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Heavy Metal Levels and Potential Ecological Risks Assessed at an Agroecosystem Site in Tropical Region

Luc Kalendele Lundemi[1], Stéphanie Salumu Neema[2], Emmanuel Kazinguvu Atibu[3*], Crispin Kyela Mulaji[3], Thierry Tabou Tangou[1], Camille Ipey Nsimanda[1], Robert Bueya Suami[4], Marie Onokoko Esako[5], Dieudonné Eyul'Anki Musibono[1], Fernando Piedade Carvalho[6]

[1]Department of Environment, Faculty of Sciences, University of Kinshasa, Kinshasa, Democratic Republic of the Congo
[2]Faculty of Oil, Gas and Renewable Energy, University of Kinshasa, Kinshasa, Democratic Republic of the Congo
[3]Department of Chemistry and Industry, University of Kinshasa, Kinshasa, Democratic Republic of the Congo
[4]Faculty of Pharmaceutical Sciences, University of Kinshasa, Kinshasa, Democratic Republic of the Congo
[5]Université Pédagogique National, Kinshasa, République Démocratique du Congo
[6]Laboratório de Protecção e Segurança Radiologica, Instituto Superior Técnico/Campus Tecnológico Nuclear, Universidade de Lisboa, Bobadela, Portugal
Email: *emmatibu@yahoo.fr

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Abstract

The concentrations and distribution of thirteen metals and metalloids were investigated in soils, sediments, and two biological matrices (the fish *Clarias gariepisnis* and the earthworm *Pontoscolex corethrurus*) from the CECOMAF agroecosystem, in Kinshasa, Democratic Republic of the Congo, in order to assess the impact of anthropogenic activities. The results revealed high concentrations of heavy metals, such as Cu, Zn, As, Cd, Pb, and Hg all above values recommended by sediment quality guidelines and their probable effect levels on biota. According to the calculated Enrichment Factor, soil and sediments ranked from moderately to heavily polluted by Cu, Zn, Cd, Pb and Hg. The Contamination Degree and other ecological risk indices indicated very high contamination and very high ecological risks posed by Cd and Hg, respectively. The Geoaccumulation Index indicated that current metal concentrations in the agroecosystem originated from anthropogenic activities, while the Spearman correlation matrix values indicated that Hg could originate from different sources and pathways than the other metals. It was concluded that metals from unchecked anthropogenic activities have negatively impacted agricultural activities and fish production at the CECOMAF agroecosystem. Action to reduce the contamination level and the ecological risks by remediating and preventing metal pollution in the CECOMAF agroecosystem site is recommended.

Keywords

Agroecosystem Site, Heavy Metals, Soil, Sediment, Ecological Risk, *Clarias gariepisnis*, *Pontoscolex corethrurus*

1. Introduction

Providing potable and clean water for human consumption and agriculture is one of the sustainable development goals of 2030 agenda (UN-Water, 2018; WHO/UNICEF, 2017). Nevertheless, contamination of water and aquatic ecosystems by toxic metals represents an escalating issue of global concern. Toxic metals can render water unsuitable for human consumption and are accumulated also by aquatic biota. Furthermore, contaminated water used for irrigation may contaminate soils and crops and, therefore, toxic metals and other pollutants may reach humans via food chains, thus representing an environmental and public health threat (Goyal et al., 2022; Carvalho, 2017a, 2017b).

Ensuring water abundance and water quality, combined with enhanced food production, is a very challenging task in the Sub Saharan regions but one of central importance. Several initiatives are trying to improve water availability and to increase food production including, for example, the Developing Green Cities initiative (FAO, 2012). These initiatives have known a reasonable success locally, and are feeding increasingly larger populations.

In the Kinshasa region, Democratic Republic of the Congo (DRC), one among several food production strategies fostered in latest years has been the development of integrated agroecosystems. In such integrated agroecosystems, the residues from vegetable production and marketing are collected, composted, and used as a fertilizer, and the river water is distributed through constructed waterways for use in crop irrigation. In several districts around Kinshasa, such agroecosystems have developed with noticeable success, such as CECOMAF, which is also a marketing center for vegetable and fruit products.

Because of population growth and expanding anthropogenic activities, in particular in the urban settings, surface waters may become increasingly polluted by the discharge of residential wastewater, sewage outlets, discharges from sites of livestock rearing, and agrochemical residues from agricultural areas. This is the case of the N'djili River section by the CECOMAF agroecosystem (Su et al., 2014). Despite the growing number of inhabitants consuming products from these integrated agroecosystems, the management and monitoring of such agroecosystems has been neglected.

The high particle suspended load of domestic wastewater discharges and in surface runoff settle on the bottom of N'djili River as a sludge which builds up with time (Park et al., 2013). This sludge may contain important quantities of toxic metals and other pollutants, which may contaminate aquatic biota, agriculture soils, and the groundwater (Goyal et al., 2022).

The concentration, distribution, and toxicity of heavy metals in soil, sediment and edible fish samples of surface waters (for example, rivers and West Atlantic Ocean Coast) of Kinshasa and Central Kongo province of the Democratic Republic of the Congo have been recently assessed (Ngweme et al., 2020; Mata et al., 2019, 2020; Suami et al., 2018). However, there is a lack of information on the concentration, distribution, and ecological toxicity of heavy metals in agroecosystems at the Kinshasa region.

Heavy metals and metalloids are natural soil constituents and their concentration may vary depending upon the rock and geological materials present at specific sites and, therefore, the background concentration of metals needs to be taken into consideration in order to assess the anthropogenic inputs of metals and the resulting environmental pollution (Zlobina et al., 2022).

Earthworms are bio-indicators of heavy metal contamination in soils. Indeed, previous studies highlighted the strong bioaccumulation of heavy metals in the earthworm tissues when they live in contaminated environments and proposed their use as bioindicators for soil contamination (Dai et al., 2004; Lourenço et al., 2011; van Vliet et al., 2005).

Freshwater fish from agroecosystems is a main aquatic product for human consumption. Many studies were performed on the accumulation of inorganic and organic pollutants in fish species. These studies demonstrated that the fish species are good bioindicators of the accumulation of pollutants in aquatic environments. The monitoring of pollutants in fish can also be used to assess and to prevent human health risks (Moiseenko & Gashkina, 2020; Rajeshkumar & Li, 2018; Suami et al., 2018; Bawuro et al., 2018; Schäfer et al., 2015; Streit, 1998).

This study is the first attempt to evaluate the concentrations and distribution of thirteen metals and metalloids and their potential ecological risks at the CECOMAF agricultural site in Kinshasa. In particular, this study focused on the quality of agricultural soils and sediments from the N'djili River at that site, and bioaccumulation of metals in two biological matrices, the common catfish from the river and earthworms from soils.

2. Materials and Methods

2.1. Description of the Sampling Site

This study was carried out in the agroecosystem of N'djili CECOMAF (CECOMAF stands for "Centre de Commercialisation des Produits Maraîchers et Fruitiers", in French) which is located between N'djili and Kimbanseke communes at the eastern part of Kinshasa, the capital city of the Democratic Republic of the Congo. The agroecosystem of CECOMAF is located at coordinates S 04˚25436' and E 015˚21660', on the right bank of the N'djili River, upstream the point of water collection by the N'djili Water Distribution Authority for the supply of water for human consumption.

CECOMAF is a marketing center for vegetable and fruit products, created in 1972 as a cooperative market for agriculture products from N'djili commune.

This CECOMAF site was created to supply the growing population with agriculture products. Due to the particularity of its geography, hydrography and soil, the N'djili commune/CECOMAF occupies the first place in vegetable production in Kinshasa and supplies also several markets of the capital with various agriculture products (Mokili, 1998).

The CECOMAF site is limited to the north by districts VI and VII of the commune of N'djili, to the south by the commune of Mont-Ngafula, to the east by the commune of Kimbanseke and to the west by the N'djili River and the commune of Kisenso (**Figure 1(A)**). The area is mostly flat and located at an average altitude of 290 m above sea level.

The aquatic system of the N'djili River in the CECOMAF section includes aquatic plants, invertebrates, and several fish species. The common North African catfish (*Clarias gariepisnis* Burchell, 1822) from the *Siluridae* family, is very abundant and may attain 1 m total length and near 40 kg wet weight. The population, fishermen, and farmers have free access to the N'djili River in CECOMAF area and the fish captures attain an estimated amount of 5 tons per year. Agriculture production from the N'djili/CECOMAF area encompasses several kinds of vegetables, such as the green amaranth (*Amaranthus viridis*). N'djili and Kimbanseke communes (**Figure 1(B)**) have a population of approximately 1.5 million inhabitants, some of whom are engaged in agriculture activities and freshwater fishing. Both agriculture and freshwater fishery products are consumed by

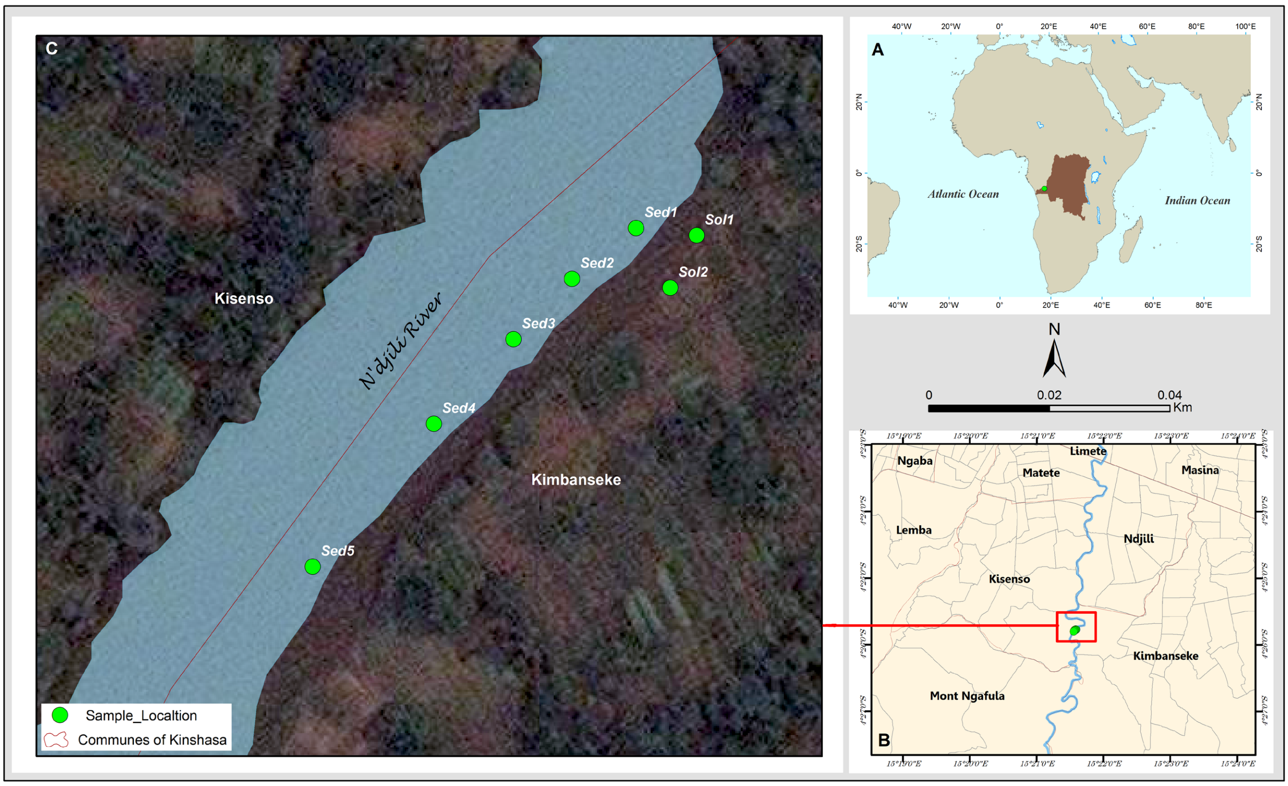

Figure 1. Map of Congo DR in Africa (A); Map of the study area CECOMAF location (in green) in Kinshasa region (B); Sampling point location of the study area in CECOMAF area (C).

the local population and account for an important fraction of the diet, although no accurate statistics are available.

2.2. Sample Collection

The sampling campaign was performed on February 2020. In the section of N'djili River at CECOMAF, bottom sediments were collected manually, using a plastic shovel and transferred into 1.5 L plastic bottle. From each sampling point, about 250 - 300 g of surface sediment was collected. The sediments were sampled at a distance of about 2 m from the shoreline and with about 1 m water depth. The sediment samples (n = 5) were labelled as Sed1, Sed2, Sed3, Sed4, and Sed5. Top soil (0 - 20 cm depth) samples (n = 2) were collected in agriculture areas by the CECOMAF site and labelled Sol1 and Sol2 (**Figure 1**, **Figure 2**).

Biota samples, including the common African catfish *Clarias gariepisnis*, (**Figure 2(D)**) from the agroecosystem of CECOMAF, and the earthworm *Pontoscolex corethrurus* from agriculture soils nearby (**Figure 2(C)**), were collected using a gill net and a shovel, respectively.

All samples were stored at 4˚C and transported refrigerated to the analytical platform of the University of Geneva for analysis.

2.3. Sample Preparation

Earthworms were washed, weighed, dissected and the tissues of several specimens

Figure 2. Image (photos by Luc Lundemi and Stéphanie Neema) describing: (A) The CECOMAF section of the N'djili river; (B) Use of pesticides on crops around the CECOMAF agroecosystem; (C) The earthworm *Pontoscolex corethrurus* collected along the studied site of CECOMAF and (D) The fish *Clarias gariepisnis* collected along CECOMAF agroecosystem.

bulked to make a sufficiently large sample. Fish were washed, weighed, dissected and the muscle tissue (filet) from several adult specimens were combined in one sample. Biological samples were then frozen and stored at −20˚C in clean polypropylene bottles until acid digestion. The digestion of samples was performed, with minor modifications, as described by Dai et al. (2004), Rashed (2001), and Sivaperumal et al. (2007). In short, a portion of tissue from each sample was freeze-dried (Adolf Kühner, Birsfelden, Switzerland) and grinded to obtain a fine powder. Then, approximately 1 g of fish sample and 100 mg of earthworm sample was digested in a suprapur HNO_3 (Nitric acid 65% Suprapur®, Merck KGaA, Darmstadt Germany)-$HClO_4$ (Perchloric acid 70%, Merck KGaA, Darmstadt Germany) mixture (3:1), in Teflon pressure vessels and heated overnight at 110˚C. The digested samples were cooled at room temperature and centrifuged to obtain a clear solution.

Soil and sediment samples were freeze-dried, grinded to a fine powder, homogenized, and sieved using a 63 µm sieve. They were then digested, as described by Thevenon & Poté (2012). In brief, approximately 10 - 15 mg of sample powder was completely digested using pure acids in Teflon vessels and a glass-ceramic hot plate. The procedure involves three heating steps with: 1) 1 mL HNO_3 (suprapure, 65%); 2) a mixture of 0.5 mL of $HClO_4$ (suprapure, 70%) with 0.5 mL HF (suprapure, 40%); and 3) additional treatment with 0.5 mL of HNO_3 (suprapure, 65%). The solvents were evaporated between each step and finally the samples were diluted in 10 mL of a 1% HNO_3 solution before chemical analysis. Analysis was performed within 24 hours after dilution. Acid dilutions were performed using ultrapure water (Millipore, Milli-Q, 18MW, Merck, Darmstadt, Germany).

2.4. Metal Analysis by ICP-MS

Twelve heavy metals and metalloids, namely scandium (Sc), chromium (Cr), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), arsenic (As), selenium (Se), cadmium (Cd), tin (Sn), antimony (Sb), and lead (Pb) were analyzed using Inductively Coupled Plasma-Mass Spectrometry (Agilent 7700x series ICP-MS, developed for complex matrix analysis). Methods used were described in detail elsewhere (Atibu et al., 2018). In short, a collision/reaction cell (Helium mode) and several interference equations were used to eliminate different spectral interferences. For the ICP-MS calibration were used standard Merk IV solutions with different concentrations (0, 0.2, 1, 5, 20, 100 and 200 $\mu g \cdot L^{-1}$), the ICP multi-element standard solution, and other mono-element solutions (Se and Sb, Merck KGaA, Darmstadt Germany) (Thevenon et al., 2013). Standard deviations of three replicate measurements were below 5%. The limit of detection (LOD) was calculated as 3 times the standard deviation of the analytical blanks, and was less than 0.001 $\mu g \cdot L^{-1}$ for all analyzed elements. The chemical blanks for the procedure were less than 2% of the sample signal. The sensitivity of the device and the reliability of the results were verified through the repeated use of the certified reference material LKSD4 (CANMET, Canada). The analysis of samples by ICP-MS was performed after dilution of sample digests with suprapur 1% HNO_3 (Nitric acid 65% Suprapur®, Merck KGaA, Darmstadt Germany).

Results of metal concentrations in soils and sediments are expressed in ppm ($mg \cdot kg^{-1}$ of sample dry weight). The results for metal concentrations in biological samples are expressed in ppm ($mg \cdot kg^{-1}$ wet weight) calculated with average values of water content in fish and earthworm muscle tissues as described in Garcia-Bravo et al. (2011).

2.5. Mercury Analysis in Soil and Sediment Samples by AAS

The Advanced Mercury Analyser (AMA 254, Altecs.r.l., Czech Rep.) which is a mercury specific Atomic Absorption Spectrometer (AAS), was used for the total Hg analysis in soil and sediment samples according to the method described by Roos-Barraclough et al. (2002). The method consists on the sample combustion, and mercury amalgamation on a gold trap, followed by the gaseous mercury measurement by AAS. The limit of detection (3 SD blank) value was determined at 0.005 $mg \cdot kg^{-1}$, and the reproducibility was better than 5%. The Hg concentrations are expressed is $mg \cdot kg^{-1}$ of dry weight (ppm).

2.6. Potential Ecological Risks

The distribution of metals between water and sediment phases was determined as the partitioning coefficient, Kd, being:

$$\text{Kd} = [\text{metal concentration in the sediment } (mg \cdot kg^{-1} \text{ of dry weight})/\text{metal concentration in the water } (mg \cdot L^{-1})]. \tag{1}$$

The accumulation of metals in the fish muscle was determined as the metal bioconcentration factor (BCF), being:

$$\text{BCF} = [\text{metal concentration in fish } (mg \cdot kg^{-1} \text{ of wet weight})/\text{metal concentration in the water } (mg \cdot L^{-1})]. \tag{2}$$

Several indices have been proposed to assess the contamination of soils and sediments by metals and potential environmental risks (Mavakala et al., 2022; Aja et al., 2021; Akanchise et al., 2020, Atibu et al., 2016).

According to Maanan et al. (2004), the natural concentrations and anthropogenic pollution of soil and sediment can be assessed through the determination of the Geo-accumulation Index (Igeo) and the Enrichment Factor (EF), respectively.

1) *Geoaccumulation Index* (*Igeo*)

The following equation was used to calculate the geoaccumulation index (Igeo):

$$\text{Igeo} = \text{Log}_2[(C_n)/1.5(B_n)] \tag{3}$$

In the above equation, "C_n" represents heavy metal (n) concentration in the sample; "B_n" represents heavy metal (n) concentration in the geochemical background; "1.5" represents the background matrix correction due to lithogenic effects.

2) *Enrichment Factor* (*EF*)

The following equation was used to calculate the Enrichment Factor, EF:

$$EF = (Metal/Sc)\ Sample/(Metal/Sc)\ Background \quad (4)$$

where "Metal" represents the concentration of any heavy metal in the analyzed sample and in the geochemical background; "Sc" represents the scandium concentration in the sample and in the geochemical background. Therefore, a geochemical normalization is performed using scandium (Sc) as a normalization factor. The UCC (Upper Continental Crust) concentration values were used as background values of heavy metals (McLennan, 2001).

3) *Contamination Factor* (*CF*)

The contamination factor (CF) was determined to quantify the contamination level of heavy metals in soil and sediment samples. The following equation was used for CF calculation (Rubio et al., 2000; Förstner et al., 1989; Hånkanson, 1980):

$$CF = (C_n/B_n) \quad (5)$$

where "C_n" represents the concentration of metal *n* in sample and "B_n" represents the concentration of the same metal in the geochemical background.

4) *Polymetallic Contamination Degree* (*CD*)

An *a priori* evaluation of the polymetallic contamination level for each sample was made by the calculation of the contamination degree (CD) using the following formula (Hånkanson, 1980):

$$CD = \Sigma CF_i \quad (6)$$

where "*i*" represents a specific heavy metal, and "CF_i" represents the contamination factor of the heavy metal *i*. Eight heavy metals were considered for the CD calculation.

5) *Ecological risk index* (*Eri*)

To evaluate the harmful impact of heavy metals on the environment and humans, the Ecological risk index (Eri) was calculated. This parameter reflects and combines the ecological sensitivity and the toxicity of the pollutants (Hånkanson, 1980; Suresh et al., 2012). The Eri is determined through the following equation:

$$Eri = Tr_i \times CF_i \quad (7)$$

where "CF_i" represents the contamination factor of a specific heavy metal *i*, and "Tr_i" represents the toxic-response factor for a given heavy metal (or the biological toxic factor) of a given heavy metal. The Tr_i values used were 40; 30; 10; 5; 5; 5; 2 and 1 for Hg; Cd; As; Co; Cu; Pb; Cr and Zn, respectively (Hånkanson, 1980; Islam et al., 2014).

6) *The potential ecological risk index* (*RI*)

The potential ecological risk index (RI) for polymetallic contamination was calculated by summing the single ecological risk index Eri. This RI index considers the synergy (combined effect) of several parameters, namely the toxic level, the concentration of heavy metals, and the ecological sensitivity of biological communities to heavy metals (Singh et al., 2002). The RI was computed using the following equation:

$$RI = \sum Eri \tag{8}$$

where "Eri" represents the ecological risk index for a specific heavy metal *i*.

2.7. Data Analysis

Spearman's rank order correlation was performed using XLSTAT (New York, USA. https://www.xlstat.com) in order to explore the relationship among compounds and their potential sources.

3. Results and Discussion

3.1. Heavy Metal Concentrations in Soils, Sediments and Biota

The metal and metalloid (Sc, Cr, Co, Ni, Cu, Zn, As, Se, Cd, Sn, Sb, Pb, and Hg) concentrations in soil and sediment samples are shown in Table 1.

Table 1. Heavy Metal content ($mg \cdot kg^{-1}$ dw in soil and sediment and $mg \cdot kg^{-1}$ ww) in the fish *Clarias gariepisnis* and in the earthworm *Pontoscolex corethrurus* analized by ICP-MS[a] and AAS.

Sample	Sc	Cr	Co	Ni	Cu	Zn	As	Se	Cd	Sn	Sb	Pb	Hg	From
Water		1.26E−03	9.00E−05		3.80E−03	6.12E−03	3.40E−04		3.00E−05			2.70E−04		Ngweme et al. 2020
Sol 1	0.1	3.8	0.6	0	**303.7**	**742.8**	**2.8**	0.4	**1.1**	0.2	0.5	**276.1**	**1.5**	
Sol 2	2.3	17.3	1.8	1.6	**281.5**	**602.4**	**3.7**	5.2	**1.2**	0.1	0.1	**270.0**	**0.3**	This work
Sed1	1.1	8.4	1.1	0.9	**182.8**	**749.8**	**3.1**	1	**1.3**	0.2	0.1	**151.1**	0.1	
Sed2	0.7	7.2	1.1	0.5	**188.3**	**804**	**2.9**	1.5	**0.8**	0.2	0.2	**272.9**	**0.9**	
Sed 3	1.5	10.7	1.3	1.2	**208.6**	**780.6**	**3.3**	1.9	**1.5**	0.2	0.1	**201.7**	**0.2**	
Sed 4	0.8	9.9	1.4	1.1	23.5	**910.6**	**3.1**	1.1	**1.2**	0.2	0.2	**309.9**	**0.3**	
Sed 5	0.6	6.3	0.8	0	15.5	114.2	**3**	1.1	**1**	0.2	0.2	**49.1**	**0.7**	
Mean Sed	0.94	8.5	1.14	0.74	123.74	671.84	3.08	1.32	1.16	0.2	0.16	196.94	0.44	
SD Sed	0.36	1.83	0.23	0.49	95.68	317.55	0.15	0.38	0.27	0.00	0.05	103.09	0.34	
% SD Sed	39	21	20	67	77	47	4.8	28	23	0	34	52	78	
Biota samples														
Fish	0	0.3	0.1	0	1.2	28.6	1	1.5	0.3	0.2	0	**0.6**		
Earth-worm	2.2	20.5	2.3	4.8	**36.5**	**591**	4.7	8.1	**2.6**	2.9	0.1	**1.7**		
FAO[b]					30	30			0.5			0.5		
EU[c]									0.1			0.2		
Kd		6.75E+03	1.27E+04		3.26E+04	1.10E+05	9.06E+03		3.87E+04			7.29E+05		This work
Kd (IAEA)			4.3E+04											IAEA, 2010
BCF		238	1111		316	4673	2941		10000			2222		This work
BCF IAEA)		40	76		230	3400	330					25		IAEA, 2010

[a]Total variation coefficients for triplicate measurements are smaller than 5% for ICP-MS analysis. The recovery values from measurements for reference material (LKSD 4) were above 97.5% for all elements the ICP-MS triplicate. In bold, values of the heavy metal's concentration high than the recommended probable effect concentration according to International Sediment Quality Guidelines for the Protection of Aquatic Life recommendation (CCME, 1999). ISQG—Interim freshwater sediment quality guidelines; PEL—probable effect levels (sediment dry weight). [b]FAO/WHO permissible level for toxic metals in fish (Joint FAO/WHO Expert Committee on Food Additives, 2000). [c]EU permissible level for toxic metals in fish (EU, 2005).

Heavy metal concentrations in the soil samples did not differ from concentrations in sediments from the N'djili River section by CECOMAF. These concentrations are significantly higher than concentrations determined in soil samples from a reference site, not receiving urban and farm wastewater discharges (Ngweme et al., 2020), thus giving a first indication that there is environmental contamination by heavy metals in the CECOMAF area (**Table 1**).

Regarding the relative concentrations in soil and sediments (mg·kg^{-1} dw), in general heavy metals followed the order: Zn (910.6) > Pb (309.9) > Cu (303.7) > Cr (17.3) > Se (5.2) > As (3.7) > Co (1.8) > Ni (1.6) > Cd (1.5) > Hg (1.5) > Sb (0.5) > Sn (0.2) > Sc (0.1).

The concentrations of Cu, Zn, As, Cd, Pb and Hg in sediments were higher than the reference values set by the sediment quality guidelines (SQGs) for protection of aquatic life and their probable effect levels (CCME, 1999). These high concentrations of Cu, Zn, As, Cd, Pb and Hg in samples are attributed to anthropic sources around the site, such as untreated urban water runoff, runoff from farmlands, fertilizer applications, etc. (Tshibanda et al., 2014; Ngweme et al., 2020). Such level of contamination is likely to put a threat to the health of the aquatic system.

Table 1 indicates also the concentrations of Sc, Cr, Co, Ni, Cu, Zn, As, Se, Cd, Sn, Sb and Pb in biological matrices, the fish (*Clarias gariepisnis*) and the earthworm (*Pontoscolex corethrurus*). Comparing metal concentrations in the fish filet against the reference limit values set by both the Food and Agriculture Organization (FAO) and the European Union (EU), it may be noted that metals in fish did not exceed the concentration limits recommended by FAO, but exceed the most stringent values adopted by the EU for protection of the human health. Therefore, humans consuming regularly this fish are exposed to a relatively high intake of Cd and Pb.

The earthworm *P. corethrurus* displayed heavy metal concentrations generally higher than those of fish *C. gariepisnis*. High metal concentrations in earthworms indicate availability of these metals from agriculture soils (Dai et al., 2004). There is no similar data for earthworms from a reference site in the DRC to make a comparison.

3.2. Environmental Distribution and Bioaccumulation of Metals

The computed values for metals partitioning between the water and sediment phases (Kd) and metals bioconcentration in fish muscle (BCF) are shown in **Table 1**. Although Kd values have been reported for many metals in freshwater ecosystems, many values have not been validated in recent literature reviews and, therefore, comparisons of our results are limited to a few chemical elements (IAEA, 2010). The same is valid for BCF values in freshwater fish, and large variations in BCF values reported have been noticed (IAEA, 2010). Many determinations were made under the conditions of temperate environments and may not apply to tropical ecosystems, in particular due to the presence of organic matter in higher amounts in tropical aquatic systems and higher temperature, which may affect the distribution and bioaccumulation of metals. Nevertheless, for the metals with available data for comparison, the determinations made in this study are in the same order of magnitude or in the range of values reported in literature (Table 1). It must be noted that high Kd values (e.g., 10^4 - 10^5) correspond to strong sediment-water partitioning, i.e., low water solubility of the metal species with most of the metal bound to the sediment. Low Kd values (10^0 - 10^2) indicate high water solubility of the metal and a minor immobilization of the metal in sediments. The BCF values calculated were noticeably high for some metals and indicate that a significant metal transfer to humans may occur through fish consumption.

3.3. Geoaccumulation Index, Enrichment Factor, and Ecological Risk Parameters

The calculated Igeo and EF parameter values are shown in Table 2. The results for both of them were interpreted according to classification scales developed in previous studies which are shown under Table 2 (Atibu et al., 2018, Thevenon et al., 2013). These scales allow for an easier ranking and interpretation of results.

The Igeo values ranged from −1.0 to 3.0 (Cu), 0.0 to 3.0 (Zn), 0.0 to 1.0 (As), 2.0 to 3.0 (Cd), 1.0 to 3.0 (Pb) and 0.0 to 4.0 for Hg. The Cr and Co Igeo values were less than −1.0 for all sampling sites, i.e., much lower than the average concentration of these two elements in the upper continental crust. This can be due

Table 2. EF and Igeo values for Cr, Co, Cu, Zn, As, Cd, Pb and Hg, in soil and sediment samples.

Sample	EF								Igeo							
	Cr	Co	Cu	Zn	As	Cd	Pb	Hg	Cr	Co	Cu	Zn	As	Cd	Pb	Hg
Sol1	11.9	6.6	1336.3	1150.8	205.3	1234.7	1518.6	2946.4	−3.0	−4.0	2.0	3.0	0.0	3.0	2.0	0.0
Sol2	2.4	0.9	53.9	40.6	11.8	58.6	64.6	25.6	−4.0	−5.0	3.0	3.0	0.0	3.0	3.0	4.0
Sed1	2.4	1.1	73.1	105.6	20.7	132.7	75.6	17.9	−2.0	−3.0	3.0	2.0	1.0	3.0	3.0	2.0
Sed2	3.2	1.7	118.4	177.9	30.4	128.3	214.4	252.6	−3.0	−4.0	2.0	3.0	0.0	2.0	3.0	3.0
Sed3	2.2	1.0	61.2	80.6	16.1	112.2	74.0	26.2	−2.0	−4.0	2.0	3.0	1.0	3.0	3.0	1.0
Sed4	3.9	1.9	12.9	176.3	28.4	168.4	213.1	73.7	−2.0	−3.0	−1.0	3.0	0.0	3.0	3.0	2.0
Sed5	3.3	1.5	11.4	29.5	36.7	187.1	45.0	229.2	−3.0	−4.0	−1.0	0.0	0.0	3.0	1.0	3.0

EF values interpretation

EF < 1	No enrichment
EF < 3	Minor enrichment
EF 3 - 5	Moderate enrichment
EF 5 - 10	Moderately severe enrichment
EF 10 - 25	Severe enrichment
EF 25 - 50	Very severe enrichment
EF > 50	Extremely severe enrichment

Igeo classification

Igeo ≤ 0	Class 0—Practically unpolluted
0 < Igeo < 1	Class 1—Unpolluted to moderately polluted
1 < Igeo < 2	Class 2—Moderately polluted
2 < Igeo < 3	Class 3—Moderately to heavily polluted
3 < Igeo < 4	Class 4—Heavily polluted

to high content of the samples in organic matter, deviating their composition from the average geochemical background. In general, samples were graded in class 0 and 1 ("practically unpolluted" and "unpolluted to moderately polluted", respectively) for Cr, Co and As. Some samples were graded in classes 2 and 3 ("moderately polluted" and "moderately to heavily polluted", respectively) namely for Cu, Zn, Cd, Pb, and Hg (**Table 2**), indicating that these elements were present in the agroecosystem in concentrations much higher than the average geochemical background.

For a specific heavy metal, the EF value can be used to identify whether site contamination has occurred and, because of the normalization introduced with the use of Sc, this parameter provides information that is more robust than Igeo. For 0.5 ≤ EF ≤ 1.5, the concentration of metal in samples is considered to acceptably match heavy metal natural concentrations in crustal sources. However, when EF ≥ 1.5 a metal enrichment has occurred and may be attributed to input of the heavy metal from anthropogenic sources to the study area (Feng et al., 2004; Zhang & Liu, 2002). In the study area, the EF values of all samples ranged between "no enrichment" (EF < 1) to "moderately severe enrichment" (EF 5-10) for Cu and Co, while for Cu, Zn, As, Cd, Pb and Hg the EF values ranged from "severe enrichment" (EF 10 - 25) to "extremely severe enrichment" (EF > 50) (**Table 2**).

The heavy metal EF values in samples followed this order: Hg (2946) > Pb (1519) > Cu (1336) > Cd (1235) > Zn (1151) > As (205) > Cr (11.9) > Co (6.6). According to these results, the anthropogenic activities did originate a clear enhancement of heavy metal concentrations in the studied site. The first five elements of this roll based on EF are the same spotted by Igeo in classes 2 and 3, and thus both parameters provide convergent information.

The results for potential ecological risk parameters, such as the Contamination Factor (CF), the Ecological Risk Factor (Eri), the Polymetallic Contamination Degree (CD) and the Ecological Risk Index (RI) are shown in **Table 3**.

According to the CF parameter, most samples displayed a very high contamination factor (6 < CF) for Cu, Zn Cd, Pb, and Hg. Soils and sediments displayed moderate contamination for As (1 < CF < 3). Low sediment contamination (CF < 1) was displayed only for Cr, Co (and some samples for Cu, as well).

Based on the parameter values for Polymetallic Contamination Degree (CD), all samples displayed either high or very high contamination (**Table 3**).

Based on the parameter values for Ecological Risk (Eri), the results indicated low or moderate risk for Cr, Co, Cu, Zn, As, and Pb, while for Cd and Hg the results indicated high, or very high ecological risk.

Parameter values for the Synergistic Effects index (RI) indicated very high ecological risk from all soil and sediment samples.

Previous analysis of heavy metals in the region included also areas non-significantly impacted by human activities that can be used for comparison with current results for the CECOMAF site (Ngweme et al., 2020). This comparison showed that metal concentrations in soil and sediments from CECOMAF were

Table 3. CF, CD, Eri and RI values for Cr, Co, Cu, Zn, As, Cd, Pb and Hg, in soil and sediment samples.

Sample	CF								CD	Eri								RI
	Cr	Co	Cu	Zn	As	Cd	Pb	Hg		Cr	Co	Cu	Zn	As	Cd	Pb	Hg	
Sol1	0.1	0.1	12.1	10.5	1.9	11.2	13.8	26.8	77	0.2	0.5	60.5	10.5	19	336	69	1072	1568
Sol2	0.5	0.2	11.3	8.5	2.5	12.2	13.5	5.4	54	1	1	56.5	8.5	25	366	67.5	216	742
Sed1	0.2	0.1	7.3	10.6	2.1	13.3	7.6	1.8	43	0.4	0.5	36.5	10.6	21	399	38	72	578
Sed2	0.2	0.1	7.5	11.3	1.9	8.2	13.6	16.1	59	0.4	0.5	37.5	11.3	19	246	68	644	1027
Sed 3	0.3	0.1	8.3	11	2.2	15.3	10.1	3.6	51	0.6	0.5	41.5	11	22	459	50.5	144	729
Sed 4	0.3	0.1	0.9	12.8	2.1	12.2	15.5	5.4	49	0.6	0.5	4.5	12.8	21	366	77.5	216	699
Sed 5	0.2	0.1	0.6	1.6	2	10.2	2.5	12.5	30	0.4	0.5	3	1.6	20	306	12.5	500	844

CF classification

CF<1	Low contamination
1 < CF < 3	Moderate contamination
3 < CF < 6	Considerable contamination
6 < CF	Very high contamination

Eri classification

Eri < 40	Low ecological risk
40 < Eri < 80	Moderate ecological risk
80 < Eri < 160	Considerable ecological risk
160 < Eri < 320	High ecological risk
Eri > 320	Very high ecological risk

RI classification

RI < 150	Low ecological risk or low ecological pollution level
150 ≤ RI < 300	Moderate ecological risk or Moderate ecological pollution level
300 ≤ RI < 600	Considerable ecological risk or severe ecological pollution level
RI > 600	Very high ecological risk or serious ecological pollution level

CD classification

8 ≤ CD < 16	Moderate contamination
16 ≤ CD < 32	High contamination
32 ≤ CD	Very high contamination

one to 44 times higher than the reference soil labelled "CESCO" by Ngweme et al. (2020). Therefore, the overall results from the ecological indices for the CECOMAF site are in line with the relative level of metal concentrations in soils and sediments.

In view of these results, the populations living near the CECOMAF agroecosystem site, namely those who use the section of the N'djili river at CECOMAF site for domestic needs, crop irrigation, and consume fish from the river are exposed to heavy metals contamination.

3.4. Correlation between Parameters

The Spearman's rank-order correlation values, which were calculated to investigate heavy metal possible sources and pathways in soils and sediments, are presented in the Table 4.

Table 4. Spearman rank order correlation for heavy metals analysed in the soil and sediment samples.

Variables	Cr	Fe	Co	Ni	Cu	Zn	As	Se	Cd	Sn	Sb	Pb	Hg
Sc	**0.964**	**0.964**	**0.865**	**0.955**	0.143	0.107	**0.955**	**0.721**	**0.703**	−0.612	−0.926	−0.179	−0.775
Cr		**0.929**	**0.955**	**0.991**	0.107	0.214	**0.955**	**0.775**	**0.649**	−0.612	−0.810	0.000	−0.685
Fe			**0.811**	**0.901**	0.036	−0.071	**0.991**	**0.667**	**0.739**	−0.612	−0.926	−0.321	−0.811
Co				**0.945**	0.018	0.324	**0.864**	**0.745**	0.473	−0.618	−0.642	0.198	−0.545
Ni					0.216	0.252	**0.927**	**0.736**	**0.673**	−0.618	−0.778	0.090	−0.655
Cu						−0.107	0.018	0.126	0.144	−0.408	0.000	0.357	0.252
Zn							−0.018	0.036	0.162	0.408	0.039	**0.607**	−0.162
As								**0.700**	**0.718**	−0.618	−0.876	−0.234	−0.773
Se									0.155	−0.618	−0.584	−0.108	−0.245
Cd										−0.103	−0.720	−0.162	−0.855
Sn											0.441	0.000	0.103
Sb												**0.501**	**0.876**
Pb													0.414

Correlation coefficients have been calculated using the log value of the parameter contents to normalize their distribution ($n = 7$, statistically significant coefficients, $p < 0.05$, are in bold).

In one hand, positive and significant correlation ($p < 0.05$) was recorded between some metal concentrations, such as Cr/As ($r = 0.955$), Cr/Se ($r = 0.775$), Cr/Cd ($r = 0.649$), Co/As ($r = 0.864$), Co/Se ($r = 0.745$), Ni/As ($r = 0.927$), Ni/Cd ($r = 0.673$), Zn/Pb ($r = 0.607$), As/Se ($r = 0.700$), As/Cd ($r = 0.718$) and, Sb/Hg ($r = 0.876$). The positive correlations observed among these metal pairs suggest that these metals could have originated from common sources and may have similar transport pathways. In the other hand, some negative and significant correlations ($p < 0.05$) were observed, namely between metal pairs Hg/Sc ($r = -0.775$), Hg/Cr ($r = -0.685$), Hg/Fe ($r = -0.811$), Hg/Co ($r = -0.545$) suggesting that Hg did not originate from the same source than other metals and may have followed different environmental pathways (Poté et al., 2008; Haller et al., 2009; Atibu et al., 2021).

4. Conclusion

Integrated agroecosystems have been fostered to increase food production and food availability, especially in the highly populated urban areas of Kinshasa. This study focused on the evaluation of heavy metal's levels and distribution, as well as potential ecological risks in an integrated agroecosystem (CECOMAF, Kinshasa) with a significant agriculture and freshwater fish production for human consumption.

Analyses of metals were made in soils, sediments and biota samples. The results for agriculture soils and sediments showed similar metal contamination in both. The concentration of heavy metals (namely Cu, Zn, As, Cd, Pb and Hg) in the study area were significantly higher than concentrations in a reference area, and exceeded the reference limits recommended by Sediment Quality Guidelines (SQGs) and their Probable Effect Levels (PELs) for the protection of aquatic biota. The results for Contamination Degree (CD) and Ecological Risk (Eri) parameters indicated that soil and sediments from the study area were very highly contaminated and, in particular, the concentrations of Cd and Hg represent a very high ecological risk. The current levels of such contaminants may jeopardize the health of aquatic system and compromise aquatic biota.

The Spearman correlation matrix values suggested different sources and/or pathways for Hg when compared with other metals. Most base metals originate in urban activities and likely reach the agroecosystem with untreated wastewater and surface runoff. Globally, the analytical results combined with values from several ecological parameters give a robust indication that there is a significant contamination of the agroecosystem by several heavy metals originating from anthropic sources.

Metals in the earthworm *Pontoscolex corethrurus* displayed high levels for Cu, Zn, and Cd, which indicate high bioavalability of these metals in agriculture soils and, therefore, a possible easy transfer of such metals from soils to native plants and crops through root absorption. This deserves further investigation.

The high concentrations of metals in fish indicated bioaccumulation from water and food in the aquatic system. High concentrations of Cd and Pb in fish, above international recommended limits, are a potential threat to human health, especially for heavy consumers of fish.

Based on these results, it is concluded that there is an urgent need to implement suitable measures to reduce environmental discharges of heavy metals, to establish limits for contaminant levels in national regulations, and to adopt measures to minimize ecological risks. In particular, and taking into consideration the relevance of urban agroecosystems to feed the population, an environmental remediation process in the CECOMAF agroecosystem and N'djili River should be foreseen. Furthermore, a management and a monitoring plan are needed to ensure the sustainability of the agroecosystem and the production of healthy food for the population.

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Conflicts of Interest

The authors declare no conflict of interest.

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