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Baseline concentrations of 11 elements as a function of land uses in surface soils of the Katangese Copperbelt area (D.R. Congo)

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Abstract: The Katangese Copperbelt area (KCA) located south-eastern of D.R. Congo presents high concentration of metal trace elements (MTE) in several soils due to a rich natural geochemical background, and intense mining activities, causing serious health issues to humans and animals. However, the lack of data on specific baseline concentrations makes it difficult to properly assess and monitor the environmental quality of soils in the region. In this study, the baseline concentration of 11 potentially toxic MTE (i.e., Mn, Zn, Cu, Co, Cr, Pb, Cd, Ti, Ni, Al, and Fe) was assessed in topsoils of the KCA, and the possible influence of land uses (croplands, forest and mining areas) was examined. Results showed the following baseline concentrations, i.e. lower and upper limits (mg.kg^{-1}) in cropland soils : Al (18.4–162.0), Cd (0.0–0.1), Co (0.1–3.5), Cu (0.8–17.7), Cr (0.0–0.2), Fe (4.7–233.8), Mn (3.5–575.6), Ni (0.1–1.9), Pb (0.2–2.4), Zn (0.1–20.3), Ti (0.0–392.6); in forests: Al (18.8–1167.1), Cd (0.02–0.48), Co (0.20–18.1), Cu (3.6–42.7), Cr (0.1–33.7), Fe (86.4–283.3), Mn (4.9–1538.9), Ni (0.05–24.2), Pb (0.3–13.7), Zn (2.0–7.0), Ti (0.2–0.8); and in mining areas: Al (7.4–241.2), Cd (0.01–164.8), Co (0.2–211.3), Cu (2.4–5485.4), Cr (0.03–0.4), Fe (5.9–481.6), Mn (7.1–95.9), Ni (0.1–1.9), Pb (0.2–390.8), Zn (1.5–5629.3), Ti (0.1–1.3). Cu and Zn were highest in mining areas demonstrating a prevalent influence of mining activities in altering the natural background of metals concentrations in the region. By contrast, croplands and forest shared a similar trend of Al and Mn contents, suggesting a mild influence of agricultural activity. Intriguingly, higher Cu and Co contents were found in forest compared to croplands. For all the three studied land uses, no straightforward relation was found between metal concentrations and soil total acidity. This study is the first attempt to establish reference values of MTE contents in the KCA soils and thus provides valuable information for legislative purposes and for soil quality assessment.

Keywords: Contamination, trace elements, metal concentration, environmental pollution

1. Introduction

In recent years, ecosystems pollution with metals has become of great concern due to the inherent adverse effects on living organisms including humans [1]. In the prospect to preventing humans and animals' exposure, assessing the environmental quality of soils represents a crucial step, for which understanding the patterns of trace elements concentrations in soils is of critical importance [2].

The behaviour of metals into soils is constrained by various geochemical processes and pH is one of the most important factors determining the forms of metal occurrence and influencing metal solubility and speciation and thus toxicity [3]. The first approach for soil

contamination evaluation is the determination of the total available trace element content [4]. In this regard, the natural background concentrations of trace elements are suggested as a reference to identify polluted soils, and eventually to propose a plan for their proper management [1,4].

Referred to as the natural range of concentration prior to any anthropogenic influence, the concept of geochemical background concentrations is scarcely considered however, and related data are quasi-inexistant for most environments. Conventionally, 95% of the expected range of background concentrations is designated as baseline concentrations, which is most often considered as reference to evaluate soil contamination levels [1].

Several sources affecting the natural background concentrations of metals in soils are listed including mining, agricultural and other industrial activities [5,6]. In the The Katangese Copperbelt area (KCA), located south-eastern of the Democratic Republic of Congo, significant deposits of copper (Cu), cobalt (Co) and other metals are found [7,8]. Hence, most environmental studies have revealed widespread soil contamination in the region [9, 10-12]. In most cases, these studies expressed the metals contents in soil referring to the so-called universal concentrations of trace elements. A reference study by [13] provided an interesting overview of metal contents in the region, but did not present the related baseline concentration. Moreover, none of these studies established the lower and upper limits of elements contents based on the baseline calculation that can be used as soil quality reference values for the KCA. This makes it difficult to adequately establish quality standards to detect and monitor sites affected by metals contamination in the region. Yet the concentrations of trace elements in soils depends on the mineralogical composition of the parent material and is influenced by weathering processes; therefore, it appears inappropriate to use universal reference concentrations [4,14]. Alternatively, a more appropriate approach would imply establishing baseline concentrations on a regional basis to be used as a reference in the identification of contaminated soils [1]. Unfortunately, no data on baseline concentrations of trace elements exist for the Katangese Copperbelt area (KCA).

The present study was initiated to (i) establish baseline concentrations of 11 potentially toxic trace elements (i.e., Mn, Zn, Cu, Co, Cr, Pb, Cd, Ti, Ni, Al, Fe) in the Katangese Copperbelt area (KCA) surface soils; and (ii) investigate their variation under 3 different land uses (croplands, forest and mining areas). The results of this research can be used as soil quality reference values in assessing anthropogenic vs. natural levels of trace elements in the KCA.

2. Materials and Methods

2.1. Study area

This study was conducted in the KCA extends from Sakania to Kolwezi (Haut- Katanga and Lualaba provinces, D.R. Congo) (Figure 1). Climate of the KCA is humid subtropical (Köppen climate classification Cwa) with one dry season (May to September) and one wet season (November to April). Annual mean rainfall is about 1300 mm, the majority falling during the wet season. Temperature fluctuates from 15–17°C (early dry season) to 31–33°C (late dry season). Mean annual temperature is about 20 °C [15,16]. The bedrock of the KCA consists of ~10,000 m thick sedimentary succession that belongs to Neoproterozoic Katangan Supergroup [17,18]. Based on the regional occurrence of two diamictites (the 'Grand Conglomérat' and 'Petit Conglomérat'), the Katangan sedimentary succession is subdivided in three lithostratigraphic groups i.e., from base to top, the Roan, Nguba and Kundelungu Groups [19,20] The Roan Group comprises siliciclastic and dolomitic rocks, whereas the Nguba and Kundelungu Groups largely consist of siliciclastic rocks,

with one major carbonate unit occurring as part of the Nguba Group (Kakontwe Limestone) [20,21]. The primary vegetation of the region is miombo woodland [22]. Strong anthropogenic degradation of the vegetation resulted in woodland being substituted by a secondary grass savannah in the peri-urban zones of the KCA [23].

2.2. Sample collection and laboratory analyses

Surface soils (0-20 cm) used in this study were collected in Lubumbashi and its surroundings between 2015 and 2021. The sampling area covers 3 land uses with a total of 490 samples (Figure 1):   agricultural land (450 samples), mining sites (24 samples), and miombo woodland (16 samples).

Analyses were performed on air-dried soil fractions (< 2 mm). The soil pH was measured potentiometrically in a 1:2.5 (W/V) suspension of 1 M KCl. Total trace element content was measured using the Inductively coupled plasma - optical emission spectrometry (ICP-OES: Optima 8300 078S1509233. The detection limits of the device used in this study is between 0.5 -10 ppm). Briefly, the traces elements analyzed in the soil samples were: Mn, Zn, Cu, Co, Cr, Pb, Cd, Ti, Ni, Al, and Fe. Digestion of the sample was performed by wet extraction with ETDA at pH 4.65 following the protocol described in the soil analysis manual [24]. All analyses were performed in triplicate. When processing the samples, we started with the approach to make analyses on all trace elements, but only 11 trace elements were detected having regard to the limit of detection of the device on the one hand, and on the other hand, they showed themselves potentially toxic according to their observed content.
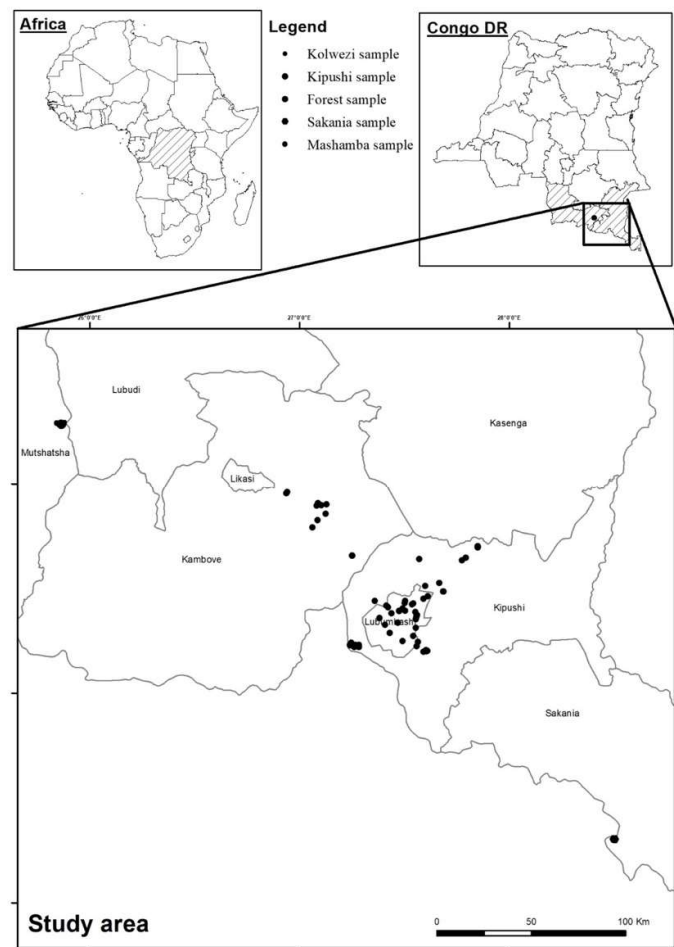
Figure 1. Sampling areas in the KCA with 3 land uses (cropland, forest and mining area).

2.4. Data processing and statistics

Principal component analysis (PCA) was performed using the package FactoMiner in R software (4.0.1) to characterize studied land uses based on measured elements [25]. The arithmetic mean (AM), arithmetic standard deviation (ASD), geometric mean (GM), geometric standard deviation (GSD), minimum, maximum, and median were calculated to compare trace element contents and pH across land uses. The AM and ASD are best estimates of geochemical abundance of an element, whereas, the GM and GSD are better maximum likelihood estimators for most geochemical data. Baseline concentrations of the 11 trace elements were calculated using GM/GSD^2 and $GM \times GSD^2$ of the samples, respectively as the lower and the upper limit of the established baseline [26].

To understand relationships between pH and studied trace elements, we transformed our data using best normalize package in R to meet the normal distribution requirement of data before we calculated Pearson correlation coefficient.

3. Results

3.1. Element contents and pH across landuses

The principal component analysis (ACP) explained over 55% of variability within the dataset (Figure 2). The largest variability in metal contents was observed in croplands. The PCA results in two groups of metals positively correlated between each other. The first group consisted of Cd, Ti, Fe, and Zn while the second group consisted of Ni, Co, Cu, Mn, and Cr. The pH was genitively correlated with Pb along the dimension 1. Aluminium was strongly associated with samples from forests and mining areas.
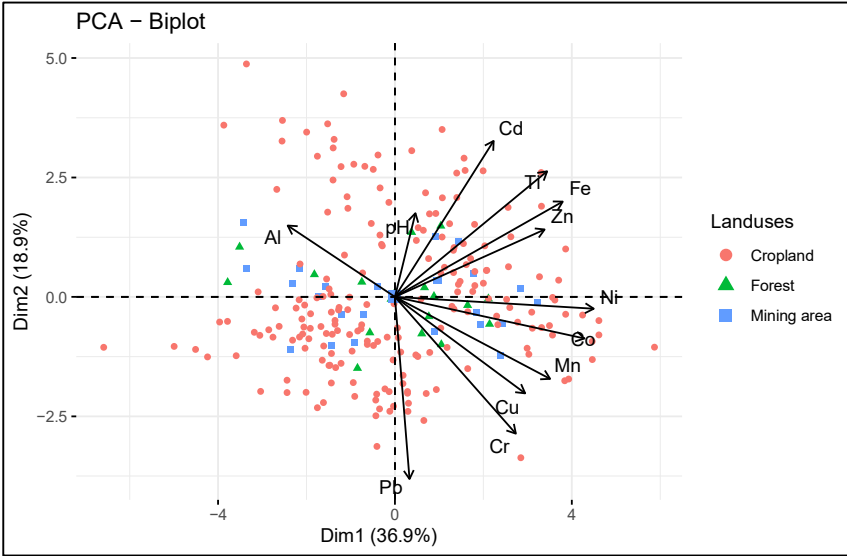

Figure 2. Principal component analysis showing variability of studied elements across 3 land uses (cropland, forest and mining area).

The lowest pH in cropland was 3.9 while the highest was 7.3 (Table 1). The mean pH in investigated cropland was 5.3 and 5.4 for arithmetic and geometric mean respectively. The minimum Al and Fe concentrations were 16.6 and 10.7 mg. kg^{-1} respectively, whereas, their respective maximum concentrations were 282.9 and 215.7 mg.kg^{-1}. Al, Fe, and Mn were the most abundant elements in studied cropland soils, with respectively 63.4, 42.2, and 57.6 mg. kg^{-1} on average.

The mean Cu and Co concentrations were 5.4 and 1.0 mg.kg^{-1}, respectively. The following elements were found in trace concentrations (maximum concentration < 5 mg.kg^{-1}), Cd, Cr, Pb, Zn and Ni.

Table 1. Descriptive statistics of element contents (mg.kg^{-1}) and pH in croplands (n=450).

	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Ti	pH
Minimum	16.6	0.0	0.1	0.0	1.1	10.7	2.8	0.1	0.0	0.0	0.1	3.9
Maximum	282.9	0.4	16.0	0.4	160.7	215.7	323.0	2.6	2.2	3.9	100.5	7.3
Median	51.5	0.1	0.5	0.1	3.4	30.4	46.7	0.2	0.7	1.4	0.9	5.2
AM	63.4	0.1	1.0	0.1	5.4	42.2	57.6	0.3	0.7	1.3	16.4	5.4
ASD	37.4	0.0	1.8	0.0	12.0	35.9	42.9	0.3	0.4	0.7	20.0	0.8
GM	54.6	0.0	0.5	0.1	3.7	33.0	44.8	0.2	0.6	1.0	2.4	5.3
GSD	1.7	1.4	2.5	1.6	2.2	2.7	3.6	2.0	2.0	4.5	12.7	1.2

GSD²	3.0	1.9	6.5	2.7	4.7	7.1	12.9	3.9	4.1	20.0	162.4	1.4
GM/GSD²	18.4	0.0	0.1	0.0	0.8	4.7	3.5	0.1	0.1	0.1	0.0	3.8
GM*GSD²	162.0	0.1	3.5	0.2	17.7	233.8	575.6	0.9	2.4	20.3	392.6	7.5

In forests, pH varied between 4.0 and 5.2 with an average of 4.3 (Table 2). The most abundant elements were Al, Fe, and Mn, with respectively 222.2, 162.6, and 163.0 mg.kg^{-1} on average. Mean Cu concentration was higher (14.6 mg.kg^{-1}) than that of Co (2.7 mg.kg^{-1}). Cu and Co were found up to maximum of 31.4 mg. kg^{-1} and 4.5 mg. kg^{-1} respectively. The maximum concentrations in Cd, Cr, Ni, Pb, Zn, and Ti were lower than 10 mg.kg^{-1}. In contrast croplands, Cr in was found up to a maximum of 44.9 mg.kg^{-1} with an average of 6.1 mg.kg^{-1}.

Table 2. Descriptive statistics of element contents (mg.kg^{-1}) and pH in forests (n=16).

	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Ti	pH
Minimum	38.9	0.0	0.1	0.4	3.2	74.3	4.3	0.2	0.3	2.0	0.2	4.0
Maximum	401.9	0.4	4.5	44.9	31.4	253.6	370.0	7.6	7.5	6.8	0.7	5.2
Median	302.5	0.1	2.7	2.1	10.5	158.6	115.5	0.5	1.3	3.8	0.4	4.2
AM	222.2	0.1	2.7	6.1	14.6	162.6	163.0	2.8	2.8	3.9	0.4	4.3
ASD	160.9	0.1	1.6	11.3	8.4	44.8	134.1	3.0	2.4	1.2	0.1	0.4
GM	148.0	0.1	1.9	2.2	12.3	156.4	86.9	1.1	1.9	3.7	0.4	4.3
GSD	2.8	2.3	3.1	3.9	1.9	1.3	4.2	4.7	2.7	1.4	1.4	1.1
GSD²	7.9	5.1	9.6	15.3	3.5	1.8	17.7	21.7	7.3	1.9	2.1	1.2
GM/GSD²	18.8	0.0	0.2	0.1	3.6	86.4	4.9	0.1	0.3	2.0	0.2	3.6
GM*GSD²	1167.1	0.5	18.1	33.7	42.7	283.3	1538.9	24.2	13.7	7.0	0.8	5.1

In the mining area, pH varied between 4.2 and 6.8 with an average of 5.8. Cu was the most abundant metal, with a maximum of 1828.0 mg.kg^{-1} and an average of 424 mg.kg^{-1} (Table 3). Following Cu, Zn was found up to a maximum of 1410.0 mg. kg^{-1} and an average of 408.2 mg.kg^{-1}. Mean Al, Fe, and Mn concentrations were respectively 60.5, 105.3, 32.4 mg. kg^{-1}. Cr, Ni, and Ti were found in lowest concentrations, with a maximum of 1.0, 1.7, 1.2 mg.kg^{-1} respectively.

Table 3. Descriptive statistics of element contents (mg.kg^{-1}) and pH in the mining areas (n=24).

	Al	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Ti	pH
Minimum	7.3	0.0	0.6	0.0	8.6	15.8	10.3	0.2	0.8	6.1	0.1	4.2
Maximum	222.8	52.2	527.2	1.0	1828.0	700.9	92.5	1.7	182.2	1410.0	1.2	6.8
Median	39.8	1.0	3.8	0.1	154.1	38.5	24.2	0.3	3.8	56.6	0.3	6.1
AM	60.5	9.0	41.2	0.1	424.0	105.3	32.4	0.6	37.7	408.2	0.4	5.8
ASD	52.8	14.3	111.9	0.2	549.4	158.2	23.3	0.5	55.7	519.4	0.3	0.7
GM	42.8	1.3	7.0	0.1	115.5	53.3	26.1	0.5	8.2	93.1	0.3	5.8
GSD	2.4	11.4	5.5	1.9	6.9	3.0	1.9	2.0	6.9	7.8	1.9	1.1
GSD²	5.8	130.6	30.2	3.6	47.5	9.0	3.7	4.1	47.8	60.5	3.7	1.3
GM/GSD²	7.4	0.0	0.2	0.0	2.4	5.9	7.1	0.1	0.2	1.5	0.1	4.5
GM*GSD²	247.2	164.8	211.3	0.4	5485.5	481.7	95.9	1.9	390.8	5629.3	1.3	7.4

3.2. Baseline concentrations of elements and pH across different land uses

The minimum and maximum baseline pH in croplands were 3.8–8.0 (Figure 3). The minimum expectable Al content (baseline) in cropland was 18.4 mg.kg^{-1} while the maximum was 162 mg. kg^{-1}. Minimum and maximum baseline Fe concentrations was 4.7–234.0 mg.kg^{-1}. Baseline Mn concentrations varied between 3.5–576.0 mg.kg^{-1}. The minimum baseline Cu and Co concentrations were 0.1 and 0.8 mg.kg^{-1} while the maximum was 4.0 and 18.0 mg.kg^{-1}.
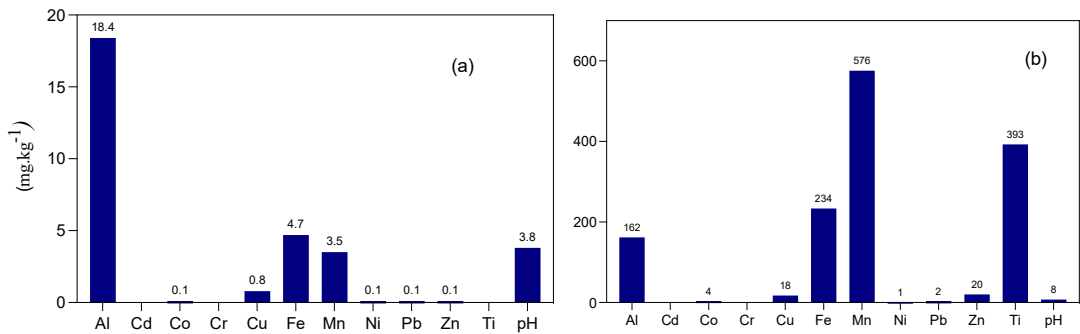
Figure 3. Minimum (a) and maximum (b) baseline concentrations of elements and pH in croplands.

In the forests, minimum-maximum baseline pH was 3.6–5.0 (Figure 4). Minimum baseline Al and Fe concentrations were respectively 18.8 and 86.4 mg.kg^{-1} while the maximum baseline concentrations were 1167 and 283 mg.kg^{-1}. Minimum and maximum baseline Mn concentrations varied between 4.9 and 1539 mg.kg^{-1}. Obtained baseline Cu concentrations was 3.6–43.0 mg.kg^{-1} while that of Co was 0.2–18.0 mg.kg^{-1}.
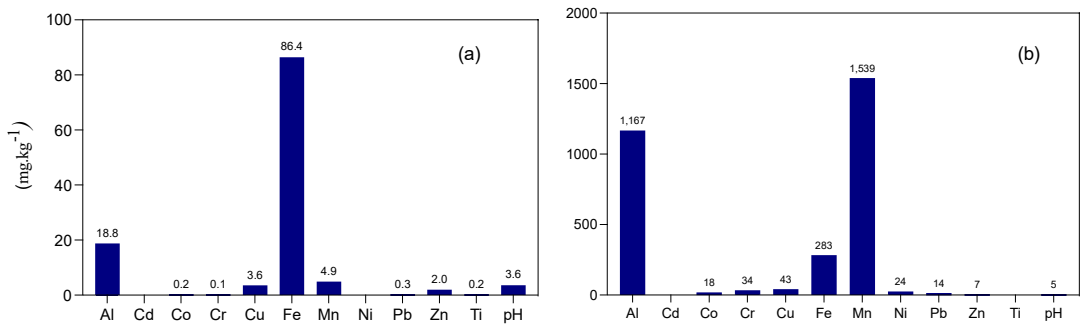

Figure 4. Minimum (a) and maximum (b) baseline concentrations of elements and pH in forests.

Baseline pH observed in the mining areas was 4.5–7.0 (Figure 5). Minimum baseline Al and Fe concentrations were 7.4 and 5.9 mg.kg-1 while the maximum baseline concentrations were 247.0 and 482.0 mg.kg^{-1} respectively. Baseline Mn and Zn concentrations were 7.1–86.0 mg.kg^{-1} and 1.5–5.629 mg.kg^{-1} respectively. Baseline Cu concentration was 2.4–5485 mg.kg-1 while that of Co was 0.2–211 mg.kg^{-1}.
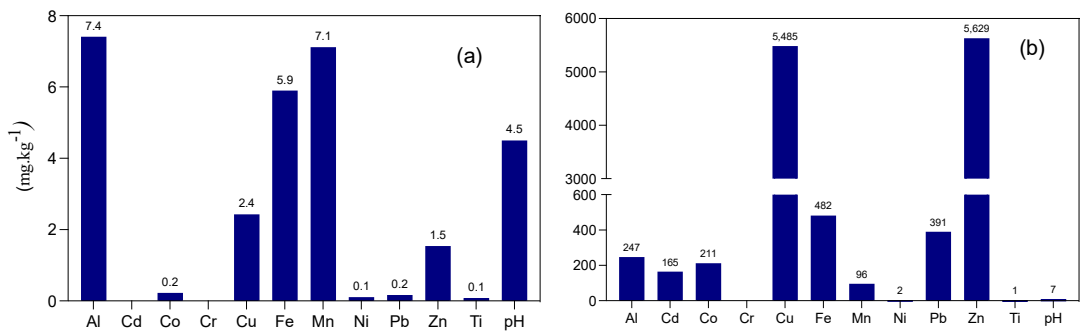
Figure 5. Minimum (a) and maximum (b) baseline concentrations of elements and pH in mining areas.

3.3. Correlations between pH and studied elements

In croplands, pH was negatively correlated with Co, Fe, and Zn but positively correlated with Cu, Al, and Zn. However, none of these correlations was statistically significant (Figure 6 a, d, g). In mining areas, pH was negatively correlated with Al while positively significantly correlated with Fe and Zn. A positive but non-statistically significant correlations of pH with Cu and Co were observed. In the forests, pH was negatively and significantly correlated with Mn and Co while positively and significantly correlated with Al.
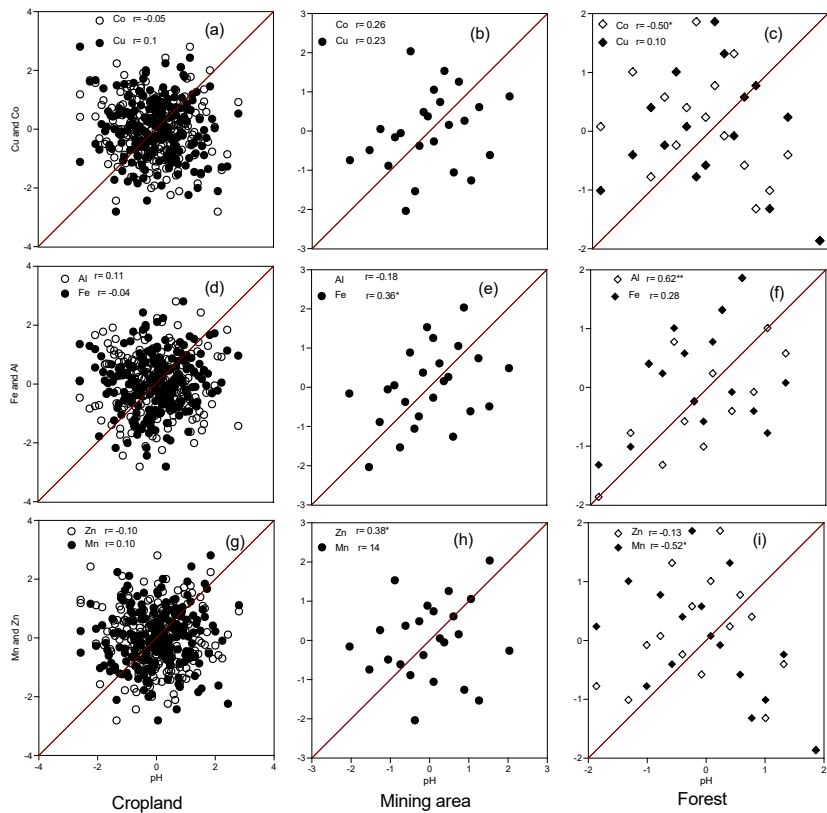

Figure 6. Correlations of pH with studied elements across land uses.

4. Discussion

Soils across investigated land uses were acidic with a mean pH lower than 6, which is in accordance with previous studies in the regions [27]. However, the baseline pH in studied land uses varied from acidic to alkaline soils, with 3.8–8.0 for croplands, 3.6–5.0 for forests, and 4.5–7.0 for mining areas (Figure 4–5). These results suggest that although studied soils are globally characterized as acidic, the pH at some locations can be neutral or alkaline as observed for mining areas and croplands respectively. Surprisingly, soils in the mining areas and croplands showed higher pH compared to forests; which would come from the difference between the soil units. The possibility that the use of lime in mining areas during waste treatment and cropland could be raised to explain the elevated pH. Additionally, we cannot rule out the possibility that organic matter decomposition and the inherent organic acidic production in forests soils may lower the pH. Compared to other land uses, croplands showed the largest range from lower and upper limit of reported pH baseline (3.8–8.0). This is indication that pH in croplands can be powerfully variable as a result of differences in soil management and cropping systems but likewise the difference may simply be due to the difference in soil units.

More interestingly, results delineated a considerable variability of element concentrations across different land use allocations (Table 1–3). The most abundant elements in croplands were Al, Fe, and Mn with respectively 63.4, 42.2, and 57.6 mg.kg^{-1} on average. One of the typical characteristics of tropical soils is strongly altered soils with low pH [28]. Coincidentally, Al, Fe, and Mn were also the most abundant elements in forests, with values averaging 222.2, 162.6, and 163.0 mg.kg^{-1} respectively. In mining areas on contrary, soils were dominated by Cu and Zn, with respectively 424 and 408.2 mg.kg^{-1}. This accumulation could be linked to the complexation of these cations with organic acids in forest soils.

We suppose that these element concentrations do not reflect their natural abundance since ongoing human activities related to agriculture and mining are likely to alter natural distribution and concentration patterns of metals in comparison to their concentrations in the underlying parent materials. However, baseline element concentrations presented here, reflect their common occurrence in the study area and can thus be used as reference values for soil quality. The high concentrations of trace elements in the soils of different sites in Katangese Copperbelt area have already been reported by several authors [12,29,30]. The upper limits of Cu and Co concentrations in forest soils were higher compared to cropland soils. This is an indication that in Katangese Copperbelt area, even non-disturbed areas can contain significantly higher Cu and Co without a direct human activity. Nevertheless, Cu, Co, Pb and Zn contents we found in forest and cropland soils were significantly lower than those reported by [13] as upper limits of their natural geochemical background (400, 50, 120 and 200 mg.kg^{-1} for total Cu, Co, Pb and Zn respectively) in the Lubumbashi region. Moreover, the ranges of trace element concentrations reported in this study are significantly wider than those reported in several studies carried out in other regions [31] [32-35]. These discrepancies could be related to differences in sample size, sampling strategy, sampling areas, and analytical methods. Indeed, contrast to [13] who used data reported in studies that focused in potentially polluted areas with sample size lower than 50, in this study, 490 samples were used, thus covering a large variability of soils.

In the Katanga Copperbelt in general and the Lubumbashi area in particular, mining activities have likely modified natural metal contents in soils, making comparisons with international or regional reference values inappropriate for environmental studies. It was reported that in all cases, total Cu, Co, Zn, and Pb concentrations are higher at the surface than at depth in Lubumbashi soils [13]. It is therefore possible that the elements concentrations observed across land uses in this study are a result of a combination of the soil

geochemical background naturally rich in these elements [36,37], and that of the intensification of anthropogenic activities [38].

We observed that pH was negatively correlated with Co, Fe, and Zn but positively correlated with Cu, Al, and Zn in croplands. However, none of these correlations was statistically significant (Figure 6 a, d, g). In mining areas, pH was negatively correlated with Al while positively significantly correlated with Fe and Zn. In the forests, pH was negatively and significantly correlated with Mn and Co while positively and significantly correlated with Al. These relationships suggest a lack of a direct and strong pH-dependence of measured elements. As pH_{KCl} measured in this study reflects total soil acidity rather actual acidity, element concentrations would likely be strongly related to pH_{water}. Nevertheless, our results showed contrasting and poor relationships of pH with metals depending on the land use. The lack of strong correlations between metals and pH was also observed by [39] who argued that their observation was possibly due to the very wide pH range, having little influence on elemental behaviour.

Conclusion

This study has provided crucial information regarding lower and upper limits of trace elements concentrations expected in surface soils of the Katangese Copperbelt area (KCA). The identified baseline data did not reflect the natural geochemical abundance of elements, suggesting a predominant influence of ongoing anthropogenic activities in the region (i.e Agriculture and Mining) over natural processes. Overall, mining activities were found to induce the greatest influence on the baseline concentrations of elements, particularly for Cu and Zn. Comparatively, Agriculture generated mild effects on the natural background concentrations of metals, as a similar trend of Al and Mn distribution was recorded in croplands and forests.

Results also provided indication of possible variability in natural background concentrations of metals in the region, with forests presenting significantly higher Cu and Co contents than croplands.

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Data Availability Statement: Data used in this study can be obtained from the corresponding author upon reasonable request.

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

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