

Detection and spatial distribution of pesticides in cocoa farming in Ivory Coast

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Abstract

Ivory Coast is the world's leading cocoa producer with more than 40% of market volumes. But its production is confronted with the scourge of innumerable pests of cocoa trees favored by the sub-equatorial and humid tropical natural environment. The main response strategy chosen is the use of pesticides. However, this practice has consequences for terrestrial and aquatic environments. Thus, the objective of this work is to quantify the concentrations of 22 pesticides from the official list of approved for cocoa cultivation in Ivory Coast in water (running and phreatic), soils and cocoa bean on ten sites across the entire production basin to understand their transfer between the different matrices.

The results show contamination at all sites and in all studied matrices and underline that fungicides are the most frequently detected pesticides. And among them pyraclostrobin is the main one. This omnipresence of fungicides is explained by the high prevalence of cryptogamic diseases causing pod rot because they constitute the main health concern, with losses of 30% or even 40% in production, compared to cocoa swollen shoot disease and insect pests. The spatial distribution reveals variability in contamination between sites: southern areas, which are wetter, generally show higher concentrations. The study highlights the need for pesticide monitoring and the importance of understanding their transfer (soil-water) in order to reduce environmental impacts, ensure sustainable production and protect one health (human and environmental).

Introduction

Ivory Coast is the world's leading cocoa producer with more than 40% of market volumes. Its economy has a strong addiction to its “brown gold”. This dependence is such that authorities and producers cannot be indifferent to the scourge of innumerable pests of cocoa trees favored by the sub-equatorial and humid tropical natural environment. These include, among other harmful plant pathogens, diseases and pests, brown rot caused by the pseudo-fungus *Phytophthora megakarya*, orange rust, mirids, bark beetles, trunk borers, etc.). In addition, the resurgence of “Swollen shoot” disease or edema of cocoa tree shoots (viral disease whose known vectors, to date, are species of mealybugs, with 5 symptoms including swelling of the branches, swelling of roots, reddish spots along the veins on young leaves, yellowish spots on adult leaves, stunting of pods) (CCC, 2014). In summary, there is a very large diversity of bio-aggressors like insects and diseases of bacterial, viral, fungal, algal or physiological origins (CABI, 2019; Keane, 1992; Keane & Putter, 1992).

In response to these threats that loom over the future, a national response strategy has been developed. This strategy is materialized by the “2QC” program (Quantity-Quality-Growth program) (CCC, 2014) over a period of 10 years from year 2014 to 2023. This program includes six strategic axes. In the first axis, the program aims to “Improve farm productivity”. Among other productivity tools, chemical methods of protecting cocoa trees with phytosanitary products are recommended (CCC, 2013, 2014). The approach consists of facilitating cocoa farmers' access to agricultural inputs such as free of charge chemical pesticides (insecticides, fungicides mainly). The objective is to intensify the fight against phytopathogens and harmful pests of the cocoa tree.

Pesticides refer to the active substance, the commercial specialty or preparation composed of one or more active substances as well as a certain number of adjuvants, solvents, inert ingredients, residual substances and metabolites which are molecules which appear during of product degradation. The intensive use of pesticides have led to a contamination of various matrices such as water, soils, sediments and food (Burby et al., 2000; Khan et al., 2022; Mitra et al., 2024). They have been monitored to evaluate their behavior in these environmental compartments.

The fate of these contaminants depends on the intrinsic characteristics of the molecules as well as environmental parameters such as volatilization, runoff, leaching, absorption or even the removal of crops (Maillard, 2014). Indeed, factors influencing these processes are climate (in particular temperature, precipitation and radiation), physical conditions (type of soil, slope and altitude), the characteristics of the active substances (volatility, degradation rate, etc.) and agricultural practices (application techniques, tillage, grass cover, etc.) (N'gnessan Bi Vami et al., 2017).

Tropical climatic conditions, with higher temperatures, precipitation, radiation and humidity, accelerate many processes such as volatilization, leaching and degradation (Lewis et al., 2016; Racke et al., 1997; Varca and Magallona, 1994). The kinetics of these processes is also increased by the characteristics of tropical soils, due to their organic matter contents and high infiltration capacity (Mottes, 2013). At the level of agricultural practices, tillage, for example, affects the hydraulic properties of the soil which, in turn, modify different hydrological processes such as leaching and runoff (Alletto et al., 2010). Erosion is also a very important phenomenon (Cheula, 2013). It should be noted that most of these processes are less well understood and fewer measurements are available in tropical regions than in temperate regions (Lewis et al., 2016). For a better understanding of the different mechanisms, additional research is necessary and specifically in Ivory Coast where the use of pesticides seems a necessity in cocoa cultivation.

However, this approach is not without consequences for the environment of the country which is classified among the “25 Biodiversity hotspots”. To our knowledge, few studies concerning the implementation of a protocol for detecting pesticides applied to cocoa trees in Ivory Coast has been carried out. It is the same for the analysis of the spatial distribution of pesticides over the cocoa growing area in water, soil and cocoa pods.

The aim of this study is therefore to provide a concomitant analysis of pesticides in water, soil and cocoa beans in diverse zones of cocoa farming in the Ivory Coast.

Materials and methods

Description of the study area

The Ivorian territory covers 332,462 km² and is bordered to the south by the Atlantic Ocean for 520 km leading to the Gulf of Guinea. Its continental part shares 3110 km of borders with 5 countries namely to the east, Ghana to the southwest Liberia to the northwest Guinea and to the north Mali and Burkina Faso

The study area is in the central and southern part of the country which includes the preferred districts for cocoa farming (see Fig. 1). Cocoa production in Ivory Coast takes place between longitudes 2.30° W and 8.30° W and latitudes 4° N and 9° N covering an area of 170,320 km². This cocoa production area represents approximately 53% of the country's territory. The climatic conditions are favorable for good growth and productivity of the cocoa tree. Thus, rainfall is bimodal with 2 rainy seasons (April-July and September-October) and 2 dry seasons (December-May and August). Cumulative rainfall averages between 1200 and 1500 mm/year with significant peaks depending on the region. The average annual temperature varies from 27°C to 31°C (Yoroba et al., 2019). The regions of East, Center-West and South-West are the main cocoa production areas in Ivory Coast. But the Central-West and South-West regions have experienced significant growth in new planted areas and their production, making them the “new cocoa belt”. These regions have thus supplanted the eastern region, pioneer of cocoa farming in Ivory Coast. Its production has remained constant or even declining due to the aging of plantations, diseases and pests of cocoa trees.

Sampling sites and techniques

In total, 10 plantations spread over 10 production districts were chosen randomly and the majority of them (i.e. 7 plantations) are located in the “new cocoa belt” because the most of the national production is concentrated there and it extends from the southwest to the west as shown by the importance of the percentages of planted areas (see Fig. 1). The 3 other sites were selected in the south-east and east where production is less important because of declining, particularly in the east due to climate change. Sampling took place over three years during the dry season.

Soils have been collected based on methods recommended by the “Codex Alimentarius Commission” (FAO & WHO, 1999) and the FAO,(2016) for sampling and determination of pesticides residues.

Cocoa pods have been harvested in accordance with the sampling methods recommended for large fruits with a unit mass greater than 250 grams (AgVic, 2022; FAO, 2016; FAO and WHO, 1999). Fresh mucilaginous beans are removed from the pods and placed in a sampling bag.

Surface water has been collected near the center of the watercourse by immersing a 500 mL HDPE (high-density polyethylene) sampling bottle, approximately 30 cm below the water surface. It is filled completely, without leaving any air in the neck and closed using a seal and a lid (Deng and Ensminger, 2022; Khadse, 2010).

For groundwater, in the case of open wells, a bucket attached to the end of a rope has been used to collect water from the bottom of the well. The bucket is immersed below the surface of the water. The bottle is filled with the contents of the bucket. For drilling with human-powered or HMP pumps (manual or pedal) (CIV Gouv., 2021; Fondation de l'Eau, 1990), the sample is collected after running the pump for about 5 minutes to let the water flow at a loss. Following this, the 500 mL HDPE bottle is filled.

Preliminary sample treatments

The soil samples are spread out in the shade to dry. Once dried, they are crushed. Then 250 g of ground material are packaged in an HDPE screw jar for laboratory analysis.

The fresh mucilaginous beans are fermented for 1 week, taking care to knead them regularly. At the end of this week of fermentation, the beans are spread out in the sun to dry. This drying can last from 1 week to 2 weeks depending on the sunshine. It aims to limit the formation of mold. Once dried, the beans are peeled and crushed. Then 250 g of crushed beans are packaged in a jar for analysis in the laboratory.

The water is filtered using a Büchner funnel or filtration under reduced pressure. The contents of the bottles are first homogenized. Then, depending on the water, the samples are filtered on 8 µm membrane before passing to 0.45 µm. The filtered water is collected in new 500 mL bottles and stored in the refrigerator at 4°C.

Analyzed pesticides molecules

The monitored twenty-two (22) molecules are from the official list of pesticides approved for cocoa cultivation in Ivory Coast (Table 1).

Table 1
Monitored pesticides in water, soils and cocoa

Insecticides			
Alpha cypermethrin	Deltamethrin	Lambda-cyhalothrin	Pyrimicarb
Bifenthrin	Chlorpyrifos-methyl	Thiacloprid	
Clothianidin	Imidacloprid	Thiamethoxam	
Fungicides			
Dimethomorph	Metalaxyl-M	Pyrimethanil	
Fenpropidin	Pyraclostrobin	Cyproconazole	
Herbicides			
Alachlor	Trifluralin	Oxadiazon	
Dichlobenil	Metamitron	Pendimethalin	

Extraction procedure of pesticide residues

Surface and groundwater samples

Water samples have been extracted by using Celite cartridges (CHROMABOND XTR). The XTR Chromabond column, composed of a highly purified inert diatomite material known as celite, is used during extraction. Its volume is 150 mL, the capacity of the aqueous solution is 50 mL, and its filling quantity is 37°500 mg. The column should not interact with the analyte to be analyzed, has high surface area providing large pore size, and high pore volume. It works in the pH range of 1 to 13, and the maximum capacity of the aqueous solution should not exceed 50 mL. First, the aqueous solution is loaded into the column constituted of diatomite material known as celite. Second, the water is adsorbed on the solid support. This process that usually takes between 10 and 15 minutes. Third, the extraction solvent is added on the top of the cartridge and will flow by gravity over the cartridge with organic compounds. (Khoury et al., 2023).

Soil samples

Soils (50 grs) were extracted twice using acetonitrile by Pressurized Liquid Extraction (PLE) using an ASE 350 (Accelerated Solvent Extraction) system at 1500 psi for 10 min at 150°C (flush 60%, purge time 150 s). Extracts were slowly concentrated to 1 mL in a fume hood before analysis.

Cocoa samples

Cocoa samples were extracted by the *QuEChERS method*. For this, 10 g of dried, hulled and crushed beans are mixed with 10 mL of HPLC grade acetonitrile and placed in a 50 mL centrifugation tube. The tube is shaken by hand for 1 min, vortexed for 1 min and finally centrifuged for 10 min at 5000 rpm. Then, 3 mL of supernatant is taken and put into a QuEChERS 26245 tube (containing 900 mg MgSO₄, 300 mg PSA, 300 mg C₁₈-EC, 45 mg GCB). The tube is shaken by hand, vortexed for 30 seconds then centrifuged for 10 min at 5000 rpm. 2 mL of supernatant are placed in a vial and evaporated until exactly 1 ml (see Fig. 2).

Quantification of pesticides residues by chromatographic analysis

Reagents and chemicals

Solvents used for the extraction of samples and analysis by chromatography are HPLC grade and were obtained from Fisher Scientific (France). Calibrations were done by using pure certified standards (> 99% purity) of individual pesticides purchased from Merck (L'Isle D'Abeau, France). Stock solutions at 1 g L⁻¹ of each molecule were prepared in acetonitrile and a work solution at 10 mg L⁻¹ was used for the realization of the calibration's curves in GC/MS and LC/MSMS.

Quantification by GC-MS

The system used is a GC Trace (Thermo Scientific) coupled to a DSQ II single quadrupole mass spectrometer (Thermo Scientific). Chromatographic separation was carried out with an Optima XLB column (30 m x 0.25 mm; 0.25 µm) from Macherey-Nagel. Helium was used as the carrier gas at 1 mL min⁻¹. The injection (3 µL) was carried out by pulsed flow (3 mL min⁻¹ for 1 minute) at 250°C in splitless mode (1 min). The temperature gradient used for the separation of the molecules is as follows: 50°C (1 min) then ramps from 20°C/min to 240°C (1 min) then 3°C/min to 270° C (11 min) then 20°C/min up to 290°C (1 min) and 10°C/min up to 320°C (5 min).

The mass spectrometer source and the transfer line temperature were maintained respectively at 210°C and 300°C.

Quantification of pesticides was done in the SIM (Single Ion Monitoring) mode. Ions selected for each pesticide are presented in Table 2 with detection and quantification limits. Trifluralin d¹⁴ was used as internal standard.

Table 2
List of molecules analysed by GC-MS

Molecule	Ion quantification	Ion qualification	LOD (ng g ⁻¹)	LOQ (ng g ⁻¹)	LOD (µg L ⁻¹)	LOQ (µg L ⁻¹)
Trifluralin d ¹⁴	267	315				
Alachlor	132	160	1.96	6.25	1.96	6.25
Bifenthrin	181	165	2.28	7.58	2.28	7.58
Chlorpyrifos-methyl	271	286	2.36	7.02	2.36	7.02
Cyproconazole	202	153	1.25	4.28	1.25	4.28
Deltamethrin	181	152	1.00	3.33	1.00	3.33
Dichlobenil	171	136	1.54	4.23	1.54	4.23
Fenpropidin	95	70	1.23	4.01	1.23	4.01
Metalaxyl-M	125	159	1.01	2.28	1.01	2.28
Metamitron	202	104	1.10	2.53	1.10	2.53
Oxadiazon	174	202	1.52	5.25	1.52	5.25
Pyrimethanil	183	198	1.12	3.28	1.12	3.28
Pyrimicarb	72	166	1.32	4.02	1.32	4.02
α-cypermethrin	181	105	1.61	5.37	1.61	5.37
λ-cyhalothrin	181	195	1.41	5.23	1.41	5.23
Dimetomorph	165	301	0.56	1.83	0.56	1.83

Quantification by HPLC-MSMS

For LC analyses, a Thermo Scientific TSQ Quantum Access triple quadrupole mass spectrometer operating in electrospray heated positive ionization (HESI+) mode coupled to a Thermo Accela 1250 pump and a Thermo Combi Pal autosampler maintained at a temperature of 5°C was used. A volume of 20 µL of the sample was injected into a Nucleodur C₁₈ Pyramid column (150 mm × 3 mm, 3 µm) at room temperature. A water/ACN mobile phase both containing 0.1% formic acid, at a flow rate of 0.3 mL min⁻¹ was used. The gradient started at 70:30 (H₂O/ACN) then increased to 50:50 (v/v) in 5 min, then to 20:80 (v/v) at 11 min and 5:95 at 18 min (hold for 6 min) and finally returned to 70:30 at 25 min (hold for 8 minutes) for a total duration of 33 min. Calibration was done by internal standard method (pendimethalin d⁵).

Detection and quantification of all compounds were performed using MRM detection mode. The source was operated in positive ionization mode with a spray voltage of 4500 V and a vaporizer and capillary temperature of 270°C and 300°C respectively. Nitrogen was used for the sheath and auxiliary gas pressure (20 and 10 arbitrary units) while argon was used for the collision pressure (1.5 arbitrary units). Two precursor product ion transitions for each analyte and internal standard were used for quantification. The selected transitions for MSMS analysis, collision energy and retention times are shown in Table 3.

Table 3
Multiple reaction monitoring (MRM) transitions, corresponding collision energies (CE), retention times (t_R) for each target compound analyzed in LC/MSMS (two MRM transitions were monitored for a given target compound, the ion product 1 was used for quantification and product ion 2 was used for qualification).

Compound	Parent Ion	Q1	Q3	t _R (min)	LOD (ng g ⁻¹)	LOQ (ng g ⁻¹)	LOD (µg L ⁻¹)	LOQ (µg L ⁻¹)
Thiametoxam	292	181	211	4.35	8	16	8	16
Clothianidin	250	132	169	5.22	2	5	2	5
Imidacloprid	256	209	211	5.48	12	21	12	21
Thiacloprid	253	126	99	7.40	27	91	27	91
Pyraclostrobin	388	194	163	14.70	5	17	5	17
Pendimethalin-d ⁵	287	212	194	17.22				
Pendimethalin	282	212	194	17.28	4	13	4	13

Results and discussions

Frequencies of detection

Parameters strongly related to frequency of detection (FD)

To understand the behavior of a pesticide in the environment, it is necessary to analyze the relationship between the frequency of pesticide detection (in water or soil) and its K_{oc} (organic carbon/water partition coefficient). However, the detection frequency also depends strongly on the persistence (half-life, DT₅₀) of the pesticide in the environment.

The K_{oc} coefficient is defined as the ratio between the concentration of the pesticide in its sorption state (i.e., bound to soil particles) and the solution phase (i.e., dissolved in soil water) (FAO, 2000a) and it measures the adsorption capacity of a given pesticide in soil.

Pesticides with a low K_{oc} value are therefore more likely to infiltrate groundwater than those with a high K_{oc} value (FAO, 2000a). Due to the wide range of K_{oc} values, the logarithm of K_{oc} is used to indicate the degree of mobility in the soil (FAO, 2000a) (see Table 4).

Table 4
Classification of pesticide mobility in soil
(according to FAO, 2000)

LogK _{oc}	Degree of pesticides mobility in soils
< 1	Very mobile
1–2	Mobile
2–3	Moderately mobile
3–4	Slightly mobile
4–5	Not very mobile
> 5	Immobile

The FD of pesticides in soils can be explained by their persistence as assessed by IUPAC PPDB (2024d) to:

- If half-life DT₅₀ field < 30 days then the molecule is non-persistent,
- If DT₅₀ field is between 30–100 days, then the molecule is moderately persistent,
- If DT₅₀ field is between 100–365 days, then the molecule is persistent,
- If DT₅₀ field > 365 days, then the molecule is very persistent.

Among the 22 analyzed molecules, 13, 13, 17 and 11 molecules were respectively detected in surface water, groundwater, soils and cacao (Table 1).

FD in surface water

In surface water, metamitron (herbicide) has a 100% frequency of detection (FD) followed by an 83% FD for bifenthrin (insecticide), λ-cyhalothrin (insecticide) and pyraclostrobin (fungicide) (Fig. 3a). Bifenthrin, is a molecule prohibited for use but its detection is linked to its authorized use for cocoa. It is fairly regularly found in these surface waters. Bifenthrin has the ability to bind tightly to the soil (K_{oc}= 236610 mL/g) where it remains immobile (IUPAC PPDB, 2024a). Its DT₅₀(field) half-life is 86.8 days, thus moderately persistent in soils. However, soil-bound bifenthrin can potentially contaminate surface water through runoff (NPIC, 2011). Apart from the explanation by its properties, its presence in waters may also be the result of continued use by cocoa producers of old stocks available for sale despite the ban.

The same could applied to λ-cyhalothrin, with a K_{oc} of 283,707 mL/g (IUPAC PPDB, 2024c).

Other fungicides have been found with significant FD of 67% and 33% respectively for cyproconazole and pyrimethanil. Fenpropidin and metalaxyl-M present a low FD < 20%.

Metamitron is frequently found in runoff water (Norat and Turnbull, 2008), because this herbicide has a low retention rate in the soil (K_{oc}= 77.7 mL/g) (IUPAC PPDB, 2024c). It will therefore contaminate surface water through run-off. Its high FD also reflects a trend towards chemical weeding with herbicides in Ivory Coast as shown by various studies (Boraud et al., 2010; Mangara et al., 2014; N’guessan et al., 2016).

The pyraclostrobin FD is due to the spreading of this fungicide in response to the scourge of pod rot caused by different species of fungi of the *Phytophthora* genus (Mpika et al., 2009).

Fungicides, due to the contribution of pyraclostrobin, are therefore the main contaminants of surface water with 52% contribution to total contamination followed by insecticides and herbicides (31% and 15% respectively) despite the detection frequency of 100% for metamitron.

Figure 3a: Classification of active materials according to their detection frequencies in surface waters

FD in groundwater

In groundwater the FD are in order of importance: cyproconazole (100%) > pyraclostrobin (88%) > oxadiazon 75%) > bifenthrin (63%) (Fig. 3b). Cyproconazole detection frequency of 100%, distinguishes groundwater from surface water where pyraclostrobin was most important (with 83%). In all groundwater (Fig. 3b), it should be noted the presence of three active ingredients prohibited for use in the EU: dichlobenil (herbicide), alachlor (herbicide) and bifenthrin (insecticide). Their detection frequencies (FD) are respectively 13%, 50% and 63%. But among these molecules, only bifenthrin (insecticide) is on the official list of molecules approved for cocoa in Ivory Coast and not the other two. On the other hand, the presence of dichlobenil (herbicide) and alachlor (herbicide) could be explained by old production practices.

If we compare these results with those of surface water, we note that in these surface water, fungicides are certainly the majority, but in a less significant proportion, with 52% contribution to the total contamination of this surface water. This can be explained by the fact that the two main fungicides quantified, i.e. pyraclostrobin and cyproconazole, are more likely to leach into groundwater rather than runoff into surface water. Indeed, we note that Cyproconazole has a solubility of 93 mg L^{-1} (with $10 < \text{solubility} < 100$). It is easily soluble in water. It has a $\text{DT}_{50} = 129$ days (with $\text{DT}_{50} > 100$ days), is persistent in the soil. It is relatively mobile, with $K_{OC} = 309 \text{ L/kg}$ ($2 < \text{Log}K_{OC} = 2.49 < 3$) according to the classification scales (FAO, 2000). However, pesticides which are very soluble in water, which are relatively persistent and which are difficult to absorb by soil particles (low K_{OC} or low K_d distribution coefficient) have the greatest potential for movement (Kerle et al., 1996). As a result, cyproconazole is highly leachable to groundwater as also attested by the PPDB (IUPAC PPDB, 2024b). As for pyraclostrobin, it has a $\text{DT}_{50} = 33.3$ days (with $\text{DT}_{50} > 30$ days) and is moderately persistent in soils with $K_{OC} = 9304 \text{ L/kg}$ and $\text{Log}K_{OC} = 3.97$ ($3 < \text{Log}K_{OC} < 4$). It is relatively mobile in soils towards groundwater (FAO, 2000). The high rainfall in the case of Ivory Coast will contribute to accentuate this leaching towards groundwater. In addition, its solubility of 1.9 mg L^{-1} (with $1 < \text{solubility} < 10$) is low. Therefore, it does not dissolve sufficiently to be carried away in runoff water (Leonard, 1990; PES & Cornell University Cooperative Extension, 2024). In addition, the low solubility of pyraclostrobin and its lipophilicity, give it resistance to rain and watering. (COMPO-France, 2005; Guston, 2010). However, it is these runoff waters which will mainly form surface water. This explains the lower proportion of fungicides in surface water than groundwater.

Figure 3b: Classification of active materials according to detection frequencies in groundwater

FD in soils

The residues most observed in soils (Fig. 3c) are pyraclostrobin and imidacloprid (FD 90%) followed by thiamethoxam and thiacloprid (FD 80%), λ -cyhalothrin, α -cypermethrin and deltamethrin (FD 50%), clothiadin (FD 40%) and bifenthrin (FD 30%).

Thiamethoxam ($\text{DT}_{50} = 39$ days), thiacloprid ($\text{DT}_{50} = 39$ days) and pyraclostrobin ($\text{DT}_{50} = 33.3$ days) are moderately persistent, but with a $K_{OC} = 3304 \text{ L/kg}$, pyraclostrobin can bind strongly to soil and be persistent (Guston, 2010). While, Imidacloprid has $\text{DT}_{50} = 174$ days and is persistent in soil, which may help explain its FD.

Figure3c: Classification of pesticides according to detection frequencies in soil

FD in cocoa beans

The active substances found in cocoa beans in the greatest percentage are cypermethrin and pyraclostrobin with respectively 63% and 75% FD (Fig. 3d). The bioaccumulation potentials of these two active substances could explain their high FD values. According to (IUPAC PPDB, 2024d), bioaccumulation is characterized as follows:

- If $\log P$ or $\log K_{OW} < 2.7$ then there is low bioaccumulation,
- If $\log P$ or $\log K_{OW}$ between 2.7–3 then there is moderate bioaccumulation,
- If $\log P$ or $\log K_{OW} > 3.0$ then there is high bioaccumulation.

Cypermethrin and pyraclostrobin have respectively a $\log K_{OW}$ of 5.5 and 3.9. They present thus a high bioaccumulation potential. In addition, they can be diffused by locosystemy (local systemy) or translaminar processes (Afssa, 2010; Bartlett et al., 2002; Carisse, 2011) and possibly a local intra-cellular diffusion.

In addition to this bioaccumulation factor, it should be noted that cocoa is rich in fat. It therefore has a natural lipophilicity which promotes the penetration of hydrophobic pesticides explaining the high FD in cocoa beans.

Hypothesis of pesticide transfer from the soil matrix to groundwater

The initial hypotheses concerning pesticide transfers from soil to groundwater can be based on the leaching potentials given by the values of the Gustafson indices (Groundwater ubiquity score: GUS) (Gustafson, 1989). GUS combines two key properties of a molecule: its persistence in the soil (DT_{50}) and its affinity for soil organic matter (adsorption coefficient K_{OC}) (Eq. 1).

$$\varvec{G} \varvec{U} \varvec{S} = \varvec{cl} \varvec{co} \varvec{g}_{10} \left(\varvec{D} \varvec{T}_{50} \right) \times \left(4 - \varvec{cl} \varvec{co} \varvec{g}_{10} \left(\varvec{K}_{\varvec{varve}} \right) \right) \quad (\text{Eq. 1})$$

where :

K_{OC} = soil organic carbon adsorption coefficient

→ measures the mobility of a molecule (low K_{OC} = very mobile). It expresses the adsorption capacity on soil organic matter. The adsorption capacity of organic matter is due to its high chemical reactivity with both mineral surfaces and organic molecules, allowing for various types of interactions with pesticides. The sorption capacities of organic matter are controlled not only by their chemical composition but also by their size, due to a greater number of sorption sites associated with a larger surface area (Benoit et al., 2008). As we have previously pointed out, the absorption of a given pesticide is greater in soils with higher organic matter content

DT_{50} = half-life in soil (in days)

→ measures pesticide persistence in soil until half of the initial concentration of the molecule is dissipated through degradation.

Pesticides can be grouped into three main classes based on their leaching potential, mobility, and persistence in soils.

The high leaching potential class ($GUS \geq 2.8$) includes only cyproconazole ($DT_{50} = 129$ days, $K_{OC} = 309$ L/kg, and $GUS = 3.19$), imidacloprid ($DT_{50} = 174$ days, $K_{OC} = 255$ L/kg, $GUS = 3.57$) and thiamethoxam ($DT_{50} = 39$ days, $K_{OC} = 68$ L/kg, $GUS = 3.45$).

Cyproconazole has a solubility of 93 mg L^{-1} (with $10 < \text{solubility} < 100$). It has a detection frequency of 20% in the analyzed soils, while it presents the highest detection frequency ($FD = 89\%$) in the sampled groundwater. This result is consistent with the high leaching potential value ($GUS = 3.19$). Kerle et al., 1996 have also shown that pesticides which are very soluble in water, which are relatively persistent and which are difficult to absorb by soil particles (low K_{OC} or low K_d distribution coefficient) have the greatest potential for movement.

Astonishingly, imidacloprid and thiamethoxam are detected with high frequency in soils, not in groundwaters, although high water solubilities (610 mg L^{-1} and 4.1 g L^{-1}).

The class with moderate leaching potential ($1.8 \leq GUS < 2.8$) is represented in this study by five molecules (pirimicarb ($GUS = 2.63$), dimethomorph (2.40), pyrimethanil (2.28), alachlor (2.03), thiacloprid (2.30) .

The transfer levels of these molecules to groundwater are generally linked to their physico-chemical properties, including adsorption onto soil organic matter with the organic carbon-water partition coefficient (K_{OC}), solubility in water, half-life in soil (DT_{50}) and also soil characteristics (e.g., texture, clay content). Pirimicarb is a non-persistent to moderately persistent insecticide in soil ($DT_{50} = 9\text{--}73.6$ days). It has a water solubility of 3100 mg L^{-1} . Pirimicarb was not found in soils, but it was detected in groundwater at a frequency of 33%. Due to its non-persistence, some may have dissipated and some leached from the soil into groundwater, given its solubility characteristics. Other studies conducted in tropical environments have shown that molecules considered to be poorly or moderately leachable are detected in groundwater. Researchers have linked these observations to the specific characteristics of the tropical environment, such as seasonal variations, preferential groundwater flow patterns, and the transport of molecules associated with soluble organic matter. (Dores, E. F. G. DE C. et al., 2023).

Alachlor, is moderately to weakly adsorbed onto organic matter ($K_{OC} = 124 \text{ mg L}^{-1}$, $50 < K_{OC} < 150$). Its half-life is 14 days in soils ($DT_{50} < 16$). This herbicide is non-persistent in soil because it dissipates rapidly through degradation. It has a moderate solubility of 240 mg L^{-1} , which facilitates its leaching by water into groundwater.

Thiacloprid was only detected in soils, suggesting that other parameter than DT_{50} and K_{OC} play a significant role in the transfer of this pesticide as rainfall, soil type, water table depth, actual degradation on the ground, such as biological degradation.

Finally, there are the **classes with low ($GUS < 1.8$) or very low ($GUS < 0$) leaching potential**. These are molecules whose properties, such as their mobility (which varies from "difficult to move" to "immobile") or K_{OC} values, tend to retain them in soils. However, analytical results show their presence in groundwater (see Fig. 3b and 3c).

According to GUS indicator, pyraclostrobin is classified as having a low leaching potential (Eq. 2) with $DT_{50} \approx 37$ days as a representative value (maximum value used in regulatory assessments) and $K_{OC} \approx 10\,000 \text{ mL/g}$ as a representative average value.

$GUS = 1,57 \times (4 - 4,00) = 1,57 \times 0 = 0,00$ (Eq. 2)

Indeed, a strong presence of pyraclostrobin is observed in soils to which it binds strongly (Guston, 2010). The actual half-life varies greatly depending on soil type, temperature, biological conditions, K_{OC} depends on the type of organic matter in the soil and can vary in practice.

The GUS based on molecules' persistence (DT_{50}) and mobility (K_{OC}) is an empirical index for estimating the risk of pesticide transfer to groundwater. It is easy to calculate and widely used but it's a static index based on average values. It does not consider climate, soil type, or agricultural practices. Moreover, it does not take pesticide metabolites into account. It needs to be supplemented by empirical data from several sampling campaigns.

Spatial distribution and contamination of the different pesticides' categories across the cocoa production basin

The objective is first to highlight the health constraints faced by different cocoa-growing regions, which lead to the use of sprays to address these various threats. Secondly, the aim is to establish a spatial distribution of pesticide families for each matrix.

Surface water

Across all the regions covered for sampling, all the surface waters that were sampled contain different molecules at varying concentrations from one sampling site to another.

The percentage of each pesticide family is reported to the total quantified pesticides on each sampling site (Fig. 4a).

The three families of pesticides are represented throughout the cocoa basin but are distributed differently depending on the latitude (Fig. 4a). Thus, around latitude 7°N, herbicides are dominant at 63% on the Daloa site for example. This plantation is old with many dead cocoa trees. The foliage of cocoa trees does not cover the undergrowth, which promotes the growth of weeds. The producer therefore treats them with herbicides. Close to 6°N of latitude, the family of insecticides is in the majority as illustrated by the sites of Soubré and Gagnoa (54%). At latitudes below 5.5°N, fungicides are predominantly present in surface waters. The analyzed sites are located in a region where temperatures and rainfall are most favorable to diseases and pests (CNRA, 2021; M'Bo et al., 2023). Fungicides are the majority in the Éholié rivers in Aboisso (83%), in the N'koua in Alépé (46%), as well as in San Pedro (38%). These three sites are in two regions identified by the CNRA (national agricultural research center) as having a high prevalence of fungal diseases and in an adjoining region: San Pedro and Comoé (CCC, 2013), and Agnéby region adjacent to the Comoé. The spatial distribution of the three pesticides families is in accordance with the climatic conditions of the Ivory Coast cocoa production basin (Ndoumbé et al., 2010).

However, it is noted that the surface waters of the entire cocoa basin have significant percentages of fungicides (from 32% in the Soubré region to 83% in that of Aboisso). The Soubré site is in the Nawa region designated by the CNRA for anti-fungal protection, which explains the proportion of 32% of fungicides, while Gagnoa in the Gôh region is not in this anti-fungal program. The Haut-Sassandra region where Daloa is located is one of the areas with a prevalence of fungal diseases identified by the CCC and requiring protection through the distribution of fungicides.

Groundwater

Fungicides are predominant in six of the groundwaters sampled including five wells in Divo (96%), Aboisso (92%), Biankouma (87%), Soubré (80%), Daloa (62%), and one borehole in Daoukro (49%) (Fig. 4b). Among these sampling points, the three sites of Aboisso, Soubré and Daloa are in regions identified by the CNRA as having a high prevalence of fungal diseases.

As for the 3 other sites, we note that that of Divo (96%), due to its geographical position (5° 50' N, 5° 22' W) is under the influence of the conditions which prevail below latitude 5.5° N with a dense humid evergreen forest where conditions are favorable for the expression of fungal diseases. Brown rot, black rot, root rot and anthracnose are fungal diseases that characterize this area of Divo (Koua et al., 2018). As for Biankouma, the area has the particularity of being in an equally humid montane evergreen forest (Kouamé and Bi Irié, 2010). The last site of this group, i.e. Daoukro (49%), is on the southern border of the Iffou region, bordering the Indénie-Djuablin region which was identified by the CNRA as having a prevalence of diseases.

Insecticides (36%) are in equal proportion to fungicides (36%) in the San Pedro region. It is indicated by the CCC and the CNRA that all regions of the production basin are concerned by the fight against insect pests. The spreading of insecticides on this cocoa plantation in San Pedro could be done to remedy infestations which cause their presence in the groundwater. On the other hand, San Pedro is in the wettest part of the production basin.

Herbicides are only most represented on the Alépé site (58%). Chemical weeding is increasingly practiced. It is practiced because it is not possible to carry out mechanical (manual) weeding. The lack of labor is one of the causes. Indeed, we are seeing an aging of cocoa farmers and the non-generational renewal which is one of the challenges that producers face (Ruf et al., 2020). Chemical weeding is therefore an alternative which has the corollary of generating the residues observed on this site.

Soils

All the soils of the 10 plantations showed the presence of residues. They vary from one site to another. This variability concerns both the molecules and the numbers of residues, but also the quantified concentrations. It is the plantations in the southern half of the basin which have the highest numbers of residues compared to the rest of the production basin, namely Aboisso > San Pedro, Soubré>Alépé> Divo.

Thus, out of ten sampling sites, six show a predominance of fungicides in regions identified by the CNRA as having significant prevalence of fungal diseases. These are respectively the regions of Sud-Comoé, Haut-Sassandra, Nawa and San Pedro (CCC, 2013). In addition to these four regions, there is the site of Alépé (78%) which is below latitude 5.5°N in the last southern half of the production basin, with a dense humid evergreen forest offering the conditions of expression of fungal diseases. On the other hand, the Biankouma area has the particularity of being located at the northern limit of the production basin, but in an equally humid montane evergreen forest (F. Kouamé and Bi Irié, 2010) (Fig. 4c).

On the other four sites that are Daoukro, Divo, Dueukoué and Gagnoa, insecticides are found respectively in proportions of 100%, 86%, 82% and 56%. The CCC and the CNRA indicate that all regions of the production basin are concerned by the fight against insect pests and as such, the spreading of insecticides on these plantations is carried out in order to remedy the infestations.

Herbicides in these soils are only represented in small proportions, such as in San Pedro (14%), Aboisso (6%), Soubré (5%), Biankouma (4%), Alépé (2%).

Finally, by comparing the results of analyses on soils to these of waters it is noted that like in the water matrix, fungicides are the majority of residues in soils.

Cocoa beans

It is known that soils and water interact with plants, by serving as a support and reservoir of water and nutrients, but also that plants have reciprocal effects on soil formation, physical structure and activities of soil biota (Ehrenfeld, 2013). As a result, molecules in soil and water are subject to the same

interactions. They have properties that will determine their transfers into plants from soil and water.

In cocoa beans, fungicides are predominant on 6 plantations: Aboisso >Alépé> Soubré> Gagnoa > San Pedro> Daoukro (Fig. 4d), among them Pyraclostrobin alone represents 80% of the total quantities of contaminants present in beans. It is noted that the Alépé site, where fungicides represents 98% of the total contaminants, is below latitude 5.5°N in the last southern half of the production basin. This is the part of the basin with dense humid evergreen forest providing the conditions for the expression of fungal diseases. For the Gagnoa area (76%), a high disease pressure has been identified by previous authors (N'guessan, 2018). The last site Daoukro (62%), is on the southern border of the Iffou region, bordering the Indénié-Djuablin region which was identified by the CNRA as having a prevalence of fungal diseases.

At the Biankouma and Daloa sites, herbicides are the main residues in cocoa. Daloa is one of the former foci of swollen shoot disease or CSSD (Cocoa swollen shoot disease) (Kouakou, 2014). Biankouma in the Tonkpi region is one of the new foci of CSSD infection (Aka et al., 2020). To combat this disease, it is necessary to eliminate weeds by alternating manual weeding and chemical weeding (at least four passes per year). For trees, the recommended technique is poisoning 1.5 meters from the ground, 30 cm wide, removing the bark down to the wood and applying a total herbicide with a brush. If the tree is not dead a few weeks later, repeat the operation at least 30 cm higher using another total herbicide (Kébé et al., 2016).

These successive uses of herbicides on this site can lead to the pesticide levels observed in cocoa beans.

Compliance with regulatory thresholds

Surface water and groundwater

The analysis of Fig. 5a shows that, pyraclostrobin is by far the active substance with the highest residue level ($0.1 \mu\text{g L}^{-1}$ at Aboisso). It is followed in decreasing order by lambda- cyalothrin ($0.03 \mu\text{g L}^{-1}$ at Soubré) and deltamethrin ($0.014 \mu\text{g L}^{-1}$ at Alepe). The detection of a banned molecule such as bifenthrin ($0.01 \mu\text{g L}^{-1}$ at Gagnoa) raises the problem of contradictions in the application of regulations on pesticides and the need for harmonization between countries producing active ingredients and importing countries. Indeed, bifenthrin is prohibited in accordance with Decision No. 2009/887/EC of 30/11/09 concerning the non-inclusion of bifenthrin in Annex I of Council Directive 91/414/EEC and the withdrawal of authorizations of plant protection products containing this substance in the European Union (EU). This active ingredient is suspected of being carcinogenic (EFSA, 2011). However, different sources tend to show that such active substances continue to be exported to other countries outside the European Union, which fuels their use in these countries (Alliot, 2024; Public Eye, 2018; Tostado & Bollmohr, 2023; Van Scharen & Camargo, 2024). This is the case in Ivory Coast where this molecule is currently on the official list of active ingredients approved for cocoa cultivation. However, the main destination market for Ivorian cocoa is indeed the European Union, which leads to say that there is a "boomerang-residues of banned chemicals returning home" (Van Scharen & Camargo, 2024).

However, despite prohibited molecules, we note that all the residue levels for the different molecules are below the regulatory threshold except on the Aboisso site where pyraclostrobin is at the limit of the regulatory threshold. Indeed, the residue levels of the different quantified molecules were compared individually to the regulatory threshold in accordance with the decree of October 9, 2023 amending the decree of December 17, 2008. It sets for the active substances of pesticides, as well as the relevant metabolites and degradation and reaction products at $0.1 \mu\text{g L}^{-1}$ per molecule and $0.5 \mu\text{g L}^{-1}$ for total molecules (Ministère de la santé et des solidarités, 2023, 2024).

For groundwaters, the total pesticides per site are all below the reference threshold of $0.5 \mu\text{g L}^{-1}$. However, when comparing the residual levels of the different molecules with the regulatory threshold of $0.1 \mu\text{g L}^{-1}$, we observe that this threshold is exceeded for pyraclostrobin. At Divo its concentration is $0.21 \mu\text{g L}^{-1}$ and at Soubré $0.12 \mu\text{g L}^{-1}$. Regarding the pyraclostrobin concentration in surface water, it can be noted that in Divo, it is $0.1 \mu\text{g L}^{-1}$ and in Soubré $0.021 \mu\text{g L}^{-1}$. These values confirm the explanations concerning the pyraclostrobin frequency of detection, namely that this molecule tends to be leached into groundwater rather than runoff into surface water. The intrinsic characteristics of the molecule (DT = 33 days in soils, Log Koc = 3.97, solubility = 1.9 mg L^{-1} and lipophilicity) combined with the physicochemical conditions of the environment (low organic matter content in ferrallitic soils and abundant rainfall) allow a transfer of pyraclostrobin from soils to groundwater.

In addition to that, fungicides are the main pesticides contaminating both groundwater and surface water, followed by insecticides and herbicides in order of importance of the concentrations observed.

Cocoa beans

Compliance of residue levels is checked by comparing them to MRLs (maximum residue limits). MRLs are defined by the Codex Alimentarius as the maximum residue limit (MRL) of a pesticide in or on a product intended for human or animal consumption, i.e. the maximum legally tolerated concentration of residues of the pesticide in question when used correctly, in accordance with good agricultural practices (FAO & WHO, 2024).

MRLs are established by the Codex Committee on Pesticide Residues (CCPR), based on recommendations made by the Joint FAO/WHO Meeting on Pesticide Residues (JMPR). MRL values can be found in the "Codex Online Database of Pesticide Residues in Foods", which defines over 4,300 maximum residue limits covering around 200 pesticides.

According to Regulation (EC) No 396/2005, all food intended for human or animal consumption in the European Union (EU) is subject to a maximum residue limit (MRL) of pesticides to protect animal health and human. European legislation regulates the MRLs that apply to different products and establishes a "default MRL" when no specific MRL exists.

The MRLs, set by the European Commission, include MRLs specific to certain products intended for human or animal consumption and a general MRL that applies when no specific MRL has been defined (a “default limit” of 0.01 mg/kg) (EUR-Lex, 2023).

For cocoa, MRLs are determined on “beans after removal of hulls”, as indicated in EC Regulation 178/2006. However, in some countries, MRLs are determined on whole beans (i.e. before removal of the shell or seed coat). This shell should not be confused with the rigid pod which encloses the beans. It is therefore the thin layer that coats the almond inside the bean. The present study followed this protocol, and the seed coats/skins were removed from the dried beans (see the materials and methods chapter).

In addition to the legislation, the International Cocoa Organization (ICCO) has published a list of strategic pesticides used in the production and conservation of cocoa. This document gives the current MRLs, as well as pesticides that “should not” be used in cocoa.(Bateman & Crozier, 2023).

In the present study, the MRLs on cocoa beans were obtained from the ICCO publication (Bateman & Crozier, 2023), but also by consulting the European Commission website and the FAO/WHO Codex Alimentarius. The results of the analyzes carried out on cocoa from the different plantations visited are shown in the Fig. 5b with the default MRL of 0.01 mg/kg.

Comparison of the analytical results with the MRL of 0.01 mg/Kg shows systematic exceedances on almost all plantations for pyraclostrobin.

In addition to this general analysis with the default MRL, the MRLs specific to each of the molecules were compared to the sample results. The two (2) findings of excess are presented in Table 5.

Table 5 Exceeding MRLs in cocoa			
Plantation sites	Active ingredients	Analytical results (mg/kg)	MRL (mg/kg)
Alépé	Pyraclostrobin	0.18	0.1
Soubré	Pyraclostrobin	0.16	0.1

Conclusion

This study showed that all four matrices (cocoa, soil, surface water and groundwater) present contamination by pesticide residues. Likewise, all the 10 sites studied are contaminated. In all regions of the production basin, most pesticide residues detected are fungicides, which reflects that they are the most used. This result is consistent with the findings of the ICCO (International Cocoa Organization) (Bateman, 2009; Bateman & Crozier, 2023) which states that fungicides are the most commonly used pesticides in primary cocoa production. It is the same point of view for Traoré & Haggblade, (2017) who indicate their important use for the cultivation of cocoa, but also Coulibaly et al., (2017) who underline that losses can reach 60% in certain regions if no phytosanitary protection is ensured.

Fungicides are followed by insecticides and herbicides. This result reflects the phytosanitary constraints currently facing cocoa cultivation in Ivory Coast. Among these threats, insect pests are of concern and chemical control measures are for the entire production area. (CCC, 2013). And since 2016, an invasion of phytophagous caterpillars (which eat leaves) has been observed in plantations. This is *Achaea catocaloides*, which today constitutes a new threat to Ivorian cocoa farming (N'guessan et al., 2022). The use of insecticides is therefore practiced for protecting production against these threats.

Herbicides are the less quantified. Their use is becoming more and more necessary. Particularly for the fight against CSSVD (cocoa swollen shoot virus disease) as an “arboricide” against cocoa trees affected by this virus.

By comparing the results obtained with other studies in Cote d'Ivoire and elsewhere, we note that in Cote d'Ivoire, in the Dohou and Guessabo lakes, the substances frequently encountered are herbicides, contrary to the results of the present study (Kouamé, 2021). But they indicate that these lakes are bordered by more diverse crops such as market gardens, rubber trees, rice, corn, banana trees (plantain), coffee trees, in addition to cocoa trees, which differentiates its study context. In another study on the Sassandra River in the Guéssabo region, involving 5 insecticides (imidacloprid, deltamethrin, lambda-cyhalothrin, cypermethrin, acetamiprid), concentrations are 0.0052, 0.0055, 0.635, 0.0129 and 0.0109 respectively $\mu\text{g L}^{-1}$ (K. V. Kouamé et al., 2023). These values are in the same orders of magnitude as the average values for deltamethrin, lambda-cyhalothrin and cypermethrin in the surface waters of the 10 sites studied here. In their analysis, they found that the Sassandra River is contaminated mainly by pyrethroids. On the other hand, for Atto & Monde, (2020), on Lake Adaou, the average of substances detected is $79.07 \mu\text{g L}^{-1}$. It is above the European Union guideline values ($5 \mu\text{g L}^{-1}$) and the values of the present study. They explain it by the water leaching from agricultural land from rubber, coffee and cocoa plantations and the pesticides volatilized after application.

On cocoa beans Biego et al., (2009) in a study relating only to a few organochlorines, showed the presence of traces of residues in cocoa beans collected at the ports of San Pedro and Abidjan as well as in the Agboville region. Among the 15 pesticides detected, 13 were banned for use in Ivory Coast and only one had conditional approval in 2009.

On the soils in Côte d'Ivoire, in the absence of specific studies on the country's cocoa growing lands, it should be noted that in neighboring Ghana, Fosu-Mensah et al., (2016a, b) found residues of organophosphates varying from 0.01 mg/kg to 0.04 mg/kg, with an average of $0.03 \pm 0.01 \text{ mg/kg}$ and organochlorines varying from 0.005 to 0.05 mg/kg, dieldrin being the most common. In Nigeria, in Ondo State (main cocoa growing region of the country), Aiyesanmi & Idowu, (2012), found organochlorines in the soils of cocoa plantations at variable concentrations including Endosulfan I and Endosulfan II, the most common with high concentrations of 350.10 mg/kg and 3.55 mg/kg respectively, in addition to heptachlor, heptachlor epoxide, aldrin, dieldrin, benzene hexachloride isomers. These organochlorines were gradually banned between 1980 and 2009 (Balestier et al., 2021).

The study located in a tropical context also made it possible to conclude that the transfers of residues occurring in the different matrices (cocoa, soils, surface water, groundwater) are a function of the physicochemical properties of the molecules, the properties of soils with a ferrallitic dominance, abundant rainfall, the management practices of phytosanitary products and the cultivation practices specific to cocoa farmers. The residues can be absorbed from the soil and reach by translocation the different parts of cocoa trees and associated crops.

Thus, this study contributed to showing that the use of pesticides for cocoa cultivation is not without risks and consequences for the environment, with public health issues both locally in Côte d'Ivoire, but also beyond national borders for importing markets. It is therefore important that a plan is designed to reduce uses, promote alternatives and put in place a mechanism for the control and regular monitoring of these residues (Traoré, 2018). At the end of the 2QC program and the chemical control measures applied as emergency measures, the authorities and the Coffee and Cocoa Council (CCC) should conduct such a reflection on the uses of pesticides and assess the impacts in the different compartments of the production basin, but also on cocoa farmers.

Declarations

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Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author contributions

Adama Soro, Maurice Millet and Gaetana Quaranta contributed to the study conception and design. Material preparation and sampling were performed by Adama Soro. Analysis were performed by Adama Soro, Sylvia Georg, Catherine Galindo and Maurice Millet. The first draft of the manuscript was written by Adama Soro, Maurice Millet and Gaetana Quaranta. All authors commented on previous versions of the manuscript and approved the last version.

Ethical approval

This is not applicable

Consent to participate

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data in the manuscript

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Figures

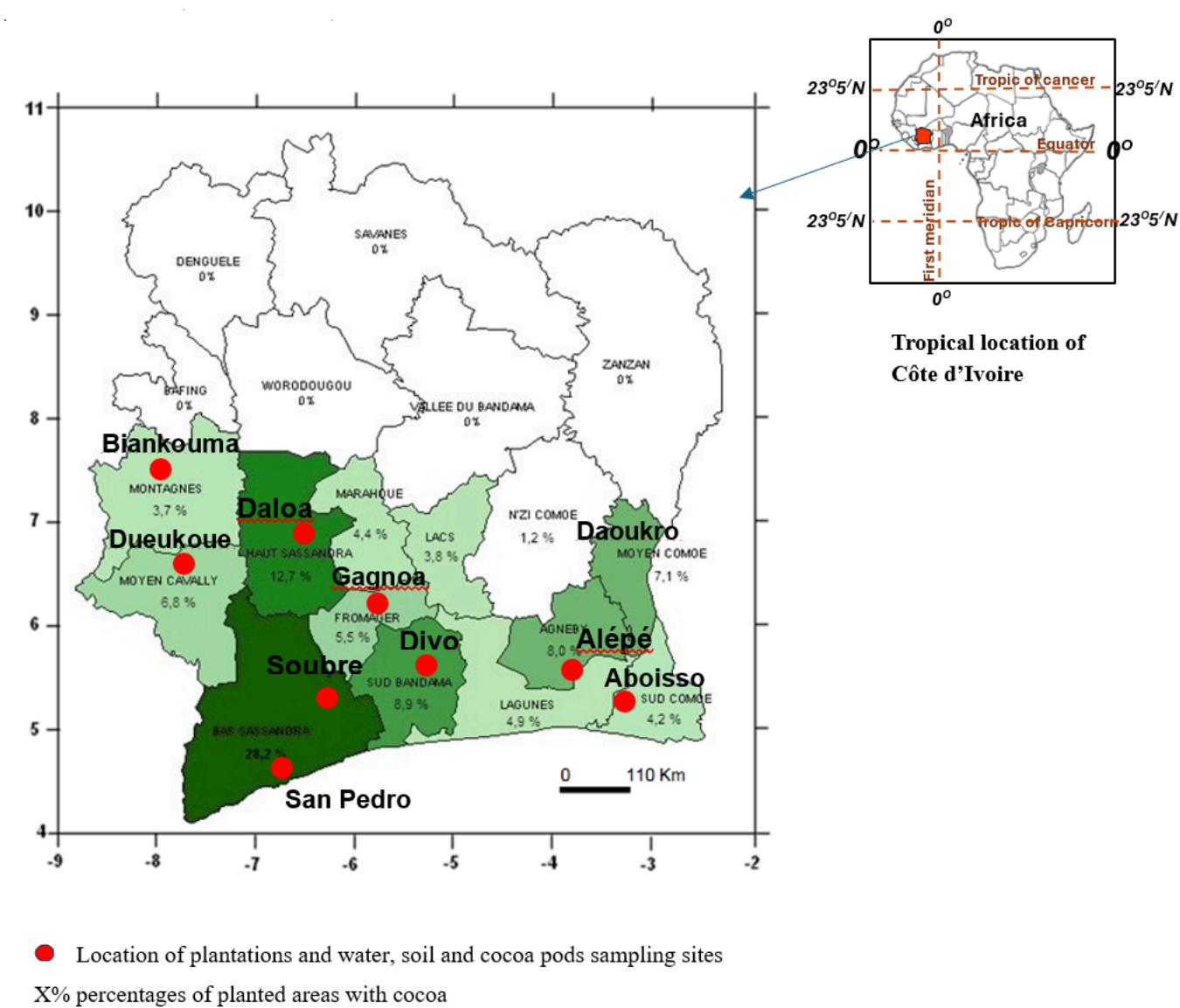


Figure 1
Delimitation of the cocoa growing zone (Direction de l'agriculture, 2003)

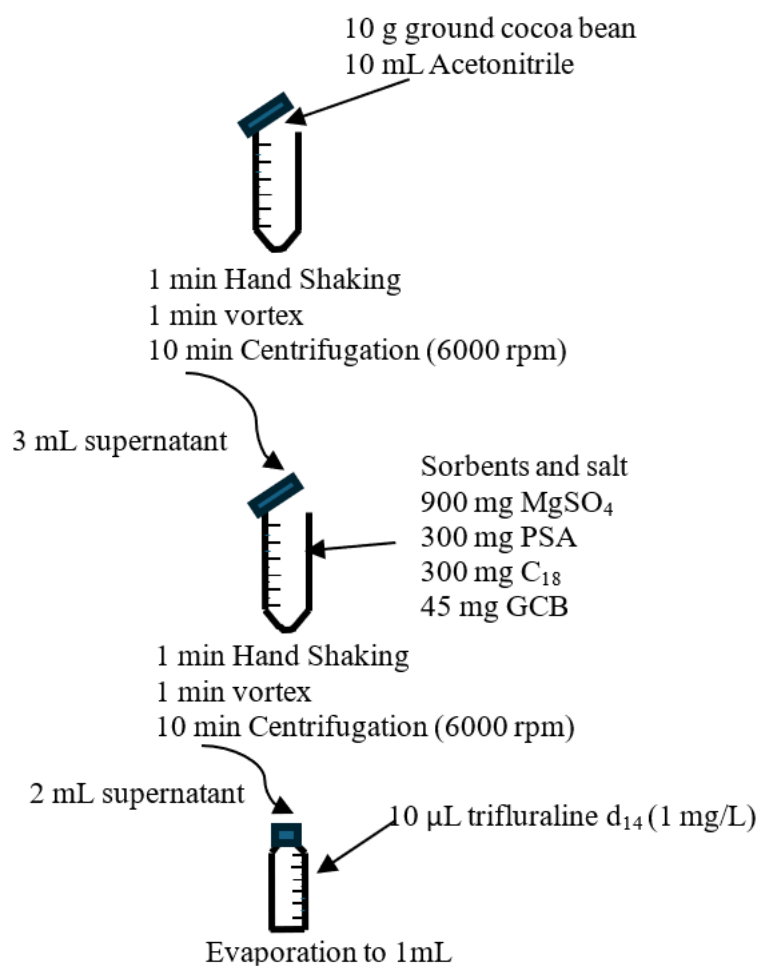
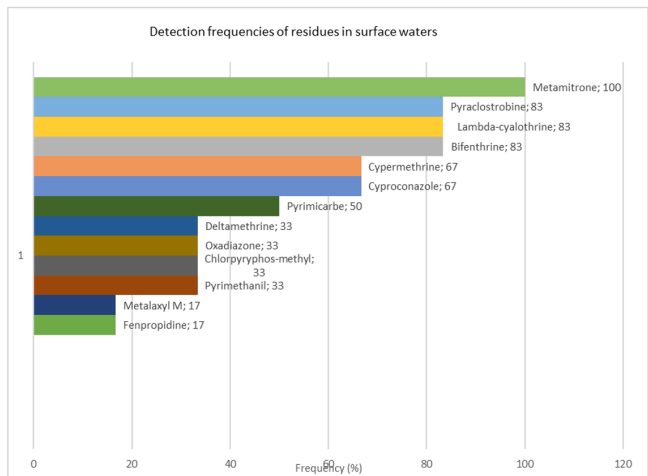
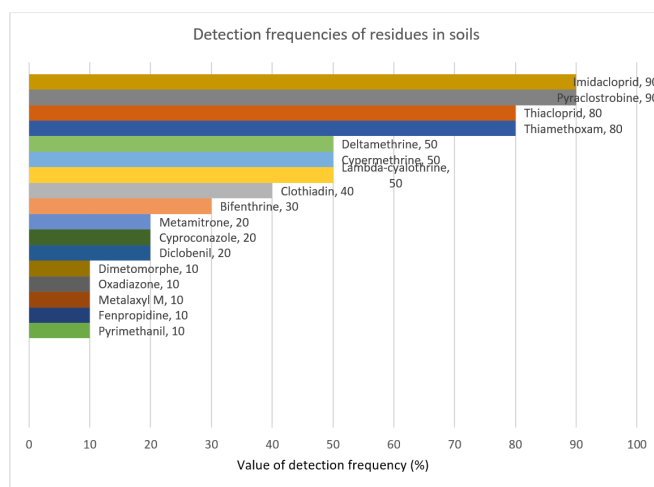


Figure 2

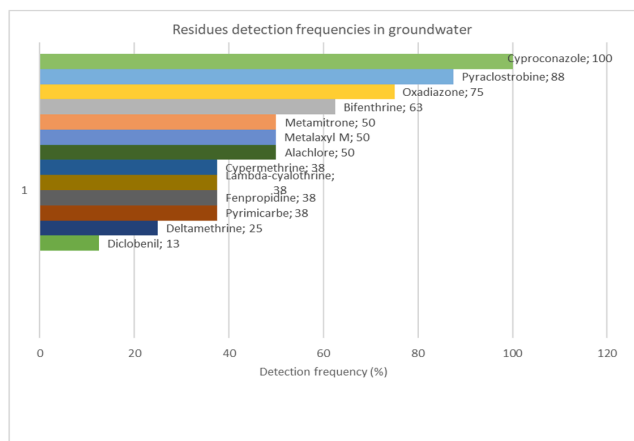
Descriptive diagram of the cocoa extraction method by QuEChERS



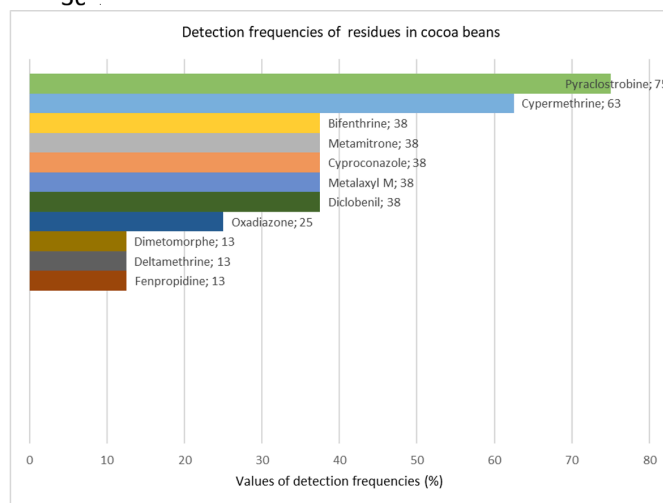
3a



3c



3b



3d

Figure 3

3a: Classification of active materials according to their detection frequencies in surface waters

3b: Classification of active materials according to detection frequencies in groundwater

3c: Classification of pesticides according to detection frequencies in soil

3d: Ranking of residue detection frequencies in cocoa

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Figure 4

4a Spatial distribution of pesticide families in surface waters

4b Spatial distribution of pesticide families in groundwater

4c Spatial distribution of pesticide families of soils

4d Spatial distribution of pesticide families in cocoa beans

Image not available with this version

Figure 5

5a Compliance of surface water residue levels with regulatory thresholds

5b Comparison of concentrations with MRLs for cocoa beans