	14	14
Trichlorfon	2006	0.002	1	1	1	1	1	1	1	1
Tricyclazole	No ADI									

The highest percentage of the ADI covered by the upper-bound dietary exposure corresponds to triazophos and is consistently between 11% (in Duala) and 16% (in Borgou) of the ADI. Triazophos was only detected once in cottonseed oil in Duala without exceeding the limit of quantification (3 µg/kg < concentration < 10 µg/kg) and never exceeded the limit of detection in any of the other centres. The upper-bound exposure almost exclusively results from the utilisation of the analytical limit as a concentration and the food consumption data, which means that the lower-bound exposure tends towards zero.

3.3.4. Dietary exposure to metals and trace elements

3.3.4.1. Lead

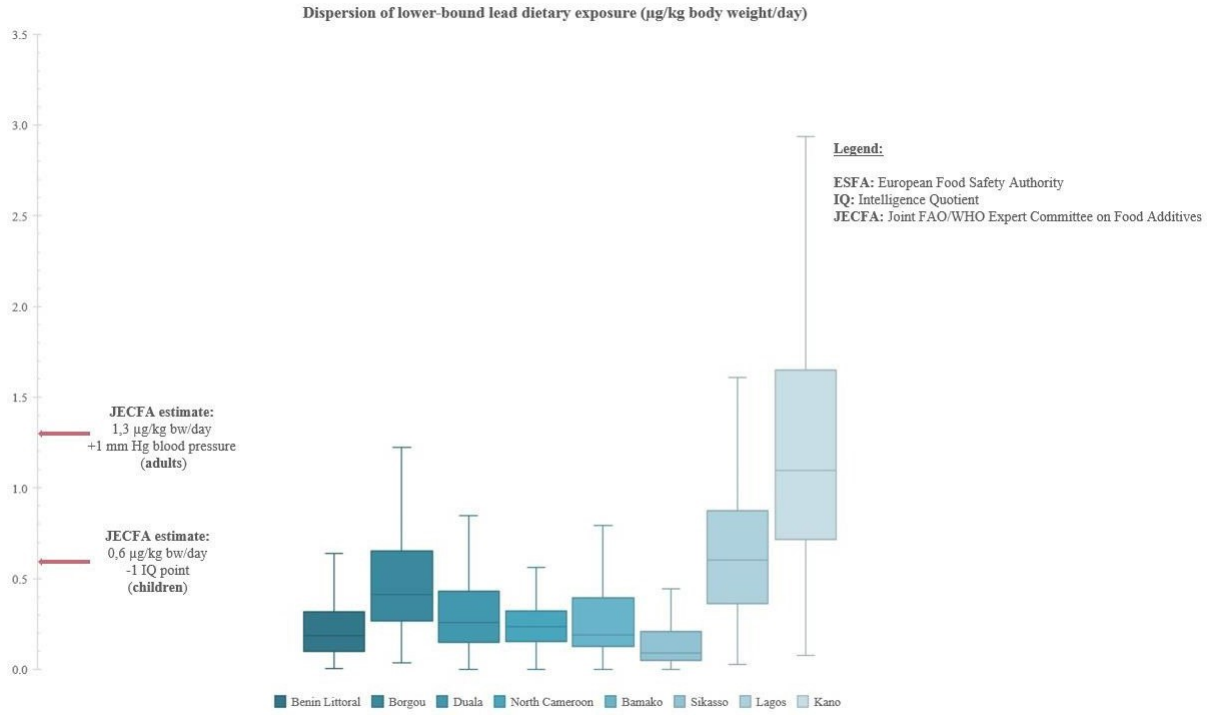


Figure 33: Box-and-whisker plot of lead lower-bound exposure in eight study centres

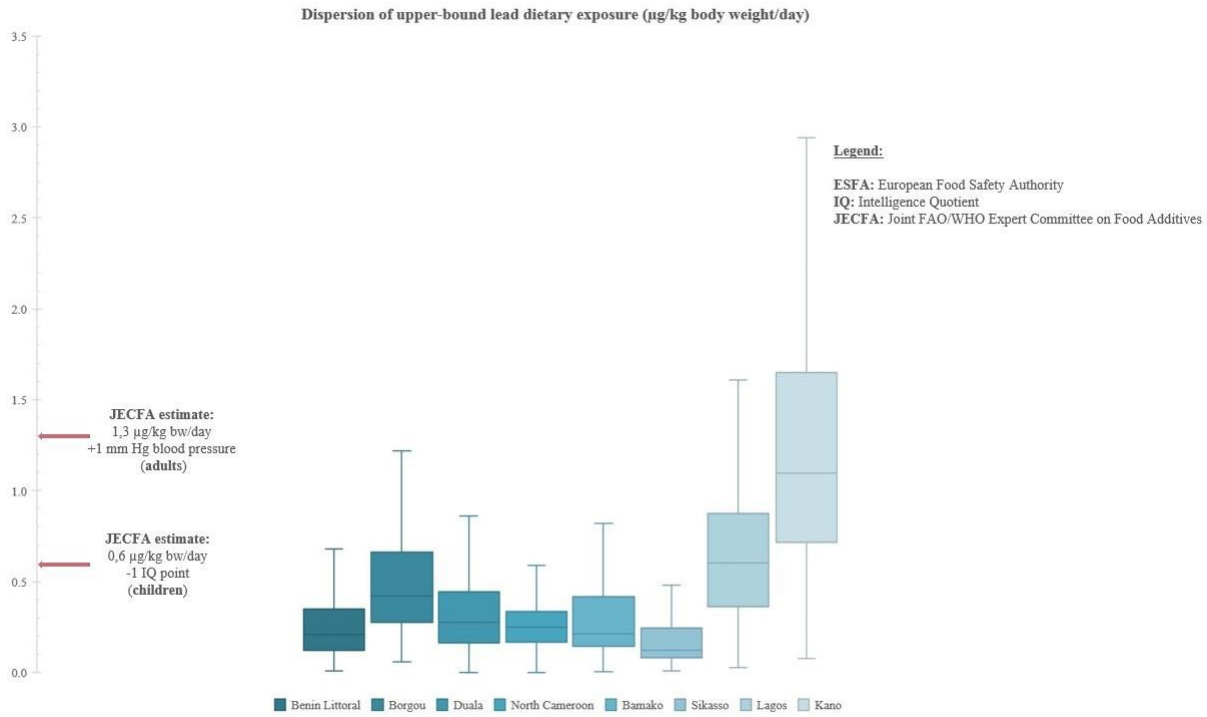


Figure 34: Box-and-whisker plot of lead upper-bound exposure in eight study centres

Table 23: Exposure to lead and extrapolation of JECFA end points.

Exposure (µg/kg bw/d)		Mean		P95		IQ point loss (children)*				mm Hg blood pressure increase (adults)*				Contributors		
		LB	UB	LB	UB	Mean (LB)	Mean (UB)	P95 (LB)	P95 (UB)	Mean (LB)	Mean (UB)	P95 (LB)	P95 (UB)	Core food	%LB	%UB
BENIN	Littoral	0.25	0.27	0.69	0.72	0.4	0.5	1.1	1.2	0.2	0.2	0.5	0.6	Cassava dry	75	69
														Cassava dry	25	25
														Maize	17	17
	Borgou	0.52	0.53	1.20	1.22	0.9	0.9	2.0	2.0	0.4	0.4	0.9	0.9	Sorghum	31	31
														Yam fresh	14	14
CAMEROON	Duala	0.32	0.34	0.85	0.88	0.5	0.6	1.4	1.5	0.2	0.3	0.7	0.7	Cassava dry	45	43
														Green leaves	24	23
														Cassava fresh	12	12
	North	0.26	0.27	0.51	0.53	0.4	0.5	0.9	0.9	0.2	0.2	0.4	0.4	Green leaves	13	13
														Maize	37	36
														Peanuts	15	15
MALI	Bamako	0.35	0.37	1.18	1.22	0.6	0.6	2.0	2.0	0.3	0.3	0.9	0.9	Cassava dry	61	59
														Rice	15	15
														Cassava dry	65	55
	Sikasso	0.16	0.20	0.56	0.60	0.3	0.3	0.9	1.0	0.1	0.2	0.4	0.5	Smoked fish	17	15
														Cassava dry	54	54
NIGERIA	Lagos	0.67	0.68	1.42	1.43	1.1	1.1	2.4	2.4	0.5	0.5	1.1	1.1	Sorghum	58	58
														Millet	22	22
	Kano	1.24	1.24	2.65	2.65	2.1	2.1	4.4	4.4	1.0	1.0	2.0	2.0			

* JECFA 2011, 73rd Report

Chronic dietary exposure was particularly high in Nigeria, especially in Kano. Using the JECFA endpoints (WHO, 2011) highly exposed households (95th percentile) from Kano may undergo a blood pressure increase of 2.0 mm Hg. In the case of children, a loss of 4.4 IQ points is to be expected. In the case of the mean exposure levels, children in Kano may undergo a 2.1 loss of IQ points and a 1.0 mm Hg increase would be expected in the case of adults. Exposure in excess of the 0.6 µg/kg body weight/day (1 IQ point loss) were observed at the 95th percentile in Borgou, Duala, Bamako, Lagos, and Kano. The difference between the lower-bound and upper-bound dietary exposures to lead are not perceptible in Figures 33 and 34 or in Table 23, which can be explained by the sensitive analytical method, combined with relatively high concentrations in the tested food items.

While analysing these results, it is important to bear in mind that we did not design the study to assess children's exposure, and therefore children's dietary exposure may be even more severe in all study centres, than the ones assessed and displayed in Table 23.

3.3.4.2. Aluminium

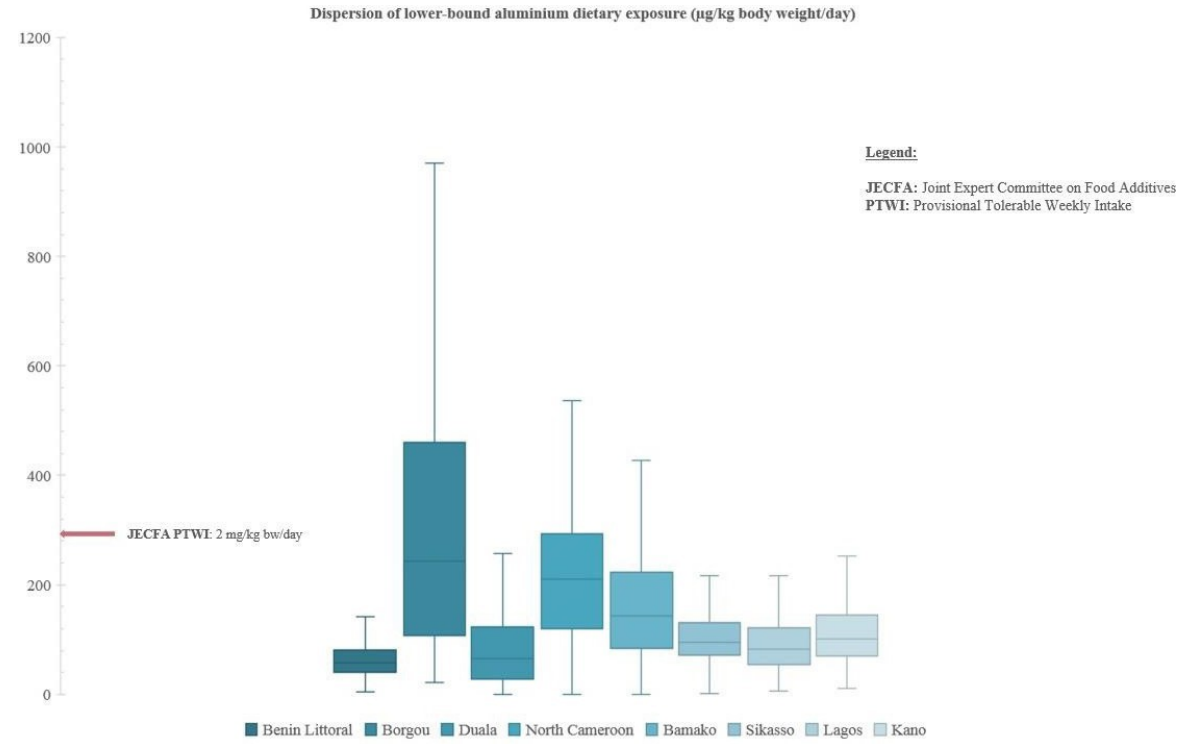


Figure 35: Box-and-whisker plot of aluminium lower-bound exposure in eight study centres

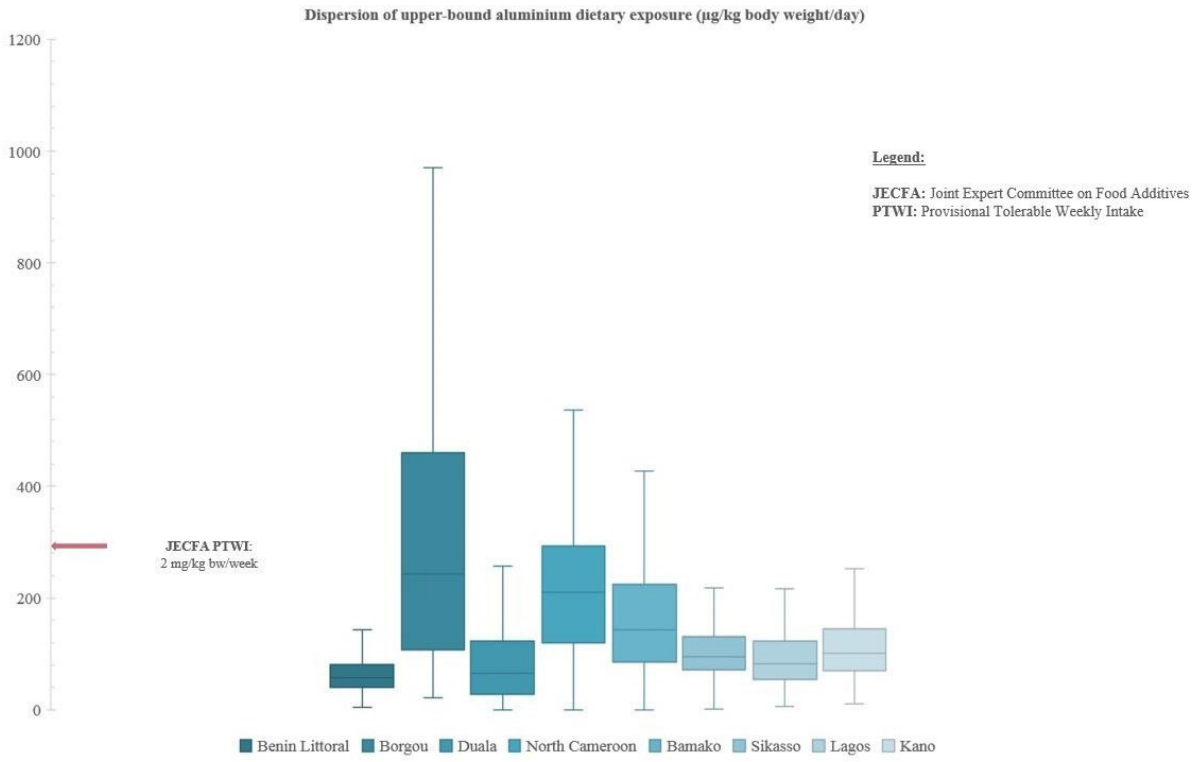


Figure 36: Box-and-whisker plot of aluminium upper-bound exposure in eight study centres

Table 24: Exposure to aluminium and exceedance of the JECFA PTWI.

Exposure (µg/kg bw/d)		Mean		P95		%> PTWI (2mg/kg bw/week)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	65	65	129	129	0.3	0.3	Maize	43	43
								Tomato	17	17
								Casava dry	17	17
	Borgou	337	337	955	955	**45**	**45**	Sorghum	57	57
								Maize	17	17
CAMEROON	Duala	92	92	273	274	4.0	4.0	Green leaves	61	61
	North	211	211	411	411	**27**	**27**	Maize	64	64
								Green leaves	14	14
								Peanuts	13	13
MALI	Bamako	166	167	386	386	**14**	**14**	Cassava dry	23	23
								Millet	26	26
								Smoked fish	10	10
								Sorghum	21	21
	Sikasso	105	105	190	190	0.3	0.3	Smoked fish	43	43
								Sorghum	15	15
								Peanuts	11	11
NIGERIA	Lagos	93	93	188	188	0.3	0.3	Cassava dry	45	45
	Kano	112	112	230	230	0.8	0.8	Sorghum	38	38
								Millet	29	29
								Maize	10	10

* JECFA 2011, 74th Report

The imperceptible difference between the lower-bound and upper-bound exposures is shown in Figures 35 and 36, as well as in Table 24. This can be explained by the censored data having virtually no impact, due to the analytical limits and the ubiquitous presence of Al in African foods, as presented in the occurrence article dealing with metals and trace elements (Jitaru et al., 2019, submitted). The JECFA PTWI of 2 mg/kg body weight/week takes nephrotoxicity into consideration and corresponds to an exposure of 286 µg/kg body weight/day, the unit that was used to assess metals and trace elements. The 95th percentile chronic dietary exposure of the populations studied in Borgou (45% of households), North Cameroon (27%), and Bamako (14%) exceeded the PTWI.

3.3.4.3. Arsenic

Table 25: Exposure to arsenic and exceedance of the JECFA PTWI.

Exposure (µg/kg bw/d)		Mean		P95		MOE/BMDL0,5 lung cancer (3 µg/kg bw/d)*				Contributors		
		LB	UB	LB	UB	Mean (LB)	Mean (UB)	P95 (LB)	P95 (UB)	Core food	%LB	%UB
BENIN	Littoral	0.29	0.31	0.73	0.76	10.4	9.6	4.1	4.0	Smoked fish	75	70
										Rice	15	14
	Borgou	0.28	0.30	0.94	0.96	10.6	9.9	3.2	3.1	Smoked fish	61	57
										Sorghum	20	18
CAMEROON	Duala	0.78	0.79	1.96	1.97	3.9	3.8	1.5	1.5	Sea fish	58	57
										Rice	20	19
										Smoked fish	19	18
	North	0.14	0.15	0.31	0.33	21.7	20.1	9.6	9.2	Sea fish	45	43
										Rice	22	22
										Smoked fish	19	18
MALI	Bamako	0.54	0.56	0.94	0.96	5.5	5.4	3.2	3.1	Rice	91	89
	Sikasso	0.12	0.15	0.38	0.40	24.0	20.3	7.9	7.4	Rice	89	78
NIGERIA	Lagos	0.43	0.45	1.28	0.00	6.9	6.6	2.3	2.3	Sea fish	71	68
										Rice	21	20
	Kano	0.25	0.26	0.59	0.61	12.1	11.5	5.1	4.9	Rice	46	44
										Sorghum	25	24
										Sea fish	18	18

* JECFA 2011, 72nd Report

The JECFA removed the former arsenic PTWI of 0.015 mg/kg body weight/week (corresponding to 2.1 µg/kg body weight/day) because the newly established BMDL0.5 (3 µg/kg body weight/day) for lung cancer incidence was of the same magnitude. Arsenic is not considered genotoxic, according to available studies, and therefore its margin of exposure does not have a typical threshold. The MOEs were all superior to 1 in the study centres, based on the total arsenic exposure assessed by the SSA, without speciation. Because arsenobetaine is by far the most abundant form in fish and the least toxic one, MOEs based on inorganic arsenic would be even higher than the ones estimated in Table 25. The slight difference between lower-bound and upper-bound exposures result from the sensitive ICP-MS analytical method.

3.3.4.4. Cadmium

Table 26: Exposure to cadmium and exceedance of the JECFA PTWI.

Exposure (µg/kg bw/d)		Mean		P95		%> PTMI (25µg/kg bw/month)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	0.05	0.05	0.10	0.11	0	0	Tomato	47	43
								Rice	20	18
								Pasta	12	11
	Borgou	0.04	0.05	0.09	0.10	0	0	Yam fresh	36	33
								Smoked fish	24	21
CAMEROON	Duala	0.08	0.08	0.17	0.17	0	0	Rice	34	34
								Green leaves	18	18
								Wheat/bread	13	12
	North	0.05	0.05	0.10	0.10	0	0	Peanut	33	31
								Green leaves	18	17
								Rice	18	16
								Maize	10	15
MALI	Bamako	0.07	0.07	0.10	0.11	0	0	Rice	61	59
	Sikasso	0.02	0.02	0.04	0.04	0	0	Rice	23	19
								Millet	22	18
								Peanut	21	17
NIGERIA	Lagos	0.09	0.09	0.18	0.18	0	0	Beef	24	24
								Rice	26	25
								Wheat/bread	19	19
	Kano	0.04	0.05	0.11	0.12	0	0	Rice	54	50
								Wheat/bread	19	13

* JECFA 2013, 77th Report

The small difference between lower-bound and upper-bound exposures result from the sensitive method, as shown by Table 26. None of the 7 291 households' exposures exceeded the JECFA (2013) cadmium PTMI of 25 µg/kg body weight/month, which corresponds to 0.83 µg/kg body/day.

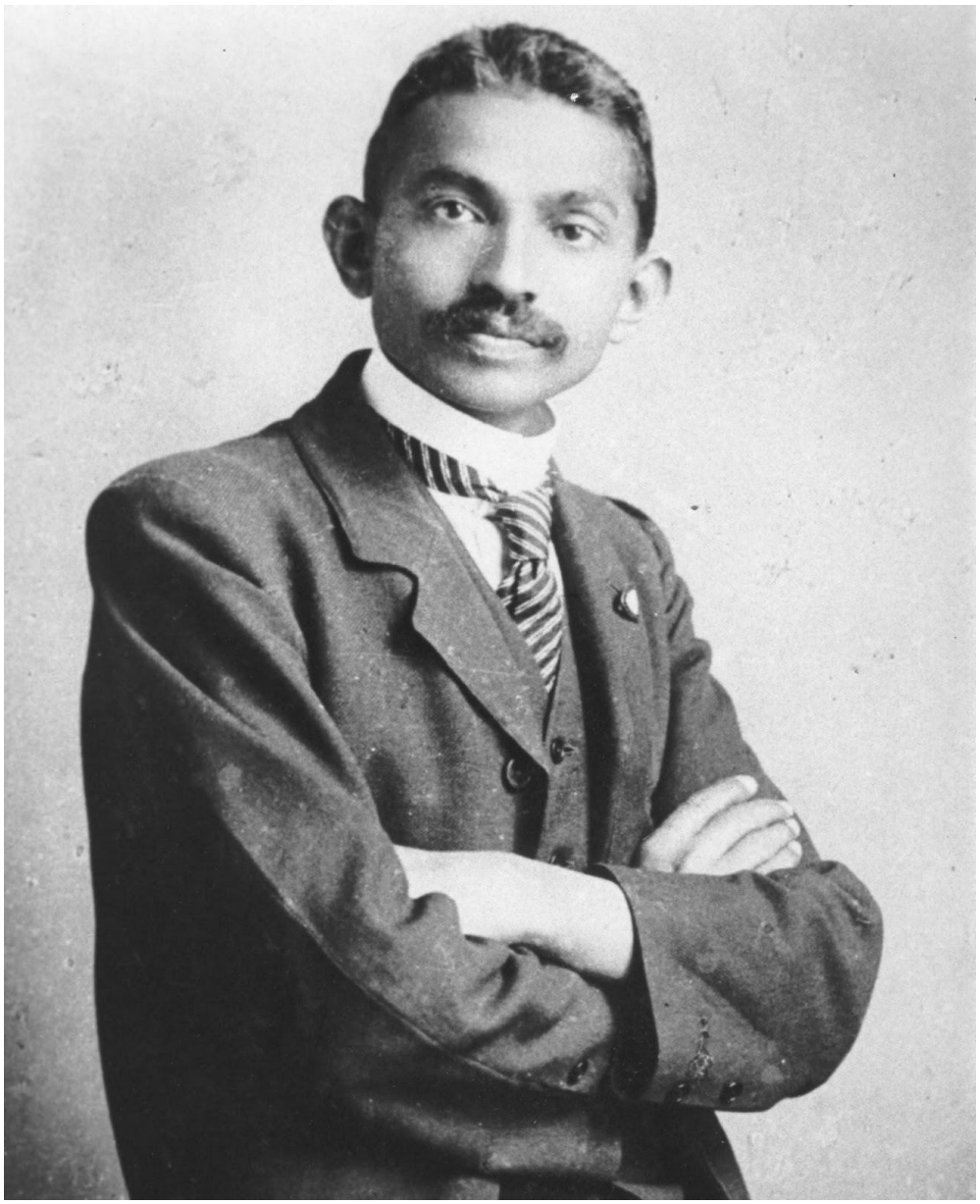
3.3.4.5. Mercury

Table 27: Exposure to mercury and exceedance of the JECFA PTWI.

Exposure (µg/kg bw/d)		Mean		P95		%> PTWI (4µg/kg bw/week)*		Contributors		
		LB	UB	LB	UB	LB	UB	Core food	%LB	%UB
BENIN	Littoral	0.02	0.15	0.06	0.28	0	0	Smoked fish	71	12
								Maize	0	32
	Borgou	0.17	0.29	0.40	0.57	1.5	4.8	Yam fresh	89	56
								Maize	0	20
CAMEROON	Duala	0.06	0.15	0.16	0.32	0.2	0.3	Salt	59	23
								Sea fish	30	11
	North	0.01	0.15	0.01	0.27	0	0	Sea fish	44	1
								Smoked fish	31	2
								Fresh water fish	25	1
MALI	Bamako	0.01	0.17	0.03	0.27	0	0	Smoked fish	100	6
								Rice	0	39
	Sikasso	0.01	0.13	0.02	0.21	0	0	Smoked fish	100	6
								Maize	0	27
								Millet	0	16
								Rice	0	17
								Sorghum	0	14
NIGERIA	Lagos	0.02	0.15	0.06	0.28	0	0	Sea fish	87	11
								Cassava dry	0	20
								Rice	0	18
	Kano	0.02	0.15	0.06	0.27	0	0	Salt	69	11
								Maize	0	14
								Millet	0	14
								Rice	0	13
								Sorghum	0	25

* JECFA 2011, 72nd Report

The JECFA PTWI applicable to inorganic mercury (4 µg/kg body weight/week) corresponds to an exposure of 0.57 µg/kg body weight/day. All total mercury exposure was below the PTWI at the 95th percentile, although in the case of Borgou, the P95 exposure was almost at the PTWI (4.8% percent of households were in excess of the PTWI) in the upper-bound hypothesis. This value was three times the proportion of households in the lower-bound hypothesis. Even though the impact of censored data on the uncertainty of the mercury measurement (lower-bound vs upper-bound) is high, it is unlikely that a major health concern is at stake.



'You may never know what results come of your action, but if you do nothing there will be no result'

Mohandas Karamchand Gandhi (2 October 1869 - 30 January 1948)

4. CONCLUSIONS

I started this document stating that consumers and public health institutions are increasingly aware of the importance of eating safe food. However, in Africa, according to a study conducted by Omari and colleagues (2018) in Ghana, the public was less worried about chemical hazards than unhygienic selling, cooking, and serving environments. Furthermore, the degree ofconcern expressed by interviewees about the safety risks of foods produced around mining sitesand aflatoxins was relatively low, compared to other food safety risks (Omari et al., 2018). Thisstudy provides new evidence of food safety risks, which can be used as a foundation to better protect consumers who need to be sensitized and informed.

4.1. Methodological Aspects of the SSA-TDS

4.1.1. Strengths

4.1.1.1. Regional approach

The fact that this TDS encompassed several African countries is unprecedented to the best of our knowledge. Although there are examples of multinational projects (the EU TDS Exposure Project, in particular), with common standard operating procedures (Pité et al., 2018) and, for the European TDS, a common food list (Akhandaf et al., 2018), they were carried out individually by each country with their own choice of analytes, laboratories, and sampling plans.

In the case of the SSA-TDS, the fact that the sampling methodology, the food classification, the number of analytes, and the analytical laboratories were shared, and that the number of samples were similar among the study centres from different countries, makes the exposure data eminently comparable.

4.1.1.2. Performances of analytical methods

We selected the analytical laboratories based on their analytical performance, the width of the chemical hazard spectrum for which they were competent, and more importantly, their capability to provide robust data even at ultra-trace levels. These very low limits of detection

and quantification were a guarantee to ensure that the uncertainty of the dietary exposure in relation to the impact of non-detects (censored data) in the upper-bound maximalist hypothesis remained low.

The combination of the 12-pooled-sample approach with these low analytical limits made the SSA-TDS a powerful tool to characterize contamination levels and patterns, even at very low concentrations. For example, the difference in detection patterns with regard to pesticide occurrence between the Yaoundé TDS (Gimou et al., 2008) and the SSA-TDS show that some analytes would probably not have been detected if the analytical limits had been poor, and if 12 subsamples of equal size had not been collected and pooled together.

4.1.1.3. Spectrum of analytes

The number of analytes covered by the SSA-TDS (872) was an undeniable asset in the food contamination characterization process. Notably, more than the half of these analytes were covered in 470 pesticides and more than a third by 295 mycotoxins and other fungal, plant, and bacterial secondary metabolites.

4.1.1.4. Extent of the analytical grid

The fact that we tested fish for pesticides is not very common in terms of the surveillance of phytosanitary substances. We screened pesticides in fish based on the assumption that environmental-level pollution was possible. The fact that we systematically tested pesticides in animal products allowed for the detection of the unexpected contamination of Malian smoked fish with six pesticides, and especially with massive amounts of chlorpyrifos. Although costlier in terms of analytical work, this approach proved to be an asset in terms of risk assessment. Such concentrations seem to reveal an intentional use of chlorpyrifos, which is, of course, not permitted by any standard in fish.

4.1.1.5. Seasonal patterns

In the case of mycotoxins and pesticide residues, the seasonal variation patterns could be captured for the two main seasons of Benin, Cameroon, Mali, and Nigeria: the rainy and dry seasons, using the approach recommended by the TDS Exposure Project (Elegbede et al., 2017). Although the mean concentration of the two seasons was used for the chronic exposure assessment (implying that the variation itself does not impact the risk assessment), capturing the seasonal variation is useful to identify when a food chemical is most prevalent, so that appropriate risk mitigation activities may be deduced from the TDS occurrence data.

4.1.1.6. Cost effectiveness

The multi-centre approach was an asset to reduce the costs of coordination, and to access state-of-the art laboratories involved in academic activities. The use of already existing HBSs was also an advantage in terms of cost, although it required time-consuming efforts to generate a food list fit for the four countries.

The pooled sample approach ensured the representativeness of the individual observed mean concentrations of the food chemicals while reducing the cost of the analytical components of the SSA-TDS. The value of increasing the number of subsamples from 12 to 15 would be somewhat limited, compared to the increase in sampling cost of 20%. The utilisation of some national samples, upon justification that the contamination is not likely to vary significantly between two study centres in the same country also allowed for a 25% decrease in the number of samples.

I need to mention that the laboratories agreed to carry out the analysis with no profit margin whatsoever, which made this achievement possible within the available budget. The second French TDS cost around 5 M€ (ANSES, 2011), the EU TDS Exposure Project 7.6 M€(European Commission, 2016), and the ongoing German Meal study 13 M€ (BfR, 2015). In comparison the SSA-TDS was carried out with a total budget of just 1 000 000 € (1 191 353 USD).

4.1.2. Limitations and weaknesses

4.1.2.1. Observed individual mean concentration of food chemicals

Because we used pooled samples and did not capture the distribution of the contamination within the study centre, the risks faced by some of the households may be underestimated (Dorne et al., 2009). This means that caution is essential when drawing conclusions regarding the safety of certain chemicals.

4.1.2.2. Consumption data derived from household budget surveys

In contrast to most TDSs carried out throughout the world and described in the context part of this manuscript, we used HBSs instead of individual food consumption data. This is probably the most critical weakness in our methodology because it means that we cannot identify within-household variability in dietary exposure patterns. In other words, it would be beneficial to identify who eats what in each household, but this is not possible with our approach.

4.1.2.3. Coverage of the population

The fact that only 7 291 households were investigated out of the 44 431 normal reporting households was based on a decision by national stakeholders (December 2014-January 2015). Although assessing both the diet and the contamination profile of a specific study centre is scientifically interesting in terms of accuracy, it means that the exposure of 37 140 available households was not considered in this study.

4.2. Perspectives

4.2.1. Threats

The pertinent utilization of the study results has implications in terms of roles and responsibilities. In particular, the communication of the results to decision makers is an absolute necessity, to be able to expect a reduction in exposure to mycotoxins, lead, aluminium, PAHs, and chlorpyrifos. Adequate knowledge, policies, strategies, and actions are required to act against the chronic dietary exposure to chemicals. Policy makers therefore need to be involved in the definition and implementation of these policies, strategies, and actions.

Targeting specific actions means better utilization of available resources. The commitment of decision makers is key to achieving resource mobilization. This is a challenging outcome, given the dire financial issues encountered in developing countries, and particularly in Africa.

A very significant threat to the implementation of mitigation measures is the currently insufficient routine monitoring and surveillance of food chemical contamination for conformity assessment, in Africa. This means that laboratory facilities need to be readily available locally. The quality management systems of the African laboratories need to comply with international standards, including by participating in inter-laboratory proficiency tests.

4.2.1.1. Need for additional research

Exposure to organophosphate was associated with neurotoxicity via acetylcholine esterase inhibition (Figueiredo et al., 2018). More recently, it has been suggested that pre-natal exposure to organophosphate pesticides such as chlorpyrifos may be responsible for cardio-metabolic disorders (Declerk et al., 2017). The investigations led by the authors suggest that the mechanism involves DNA-methylation in children carrying the paraoxanase 1 192R-allele,

substituting an arginine to glutamine at position 192. Studying the polymorphism of the Malian population, as well as epidemiological data concerning heart disease, would be a welcome contribution to our knowledge, in the context of the high exposure to chlorpyrifos in Mali.

4.2.1.2. Need for codes of practice and standards

Conformity implies the existence of adequate national standards, possibly based on Codex standards. There is an import threat in relation to the absence of adequate standards for the routine monitoring and surveillance of incriminated food commodities. Codex maximum limits are also currently unavailable (aflatoxin in maize) or insufficiently protective in the African context (maize did not exceed the fumonisin maximum limits in samples, although the exposure exceeded the JECFA PMTDI). The implementation of a lower standard without developing new Codex codes of practice such as the one currently applicable for peanuts (Codex Alimentarius, 2004), could jeopardize food security in terms of availability of food. Instead of destroying non-conforming commodities as a consequence of lowering the maximum limit for fumonisins, a stepwise approach is necessary.

First the need for new codes of practice must be assessed. The fact that oil seeds were not included in the Codex Code of Practice for the reduction of mycotoxins, makes it difficult to tackle the issue of the presence of mycotoxins in edible oils. The same goes for PAHs in smoked fish and edible oils, which the Codex does not currently regulate.

4.2.1.3. Need for health-based guidance values

The absence of JECFA or JMPR health-based guidance value may occasionally be compensated by references from other entities e.g., EFSA. This was the case for citrinin (EFSA,2012), as well as poly and perfluorinated substances (EFSA, 2012).

The fact that the JMPR (FAO, 2004) and EFSA (EFSA, 2014b) ADIs differ by a 10-fold factor in the case of chlorpyrifos poses an interpretation issue concerning the risk assessment completed in Mali, where smoked fish tuned out to be the main contributor to chlorpyrifos dietary exposure (Table 16). In order to illustrate this, Figure 37 shows that, in Mali, the fraction of the populations' exposure exceeding the EFSA ADI is high, but the fraction of the same population that exceeds the JMPR ADI is low.

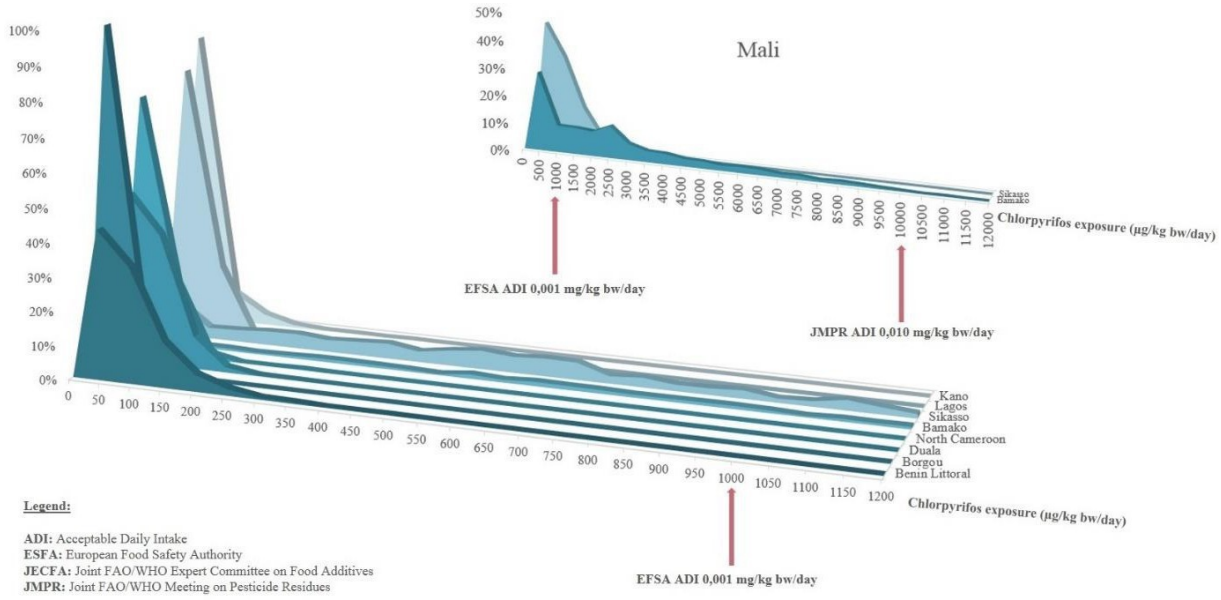


Figure 37: Comparison of JMPR and EFSA ADIs in relation to the distribution of chlorpyrifos dietary exposure

These observations call for advocacy for the timely updating of the JMPR ADI in light of the recent assessment of the most sensitive chlorpyrifos endpoint, namely the red blood cell cholinesterase (EFSA, 2014b).

4.2.2. Opportunities

4.2.2.1. Immediate action to tackle the presence of chlorpyrifos in smoked fish

Following the release of the information, the Malian authorities organized a workshop and a visit to fish markets in March 2019 to discuss the issues with national stakeholders. The data generated during this study may help to find pragmatic solutions to solve the problem. Although the report of the working group is not yet available, it will contribute to documenting and taking appropriate mitigation measures to significantly reduce the exposure of Malians to pesticides allegedly used to preserve smoked fish at ambient temperatures.

4.2.2.2. Long-term mitigation measures

The risk assessment was completed, although additional knowledge may be necessary to better understand contamination throughout the food chain. In particular, the origin of the lead and aluminium contamination will require further investigation to understand how exposure to lead and aluminium can be reduced. Many of the problems identified could find a solution in the application of the codes of practices provided by the Codex (Codex, 2003, 2004, 2009, 2013).

4.2.2.3. Food diversification

Maize, which was not extensively produced in Africa until a century ago (Cherniwchan and Moreno-Cruz, 2019), now represents a significant portion of the total diet of the study countries; Benin: 41%, Cameroon: 20%, Mali: 8%, and Nigeria: 13% (Ingenbleek et al., 2017). Unfortunately, maize is also a major contributor of dietary exposure to several mycotoxins, e.g., aflatoxins and fumonisins. Diversifying the diet, by promoting other crops such as fonio, millet, sorghum, and teff (Fahey, 1998) might contribute to lowering the risk for consumers.

4.2.2.4. Individual food consumption data

If individual food consumption data were available in the countries where the assessments were completed, a full probabilistic dietary exposure assessment would be possible. The FAO/WHO GIFT platform may support this kind of opportunity in the near future (Leclercq et al., 2019).

4.2.2.5. Burden of disease approach

The identification of dietary exposure in excess of the HBGVs may be an opportunity to raise awareness about the burden of food-borne diseases, in Africa and beyond. To take the results of this study a step further, the burden of disease approach, using the disability-adjusted life year would give even more weight to the data generated in the framework of the first ever multi-centre TDS carried out in Sub-Saharan Africa.

The identification of 24 substances exceeding the HBGVs represents an opportunity for improving food safety. Efforts to better protect consumers can be targeted at these analytes, which represent only 3% of the 872 screened analytes.

The identification of priorities in terms of regulations (Codex Alimentarius and national standards) and codes of practice is the main outcome of this study. There is therefore an outstanding opportunity to seize, in order to include the contribution of four African countries in the standard-setting process. The replication of this methodology in other locations of Benin, Cameroon, Mali, and Nigeria may be carried out. This could also be the case in other African countries, and in other developing countries as well. Additional substances, such as bisphenols and phthalates, which were not screened in this survey, may be considered in the future.

In all cases, the risk mitigation of food safety hazards will require a great deal of collective expertise (Montet et al., 2019).

Summary in French

Cette étude de l'alimentation totale a été financée par le Fonds pour l'Amélioration des Normes et le Développement du Commerce (FANDC) via l'Agence des Nations Unies pour l'Alimentation et l'Agriculture (FAO) et l'appui technique de l'Organisation Mondiale de la Santé (OMS), de 2014 à 2018.

Il y a donc eu convergence entre la mise en œuvre du projet FAO/FANDC, dans laquelle j'ai été impliqué, et la réalisation de ce travail de thèse.

Dans quelle mesure les aliments consommés en Afrique Subsaharienne sont-ils sûrs ?

C'est à cette question que j'ai essayé de répondre, en limitant le champ de l'étude à certains résidus et contaminants alimentaires et à quelques localités de quatre pays de la région Africaine.

Pour ce faire, quatre grandes étapes ont été mises en œuvre. Tout d'abord, la production de données de consommation a permis d'obtenir une estimation de la distribution de la consommation alimentaire des ménages. Ensuite, des données d'occurrence ont été générées, de façon à pouvoir caractériser les niveaux moyens de concentration des substances retenues dans les aliments les plus consommés. Alors, l'exposition par voie alimentaire des populations de l'étude pouvait être dérivée du croisement des données de consommation par celle de la contamination des denrées alimentaires. Enfin, un exercice de caractérisation du risque a été initié, sur une sélection de substances détectées, sur la base d'une comparaison entre les niveaux d'exposition par voie alimentaire et les valeurs toxicologiques de référence disponibles.

Données de consommation

La consommation alimentaire au Bénin, Cameroun, Mali et Nigéria a été estimée en partant des données sources d'enquêtes de budget totalisant 72 979 ménages dans les quatre pays. Les données d'achat d'aliments de ces ménages ont été transformées en données de consommation d'aliments tels que consommés en tenant compte de données de prix unitaire, de fraction comestible et de rendement de cuisson.

La consommation de chaque ménage a été normalisée par unité de consommation (équivalent adulte masculin de 60 kg ou EAM) et sélectionnés sur la base d'une consommation énergétique comprise entre 1 200 et 5 100 kcal/EAM/j. Au total la consommation alimentaire de 44 431 ménages normo-évaluants a ainsi été estimée.

Une classification constituée de trois strates et représentant chacune 100% de l'alimentation a été retenue avec des niveaux d'agrégation différents. Le niveau le plus éclaté de la classification des aliments est constitué des 163 à 284 produits alimentaires, tels que collecté par les instituts nationaux de la statistique du Bénin (INSAE), Cameroun (INS), Mali (INSTAT) et Nigéria (NBS). Les produits alimentaires n'étant pas harmonisés entre les institutions concernées, le choix a été fait de créer un niveau, plus agrégé et commun aux quatre pays : les sous-groupes d'aliments ou 84 aliments principaux. C'est ce niveau intermédiaire de la classification qui a été choisi pour élaborer des échantillons, analysés par les laboratoires. Enfin, la strate la plus agrégée de la classification alimentaire est constitué de groupes d'aliments, à savoir *les céréales, les tubercules, les légumineuses, les légumes, les fruits, les noix et oléagineux, les viandes, les œufs, le poisson, les produits laitiers, les huiles et matières grasses, les boissons et les divers*.

Des profils de consommation alimentaires ont pu être mis en avant, avec en particulier des différences en ce qui concerne la consommation des aliments de base, plus riches en céréales dans les centres sahéliens que dans les centres côtiers.

Liste principale des aliments

Une liste d'aliments fortement consommés et devant servir à la constitution d'un plan d'échantillonnage a été élaborée dans chaque pays de façon à optimiser la couverture de la ration moyenne totale en poids (plus de 90%) avec un minimum d'échantillons.

Chaque groupe d'aliment s'est trouvé représenté dans la liste d'aliments à échantillonner et couvert par des sous-groupes à hauteur de 50% minimum (lorsque le groupe d'aliments représente moins de 1% de la ratio totale moyenne en poids). Le taux de couverture des groupes d'aliments qui représentent plus de 1% de la ration totale moyenne a été au minimum de 90%.

Plan d'échantillonnage

Huit centres d'étude ont fait l'objet de prélèvements d'échantillons d'aliments, représentatifs de l'alimentation des 7291 ménages normo-évaluants, résidents dans les localités du Littoral et du Borgou (Bénin), Douala et le Nord (Cameroun), Bamako et Sikasso (Mali) ainsi que Lagos et Kano (Nigéria). Au total 4020 échantillons ont été prélevés, préparés conformément aux habitudes alimentaires pour former 335 composites de même nature, c'est-à-dire à travers l'agrégation de 12 échantillons de même poids et appartenant au même sous-groupe d'aliment. Les critères de sélection des 12 sous-échantillons ont été déterminés pour refléter, autant que

possible les origines des produits importés, les proportions de la consommation nationale moyenne des produits alimentaires au niveau le plus éclaté de la classification, et enfin les principaux lieux d'achat d'aliments (marchés). L'essentiel des échantillons a été prélevé simultanément dans les 4 pays pendant la saison des pluies, c'est-à-dire au mois d'octobre 2017. Cette première vague de collecte d'échantillons a inclus la totalité des 13 groupes d'aliments. Une deuxième vague d'échantillons se limitant à cinq groupes d'aliments (*céréales, tubercules, légumineuses, légumes et fruits*) a été collectée en février 2018, pendant la saison sèche ou harmattan.

Plan d'analyse

Une liste d'analytes constituée de 872 substances a été retenue et croisée avec une grille analytique déterminant, de façon systématique, quels analytes devaient être recherchés dans quelles matrices alimentaires.

La liste d'analytes a *in fine* contenu 470 pesticides, 295 mycotoxines et autres métabolites secondaires fongiques, bactériens et végétaux, 30 éléments traces métalliques (ETM), 16 hydrocarbures aromatiques polycyliques (HAP), 35 polychlorodibenzodioxines/furanes (PCDD/F) et polychlorobiphényles (PCB), 14 composés perfluorés (PFAS) et 12 retardateurs de flamme bromés (RFB).

Tous les analytes ont été recherchés dans les échantillons prélevés pendant la saison des pluies ; les pesticides et les mycotoxines ont fait l'objet de caractérisation également pendant la saison sèche, ceci pour caractériser un éventuel impact saisonnier et ou climatique sur la contamination. Le choix de prélever deux saisons, dans le cas des pesticides a été influencé par le caractère saisonnier des récoltes et de l'application des substances phytosanitaires, tandis que le choix des mycotoxines est lié aux conditions de croissance et de stress des moisissures, susceptibles de varier en fonction des conditions climatiques et environnementales, et d'impacter la production mycotoxinique.

Les échantillons ont été transportés congelés par voie aérienne en présence de neige carbonique vers quatre laboratoires européens et analysés à l'aide de méthodes analytiques validées et accrédités repossant exclusivement sur la spectrométrie de masse.

Données de contamination

Pesticides

Les mesures analytiques ont permis la détection de 40 pesticides au total sur les 4 pays, dont la moitié n'a été détectée qu'une seule fois. Les composites ne contenaient pas de pesticide au-delà de la limite de détection dans 54% des cas. Les autres échantillons contenaient de 1 à 8 pesticides au-delà de la limite de détection, dont six pesticides représentaient 75% des détections. Ces six pesticides appartiennent au groupe des organophosphorés (chlorpyrifos, dichlorvos, profenofos) et des pyréthrinoïdes (cyperméthrine, lambda-cyhalothrine et permethrine). Une forte concentration de chlorpyrifos a été mesurée dans des échantillons de poisson fumé collecté dans les deux centres d'étude du Mali, soit 18 et 5 mg/kg (Bamako et Sikasso, respectivement). Un profil saisonnier a été identifié en ce qui concerne les pesticides, avec des détections plus fréquentes pendant la saison des pluies que pendant la saison sèche, le plus souvent à l'état de traces.

Mycotoxines

L'occurrence de métabolites secondaires, dont les mycotoxines a permis la détection de 164 métabolites, dont la somme des aflatoxines B1, B2, G1, et G2 (AF_{tot}). Tandis que les aflatoxines ont été détectées dans 22% des échantillons, les aliments les plus contaminés par les AF_{tot} étaient l'arachide, l'huile d'arachide et le maïs. Pendant la saison humide, les concentration moyennes étaient significativement plus élevées dans le maïs. L'aflatoxine B1, la plus puissante des mycotoxines représente dans les échantillons analysés entre 76 et 88% de AF_{tot} en moyenne,selon les matrices considérées. La concentration des AF_{tot} a atteint 246 µg/kg dans un composite d'arachide tel que consommé. Le Codex Alimentarius recommande une teneur maximale de 15 µg/kg pour des arachides destinées à des transformations avant consommation, et en mars 2018, le comité FAO/OMS des normes Codex sur les contaminants dans les aliments a proposé une limite maximum de 4 µg/kg, alignée sur celle de l'UE. Le niveau actuellement en vigueur de 15 µg/kg pour les AF_{tot} est dépassé dans 100% des huiles d'arachide, 50% des arachides et 19% des échantillons de maïs.

Les fumonisines ont été détectées dans 14% des échantillons. La somme des fumonisines B1, B2, B3 et B4 (Fum_{tot}) ont dépassé 10 µg/kg dans 94% des composites de maïs, mais n'ont jamais excédé la limite du Codex de 2 mg/kg. Les concentrations de Fum_{tot} dans le maïs ont été en moyenne entre 8 et 14 fois plus élevées que celles retrouvées dans le sorgho. Elles ont été supérieures de 13 à 19 fois que celles quantifiées dans le manioc ayant été soumis à une opération de séchage.

La sterigmatocystine (STC), qui n'est actuellement régulée ni par le Codex ni par l'UE, a été détectée dans 15% des échantillons. La concentration de la STC s'est avérée plus élevée dans deux composites d'huile d'arachide (Littoral du Bénin : 8,3 µg/kg ; Kano : 8,7 µg/kg) et de coton (Nord Cameroun : 9,2 µg/kg). C'est également le cas de l'huile de palme de Lagos (5,3 µg/kg), et du mil (Bamako : 4,8 µg/kg) du sorgho (Sikasso : 2,4 µg/kg).

L'ochratoxine A (OTA) a été détectée dans 10% des échantillons. La plus forte concentration d'OTA a été quantifiée dans un échantillon composite d'huile de palme rouge artisanale (35,4 µg/kg). Le riz et sorgho contenaient également de l'ochratoxine A au-delà de la limite de détection (31% des composites de riz et 20% des composites de sorgho). L'OTA n'est réglementé actuellement ni en ce qui concerne les huiles alimentaires ni en ce qui concerne le riz ou le sorgho.

La citrinine (CIT) qui n'est pas régulée par le Codex Alimentarius a été retrouvée dans 19% des échantillons. Les limites de détection et de quantification étaient de 0.75 et 2.5 µg/kg, respectivement. Les plus fortes concentrations de CIT ont été retrouvées dans le maïs.

La zéaralenone (ZEA) et le déoxynivalenol (DON) ont été chacun détectés dans 6% des échantillons et n'ont jamais dépassé les normes EU et Codex, respectivement.

Eléments traces et métalliques

Alors que le plomb et l'aluminium se sont avérés quasi-ubiquitaires dans les différents groupes d'aliments des différents pays, l'arsenic total et le mercure total ont été principalement quantifiés dans le poisson.

Au Nigéria, les fréquences de concentrations de plomb au-delà de la limite de quantification (entre 1 et 5 µg/kg, selon les matrices) ont été plus importantes que dans les autres pays, notamment avec 100% des échantillons au-dessus de la limite de quantification pour huit groupes d'aliments et plus de 50% de quantification dans quatre sur un total de 13 groupes d'aliments.

Les teneurs en arsenic total se sont avérées plus élevées dans les échantillons de poisson et particulièrement dans le poisson de mer par rapport au poisson d'eau douce. Les concentrations d'arsenic total dans le riz étaient toutes inférieures à la limite maximum proposée par le Codex ainsi que par l'Union Européenne (0,20 mg/kg) applicable à l'arsenic inorganique.

Les teneurs en mercure total de tous les échantillons se sont révélés inférieures au limites maximum proposées par le Codex pour le methylmercure dans le poisson (0,5 mg/kg et 1,0 mg/kg pour les poissons prédateurs).

Selon les pays 10 à 12 groupes d'aliments sur 13 ont 100% des échantillons présentent des valeurs de concentrations en aluminium au-delà de la limite de quantification, qui vont de 0.03 à 0.17 mg/kg selon les matrices. Le sésame a été la denrée la plus chargé en aluminium (662 mg/kg) de notre échantillonnage (Sikasso).

Le niveau moyen de cadmium le plus élevé a été caractérisé dans le groupe du poisson avec 0.036 mg/kg ; aucune limite maximale n'est proposée par le Codex pour cette catégorie, tandis que l'Union Européenne prévoit des limites pour le cadmium dans le poisson de 0.050 à 0.250 mg/kg, selon les espèces considérées.

Hydrocarbures aromatiques polycycliques

Les hydrocarbures aromatiques polycycliques (16) recherchés ont été retrouvés à des concentrations importantes dans les poissons fumés et les huiles alimentaires. Les profils de contamination font clairement ressortir le procédé de combustion comme source dans les deux cas. Tous les composites de poisson excédaient la limite du règlement EU 835/2011 de 2 µg/kg de benzo[a]pyrène et de 12 µg/kg pour la somme du benzo(a)pyrène, benz(a)anthracène, benzo(b)fluoranthène et du chrysène. Selon les composites, le dépassement est de 2 à 50 fois la limite maximum En termes de dépassement, les huiles ont dépassé la limite réglementaire dans 40% des cas.

Polluants organiques persistants et composés perfluorés

Les dioxines (PCDD/F), les polychlorobiphényles (PCB), les retardateurs de flamme bromés (PBDE, HBCD, PBB) et composés perfluoroalkylés ont rarement été détectés et dans ces rares cas, les teneurs se sont avérés très faibles par rapport à ce qui a été constaté en Europe.

Exposition par voie alimentaire et caractérisation du risque

L'exposition de 39 pesticides ne posent pas de problème, y compris dans le cas de l'hypothèse maximaliste, dans laquelle la concentration des échantillons non détectés équivaut à la limite analytique traduisant d'une part l'adéquation des performances des méthodes utilisées et un risque sanitaire associé non préoccupant.

Dans le cas du chlorpyrifos, et dans le cas des deux centres d'étude du Mali uniquement (Bamako et Sikasso), nous avons constaté de possibles dépassements de la dose journalière acceptable (DJA) proposée par le JMPR (WHO, 2004) et l'AESSA (EFSA, 2014b), en raison de la concentration massive de chlorpyrifos quantifiée dans le poisson fumé. Compte tenu du fait que les DJA diffèrent d'un facteur 10, et que 10 ans séparent les deux évaluations, en se basant sur la plus récente des deux (AESSA), 21-22% des ménages de Sikasso et 61-62% des ménages de Bamako dépassent la DJA applicable au chlorpyrifos.

Pour l'aflatoxine B1, les populations les plus exposées sont situées au Nord du Cameroun et au Bénin (Littoral et Borgou), où les principaux contributeurs alimentaires ont été le maïs, suivi de l'arachide et l'huile d'arachide. De façon intéressante, bien que les concentrations en aflatoxine B1 dans l'arachide et l'huile d'arachide sont plus élevées que dans le maïs, celui-ci est en moyenne beaucoup plus consommé, ce qui explique sa forte contribution à l'exposition.

Le maïs est aussi le principal contributeur de l'exposition aux fumonisines. Les populations les plus exposées aux Fum_{tot} résident au Nord du Cameroun et au Bénin (Littoral et Borgou), et 72-75% de ces ménages dépassent la dose journalière tolérable de 2 µg/kg poids corporel/jour (JECFA).

L'exposition à la STC, substance génotoxique et cancérogène pour le foie était plus importante au Mali (Bamako et Sikasso) et au Nord du Cameroun que dans les autres centres. Les aliments les plus fortement contributeurs de l'exposition ont été identifiés comme étant le maïs, le sorgho, le mil et l'huile de coton. Les niveaux d'exposition à la STC correspondent à des marges d'exposition, par rapport à la BMDL10 (0,16 mg/kg poids corporel/jour), au 95^{e} percentile comprises entre 5 369 et 7 484, c'est-à-dire en deçà des marges d'exposition considérées comme adéquates (supérieures à 10 000) par le JECFA.

La proportion des ménages du Bénin (Littoral et Borgou), du Nord du Cameroun, de Sikasso et Kano plus exposée que la dose hebdomadaire tolérable de 0,112 µg/kg poids corporel/jour de l'OTA oscille entre 14 et 34%. Les principaux contributeurs de cette exposition varient entre les centres, il s'agit de l'huile de palme rouge, le maïs, le sorgho et le riz.

Entre 98 et 100% des ménages du Bénin dépassent dans notre étude la dose journalière tolérable de 0,2 µg/kg poids corporel/jour pour la CIT. C'est également le cas de 60 à 62% des ménages résident dans le Nord du Cameroun et de 30 à 31% des ménages de Douala et Kano et ce principalement en raison de la contribution du maïs à l'exposition.

Nous avons conclu que les expositions à la ZEA et au DON ne posaient pas de problème majeur vu la marge qui sépare les plus fortes valeurs d'exposition et les DJT correspondantes du JECFA.

L'exposition à l'aluminium est plus élevée dans le Borgou avec 45% des ménages dépassant le dose hebdomadaire tolérable de 2 mg/kg poids corporel/semaine. C'était également le cas pour les ménages du Nord du Cameroun (27%) et de Bamako (14%), dans tous les cas les contributeurs principaux à l'exposition à aluminium étant les céréales (sorgho, maïs et mil).

L'exposition au plomb s'est avérée plus importante au Nigéria que dans les autres pays étudiés.

L'exposition aux 13 hydrocarbures aromatiques polycycliques génotoxiques et cancérogènes à la fois *in vitro* et *in vivo* a été comparée à la BMDL10 applicable au seul benzo[a]pyrène, selon l'approche dite de substitution et recommandée par le JECFA. Les marges d'exposition des percentile 95 des centres étaient toutes inférieures à 10 000, en raison de la contribution du poisson fumé et des huiles de cuisson, ce qui traduit un risque sanitaire non négligeable.

List of acronyms

ADI: acceptable daily intake

AFB: aflatoxin B

AME: adult male equivalent

Anses: French Food Safety Agency

BaP: benzo[a]pyrene

BFR: brominated flame retardant

BMDL: benchmark dose lower confidence limit

CI: confidence interval

CIT: citrinin

DALY: disability adjusted life years

DNA: deoxyribonucleic acid

DON: deoxynivalenol

EFSA : European Food Safety Authority

FAO: Food and Agriculture Organization of the United Nations

FB: fumonisin B

FDA: Food and Drug Administration

FUM_{tot}: total fumonisins

GC: gas chromatography

HBAg: hepatitis B antigen

HBGV: health-based guidance values

HBS: household budget survey

ICP: inductively coupled plasma

IQ: intelligence quotient

IQR: interquartile range

ISO: International Organization for Standardization

JECFA: Joint FAO/WHO Expert Committee on Food Additives

JMPR: Joint FAO/WHO Meeting on Pesticide Residues

LB: lowerbound

LC: liquid chromatography

LOD: limit of detection

LOQ: limit of quantification

MOE: margin of exposure

MS: mass spectrometry

NCHS: National Center for Health Statistics

NIV: nivalenol

NOAEL: No observed adverse effect limit

OTA: ochratoxin A

P95: 96th percentile

PAH: polycyclic aromatic hydrocarbon

PBDE: polybrominated diphenyl ether

PCB: polychlorinated biphenyls

PCDD/F: polychlorinated dibenzo dioxins/furans

PFAS: per and poly fluoroalkyl substances

PFOA: perfluorooctanoic acid

PFOS: perfluorooctane sulfonic acid

PMTDI: provisional maximum tolerable daily intake

POP: persistent organic pollutant

PTWI: provisional tolerable weekly intake

RfD: reference dose

RFB: Retardateur de flame bromé

SD: standard deviation

SOP: standard operating procedure

SSA-TDS: Sub-Saharan Africa TDS

STC: sterigmatocystin

STDF: Standard and Trade Development Facility

TDI: tolerable daily intake

TDS: total diet study

UB: upperbound

WHO: World Health Organization

ZEA: zearalenone

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Titre : Exposition par voie alimentaire de populations du Bénin, Cameroun, Mali et Nigéria, à un large panel de substances chimiques

Mots clés : Etude de l'Alimentation Totale, Exposition par voie alimentaire, Evaluation du risque chimique.

Résumé : L'Etude de l'Alimentation Totale en Afrique Subsaharienne a couvert plus de 90% de l'alimentation moyenne de populations correspondant à 7291 ménages et 4020 échantillons d'aliments, représentatifs des habitudes alimentaires locales et préparés tels que consommés. Les polluants organiques persistants tels que les dioxines, les polychlorobiphényles, les retardateurs de flamme bromés ainsi que les composés perfluorés ont été quantifiés à des teneurs très faibles. Une sélection de 68 substances, dont l'exposition a été étudiée de façon semi-probabiliste a permis de mettre en évidence que 24 de celles-ci sont, de façon individuelle ou en groupe, susceptibles de représenter un fardeau de santé publique. L'Aflatoxine B1, compte tenu des prévalences du virus de l'hépatite B, est susceptible de générer chaque année des cas de cancer du foie, à raison de 0,2 (Douala) - 23,9 (Nord Cameroun) cas additionnels pour 100 000 habitants. La co-exposition à l'aflatoxine B1, à la sterigmatocystine et aux fumonisines est particulièrement préoccupante dans certains centres de l'étude. La co-exposition à des niveaux importants à l'ochratoxine A, à la citrinine et à de l'aluminium, qui sont toutes des substances néphrotoxiques, a été relevé au Bénin. Des teneurs fortes de chlopyrifos dans le poisson fumé consommé au Mali alertent les autorités sur le risque chronique et aigu.
Les expositions aux treize hydrocarbures aromatiques polycycliques dans le poisson fumé et les huiles de cuisson ne permettent pas des marges d'exposition sécurisantes eu égard à leur caractère génotoxique et cancérogène.
L'exposition au plomb, en particulier au Nigeria révèle un risque d'augmentation de la pression artérielle chez les adultes et des pertes de points de QI chez les enfants. Les données générées aideront les gestionnaires de risques à mieux protéger le consommateur. Une étude visant spécifiquement l'exposition du nourrisson et du jeune enfant permettrait d'aller plus loin dans la connaissance du risque chimique en Afrique Subsaharienne.

Title: Dietary exposure of populations from Benin, Cameroon, Mali and Nigeria to a wide spectrum of food chemicals

Keywords: Total Diet Study, Dietary exposure, Chemical risk assessment.

Abstract: The Total Diet Study in Sub-Saharan Africa covered 90% of the average diet of 7291 households with 4020 samples of typical foods prepared as consumed. Persistent organic pollutants such as dioxins, polychlorinated biphenyls and brominated flame retardants, as well as perfluorinated compounds were quantified at very low concentrations. The exposure of a selection of 68 chemicals was assessed, individually or in group, following a semi-probabilistic approach. Among these,
24 substances were characterized as a potential public health concern. The co-exposure to aflatoxin B1 and hepatitis B is likely to induce a morbidity factorin additional cases of liver cancer/100 000 inhabitants/year ranging from 0.2 (Duala) to 23.9 (North Cameroon). The co-exposure to aflatoxin B1, sterigmatocystin and fumonisins is of concern in some of the studied areas.
The co-exposure to ochratoxin A, citrinin and aluminium, each of which are nephrotoxic, represents a concern identified in Benin. High chlorpyrifos concentrations in smoked fish from Mali represent chronic and acute risks for consumers. The 13 genotoxic polycyclic aromatic hydrocarbons in smoked fish and edible oils do not allow for safe margins of exposure.
Lead exposure, particularly in Nigeria, might elevate blood pressure in adults and affect children neurodevelopment.
The generated data will inform the risk managers and help them to better protect consumers. Additional studies, more specifically focussing on theinfant and young child would be a beneficial perspective in terms of contribution to knowledgeabout food chemical risks in Sub-Saharan Africa.