Advances in Materials Physics and Chemistry, 2024, 14, 25-46
https://www.scirp.org/journal/ampc
ISSN Online: 2162-5328
ISSN Print: 2162-531X

Evaluation of Agricultural Contamination Level by Heavy Metals and Pesticides in Sediments of Rivers and Water Bodies for Shrimp Fishing in the Lower Ouémé: Case of the Aguigadji, Ahlan and Sele Stations in Benin

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How to cite this paper: Orou Goura, S., Gouissi, F.M., Hounkpatin, A.S.Y., Yessoufou, W.B., Biaou, T.S. and Fassinou, N.M. (2024) Evaluation of Agricultural Contamination Level by Heavy Metals and Pesticides in Sediments of Rivers and Water Bodies for Shrimp Fishing in the Lower Ouémé: Case of the Aguigadji, Ahlan and Sele Stations in Benin. *Advances in Materials Physics and Chemistry*, **14**, 25-46.
https://doi.org/10.4236/ampc.2024.142003

Received: January 15, 2023
Accepted: February 2, 2024
Published: February 5, 2024

Abstract

This study assessed sediment contamination by heavy metals and pesticide active ingredients linked to chemical inputs used in agricultural activities in the lower Ouémé. Pesticide residues from the organochlorine, pyrethroid and organophosphorus families were investigated by gas chromatography, and heavy metals (Cd, Pb, As, Ni, Zn, Fe, Mg, Cr and Hg) by atomic absorption spectrophotometry. The metallic pollution indices, the contamination factor (CF) and the ecological risk index were calculated. The results revealed 8 active ingredients in the rainy season and 9 in the dry season. Glyphosate was the active ingredient with the highest concentration at all stations, 9.65 ± 0.84 mg/kg recorded in the dry season at the Aguigadji station. All glyphosate values were above the EQS. DDT, Atrazine and Endosulfan also showed high concentrations in the dry and rainy seasons. Emamectin, Abamectin and Lambda Cyhalothrin also showed high concentrations in the dry season at Aguigadji, Ahlan and Sele. Only glyphosate was recorded at the control station (Toho), but in very low concentrations. Lead showed the highest concentrations at all the stations, 265.96 ± 21.02 mg/Kg in the rainy season and 255.38 ± 79.09 mg/Kg in the dry season, all detected at the Aguigadji station and above the EQS. Zn, Ni, Fe, Cu and Cr were all representative in both the dry and rainy seasons at the contaminated stations. Manganese showed high concentrations in the rainy season. Pb showed very high contamination (FC

≥ 6) at the Aguigadji and Ahlan stations and significant contamination (3 ≤ FC < 6) at the Sele station in both the rainy and dry seasons. Ni, Hg and Cd, showed either moderate or significant contamination at the contaminated stations. The risk values showed a considerable ecological Ri (190 ≤ Ri < 380) in the rainy season and a moderate ecological Ri (95 ≤ Ri < 190) in the dry season at these contaminated stations.

Keywords

Pesticides, Heavy Metals, Sediments, Shrimp Fishing Area, Lower Ouémé

1. Introduction

In rural areas, the intensive use of fertilisers and pesticides associated with the intensification of agricultural activities is the main source of heavy metal and pesticide residue contamination of aquatic environments [1] [2] [3] [4]. This is the case in the lower Ouémé region, where large quantities of mineral fertilisers and chemical pesticides are used to satisfy the ever-increasing demand for market garden produce and food crops. Given the intensity of farming activities around watercourses, the latter are faced with various pollution problems, the main one being agricultural pollution from the inputs used. In fact, after phytosanitary use, these chemical inputs are dispersed in various environmental matrices, including the sediments of aquatic environments, where they accumulate. Sediments are therefore reservoirs for metals and pesticides in the environment. Even at very low concentrations, some pesticides have toxic properties [5]. According to Benguedda-rahal in 2012, the study of sediments is of interest because they are indicators of contamination in aquatic environments, due to their ability to bind pollutants, especially trace elements [6]. They are therefore of particular interest when it comes to taking appropriate measures for the sustainable management of an aquatic ecosystem [7]. In Benin, studies have been carried out to assess the level of contamination of sediments by pesticides and heavy metals. No study has been conducted on assessing the pollution levels of aquatic ecosystem compartments in the Lower Ouémé area on rivers and water bodies where shrimp fishing is practiced. The aim of this study is therefore to evaluate the levels of contamination of sediments in shrimp fishing areas in the lower Ouémé. The aim is to provide recent data on pesticide residues and metal elements levels in sediments, with a view to the sustainable management of aquatic ecosystems in the study area.

2. Material and Methods

2.1. Study Area

This study was carried out in the lower Ouémé basin in the districts of Kétou, Zagnanado and Ouinhi. This area extends between 8˚15' - 6˚33' north latitude and 2˚00' - 1˚50' east longitude (**Figure 1**).

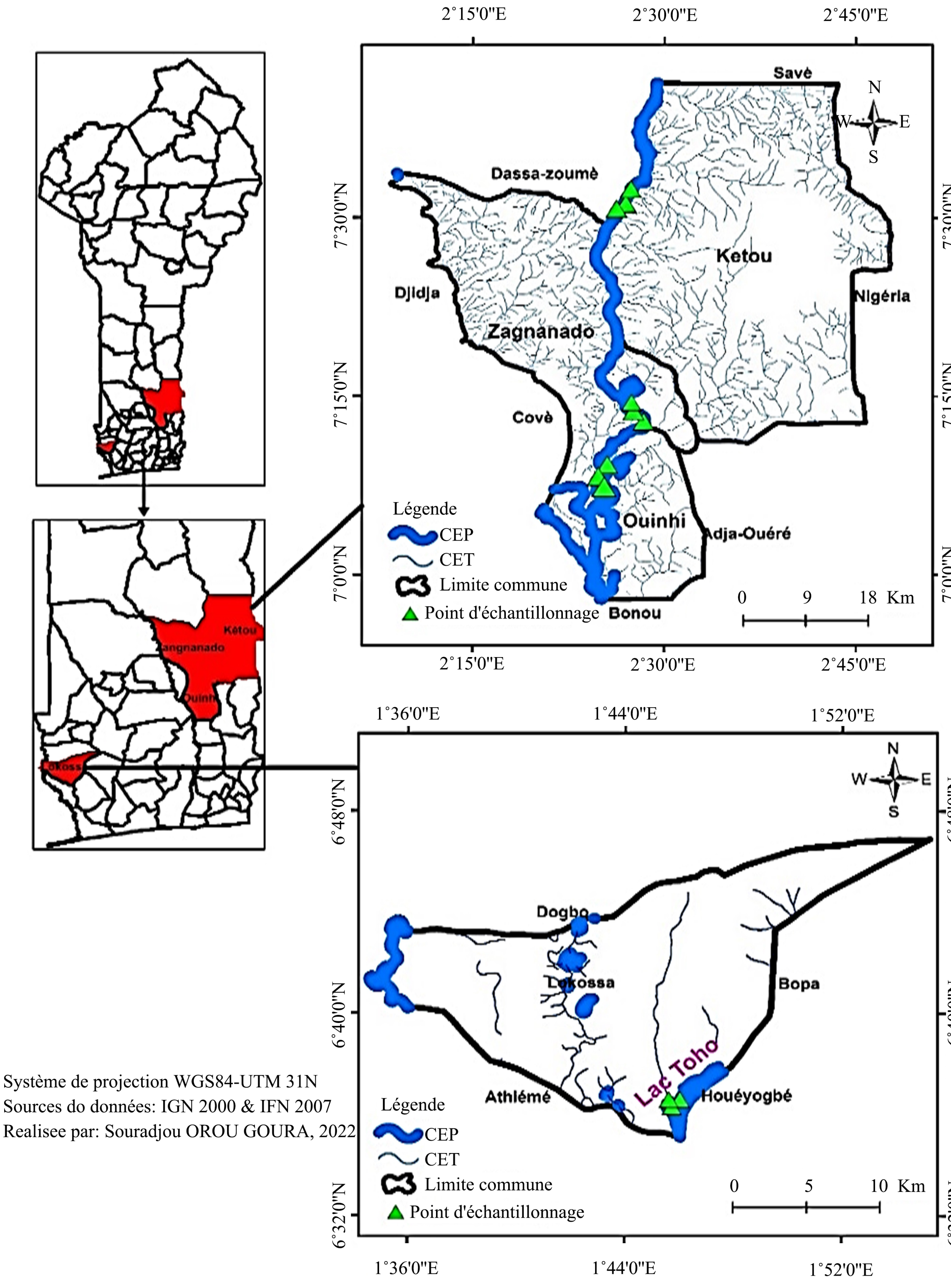


Figure 1. The map of the study area showing sampling stations.

The lower Ouémé basin is the weir of the upper Ouémé upstream [8]. This region covers the districts of Kétou, Zagnanado and Ouinhi. These three communes belong to different agro-ecological zones but share the same Sudano-Guinean climate (Table 1).

The first 3 study stations (Aguigadji, Ahlan, Sele) were chosen primarily because of their accessibility, the intensive farming activities carried out mainly along rivers and bodies of water and, above all, because they are shrimp fishing areas. As for the Toho station, the choice was based on its agricultural activities and its shrimp fishing activities. It is considered the reference station in this study.

2.2. Sediment Sampling

Two series of samples and measurements were taken, one over three months (May-June-July) in the wet season of 2021 and the other over three months (January-February-March) in the dry season of 2022. Following standard procedures, sediments were collected from the top five (5) centimetres of the benthos and packaged in aluminium foil lined with a plastic bag. For each sample, three (3) sediment samples were taken at three different locations, one (01) in the middle and the other two (02) moving away towards the banks of the watercourses. The samples were then mixed to form a single composite sample of 500 g of sediment for each month at each station. At each sampling station, one sample was taken for pesticides and another for heavy metals. These samples were then automatically labelled and stored at 4˚C with ice in a cooler before being transported to the various laboratories. So each month we had four (4) sediment samples for each group of parameters to be analyzed, making a total of twelve (12) samples in three (3) months. A total of 24 samples (12 in the dry season and 12 in the rainy season) were therefore collected for pesticide measurements and a further 24 for heavy metal measurements. On each occasion, the samples were transported to the GSA (Ghana Standard Authority) in Ghana, to the pesticide analysis laboratory and the general chemistry laboratory respectively, where they were coded before being placed in a refrigerator.

2.3. Methods for Analyzing Pesticide Residues and Heavy Metals

Multi-residue analyses of pesticides and heavy metals were carried out at the GSA (Ghana Standard Authority) national laboratory to determine the levels in sediments in the so-called polluted zones and the control zone.

Table 1. Coordinates of sampling stations in shrimp fishing areas.

Stations	GPS	
Aguigadji	7˚28'60"N	2˚25'60"E
Ahlan	7˚12'55"N	2˚28'12"E
Sele	7˚09'23"N	2˚25'34"E
Toho	6˚36'00"N	1˚46'60"E

2.4. Analysis of Pesticide Residues

The equipment used was a Varian CP-3800 GC-ECD gas chromatograph with a CombiPAL autosampler. The analyses began with an extraction. This consisted of weighing 10.0 g ± 0.1 g of the crushed homogeneous sediment sample into a 100 ml separation flask. Add 10 ml of acetonitrile, cap the flask and sonicate for 2 min. Then add 10 ml acetonitrile, cap the flask and place it on a horizontal mechanical shaker and shake continuously for 30 minutes. The mixture is left to stand for 10 minutes to separate the layers. Pipette a 10 ml aliquot of the organic phase (upper layer) into a 50 ml round-bottomed flask. Evaporate to approximately 2 ml to purify the extract. Next comes the extra purification step. This involves packing a "Silica" cartridge (1000 mg/6ml), which has a 1 cm thick layer of anhydrous magnesium sulphate on top, with (10 ± 0.2 ml) acetonitrile and loading the extract onto the cartridge. The eluent is collected in a 50 ml pyriform flask. The column is eluted with 20 ml acetonitrile and the eluent collected is concentrated to dryness using the rotary evaporator at less than 40˚C. Redissolve in ethyl acetate by adding 1 ml by pipetting. Add 20 µl of 1% polyethylene glycol in ethyl acetate (v/v). The extract is transferred to a 2 ml standard opening vial prior to quantification by GC-ECD and GC-PFPD.

2.5. Analysis of Heavy Metals

Metals were measured by Atomic Absorption Spectrometry (AAS) using two spectrometric methods: one is ISO 8288 for Mercury (Hg), Cadmium (Cd), Lead (Pb), Copper (Cu), Chromium (Cr), Zinc (Zn), Arsenic (As) and Nickel (Ni) and the other is ISO 6332 for the detection of Iron (Fe) in particular.

2.6. Statistical Analysis Methods

2.6.1. Pesticide Data Analysis

The raw pesticide data on sediments in both the dry and wet seasons were entered into tables in Microsoft Office EXCEL 2013 and subjected to R software for the calculation of averages and numerical classification, and to Principal Component Analysis (PCA) in order to establish correlations between the various parameters and the stations, taking the seasons into account.

2.6.2. Heavy Metal Data Analysis

Raw data on heavy metal content in sediments in both dry and wet seasons were entered into tables in Microsoft Office EXCEL 2013 and submitted to R software to calculate averages and produce graphs. Contamination factors (CF) and ecological risk indices (Ri) were determined using the respective formulae below.

2.6.3. Estimation of Contamination Intensity

To assess the intensity of trace metal contamination in the sediments, the contamination factors (CF) and the ecological risk index (Ri) were determined.

- **Contamination factors (CF) for metallic elements.**

  The contamination factor is determined by the following formula:

$$\text{CF} = \frac{\text{Concentration of the element}}{\text{References concentration}}$$

The CF values were interpreted according to Hakanson in1980 [9]:

- FC ≤ 1 indicates low contamination;
- 1 ≤ FC < 3 is moderate contamination;
- 3 ≤ FC < 6 is important contamination;
- FC ≥ 6 is very high contamination.

Table 2 shows the reference concentrations used according to Canadian standards for trace element pollution in sediments [10].

❖ **Ecological risk index (Ri)**

Established by Hakanson in 1980 [9]. This index is used to assess the ecological risk of sediments. It has been used by other authors to determine the ecological risk of contaminants such as metals in soil and sediments [11].

$$\text{Ri} = \sum_{i=1}^{i} E_r^i \quad \text{with } E_r^i = \text{Tr} \times \text{CF}$$

Ri is the ecological risk index; Tr is the toxic reaction factor; FC is the contamination factor; Er is the potential ecological risk for each metal. The toxic reaction factors (Tr) for the trace elements studied (As, Cd, Cu, Cr, Hg, Pb, and Ni) according to Hilton *et al.* in 1985 [12] are 10; 30; 5; 2; 40; 5; 6, respectively.

Table 3 shows the interpretation of the ecological risk values for sediments according to Hakanson in 1980 [9].

3. Results

3.1. Sediments Contaminations by Pesticides

In the laboratory, thirty (31) pesticide active ingredients (Aldrin, Abamectin, Acetamipride, Atrazine, Buthaclor, Chlordane-gamma, dichlorodiphenyldichloroethane (DDD), dichlorodiphényltrichloroéthane (DDT), Lambda Cyhalothrin, Cypermethrin, Chlorpryphos ethyl, Emamectin, Deltamethrin, Dieldrine,

Table 2. Canadian standard values for trace element pollution in sediments.

Metallic elements	Zn	Pd	As	Mg	Cd	Ni	Hg	Fe	Cu	Cr
SQG (CCME, 2001)	123	35	5.9	-	0.6	18	0.17	-	35.7	37.3

Table 3. Characterisation of potential ecological risk (Er) and ecological risk index (Ri).

Er value	Potential ecological risk	Ri value	Ecological risk index
Er < 40	Low	**Ri < 95**	Low
40 ≤ Er < 80	Moderate	95 ≤ Ri < 190	Moderate
80 ≤ Er < 160	Considerable	190 ≤ Ri < 380	Considerable
160 ≤ Er < 320	High	380 ≥ Ri	Higher
320 ≥ Er	Higher		

Endosulfan, Endrine, Imazethapyr, Fenthion, Glyphosate, Heptachlor, Isopropalamine, Lindane, Malathion, Monocozeb, Nicosulfiron, Oxyfluorfene, Parathion, Pendimethaline, Permethrine, Propisochlor) were researched. These active ingredients belong to the organochlorine, synthetic pyrethroid and organophosphate pesticide families.

The results of the analyses revealed eight (8) active pesticide ingredients in the sediments during the rainy season and nine (9) during the dry season. The average concentrations of pesticides found in the sediments of the lower Ouémé and Lake Toho are shown in Table 4 and Table 5. Calculations of mean concentrations showed higher values of glyphosate at all stations not only in the rainy season but also in the dry season. The highest concentration of glyphosate was 9.65 ± 0.84 µg/kg, recorded in the dry season at the Aguigadji station. This active ingredient was the only one detected at the control station (Toho) in both the

Table 4. Results of analysis of pesticide residues in sediment samples taken from the lower Ouémé and Toho river.

Detected active ingredients	Aguigadji	Ahlan	Sele	Toho
		RAINY SEASON		
Aldrin	0.02 ± 0.011	0.03 ± 0.005	nd	nd
Atrazine	0.83 ± 0.50	0.7 ± 0.78	0.19 ± 0.09	nd
Abamectin	0.12 ± 0.01	0.13 ± 0.03	0.05 ± 0.02	nd
Cyperméthrin	0.12 ± 0.06	0.06 ± 0.04	0.13 ± 0.05b	nd
DDT	3.13 ± 0.81	3.09 ± 1.63	0.08 ± 0.01	nd
Emamectin	0.002 ± 0.001	0.08 ± 0.3	0.10 ± 0.05	nd
Endosulfan	3.07 ± 0.91	2.07 ± 0.73	0.38 ± 0.49	nd
Glyphosate	6.98 ± 1.85	6.36 ± 1.32	4.19 ± 2.08	0.003 ± 0.002
Lambda Cyhalothrin	0.057 ± 3.06	0.35 ± 0.20	0.053 ± 0.024	nd
		DRY SEASON		
Aldrin	0.41 ± 0.28	0.28 ± 0.06	nd	nd
Abamectin	1.26 ± 0.61	1.03 ± 0.66	2.16 ± 0.53	nd
Butachlor	0.18 ± 0.15	0.1 ± 0.07	0.03 ± 0.02	nd
Cyperméthrin	0.11 ± 0.06	0.05 ± 0.03	0.6 ± 0.21	nd
Emamectin	0.93 ± 0.52	1.47 ± 0.97	1.86 ± 0.49	nd
Atrazine	2.76 ± 2.33	2.33 ± 0.46	0.53 ± 0.47	nd
DDT	2.13 ± 0.82	4.43 ± 1.70	1.38 ± 0.43	nd
Lambda-Cyhalothrin	2.7 ± 3.68	1.05 ± 0.09	3.38 ± 1.26	nd
Glyphosate	9.65 ± 0.84	7.36 ± 1.32	6.86 ± 0.54	0.0003 ± 0.0005
Endosulfan	4.07 ± 0.90	3.40 ± 0.51	1.05 ± 0.03	nd

nd: no detected.

Table 5. Environmental quality standards (EQS) accepted for some pesticides in freshwater sediments.

Active ingredients	Environmental quality standards (mg/kg)
Aldrin	0.08
Atrazine	-
Abamectine	-
Butachlor	-
Cyperméthrin	-
DDT	0.00477
Emamectin	-
Endosulfan	0.006
Glyphosate	0.5
Lambda-Cyhalothrin	-

dry and rainy seasons, and at very low concentrations (**Figure 2**). In the rainy season, DDT, Atrazine and Endosulfan were also more representative at the Aguigadji and Ahlan stations, almost non-existent at the Sele station and not detected at the Toho station (control). In the dry season, these active ingredients also showed high concentrations at the Aguigadji, Ahlan and Sele stations (so-called contaminated stations) and none at the Toho station (control). Emamectin, Abamectin and Lambda Cyhalothrin also showed remarkably high concentrations in the dry season at the so-called contaminated stations. In this study, Environmental Quality Standards (EQS) were used to assess the ecological status of the water. The concentrations of most of the pesticides detected are well above the EQS. The highest average value for this campaign was 9.65 ± 0.84 mg/kg for glyphosate, obtained in the dry season at the Aguigadji station. The highest value recorded in the rainy season was 6.98 ± 1.85 mg/kg. All these values are above the EQS set at 0.5 mg/kg. Concentrations of this active ingredient exceeded the EQS at all the so-called contaminated stations in both the dry and wet seasons.

The concentration of glyphosate obtained at the control station remained below the EQS in both the dry and wet seasons. The other molecules (DDT, Endosulfan and Aldrin) also showed concentrations above the EQS. From all these results we can say that the sediments of the lower Ouémé have a poor ecological status. **Figure 3** shows the results of the principal component analysis (PCA). This analysis enabled us to examine the correlation between the variables and also to classify the stations according to season. The Dim 1 dimension contributed 46.94% of the inertia and the Dim 2 dimension 17.14%. The two dimensions Dim 1 and Dim 2 give a total contribution to the information of 64.08%. The Dim 1 dimension is defined on the positive side by glyphosate, butachlor, aldrin, DDT, endosulfan and atrazine. Dim 2 is characterised by cypermethrin, abamectin, lambda cyhalothrin and emamectin (**Figure 3(b)**). Four groups were thus highlighted by the sediment graph (**Figure 3(a)**).

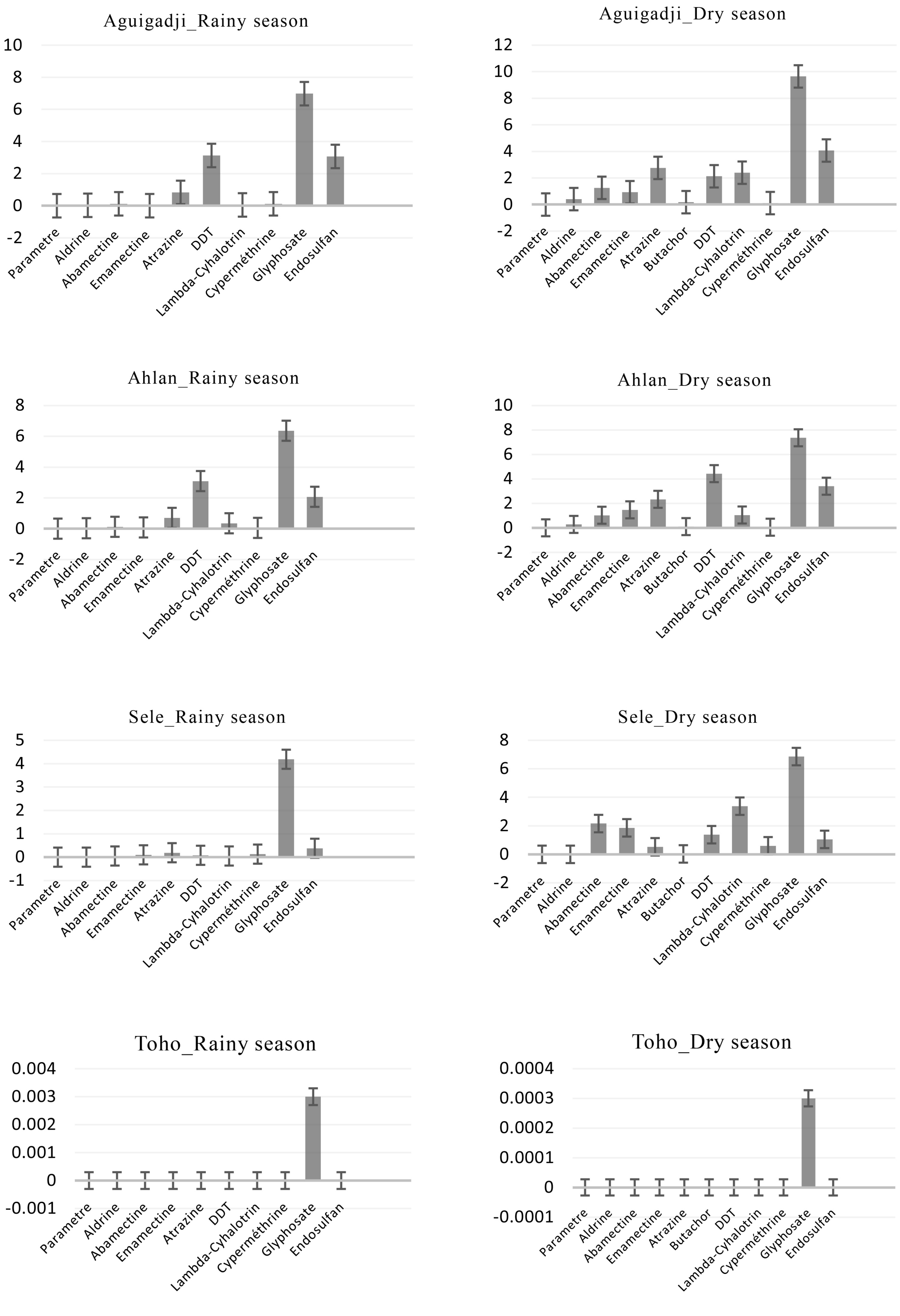
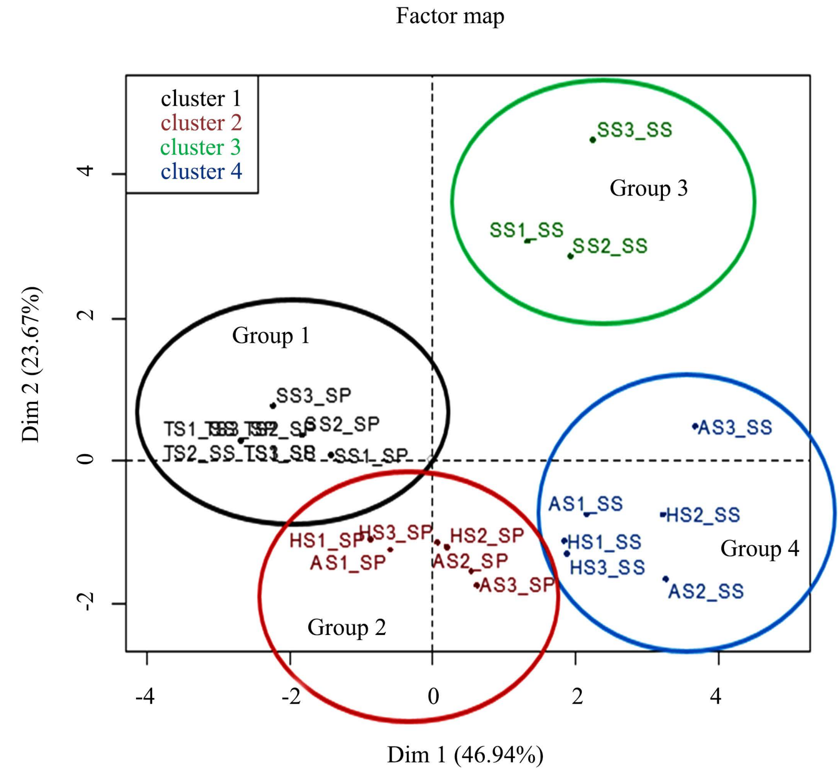
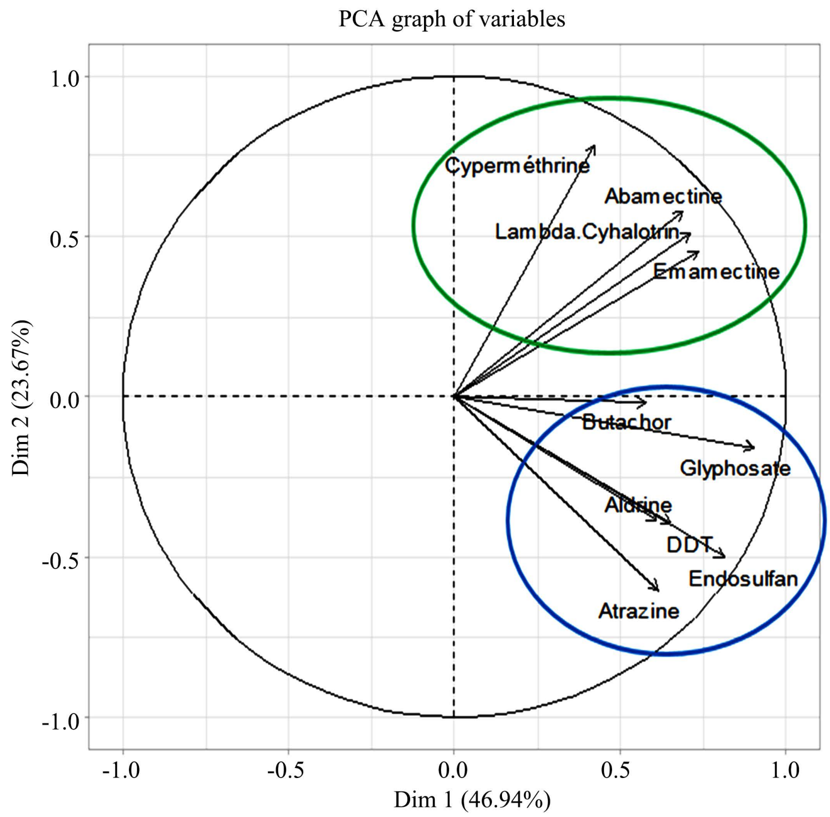


Figure 2. Seasonal variation in concentration of pesticides residues in sediements.



(a)



(b)

Figure 3. (a) and (b): Results of Principal Component Analysis (PCA).

- Group 1 is characterised by stations where samples are very weakly correlated with all pesticide active ingredients. In particular, this mainly concerns samples taken at the control station (Toho) in the dry and rainy seasons and also at the Sele station in the rainy season.
- Group 2 is characterised by stations that are weakly correlated with glyphosate, butachlor, aldrin, DDT, endosulfan and atrazine. These stations are Aguigadji and Ahlan, mainly in the rainy season.
- Group 3 is characterised solely by dry-season sampling at the Sele station, which is strongly correlated with cypermethrin, abamectin, lambda-cyhalothrin and emamectin, with high values for these active ingredients.
- Group 4 is characterised solely by dry season samples from the Aguigadji and Ahlan stations, which are strongly correlated with glyphosate, butachlor, aldrin, DDT, endosulfan and atrazine.

3.2. Sediments Contaminations by Heavy Metal

The results of the chemical analyses of the sediments in the dry and rainy seasons gave the average concentrations of metallic elements shown in Table 6. From the analysis of the results of the metallic elements in the sediments, lead (Pb), zinc (Zn), nickel (Ni), iron (Fe), copper (Cu) and chromium (Cr) were the most representative in both the dry and rainy seasons at the so-called contaminated stations. Pb was the most highly represented metal, followed by Zn, Ni, Fe, Cu and Cr, which were all representative in both the dry and rainy seasons at the contaminated stations (Aguigadji, Ahlan and Sele) and almost absent at the control station (Toho). In the rainy season, the highest lead concentration was 265.96 ± 21.02 mg/Kg and in the dry season 255.38 ± 79.09 mg/Kg, all detected at the Aguigadji station. This station apparently showed higher lead values from one season to the next. These values are higher than the EQS (91.3 mg/Kg). These concentrations were higher in the dry season than in the rainy season at all the stations (Figure 4 and Figure 5). The elements with low concentrations

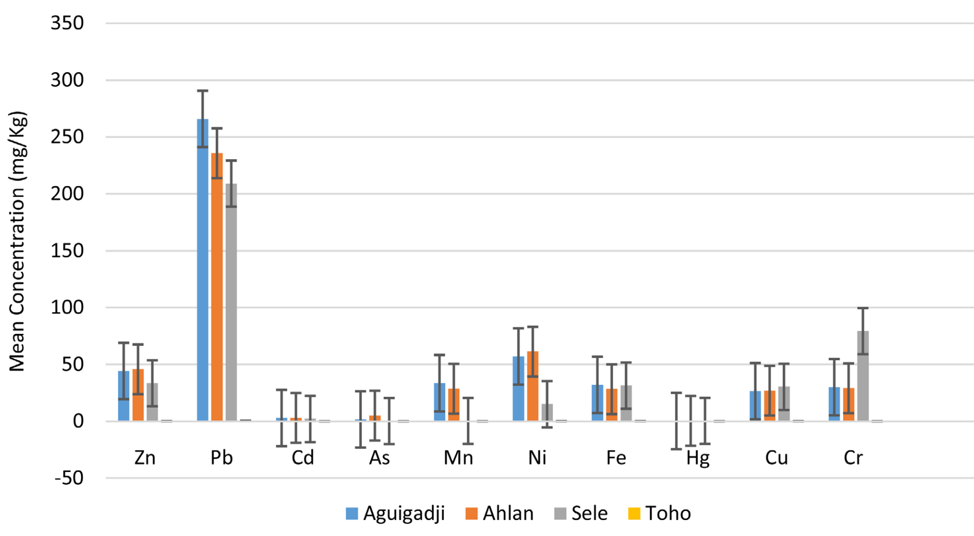


Figure 4. Concentrations of heavy metals in sediement during the rainy season.

Table 6. Concentrations of metallic elements in sediments in the rainy and dry season.

DE	Stations							
	Aguigadji		Ahlan		Sele		Toho	
	Rainy season	Dry season	Rainy season	Dry season	Rainy season	Dry season	Rainy season	Dry season
Zn	44.31 ± 2.82	31.92 ± 6.21	45.70 ± 0.88	30.64 ± 3.98	33.49 ± 2.54	31.2 ± 4.88	0.06 ± 0.02	0.02 ± 0.008
Cd	3.02 ± 0.40	2.32 ± 0.08	3.00 ± 0.56	2.12 ± 0.03	2.10 ± 0.64	1.97 ± 0.35	nd	0.06 ± 0.05
As	1.76 ± 0.96	0.4 ± 0.25	5.08 ± 3.10	0.45 ± 0.32	0.27 ± 0.24	0.18 ± 0.04	nd	0.03 ± 0.03
Pb	265.96 ± 21.02	255.38 ± 79.09	235.70 ± 25.94	231.48 ± 20.04	209.05 ± 12.74	188.67 ± 34.41	0.43 ± 0.34	0.24 ± 0.17
Mn	33.57 ± 13.79	2.22 ± 1.29	28.70 ± 4.79	2.63 ± 0.88	0.4 ± 0.35	0.04 ± 0.29	nd	nd
Ni	57.05 ± 3.34	53.35 ± 8.01	61.98 ± 11.22	56.88 ± 3.01	15.07 ± 4.93	11.50 ± 1.12	0.06 ± 0.03	0.03 ± 0.02
Fe	32.02 ± 0.80	26.01 ± 4.32	28.29 ± 6.39	25.38 ± 4.67	31.45 ± 1.58	23.85 ± 0.95	0.07 ± 0.004	0.05 ± 0.34
Hg	0.25 ± 0.10	0.04 ± 0.005	0.46 ± 0.22	0.08 ± 0.001	0.43 ± 0.40	0.03 ± 0.02	nd	nd
Cu	26.54 ± 1.92	18.31 ± 5.58	26.94 ± 2.04	25.19 ± 4.66	30.35 ± 2.37	31.25 ± 0.98	0.05 ± 0.02	0.02 ± 0.008
Cr	30.4 ± 1.25	28.06 ± 1.96	29.04 ± 2.93	28.31 ± 5.45	79.31 ± 7.001	29.64 ± 9.63	nd	nd

DE: Detected Elements; nd: no detected.

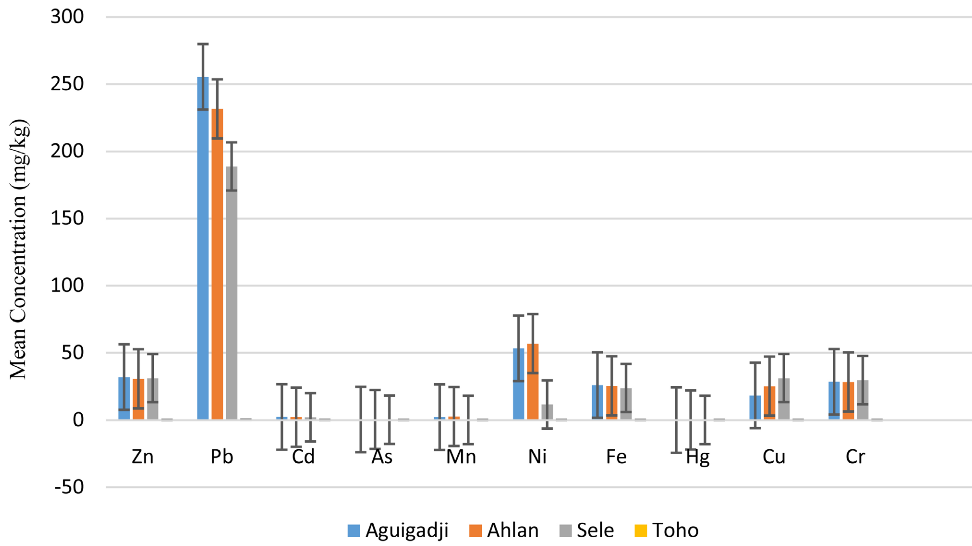


Figure 5. Concentrations of heavy metals in sediement during the dry season.

in the dry and rainy seasons were cadmium (Cd), arsenic (As), mercury (Hg) and manganese (Mg) at all stations. Manganese (Mg) was an exception, with high concentrations in the dry season at the Aguigadji and Ahlan stations (**Figure 4**). In addition, the average concentrations of heavy metals at all the so-called contaminated stations were well below those at the Toho station (control) (**Figure 4** and **Figure 5**), both in the dry and rainy seasons. Certain metallic elements including Mg, Mercury (Hg) and Chromium (Cr) were not detected at this station in either the dry or rainy seasons. In addition to these elements, Cadmium (Cd) and Arsenic (As) were also not detected in the rainy season at the control station (**Table 6** and **Table 7**).

Table 7. Accepted environmental quality standards for some heavy metals in freshwater sediments.

Métaux lourds	Zn	Pd	As	Mg	Cd	Ni	Fe	Hg	Cu	Cr
NQE	315	91.3	17	1100	3.5	75	43.766	0.486	197	90

EQS: Environmental Quality Stand.

3.3. Values of Contamination Facteurs (CF) for Metallic Elements

- **Contamination factor (CF)**

The Table 8 shows the seasonal contamination factor (CF) values for trace elements in sediment at each station.

The table shows that in the rainy season, the Aguigadji and Ahlan stations showed sediment CF values of less than 1 for Zn, As, Cu and Cr and higher values for Pb, Cd, Ni and Hg. At the Sele station, only Pb, Cd and Hg showed higher values. At all three of these so-called contaminated stations, there was significant contamination ($3 \leq FC < 6$) in Cd. A very high level of Pb contamination ($FC \geq 6$) in the sediments at the Aguigadji and Ahlan stations, which is significant ($3 \leq FC < 6$) at the Sele station. Significant Ni contamination was also noted at the Aguigadji and Ahlan stations. The sediments at all the so-called contaminated stations were also affected by moderate Hg contamination.

In the dry season, the so-called contaminated stations (Aguigadji, Ahlan and Sele) also showed values greater than 1 for Pb and Cd. The Cd values indicate significant contamination of the sediments by this element. The Pb values show very high contamination at the Aguigadji and Ahlan stations, followed by significant contamination at the Sele station. The values recorded in the dry season are mostly all lower than those in the rainy season. Ni also showed moderate contamination ($1 \leq FC < 3$) at the Aguigadji station and significant contamination ($3 \leq FC < 6$) at the Ahlan station. As for the Toho station considered as a control, there was low contamination of all metallic elements during all seasons.

3.4. Ecological Risk Index (Ri) Values

Table 9 shows the respective values of the potential risk factors (Er) and the ecological risk index (Ri) for trace elements in sediments in the wet and dry seasons. The results of the evaluation of the ecological risk index in relation to trace element contamination of sediments show potential risk factor (Er) values of less than 40 for the majority of metallic elements in both the dry and rainy seasons at all stations, with the exception of Cd and Hg, which showed high values at the so-called contaminated stations (Aguigadji, Ahlan and Sele). The respective values for Cd were 150.9, 150.3 and 105 in the rainy season and 115, 105.9 and 98.4 in the dry season. Hg showed high values only in the rainy season, at 56.4, 108 and 100.8 respectively. All these Cd and Hg values presented a potentially considerable ecological risk, except for Hg at the Aguigadji station (56.4), which showed a moderate ecological risk in the rainy season. The control station (Toho) showed values between 0 and 0.05 in the rainy season and between 0 and 3 in the dry season. These values are very low and equate to a very low potential ecological risk. These Er values led to high Ri values of 272.41, 326.26 and 249.52 in the wet season, compared with 184.63, 184.41 and 142.16 in the dry season at the Aguigadji, Ahlan and Sele stations respectively. These values showed a considerable ecological risk ($190 \leq Ri < 380$) in the rainy season and a moderate ecological risk ($95 \leq Ri < 190$) in the dry season at these so-called contaminated stations. The Toho station, on the other hand, showed Ri values < 95, which justifies a low ecological risk for sediments from this station.

Table 8. FC contamination factors for metallic elements.

	Zn	Pb	Cd	As	Ni	Hg	Cu	Cr
				Rainy Season				
Aguigadji	0.36	7.59	5.03	0.29	3.16	1.41	0.74	0.8
Ahlan	0.37	6.73	5.01	0.86	3.40	2.7	0.75	0.78
Sele	0.27	5.97	3.5	0.04	0.83	2.52	0.85	2.12
Toho	0.0004	0.01	0	0	0.003	0	0.001	0
				Dry Season				
Aguigadji	0.26	7.28	3.86	0.06	2.96	0.25	0.51	0.76
Ahlan	0.25	6.61	3.53	0.07	3.16	0.47	0.7	0.75
Sele	0.25	5.39	3.28	0.03	0.63	0.17	0.87	0.79
Toho	0.0001	0.007	0.1	0.005	0.001	0	0.005	0

Table 9. Potential risk factors (Er) and seasonal ecological risk index (Ri) for sediments in the lower Ouémé river.

	Er								Ri
	Zn	Pb	Cd	As	Ni	Hg	Cu	Cr	
				Rainy Season					
Aguigadji	-	37.95	150.9	2.9	18.96	56.4	3.7	1.6	**272.41**
Ahlan	-	33.65	150.3	8.6	20.4	108	3.75	1.56	**326.26**
Sele	-	29.85	105	0.4	4.98	100.8	4.25	4.24	**249.52**
Toho	-	0.05	0	0	0.01	0	0.005	0	**0.073**
				Dry Season					
Aguigadji	-	36.4	115	0.6	17.76	10	2.55	1.52	**184.63**
Ahlan	-	33.05	105.9	0.7	18.96	18.8	3.5	1.5	**182.41**
Sele	-	26.95	98.4	0.3	3.78	6.8	4.35	1.58	**142.16**
Toho	-	0.035	3	0.05	0.006	0	0.025	0	**3.116**

4. Discussion

A significant number of active ingredients were detected in the sediments of the lower Ouémé in both the wet and dry seasons. This result is similar to that of Yao *et al.* in 2018, who detected numerous active ingredients from herbicide families in the sediments of the Ebrié lagoon in Côte d'Ivoire [13]. The work of Kuranchie-Mensah *et al.* in 2012 [14], Gbaguidi *et al.* in 2011 [15] and Youchaou Tawaye *et al.* in 2021 also revealed these molecules in the River Densu in Ghana, the River Agbado in Benin and two aquatic ecosystems in Niger, respectively [16]. According to the author, this may also be due to the excessive use of synthetic pesticides in this area, which according to Lafia in 1996 [17] is a consequence of farmers' ignorance and illiteracy.

In both the dry and rainy seasons, glyphosate was the most representative active ingredient compared with all the others at all the stations. Glyphosate is the active ingredient in most of the total herbicides used in the study area. According to the FAO in 2004 [18], it is the most widely used herbicide in the world today. High concentrations of this active ingredient are evidence of the extensive use of herbicides in the environment. It belongs to the aminophosphate family, which is much more abundant in sediments than other pesticide residues [15]. Work by the same authors has also identified glyphosate in the waters of the Agbado River in Benin. Our results are similar to their findings following the detection of glyphosate in both dry and rainy seasons in the river sediments. Moreover, glyphosate is the only pesticide found in the sediments of the control station. The presence of glyphosate at this station is thought to be due to certain weed control practices in urban areas using plant protection products, which could lead to contamination of the water and then the lake sediments through run-off. This is in line with Coats in1991, who states that pesticides accumulate in sediments as a result of soil leaching [19].

DDT, atrazine and endosulfan were the other active ingredients that also showed remarkable concentrations and were detected at the Aguigadji, Ahlan and Sele stations (contaminated stations) and absent at the control station (Toho) in the wet and dry seasons. This result is contrary to that of Adeshina *et al.* in 2019 who found DDT and endosulfan in Owena River sediments in Nigeria only in the dry season [20]. Similar to our results, Gbaguidi *et al.* in 2011 [15] also detected atrazine only in the dry season in the sediments of the Agbado River. Atrazine is a member of the triazine family. According to the CNAC in 2007, the reported use of atrazine in Benin is as a herbicide for agricultural purposes (weeding crop fields) [21]. Atrazine concentrations obtained in the dry season are higher and almost non-existent in the rainy season at all the contaminated stations (Aguigadji, Ahlan and Sele) where it exists, with concentrations of 2.76 ± 0.40, 2.33 ± 0.46 and 0.52 ± 0.47 μg/kg respectively. This observation is in line with the assertions of Brignon and Gouzy (2007), who according to these authors, the adsorption coefficient (Koc) of atrazine is also relatively low, varying from 38 to 170, hence the probable absence of triazines in sediments during the rainy season [22].

Endosulfan concentrations were higher at the Ahlan and Aguigadji stations than at the Sele station in both the wet and dry seasons. Adam *et al.* in 2010 also found this molecule in the sediments of the Gogounou, Kandi and Banikoara cotton belt in Benin [23], but at relatively low doses, unlike the Densu river, where the highest concentration was that of endosulfan compared with the other active ingredients detected [14]. The highest concentration of endosulfan was 4.07 ± 0.90 μg/kg and was detected at the Aguigadji station in the dry season. DDT concentrations were remarkably high at all the so-called contaminated stations, but much higher at the Aguigadji and Ahlan stations in both the dry and wet seasons. Teklit in 2016 also found DDT residues in the Tekeze dam in both the dry and wet seasons [24]. Yehouenou-Pazou *et al.* in 2013 also found DDT in the sediments of Lake Nokoué at levels above the detection limit. The highest DDT concentration is 4.43 ± 1.70 μg/kg and was recorded at the Ahlan station in the dry season [25].

Pesticides such as Emamectin, Abamectin and Lambda-Cyhalothrin also showed remarkably high concentrations in the dry season. Cypermethrin and lambda-cyhalothrin are mainly pyrethroid pesticides imported into Benin for vector control [21]. The compounds in this family have very low volatility and are chemically unstable (half-life or DT50 of between 5 days and 13 weeks in the soil) [26]. They were detected at all the contaminated stations, but with much higher concentrations at the Sele station in the dry season. These results are similar to those of Adam *et al.* in 2010, who found pyrethroid residues, particularly lambda-cyhalothrin, in sediments in the cotton belt of Gogounou, Kandi and Banikoara in Benin [23]. These results are contrary to those of Gbaguidi *et al.* in 2011, who did not detect any pyrethroid active ingredients in the sediments of the Agbado river in either the wet or dry seasons [15]. Pesticide concentration levels in the contaminated stations (Aguigadji, d'Ahlan and Sele) and a comparison with pollution levels in the control station (Toho) show that sediments taken from the lower Ouémé are contaminated with pesticide residues. This is further evidence of the extent of pesticide use in this region. The other active ingredients found in the sediments, such as DDT and atrazine, which were not included in the list of pesticides used in the region, could be the result of contaminated water discharged upstream. However, this could also be due to the persistence of pesticides in the dry season [27]. Concentrations of active ingredients remain higher in the dry season than in the rainy season at all the stations. This result is contrary to that of Yao *et al.* in 2018, who found higher concentrations in the rainy season in the Ebrié River in Côte d'Ivoire [13]. The other sources of dry-season pollution leading to higher concentrations of these active ingredients could be the intensive market gardening activities carried out near water in the dry season in the region, mainly at the Sele station. On the other hand, the direct use by fishermen of organochlorine pesticides, in particular endosulfan, DDT and endrin, to catch fish during the low-water period [25] [28], which, following spillage, contaminates the water and subsequently the sediment. In the rainy season, this is justified by the fact that sediment contamination is caused first and foremost by surface water, which has already been contaminated by run-off from farms. This led Ouattara *et al.* in 2012 in a similar study to demonstrate that the abusive use of plant protection products in agriculture leads to the aquatic environment through runoff and leaching from farmland [29]. Principal component analysis (PCA) was used to classify the stations into four groups according to season and sampling. Group 1 (control (Toho) in the dry and rainy seasons and Sele in the rainy season), which is influenced by very low pesticide residue values. Group 2 (Aguigadji and Ahlan stations, mainly in the rainy season) is influenced by pesticide residue concentrations similar to those in group 1 at some stations and slightly higher levels of glyphosate, butachlor, aldrin, DDT, endosulfan and atrazine at others. Group 3 (Sele in the dry season) is influenced by higher concentrations of cypermethrin, abamectin, lambda-cyhalothrin and emamectin, with the last three being strongly correlated. This means that these micropollutants have the same affinity [30]. These active ingredients would therefore be governed by the same mechanism, which may be adsorption, allowing them to be retained in sediments. As for group 4 (Aguigadji and Ahlan in the dry season), it is strongly influenced by higher concentration values of glyphosate, butachlor, aldrin, DDT, endosulfan and atrazine. In addition, the results of the PCA revealed that the Sele, Aguiguadji and Ahlan stations are really polluted. This result could be due to the intensity of agricultural activities and, consequently, the abusive use of pesticides in the region. As for the control (Toho), it remains free of any pesticide pollution, with the exception of the detection of glyphosate, but in very low concentrations. In the rainy season, this is justified by the fact that sediment contamination is caused first and foremost by surface water, which has already been contaminated by run-off from farms. This led Ouattara *et al.* in 2012 in a similar study to demonstrate that the abusive use of plant protection products in agriculture leads to the aquatic environment through runoff and leaching from farmland [29]. 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From the analysis of the results of the metallic elements in the sediments, Lead (Pb), Zinc (Zn), Nickel (Ni), Iron (Fe), Copper (Cu) and Chromium (Cr) were the most representative in both the dry and rainy seasons at the contaminated stations. Cadmium (Cd), arsenic (As), mercury (Hg) and manganese (Mg) were mostly represented with low concentrations compared to the previous ones in both dry and rainy seasons. In the country and in the sub-region, authors have also identified heavy metals in sediments. These include scientific works [23] [31] [32] [33], which found a range of heavy metals in sediments from the Gogounou, Kandi and Banikoara cotton belts in Benin, the Weija water reserve in Ghana, the lake at the Nangbéto hydroelectric dam in Togo and sediments from the N'zi river in Côte d'Ivoire. Lead concentrations were highest at all the stations, with the highest concentration of 265.96 ± 21.02 mg/Kg recorded at the Aguigadji station during the rainy season. This result is in line with the findings of Ablain in 2002, who said that lead is not very soluble and would therefore accumulate relatively more in sediments due to its low solubility in water [34]. This result corroborates that of Adam *et al.* in 2010, who also recorded higher lead concentrations than other elements in sediments in the cotton belt (Gogounou, Kandi and Banikoara) [23]. The results of studies by Lawani *et al.* in 2014 also recorded high concentrations of Lead in sediments at several stations on Lake Nokoué in Benin [35]. In the wet season, all metal elements showed higher concentrations compared to those recorded in the dry season at all stations. This result corroborates that of Adje *et al.* in 2021 who recorded metal element values that were mostly higher in the rainy season than in the dry season in a lake in Togo [32]. The other metallic elements that showed relatively high concentrations after lead were Zinc (Zn), Nickel (Ni), Iron (Fe), Copper (Cu) and Chromium (Cr). This result seems normal if we consider, in part, that metals from cotton-growing soils [19] in particular and other crops in general accumulate in the sediments of adjacent waters. This result is similar to the trends of Ansah *et al.* in 2018 who found high concentrations of Copper (Cu), Zinc (Zn), Nikel (Ni), Iron (Fe) and Chromium (Cr) in the sediments of the Weija water reserve in Ghana [31]. In the rainy season, the Aguigadji, Ahlan and Sele stations showed sediment CF values of less than 1 for Zn, As, Cu and Cr. This result is similar to the trends recorded in a lake in Togo, where the CF of the metallic elements all remained below 1 in both the dry and rainy seasons [32]. The FC contamination factor showed very high levels of Pb, high levels of Cd and Ni and moderate levels of Hg at contaminated stations in both the dry and rainy seasons. This result is similar to that of Djeddi *et al.* in 2018 [7] in Algeria, which revealed the presence of lead in the sediments of most of the stations at the Béni Haroun dam, but with moderate contamination during the seasons of the year. FC values for the elements Cd and Hg also showed moderate contamination of sediments at the Moofoué station on the N'zi river in Côte d'Ivoire [33]. All these Pb and Hg values presented a potentially considerable ecological risk, except for Hg at the Aguigadji station, which showed a moderate ecological risk in the rainy season. These results are contrary to those of Adje *et al.* in 2021 [32], who found low ecological risks for sediments at all stations in the lake of the Nangbéto Togo hydroelectric dam in both the dry and wet seasons.

5. Conclusion

This study investigated the concentrations of pesticide residues and heavy metals in sediments from shrimp fishing areas in the lower Ouémé. Pesticide molecules were present in the sediments, with the majority being glyphosate, which showed higher concentrations at all the stations in both the dry and rainy seasons. These concentrations were higher than the environmental quality standard (EQS) at the so-called contaminated stations. Other active ingredients, including DDT, Atrazine, Endosulfan, Emamectin, Abamectin and Lambda Cyhalothrin also showed high concentrations at contaminated sites. Lead was the metallic element most represented in the sediment samples. Lead concentrations remained above the EQS and led to either very high levels of contamination, or high or moderate levels of contamination at contaminated sites, but with moderate ecological risks in the dry season and considerable risks in the wet season. Elements such as Cd, Ni and Hg also showed remarkable concentrations at the contaminated stations. The Toho station remained free of any high concentrations of pesticides and metallic elements.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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