Monitoring and Dietary Risk Assessment of 81 Pesticide Residues in 11 Local Agricultural Products from the 3 Largest Cities of Cameroon

Yamdeu Joseph Hubert Galani[1,2*], Michael Houbraken[2], Abukari Wumbei[2], Joseph Fovo Djeugap[3], Daniel Fotio[4], Yun Yun Gong[1] and Pieter Spanoghe[2]

[1] School of Food Science and Nutrition, University of Leeds, Leeds, LS2 9JT, United Kingdom

[2] Department of Plants and Crops, Faculty of Bioscience Engineering, Ghent University, Coupure Links 653, 9000, Ghent, Belgium

[3] Department of Plant Protection, Faculty of Agronomy and Agricultural Sciences, University of Dschang, P.O. Box 222 Dschang, Cameroon

[4] Inter-States Pesticides Committee of Central Africa, P.O. Box: 16344 Yaounde, Cameroon

*Correspondence: j.h.galaniyamdeu@leeds.ac.uk, josephgalani@gmail.com; +44 113 343 77 24; ORCID: 0000-0003-4841-7414

Highlights

- Residues of 58 pesticides were found in samples of 11 foods from Cameroon.
- Half of the positive pesticides, among which 5 banned, were above their EU MRLs.
- White pepper, maize, Egusi seeds, soybeans and groundnuts were of most concern.
- Hazard quotient of Carbofuran in groundnuts was 22% above the safe value.
- Most of the food items were safe, but the increasing health risk needs attention.

Abstract

This study monitored 81 pesticides residues in 160 samples of 11 dry agricultural products collected in the 3 largest cities of Cameroon, extracted using QuEChERS method and analysed by liquid chromatography tandem mass spectrometry (LC-MS/MS). Residues of 58 (71.6%) compounds were found in the samples, the most distributed pesticides were Imazalil, Triadimenol and Pyrimethanil, and those with the highest average concentrations were Cymoxanil, Thiamethoxam and Thifensulfuron. Half of the positive pesticides were above their European Union maximum residue limits (MRLs) among which Carbaryl, Carbofuran, Malathion, Metalaxyl and Propoxur are pesticides banned in the country. All the 11 food items contained pesticides, the highest contamination rates (12.8% to 5.0%) were found in white pepper, maize, Egusi seeds and groundnuts, while groundnuts, Egusi seeds, maize and soybeans showed the highest residue concentrations (1.46 to 1.37 mg/kg). Pesticide contamination rates were similar in the 3 sampling cities, but Bafoussam and Yaounde had more samples above the MRLs than Douala. Using the food consumption data for Cameroon from the recent Sub-Saharan Africa Total Diet Study, dietary exposure was calculated and potential health risk of Cameroonian consumers was evaluated. Hazard quotient of Carbofuran in groundnuts was 22% above the safe value, the remaining food items could be considered safe for individual pesticide residues, although Triazophos and Metribuzin in maize were of concern. Groundnuts (0.531) and maize (0.443) showed high hazard index, with 17 highly contributing compounds, but there is no reason to be concerned about cumulative exposure to residues from the food items. While the food items are in general safe to eat, to minimize the increasing human health risk of consumers and ensure approval of Cameroon export produces on international market, this study suggests that authorities must regulate the usage of agrochemicals, strengthen the controls for effective implementation of the pesticide bans and implement strong control of obsolete pesticide stocks in the country.

Keywords: Food safety; Pesticide residues; QuEChERS method; Risk assessment; Hazard quotient; Hazard index.

1. Introduction

Pesticides are intensively used in Cameroon by farmers and traders to protect their plants and products during production and postharvest storage (Galani et al., 2018; Kenko, Asanga, Tchamadeu, & Mpoame, 2017; Mahob et al., 2014; Manfo et al., 2012; Matthews, Wiles, & Baleguel, 2003; J. Sonchieu, Ngassoum, Tchatchueng, Srivastava, & Srivastava, 2013; D. Tarla, Manu, Tamedjouong, Kamga, & Fontem, 2015). Despite the numerous advantages of pesticide use in agriculture, their harmful residues can be found in all environmental compartments, but the highest risk for consumers is through consumption of residues in food (Price, 2008). Since certain pesticides are hazardous and toxic to human health, their residues remaining in or on food can pose danger to humans and may cause certain diseases (Aktar, Sengupta, & Chowdhury, 2009): therefore, there is a need for scientific evaluation and control of these products. To avoid the health hazard caused by pesticide residues, regulatory authorities in many countries have established maximum residue limits (MRLs) for various agricultural products. However, the above MRL residue violations often indicate breaches of Good Agricultural Practices but only in very rare circumstances represent cases of health concern. The potential health risks posed by pesticide residues in foods can best be assessed by developing estimates of dietary exposure to pesticides and comparing the exposure estimates to toxicological reference values of health concern like Chronic Reference Dose (RfD), Acceptable Daily Intake (ADI) or Provisional Tolerable Daily Intake (PTDI) (Winter, 2015). An estimate of dietary exposure to pesticide residues in foods is obtained based on the average food consumption per person per day, average adult weight, and level of pesticide residue in food.

Reports show that numerous factors contributing in possible high exposure of consumers such as intensive utilisation and limited knowledge about pesticide use, are found in Cameroon. In fact, because farmers did not receive sufficient training on pesticide application and proper assistance from agricultural extension agents, inappropriate use of pesticides by Cameroonian farmers have been documented in many studies from different parts of the country (Gama, Folorunso, & Adeola, 2016; Kenko et al., 2017; Manfo et al., 2012; Nchare, 2007; Jean Sonchieu, Benoit Ngassoum, Bosco Tchatchueng, Srivastava, & Srivastava, 2010; Jean Sonchieu, Fointama, Akono, & Serri, 2019; Tandi, Wook, Shendeh, Eko, & Afoh, 2014; D. Tarla et al., 2015; Tayoh, Kiyo, & Nkemnyi, 2016). The situation is aggravated by the high number and various types of obsolete pesticides accumulated over the years in the country (D. N. Tarla et al., 2014), and with little or no control around these pesticide stocks, they could be a source of severe acute or chronic pollution (Gimou, Charrondiere, Leblanc, & Pouillot, 2008) or can be used fraudulently (Kenko et al., 2017; Jean Sonchieu, Srivastava, Ngassoum, Tchatchueng, & Srivastava, 2017). For instance, banned compounds including dichlorodiphenyltrichloroethane (DDT) and its metabolites were found in food samples from Cameroon (Galani et al., 2018; Jean Sonchieu et al., 2010). Moreover, Cameroonian cocoa (one of the major export crop product of the country) was banned from European market because of the presence of Metalaxyl residues beyond the European Union MRL of 0.1 mg/kg. As a result, in December 2016, Cameroon Government had prohibited importation, commercialisation and use of metalaxyl-based pesticides which were intensively used in cocoa to control black pod disease (MINADER, 2016).

Despite this increased concern on dietary risk linked to use of agro-pesticides in Cameroon, only few emphasis have been put on assessing how the growing use of pesticides in the country can impact on food safety and consumers health, probably because of lack of food consumption data. The last study in Cameroon dated 12 years ago and showed low dietary exposure to pesticide residues in the capital city, Yaoundé, but the authors recommended that further investigations using more sensitive analytical methods should be planned (Gimou et al., 2008). But more recently, it was found that maize, cowpea and millet samples from northern Cameroon contained pesticide residues above the MRLs (J. Sonchieu et al., 2013; Jean Sonchieu et al., 2010). Additionally, our previous report (Galani et al., 2018) on evaluation of pesticides residues in 12 agricultural products from western highlands of Cameroon, the main food basket of the country, showed that all the samples contained at least one pesticide, 21 pesticides (34.4%) exceeded their European Union MRLs. Residues of Acetamiprid, Pirimiphos-methyl and Dimethoate were largely found in meal samples from Douala and Garoua in Cameroon (Ingenbleek et al., 2019). All these studies suggested a potential high human dietary exposure and highlighted the necessity of continuous monitoring and dietary risk assessment of pesticide residues in Cameroon.

Cocoa, coffee, palm oil, maize, beans, cassava, groundnuts, plantains and bananas are among the most common food items produced in Cameroon. Other items like soybean, chili pepper, Egusi seeds and white pepper are also largely produced and consumed in the country (INS, 2015). Because dried foods are concentrated and therefore may contain higher pesticide residue levels compared to fresh agricultural products, more attention is being given to dried foods in pesticides monitoring programs (Seo et al., 2013). However, there is no report on pesticide residue levels in food commodities harvested in Cameroon and consumed in the major cities of the country. Besides, there is no recent information on health risk assessment of pesticide residues from dietary exposure in the country. Hence, in continuation of the work we previously initiated (Galani et al., 2018), the present study determined residues of 81 pesticides of diverse chemical groups in 160 samples of 11 agricultural products collected in the 3 largest cities (Bafoussam, Douala and Yaounde), using QuEChERS extraction method and analysis by liquid chromatography tandem mass spectrometry (LC-MS/MS). This will allow to monitor the compliance of these foods with the legal limits for targeted pesticide residues. Then we used the residue levels and food consumption data for Cameroon from the Sub-Saharan Africa Total Diet Study (SSA-TDS) (Ingenbleek et al., 2017) to assess the dietary exposure and evaluate the potential health risk to the Cameroonian consumers.

2. Materials and Methods

2.1. Sample collection and residue analysis

Samples of 11 dry agricultural produces i.e., groundnut, soybean, kidney bean, black bean, cowpea, chili pepper, Egusi seed, coffee bean, cocoa bean, maize and white pepper were collected in December 2017 in the main markets of Bafoussam, Douala and Yaounde. Five samples of each food were collected in each market, except coffee which was not available in Yaounde, hence a total of 160 samples. Approximately 300 g of each food was sampled in hard paper envelope within polyethylene plastic bags, sealed, labelled, transported to the Laboratory of Crop Protection Chemistry at Ghent University, Belgium, and kept at 20 °C until grinding, extraction and analysis.

The active ingredients to be screened were selected based on the list of registered agricultural pesticides authorised in Cameroon and the list of banned compounds in the country, established by the Ministry of Agriculture and Rural Development, and recommended to be used on the sample crops (MINADER, 2013). Pesticides extraction and dispersive solid phase extraction (d-SPE) clean-up were performed using the QuEChERS method, and analysis was done by LC-MS/MS previously developed and validated by (Galani et al., 2018). Details of the 81 selected compounds belonging in 28 chemical classes, and the LC-MS/MS parameters can be found in (Galani, Houbraken, Van Hulle, & Spanoghe, 2019). The compliance of quantified pesticides with existing regulations was checked by comparing their residue level with European Union MRLs taken from the EU Pesticides Database (European Commission, 2020).

2.2. Deterministic dietary exposure

Based on the monitoring results of the pesticide residues, human dietary exposure was estimated in order to determine the degree of risk associated to the detected pesticide residues in food samples. To estimate dietary exposure to specific residue for each food, the daily amount of the consumed food by an adult (kg/day) was multiplied with the residue level (expressed in mg/kg) in the food to obtain the pesticide residue intake. The exposure in term of estimated daily intake (EDI, in mg/kg bw per day) was then calculated by dividing the residue intake with the body weight (kg) of an adult.

$$\text{Estimated daily intake (EDI)} = \frac{\text{Food consumption x Pesticide residue amount}}{\text{Body weight}}$$

In the calculation of dietary exposure to pesticide residues, the major sources of uncertainties are food consumption level, body weight, sampling, analytical bias and variation, processing factor and left-censored data (EFSA, 2006). The consumption data for each food was obtained by extracting the mean daily consumption for an adult for Cameroon from the SSA-TDS, which was developed to be used in the completion of quantitative risk assessments with regard to food chemicals (Ingenbleek et al., 2017). Processing factors, that represent the fraction of the chemical lost from the raw food or agricultural commodity during processing steps such as washing, peeling, grinding and cooking, were not taken into account in the determination of exposure to pesticide residues in this work. The consumption values (in g/day) were 36.4 for groundnuts, 1.0 for soybeans, 81.4 for beans, 1.6 for peas, 2.7 for chili pepper, 384.9 for maize and 1.3 for white pepper. For cocoa, consumption value of chocolate (0.7 g/day) was used and for Egusi seed the value from the ‘’Other Vegetables’’ group (1.4 g/day) was taken. For coffee, the value 0.065 g/day was computed by averaging the consumption values of Douala (0.03 g/day) and North Cameroon (0.1 g/day). The average body weight of a Cameroonian adult (64.83 kg) was used (Walpole et al., 2012).

For handling the data reported to be below the limit of detection (LOD) or non-detected (ND) (left-censored) data, the substitution method according to three scenarios was applied (EFSA, 2010): the upper bound which considered non-detected sample values equal the limit of detection (NQ = LOD); medium bound for which non-detected values equal half of the limit of detection (NQ = 1/2 LOD); and lower bound for which non-detected values equal zero (NQ = zero). Therefore, the EDI of each pesticide from consumption of each food was assessed at upper, medium and lower bound scenarios.

2.3. Chronic dietary risk assessment

The risk for the long-term exposure of individual pesticide residues was assessed by using the Hazard Quotient (HQ), calculated by dividing the exposure (EDI in mg/kg bw per day) with the corresponding toxicological reference value, the acceptable daily intake (ADI, in mg/kg bw per day) (EPA, 2011).

$$\text{Hazard Quotient (HQ)} = \frac{\text{Estimated daily intake}}{\text{Acceptable daily intake}}$$

The ADI is an estimate of the daily maximum intake of a substance over a lifetime that will not result in adverse effects at any stage in human life span. The ADI values for pesticides were taken from the European Food Safety Authority (EFSA) Pesticides Database or, for pesticides not approved in the European Union, from the Pesticide Properties DataBase (PPDB). A value of HQ<1 indicates that lifetime consumption of commodity containing the measured level of pesticide residues could not pose health risks.

As residues of many pesticides were found in each commodity, the cumulative risk assessment of the combined exposure from a given commodity was performed by using the Hazard Index (HI) method, which is calculated by summing the HQs of the individual pesticide residue.

$$\text{Hazard Index (HI)} = \sum\nolimits_{i=1}^{n} \text{HQi}$$

A HI value is greater than 1 indicates that the concerned commodity should be considered a risk to the consumers, whereas an HI index below 1 indicates that its consumption should be considered safe (El Hawari, Mokh, Al Iskandarani, Halloum, & Jaber, 2019; Gad Alla, Loutfy, Shendy, & Ahmed, 2015; Reffstrup, Larsen, & Meyer, 2010).

2.4. Data analysis

Descriptive statistics were generated. For each pesticide, the percentage of occurrence in foods, above the MRL and in sampling locations was calculated. The lowest, highest, mean and median residue content were computed. For each food item, the percentage of positive analyses, quantified residues, analyses above MRL and pesticides above MRL were determined. The different in HI between the lower, medium and upper bound scenarios was assessed determined by a two-tailed F-test at 0.05 level of significance, using the data analysis function of Microsoft Excel 2019.

3. Results and Discussion

3.1. Distribution of pesticide residues found in food samples

Residues of 81 pesticides were assessed by LC-MS/MS in 11 highly consumed agricultural products from the three largest cities of Cameroon. A total of 58 (71.6%) compounds were detected and quantified in the 160 samples (Table 1). Herbicides (30.2%), fungicides (30.2%) and insecticides (25.4%) were the most found, whereas acaricides and nematicides accounted only for 6.3% and 7.9%, respectively. In our previous initial work (Galani et al., 2018), only 44 (54.3%) of the 81 compounds were found in the samples of same food items originating from the western highlands of Cameroon. Herbicides represented 16.2%, 30.9% were fungicides, and insecticides were 39.7%. As a comparison, food samples in the present study contained more pesticide compounds, and more herbicides but less insecticides were found.

The most distributed pesticide was Imazalil which was found in 24.4% of the samples, followed by Triadimenol (21.9%) and Pyrimethanil (17.5%). On the other hand, Chlorpyrifos, Cyanazine, Fenbuconazole, Monocrotophos and Spiroxamine each were found in only one sample. Each of Imazalil, Ethoprophos, Thiofanate-methyl, Pirimiphos-methyl were quantified in 7 food items while 12 compounds were found in only one food item. Twenty-five pesticides were quantified in food items from the 3 locations, 19 were found in 2 locations, and 14 compounds were found in only one location. The highest contaminations found were Cymoxanil in groundnuts from Bafoussam (1.46 mg/kg), Atrazine in Egusi seeds from Douala (1.41 mg/kg), Metribuzin in maize from Douala (1.39 mg/kg) and Pyrimethanil in soybeans from Douala (1.37 mg/kg). But in average, Cymoxanil (0.56 mg/kg), Thiamethoxam (0.34 mg/kg) and Thifensulfuron (0.20 mg/kg) were the compounds found with the highest concentration. In the opposite, Iprodione, Diuron and Fenbuconazole were found at concentrations lower than 0.001 mg/kg.

In general, the monitoring results show many disparities with the findings of our previous work (Galani et al., 2018). Imazalil, Triadimenol and Pyrimethanil are the most distributed pesticide in this study, while in the 2018 study, Imazalil was found only in 1 of the 72 samples, Triadimenol was not detected, and Pyrimethanil was positive in only 4 samples. On the other hand, while Malathion was previously the most found pesticide in the samples (97.2% of positive samples), it was rather less detected (11.3%) in the present work. Cymoxanil, found with the highest concentration here was not detected in any sample during the previous study. Concentration-wise, the mean residue content in this study varied from 0.0006 to 0.6101 mg/kg, while it was between 0.0004 and 0.95 mg/kg in the previous work. Regarding the highest residue content, values between 0.0006 and 1.46 mg/kg were recorded here, while the range was 0.0004-5.52 mg/kg in the previous study. So in general, monitoring results show that the content of pesticides in the food samples is lower than the previous work. These differences can be explained by yearly and seasonal variations in pesticide usage, and the different origins of the samples.

In cowpea samples from markets of Ngaoundere in the northern part of Cameroon, levels of Dichlorvos, Methylparathion, Malathion, Profenofos, Diazinon and Chlorpyrifos ranged from 0.02 to 5.4 mg/kg in peripheral zone, while 0.02 to 4.62 mg/kg of Dichlorvos, Methyl-Parathion, Malathion, Profenofos and Chlorpyrifos were found in the urban area (J. Sonchieu et al., 2013). These values are higher than the corresponding pesticide values found in our work. In another study of pesticide residues in composite foods from Benin, Cameroon, Mali and Nigeria, Chlorpyrifos (22.4%) and Profenofos (5.8%) were among the most detected compounds. In samples of Cameroon, Acetamiprid was detected in 11 samples, with higher concentrations in leafy vegetables from Garoua (0.094-0.241 mg/kg), which is greater than the concentrations obtained in the present study. On the other hand, Acetamiprid was also found in two bean samples from Garoua (0.019-0.023 mg/kg), and tomato samples from Douala (0.014 mg/kg). Pirimiphos methyl was detected in wheat bread and pasta from Bamako (Mali), Douala and Cotonou (Benin) between 0.003 and 0.181 mg/kg). Dimethoate was also quantified in tomatoes from Douala at 0.031 mg/kg (Ingenbleek et al., 2019). All these residue amounts were lower than the values obtained in our study. These differences can be due to many factors, including sampling location, sampling season, food type and food processing.

3.2. Compliance of the quantified pesticides with European limits

Of the 58 quantified pesticides, 30 (51.7%) were above their existing European Union MRLs. These include all the 3 quantified samples with Thiamethoxam (white pepper from

Bafoussam). More than 50% of the positive samples for Hexaconazole, Triazophos, Cymoxanil, Simazine, Fenpropimorf and Linuron were above their MRLs. On the other hand, 22 quantified pesticides with existing MRLs were found below the limits. Additionally, 5 banned pesticides in the country (Carbaryl, Carbofuran, Malathion, Metalaxyl and Propoxur) were found in the samples. More importantly, the proportion of these banned pesticides above their MRLs were as high as 44.4% for Malathion, 33.3% for Metalaxyl, 25.0% for Carbofuran, 14.3% for Carbaryl, but only 9.1% for Propoxur (Table 1). These results are higher than our previous findings, in which only 34.4% of these pesticides were found above their existing MRL values and in 38% of the positive analyses (Galani et al., 2018). However, 75% of samples containing pesticide residues above MRLs were found in maize, cowpea and millet from northern Cameroon (Jean Sonchieu et al., 2010). These can be justified by lack of Good Agricultural Practices (GAP) leading to appropriate applications of pesticides by farmers, because of insufficient training and deficient assistance from agricultural extension agents (Mahob et al., 2014; D. Tarla et al., 2015), hence the necessity of actions to be taken by regulatory authorities to regulate usage of agrochemicals in the country (Galani et al., 2018; J. Sonchieu et al., 2013; D. N. Tarla et al., 2014). Percent of positive samples above MRL for Metalaxyl has increased, from 1.4% in our previous study, to 33.3% here, despite the ban of its importation and usage in 2016 by Cameroonian authority (MINADER, 2016). This suggests that the authority needs to strengthen the controls for effective implementation of the Metalaxyl ban. On the other hand, certain of these banned compounds found in our study were prohibited 7 to 10 years ago: Carbaryl, Malathion and Propoxur in 2008, Carbofuran in 2013 (MINADER, 2013). These pesticides are not persistent molecules like organochlorine and organophosphate pesticides, therefore, presence of their residues in foods today cannot be due to environmental persistence, but may come from illegal use. In fact, stocks of more than 200,000 kg and 300,000 L of obsolete pesticides accumulated over the years were inventoried in the country (D. N. Tarla et al., 2014) and because of their efficacy, cheap price on the black market and limited control by the authority, they are illegally used on crops and produce (Jean Sonchieu et al., 2017). This highlights the necessity of strong control of stock of obsolete pesticides in Cameroon.

3.3. Contamination of the food items with pesticide residues

All the food items were contaminated with pesticides (Table 2). White pepper (12.8% of positive results) was by far the most contaminated food item, followed by maize (5.3%), Egusi seeds (5.3%) and groundnuts (5.0%). Cowpea was the least contaminated item with only 1.5% of the samples tested positive. Similarly, residues of 34 pesticides (42.0%) were found in white pepper samples, 25 residues (30.9%) in maize and 21 (25.9%) in groundnuts, while only 6 compounds (7.4%) were found in coffee samples. Conversely, food items with the highest samples having residues above MRLs were kidney beans (2.0%), groundnuts (1.9%) and maize (1.2%), whereas no compound above its MRL was found in coffee. Ten pesticide compounds above their MRLs were found in groundnuts, and 7 in chilli pepper, kidney beans and maize, each. In the previous study, chili pepper, white pepper, kidney beans and soybeans were the most contaminated samples and in general, the most critical food commodities (with the highest residue concentrations) were kidney beans, soybeans, chili pepper, and maize (Galani et al., 2018).

3.4. Distribution of pesticide residues among the sampling locations

The distribution of contaminated samples among the three cities showed some similarities. In Yaounde and Douala each, 190 (32.5%) analyses were positive, while in Bafoussam, it was 34.9%. However, 40.6% of positive samples in Yaounde and Bafoussam each contained residues above the MRLs, while the value was only in 18.9% in Douala (Figure 1). A recent study found that the contamination of organophosphate residues in composite foods from Cameroon varied with the location (Douala vs Garoua) and with the season of sample collection (rainy vs dry season) (Ingenbleek et al., 2019). These results reflect the differences in the knowledge of farmers on pesticides, and in practices of pesticide usage in the different parts of the country presented in previous reports (Gama et al., 2016; Kenko et al., 2017; Manfo et al., 2012; Nchare, 2007; Jean Sonchieu et al., 2010; Tandi et al., 2014; D. Tarla et al., 2015; Tayoh et al., 2016). These dissimilarities can also be due to the differences in agroecology of these areas, which impose different agricultural constrains that necessitates different patterns of pesticide usage in each area. In fact, Bafoussam is located in western highlands agroecological zone of Cameroon, while Douala is in the humid forest with monomodal rainfall zone, and Yaounde in the humid forest with bimodal rainfall zone. Therefore, farming practices and type and prevalence of pests and diseases differ among these zones. Moreover, farmer's knowledge on pests and diseases, and crop protection practices varied among the different agroecological zones of Cameroon (Okolle, Afari-Sefa, Bidogeza, Tata, & Ngome, 2016; Oyekale, 2018). All these could result in variations in pesticide applications, hence the difference in types and contents of residues found in food samples.

3.5. Dietary chronic risk assessment of individual pesticide residues

In general, there was no significant difference in the lower, medium and upper bound scenarios in the risk assessment analysis (F-test, $p<0.05$). The HQ values ranged between 0.001 and 1.220. The highest HQ values were 1.22 for Carbofuran in groundnuts, followed by 0.71 for Triazophos in maize and 0.64 for Metribuzin in maize. These 3 values were identical under the upper, middle and lower bound scenarios, as they were computed from samples with quantified pesticides. All the remaining values were below 0.38. This suggest that most of the food items can be considered safe for the tested individual pesticide residues for Cameroonian adult consumer. However, Triazophos and Metribuzin in maize are of concern and need to be monitored. Carbofuran is one of the most toxic carbamate pesticides, classified by WHO in the category of highly hazardous insecticides, class Ib (WHO, 2010). It is extremely toxic to humans and animals through cholinesterase inhibition, with reported teratogenic and embryotoxic effects (Saini, Kumar, Hira, & Manna, 2017). Carbofuran was banned in Cameroon seven years ago (MINADER, 2013). The presence of residues of this pesticide in foods from Cameroon at levels above toxicological limits raises the necessity of its regulation in Cameroon.

Our results contrast with the previous findings in Yaounde Cameroon of (Gimou et al., 2008), which reported low dietary exposure and risk to 46 pesticide residues, with mean exposures using the upper bound estimate represent from 0.24% (cypermethrin) to 3.03% (pirimiphos methyl) of the ADI, that is, HQ of 0.0024 to 0.0303. This suggests that for the pesticides considered in these studies, in the last 12 years, the majority of foods consumed in large cities of Cameroon remained safe. However, there is a huge increase of the HQ values, which is a concern. Similar observation were made in other countries. In a total diet study on pesticide residues in France, exposure levels were below the ADI for 90% of the pesticides under the two scenarios. Only dimethoate intakes exceeded the ADI under the lower bound scenario, which tends to underestimate exposure levels. Under the upper bound scenario that overestimates exposure, a chronic risk could not be excluded for dithiocarbamates, ethoprophos, carbofuran, diazinon, methamidophos, disulfoton, dieldrin, endrin and heptachlor (Nougadère et al., 2012). The monitoring and risk assessment of pesticide residues in commercially dried vegetables in Seoul, Korea showed that EDIs ranged from 0.1% of the ADI for bifenthrin to 8.4% of the ADI for cadusafos, suggesting that the detected pesticides could not be considered a serious public health problem (Seo et al., 2013). The HQ values for pesticide residues in green peppers and cucumbers for Turkish population ranged from 0.0003 to 0.0143%, and from 0.0001 to 0.0103%, respectively, suggesting that the consumption of these vegetables is safe (Golge, Hepsag, & Kabak, 2018). A health risk assessment of detected pesticides in apples in Lebanon showed HQ values in the range of 0.1–8% of the ADI's, indicating no risk to human health (El Hawari et al., 2019). It can be seen that as compared to similar studies in other countries, the risk in Cameroon is higher and this should be addressed.

3.6. Cumulative risk assessment of multiple pesticide residues in each food item

The HI values representing risk assessment of cumulative pesticides in each food item are compiled in table 2. All the HI values were below 1, indicating that there may be not health risk for Cameroonian adults consuming these food items. However, HI of groundnuts (0.531) and maize (0.443) suggest that these two food items should be cautiously monitored. In fact, the majority of dishes consumed by urban and rural Cameroonian populations are based on cereals, legumes and tubers, and most of consumed meals are composite dishes, containing many food items analysed in this work (Kouebou et al., 2013; Ponka et al., 2006; Sharma et al., 2007; Sop et al., 2008). In this case and without taking into account the processing factor, the cumulative risk would show that composite meals may not be safe. Similar observations were reported in studies from different countries. In a study of Greek population dietary chronic exposure to pesticide residues in fruits, vegetables and olive oil, it was found that Pyrethrins and Organochlorines (HI value of 0.052 and 0.087, respectively) presented a negligible hazard for the consumers. Organophoshates (HI=0.343) and carbamates (HI=0.389) were not expected to constitute a risk but required further attention. The authors recommended a long term monitoring program from different areas and different times of sampling for more accurate conclusions (Tsakiris, Toutoudaki, Kokkinakis, Paraskevi, & Tsakiris, 2011).

The major contributors of each quantified pesticide residues to the hazard index of each commodity is represented in Figure 2. Compounds with up to 1% contribution were represented individually, while all those with values less than 1% were grouped as ‘’others’’. It can be observed that for the majority of commodities, a large number of compounds (10 to 15) contribute to HI, the highest being 18 pesticides for coffee. While for cocoa, Egusi seeds and groundnuts only 3 to 4 pesticides are the highest contributors. The most common pesticides with the highest percent contribution to HI were Carbofuran (97%, 92%, 91%, 39%, 14%, 11% and 10%), Monocrotophos (42%, 26%, 11% and 11%), Methiocarb (36% and 36%), Ethoprophos (34%), Penconazole (33%), Fenpropimorf (32% and 13%), Pirimiphos-methyl (25%), Cadusafos (31%), Carbaryl (24%), Simazine (18%), Hexaconazole (18%), Triazophos (15%, 11%), Acetamiprid (13%), Oxamyl (12%), Metribuzin (11%), Thiamethoxam (11%) and Dimethoate (10%). In Turkey, it was found that the major contributors to HI for green pepper and cucumber were Propamocarb and Chlorpyrifos, but here also, the HI values showed that there is no reason of concern about cumulative exposure to their residues from these vegetables (Golge et al., 2018).

4. Conclusion

In the last 12 years, this is the largest sampling and the first study of monitoring and risk assessment of pesticide residues in Cameroon. We found 58 compounds in the samples, half of the positive analyses were above their existing EU MRLs and 5 pesticides banned in the country. Imazalil, Triadimenol and Pyrimethanil were the most distributed pesticides in the majority of samples, while Cymoxanil, Thiamethoxam and Thifensulfuron were the compounds with the highest average concentration. Pesticides were present in all the 11 food items, with white pepper, maize, Egusi seeds, and groundnuts showing the highest contamination rates. Except Carbofuran in groundnuts, for an adult Cameroonian, the studied food items could be considered safe for individual pesticide residues, although Triazophos and Metribuzin in maize necessitate further attention. Cumulative risk assessment also showed that the food items could be safe, albeit special attention should be paid on groundnuts and maize. This study underlines the urge for Cameroonian regulatory authorities to monitor the usage of agrochemicals in the country, strengthen the controls for effective implementation of the pesticide bans and implement strong control of obsolete pesticide stocks in Cameroon. These measure are critical not only for minimizing the increasing human health risk for Cameroonian consumers, but also to ensure acceptance of Cameroon export produces on international market.

Acknowledgement

This work was supported by Islamic Development Bank's (IDB) Merit Scholarship for High Technology Programme (Grant number 600035046). The wonderful laboratory assistance of Lilian Goeteyn is gratefully acknowledged.

References

Aktar, W., Sengupta, D., & Chowdhury, A. (2009). Impact of pesticides use in agriculture: Their benefits and hazards. *Interdisciplinary Toxicology*, *2*(1), 1–12. https://doi.org/10.2478/v10102-009-0001-7

EFSA. (2006). Guidance of the scientific committee on a request from EFSA related to uncertainties in dietary exposure assessment. *EFSA Journal*, *438*, 1–54.

EFSA. (2010). Management of left-censored data in dietary exposure assessment of chemical substances. *EFSA Journal*, *8*(3), 1557–1653. https://doi.org/10.2903/j.efsa.2010.1557

El Hawari, K., Mokh, S., Al Iskandarani, M., Halloum, W., & Jaber, F. (2019). Pesticide residues in Lebanese apples and health risk assessment. *Food Additives and Contaminants: Part B Surveillance*, pp. 1–9. https://doi.org/10.1080/19393210.2018.1564370

EPA. (2011). *EPA's National-scale Air Toxics Assessment An Overview of Methods for EPA's National-Scale Air Toxics Assessment*. Durham, North Carolina. Retrieved from https://www.epa.gov/sites/production/files/2015-10/documents/2005-nata-tmd.pdf

European Commission. (2020). EU Pesticides database. Retrieved February 1, 2020, from http://ec.europa.eu/food/plant/pesticides/eu-pesticides-database/public/?event=homepage&language=EN

Gad Alla, S. A., Loutfy, N. M., Shendy, A. H., & Ahmed, M. T. (2015). Hazard index, a tool for a long term risk assessment of pesticide residues in some commodities, a pilot study. *Regulatory Toxicology and Pharmacology*, *73*(3), 985–991. https://doi.org/10.1016/j.yrtph.2015.09.016

Galani, J. H. Y., Houbraken, M., Van Hulle, M., & Spanoghe, P. (2019). Comparison of electrospray and UniSpray, a novel atmospheric pressure ionization interface, for LC-MS/MS analysis of 81 pesticide residues in food and water matrices. *Analytical and Bioanalytical Chemistry*, *411*(20), 5099–5113. https://doi.org/10.1007/s00216-019-

01886-z

Galani, J. H. Y., Houbraken, M., Wumbei, A., Djeugap, F. J., Fotio, D., & Spanoghe, P. (2018). Evaluation of 99 pesticide residues in major agricultural products from the western highlands zone of Cameroon using QuEChERS method extraction and LC-MS/MS and GC-ECD analyses. *Foods*, *7*(11), 184–201. https://doi.org/10.3390/foods7110184

Gama, E. N., Folorunso, S. T., & Adeola, S. S. (2016). Analysis of factors affecting the technical efficiency of cocoa producers in the south-west region of Cameroon. *Journal of Agricultural Research and Development*, *14*(2), 89. https://doi.org/10.4314/jard.v14i2.9

Gimou, M.-M., Charrondiere, U. R., Leblanc, J.-C., & Pouillot, R. (2008). Dietary exposure to pesticide residues in Yaoundé: The Cameroonian total diet study. *Food Additives & Contaminants: Part A*, *25*(4), 458–471. https://doi.org/10.1080/02652030701567475

Golge, O., Hepsag, F., & Kabak, B. (2018). Health risk assessment of selected pesticide residues in green pepper and cucumber. *Food and Chemical Toxicology*, *121*, 51–64. https://doi.org/10.1016/j.fct.2018.08.027

Ingenbleek, L., Hu, R., Pereira, L. L., Paineau, A., Colet, I., Koné, A. Z., … Le Bizec, B. (2019). Sub-Saharan Africa total diet study in Benin, Cameroon, Mali and Nigeria: Pesticides occurrence in foods. *Food Chemistry: X*, *2*(December 2018), 100034. https://doi.org/10.1016/j.fochx.2019.100034

Ingenbleek, L., Jazet, E., Dzossa, A. D., Adebayo, S. B., Ogungbangbe, J., Dansou, S., … Leblanc, J. C. (2017). Methodology design of the regional Sub-Saharan Africa Total Diet Study in Benin, Cameroon, Mali and Nigeria. *Food and Chemical Toxicology*, *109*, 155–169. https://doi.org/10.1016/j.fct.2017.08.017

INS. (2015). *Annuaire Statistique du Cameroun, Edition 2015. Chapitre 14 : Agriculture. Institut National de la Statistique.* Yaounde.pp 232-255. Retrieved from http://www.stat.cm/downloads/2016/annuaire2016/CHAPITRE14_AGRICULTURE.pdf

Kenko, N. D. B., Asanga, B. F. P., Tchamadeu, N. N., & Mpoame, M. (2017). Environmental and human health assessment in relation to pesticide use by local farmers and the Cameroon development corporation (CDC), Fako division, South-West Cameroon. *European Scientific Journal*, *13*(21), 454–473. https://doi.org/10.19044/esj.2017.v13n21p454

Kouebou, C. P., Achu, M., Nzali, S., Chelea, M., Bonglaisin, J., Kamda, A., … Kana Sop, M. M. (2013). A review of composition studies of Cameroon traditional dishes: Macronutrients and minerals. In *Food Chemistry* (Vol. 140, pp. 483–494). https://doi.org/10.1016/j.foodchem.2013.01.003

Mahob, R. J., Hoopen, G. M. T. E. N., Dibog, L., Nyassé, S., Rutherford, M., Mbenoun, M., … Bilong, C. F. B. (2014). Pesticides use in cocoa sector in Cameroon : characterization of supply source , nature of actives ingredients , fashion and reasons for their utilization. *International Journal of Biological and Chemical Sciences*, *8*(5), 1976–1989.

Manfo, F. P. T., Moundipa, P. F., Déchaud, H., Tchana, A. N., Nantia, E. A., Zabot, M.-T., & Pugeat, M. (2012). Effect of agropesticides use on male reproductive function: A study on farmers in Djutitsa (Cameroon). *Environmental Toxicology*, *27*(7), 423–432. https://doi.org/10.1002/tox.20656

Matthews, G., Wiles, T., & Baleguel, P. (2003). A survey of pesticide application in Cameroon. *Crop Protection*, *22*(5), 707–714. https://doi.org/10.1016/S0261-2194(03)00008-5

MINADER. (2013). *Liste des pesticides homologués au Cameroun au 31 juillet 2013. Liste réservée au Grand Public. Commission Nationale d’Homologation des Pesticides à Usage Agricole. Ministère de l’Agriculture et du Développement Rural (MINADER), Cameroun*.

MINADER. (2016). *Décision N°01326/16/D/MINADER/SG/DRCQ/SDRP/SRP portant interdiction de l’importation et de l’homologation des formulations de pesticide à base de métalaxyl au Cameroun. Ministère de l’Agriculture et du Développement Rural (MINADER)*.

Nchare, A. (2007). *Analysis of factors affecting the technical efficiency of arabica coffee producers in Cameroon*. *African Economic Research Consortium* (Vol. 14). Faculty of Agriculture, University of Ilorin. https://doi.org/10.4314/jard.v14i2.9

Nougadère, A., Sirot, V., Kadar, A., Fastier, A., Truchot, E., Vergnet, C., … Leblanc, J. C. (2012). Total diet study on pesticide residues in France: Levels in food as consumed and chronic dietary risk to consumers. *Environment International*, *45*(1), 135–150. https://doi.org/10.1016/j.envint.2012.02.001

Okolle, N. J., Afari-Sefa, V., Bidogeza, J. C., Tata, P. I., & Ngome, F. A. (2016). An evaluation of smallholder farmers’ knowledge, perceptions, choices and gender perspectives in vegetable pests and diseases control practices in the humid tropics of Cameroon. *International Journal of Pest Management*, *62*(3), 165–174. https://doi.org/10.1080/09670874.2016.1184772

Oyekale, A. S. (2018). Cocoa farmers' compliance with safety precautions in spraying agrochemicals and use of personal protective equipment (PPE) in Cameroon. *International Journal of Environmental Research and Public Health*, *15*(2), 327–344. https://doi.org/10.3390/ijerph15020327

Ponka, R., Fokou, E., Fotso, M., Tchouanguep, F. M., Leke, R., Souopgui, J., & Bih, M. A. (2006). Composition of dishes consumed in Cameroon. *International Journal of Food Science and Technology*, *41*(4), 361–365. https://doi.org/10.1111/j.1365-2621.2005.01072.x

Price, C. (2008). Implications of pesticide residues in inter-rated ditchduke farming. *Central Thailand Aquiculture News*, *32*, 23.

Reffstrup, T. K., Larsen, J. C., & Meyer, O. (2010). Risk assessment of mixtures of pesticides. Current approaches and future strategies. *Regulatory Toxicology and Pharmacology*, *56*(2), 174–192. https://doi.org/10.1016/J.YRTPH.2009.09.013

Saini, R., Kumar, P., Hira, S. K., & Manna, P. P. (2017). Evaluation of carbofuran-mediated toxicity against human lymphocytes and red blood cells in simulated wastewater degraded by coagulation–flocculation. *Environmental Science and Pollution Research*, *24*(18), 15315–15324. https://doi.org/10.1007/s11356-017-9098-y

Seo, Y. H., Cho, T. H., Hong, C. K., Kim, M. S., Cho, S. J., Park, W. H., … Kim, M. S. (2013). Monitoring and risk assessment of pesticide residues in commercially dried vegetables. *Preventive Nutrition and Food Science*, *18*(2), 145–149. https://doi.org/10.3746/pnf.2013.18.2.145

Sharma, S., Mbanya, J. C., Cruickshank, K., Cade, J., Tanya, A. K. N., Cao, X., … Wong, M. R. K. M. (2007). Nutritional composition of commonly consumed composite dishes from the Central Province of Cameroon. *International Journal of Food Sciences and Nutrition*, *58*(6), 475–485. https://doi.org/10.1080/09637480701288454

Sonchieu, J., Ngassoum, M. B., Tchatchueng, J. B., Srivastava, A. K., & Srivastava, L. P. (2013). Contamination of cowpea and by-products by organophosphorous pesticide residues in

Ngaoundere markets: Dietary risk estimation and degradation study. *African Journal of Food Science*, *7*(5), 92–102. https://doi.org/10.5897/AJFS2013.1013

Sonchieu, Jean, Benoit Ngassoum, M., Bosco Tchatchueng, J., Srivastava, A. K., & Srivastava, L. P. (2010). Survey of pesticide residues in maize, cowpea and millet from northern Cameroon: part I. *Food Additives and Contaminants: Part B*, *3*(3), 178–184. https://doi.org/10.1080/19393210.2010.503329

Sonchieu, Jean, Fointama, E., Akono, N. E., & Serri, B. (2019). Farmers' knowledge, attitude and practices on pesticide safety: a case study of vegetable farmers in Mount-Bamboutos agricultural area, Cameroon. *Agricultural Sciences*, *10*(08), 1039–1055. https://doi.org/10.4236/as.2019.108079

Sonchieu, Jean, Srivastava, A. K., Ngassoum, B. M., Tchatchueng, J. B., & Srivastava, L. P. (2017). Contamination of cowpea (*Vigna unguiculata* (L.) Walp) and derived products by residues of banned pesticides in Cameroon: health risk estimation. *Journal of Pesticide*, *104*, 147–160. Retrieved from https://sites.google.com/site/internationalindexing/home/journal-of-pesticide

Sop, M. M. K., Gouado, I., Mofor, C. T., Smriga, M., Fotso, M., & Ekoe, T. (2008). Mineral content in some Cameroonian household foods eaten in Douala. *African Journal of Biotechnology*, *7*(17), 3085–3091. https://doi.org/10.5897/AJB08.126

Tandi, T. E., Wook, C. J., Shendeh, T. T., Eko, E. A., & Afoh, C. O. (2014). Small-scale tomato cultivators' perception on pesticides usage and practices in Buea Cameroon. *Health*, *06*(21), 2945–2958. https://doi.org/10.4236/health.2014.621333

Tarla, D., Manu, I. N., Tamedjouong, Z. T., Kamga, A., & Fontem, D. A. (2015). Plight of pesticide applicators in Cameroon: case of tomato (*Lycopersicon esculentum* Mill.) farmers in Foumbot. *Journal of Agriculture and Environmental Sciences*, *4*(2), 87–98. https://doi.org/10.15640/jaes.v4n2a10

Tarla, D. N., Tchamba, N. M., Baleguel, N. P., Fontem, D. A., Baleguel, P. D., & Hans, D. (2014). Inventory of obsolete pesticide stockpiles in Cameroon. *Scholarly Journal of Agricultural Science*, *4*(1), 43–50.

Tayoh, L. N., Kiyo, M. L. I., & Nkemnyi, M. F. (2016). Chemical fertilizer application and farmers perception on food safety in Buea, Cameroon. *Agricultural Science Research*

Journal, *6*(12), 287–295.

Tsakiris, N. I., Toutoudaki, M., Kokkinakis, M., Paraskevi, M., & Tsakiris, M. A. (2011). A risk assessment study of Greek population dietary chronic exposure to pesticide residues in fruits, vegetables and olive oil. In Margarita Stoytcheva (Ed.), *Pesticides - Formulations, Effects, Fate* (pp. 253–268). eBook (PDF) ISBN: 978-953-51-4524-0: IntechOpen. https://doi.org/10.5772/14065

Walpole, S. C., Prieto-Merino, D., Edwards, P., Cleland, J., Stevens, G., & Roberts, I. (2012). The weight of nations: An estimation of adult human biomass. *BMC Public Health*, *12*(1), 439. https://doi.org/10.1186/1471-2458-12-439

WHO. (2010). *The WHO recommended classification of pesticides by hazard and guidelines to classification 2009*. (WHO, Ed.), *World Health Organization*. Geneva, Switzerland: Press, WHO. Retrieved from https://apps.who.int/iris/bitstream/handle/10665/44271/9789241547963_eng.pdf

Winter, C. K. (2015). Chronic dietary exposure to pesticide residues in the United States. *International Journal of Food Contamination*, *2*(1), 11. https://doi.org/10.1186/s40550-015-0018-y

Table 1. Distribution of 58 quantified pesticide residues in 11 food items from the 3 largest cities of Cameroon.

Sr. No.	Analyte	Application	Banned?	Number of positive samples	Percent of positive samples (%)	Number of positive food items	Number of positive locations	Lowest content (mg/kg)	Highest content (mg/kg)	Mean content (mg/kg)	Median content (mg/kg)	Number of samples > MRLs	Percent of positive samples > MRLs (%)
1	2,4-D	Herbicide	No	4	2.5	2	2	0.0004	0.0029	0.0016	0.0016	0	0.0
2	Acetamiprid	Insecticide	No	12	7.5	4	2	0.0041	0.1916	0.0471	0.0111	4	33.3
3	Ametryn	Herbicide	No	13	8.1	6	3	0.0013	0.0218	0.0087	0.0076	NA	NA
4	Atrazine	Herbicide	No	7	4.4	5	3	0.0078	1.4160	0.2180	0.0177	0	0.0
5	Azoxystrobin	Fungicide	No	7	4.4	3	2	0.0084	0.7164	0.1614	0.0749	0	0.0
6	Bentazon	Herbicide	No	13	8.1	5	3	0.0051	0.0734	0.0290	0.0225	1	7.7
7	Boscalid	Fungicide	No	7	4.4	3	3	0.0090	0.0614	0.0313	0.0143	0	0.0
8	Butachlor	Herbicide	No	8	5.0	2	1	0.0012	0.2870	0.1020	0.0525	NA	NA
9	Cadusafos	Insecticide/ Nematicide	No	7	4.4	2	2	0.0062	0.0451	0.0210	0.0179	3	42.9
10	Carbaryl	Insecticide	Yes	7	4.4	3	2	0.0064	0.0471	0.0189	0.0078	1	14.3
11	Carbendazim	Fungicide	No	13	8.1	4	3	0.0021	0.4090	0.0623	0.0224	1	7.7
12	Carbofuran	Insecticide	Yes	12	7.5	5	3	0.0017	0.3562	0.0966	0.0122	3	25.0
13	Chlorotoluron	Herbicide	No	10	6.3	3	2	0.0066	0.1622	0.0404	0.0104	3	30.0
14	Chlorpyrifos	Insecticide	No	1	0.6	1	1	0.0016	0.0016	0.0016	0.0016	0	0.0
15	Cyanazine	Herbicide	No	1	0.6	1	1	0.0076	0.0076	0.0076	0.0076	0	0.0
16	Cymoxanil	Fungicide	No	8	5.0	2	3	0.0006	1.4617	0.6101	0.5675	5	62.5
17	Diuron	Herbicide	No	8	5.0	2	3	0.0009	0.4280	0.0633	0.0010	2	25.0
18	Epoxiconazole	Fungicide	No	17	10.6	6	3	0.0034	0.1830	0.0328	0.0142	3	17.6
19	Ethoprophos	Insecticide/ Nematicide	No	25	15.6	7	3	0.0009	0.0127	0.0051	0.0056	0	0.0
20	Fenamiphos	Nematicide	No	8	5.0	2	3	0.0006	0.0036	0.0019	0.0019	0	0.0
21	Fenbuconazole	Fungicide	No	1	0.6	1	1	0.0006	0.0006	0.0006	0.0006	0	0.0
22	Fenpropimorf	Fungicide	No	17	10.6	6	3	0.0014	0.1228	0.0308	0.0115	9	52.9
23	Hexaconazole	Fungicide	No	7	4.4	3	2	0.0160	0.1955	0.0561	0.0210	5	71.4

24	Hexythiazox	Acaricide	No	7	4.4	3	2	0.0020	0.0171	0.0091	0.0077	0	0.0
25	Imazalil	Fungicide	No	39	24.4	7	3	0.0004	0.0856	0.0139	0.0021	3	7.7
26	Imidacloprid	Insecticide	No	4	2.5	1	1	0.0181	0.0231	0.0210	0.0214	0	0.0
27	Iprodione	Fungicide	No	7	4.4	2	2	0.0005	0.0479	0.0076	0.0010	1	14.3
28	Linuron	Herbicide	No	2	1.3	1	1	0.0085	0.0504	0.0295	0.0295	1	50.0
29	Malathion	Insecticide	Yes	18	11.3	5	3	0.0027	0.0739	0.0285	0.0185	8	44.4
30	Metalaxyl	Fungicide	Yes	15	9.4	5	3	0.0031	0.0497	0.0181	0.0093	5	33.3
31	Methiocarb	Acaricide /Insecticide	No	3	1.9	1	1	0.0064	0.0070	0.0068	0.0069	0	0.0
32	Metribuzin	Herbicide	No	9	5.6	4	3	0.0004	1.3898	0.1604	0.0044	1	11.1
33	Metsulfuron-methyl	Herbicide	No	9	5.6	2	2	0.0009	0.0184	0.0086	0.0080	3	33.3
34	Monocrotophos	Acaricide	No	1	0.6	1	1	0.0430	0.0430	0.0430	0.0430	0	0.0
35	Oxamyl	Insecticide/ Nematicide	No	5	3.1	1	1	0.0131	0.0378	0.0207	0.0151	0	0.0
36	Penconazole	Fungicide	No	6	3.8	2	1	0.0055	0.0184	0.0096	0.0083	0	0.0
37	Pendimethalin	Herbicide	No	3	1.9	1	1	0.0089	0.0136	0.0114	0.0116	0	0.0
38	Pirimicarb	Insecticide	No	3	1.9	2	2	0.0013	0.0073	0.0036	0.0024	0	0.0
39	Pirimiphos-methyl	Insecticide	No	20	12.5	7	3	0.0016	0.5745	0.0557	0.0082	4	20.0
40	Prochloraz	Fungicide	No	5	3.1	2	2	0.0025	0.0238	0.0074	0.0030	0	0.0
41	Propanil	Herbicide	No	13	8.1	4	3	0.0078	0.0423	0.0216	0.0212	2	15.4
42	Propazine	Herbicide	No	7	4.4	1	2	0.0041	0.1728	0.0811	0.0716	NA	NA
43	Propiconazole	Fungicide	No	7	4.4	2	2	0.0012	0.1609	0.0542	0.0048	3	42.9
44	Propoxur	Insecticide	Yes	11	6.9	3	3	0.0056	0.0523	0.0220	0.0177	1	9.1
45	Pyrimethanil	Fungicide	No	28	17.5	6	3	0.0005	1.3716	0.0632	0.0058	4	14.3
46	Simazine	Herbicide	No	13	8.1	4	3	0.0024	0.1171	0.0359	0.0257	7	53.8
47	Spiroxamine	Fungicide	No	1	0.6	1	1	0.0351	0.0351	0.0351	0.0351	0	0.0
48	Tebuconazole	Fungicide	No	7	4.4	2	2	0.0032	0.0979	0.0314	0.0116	2	28.6
49	Tebufenozide	Insecticide	No	5	3.1	2	2	0.0063	0.0168	0.0125	0.0119	0	0.0
50	Tebuthiuron	Herbicide	No	8	5.0	3	2	0.0010	0.0117	0.0040	0.0028	NA	NA
51	Terbuthryn	Herbicide	No	15	9.4	3	3	0.0011	0.0372	0.0090	0.0043	NA	NA
52	Terbuthylazine	Herbicide	No	17	10.6	4	3	0.0009	0.0263	0.0072	0.0032	0	0.0
53	Thiacloprid	Insecticide	No	11	6.9	2	2	0.0017	0.0066	0.0031	0.0025	0	0.0

54	Thiamethoxam	Insecticide	No	3	1.9	1	1	0.3408	0.4374	0.3730	0.3409	3	100.0
55	Thifensulfuron	Herbicide	No	6	3.8	2	2	0.1019	0.2960	0.1983	0.2069	NA	NA
56	Thiofanate-methyl	Fungicide	No	22	13.8	7	3	0.0004	0.0689	0.0168	0.0025	5	22.7
57	Triadimenol	Fungicide	No	35	21.9	5	3	0.0005	0.0271	0.0059	0.0017	9	25.7
58	Triazophos	Acaricide/ Nematicide	No	6	3.8	3	1	0.0111	0.1192	0.0328	0.0154	4	66.7

NA: Not Applicable, because of no existing MRLs.

Table 2. Distribution of pesticide residues and hazard index of 11 food items from the 3 largest cities of Cameroon.

Sr. No.	Food item	Number of samples	Number of analyses	Positive analyses number	Positive analyses rate (%)	Quantified pesticides number	Quantified pesticides rate (%)	Analyses above MRLs number	Analyses above MRLs rate (%)	Pesticides above MRLs number	Pesticides above MRLs rate (%)	Hazard index
1	Black beans	15	1215	29	2.4	13	16.0	4	0.3	3	3.7	0.021
2	Chili pepper	15	1215	48	4.0	17	21.0	12	1.0	7	8.6	0.002
3	Cocoa	15	1215	29	2.4	11	13.6	4	0.3	3	3.7	0.007
4	Coffee	10	810	16	2.0	6	7.4	0	0.0	0	0.0	<0.001
5	Cowpea	15	1215	18	1.5	7	8.6	4	0.3	2	2.5	0.001
6	Egusi seeds	15	1215	65	5.3	16	19.8	NA	NA	NA	NA	0.008
7	Groundnuts	15	1215	61	5.0	21	25.9	23	1.9	10	12.3	0.531
8	Kidney beans	15	1215	47	3.9	16	19.8	24	2.0	7	8.6	0.063
9	Maize	15	1215	65	5.3	25	30.9	14	1.2	7	8.6	0.443
10	Soybeans	15	1215	50	4.1	13	16.0	12	1.0	4	4.9	<0.001
11	White pepper	15	1215	156	12.8	34	42.0	9	0.7	4	4.9	0.001

NA: Not Applicable, because of no existing MRLs.

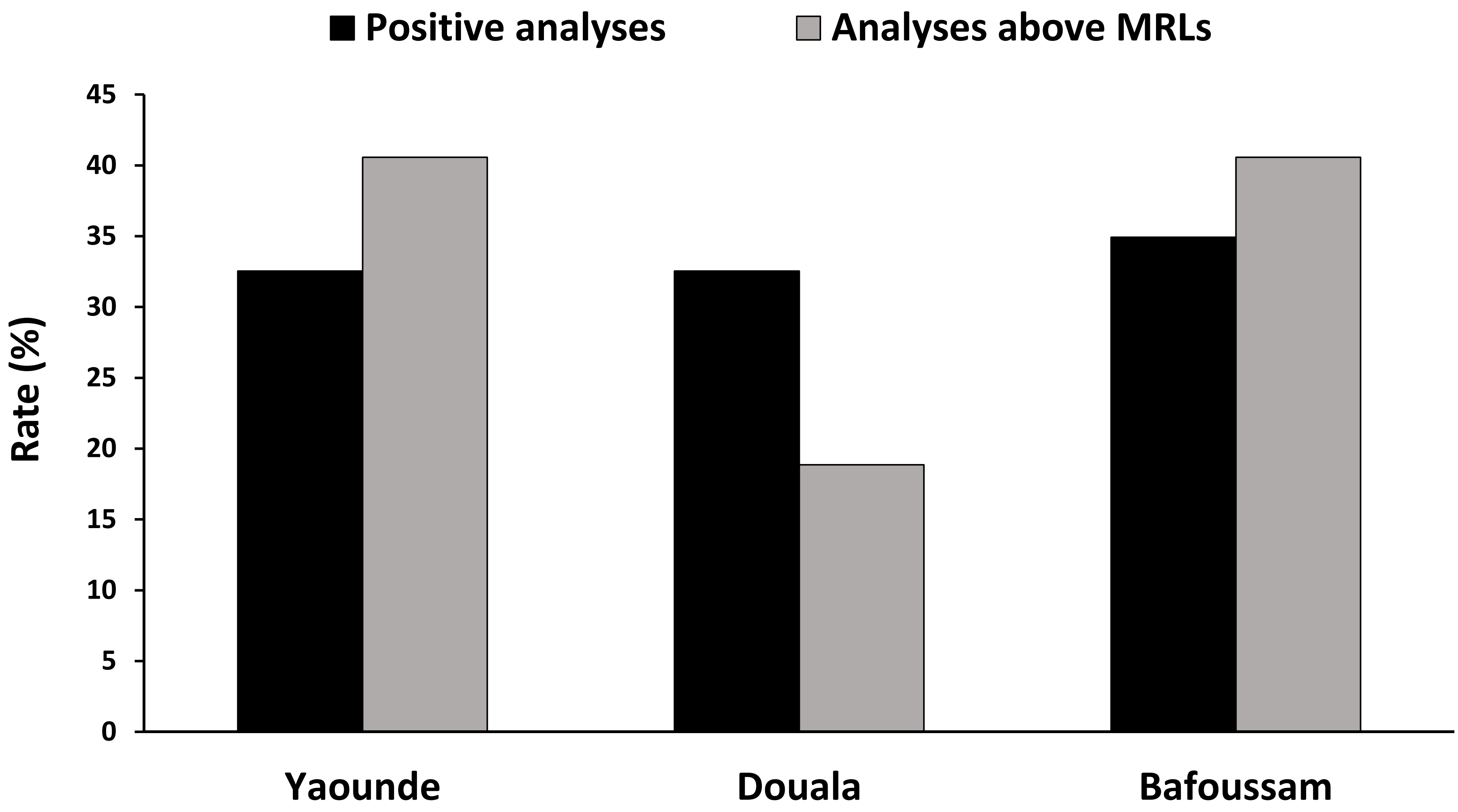
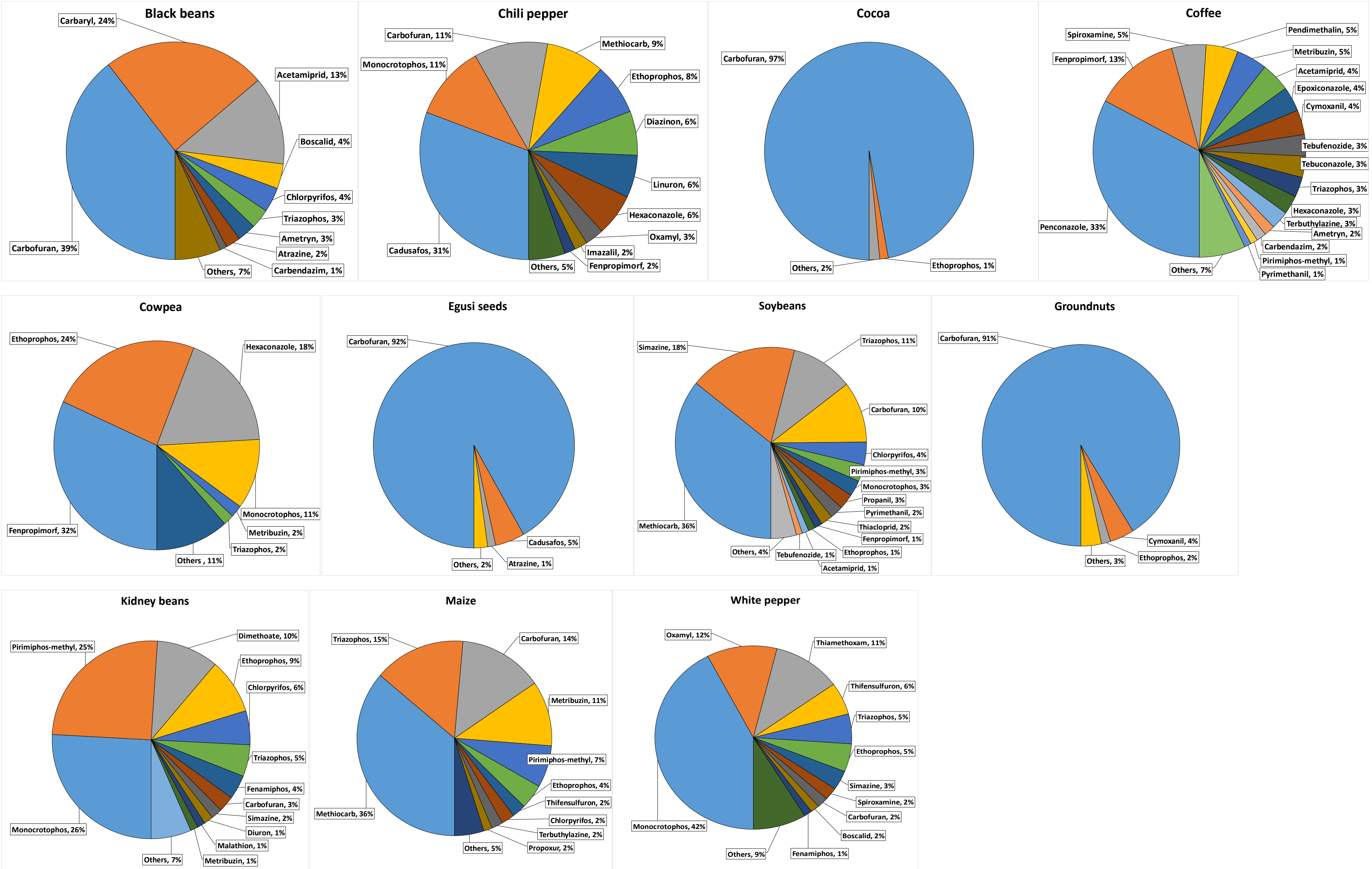


Figure 1. Distribution of quantified pesticide residues among the 3 sampling cities in Cameroon.



Black beans
Carbaryl, 24%
Acetamiprid, 13%
Boscalid, 4%
Chlorpyrifos, 4%
Triazophos, 3%
Ametryn, 3%
Atrazine, 2%
Carbendazim, 1%
Others, 7%
Carbofuran, 39%
Chili pepper
Carbofuran, 11%
Monocrotophos, 11%
Methiocarb, 9%
Ethoprophos, 8%
Diazinon, 6%
Linuron, 6%
Hexaconazole, 6%
Oxamyl, 3%
Imazalil, 2%
Fenpropimorf, 2%
Others, 5%
Cadusafos, 31%
Cocoa
Carbofuran, 97%
Others, 2%
Ethoprophos, 1%
Coffee
Spiroxamine, 5%
Fenpropimorf, 13%
Pendimethalin, 5%
Metribuzin, 5%
Acetamiprid, 4%
Epoxiconazole, 4%
Cymoxanil, 4%
Tebufenozide, 3%
Tebuconazole, 3%
Triazophos, 3%
Hexaconazole, 3%
Terbuthylazine, 3%
Ametryn, 2%
Carbendazim, 2%
Pirimiphos-methyl, 1%
Pyrimethanil, 1%
Others, 7%
Penconazole, 33%
Cowpea
Ethoprophos, 24%
Hexaconazole, 18%
Monocrotophos, 11%
Metribuzin, 2%
Triazophos, 2%
Others , 11%
Fenpropimorf, 32%
Egusi seeds
Carbofuran, 92%
Cadusafos, 5%
Atrazine, 1%
Others, 2%
Soybeans
Simazine, 18%
Triazophos, 11%
Carbofuran, 10%
Chlorpyrifos, 4%
Pirimiphos-methyl, 3%
Monocrotophos, 3%
Propanil, 3%
Pyrimethanil, 2%
Thiacloprid, 2%
Fenpropimorf, 1%
Ethoprophos, 1%
Acetamiprid, 1%
Tebufenozide, 1%
Others, 4%
Methiocarb, 36%
Groundnuts
Carbofuran, 91%
Cymoxanil, 4%
Ethoprophos, 2%
Others, 3%
Kidney beans
Pirimiphos-methyl, 25%
Dimethoate, 10%
Ethoprophos, 9%
Chlorpyrifos, 6%
Triazophos, 5%
Fenamiphos, 4%
Carbofuran, 3%
Simazine, 2%
Diuron, 1%
Malathion, 1%
Metribuzin, 1%
Others, 7%
Monocrotophos, 26%
Maize
Triazophos, 15%
Carbofuran, 14%
Metribuzin, 11%
Pirimiphos-methyl, 7%
Ethoprophos, 4%
Thifensulfuron, 2%
Chlorpyrifos, 2%
Terbuthylazine, 2%
Propoxur, 2%
Others, 5%
Methiocarb, 36%
White pepper
Oxamyl, 12%
Thiamethoxam, 11%
Thifensulfuron, 6%
Triazophos, 5%
Ethoprophos, 5%
Simazine, 3%
Spiroxamine, 2%
Carbofuran, 2%
Boscalid, 2%
Fenamiphos, 1%
Others, 9%
Monocrotophos, 42%

Figure 2. Relative contribution of pesticide residues to the hazard index 11 food items from the 3 largest cities of Cameroon.