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Assessment of Heavy Metal Pollution of Agricultural Soil, Irrigation Water, and Vegetables in and Nearby the Cupriferous City of Lubumbashi, (Democratic Republic of the Congo)

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Abstract: Lubumbashi (DR Congo)—the capital of copper mining—has been considered as one of the richest mining regions of the world for more than a decade. These riches have brought along multiple mining companies responsible for soil, river water and vegetable pollution, as in many African cities. The aim of the present study was to quantify and evaluate the pollution levels and the potential sources of soil, irrigation water and vegetable contamination by the metals As, Cd, Cr, Cu, Pb, Co and Zn in the urban gardens of Lubumbashi (DR Congo). The contamination, pollution and enrichment levels of the gardens were determined based on different indices in order to rank the soils. The results show that soils, waters and vegetables present contamination levels that represent a serious concern for human health. All soils presented contamination indices ranging from low (72% of the soils) to very high (3.4% of the soils) metal (copper, lead, zinc) contamination. The Cu and Cd contents varied between 1355 mg/kg et 236 mg/kg, much higher than the World Health Organisation (WHO) thresholds (100 mg/kg for Cu and 2 mg/kg for Cd). Moreover, the water used for crop and garden irrigation presented high Pb (57% of the waters), Fe (52%), Cu (19%) and Cd (10%) contamination levels, above the Association Française de Normalisation (AFNOR) U4441 toxicity thresholds (2 mg/kg for Cu; 0.1 mg/kg for Fe and 0.01 mg/kg for Pb) for crop irrigation. Finally, the vegetables produced in these gardens and sold in the local markets had very high metal content (47% contained Cu; 100% contained copper and cobalt) above the WHO standard (10 mg/kg for Cu, 2 mg/kg for Cd and 1 mg/kg for Co) for human consumption. In the face of these issues, it would be preferable to consider cheaper, more sustainable techniques that reduce soil-to-plant metal transfer.

Keywords: spatial variability; pollution indices; market gardens; Lubumbashi

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1. Introduction

Lubumbashi, the "capital of copper "located in the southeast of the Democratic Republic of the Congo, has been considered as one of the richest mining regions of the world for more than a decade. In recent years, the Haut-Katanga province and more particularly the city of Lubumbashi have witnessed the expansion of the mining industry and the subsequent construction of numerous ore extraction plants. Today, numerous studies show a link between human activities (e.g., mining activities, landfills) and environmental contamination, especially soil, water and plant contamination [1–4]. In Lubumbashi, these companies are the source of the contamination of agricultural and household soils, market garden products and river waters [5,6]. The extraction processes of these mining activities—especially metalworking and pyrometallurgy—have contributed to soil and water contamination from atmospheric emissions and mining effluents, in contrast to the sites located a distance aways from the pollution cone [7–9]. These phenomena are the source of diverse health and environmental issues [7,8,10]. In Pakistan, for example, similar situations to those noted in Lubumbashi have been observed [11,12]. The authors showed that the application of mining effluents as organic amendments was the main cause of soil contamination in market gardens. Furthermore, the studies by [12–14] showed that the vegetables produced in the market gardens and sold in Lubumbashi markets presented high levels of trace metal elements (TMEs) above the WHO toxicity threshold. Southeast of Casablanca (Morocco), crop irrigation with industrial effluents containing high loads of heavy metals was found to contaminate five vegetable crops with As, Cd, Cr and Cu [15–17]. Similar situations have been observed in Lubumbashi, where market gardeners use water with high loads of metals to water their crops. This represents a notable hazard for vegetable consumption and human health, as very high metal contents have been detected in water and vegetables alike [9,18,19]. Another issue under strong criticism in the city of de Lubumbashi is the growing number of poorly managed \landfills, which are potential sources of soil and urban market garden contamination. Very high heavy metal concentrations have been found in the soils of former landfills [20–22].

Similar situations to Lubumbashi have been observed in the agricultural region of Sri Lanka where heavy metal-rich effluents (Cd, Fe, Pb) discharged into watercourses have caused chronic renal disease in around 5000 people aged 5 to 50. This disease was due to the consumption of rice irrigated with water containing a strong load of cadmium. Heavy metal contamination levels ten times as high as the toxicity threshold were found in the inhabitants' urine [23–26]. In the same vein, the studies by [25,27] carried out in Lubumbashi showed that contamination of pregnant women by uranium and manganese led to the birth of three babies presenting malformations known as holoprosencephaly. Very high levels of uranium and manganese were found in their mothers' urine and blood. Therefore, the people currently living in and nearby Lubumbashi are going through a health crisis.

In Lubumbashi, [28] showed that vegetables mainly come from 23 market gardens and are sold in four main markets. This study stands out from other research in that the authors succeeded in analysing the different components (soils, water, plants) of market gardens separately. However, a study encompassing all three environmental components within the global setting of Lubumbashi urban market gardens has never been undertaken.

In this context, the aim of the present study was to quantify and evaluate the pollution levels and the potential sources of soil, irrigation water and vegetable contamination by the metals As, Cd, Cr, Cu, Pb, Co and Zn in the urban gardens of Lubumbashi (DR Congo). The first step consisted of identifying market gardens and markets. Then, the concentrations in MTEs—Al, As, Cd, Co, Cu, Fe, Pb and Zn—in the vegetables were analysed to determine the safety of urban market products in Lubumbashi.

2. Materials and Methods

2.1. Identification of the Market Gardens and Urban Markets of Lubumbashi

Based on FAO mapping carried out in 2008 [28] and on a separate survey led within the framework of this study, 40 market gardens and 33 Lubumbashi markets meeting our selection criteria were identified (Appendix A.1).

The survey carried out within the framework of the present study led to the identification of 17 new market gardens and 29 new markets in addition to those listed by the FAO [8,16]. Among the market gardens, 29 were selected following the criteria of the present study, as detailed below. These market gardens are the main local vegetable suppliers of the city of Lubumbashi (Appendix A.2).

The criteria for including market gardens in our list were that they should be farmed by at least five market gardeners and that at least three of the most commonly cultivated vegetables in Lubumbashi be grown, among *Brassica chinensis*, *Amaranthus vulgaris*, *Brassica* *oleracea var. capitata, Lycopersicum esculentum Mill, Allium porrum, Lactuca sativa, Allium cepa., Brassica carinata, Abelmoschus esculentus, Daucus carota, Beta vulgaris, Petroselinum crispum, Solanum melongena* L., *Apium graveolens var. dulce, Beta vulgaris subsp. vulgaris, Brassica oleracea var. botrytis, Solanum tuberosum, Raphanus sativus, Hibiscus sabdariffa* L., and *Cucumis sativus* L. Moreover, the markets had to be (i) located in Lubumbashi, (ii) run by at least five vegetables sellers, and (iii) selling local vegetables within the market gardens of Lubumbashi [15].

For further analysis of the market gardens on the list, the geographic coordinates were recorded using a global positioning system (GPS; Garmin Montana 680t) and treated with the mapping software program ArcGIS 10.5 registered in the geographic coordinate system (GCS-WGS 84). The same procedures were applied to map the Lubumbashi markets where these vegetables are sold.

2.2. Sampling Methods

2.2.1. Soils

Soil samples were collected from five different points of each urban and peri-urban market garden, at 0–20 cm depth, and each batch of five samples was pooled to form a composite sample kept and analysed in the laboratory. Each composite sample was open-air-dried for 25 days, and then ground in a porcelain mortar and sieved to 2 mm.

2.2.2. Water Samples

Only 21 out of the 40 identified market gardens had an easily accessible water collection point for determining the safety of the waters used to irrigate the crops. Five 100-mL water samples were collected and pooled to form a composite sample. The samples were collected between March and May 2019, a favourable period for market gardening in Lubumbashi. They were kept in a refrigerator at 4 °C for seven days and then sent to the laboratory of the Office de Contrôle du Congo (OCC/DR Congo).

2.2.3. Vegetables

The study was focused on four vegetables—*Brassica chinensis*, *Brassica carinata*, *Amaranthus vulgaris* and *Spinacia oleacera*—for the following reasons:

The vegetables had to be identified among the 20 species grown in the city of Lubumbashi within the framework of the different projects run in its market gardening sector (HUP, 2000).

They had to be grown intensively in Lubumbashi [11].

Previous studies had to show that they presented a risk of MTE contamination [11,29].

To collect the vegetable samples, we first questioned the sellers to determine where the vegetables had been produced, and only those from Lubumbashi market gardens were purchased. Then, composite samples were formed, the vegetables were washed under city tap water to remove dust particles, and oven-dried at 105 °C for 24 h. The dried samples were ground in a porcelain mortar, and 100 g of powder were kept for analysis.

Metal quantification in the soils, waters and vegetables was conducted.

The chemical analyses aimed at determining the total heavy metal (Al, As, Cd, Co, Cu, Fe, Pb and Zn) concentrations in the soils of Lubumbashi market gardens were carried out using a portable X-ray fluorescence spectrophotometer (XRF, Olympus Delta Classic Plus, model DCC-4000) calibrated with stainless steel alloy 316 [18]. The soil exchangeable Cu, Co and Pb concentrations were determined by $CaCl_2$ 0.01 M extraction, and the heavy metal concentrations were determined by ICP OES atomic absorption spectrometry (AAS, VARIAN 220, Agilent Technologies, Santa Clara, CA, USA) [30].

The heavy metal (Al, As, Cd, Co, Cu, Fe, Pb and Zn) contents of the water samples were determined by inductively coupled plasma mass spectrometry (ICP-MS) [20,31]. The heavy metal (Cu, Co, Cd and Pb) contents of the vegetables were determined by acid mineralisation with HNO_3+$HClO_3$, and measurements were made by flame atomic absorption spectroscopy (AAS, VARIAN 220, Agilent Technologies, Santa Clara, CA, USA) [32].

2.3. Indices of Agricultural Soil Contamination and Pollution

2.3.1. Contamination Factor

The contamination factor can be calculated as the ratio of the measured concentration of a given metal in the soil to the background value of that metal expressed as a percentage (Al, As, Cd, Co, Cu, Fe, Pb and Zn) [33,34] (Appendix A.3):

$$\mathrm{CFi} = \frac{\mathrm{Cmetal(sample)}}{\mathrm{Cmetal(background)}}$$

Consequently, CF_i designates the contamination factor of metal I, and four classes can be established: CF < 1 = low contamination, $1 \leq$ CF < 3 = moderate contamination, $3 \leq$ CF < 6 = high contamination, and CF $\geq$ 6 = very high contamination. The soil geochemical background values used in the present study are those of the soils of the city of Lubumbashi [35].

2.3.2. Soil Pollution Load Index

The pollution load index (PLI) is used to determine the level of the pollution load of all MTEs (Al, As, Cd, Co, Cu, Fe, Pb and Zn) on the sampled sites. It is the geometric mean of the contamination factors, according to the following formula [36]:

$$PLI = \sqrt[n]{CFix\ CFjx \ldots CFn}$$

where n is the number of MTEs in the present study. A $PLI \leq 1$ highlights MTE pollution loads close to the background geological concentration, a $PLI = 1$ highlights a low pollution level, while a $PLI > 1$ highlights significant soil pollution (Appendix A.5).

2.3.3. Enrichment Factor

The enrichment factor is the ratio of the concentration of a given metal (Cx) to the concentration of the reference element (CFe) in a given sample divided by the ratio of the elemental concentration of a given element to the concentration of the reference element in the Earth's crust [37]. However, for this study, the EF was evaluated to determine the level of contamination and the influence of anthropogenic activities in the urban vegetable garden soils of Lubumbashi. Thus, the geochemical normalization of the data for heavy metals has a conservative Al (Sinex and Wendy), Al [38,39], Fe [40–42]. To determine the enrichment factor of vegetable soils, Fe was used as a conservative tracer to distinguish natural from anthropogenic components:

$$EF = \frac{\mathrm{Cx/CFe(Sample)}}{\mathrm{Cx/CFe(Control)}}$$

Thus, seven classes were distinguished depending on the enrichment factor, namely EF < 1, $1 \leq$ EF < 3, $3 \leq$ EF < 5, $5 \leq$ EF < 10, $10 \leq$ EF < 25, $25 \leq$ EF < 50 and EF > 50, meaning that MTE enrichment can be null, low, moderate, moderately high, high, very high and exceptionally high, respectively [43] (Appendix A.4).

2.4. Habit Description

The market gardens of Lubumbashi are diverse in their particular characteristics. However, we have classified them according to their likely sources of heavy metal contamination. Thus, the market gardens with a water supply source were put into one group (Table 1).

Table 1. Description of the ecosystem at study locations near pollution sources.

Market Gardens	Habitat Description	Sources of Pollution in Gardens
Bongonga	Mining Effluents/Dump sites	Kafubu River Middle
Chem-chem	Mining effluents/MET-rich subsoil	Kafubu River Downstream
Daipen/Kashamata	Mining Effluents/Dumpsites	Kafubu River Upstream
Kabetsha	Mining Effluents/Dumpsites	Tshamilemba River
Kafubu	Mining effluents/MET-rich subsoil	Kafubu River Middle
Kalebuka	Mining Effluents/Dumpsites	Kafubu River Middle
Kalubwe	Mining Effluents/Dumpsites	Middle Lubumbashi River
Kalulako	Mining effluents/MET-rich subsoil	Kafubu River Downstream
Kamakanga	Metal-rich subsoil	Kafubu River Middle
Kamatete	TME-rich subsoil/Mining effluents	Lubumbashi River Upstream
Kamilombe	Mining effluents/MET-rich subsoil	Kafubu River Downstream
Kamisepe	Mining Effluents	Lubumbashi River Upstream
Kantumbwi	Mining Effluents/Dumpsites	Middle Lubumbashi River
Kasungami	Mining Effluents/Dumpsites	Kafubu River Upstream
Katemo	Effluents miniers/Metal-rich subsoil	Kafubu River Downstream
Kawama	MTE-rich subsoil	Kafubu River Downstream
Kikula/Sambwa	Metal-rich subsoil	Kafubu River Downstream
Kilobelobe	Mining Effluents/Dumpsites	Kafubu River Middle
Kinsense	Mining Effluents/Dumpsites	Tshamilemba River
Kinsevere (Manoah)	Effluents miniers/Metal-rich subsoil	Kiswishi River
Kitanda	TME-rich subsoil/Mining effluents	Kafubu River Downstream
Luano	MTE-rich subsoil	Luano River
Maendeleo	Dumpsites	Well
Mashimikila	Effluents/Metal-rich subsoil	Kafubu River Downstream
Mwenda	Metal-rich subsoil	Kafubu River Downstream
Penga-penga	Mining effluents/MET-rich subsoil	Régideso
Tingi-Tingi	Mining Effluents/Dumpsites	Tingitingi River
Tshamalale	Mining Effluents/Dumpsites	Lubumbashi River Upstream
Tshamilemba	Mining Effluents/Dumpsites	Tshamilemba River

2.5. Data Analyses

The data were analysed using Minitab 21.3.1.0 statistical software. The one-level analysis of variance was used to determine the significance levels of the different sources of soil contamination in the market gardens as well as the vegetables sold in the urban markets of Lubumbashi. The Tukey test at the 5% level was used to compare two means.

3. Results

3.1. Identification of the Market Gardens and Urban Markets of Lubumbashi

A total of 40 market gardens and 33 markets initially met our criteria. The survey carried out within the framework of the present study allowed us to identify 17 new market gardens and 29 new markets in addition to those selected by the FAO [28,44].

The 29 markets with at least five local vegetable sellers selected within the framework of the present study were used to evaluate the safety of the vegetables sold and consumed by Lubumbashi people (Figure 1).

3.2. Characterisation of MTE-like Pollutants in Market Garden Soils, Waters and Vegetables

The one-way analyses of variance revealed that there was no significant difference between the different potential sources of contamination ($p > 0.05$) and the levels of heavy metals in the soils.

Laboratory analyses showed that around 80%, 21%, 17% and 7% of the soils from the 29 selected market gardens were contaminated with copper, zinc, cadmium and lead, respectively. The cobalt concentration was not measured due to a technical problem (Table 2).

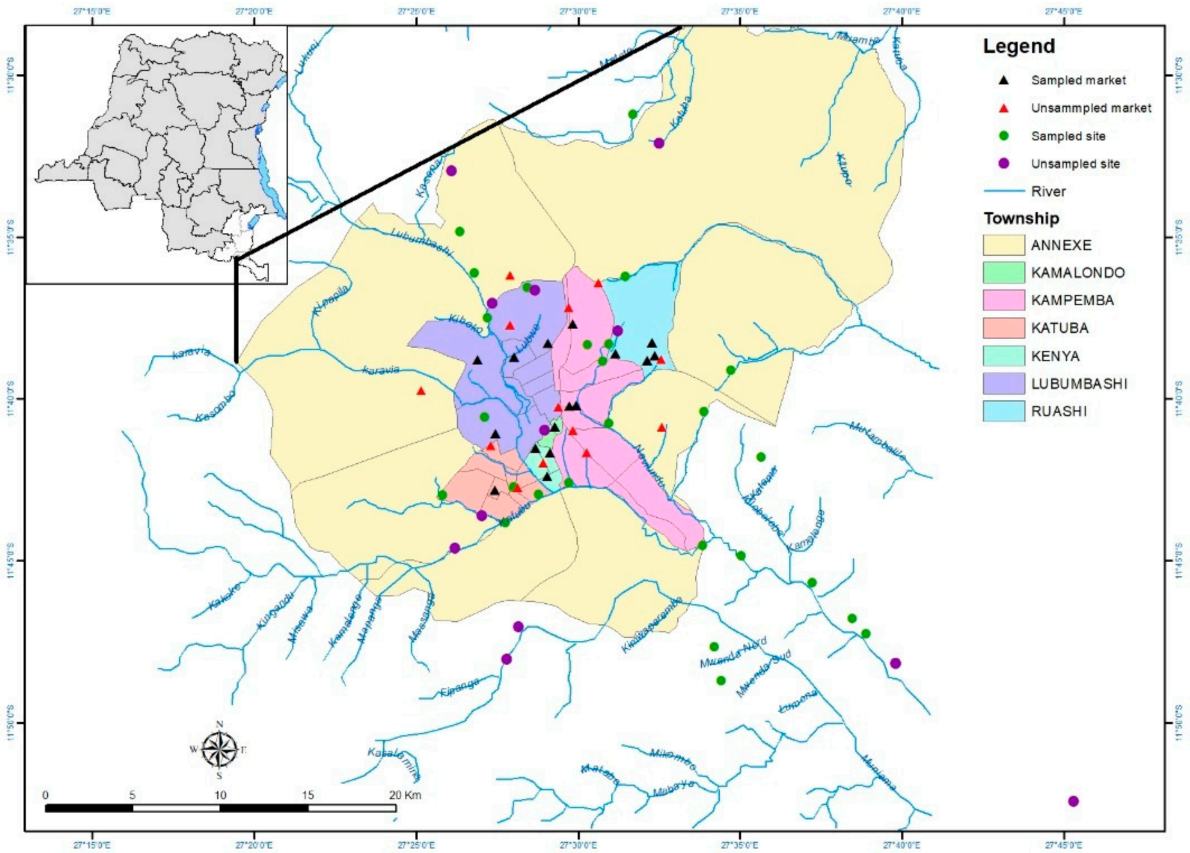

Figure 1. Map of the market gardens and urban markets of Lubumbashi (Mununga, K.F, 2022).

Table 2. XRF determination of metals in 29 urban and peri-urban market gardens of Lubumbashi (mg/kg).

Contamination Source Categories	Fe	Cd	Cu	Pb	Cr	Zn
Kafubu River Middle	2.762 ± 0.94 [a]	47.2 ± 94.40 [a]	170.6 ± 104.66 [a]	9.6 ± 19.20 [a]	35.8 ± 31.68 [a]	118.4 ± 98.96 [a]
Kafubu River Downstream	3.092 ± 1.27 [a]	13.33 ± 20.88 [a]	293.82 ± 297.82 [a]	44.22 ± 67.18 [a]	31.44 ± 29.26 [a]	234.61 ± 254.27 [a]
Kafubu River Upstream	2.24 ± 0.33 [a]	0.00 ± 0.00 [a]	266.33 ± 165.59 [a]	68.00 ± 82.43 [a]	49.66 ± 5.24 [a]	169.00 ± 169.94 [a]
Tshamilemba River	2.78 ± 0.66 [a]	0.00 ± 0.00 [a]	144.66 ± 92.17 [a]	11.33 ± 16.03 [a]	32.00 ± 22.76 [a]	70.33 ± 46.58 [a]
Middle Lubumbashi River	3.69 ± 1.36 [a]	13.33 ± 18.86 [a]	359.33 ± 351.5 [a]	0.00 ± 0.00 [a]	51.00 ± 5.10 [a]	126.33 ± 123.83 [a]
Lubumbashi River Upstream	2.71 ± 0.69 [a]	0.00 ± 0.00 [a]	154.66 ± 74.02 [a]	8.66 ± 12.26 [a]	21.00 ± 29.70 [a]	81.66 ± 42.90 [a]
Effects of sources of contamination (p-value)	0.74	0.738	0.74	0.455	0.598	0.995

Superscript letters indicate pairwise differences in means.

3.2.1. Contamination Factors, Enrichment Factors and Pollution Indices of the Soils

Based on the calculated soil contamination factors of Lubumbashi market gardens, we determined four MTE contamination classes, namely low contamination with iron, copper, lead and zinc (96.55%, 72.41%, 93.10% and 62.07% of the soils, respectively); moderate contamination with copper, lead and zinc (27.59%, 6.9% and 27.59% of the soils, respectively); high contamination with iron and zinc (3.45% and 6.9% of the soils, respectively); and very high contamination with zinc (3.45% of the soils).

The enrichment factor revealed five enrichment classes, i.e., no enrichment (41.38%, 79.31% and 34.48% of the soils for copper, lead and zinc, respectively); low enrichment (48.28%, 13.79% and 37.93% of the soils for the same metals); moderate enrichment (3.45%, 3.45% and 13.79% of the soils); medium-high enrichment (6.9%, 3.45% and 10.34% of the soils); and high enrichment (3.45% of the soils, for zinc only).

Finally, the soil pollution index was calculated to determine the pollution levels of the soils of the market crops of the city of Lubumbashi. Two classes were established, namely unpolluted gardens (79.31%) and severely polluted gardens (20.69%), whatever the MTE. We considered soil contamination as any increase of components inducing a detectable negative effect on soil functioning (Table 3; Figures 2–4), while we considered soil pollution as an increase of components within a given environment that gradually becomes severe and deleterious and perturbs the functioning of soils up to their degradation [45].

Table 3. Contamination, enrichment and pollution levels of the soils of 29 urban market gardens of Lubumbashi.

Market Gardens	Contamination Factor				Enrichment Factor			Soil Pollution Index (PLI)
	Fe	Cu	Pb	Zn	Cu	Pb	Zn	
Bongonga	0.45	0.13	0	0.17	0.29	0	0.37	0.21
Chem-Chem	0.55	2.97	2.69	8.17	5.4	4.87	14.82	2.45
Daipen/Kashamata	0.29	0.45	0.24	0.33	1.56	0.84	1.16	0.32
Kafubu	0.32	0.32	0	1.01	1.01	0	3.18	0.47
Kabetsha	0.3	0.25	0	0.23	0.84	0	0.79	0.26
Kalebuka	0.15	0.5	0	0.29	3.28	0	1.95	0.28
Kalubwe	0.67	1.87	0	1.67	2.78	0	2.49	1.28
Kalulako	0.25	0.52	0	0.47	2.11	0	1.9	0.4
Kamakanga	0.45	0.17	0	0.24	0.38	0	0.54	0.27
Kamatete	0.37	0.11	0	0.19	2.52	0	6.96	1.49
Kamilombe	0.57	1.44	0	3.98	1.81	0	1.61	0.36
Kamisepe	0.25	0.46	0	0.41	0.4	0	0.64	0.16
Kantumbwi	0.24	0.1	0	0.16	0.87	0	0.82	0.23
Kasungami	0.26	0.22	0	0.21	0.28	0	0.45	0.22
Katemo	0.44	0.12	0	0.19	2.32	0.84	2.58	0.85
Kawama	0.56	1.31	0.47	1.46	2.08	1.03	2.31	0.32
Kikula/Sambwa	0.21	0.44	0.22	0.49	1.51	1.16	3.15	0.77
Kilobelobe	0.5	0.76	0.58	1.58	1.19	0.83	1.51	0.55
Kinsense	0.5	0.59	0.41	0.76	0.26	0.22	0.48	1.84
Kinsevere (Manoah)	4.52	1.17	0.98	2.19	2.71	0.85	5.41	1.26
Kitanda	0.67	1.81	0.57	3.62	1.39	1.78	3.05	0.91
Luano	0.55	0.76	0.97	1.67	2.97	6.13	6.23	1.19
Maendeleo	0.36	1.08	2.24	2.27	6.89	2.67	3.85	0.96
Mashimikila	0.33	2.29	0.89	1.28	0.84	0	1.35	0.18
Mwenda	0.17	0.14	0	0.23	0.69	0	0.48	0.4
Pengapenga	0.58	0.4	0	0.28	1.01	0	1.06	0.36
Tingi-Tingi	0.35	0.35	0	0.37	0.3	0	0.51	0.2
Tshamalale	0.48	0.45	0.32	0.77	0.94	0.66	1.6	0.48
Tshamilemba	0.33	0.11	0	0.18	0.34	0	0.55	0.19

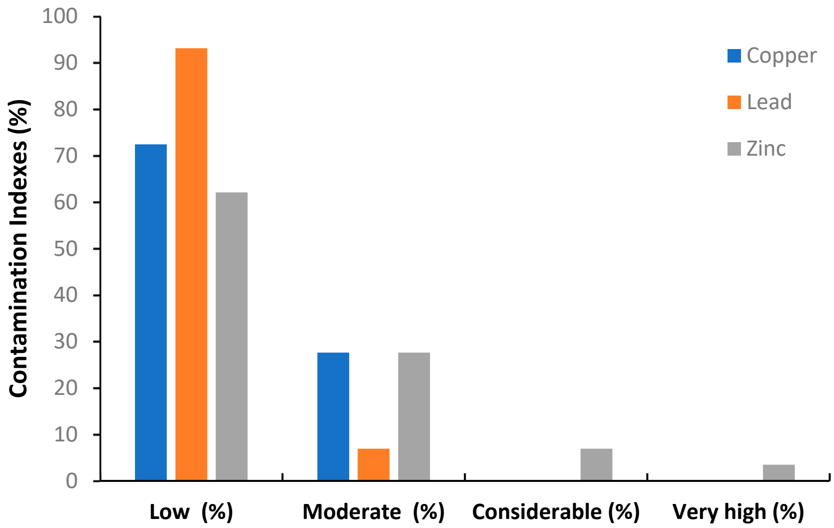

Figure 2. Distribution of the market gardens of Lubumbashi according to the contamination factors.

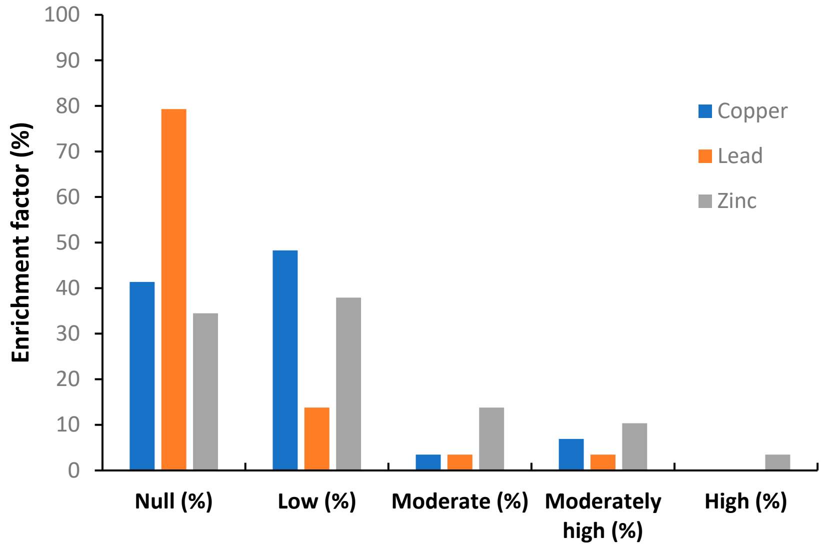

Figure 3. Distribution of the market gardens of Lubumbashi according to the enrichment factors.

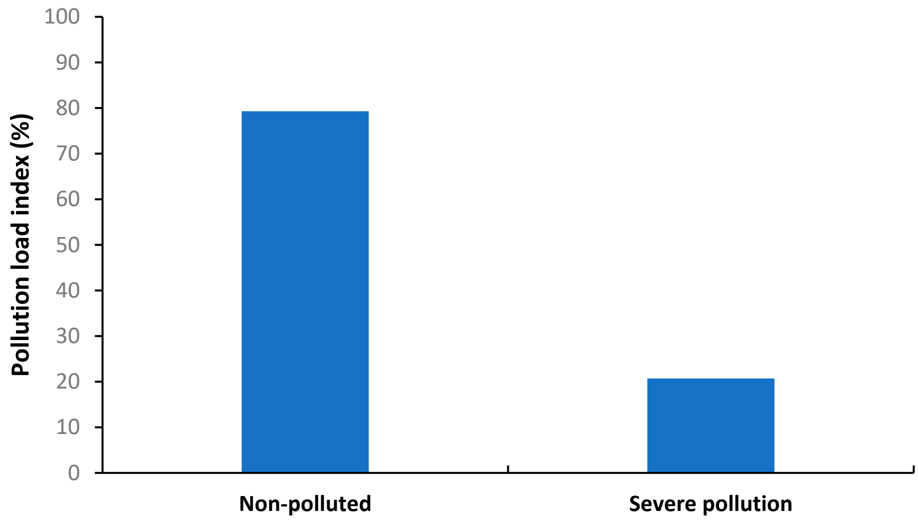

Figure 4. Distribution of the market gardens of Lubumbashi according to their pollution load index.

The maps show that among all the market gardens of Lubumbashi, those located along the northeast axis of the city present a medium-high contamination level, higher than those located along the southeast axis. MTE concentrations tend to decrease at a distance from the epicentres. Nevertheless, the presence of several mining facilities is linked to pollution hotspots. No clear trend was identified to describe the distribution of the other metals. However, these spatial representations highlight that nearly all the soils of Lubumbashi market gardens present some degree of MTE pollution, enrichment and contamination (Figures 5–8).

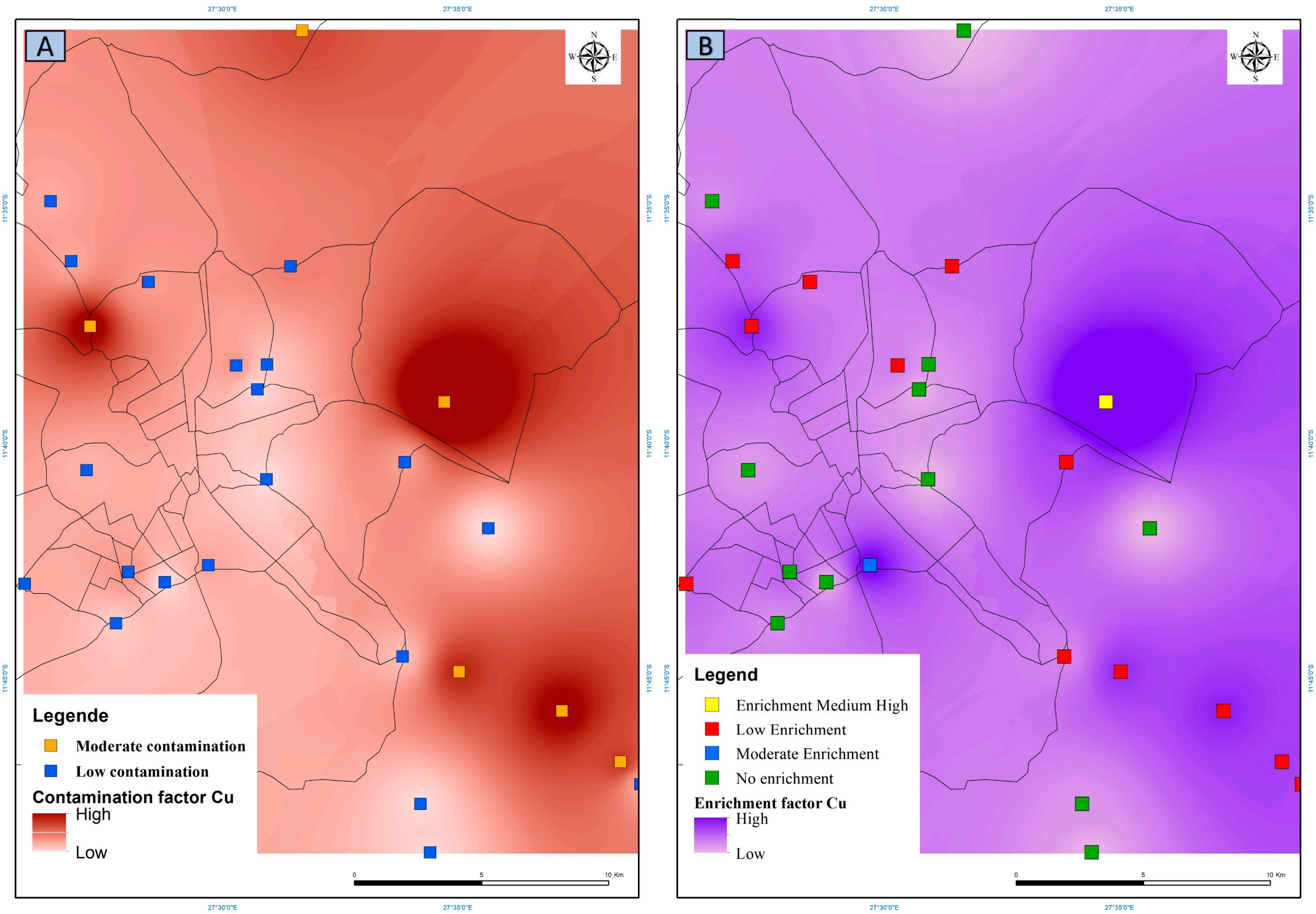

Figure 5. Spatial distribution of metals (Cu) in the market garden soils according to their contamination factor (**A**) and enrichment factor (**B**).

3.2.2. Quality of Crop Irrigation Water in the 21 Selected Market Gardens

The laboratory results showed that nearly 57%, 52%, 19%, 10% and 5% of the water sources of the gardens were contaminated by lead, iron, copper, cadmium/cobalt and zinc, respectively. As for arsenic, no pollution effect was noted in any of the market gardens in and near Lubumbashi (Table 4).

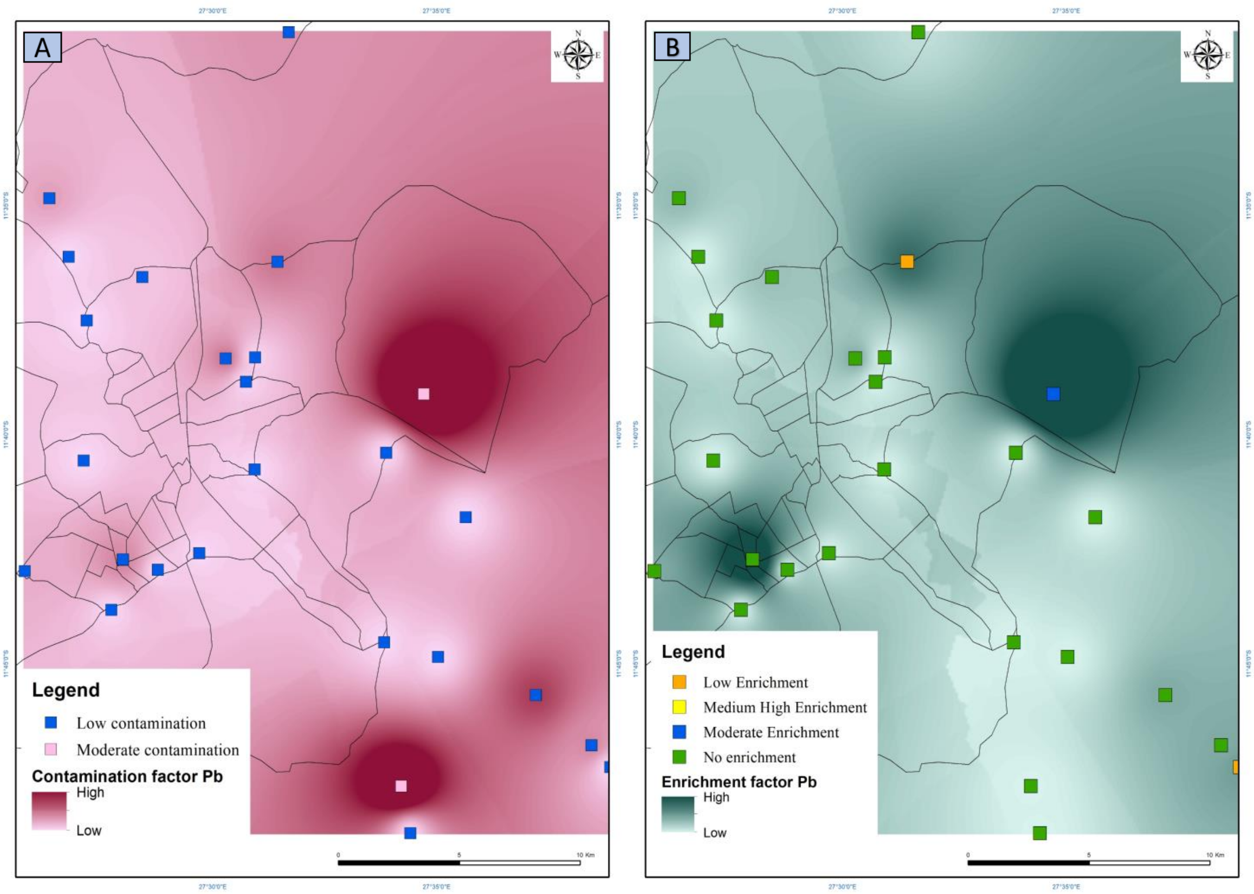

Figure 6. Spatial distribution of metals (Pb) in the market garden soils according to their contamination factor (**A**) and enrichment factor (**B**).

Table 4. Trace metal elements (mg/L) in the irrigation waters of the market garden crops of Lubumbashi gardens. Legend: Cd, cadmium; Cu, copper; Pb, lead; Co, cobalt; Al, aluminum; As, arsenic; Fe, iron; Zn, zinc; bold figures, contents above the standard.

Market Gardens	Cd	Cu	Pb	Co	As	Fe	Zn
Daipen Kisanga	0.003	0.012	0	0.012	0	0.316	0
Kafubu	0.001	3.01	0.028	0.006	0	0.381	0
Kalanda	0.002	2.51	0.049	0.012	0.02	1.175	0.002
Kalebuka	0.248	9.25	0.451	0.86	0	4.601	10.16
Kalubwe	0.002	0.019	0	0.007	0	0.782	0
Kalulako	0.001	0.129	0.038	0.018	0.027	0.185	0.065
Kamakanga	0	0.009	0.116	0.012	0	0.154	0
Kamasaka	0.002	0.014	0.017	0.007	0.022	0.144	0
Kamisepe	0.002	0.009	0.045	0.006	0.034	0.146	0
Kasungami	0.004	0.022	0.012	0.01	0.046	0.182	0.002
Katemo	0.002	0.011	0	0.008	0	0.097	0
Kawama	0.003	0.008	0.052	0.01	0	0.032	0
Kinsense	0	0.026	0.032	0.019	0.059	0.217	0.043
Kitanda	0.002	0.009	0	0.008	0.085	0.529	0.002
Maendeleo	0	0.019	0.043	0.011	0.07	0.242	0.028
Mashimikila	0.001	0.033	0.038	0.01	0	2.432	0.013
Penga-Penga	0.004	0.007	0.06	0.009	0	0	0
Sambwa	0.002	0.007	0.048	0.002	0	0.019	0
Tingi-Tingi	0	0.02	0	0.007	0	0.203	0.023
Tshamalale	0.002	0.035	0	0.005	0	1.238	0.017
Tshamilemba	0.001	2.031	0.048	0.005	0.008	0.011	0
Toxicity threshold (mg/L)	0.003	2	0.01	0.05	0.1	0.1	2

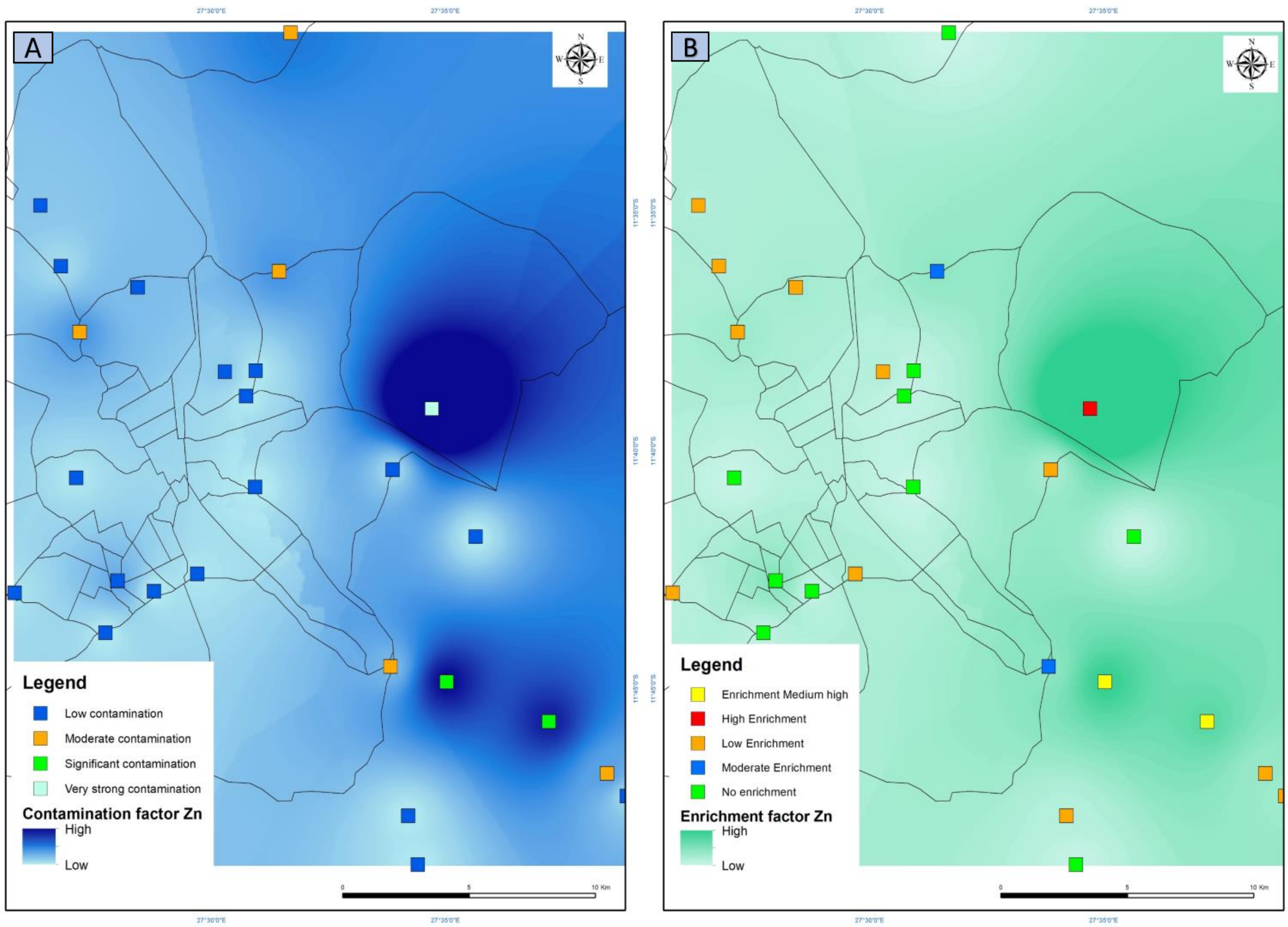

Figure 7. Spatial distribution of metals (Zn) in the market garden soils according to their contamination factor (**A**) and enrichment factor (**B**).

The analyses showed that river waters were generally more or less contaminated than well waters: 33.3%, 23.8%, 9.5% and 4.7% of the river waters were contaminated with iron, lead, copper and cadmium, respectively, while 33.3%, 28.57% and 9.5% of the well waters were contaminated with lead, iron and copper, respectively, and 14.2%, 9.5% and 4.7% of rainwaters were contaminated with lead, iron and cadmium, respectively.

The results and their mapping showed that similar to soils, most of the contaminated waters of Lubumbashi market gardens were found along the northeast (Ruashi–Kafubu) axis of the city. Moreover, these gardens are located near a pollution cone and close to effluents discharged into the rivers (Figures 9–12). The waters of the other market gardens located along the Lubumbashi–Kasumbalesa and Lubumbashi–Kimbeimbe axes showed medium contamination levels.

3.2.3. Safety of the Vegetables Sold in the 33 Selected Markets of Lubumbashi

The laboratory results showed that nearly all the vegetables sold on the 33 selected markets of Lubumbashi were contaminated with heavy metals, including Cd, Cu and Co. The analysis of variance showed that the use of several vegetables did not significantly influence the heavy metal concentrations ($p > 0.05$). However, the heavy metal concentrations remained above the contamination thresholds (Table 5). Only the Pb contents were below the WHO and AFNOR U4441 standards for vegetable consumption by humans. Among the four selected vegetables sold on Lubumbashi markets, *Brassica chinensis* presented the highest heavy metal (Cu, Cd and Co) contents, followed by *Amaranthus vulgarus, Spinacia oleracea* and *Brassica carinata* (Table 5).

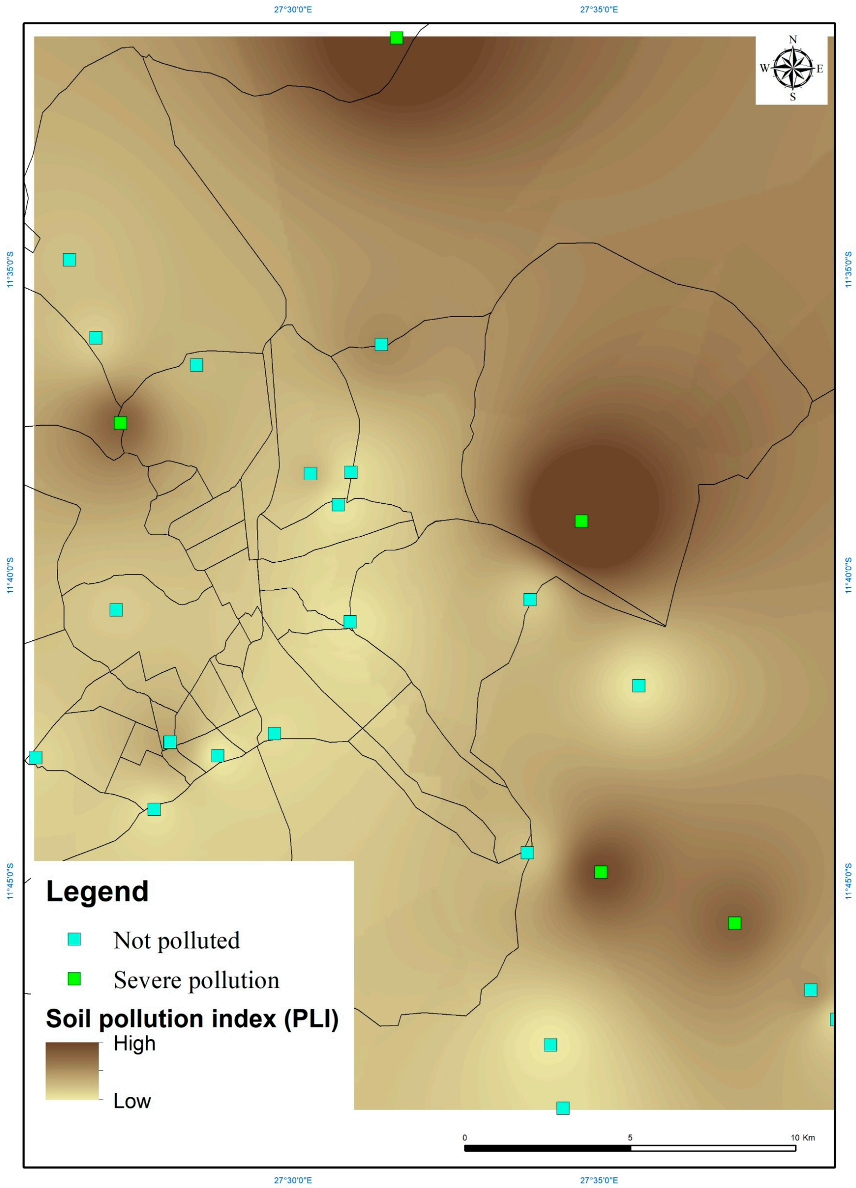

Figure 8. Spatial distribution of metals in the market garden soils according to their Soil pollution index.

Table 5. Mean heavy metal concentrations (mg/kg) in 4 crops sold in the 33 selected markets of Lubumbashi (AFNOR U4441 standard).

Plant Species	Co	Cd	Pb	Cu
Amaranthus vulgarus	4.41 ± 4.15 [a]	3.11 ± 3.56 [a]	1.27 ± 0.94 [a]	51.94 ± 61.17 [a]
Brassica chinensis	4.18 ± 2.65 [a]	2.03 ± 1.86 [a]	2.82 ± 2.94 [a]	44.93 ± 31.03 [a]
Spinacia oleracea	5.51 ± 11.48 [a]	1.25 ± 1.53 [a]	0.96 ± 1.22 [a]	21.25 ± 26.62 [a]
Brassica carinata	9.46 ± 18.33 [a]	0.82 ± 0.79 [a]	1.57 ± 2.49 [a]	27.53 ± 33.28 [a]
Species effects (*p*-value)	0.834	0.518	0.685	0.502

Superscript letters explain the differences in the pairwise means.

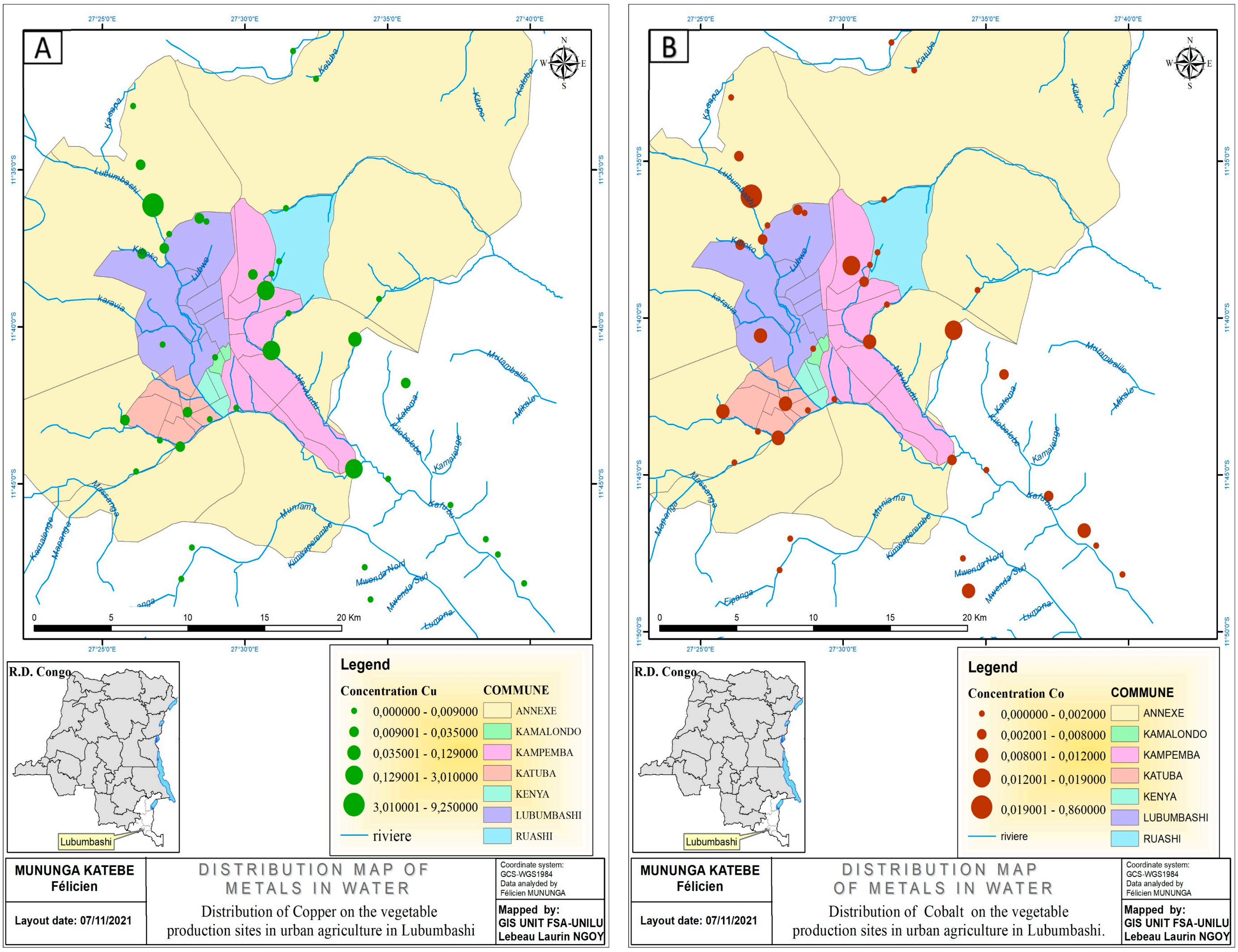

Figure 9. Spatial distribution of metals (Cu and Co) in the waters of Lubumbashi market gardens.

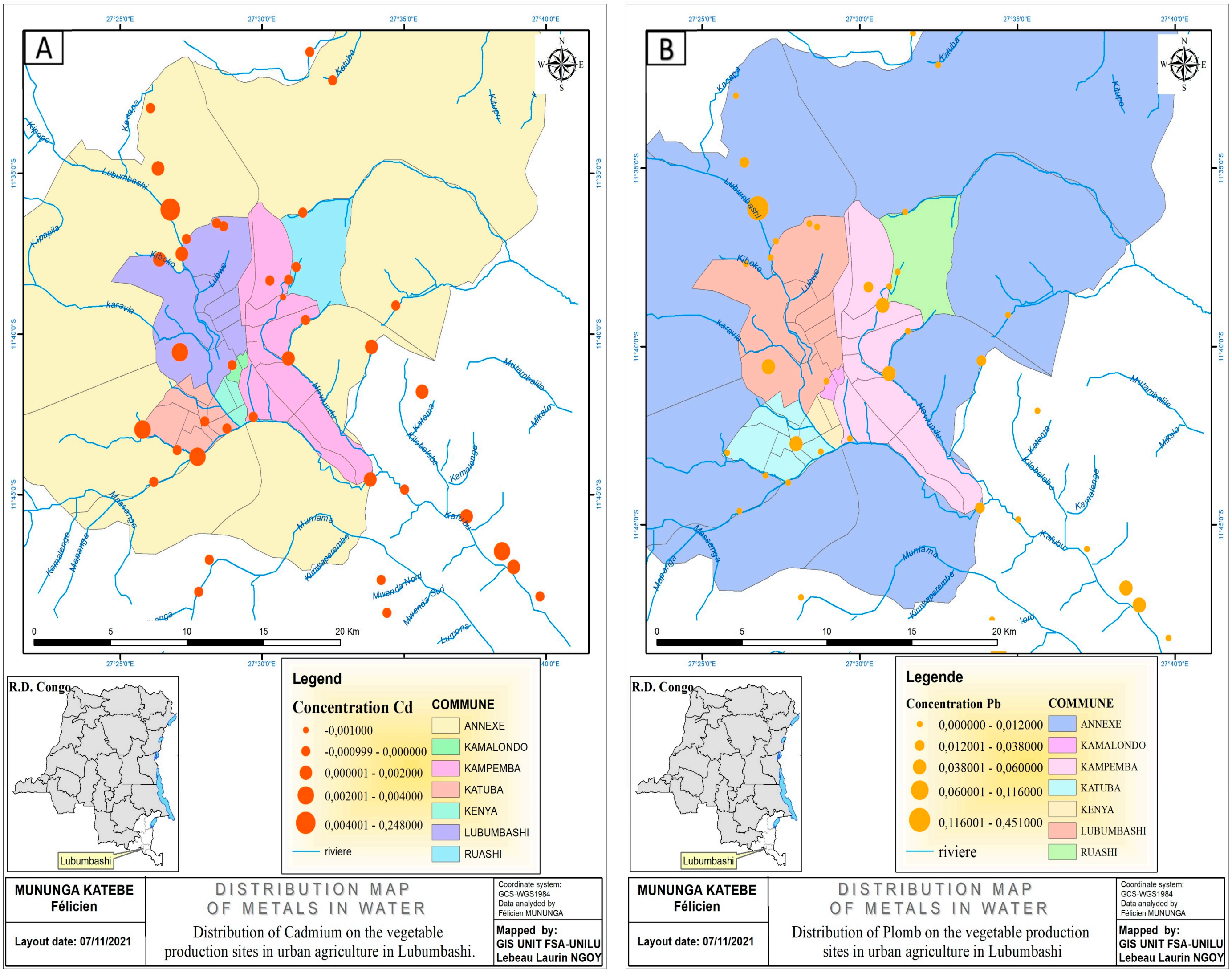

Figure 10. Spatial distribution of metals (Cd and Pb) in the waters of Lubumbashi market gardens.

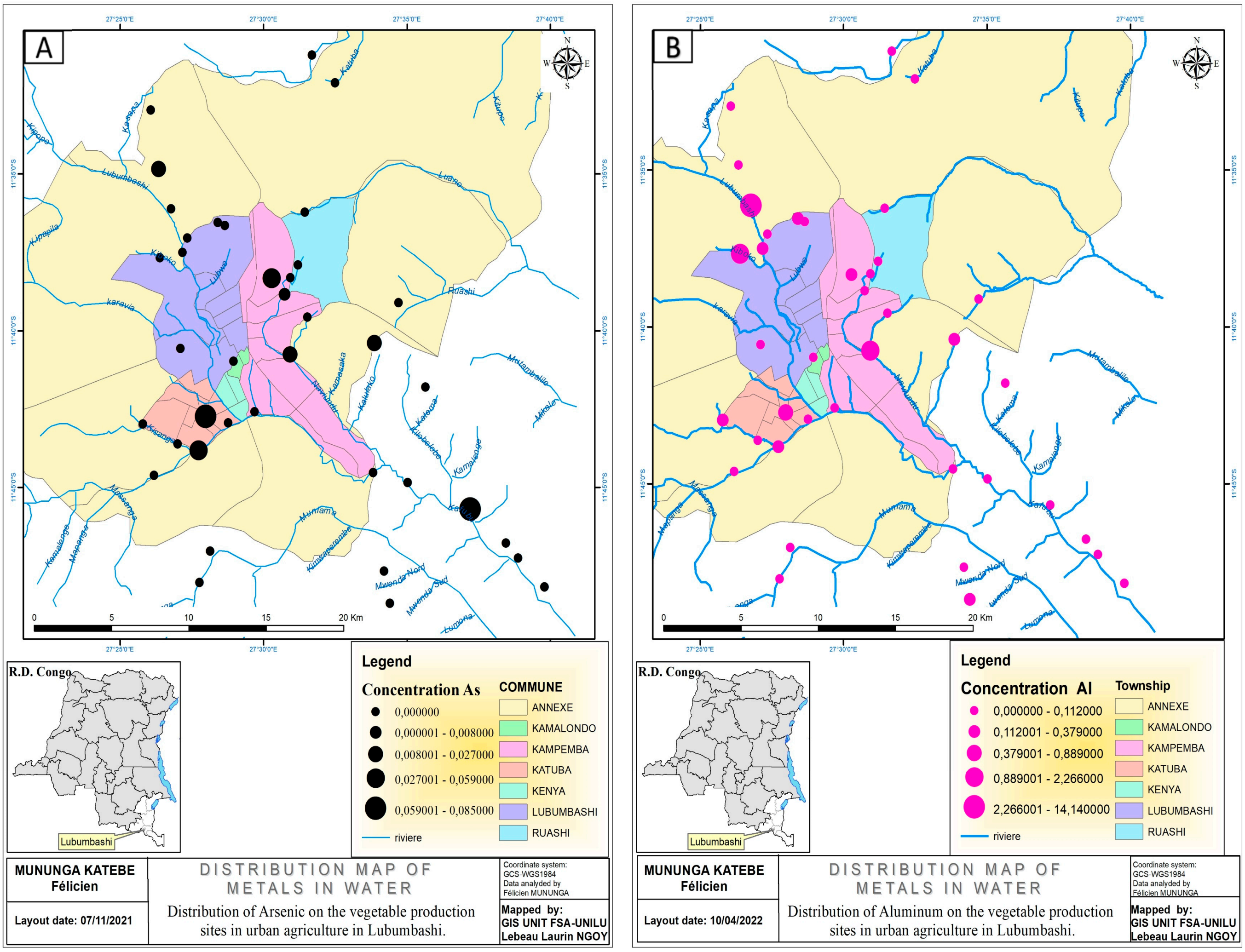

Figure 11. Spatial distribution of metals (As and Al) in the waters of Lubumbashi market gardens (**A**,**B**).

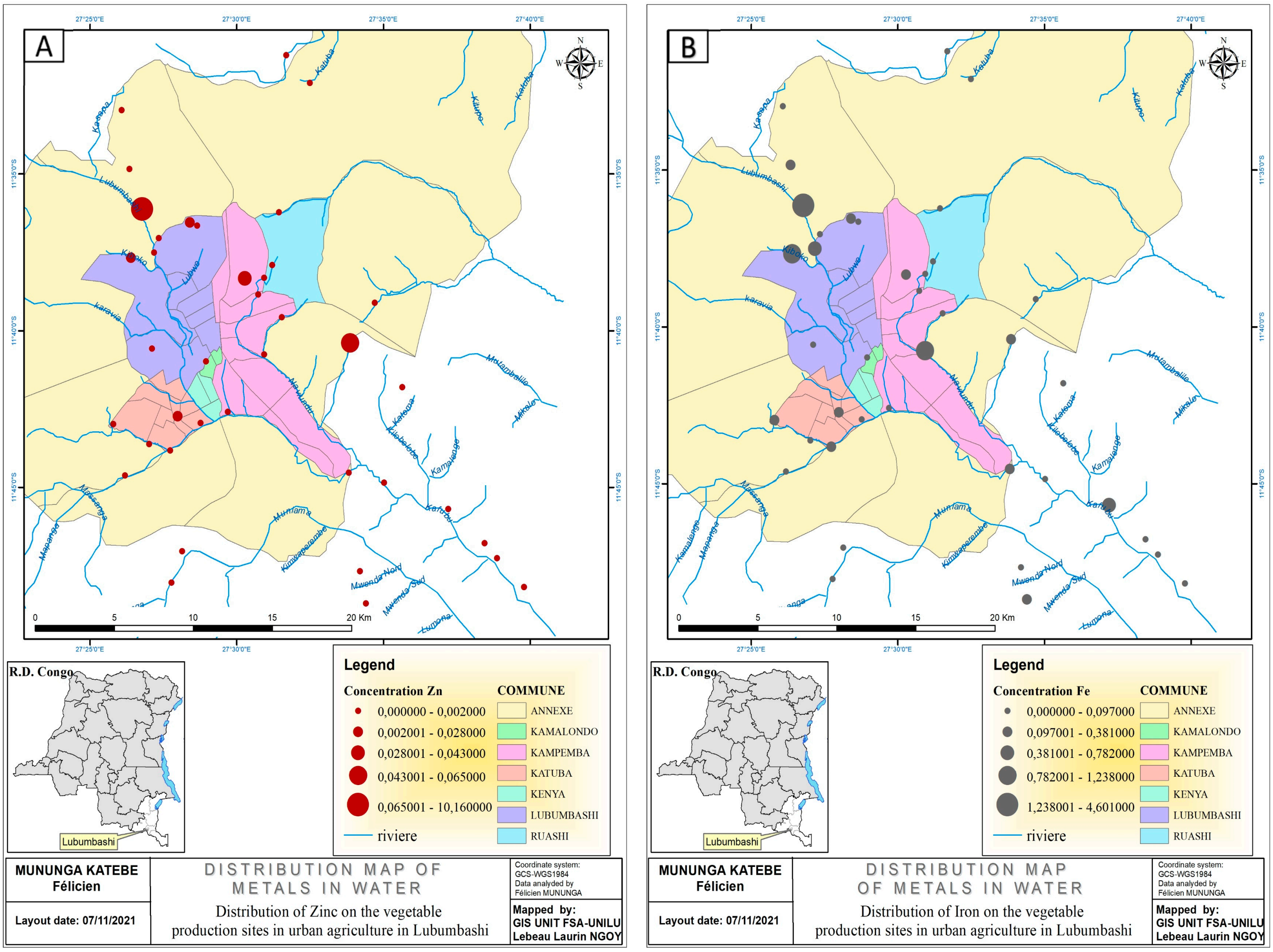

Figure 12. Spatial distribution of metals (Zn and Fe) in the waters of Lubumbashi market gardens (**A**,**B**).

4. Discussion

4.1. Assessment of Pollution Indices, Contamination Factors and Soil Enrichment Factors in Market Gardens

We determined the extent of contamination in the various agricultural and non-agricultural soils of Lubumbashi [11,46–48]. Our study is the first in the area to have data that can allow us to specifically identify each soil and its level of heavy metal pollution because none of these regional studies has been able to assess the level of pollution, contamination and enrichment of each of the soils in this regard. Our results reveal that the soil pollution index shows nearly 80% of the gardens with no pollution risk, and 21% of the market gardens are polluted. This phenomenon can be explained by the fact that market gardens close to industrial activities show severe pollution compared to the soils of gardens far from these activities [49,50]. Most anthropogenic activities practiced in the area are hydrometallurgy and pyrometallurgy to process ore; these discharge heavy metal-laden effluent into the rivers that serve as irrigation water reservoirs for urban market gardeners and pollute the agricultural soils [19,51,52]. These hydrometallurgical or pyrometallurgical treatment processes have led to heavy metal contamination of stream sediments near a gold mine in Ghana. Similar situations have been observed in Bangladesh, where [53,54] have shown that the richness of the subsoil is responsible for metal contamination of the water table on the one hand and mining activities on the other. The market gardens of Lubumbashi are close to watercourses that receive metal-rich effluents, which would cause soil pollution by watering the crops with these waters. This would be explained by the fact that organic amendments are added to the soils of the latter from former residential and mining dumps used in urban agriculture in Lubumbashi [20,55]. Based on the soil pollution index, our results corroborate those found by [56,57] who showed that soils close to a pollution source had a higher pollution index than soils far from the pollution source. In all cases, the concentrations found in the water of urban market gardens came from the exploitation of Zn-enriched deposits. Similar situations were observed in Kolwezi (DR Congo), where the Dilala and Luilu rivers had high levels of heavy metals. These high concentrations of metals in the water of these rivers were due to the former activities of abandoned mining companies that were active in the city of Kolwezi [16,58,59].

4.2. Chemical Quality of the Irrigation Water of the Market Gardens

The use of groundwater and recycled wastewater is the basis for the presence of metals in soils, water and plants [17,60,61]. These corroborate our findings that 57% of the water in the market gardens of Lubumbashi is contaminated with lead, 52% with iron, 19% with copper, 10% with cadmium and 5% with aluminum. River water is often overloaded with metals released into the environment by mining companies [62–64]. Furthermore, the market gardens along the Kafubu River have a high degree of Cu, Pb and Fe pollution in contrast to the other axes along which the other market gardens are located. This is thought to be due to the fact that the land on which these rivers intersect is cupro-cobalt bearing rock [65,66]. In addition, mineral processing effluents discharged by mining companies into the rivers are the main causes of water contamination/pollution. These very high metal concentrations create health and environmental problems [23,58,67] for crop watering. High lead concentrations would mainly come from mining companies' releases to the atmosphere and water, as well as from plants [7,10,12]. Similar situations were observed by [68] in the Nile Delta in Egypt, where high concentrations of Cr, Co, Cu, Pb, Ni and Zn found in water resulted in the production of clover plants contaminated with these heavy metals, as these vegetables were watered with river water and landfill water loaded with metals had rendered plants grown on these soils unusable.

4.3. Safety of the Vegetables Sold in the 33 Selected Lubumbashi Markets

The safety of vegetables is a function of the quality of the agricultural soils and the irrigation water with which they were produced [69]. Thus, from our results, it appears that almost all of these four vegetables sampled in the markets of Lubumbashi are contaminated with copper and cobalt, and nearly 47% are contaminated with cadmium. This situation is due to the fact that the soils on which these vegetables grow are mostly contaminated by metals from mining companies, on the one hand, and from the natural richness of the subsoil, on the other hand. Our results corroborate those found in Kolwezi by [26,69,70] who found that effluent from businesses discharged into rivers was responsible for contamination of vegetables produced and consumed in the city. Since urban agriculture in Lubumbashi is usually practiced in the dry season and on the banks of rivers, the water used to water the crops is loaded with metal pollutants, which is a very serious environmental problem. Observations made by [10,16] report that effluents discharged into the rivers of Lubumbashi and the Tshamilemba canal by mining companies dating back more than a decade were the main source of contamination of the water in these rivers, as the levels of contamination found were 200 times higher than the recommended toxicity threshold for soils, and this leads to contamination of the plants growing in them. In addition, this contamination of vegetables is thought to come from the former urban dumps of Lubumbashi that are used as market gardens in the dry season and are a reservoir of metal pollutants due to the various wastes they collect [71]. These results corroborate those found in Kolwezi by [71] who report that landfills are among the most dangerous main locations for pollution, as levels found in the soil of a former landfill were above WHO standard values. Metal concentrations found in biomass were three times the standard for vegetable consumption [11,25,72]. Our results confirm those of [73,74] that showed that the pollutant quality of soils was the basis for contamination of the six vegetables grown on these soils in the Pakistan region. Thus, of all the vegetables studied in this work, it appears that Chem-Chem amaranths are the most contaminated with heavy metals, followed by Brassica carinata. This would be justified by the simple fact that the soils of the market gardens and the water used to irrigate the crops at the Chem-Chem site are the most polluted in the city of Lubumbashi.

5. Conclusions

Our study indicated persisting heavy metal (Cu, Cd, Pb, Co and Zn) contamination of the soils, waters and market garden vegetables of Lubumbashi. Chemical analysis of heavy metals in the soils and waters revealed that the main contamination/pollution sources are most probably the naturally metal-rich soils and the metal-loaded effluents discharged into watercourses by mining companies. The quality of the soil and water of many market gardens of Lubumbashi remains poor for producing uncontaminated vegetables, and our results show that the vegetables sold on Lubumbashi markets are unfit for human consumption. In view of the currently proposed phytoremediation techniques that do not fully solve soil and vegetable pollution issues, other more advanced remediation techniques such as the composting of organic matter mixed with appropriate amounts of limestone to limit soil-to-plant heavy metal transfer should be envisaged.

Author Contributions: F.M.K.: Conceptualisation, methodology, data analysis, writing; P.R.: contribution to writing, data analysis, critical revision of the manuscript; G.C.: contribution to writing, critical revision of the manuscript, project manager; M.N.S. and M.M.M.: methodology, critical revision of the manuscript; M.H.J.: supervision, contribution to writing and to critical revision of the manuscript. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

Appendix A

Appendix A.1. Names of the 40 Identified Urban Market Gardens of Lubumbashi

Table A1. Urban and peri-urban gardens identified in Lubumbashi.

Urban and Peri-Urban Market Gardens
Bombeki
Bongonga
Camps-assistant
Camps-scout
Campus-Unilu
Chem-chem
Daipen-Kisanga
Ferme konde
Ferme nkonde
Inera-Salongo
Kabetsha
Kafubu
Kakonkania
Kalebuka
Kalubwe
Kalulako

Table A1. *Cont.*

Urban and Peri-Urban Market Gardens
Kamakanga
Kamatete
Kamilombe
Kamisepe
Kantumbwi
Kasamba
Kashamata
Kashimbala
Kasungami
Katemo
Kawama
Kilobelobe
Kinsense
Kitanda
Luano
Maendeleo
Kinsevere/Manoah
Mwenda
Naviundu
Penga-penga
Sambwa
Tingi-Tingi
Tshamalale
Tshamilemba

Appendix A.2. Names of the Markets of the City of Lubumbashi (Democratic Republic of the Congo)

Table A2. Urban markets of Lubumbashi.

Market	Municipality
ANTENNE RUASHI	RUASHI
CAMP PREFABRIQUE	KAMPEMBA
DOUBLE POTEAU	LUBUMBASHI
EUREKA	LUBUMBASHI
KABULAMENSHI	LUBUMBASHI
KALEBUKA	ANNEXE
KAMATETE	ANNEXE
KANSOKO	ANNEXE
KARAVIA	ANNEXE
KASANGULU	LUBUMBASHI
KATUBA 1	KATUBA
KATUBA 2	KATUBA
KENYA (ZONE)	KENYA
KIGOMA	KAMPEMBA
KILOBELOBE	ANNEXE
KIMUTI	KAMPEMBA
LIDO	LUBUMBASHI
MANOAH NSOKO	RUASHI
MARCHE CINE	ANNEXE
MÉTÉO	ANNEXE
MIMBULU	KATUBA
MOISE	ANNEXE
MWIMBILA	KENYA
M'ZEE	LUBUMBASHI
NJANJA	KENYA
PANDE	KAMPEMBA
PEAGE KIMUTI	KATUBA
RADEM	KAMPEMBA
RAIL	KAMPEMBA
ROSE TSHAKWIZA	ANNEXE
TABAC	KAMPEMBA
TSHAMALALE	ANNEXE
ZAMBIA	RUASHI

Appendix A.3. Contamination Levels of Urban and Peri-Urban Garden Soils according to the Contamination Factor

Market Gardens	Fe Contamination Level	Cu Contamination Level	Pb Contamination Level	Zn Contamination Level
Bongonga	Low contamination	Low contamination	Low contamination	Low contamination
Chem-chem	Low contamination	Moderate contamination	Moderate contamination	Very strong contamination
Daipen/Kashamata	Low contamination	Low contamination	Low contamination	Low contamination
Kabetsha	Low contamination	Low contamination	Low contamination	Low contamination
Kafubu	Low contamination	Low contamination	Low contamination	Moderate contamination
Kalebuka	Low contamination	Low contamination	Low contamination	Low contamination
Kalubwe	Low contamination	Moderate contamination	Low contamination	Moderate contamination
Kalulako	Low contamination	Low contamination	Low contamination	Low contamination
Kamakanga	Low contamination	Low contamination	Low contamination	Low contamination
Kamatete	Low contamination	Low contamination	Low contamination	Low contamination
Kamilombe	Low contamination	Moderate contamination	Low contamination	Significant contamination
Kamisepe	Low contamination	Low contamination	Low contamination	Low contamination
Kantumbwi	Low contamination	Low contamination	Low contamination	Low contamination
Kasungami	Low contamination	Low contamination	Low contamination	Low contamination
Katemo	Low contamination	Low contamination	Low contamination	Low contamination
Kawama	Low contamination	Moderate contamination	Low contamination	Moderate contamination
Kilobelobe	Low contamination	Low contamination	Low contamination	Moderate contamination
Kinsense	Low contamination	Low contamination	Low contamination	Low contamination
Kinsevere-Manoah	Significant contamination	Moderate contamination	Low contamination	Moderate contamination
Kitanda	Low contamination	Moderate contamination	Low contamination	Significant contamination
Luano	Low contamination	Low contamination	Low contamination	Moderate contamination
Maendeleo	Low contamination	Moderate contamination	Low contamination	Moderate contamination
Mwenda	Low contamination	Low contamination	Moderate contamination	Low contamination
Penga-penga	Low contamination	Low contamination	Low contamination	Low contamination
Sambwa	Low contamination	Low contamination	Low contamination	Low contamination
Tingi-Tingi	Low contamination	Low contamination	Low contamination	Low contamination
Tshamalale	Low contamination	Low contamination	Low contamination	Low contamination
Tshamilemba	Low contamination	Low contamination	Low contamination	Low contamination

Appendix A.4. Metal Enrichment Levels of the Urban and Peri-Urban Market Gardens of Lubumbashi

Market Gardens	Cu	Pb	Zn
Bongonga	No enrichment	No enrichment	No enrichment
Chem-chem	Enrichment Medium High	Moderate Enrichment	High Enrichment
Daipen/Kashamata	Low Enrichment	No enrichment	Low Enrichment
Kabetsha	No enrichment	No enrichment	No enrichment
Kafubu	Low Enrichment	No enrichment	Moderate Enrichment
Kalebuka	Moderate Enrichment	No enrichment	Low Enrichment
Kalubwe	Low Enrichment	No enrichment	Low Enrichment
Kalulako	Low Enrichment	No enrichment	Low Enrichment
Kamakanga	No enrichment	No enrichment	No enrichment
Kamatete	No enrichment	No enrichment	Low Enrichment
Kamilombe	Low Enrichment	No enrichment	Enrichment Medium high
Kamisepe	Low Enrichment	No enrichment	Low Enrichment
Kantumbwi	No enrichment	No enrichment	No enrichment
Kasungami	No enrichment	No enrichment	No enrichment
Katemo	No enrichment	No enrichment	No enrichment
Kawama	Low Enrichment	No enrichment	Low Enrichment
Kilobelobe	Low Enrichment	Low Enrichment	Moderate Enrichment
Kinsense	Low Enrichment	No enrichment	Low Enrichment
Kinsevere-Manoah	No enrichment	No enrichment	No enrichment
Kitanda	Low Enrichment	No enrichment	Enrichment Medium high
Luano	Low Enrichment	Low Enrichment	Moderate Enrichment
Maendeleo	Low Enrichment	Medium High Enrichment	Enrichment Medium high
Mwenda	No enrichment	No enrichment	Low Enrichment
Penga-penga	No enrichment	No enrichment	No enrichment
Sambwa	Low Enrichment	Low Enrichment	Low Enrichment
Tingi-Tingi	Low Enrichment	No enrichment	Low Enrichment
Tshamalale	No enrichment	No enrichment	No enrichment
Tshamilemba	No enrichment	No enrichment	No enrichment

Appendix A.5. Pollution Levels of the Urban and Peri-Urban Market Gardens of Lubumbashi

Market Garden Name	Pollution Level
Bongonga	Not polluted
Chem-chem	Severe pollution
Daipen/Kashamata	Not polluted
Kabetsha	Not polluted
Kafubu	Not polluted
Kalebuka	Not polluted
Kalubwe	Severe pollution
Kalulako	Not polluted
Kamakanga	Not polluted
Kamatete	Not polluted
Kamilombe	Severe pollution
Kamisepe	Not polluted
Kantumbwi	Not polluted
Kasungami	Not polluted
Katemo	Not polluted
Kawama	Not polluted
Kilobelobe	Not polluted
Kinsense	Not polluted
Kinsevere-Manoah	Severe pollution
Kitanda	Severe pollution
Luano	Not polluted
Maendeleo	Severe pollution
Mwenda	Not polluted
Penga-penga	Not polluted
Sambwa	Not polluted
Tingi-Tingi	Not polluted
Tshamalale	Not polluted
Tshamilemba	Not polluted

Appendix A.6. XRF Determination of Metals in 29 Urban and Peri-Urban Market Gardens of Lubumbashi (mg/kg). Limits of Quantification: 0.05 mg/kg

Market Gardens	Fe	Cd	Cu	Pb	Cr	Zn
Bongonga	3.31	0.05	59	0.05	0.05	30
Chem-Chem	4.08	45	1.355	221	67	1.47
Daipen/Kashamata	2.13	0.05	204	20	45	60
Kabetsha	2.19	0.05	113	0.05	45	42
Kafubu	2.34	236	146	0.05	0.05	181
Kalebuka	1.12	0.05	226	0.05	38	53
Kalubwe	4.97	40	850	0.05	58	301
Kalulako	1.84	0.05	239	0.05	0.05	85
Kamakanga	3.33	0.05	78	0.05	66	44
Kamatete	3.55	0.05	206	26	0.05	138
Kamilombe	4.23	56	656	0.05	69	716
Kamisepe	1.87	0.05	208	0.05	63	73
Kantumbwi	1.81	0.05	45	0.05	46	28
Kasungami	1.9	0.05	102	0.05	47	38
Katemo	3.23	19	56	0.05	0.05	35
Kawama	4.18	0.05	598	39	0.05	262
Kikula/Sambwa	1.57	0.05	201	18	45	88
Kilobelobe	3.71	0.05	344	48	75	284
Kinsense	3.7	0.05	270	34	51	136
Kinsevere (Manoah)	33.43	0.05	535	81	0.05	394
Kitanda	4.96	0.05	826	47	59	652
Luano	4.05	0.05	346	80	46	300
Maendeleo	2.7	0.05	493	184	57	409
Mashimikila	2.46	0.05	1.043	73	0.05	230
Mwenda	1.28	0.05	66	0.05	43	42
Pengapenga	4.31	0.05	183	0.05	49	50
Tingi-Tingi	2.57	0.05	160	0.05	45	66
Tshamalale	2.73	0.05	50	0.05	0.05	34
Tshamilemba	2.46	0.05	51	0.05	0.05	33
Toxicity threshold (mg/kg)	NT	2	100	100	150	300

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