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Dietary risk assessment of pesticides from vegetables and drinking water in gardening areas in Burkina Faso

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HIGHLIGHTS

- Developed multiresidue analysis successfully quantified 31 pesticides in vegetables.
- 16 different active ingredients have been found in vegetables and water samples.
- Dietary surveys allowed to derive local consumption estimates of staples food.
- Dietary intake of pesticides presented acute and chronic risks for children and adults.
- Chronic risks from one endocrine disruptor and probable carcinogen were detected.

GRAPHICAL ABSTRACT

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ABSTRACT

Vegetables and water samples have been collected around the lake of Loumbila in Burkina Faso. Pesticides residues in food commodities were analyzed using a modified QuEChERS extraction method prior analysis on GC-MS and UPLC-MS/MS of 31 pesticides. Maximum Residue Limits (MRLs) were exceeded in 36% of the samples for seven pesticides: acetamiprid, carbofuran, chlorpyrifos, lambda-cyhalothrin, dieldrin, imidacloprid and profenofos. Exceedance of MRLs suggests a risk for the consumers and limits the opportunities of exportation. In order to define estimated daily intake, dietary surveys were conducted on 126 gardeners using a 24 hours recall method. Single pesticide and cumulative exposure risks were assessed for children and adults. Risk was identified for: chlorpyrifos and lambda-cyhalothrin in acute and chronic exposure scenarios. Hazardous chronic exposure to the endocrine disruptor and probable carcinogen dieldrin was also detected. In the studied population, cumulative dietary exposure presented a risk (acute and chronic) for children and adults in respectively >17% and 4% of the cases when considering the worst case scenarios. Processing factor largely influenced the risk of occurrence suggesting that simple washing of vegetables with water considerably reduced the risk of hazardous exposure.
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1. Introduction

Pesticides are widely applied in agriculture for pests' control to improve yield. Previous studies and field surveys have underlined the lack of knowledge regarding good agricultural practices and the use of unsuitable and obsolete pesticides in Burkina Faso (Ouédraogo et al.,

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2011). Improper selection and use of pesticides on foodstuffs can result in undesirable levels of residues even after processing (Kaushik et al., 2009; Keikotlhaile et al., 2010; Reiler et al., 2015). Although, accidental intake, self-harm and occupational exposure are considered to be the major routes of exposure to pesticides in Burkina Faso (Toe, 2010), dietary exposure is assumed to be five orders of magnitude higher than other routes, such as air and drinking water (Jolliet et al., 2003). Hence pesticide residues in food might also constitute an important risk to human's health. To prevent health hazard and unnecessary exposure, Maximum Residue Limits (MRLs) and Admissible Daily Intake (ADI) have been implemented at national levels and internationally for example in the Codex Alimentarius (WHO/FAO, 2017) or in the European Pesticides database (European Union, 2017). Although pesticide residues contamination in foodstuffs have been monitored for decades in most developed countries, vegetables in developing countries are not much investigated for pesticide contamination (Bempah et al., 2016). Studies conducted on food produced in West Africa have identified exceedance of MRLs and ADI suggesting a risk for consumers and the need for suitable monitoring and controls of food products (Bempah et al., 2011; Mawussi et al., 2009).

In Burkina Faso, vegetables are mainly grown during the dry season (January–June). In order to guaranty continuous access to water, gardening areas are located on lakes' coasts. Pesticides application, washing of clothes and equipment used for spraying can lead to contamination of this resource used for drinking water. Three sources of drinking water have been identified during field surveys: surface water (lake), traditional wells and boreholes (Lehmann et al., 2017). Traditional wells consist of hand dug wells located in the middle of the fields. Their primary function is to minimize the distance to access water for watering but they are also used by the gardeners for drinking water. Boreholes water is consumed at household level while surface and wells water is generally consumed during work directly on the field. Surface water and traditional wells can be considered as unsafe water sources because of the proximity with cultivated crops, the submersion by the lake during the rainy season and the absence of protection measures (buffer zone, structure, cover, etc.).

The present study assessed dietary exposure to pesticide from drinking water and consumption of vegetables produced in larger quantities in gardening areas in Burkina Faso (i.e.: tomatoes, cucumber, sorrel (*Hibiscus sabdariffa*), okra (*Abelmoschus esculentus*) and two varieties of eggplant (*Solanum melongena* L. and *Solanum aethiopicum*)).The prerequisite for exposure assessment from food is the characterization of the population diet. Various methods exist which include diet history, diet recall, food frequency questionnaires, etc. (FAO/WHO, 2009). The majority of dietary studies use national consumption estimates which might not be fully representative of local trends. In this study, questionnaire surveys have been conducted on the field using the modified 24 hours recall method (24 HR) proposed by Gibson and Ferguson (1999). 24 HR method has the advantage to be faster, less-invasive and easier for both the investigator and the respondent than traditional food frequency or weighed food records. Collected data was aggregated to derive vegetables and water consumption. Acute and chronic risk assessments were subsequently performed by comparison of single pesticide and cumulative exposure to Acute Reference Dose (ARfD) and ADI.

Pesticides used in gardening are mainly neonicotinoids, pyrethroids and organophosphates. In parallel to the present study, a three years survey (2014–2016) directed toward gardeners, local resellers and health care centers (~600 persons) has been conducted in four gardening areas in Burkina Faso (Loumbila, Dem, Nariarlé basin, and Ziga). The questionnaires aimed at defining the local agricultural practices (pesticides used, application rate, spraying equipment, etc.). Target substances (i.e. 31 substances analyzed) have been retained based on their presence on the field and their identification in previous studies (Bassole and Ouédraogo, 2007; Gomgnimbou et al., 2009; Ouédraogo et al., 2011; Toe, 2010). Multiresidue extraction procedure has been developed and applied to determine pesticides levels in vegetables.

This study proposes a comprehensive assessment of the dietary intake of pesticides from vegetables and water sources in a gardening area of a Sahelian country and the resulting risk for children and adults.

2. Material and method

2.1. Site description

The study was conducted in March–April 2015 and 2016 in 3 villages Pousghin, Nabdogo, and Noungou located on the shores of the lake Loumbila (Supplementary Information (SI) Fig. S1). Loumbila Reservoir has a long historic in gardening as one off the pioneers in this domain. It is located in one of the most intensive gardening area in Burkina Faso and supplies 1/3 of the capital drinking water. Growing interest has been observed in the past decade for preservation of this valuable resource with pesticides being one of the greater concerns.

2.2. Field investigations

2.2.1. Dietary survey and consumption data

In order to assess daily consumption of the gardening commodities, a dietary survey was conducted on 126 persons using the modified 24 hours recall method (24 HR) proposed by Gibson and Ferguson (1999). Surveys were conducted during nine consecutive days in order to cover every weekdays and any special events (i.e. market day, weekend, etc.). Local kitchen utensils were used to help respondents and investigators in quantity assessment. This approach has been developed for application in rural areas in developing countries and has proved to reduce bias and memory lapses from the respondents. Attendants were randomly selected on the field or at household level (SI Fig. S1). Priority was given to women as in rural areas they are usually in charge of cooking. Individual portions have been directly derived from respondents' answers (i.e. individual diet). Single 24 HR can be also used to derive average intake of a group with the assumption that the subjects are representative of the study population (Gibson, 2005). The difference was made between products consumed raw and cooked (labeled as sauces) considering different recipes and their composition. It was noted that vegetables portions could vary between recipes and that some items were not consumed daily. As an example, when eaten raw, the full vegetable unit (ex: tomato, *Solanum aethiopicum*, etc.) is generally consumed and a larger portion is ingested at once. Thus giving items a similar weight may lead to an overestimation of the average intake. This issue was handled by Carriquiry (2003) by introducing the propensity-to-consume items. Nevertheless, a single 24 HR does not provide sufficient data to apply the statistical approaches proposed. Therefore, the propensity-to-consume items was derived from their frequency of reporting in dietary surveys, assuming subjects' representativeness of the studied population. Data aggregation allowed to derive weighted average portion estimates (WAPE) for the whole studied population. Distinction has been made between drinking water sources for average water consumption estimation. Boreholes and lake water data were aggregated while traditional wells were considered apart.

WAPE calculation included occurrence of the studied commodities in local recipes and occurrence of the final dishes in the diet of the study population. For a given food commodity/water source i, WAPE was calculated as follow:

$$WAPE_i = \sum_{1}^{n} \overline{F}_n Pu_n Pp_n \quad (1)$$

where, $\overline{F}_n$ is the average portion of a given food commodity in a recipe/volume of water from a source n (g/pers), Pu_n the probability that the food commodity is used in the given recipe ($Pu_n = 1$ for water consumption estimation) and Pp_n the probability of occurrence of the given recipe/use of the water source in the studied population diet.

A worthy issue is that data collected from field surveys always present gaps. They have been attributed to the willingness to answer of the respondent and the understanding of the translator and the surveyor. Only data allowing estimation of individual portions were considered in WAPE's calculations and further risk assessment ($n = 70$). For other calculations, the full dataset was considered ($n = 126$).

2.2.2. Sampling procedure of gardening commodities and water

The gardening areas cultivated around Loumbila Lake reach up to 3.47 km^2. Studied vegetable species were selected based on their respective surfaces of cultivated land and the likeliness of pesticide treatment before harvest. Gardeners reported that onion, carrot and garlic were generally less subject to pesticide treatment as the edible part grows under the soil surface. Based on these criteria: tomato, cucumber, sorrel (*Hibiscus sabdariffa*), okra (*Abelmoschus esculentus*) and two varieties of eggplant (*Solanum melongena* L. and *Solanum aethiopicum*) have been retained as target species for the present study.

In rural areas in Burkina Faso, diet have found to be generally poor and monotonous (Savy et al., 2007). Nevertheless, seasonality and particularly food shortage periods might influence dietary diversity (Mathilde Savy et al., 2006). Fresh vegetables are expected to present the higher levels of pesticide residues. As the objective of the present research was to study pesticides exposure from vegetables consumption, growing/harvesting period was retained as the more appropriate. Vegetables samples were collected by simple random sampling directly on the plot in the three villages located on the lake shores and on local market stalls in March–April 2015 and 2016.

Laboratory samples size was defined according to the European Commision Directive 2002/63/EC (2002) in order to obtain representative samples to determine compliance with MRLs for pesticides. Samples collected on the field were wrapped in aluminum foil and placed in opaque plastic bags. They were stored at 4 °C for transportation and −20 °C at the laboratory for conservation. In total, 59 samples of vegetables were collected (i.e. tomato: $n = 17$, cucumber: $n = 11$, sorrel: $n = 10$, okra: $n = 7$, *Solanum melongena L.*: $n = 8$ and *Solanum aethiopicum*: $n = 6$). For each collected sample, the gardener responsible of the plot was asked to answer a questionnaire about his own agricultural practices (pesticides used, application rate, water used, etc.).

During the three years monitoring study presented elsewhere (Lehmann et al., 2017), 70 surface water samples have been collected around the lake and more precisely near water inputs and areas with the highest gardening activity. In addition, 27 traditional wells and 9 boreholes were randomly sampled in the same areas where food samples have been collected.

Sampling points' location for food and water analysis is presented in Supplementary information section S1.

2.3. Chemical analysis

2.3.1. QuEChERS extraction

Pesticides residues in food commodities were extracted using a modified AOAC 2007.01 QuEChERS (Quick Easy Cheap Rugged and Safe) extraction method. Composite samples were chopped into small pieces, and mixed. Ten grams of homogenized sample were added to a 50 ml centrifuge tube and an isotopic dilution was performed with the addition of 0.2 ml of labeled surrogates' solution (SI Table S2). Solvent was allowed to evaporate prior addition of 10 ml of 1% acetic acid (HOAc) in acetonitrile and extraction for 5 min in an ultrasonic bath. QuEChERS methods have been designed for matrix with at least 75% water content (Correia-Sá et al., 2012). Water addition was needed for okra (5 ml) and sorrel (10 ml). Then, 4 g $MgSO_4$ and 1 g NaOAc were added and the mixture was shaken vigorously for 1 min, vortexed for 1 min and centrifuged for 5 min at 3000 rpm. As the reaction with $MgSO_4$ is exothermic, the tubes where cooled in a water bath at room temperature. 6 ml of the supernatant were subsequently transferred for clean-up in a 12 ml dispersive SPE (dSPE) tube packed with 420 mg of Supel™QuE *Z*-Sep/C18 sorbent (Sigma-Aldrich, Switzerland) and vortexed for 1 min. After 5 min centrifugation at 4000 rpm, 4 ml of the supernatant were concentrated to 0.4 ml. An aliquot of 0.2 ml was evaporated to dryness and reconstituted in 0.2 ml of the mixture methanol:water (5:95,v/v) with 0.1% formic acid prior UPLC-MS/MS analysis. The remaining 0.2 ml were evaporated to dryness and reconstituted in 0.2 ml of isooctane prior GC-MS analysis.

All samples were unprocessed vegetables directly collected on the plot or on market stalls. In traditional cuisine, cucumber but also *Solanum melongena L* are peeled before being consumed. Therefore, these vegetables were peeled prior homogenization and the skin and edible fraction were analyzed separately.

2.3.2. Water analysis

Details about the solid phase extraction procedure of water samples have been presented elsewhere (Lehmann et al., 2017). Briefly, water was filtered through 0.7 μm glass fiber filters (GF/F Whatman; Florham Park, NJ) prior to spiking with appropriate labeled surrogates and extraction. A 1-l water sample was pumped through a 200 mg water Oasis HLB cartridge preconditioned with 10 ml ethyl acetate, 10 ml methanol and 5 ml water. The cartridge was subsequently eluted with the following fractions 5 ml methanol, 5 ml methanol: ethyl acetate (1:1) and 5 ml ethyl acetate: hexane (1:4). After combination, the fractions were concentrated prior separation and analysis on GC-MS and UPLC-MS/MS.

2.3.3. Apparatus and chemicals

Standards of analytes and deuterated compounds were purchased from Sigma-Aldrich (Switzerland), Dr. Ehrenstorfer (Germany), and Toronto Research Chemicals (Canada). Individual solutions of each analyte and deuterated compound and their dilution were prepared in appropriate solvent prior preparation of the stock solutions respectively in acetone and methanol (Tables S1 and S2) and stored at −20 °C. Appropriate dilutions of these standards solutions were used to prepare calibration curves for further analysis on GC-MS and UPLC-MS/MS.

Ethyl acetate and methanol HPLC grade were acquired from Carlo Erba Reagents (France), formic acid from Sigma-Aldrich (Switzerland), acetone for residues analysis from Acros Organics (Belgium) and acetonitrile and n-hexane from Biosolve Chimie SARL (France). Anhydrous magnesium sulfate and sodium acetate (NaOAc) were purchased from Sigma-Aldrich, (Switzerland).

The gas chromatography analyses were performed on a Thermo Scientific Trace 1310 gas chromatograph coupled with a Thermo Scientific ISQ Single Quadrupole MS (Waltham, MA, USA). The UPLC system consisted of a UPLC Waters Acquity coupled to a Acquity Xevo TQ-S tandem quadrupole MS. Operating parameters are detailed in supplementary information (SI Section S2).

2.3.4. Quality control and quality assurance

In order to evaluate the efficiency of the analytical procedure a recovery assay was conducted. Blank samples of tomato, cucumber, eggplant (*Solanum melongena* L.) and okra were spiked in triplicates at ~10 μg/kg and ~50 μg/kg. Over the 31 pesticides analyzed, 25 presented recovery rates in the range of 47%–155% for the four vegetables species. The others substances (i.e. 6 pesticides) presented lower recovery rates for certain vegetable species. Nevertheless, they were kept in the multiresidue analysis due to low variability of the obtained results (i.e. low relative standard deviation between replicates). Detailed multiresidue extraction recoveries of studied commodities are presented in supplementary information (Table S7). The limit of detection (LOD) and limit of quantification (LOQ) for selected target analytes have been defined as the analyte concentration that produced a peak with a signal-to-noise ratio of respectively 3 and 10 (Table 1). They have been determined experimentally by measuring the coincident instrumental response of standard pesticide solutions and procedural blank or negative samples.

Table 1
LOD and LOQ for target pesticides in vegetables.

Active ingredient	LOD [μg/kg]	LOQ [μg/kg]	Active ingredient	LOD [μg/kg]	LOQ [μg/kg]
Carbamate			***Organochlorine***		
Carbofuran	0.05	0.17	alpha-Chlordane	0.07	0.24
			gamma-Chlordane	0.06	0.2
Neonicotinoid			Dieldrin	5.89	19.64
Acetamiprid	0.03	0.09	alpha-Endosulfan	4.50	15
Imidacloprid	0.05	0.17	beta-Endosulfan	5.17	17.24
			Endosulfan Sulfate	6.36	21.19
Pyrethroid			Endrin	5.87	19.57
Lambda-Cyhalothrin	2	6.68	alpha-HCH	15.00	50
alpha-Cypermethrin	6.74	22.45	gamma-HCH	15.00	50
beta-Cypermethrin	7	23.35	alpha-Heptachlor epoxyde	3.00	10
Deltamethrin	3.26	10.86	beta-Heptachlor epoxyde	3.00	10
			Hexachlorobenzene	3.00	10
Tetranortriterpenoid			trans-Nonachlor	0.07	0.24
Azadirachtin	4.11	13.7			
			Organophosphate		
Triazine			Chlorpyrifos	11.19	37.3
Atrazine	0.04	0.14	Chlorpyrifos-methyl	15.4	51.32
Atrazine desethyl	0.01	0.04	Diazinon	2.43	8.11
Atrazine desisopropyl	0.05	0.17	Omethoate	0.06	0.21
			Profenofos	2.1	7
Urea			Triazophos	0.02	0.06
Diuron	2.5	8.35			

2.4. MRL compliance and risk assessment

Compliance with MRLs was evaluated as the ratio of pesticide residues measured in or on food commodities and MRL values. As the entire foodstuff must be considered (Reg. EC No 396/2005, 2005), the sum of levels measured in edible fraction and the skin was considered when they have been separately analyzed.

The ARfD and ADI were used as predicted no effect levels for acute and chronic consumer's exposures respectively (Reiler et al., 2015). The Estimated Daily Intake (EDI) of a given pesticide was derived from Renwick (2002) and is expressed here as:

$$EDI_{pest} = \frac{\sum (Cpest \times Fc \times Fp)}{bw} \quad (2)$$

where *Cpest* is the concentration of a given pesticide residue on food, *Fc* is the food consumption, *Fp* the food processing factor and *bw* the body weight. For each detected pesticide, hazard quotient (HQ) defined as the ratio of pesticide intake to ARfD or ADI were then use to derive the resulting risk. A HQ exceeding the unity (>100% of ARfD or ADI) indicates a risk.

$$HQ_{acute} = \frac{EDI}{ARfD} \quad (3)$$

$$HQ_{chronic} = \frac{EDI}{ADI} \quad (4)$$

A number of methods have been developed for cumulative risk assessment of pesticides in food. Cumulative effects of pesticides were subsequently evaluated using Hazard Index (HI) presented in previous studies (Boobis et al., 2008). When more than one residue are present, HQ of pesticides with common mode of action were summed to account for cumulative toxicity.

$$HI = \sum_i^n HQ_i \quad (5)$$

MRL, ARfD and ADI values have been extracted from the EU – Pesticides database (European Union, 2017). HQ and HI have been calculated for WAPE and individual diets considering median and maximal residue levels on commodities for acute risk assessment. As it supposed to represent a life time exposure, chronic hazard was calculated for WAPE and individual diets considering only median residue levels on commodities. The adult body weight is estimated to be 53 kg for women with at least one child under 5 years old (Savy et al., 2006). For children aged 11–16 years, WHO (2011) proposed the parametric value of 32 kg for pesticide exposure assessment. Comparison with other studies suggested that these values complied respectively with average body weight of adults and children in rural areas of Burkina Faso (Wood, 2000). Considered scenarios for risk assessment are presented in Table 2.

3. Results

3.1. Pesticides residues and MRL compliance

Over the 31 pesticides analyzed in this study, 16 were detected in food or water samples (>LOD). Median and maximum concentrations in edible fractions (peeled vegetables) and drinking water used for risk evaluation are presented in Table 3.

Table 2
Scenarios considered for dietary risk assessment.

Risk	Reference dose	Population	Body weight	Scenario name	Food consumption	Residue level
Acute risk	ARfD	Children	32	CH_AR_1	WAPE	Median
				CH_AR_2	WAPE	Maximum
				CH_AR_3	Individual diet	Median
				CH_AR_4	Individual diet	Maximum
		Adult	53	A_AR_1	WAPE	Median
				A_AR_2	WAPE	Maximum
				A_AR_3	Individual diet	Median
				A_AR_4	Individual diet	Maximum
Chronic risk	ADI	Children	32	CH_CR_1	WAPE	Median
				CH_CR_2	Individual diet	Median
		Adult	53	A_CR_1	WAPE	Median
				A_CR_2	Individual diet	Median

Table 3
Levels of pesticides in vegetables and water samples (*N.D.*: Not detected, <LOQ: LOD < residue level < LOQ).

Pesticides	Authorized by the CSP[a]		Tomatoes	Sorrel	*Solanum melongena L*	*Solanum aethiopicum*	Okra	Cucumber	Lake	Well
			[µg/kg]						[µg/l]	
Acetamiprid	Yes	Median Maximum	1.52 25.14	109.31 3055.39	1.85 1.85	0.44 1.67	3.31 145.08	1.01 2.73	0.0018 0.0302	0.0066 0.042
∑ *Atrazine*[b]	No	Median Maximum	*<LOQ*	0.29 0.29	*<LOQ*	*N.D.*	*<LOQ*	*N.D.*	0.0237 0.4942	0.0032 0.0347
Azadirachtin	No	Median Maximum	*<LOQ*	*N.D.*	*N.D.*	*N.D.*	*N.D.*	*N.D.*	0.1126 0.1126	0.2956 0.4879
Carbofuran	No	Median Maximum	1.13 1.13	*N.D.*	0.47 0.47	2.16 3.5	*N.D.*	0.29 0.29	0.1097 0.1097	0.0106 0.0106
Chlorpyrifos	Yes	Median Maximum	90.6 667.45	100.82 590.48	*N.D.*	*N.D.*	*<LOQ*	*<LOQ*	0.0653 0.0858	0.0448 0.2022
λ-Cyhalothrin	Yes	Median Maximum	145.84 174.34	50.33 1661.77	*<LOQ*	40.92 41.63	292.48 330.71	9.59 10.38	*N.D.*	0.0294 0.0294
∑ *Cypermethrin*[c]	Yes	Median Maximum	77.13 77.13	123.78 631.28	*<LOQ*	*N.D.*	184.31 184.31	*N.D.*	0.2197 0.8392	*N.D.*
Dieldrin	No	Median Maximum	*<LOQ*	*<LOQ*	297.31 571.61	*N.D.*	*N.D.*	*<LOQ*	0.1069 0.1069	*N.D.*
alpha-Endosulfan	No	Median Maximum	*N.D.*	26.2 36.8	*N.D.*	*N.D.*	*N.D.*	*N.D.*	*N.D.*	*N.D.*
Imidacloprid	Yes	Median Maximum	5.92 152.65	41.17 159.45	7.87 39.21	20.45 100.62	52.39 193.97	0.28 0.46	0.0039 0.2355	0.0096 0.384
Omethoate	No	Median Maximum	*N.D.*	3.74 7.09	*N.D.*	*N.D.*	*N.D.*	*N.D.*	*N.D.*	*N.D.*
Profenofos	Yes	Median Maximum	14.22 73.62	618.63 2999	*N.D.*	5.04 5.04	132.96 208.76	*3.18* *3.18*	*N.D.*	0.049 0.1742
Triazophos	No	Median Maximum	0.49 0.49	0.52 0.96	*<LOQ*	*N.D.*	*N.D.*	*N.D.*	0.0007 0.0225	0.0018 0.0018

[a] Active ingredient authorized in specified commercial formulations by the Sahelian Pesticides Committee (CSP) for application in gardening.
[b] Sum of atrazine and its metabolites desethylatrazine and deisopropylatrazine.
[c] Sum of isomers: alpha-Cypermethrin and beta-Cypermethrin.

13 target pesticides have been detected in water. It is noteworthy that for some samples, threshold limits proposed by the European Directive 98/83/EC (1998) on the quality of water intended for human consumption (i.e. 0.1 µg/l for single pesticide and 0.5 µg/l for the sum of pesticides) were exceeded (Lehmann et al., 2017).

Residues from 14 different pesticides have been quantified on food commodities. MRL compliance was verified for each sample and pesticide using the appropriate limit value. Over 59 vegetable samples 21 exceeded the MRLs for seven pesticides: acetamiprid ($n = 1$), carbofuran ($n = 1$), chlorpyrifos ($n = 3$), cyhalothrin lambda ($n = 5$), dieldrin ($n = 6$), imidacloprid ($n = 4$) and profenofos ($n = 11$). Percentage of MRLs exceedance range from 100% to 2.99 10^4% (see details in SI Section S4.).

3.2. Consumption data

The majority of the respondents reported taking three meals per day (76%). Extreme values (1 or 4 meals) were always associated with a particular event (sickness, lack of money, celebration day, etc.). If most of the meals were home-made (~60%), a large fraction (~35%) was taken in local restaurants (named kiosk or maquis). Every respondent affirmed consuming at least one of the studied vegetables each day. Tomatoes, sorrel and okra were the preponderant studied commodities in the local diet. Tô is a traditional porridge usually made with millet. Over a total of 45 traditional dishes, Tô with vegetables sauce (okra or sorrel) was the most common dish (30%) followed by rice served also with vegetables sauce (16%). Raw vegetables such as cucumber (3.6%), *Solanum aethiopicum* (~3%) and tomato (2%) were also reported as part of the diet (SI Fig. S2).

After harvest, food generally undergoes processing steps until it becomes the final commodities. Processing has been proved to influence residual pesticide levels in food (Kaushik et al., 2009; Keikotlhaile et al., 2010; Liang et al., 2014). In the study area, vegetables undergone no particular process before being cooked at household level. Hence, local recipes were used to define the suitable processing factor in Eq. (2). Cucumber and eggplant (*S.melongena L*) were usually peeled. In certain cases sun dried okra was used for sauce preparation. Vegetables were usually washed with water before being consumed except when eaten raw directly on the field during harvest. In the local diet, vegetables are boiled in 89% of the dishes and fried in 11%.

Effect of peeling on pesticides residues has been assessed for cucumber and eggplant (*S.melongena L*). Results are presented in Table 4. Average of the residual amounts are presented with no distinction between commodities due to low number of samples. No processing factor has been applied for peeling in risk assessment as only pesticides levels in edible fraction have been considered.

Table 4
Percentage of the total amount of pesticides (i.e. with skin) remaining in cucumber and eggplant (*S. melongena L*) after peeling (data not sufficient to derive impact of processing on profenofos).

	Mean	n	SD	Min	Max
Acetamiprid	35%	10	11%	21%	55%
Atrazine	0%	1	–	–	–
Carbofuran	53%	2	–	53%	100%
lambda-Cyhalothrin	16%	2	–	11%	20%
Dieldrin	57%	1	–	–	–
Imidacloprid	18%	8	12%	1%	40%

Impact of processing on pesticide residues levels was proved to be highly dependent on pesticide, crop and process combination (Kaushik et al., 2009). Since data were only available for a limited number of these combinations of concern in the study area, pesticide-generic food processing factor of 0.6 was applied as proxy. This value was considered as conservative (protective) as the sum of all processes undergone (i.e. drying, washing, boiling and frying) are expected to reduce pesticide levels to a larger extent (Keikotlhaile et al., 2010; Liang et al., 2014).

24HR surveys were subsequently used to derived consumed portions for each respondent (not presented) and WAPE using Eq. (1) (SI Section S5). Meals were generally prepared by one person for many (ex: family, restaurant, etc.). When the meal was shared, equal repartition between participants was assumed with no distinction between adults and children. When water origin was not associated with wells or boreholes, it was considered that it originated from the lake. The average water consumption from the lake and traditional wells were respectively 1.74 l/pers/day and 2.29 l/pers/day.

3.3. Dietary risk assessment

3.3.1. Acute risk

For children and adults, WAPE exposure yielded no acute single pesticide nor cumulative exposure risk neither for median nor maximum residues levels (CH_AR_1&2 and A_AR_1&2). Same results were obtained when considering individual dietary exposures and median pesticides residues levels (CH_AR_3 and A_AR_3).

Acute risk was identified only in individual diets in worth case scenarios (CH_AR_4 and A_AR_4). In these scenarios, HQ_{acute} exceeded the unity for chlorpyrifos and lambda-cyhalothrin (Fig. 1). Dietary exposure of children presented a risk related to chlorpyrifos concentrations in 7 diets and lambda-cyhalothrin in 4 diets (i.e. 16% of the studied population). Chlorpyrifos hazard (HQ > 1) was found to be linked to consumption of raw tomatoes directly on the field. For adults, dietary risk was identified in only one diet for chlorpyrifos as well as for lambda-cyhalothrin (i.e. 3% of the studied population).

The cumulative risk of the organophosphates & carbamates and pyrethroids groups indicated a risk for children for respectively 9 (plus 1 value close to unity with 0.95) and 4 individual dietary exposure (Fig. 1). For the same pesticides groups, the HI_{acute} indicated a risk in respectively 3 and 1 diets for adults. HI_{acute} was also close to the unity in 3 diets (0.97, 0.98 and 0.98) for the organophosphates & carbamates pesticides. Except for these 3 diets, all the population presented a $HI_{acute} < 0.85$. In their respective group, chlorpyrifos alone accounted for at least 70% and up to 94% and lambda-Cyhalothrin ~99% of these HI_{acute} values. For children and adults, consumption of raw products were responsible of the observed risks in respectively 54% ($n = 7$) and 75% ($n = 3$) of the cases.

3.3.2. Chronic risk

No chronic risk was observed when considering exposure from WAPE for children and adults (CH_CR_1 and A_CR_1). On the other hand, individual dietary intake exhibited a chronic risk for organophosphorus, organochlorine and pyrethroids pesticides for children (CH_CR_2). Hazard quotients related to chlorpyrifos exposure exceeded the unity for 6 individual diets (i.e. 16% of the studied population). lambda-Cyhalothrin and dieldrin intake showed both a chronic risk in respectively 2 and 3 individual diets. Organophosphates and carbamates: ($n = 6$, plus one diets with $HI_{chronic} = 0.98$), organochlorines ($n = 3$) and pyrethroids ($n = 3$) groups presented all chronic risks for cumulative exposure for children (Fig. 2). Adults presented a chronic risk (A_CR_2) related only to dieldrin exposure in 3 diets (i.e. 4% of

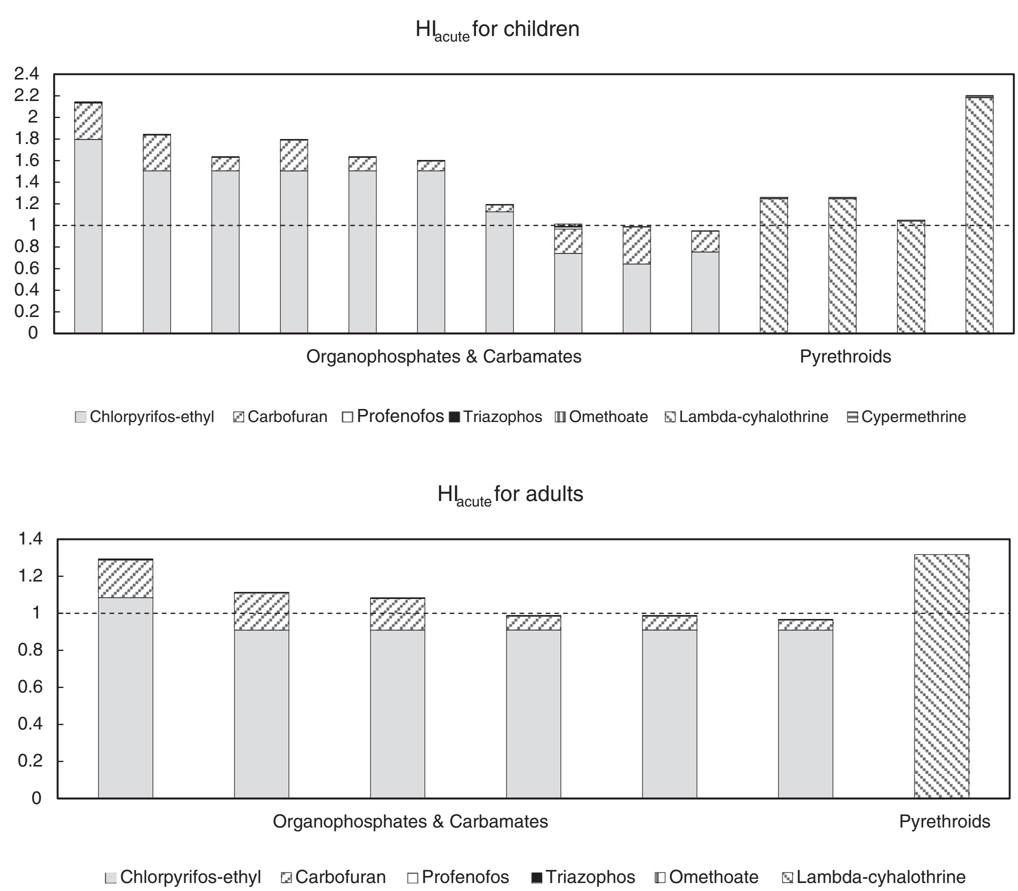

Fig. 1. Acute hazard indexes exceeding/close to the unity calculated for children and adults based on individual diets and maximum pesticides residues levels (CH_AR_4 and A_AR_4).

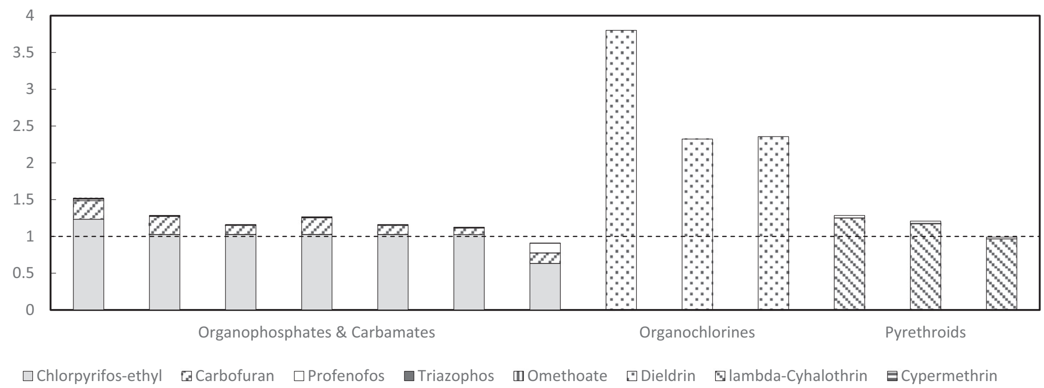

Fig. 2. Chronic hazard indexes exceeding/close to the unity calculated for children based on individual diets and median pesticides residues levels (CH_CR_2).

the studied population). Cumulative exposure did not show supplementary risk. The rest of the population presented a $HI_{chronic}<0.93$.

4. Discussion

Carbamate, neonicotinoids, organochlorines, organophosphates, pyrethroids, triazines and tetranortriterpenoid have been detected. All the vegetable samples had at least one detectable pesticide residue. Every commodity presented an average of 3 pesticide residues except sorrel which exhibited the higher contamination with an average of 6. Higher contamination of leafy vegetables was also observed in Ghanaian markets by Osei-Fosu et al. (2014).

In the present study, 36% of samples did not comply with MRLs. These findings underlined the lack of knowledge regarding Good Agricultural Practices (GAP) and pesticides use already observed in previous studies (Ouédraogo et al., 2011). Poor correlation between pesticides reported by gardeners during surveys and residues detected in samples illustrated this problematic highly linked with illiteracy and low level of education in rural areas. On the other hand, MRLs exceedance could result in prejudicial economic limitations. In Burkina Faso, the fruit and vegetable sector has been retained as a leading sector in the government strategy for rural development. The growing demand of vegetables in the developed countries is associated with reinforced controls. Exports of horticultural products from developing nations have already been rejected by international markets because of residual levels of pesticides (Bempah et al., 2011).

24HR surveys successfully provided consumption estimates of staple food items. Comparison with GEMS (Global Environment Monitoring System) Food Clusters proposed by WHO (2013) underlined the pertinence of local dietary surveys. As an example, the water consumption estimation of 0.4 g/day proposed for Burkina Faso (cluster G13) was surprisingly low. This value did not comply with field observations. Under the warm temperatures of the dry season, surveyed individuals were found to consume at least 3 times more than this prediction. GEMS vegetables/legumes consumption estimate (78.9 g/d) compared favorably with average processed items consumption (80 g/d) but less when consumption of raw products was included (220 g/d). Eating raw vegetables while working on the field is a common practice in the study area (reported by ~38% of the respondents).

Food consumption estimations combined with pesticides residues levels have allowed to derive dietary intake of target substances. Dietary risk was identified for organophosphates & carbamates, organochlorine and pyrethroids groups. Risk for lambda-cyhalothrin was associated with higher consumption of sorrel and exposure to dieldrin was induced by detection of this pesticide on eggplant (*Solanum melongena L*). Chlorpyrifos hazard was mainly linked to higher consumption of tomatoes.

Chlorpyrifos and lambda-cyhalothrin exposure exhibited an acute risk in single pesticide risk assessment (HQ > 1) for both children and adults (CH_AR_4 & A_AR_4). Unsuitable use of pesticides can induce hazardous residues levels on food even after processing. Time before harvest, mixture concentration and frequency of application recommendations were not respected and could vary greatly. Hazardous exposure of children to chlorpyrifos was found to be linked to raw tomatoes consumption in 70% of the cases. For lambda-cyhalothrin sorrel consumption was associated to hazardous exposure. Even though hazards were associated with maximal concentrations, the fact that consumption of a single commodity exceeds the ARfD for a given pesticide suggests that acute intoxication is likely to occur. Thus worst case scenario should not be underestimated.

Chronic risks were identified for dietary exposure to organophosphates, organochlorines and pyrethroids. Long-term exposure covers average daily exposure over the entire life-time. The detected risks are particularly of concerns as dieldrin and chlorpyrifos have been recognized as endocrine disruptor chemicals (WHO/UNEP, 2012) and dieldrin is also a probable human carcinogen (U.S. EPA, 1988). To date, lambda-cyhalothrin has not been classified as endocrine disruptor but endocrine-mediated mode of action could not be ruled out. It was also included in the list of candidates for substitution in Europe knowing that this substance was more toxic than those of the majority of the approved active substances within the group of insecticides (European Union, 2017). In the study area, the potential health burden suggests also that substitution must be undertaken.

Exposure to pesticide mixtures in cumulative risk assessment (HI) was associated with larger intake of pesticides with the same mode of action thus resulting in higher hazard for the consumer. Nevertheless it is worth noting that only chlorpyrifos, lambda-cyhalothrin and dieldrin presented a HQ close/exceeding the unity. Other pesticides were found in concentrations yielding smaller HQ values (lower pesticides concentrations detected or lower consumption of concerned commodities). Chlorpyrifos, lambda-cyhalothrin and dieldrin do not share the same mode of action thus they are not considered in the same cumulative assessment group (CAG). Under these conditions HIs which correspond to the sum of HQs from the same CAG will not yield a value significantly different from the HQs of these three pesticides. Nevertheless, for individual diets presenting HQs close to the unity, summing by similar CAQ yielded HIs exceeding the threshold value of one. Joint use of several trademarks alone or in combination for a single plot was reported by 50% of the gardeners in the study area. Multiresidue analysis was found to be a robust tool in the present context as screening of a large list of target substances allowed to detect residues of pesticides not identified by investigators nor gardeners during field surveys. As aforementioned lack of knowledge and illiteracy but also poor labeling quality (i.e. written in foreign language, absence, etc.) and counterfeiting could have led to misinterpretations.

Acute risks were associated with maximum levels of pesticides and higher consumption of a given type of monitored food. In chronic risk assessment, only median pesticide residues levels were considered thus the difference relied also on serving size. Every individual does not necessarily consume a given item each day and in similar proportions. For this reason deterministic approach or average portions based on a single 24 HR might underestimate or overestimate individual dietary intake. To overcome this shortage, the assumption was made that between-persons variation was representative of the propensity-to-consume a given item in the population. This assumption is supported by the fact that every respondent presented a similar socio-economic background, lived in the same area and global diet in rural areas is generally poor and monotonous (Savy et al., 2003). Calculated WAPE might be therefore closer to the usual dietary intake. In absence of extreme intake values, these average estimates yielded no acute nor chronic risk. Replicated 24 HR and food frequency questionnaire (FFQ) could be used in further studies to validate the estimation of propensity-to-consume items and refined the presented assessment.

The type of pesticide used was also of concerns. Over the 16 pesticides detected, 7 were not authorized in gardening (Table 3) by the Sahelian Pesticide Committee (CSP) among which only one was authorized for cotton production (azadirachtin). Thus 6 pesticides were not authorized in the CILSS (Permanent Interstates Committee for Drought Control in the Sahel) Member States with endosulfan and dieldrin being also banned on an international level. Burkina Faso has ratified the Stockholm convention which ban the use and production of these persistent organic pollutants (entry into force March 2005). Production, selling and use of pesticides initially included in annex A of the Stockholm convention (i.e. aldrine, chlordane, dieldrine, endrine, heptachlore, mirex and toxaphene) are prohibited since 1996 in Burkina Faso. Though, national inventories on persistent organic pesticides conducted in 2001 and 2004 reported uses of wood protection products containing aldrin and dieldrin across the country (Ministère de l'Environnement et du Cadre de Vie, 2007). The prohibition of endosulfan is more recent as entered into force since October 2012 (Stockholm Covention on Persistent Organic Pollutants, 2017). Endosulfan was detected only in sorrel samples collected in 2016 which could suggest a recent use or its confinement in one area (Noungou village sampled only in 2016). Trademarks containing endosulfan such as *Rambo*, *Endocoton 500* and Caïman *Rouge* have been identified during field surveys and must be removed from the market as soon as possible. It is also noteworthy that contaminations might also originate from other activities (ex: cotton or cereals production). For example, because of the associated costs, the use of atrazine in small-scale gardening is not common. None of the respondent reported using herbicides. The presence of atrazine on vegetables might be a consequence of the contamination of the lake water by other activities located upstream (Lehmann et al., 2017). Moreover, atrazine was detected on sorrel which is a leafy vegetable. Application of an herbicide directly on this culture would result in the death of the plant which suggests that it was not intentionally applied. The findings of this study underline the lack of incentive to comply with law and that monitoring on regular basis would help policy, health and environmental impact assessment.

Most of the risks came from the consumption of raw products (larger consumption of a product in a single portion). Processing factor of 1 was assigned to this practice as most of the vegetables were peaked on the plant and directly consumed. Risk assessment estimates are supportive that applying a processing factor of 0.6 would considerably reduce the risk for organophosphates and pyrethroids. Studies have shown that this reduction could be achieved simply by washing vegetables with water (Liang et al., 2014). A similar conclusion was made when changing processing factor of the other processed food items (i.e. fixed value of 0.6) to 1. The risks detected under these conditions suggested that cleaning vegetables with water before eating them considerably reduced health hazard. Further refinements could include the definition of specific processing factors based on local processes and pesticides used.

Except for atrazine and carbofuran, dietary exposure from vegetables was higher than water consumption. It is also noteworthy that the present study focused only on pesticides exposure resulting from consumption of the major vegetables cultivated in the study area. Local diets also include other cultivated commodities subject to pesticide applications (i,e. maize, red pepper, coffee, cowpea, onion, carrot, etc.) that can increase the daily intake of chemicals (Mekonen et al., 2014). Veterinary treatment of cattle and exposure of fishes in contaminated environment have also been studied in West Africa and suggest possible dietary risks (Adakal et al., 2013; Lawrence et al., 2015). Finally, 91% of the respondents were gardeners, therefore occupational exposure will also add to the dietary intake yielding a larger health hazard.

5. Conclusion

The developed multiresidue analysis using QuEChERS extraction method allowed to successfully quantify 31 target substances in selected vegetables. Residues from 16 different active ingredients were found in food and water samples. MRLs and ADI exceedance are in accordance with previous studies conducted in West Africa (Mawussi et al., 2009). Nevertheless, comprehensive monitoring programs are still lacking and to our knowledge the present study constitutes a premiere in Burkina Faso. In rural areas of this country, diet has found to be generally poor and monotonous (Savy et al., 2003). Based on this assumption, the present study could be seen as a preliminary assessment of dietary exposure to pesticide trough vegetables in rural areas of Burkina Faso. In the studied population, the worst case scenarios of dietary cumulative exposure of children and adults presented an acute risk in respectively 19% ($n = 13$) and 6% ($n = 4$) of the cases (CH_AR_4 & A_AR_4). These estimates fell at 17% ($n = 13$) and 4% ($n = 3$) when considering chronic risk (CH_CR_2 & A_CR_2). Precautions must be taken, to reduce dietary exposure especially for children. These include regulation and recommendations enforcement at every scales, from national policy application to the respect of good agricultural practices on the field. More incentive on law application and training of the operators are prerequisites to improve consumer's safety.

Declaration of interest

All authors declare having no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.05.285.

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