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Assessment of average exposure to organochlorine pesticides in southern Togo from water, maize (*Zea mays*) and cowpea (*Vigna unguiculata*)

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Drinking water, cowpea and maize grains were sampled in some potentially exposed agro-ecological areas in Togo and analysed for their contamination by some common organochlorine pesticides. A total of 19 organochlorine pesticides were investigated in ten subsamples of maize, ten subsamples of cowpea and nine subsamples of drinking water. Analytical methods included solvent extraction of the pesticide residues and their subsequent quantification using gas chromatography-mass spectrometry (GC/MS). Estimated daily intakes (EDIs) of pesticides were also determined. Pesticides residues in drinking water (0.04–0.40 µg1⁻¹) were higher than the maximum residue limit (MRL) ($0.03 µg1^{-1}$) set by the World Health Organization (WHO). Dieldrin, endrin, heptachlor epoxide and endosulfan levels ($13.16-98.79 µg kg^{-1}$) in cowpea grains exceeded MRLs applied in France ($10-50 µg kg^{-1}$). Contaminants' levels in maize grains ($0.53-65.70 µg kg^{-1}$) were below the MRLs ($20-100 µg kg^{-1}$) set by the Food and Agriculture Organization (FAO) and the WHO. EDIs of the tested pesticides ranged from 0.02% to 162.07% of the acceptable daily intakes (ADIs). Population exposure levels of dieldrin and heptachlor epoxide were higher than the FAO/WHO standards. A comprehensive national monitoring programme on organochlorine pesticides should be undertaken to include such other relevant sources like meat, fish, eggs and milk.

Keywords: drinking water; maize (*Zea mays*); cowpea (*Vigna unguiculata*); organochlorine pesticides residues; estimated daily intake (EDI)

Introduction

The Stockholm Convention on persistent organic pollutants (POPs) (Programme des Nations Unies pour l'Environnement (PNUE) 2001) was adopted in May 2001 by the international community as a response to the urgent need to reduce or eliminate sources and releases of these hazardous chemicals in order to protect human health and the environment from their adverse effects. Togo is party to the Stockholm Convention and has elaborated a national plan for the implementation of this treaty. The drawing of the national environmental contamination baseline is among others required in view of the evaluation of the effectiveness of the POPs reduction or elimination measures adopted by the country.

POPs include organochlorine pesticides like DDT, aldrin, dieldrin, endrin, heptachlor, which have been widely used to control agricultural pests or disease vectors in Togo. Production and use of most POPs pesticides are banned in many countries throughout the world. Togo has taken a recent regulation to integrate this in the national law (Kolani et al. 2003). POPs have been largely replaced by less persistent molecules as organophosphates, carbamates and pyrethroids. However, endosulfan, a persistent toxic substance, not yet listed in the POPs Convention, is still authorized in Togo for agricultural use in coffee (*Coffea* sp.) and cotton (*Gossypium* sp.) production. This practice leads to its illegal use by poor farmers for cowpea (*Vigna unguiculata*) production. In addition, crop rotation between cotton and maize (*Zea mays*), potentially exposes maize grains to POPs pesticides contamination (Schroll et al. 1994; Fismes et al. 2002).

Organochlorine pesticides resist chemical and biological degradation (Megharaj et al. 1997; Gevao et al. 2000), consequently they bioaccumulate in the food chain (Bernhoft et al. 1997; Ruus et al. 1999; Senthilkumar et al. 1999; Borga et al. 2001; Fisk et al. 2001). Humans may be exposed by the oral route to pesticides through both food and drinking water (Adeyeye and Osibanjo 1999; Badach et al. 2000; Harris et al. 2001; Chun and Kang 2003). Chronic low exposure levels to bioaccumulative pesticide residues can result in various severe pathologies or adverse effects including cancer, diabetes, endocrine

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disruption, reproductive failure or developmental effects (Fry 1995; Romieu et al. 2000; Chandra et al. 2001; Picard et al. 2003). The level of human exposure is related to the daily intake of contaminated foods and drinks, associated with the frequency of consumption of these products (Dagher et al. 1999). The population exposure risk could be high in Togo where staple foods like maize and cowpea grains are assumed to be potentially contaminated.

Economically, maize and cowpea have an important value in Togo. Their production is widely distributed throughout the country. In addition, rural populations depend on well and river water for drinking and other domestic water uses. The proof of significant contamination of drinking water and miscellaneous foodstuff in Togo by POPs pesticides was established by earlier work (Djaneyé-Boundjou et al. 2000). However, limited information is available on the health risk based on the exposure of the population to pesticides by food and water intake.

The objective of this study is to test the content of some common organochlorine pesticides in drinking water and in maize and cowpea grains, which are local staple foods, and to assess the health risk through estimated daily intakes as compared with acceptable daily intakes set by the Food and Agricultural Organization/World Health Organization (FAO/ WHO).

Materials and methods

Sampling areas and samples collection

Maize and cowpea grain samples were collected from ten different important local markets in the 'Région des Plateaux' in Togo (Figure 1). This part of the country is the main producer of cotton, coffee and cocoa (*Theobroma cacao* L.), which are the principal cash crops. Moreover, food crops like leguminous plants and cereals are also produced in this region. Grains of the two species (maize and cowpea), not subjected to post-harvest pesticides treatments, were sampled from new harvest in August 2004 to ensure they were representative of the region, and free of pesticides used in post-harvest protection.

At each of the ten markets, five subsamples of 100 g of the different species were collected directly from five different farmers. The grain sample, representative of the region, was obtained by pooling these individual samples. Water samples were collected from the Anié River and the Mono River in the cotton production area and from five wells in the town of Adéta in the coffee and cocoa production area (Figure 1). Each final water sample (2.5 litres of volume) was a composite of five subsamples of 0.5 litres per site. All water samples were collected in early September 2004 in glass bottles and refrigerated at 4° C until extraction (48 h).

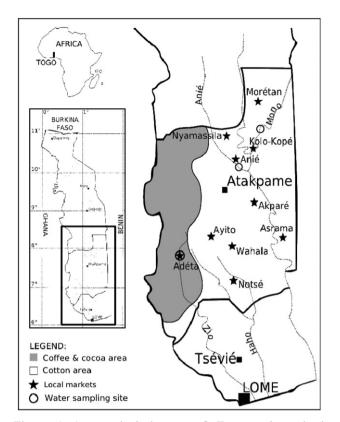


Figure 1. Agro-ecological map of Togo and retained sampling sites.

Reagents and solvents

Analytical grade reagents and solvents were used. Standard Pesticides-Mix 20 with 97.00–99.50% purity from Dr Ehrenstorfer (Augsburg, Germany) was provided by Cluzeau Info Labo (Sainte-Foyla-Grande, France). Florisil[®] cartridges used for purification were purchased from Waters Cooperation (Milford, MA, USA). Dichloromethane, hexane, acetone and anhydrous sodium sulphate were obtained from SDS (Solvant Documents Synthèse, Peypin, France). Diatomaceous earth (Hydromatrix[®]) was from Varian (Palo Alto, CA, USA).

Extraction of the pesticides residues

Pesticide residues in water samples were extracted by liquid–liquid extraction with dichloromethane (Hatrik and Tekel 1996). The 2.5-litre sample was fractioned in three subsamples which were placed in a separating funnel and mixed with 100 ml dichloromethane. The mixture was hand shaken for 30 min and allowed to settle for phase separation. The lower aqueous phase in the separating funnel was taken and extracted twice using 50 ml of dichloromethane. The combined organic extracts from each sample were filtered twice through a bed of anhydrous sodium sulphate to eliminate residual water and concentrated to dryness using a Buchi rotary evaporator at $35-40^{\circ}$ C. The dried extract was then dissolved in 1.5 ml hexane and kept in glass vials until GC/MS analysis.

Maize and cowpea grains were milled into flour which was subjected to extraction using an accelerated solvent extractor ASE 200 from Dionex (Salt Lake City, UT, USA) (Gan et al. 1999). Each sample of 5.36 g flour was mixed with 3.33 g of diatomaceous earth (Hydromatrix[®]), packed in a 22 ml cell and extracted for 5 min with acetone/dichloromethane (3:1 v/v) at 110° C, 1500 psi with an intermediate flush volume of 60% and for two cycles. The extract was washed with 30 ml of aqueous NaCl solution $(0.060 \text{ g ml}^{-1})$, dried on anhydrous sodium sulphate and concentrated to about 2.5 ml in a rotary evaporator at 38°C before clean-up on a Florisil Sep-Pack[®] Cartridges using adapted method (Müller et al. 2000). Elution was performed with 20 ml hexane/diethylether (94:6 v/v), 10 ml hexane/acetone (9:1, v/v) and 20 ml hexane/acetone (1:1 v/v), respectively. The rate of elution was about $0.5 \,\mathrm{ml}\,\mathrm{min}^{-1}$. The eluates were collected separately and evaporated individually using a Buchi rotary evaporator to about 1.0 or 1.5 ml residual solution ready to use for GC/MS analysis.

Analysis and quantification

All samples were analysed on a Hewlett-Packard 5890 Series II GC coupled to an HP MSD 5971 mass spectrometer operating in selected ion-monitoring (SIM) and electron-ionization modes (70 eV). The ions used in SIM were: γ -hexachlorocyclohexane (γ -HCH) (170, 181, 183, 219); heptachlor (100, 272, 135); aldrin (66, 79, 261, 263); heptachlor epoxide (81, 263, 353); 2,4-DDE and 4,4-DDE (246, 248); α -endosulfan (135, 197, 207, 241); dieldrin (79, 81, 261, 263, 279); 2,4-DDD and 4,4-DDD (165, 235, 237); endrin (81); β -endosulfan (197, 241); 2,4-DDT and 4,4-DDT (165, 235, 237). Chromatographic conditions were HP5MS capillary column $(30 \text{ m} \times 0.25 \text{ mm} \text{ i.d.} \times 10^{-3} \text{ capillary})$ 0.25 µm film thickness), injection in splitless mode, 1 µl of extract. The oven temperature was programmed as follows: starting temperature at 45°C for 1 min, followed by a first ramp of 35°C min⁻¹ up to 190°C, a second ramp at 3°C min⁻¹ up to 250°C held for 3 min and a third ramp at 20°C min⁻¹ to 280°C held for 15 min. The injector and the detector were maintained at 280°C and 300°C, respectively. The carrier gas was high purity helium at a constant flow rate of 0.8 ml min⁻¹. Identification of organochlorine pesticides was performed using a solution of pesticide standards. They were quantitatively determined by external standard method, comparing the peak area of each detected constituent with the same individual component in the corresponding calibration curve.

Analytical quality assurance

In preliminary validation studies the samples of water, maize and cowpea were spiked with a solution containing a mixture of the pesticide standards. Blank and recovery experiments were run for the various sample types using standard methods (Åkerblom 1995; Adou et al. 2001). In most cases the detected pesticide recoveries ranged between 70% and 115% with an acceptable repeatability standard deviation (SD) < 12%. Data were not corrected according to the recovery rate. The limit of detection (LOD) was set at a signal-to-noise ratio equal to three and the limit of quantification (LOQ) was determined to be three times the LOD.

Determination of estimated daily intake (EDI)

The dietary intake of pesticide is obtained by multiplying the residue level in food by the estimated amount of that food consumed per day. The total dietary intake of the pesticide residue is then obtained by summing up intakes from all foods and drinking water containing this residue (WHO 1997). In this study the average quantity of the edible parts of maize and of cowpea ingested per day by an adult of 55 kg was estimated via the method previously described by Sawaya et al. (1999). The estimated values were 350 g for maize and 45 g for cowpea according to the inquiry data collected from 50 households in five localities in 'Région des plateaux'. The volume of drinking water daily intake per person depends on physical activity: exercises intensity and duration, heat acclimatization, and individual sweat rate (Montain et al. 1995). The WHO estimated that adult females needed 2.21 day^{-1} and males 2.91 day^{-1} under average conditions. Under tropical climate, the manual labour increases the need in drinking water up to 4.5 litres for both men and women (Howard and Bartran 2003). To obtain the mean quantity of daily drinking water intake, the average value (litres) of these data was used:

$$\frac{(2.2+2.9+4.5+4.5)}{4} = 3.52$$
 litres.

Adjusted total estimated daily intake (EDI) was calculated using the pesticide residue data, the food and drinking water consumption and the correction factors that take into account the effects of food processing and cooking on residue levels (WHO 1997). In the absence of domestic correction factors, nonadjusted EDI was calculated as follows:

$$\begin{split} & \text{EDI}_{\text{FOOD}} \, (\mu g \, \text{kg}^{-1} \text{ body weight } \text{day}^{-1}) \\ &= \text{RL} \, (\mu g \, \text{kg}^{-1}) \times \, \text{FC}(\text{kg} \, \text{kg}^{-1} \text{ body weight } \text{day}^{-1}), \end{split}$$

where RL is the residue level in food consumed; and FC is the quantity of food consumed.

 $EDI_{DRINKING WATER}(\mu g kg^{-1} body weight day^{-1})$ $= RL(\mu g l^{-1}) \times DW(l k g^{-1} body weight day),$

where RL is the residue level in drinking water; and DW is the volume of drinking water intake.

$$EDI_{TOTAL} = \Sigma EDI_{FOOD} + EDI_{DRINKINGWATER}$$
.

Results and discussion

The average recoveries for all compounds ranged between 70% and 115%. Repeated analysis gave a standard deviation <12%. Detection limits varied for the different pesticides and samples. They were generally in the range $0.002-0.006 \text{ ug l}^{-1}$ in water.

 $0.2-1 \,\mu g \, kg^{-1}$ in maize (dry weight), and $0.5-3 \,\mu g \, kg^{-1}$ in cowpea (dry weight).

Levels of pesticide residues in tested food and water samples

Cowpea grains, maize grains and drinking water samples analysed for pesticide residue level showed that all samples were contaminated by DDT and its metabolites (DDE, DDD), y-HCH, aldrin, dieldrin, endrin, α - and β -endosulfan, heptachlor and heptachlor epoxide. The average concentrations of the pesticides detected are shown in Tables 1 and 2.

Regarding drinking water contamination, the DDT residue levels in Anié River $(0.11 \,\mu g l^{-1})$ and Mono River $(0.15 \mu g l^{-1})$ were found to be higher than the maximum residue limit $(0.1 \,\mu g \, l^{-1})$ of European Union Council Directive (98/83/EC) (European Union 1998). However, these values were below the MRL (2 ugl^{-1})

Table 1. Means, standard deviations of organochlorine pesticide residues detected in drinking water of Anié River, Mono River ad wells in Adéta town ($\mu g 1^{-1}$).

Detected pesticide	Anié River	Mono River	Well at Adéta	Guideline values of the World Health Organization (WHO)	European Union Council Directive
2,4-DDE	0.03 ± 0.00	0.05 ± 0.00	0.02 ± 0.00		
4,4-DDE	0.02 ± 0.00	-	0.00		
2,4-DDD	0.00	-	0.00		
4,4-DDT	0.11 ± 0.01	0.15 ± 0.01	0.03 ± 0.00	2	0.1
Aldrin	_	0.07 ± 0.00	_	0.03	0.03
Dieldrin	0.04 ± 0.00	0.06 ± 0.00	0.02 ± 0.00	0.03	0.03
Endrin	_	0.13 ± 0.01	_		0.1
Heptaclor	0.24 ± 0.01	0.11 ± 0.01	0.33 ± 0.01	0.03	0.03
Hepta epoxide	0.17 ± 0.01	0.15 ± 0.01	0.09 ± 0.00	0.03	0.03
α-Endosulfan	0.32 ± 0.01	0.29 ± 0.01	0.05 ± 0.00		0.1
β -Endosulfan	0.25 ± 0.01	0.40 ± 0.02	0.02 ± 0.00		0.1

Table 2. Means, standard deviations of organochlorine pesticide residues detected in maize and cowpea grains ($\mu g k g^{-1}$).

Detected pesticides	Cowpea (Vigna unguiculata)	French ^a standard	Maize (Zea mays)	Limit values ^b of the Food and Agricultural Organization/World Health Organization (FAO/WHO)
γ-HCH	4.95 ± 0.5	10	6.35 ± 0.8	500
2,4-DDE	14.57 ± 0.9		2.18 ± 0.2	
4,4-DDE	15.76 ± 1.2		5.29 ± 0.3	
2,4-DDD	11.66 ± 1.0		_	
4,4-DDD	4.71 ± 0.4		_	
4,4-DDT	12.49 ± 1.1	50	21.79 ± 1.3	100
Aldrin	6.00 ± 0.7	10	0.52 ± 0.01	20
Dieldrin	39.50 ± 1.2	10	18.09 ± 1.2	20
Endrin	13.16 ± 1.1	10	_	20
Heptaclor	3.92 ± 0.6	10	1.72 ± 0.02	20
Hepta epoxide	48.88 ± 1.2	10	17.65 ± 1.1	20
α-Endosulfan	98.80 ± 1.3	50	34.74 ± 1.2	100
β -Endosulfan	88.51 ± 1.2	50	65.71 ± 1.2	100

Notes: ^aMaximum residue limits ($\mu g k g^{-1}$) applied in France. ^bMaximum residue limits ($\mu g k g^{-1}$) prescribed jointly by the FAO/WHO.

set by the WHO (1998). The residual level of DDT $(0.03 \,\mu g \,l^{-1})$ in well water at Adéta was below the MRL of the European Union Council Directive and WHO limit (0.1 and $2\mu g l^{-1}$, respectively) (Table 1). The concentration of aldrin $(0.07 \,\mu g \, l^{-1})$ in water of Mono River exceeded the MRL $(0.03 \,\mu g \, l^{-1})$ of the European Union Council Directive. It was the same for endrin contamination $(0.13 \,\mu g \, l^{-1})$. Average residues of dieldrin $(0.04-0.06 \,\mu g l^{-1})$, heptachlor $(0.11-0.24 \,\mu g l^{-1})$, and heptachlor epoxide $(0.15-0.17 \mu g l^{-1})$ in freshwater from Anié and Mono rivers exceeded the MRL $(0.03 \,\mu g l^{-1})$ of the European Union Council Directive and WHO. Likewise, α -endosulfan (0.29–0.32 µg1⁻¹) and β -endosulfan (0.25–0.40 µg l⁻¹) residues were higher than the MRL of the European Union $(0.1 \,\mu g l^{-1})$. In well water from Adéta, the dieldrin residue level $(0.02 \,\mu g \, l^{-1})$ was close, but below the MRL $(0.03 \mu g l^{-1})$. In the same manner, the residue levels of α -endosulfan (0.05 µg1⁻¹) and β -endosulfan $(0.02 \,\mu g \, l^{-1})$ were below the MRL $(0.1 \,\mu g \, l^{-1})$. Conversely, the residue levels of heptachlor $(0.33 \,\mu g l^{-1})$ and heptachlor epoxide $(0.09 \,\mu g l^{-1})$ in this well water exceeded the MRL $(0.03 \,\mu g \, l^{-1})$. Similar values $(>0.1 \,\mu g \, l^{-1})$ have been reported by Awofolu and Fatoki (2003) in freshwater of Buffalo, Keiskamma, Swartkops and Tyume rivers in South Africa. Well water pesticide concentrations in Adéta were lower than those reported by Cissé et al. (2003) in well water in Senegal (2, 4-DDT: $5.58 \,\mu g l^{-1}$; 4, 4-DDT: 9.72 μ gl⁻¹; α -endosulfan: 1.26 μ gl⁻¹; β -endosulfan: $1.84 \,\mu g l^{-1}$; aldrin: $1.70 \,\mu g l^{-1}$; heptachlor: $3.43 \,\mu g l^{-1}$). In Tanzania, Kishimba et al. (2004) found DDT at 9 ugl^{-1} in well water. In the neighbouring Ghana. Ntow (2005) detected endosulfan at a 0.036 ng l^{-1} level in water from Lake Volta.

In Togo, the sources of drinking water for rural populations are wells located within the household areas and the rivers close to farms and/or villages. The presence of pesticide residues in well water presumably supported the assumption of the vertical mobility of organochlorine pesticides from various sources in the soil profile (Piao et al. 2004). The overall amount of DDTs, aldrin, dieldrin, heptachlor, heptachlor epoxide α -endosulfan and β -endosulfan are $1.19 \,\mu g \, l^{-1}$ in Anié River, $1.42 \,\mu g \, l^{-1}$ in Mono River and $0.58 \,\mu g \, l^{-1}$ in well water from Adéta. These values exceeded the MRL $(0.5 \,\mu g \, l^{-1})$ applied in European countries.

The average residue levels of DDT and its metabolites $(2.18-21.79 \,\mu\text{g}\,\text{kg}^{-1})$, γ -HCH $(6.35 \,\mu\text{g}\,\text{kg}^{-1})$, aldrin $(0.52 \,\mu\text{g}\,\text{kg}^{-1})$, dieldrin $(18.09 \,\mu\text{g}\,\text{kg}^{-1})$, heptachlor $(1.72 \,\mu\text{g}\,\text{kg}^{-1})$, heptachlor epoxide $(17.65 \,\mu\text{g}\,\text{kg}^{-1})$, α -endosulfan $(34.74 \,\mu\text{g}\,\text{kg}^{-1})$ and β -endosulfan $(65.71 \,\mu\text{g}\,\text{kg}^{-1})$ found in maize grains were below the maximum residue limits recommended by the FAO/WHO (Table 2). In cowpea grains, the average residual levels of DDT and its metabolites (4.71–15.76 μ g kg⁻¹), γ -HCH (4.96 μ g kg⁻¹), aldrin (6.00 μ g kg⁻¹) and heptachlor (3.92 μ g kg⁻¹) were also below the MRLs allowed in France (Ministère de l'Agriculture et de la Pêche France (MAPF) 2002). Conversely, the mean residue levels of dieldrin, endrin, heptachlor epoxide and α - and β -endosulfan in cowpea grains exceeded the MRLs applied in France. Their concentrations ranged from 13.16 to 98.80 μ g kg⁻¹ (Table 2). Previous studies on food contamination by pesticides in Togo (Djaneyé-Boundjou et al. 2000) showed higher residues levels in edible grains (maize, sorghum, rice and cowpea) DDT (20–300 μ g kg⁻¹), endrin (50–70 μ g kg⁻¹), dieldrin (10–63 μ g kg⁻¹), aldrin (3–27 μ g kg⁻¹) and heptachlor (49–490 μ g kg⁻¹).

The experimental data in Table 2 indicate that cowpea grains were more contaminated than maize grains in general. This is consistent with continued direct use of the tested pesticides in cowpea cultivation to control various insect pests (Oloude et al. 1995). Furthermore, soil contamination due to pesticides persistence is known to lead to uptake by subsequent crop after a previous use (Schroll et al. 1994; Fismes et al. 2002). This contamination route is likely the one leading to maize grain contamination as found in this work since this crop is not directly treated by pesticides in local cropping practices.

Assessment of exposure

The presence of organochlorine pesticides in drinking water and food indicates a current exposure of humans and the environment to these hazardous compounds. The estimated daily intake (EDI) of individual pesticide varied within the range of 0.004- $0.52 \,\mu g \, kg^{-1} \, day^{-1}$. These findings are comparable with those $(0.007-0.7 \,\mu g \, kg^{-1} \, day^{-1})$ previously mentioned for Russia (Sofina et al. 1991/1999; Barkatina et al. 1999). The EDI of DDTs $(0.25 \,\mu g \, kg^{-1} \, day^{-1})$ was significantly lower than previous literature reports: $1.42 \,\mu g \, kg^{-1} \, da y^{-1}$ in Japan (Nakagawa et al. 1995) and $1.22 \,\mu g \, kg^{-1} \, day^{-1}$ in Spain (Herrera et al. 1996). The highest EDIs were found in this study for dieldrin $(0.149-0.155 \,\mu\text{g kg}^{-1} \,\text{day}^{-1})$ and heptachlor epoxide $(0.158-0.162 \,\mu\text{g kg}^{-1} \,\text{day}^{-1})$. These values represented 149-155% of the acceptable daily intake (ADI) for dieldrin and 158.17-162.07% of ADI for heptachlor epoxide. The exposure level of the Togolese population to dieldrin and heptachlor epoxide exceeded the minimum level leading to health risk set by the FAO/ WHO (1998).

It is known that fatty food like fish, meat, eggs and milk accumulate organochlorine pesticides and may be making a more significant contribution to residue exposure than water, maize or cowpea. However, in the investigated areas, the consumption of fish and meat is very limited. In addition, milk and eggs are scarcely part of the diet.

Conclusion

Drinking water, maize and cowpea grains contamination by various organochlorine pesticides (most of which are currently termed as persistent organic pollutants) such as y-HCH, DDTs, aldrin, dieldrin, endrin, heptachlor, heptachlor epoxide, and endosulfan were determined to estimate their human daily intake in cash crop (cocoa, coffee, cotton) cultivation areas in Togo. The findings indicated that concentrations of organochlorine pesticides detected in water from the Anié River (cotton area), the Mono River (cotton area) and wells at Adéta (cocoa and coffee area) were higher than the maximum residue limits recommended by the European Union and the WHO. In comparison with the French standard, the cowpea grains tested were highly contaminated by dieldrin, endrin, heptachlor epoxide, and endosulfan. Dieldrin and heptachlor epoxide were the most worrying of the tested pesticides due to their higher EDI values. The experimental findings support the assumption that maize is more likely to be the principal route of pesticide intake by humans in the investigated areas and very probably throughout the country. Nevertheless, daily pesticide intake of even small amounts of fatty food like fish, meat, eggs and milk is an issue of concern to take into account in a broader monitoring programme of organochlorines related to food security. This study is a useful starting point for exposure assessment and the evaluation of dietary exposure from different relevant sources making up the total exposure in studied localities.

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