

Characterization of Trace Metal Elements from Mine Tailings as Major Pollutants in Adjacent Soils: Migration and Impact Study (The Former Kipushi Concentrator Tailings Case)

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Abstract— This research investigation was conducted to identify mine tailings from the Former Kipushi Concentrator, which has been replenished for more than 50 years in a sizable open park in Kipushi's city (D.R. Congo), as a possible source for the dispersal of metallic trace elements including Cu, Pb, Co, Fe, Mn, Cd and Zn in the nearby soils. The stockpiled tailings at Kipushi contain pyrite and metal sulfides that had undergone partial oxidation. As a result, they are constantly a risk of physico-chemical instability and leakage, which could be reflected in the degradation of the surrounding environment such as vegetation, soil, rivers etc. The main purpose of this paper is to assess the contamination of soils in term of trace metal elements total contents and to make the link with the potential source of propagation which is the mine deposits. In order to assess the risks of pollution and consider the most effective preventive measures, it will be crucial to define the distribution of copper, cobalt, and other metals (metalloids) in the soil. To accomplish these goals, it has been necessary to conduct systematic stratified sampling, proper sample preparation, rock loading characterization, chemical analysis, and statistical analysis to gauge the spread of metals more accurately from mine wastes to surrounding natural soils. Results showed that the average concentrations of metallic trace elements in soils gradually decrease as we move further away from mine wastes park whereas the results regarding rock loading characterization revealed the inverse pattern. In the three zones studied (mine tailings, lightly vegetated soil, and densely vegetated soil or control soil) we had an average Copper concentration of 1291.3 mg/kg for the first zone, 1200.3 for the second and 187.2 mg/kg for the third. In terms of Cobalt, values were 76.79 mg/kg, 61.58 mg/kg, and 32.48 mg/kg, respectively. The average lead concentration ranged between 767.7 mg/kg and 574.5 mg/kg in the first two zones before falling to 29.5 mg/kg in the third. This kind of research allows the feasibility to estimate the potential for environmental contamination by determining how Copper, Cobalt, and other metallic trace elements mine tailings pile of the affect the neighboring natural soils.

Index Terms—Tailings, trace metal elements (TMEs), rock loading, vegetation, soil, contamination, mining (key words)

I. INTRODUCTION

Mining and extractive metallurgy operations always result in large amounts of mine tailings with considerable concentrations of metals and metalloids, which are one of the most important sources of environmental contamination through the release of trace metallic elements into it[1, 2]. To explore the dispersion and effects of heavy metals in soil and water systems, studies of these substances have frequently been conducted close to mines and mine tailings[3]. In addition, natural soils can be greatly affected by tiny dust particles carrying metallic pollutants from mine wastes[4, 5].

Unlike climate change, soil pollution was not considered as a "mainstream" issue, even if it could still have a significant impact on the environment and people's health. This lack of interest can be partially attributed to the fact that it is a relatively recent issue that wasn't taken into consideration after the second War World II[6]. In addition, World Soil Day was just established in 2002[7]. Compared to air and water, the concern for soil remediation has been delayed because the soil has too long been viewed as a kind of unlimited container for our waste and pollutants produced either industrially or from urban activities[8, 9]. In fact, the purity of the air we breathe and the water we drink is directly influenced by the soil, which serves as the common point amid the hydrosphere, biosphere, and atmosphere[7, 10]. The industrial legacy of the mining city of Kipushi in the Democratic Republic of the Congo has a negative impact on both the environment and the quality of life. Industrial effluents have long been a source of environmental degradation in mining communities in Katanga, particularly in the soil and surface water[11]. We still count polluted sites covering several hectares in this area, so understanding and protecting this environment are critical issues today. These metallic trace-contaminated sites, which are typically found close to natural areas and runoff water, have a significant effect on the natural environment such as soils[12, 13].

This paper seeks to describe the level of TMEs contamination in the adjacent soils to the mine tailings site. Are these discharges a possible source of pollution, and do they affect the soils nearby and is there any evolution in the dissemination of those TMEs? These are the inquiries that this work will attempt to address. Kipushi, a mining city, is situated in the province of Katanga in the southern Democratic Republic of the Congo, 600 meters from the Zambian border and 30 kilometers southwest of Lubumbashi. Like southern Katanga, Kipushi has a tropical climate. It has two distinct seasons: a wet season and a dry season. Kipushi, a small mining community, is a part of the Kafubu river basin[14]. To processing the Copper-Zinc sulphide ores from the Kipushi mine,

this concentrator was put into service in 1935. It first treated these ores using straightforward conventional flotation, followed by differential flotation, which was primarily done in two stages with Copper ores that were rich in Zinc [12]. The concentrator ceased routine operation in 1993 for material and financial reasons. The processed Copper-Zinc ores had a gangue that was primarily dolomitic and contained 2 to 4 percent copper and 8 to 13 percent Zinc. Huge amounts of rejects were produced as a result of the differential Copper-Zinc flotation at OKC starting in the 1980s, 3,000 to 3,400 t/d for 4,000 t/d of ore fed[12].

These flotation wastes have been stored on the surface since the 1960s, many years after the concentrator was put into operation. As the concentrator's capacity increased, three parks were built downstream of the mine complex to create the tailings ponds in the Kipushi River valley. After decantation, the solid particles were retained in these dumping areas. The deposit's chemical and mineral makeup indicated that it was polymetallic. The Katanga region is well-known for its copper and cobalt ore deposits, which exhibit enrichment in these two metals as well as other significant metals and metalloids (Zn, U, As, Cd and Pb)[15]. Chemicals like Zn, Co, As, Mn, Cd, Fe, Pb, Hg, Cu, Al, alkalies and acids etc. are present in industrial wastes and contribute heavily to the soil pollution influencing the soil's physical, chemical, and biological features[16, 17]. This affects the soil's productivity[7, 18], for example in terms of vegetation's variation difference as we noticed it around mine tailings from the OKC. The Kipushi deposits are one of the deposits used to extract these elements from the Katanga Copperbelt[15, 19]. By analyzing the propagation of Copper, Cobalt, Iron, Lead, Manganese, Zinc, Cadmium from the second tailings pile of the former Kipushi Concentrator on the surrounding natural soils, this paper enables us to assess the punctual environmental pollution.

II. MATERIAL AND METHOD

Instrumentation and reagents

In order to geolocate the study site, to collect samples, and to conduct spectrometric analysis, the following instruments were used: a handheld GPS Units, a sieve, a decimeter, a straw mattress, a mini crusher, a ceramic crucible and pestle, aluminum foil, a pHmeter, a beaker, a pickler, filter paper and a funnel, a spatula, an oven (scientific and labex), an analytical balance, a stove, an atomic absorption spectrometer (AAS) and an atomic emission spectrometer-induced coupled plasma (AES-ICP). All remaining chemicals that we utilized, including HF, HClO₄, HNO₃, and distilled water, have been found in the laboratory.

METHOD

Material and Sampling

Soil samples from the 3 studied zones served as material in this work. The "stratified" sampling method was performed to characterize the distribution of copper, cobalt, and other related TMEs. Vertical and Lateral distributions were done. Vertically, sampling was carried out from the bottom to the top of the owl's profile, and the layers were measured at a centimeter-scale evaluation. The choice of layers was determined based on the color and texture differences. 15 samples were therefore collected, 3 from profile A layers, 5 from profile B, and 7 from the seven layers of profile C. Each packaged and labeled sample weighed at least 500 g. Horizontally (Laterally), surface sampling was done in each region under investigation. 3 punctual samples were obtained at 10 cm deep from each profile (stratum). Figure below shows the sampling zones related to lateral and vertical samplings.



Figure 1. lateral and vertical sampling views on the studied profiles

The studied site is about 30 kilometers from the Kipushi mining city boundaries (11° 45' 54' South, 27° 14') in the Katanga Province located in the South-East of the Democratic Republic of Congo and belonging to the natural region rich in copper ores called Copperbelt.

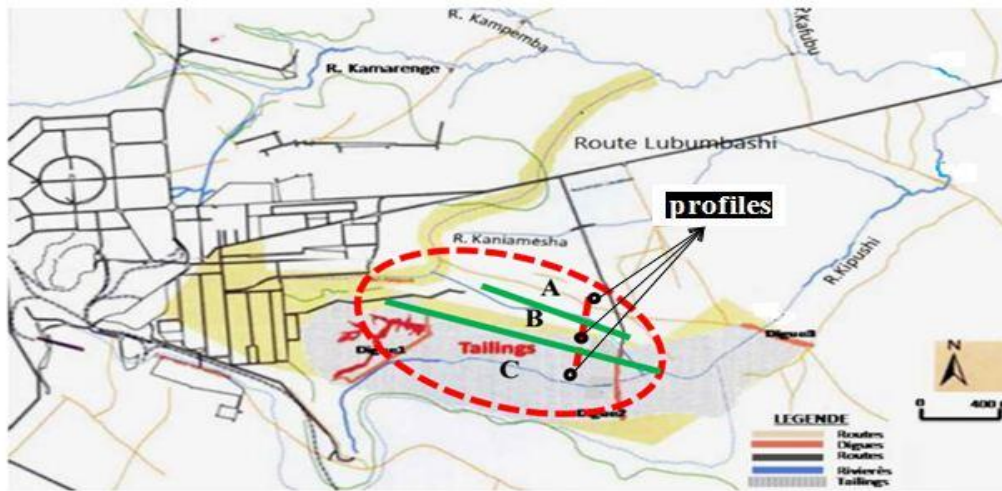


Figure 2. Location (map) of the sampling site adapted and modified from [19]

Rock loading determination

The objective, here, was to estimate the quantity of fine soil which feeds vegetation in the 3 areas under study. The following procedures must be followed when crushing the soil samples in order to determine the stony load: 500 g of the sample should be weighed, crushed before being run through a sieve (mesh ≤ 2 mm). The collected fraction should then be weighed, and the percentage should be calculated by comparing the weights of each fraction with the original sample.

pH measurement

To determine pH values, 10.0 g of soil precisely were measured out, poured each weight into a beaker, and mixed into 50 ml of pure water. A magnetic stirrer was used to swirl the liquids for 30 minutes. A filter was put on top of a funnel for filtration of the mixture. A buffer solution was used to calibrate the pH-meter before its use.

Chemical analysis of soils

Pre-conditioned soil samples were sent to the mineral analysis laboratory of the Congolese Office of Control/Lubumbashi for the determination of total metal concentrations and pH values. Measurement of organic matter and moisture content were done as well but will not be discussed in this work. The following steps were used to carry out the chemical characterization of the TMEs from soils: In a Teflon beaker, 0.25g of the finely ground sample was weighted and stirred in 2ml of HF, 4ml of perchloric acid, and 10ml of aqua regia. Then the solution was gently boiling until dry for 20 minutes. The Beaker was taken off from the plate and filled with 2ml of HCl and then reheated until dry. The beaker was filled with 20ml of purified water and 5ml of HCl; and the salts were heated for 5 to 10 minutes to be dissolved. After cooling down, the solution was transferred to a 250ml flask and brought to the mark with distilled water. The solution was then homogenized and diluted. In a 100ml flask, 2ml of solution and 2ml of HCl were added to make the medium acidic (2%) and brought to the gauge line. Finally, a certain volume from the resulted solution was analyzed through spectrometers (AES-ICP).

Statistical analysis

The characteristic differences of the strata for each studied parameter were evaluated by analysis of variance. The software Statistix 8.0. allowed us to perform this test. Correlations among variables were assessed using Pearson’s correlation method at a significance level of $p < 0.01$ by using Statistica 2007 software.

III. RESULT AND DISCUSSION

Vertical distribution of TMEs: Profile A

Rock loading measurement

The results showed that the stony load increases as we move deep down as shown in figure 3. This can be explained by the fact that the soil of Kipushi and southern Upper Katanga in general is lateritic.

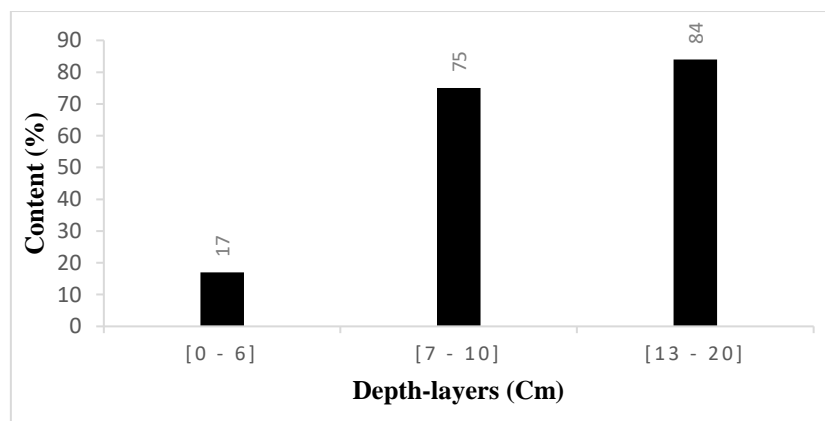


Figure 3. Rock loading variation

Total TMEs concentrations measurement

Table 1. Total concentrations of TMEs

Total Concentrations (mg/kg)							
Layers (cm)	Cu	Co	Cd	Zn	Pb	Fe	Mn
[0 - 6]	490,5	87,04	17,86	3340	140,6	24290	773,7
[7 - 0]	150	27,83	7,09	1010	0	12450	126,1
[13 - 23]	302,9	24,92	3,101	942	0	12870	88,86

When looking at the results in the **Tab. 1**, we can observe that for most of the elements, the concentrations drop with depth. In comparison to the other factors in this soil, copper, zinc, and iron have very high values. Apart from copper, whose values somehow deviate from the group for this profile, we can also see that they all have values with comparable trends. Note that the copper-zinc sulphide ores were exploited at the FCK, while the Katanga Copperbelt region is rich in copper-cobalt ores basically[16].

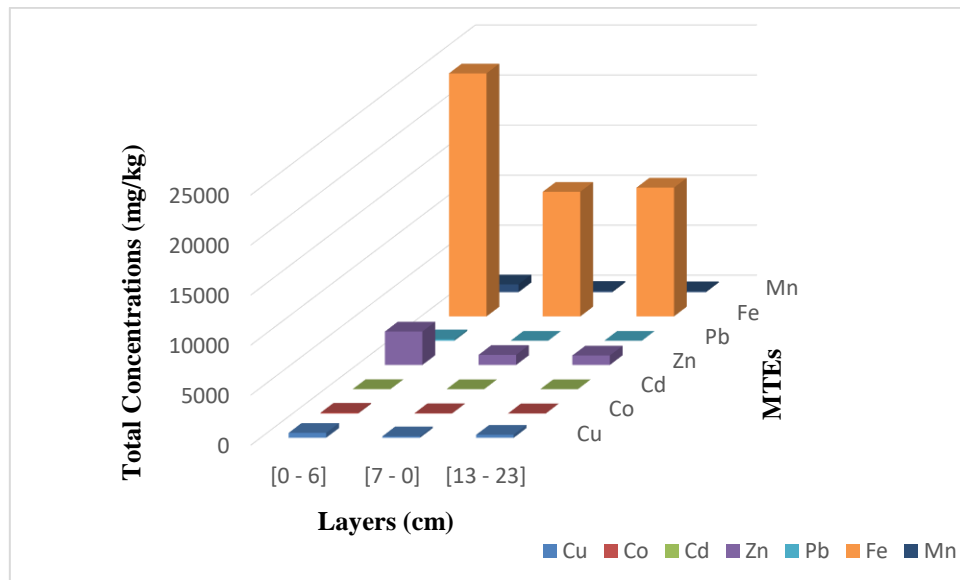


Figure 4. Total Concentrations (mg/kg) of TMEs: Profile A

Vertical distribution of TMEs: Profile B

Rock loading measurement

According to the data in the Figure below, the stony load content is 0% across the profile's uppermost layer before increasing to 86% on the last layer. The lateritic layer is where this last layer of profile B stops, while the layers above are primarily comprised of deposits as shown in the **Figure 2**.

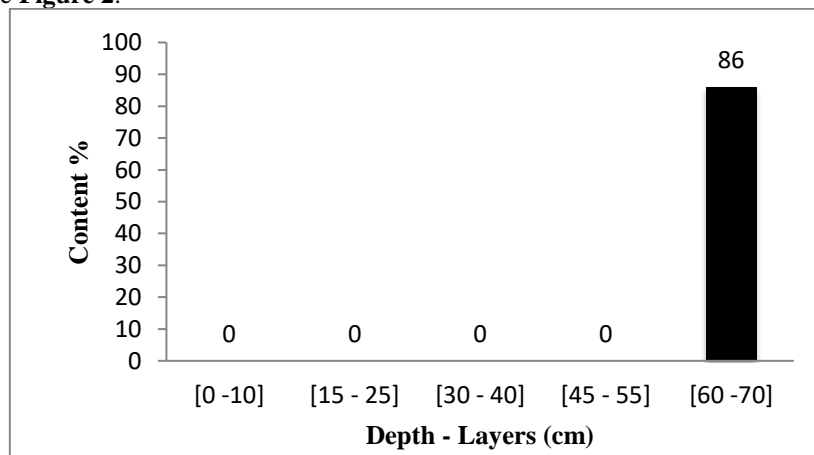


Figure 5. Rock Load Variation

Table 2 Total concentrations of TMEs

Total Concentrations (mg/kg)							
Layers (cm)	Cu	Co	Cd	Zn	Pb	Fe	Mn
[0 - 7]	2518	113,4	126,4	4500	126,3	45220	1375
[15 - 25]	1190	51,85	114,2	4710	45,85	54490	1246
[30 - 40]	1330	98,54	86,07	4880	1,204	67590	1150

[45 - 55]	1333	74,54	164,5	4880	1821	29750	1166
[60 - 70]	2048	87,07	99,39	5390	274,5	26230	1102

Like the heavily vegetated soil layers, we see that iron and zinc always have the highest concentration values over the entire depth. Zinc concentrations are essentially stable, whereas iron concentrations change with depth by getting lower. While Copper concentrations are comparable to those of manganese, Cadmium and Cobalt exhibit similar trends. Compared to the other group factors, lead exhibits a random and more specific pattern. In relation to iron, a significant element for which very high levels can still be found in the soil, Copper concentrations of more than 1000 mg/kg present a state of pollution.

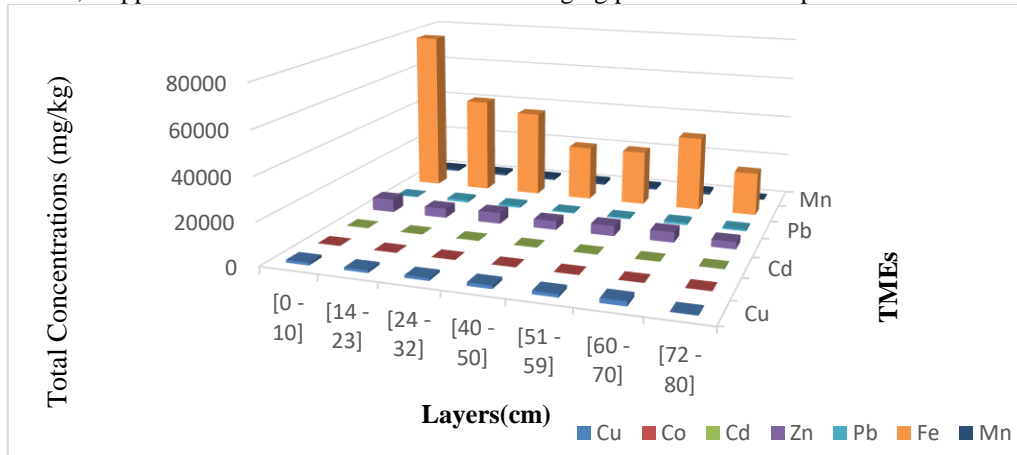


Figure 6 Total Concentrations (mg/kg) of TMEs: Profile C

Vertical distribution of TMEs Profile C

Table 3 Variations in the concentrations of the trace metals studied in the bare vegetation zone.

Total Concentration (mg/kg)							
Layer (cm)	Cu	Co	Cd	Zn	Pb	Fe	Mn
[0 - 10]	1273	102,5	127,8	6350	374,1	77940	1109
[14 - 23]	1259	66,73	99,08	4530	1169	46170	1278
[24 - 32]	1228	73,93	151,2	5290	990,4	41850	1216
[40 - 50]	1554	38,77	95,85	4140	470,1	26390	1380
[51 - 59]	1557	63,74	76,18	4850	483,6	26550	1191
[60 - 70]	2088	39,47	155,6	4970	1049	35890	1271
[72 - 80]	177,9	10,58	12,81	3440	776,2	20760	160,1

Regarding concentration values that are close to each other, the vertical distribution of the elements studied in the area without vegetation (waste park of the old Kipushi concentrator) is much more like that of the area with clear vegetation; that is, after the area without vegetation, it is the area with clear vegetation that gives high total contents of the studied metals. We can also see that the pieces are scattered virtually identically on this profile. Once more, Copper exhibits values over 1000 mg/kg; for instance, it reaches 2088 ppm in the second-to-last layer before falling to 177,9 ppm in the last layer. Regarding the iron, it exhibits more very high values in this area, qualifying it as a contaminant as well. In both the previous and current profiles, the Copper curve exhibits patterns that are comparable to those of Iron, i.e., Copper exhibits the same patterns as iron while it is dropping or increasing.

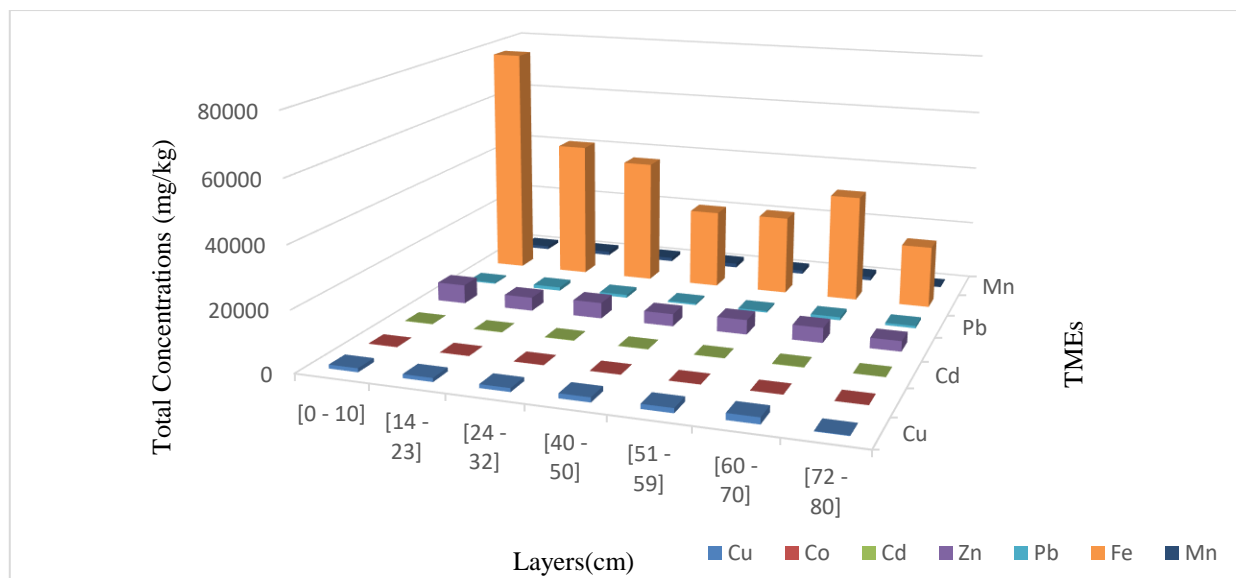


Figure 7 Total Concentrations (mg/kg) of TMEs: Profile C

Lateral (horizontal) distribution of TMEs

Table 4 lists the findings about lateral distribution of Copper, Cobalt, and other examined metals in areas with dense, moderate, and in the one without vegetation (Profiles: A, B and C respectively). The average concentration values resulting from samples obtained in triplicate from each stratum are shown in the table below. Analysis of variances (ANOVA) results. It should be mentioned on bare deposit and soil with moderate vegetation no rock load was found while 83% of rock load was recorded on the control soil. The a, b, and c letters stand for the ANOVA outcome. Simply put, the presence of two of them indicates that the region has an intermediate character. The results of the ANOVA performed on the amounts of metallic elements in the 3 strata under study are shown in Table 4. This study determines if the chosen regions vary from one another in relation to each parameter under consideration. As a result, we noticed that the average concentrations of Copper, Lead, Cobalt, and Cadmium allowed us to separate the bare deposit to the regular soil, with the discharges being substantially concentrated in copper compared to the normal soil. Given that both discharges are byproducts of mine process, and that Cu and Co were the materials extracted by this plant, the outcome is easily explained.

Because this element is connected to them, it may be possible to explain why Cd is at that place. Fe identified two zones that were notably distinct from one another, with the thinly vegetated discharge serving as a transitional zone between the naked discharge and the typical soil. The area of great concentration is the naked discharge. The most intriguing finding is that given by Pb, which shows how radically different the three strata are from one another. The naked discharge has the greatest mean value (767.7±61.18), followed by the open vegetated discharge (574.5±46.88), while the control soil has the lowest Pb content (29.5±35.083) ppm. According to our study, which was based on the usage of the land in terms of vegetation like predicted by our sight, Lead would thus be the primary factor that distinguishes the three layers. Based on the mineralization of the Kipushi Mine's orebody, which has average (%) concentrations of 0.17 of arsenic, 2.12 of copper, 0.49 of lead, 11.57 of zinc, 13.14 of sulfur, and 8.21 of iron, this might be explained[20]. However, the three zones were barely distinguishable from one another by Zn, Mn, and pH values[9]. Copper, Cobalt, Lead, Manganese, Zinc, Cadmium, and Iron were all measured. The elements at play were Copper and Cobalt, while the related elements were Cadmium and Zinc. The effects of Fe and Mn on the distribution of TMEs in the soil were evaluated. The components of Fe and Mn (hydroxides and oxides) in soils adsorb the MTEs in their structures, decreasing their mobility. They are parts of the TMEs carrier phases in soils. Given that Iron is a significant metal and one of the most significant components of the earth's crust, this may have explained why its values were found to be high[4]. It should be mentioned that the pH values of the studied soils were found to be acidic, with an average value of 5.5. This finding demonstrates that each profile under study has acidic soil, which has a high capacity to mobilize trace elements as was examined in several earlier works[9].

Mean concentration (mg/kg) and standard deviations (SD)								
Studied Zones	Cu	Co	Cd	Zn	Pb	Fe	Mn	pH
Profile A	187±59,73 ^b	32,48±13,71 ^b	48,34±33,29 ^b	4346,7±1972 ^a	29,53±5,083 ^c	29273±12411 ^b	565,5±370,6 ^a	5,933±0,115 ^a
Profile B	1200±112 ^a	61,58±6,992 ^a	105,3±3,732 ^a	5716,7±116 ^a	574,5±46,88 ^b	42990±2548 ^{ab}	923,9±14,38 ^a	5,668±0,231 ^a
Profile C	1291±17,21 ^a	76,79±2,701 ^a	119,1±10,72 ^a	6910±1385 ^a	767,7±61,18 ^a	61930±13515 ^a	954,8±11,28 ^a	5,467±0,252 ^a

Table 4 Analysis of the mean concentration carried out on the three studied strata (Anova One Way)

Parameters Correlations

It is always important to consider the different interactions that characterize the environment being investigated to forecast how particular elements will behave in concert in a substrate with established features. The normal soil (profile A) samples were utilized as references for the correlation but were not used in the actual study. Only the discharged soil (profiles B and C) samples were used. Table 5 displays the findings from this investigation. Thus, it was discovered that Cu had a somewhat positive correlation with Cd and Co ($r = 0.52$) and a substantial positive correlation with Mn ($r = 0.75$). This finding would clarify why only two of the three research regions were stratified by the three metal elements. The genesis of these three components would be the same.

Table 5 Correlations among studied factors ($p \leq 0,05$)

Correlations between factors								
Factors	Cu	Co	Cd	Zn	Pb	Fe	Mn	pH
Cu	1,00							
Co	0,52	1,00						
Cd	0,52	0,44	1,00					
Zn	0,05	0,38	0,34	1,00				
Pb	-0,21	-0,27	0,36	0,05	1,00			
Fe	-0,04	0,56	0,26	0,53	-0,28	1,00		
Mn	0,75	0,46	0,64	-0,02	-0,09	0,12	1,00	
pH	0,31	0,34	0,16	-0,28	-0,21	-0,01	0,31	1,00

In addition to Cu, Co also demonstrated an excellent correlation with Fe ($r = 0.56$). In contrast, Cd also had a positive correlation with Mn ($r = 0.64$) in addition to Cu. Fe and Zn have a $r = 0.53$ positive correlation. There was no correlation between Pb and pH and any other soil parameter. Other variables unrelated to mining activities may be able to explain how they are distributed [21]. The "statistica" program was used to investigate the correlations between the various parameters, and it provides a 95% correlation significance between the associated values. Only five significant relationships were shown by the statistical findings, and these were those between: Cobalt is 52% correlated with Copper; Cadmium is 52% correlated with Copper; Iron is 56% correlated with Cobalt and 53% correlated with Zinc; and finally, Manganese is 64% correlated with Cadmium [4, 20].

IV. CONCLUSION

The discharges from the former Kipushi Concentrator, having undergone physicochemical destabilization, are potentially dangerous and can be characterized as a source of pollution for the nearest environment first and the most distant to varying degrees; their fine granulometry allows them to be moved by the wind up to several kilometers. The results of the different statistical tests of distribution sufficiently demonstrate that the total concentrations and pH values for the samples from the deposition zone are like the nearest zone (vegetated ground), but the control zone (with abundant vegetation) shows the opposite reality. This part of the situation could better explain why the vegetation is shown in 3 distinct ways depending on which of the 3 research regions we are in.

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