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Heavy metals impacted soils from dumped municipal solid waste in Buterere - Burundi: Health risk assessment

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ABSTRACT: Municipal solid waste (MSW) landfills without any soil protection constitutes an environmental risk factor in particular heavy metal pollution. These sites, located close to inhabitants, can induce contamination through the food chain. The present work aims to be a contribution in measuring heavy metals concentrations generated by unsorted waste dumped from 1983 until 2008 on Buterere open air landfill in Bujumbura to assess soil contamination, pollution levels and health risk. Materials used are soil samples collected from 15 points at the Buterere site. The physicochemical characterization of the soil was carried out by thermogravimetry (TGA) and X-ray diffraction (X-RD). The determination of heavy metals was carried out by X-ray fluorescence (X-RF) and Inductively Coupled Plasma atomic Spectroscopy-Optical Emission (ICP-OES). Chronic carcinogenic and non-carcinogenic risk indices were evaluated based on experimental results in order to formulate adequate prospects for remediation. Results show that the heavy metals are not homogeneously dispersed on Buterere site but their concentration levels do generally exceed the limit values (World Health Organization, European Union). A significant correlation ($p < 0.05$) was demonstrated for the simultaneous presence of the micropollutants of Cd, Cu, Fe and Zn in the study area. Based on these results, the health risk assessment reveals that the Buterere site can induce carcinogenic and non-carcinogenic nuisances to the population in its and therefore remedial measures are proposed.

KEYWORDS: MSW, heavy metals, soil pollution, contamination, health risk.

1 INTRODUCTION

Municipal Solid Waste (MSW) landfill, without any prior treatment and without any protection of the ground, constitutes a major risk of contamination of soil and groundwater by heavy metals [1]. MSW landfill leachate is a potential source of pollutants, dangerous for the environment [2]; [3]. The use of MSW issued from a Beijing landfill was evaluated and the presence of heavy metals in the soil has been demonstrated.4 According to this study, the concentrations of some heavy metals were very high, exceeding consequently the norm, and, experiments have shown a significant eco-toxicological impact on plant growth [4]. An investigation on the temporal evolution (1987-2015) of the Naameh landfill in Lebanon was carried out using the Multispectral Spaceborne Imagery (MSI) method and showed that the landfill has a negative effect on the surrounding vegetation [5]. The analysis of temporal images of landfills by satellites has shown a progressive reduction of the surrounding vegetation.

In urban areas, closed and technically arranged dumping sites constitute land resources (agriculture, housing) and have harmful effects on the environment [4]. This is the case of the closed landfill of El Yahoudia in Tunisia, 30 ha of which (out of 100ha in total) have been transformed into an urban leisure space [5]. The issue of the presence of heavy metals had been confirmed in raw foods from open markets in two African cities (Kinshasa and Johannesburg); the results showed the presence of heavy metals with contents exceeding the admissible values, without however specifying their origin, which constitutes a challenge [6]. However, heavy metals classified as most dangerous for the environment do not undergo degradation during physicochemical processes and therefore persist for long periods [7]; [8]. They affect then the biogeochemical cycles and do accumulate in living organisms, including humans, through the food chain where they can disrupt biological reactions and induce various pathologies or even death [7]. Ingestion, inhalation and skin contact are the main routes of human exposure to heavy metals present in the soil [8].

Measuring the content of heavy metals in the soil is useful in order to assess their dispersion and the consequences for the environment due to natural and anthropogenic factors [9]. In fact, the importance of their impacts is a function of several natural factors such as precipitation, temperature, climate and relief as well as anthropogenic factors such as agriculture, livestock or urbanization [1]; [10]; [11].

In order to support the existing literature, there is a need to overcome the problems linked to MSW specifically in Burundi, one of Great Lakes' countries given that this region has climatic particularities [12]; [13]. The purpose of this study is to evaluate the heavy metal contents of the soils at the Buterere site, where MSW from the city of Bujumbura, the Capital of Burundi, have been dumped for more than 25 years. By carrying out the physicochemical characterization of samples of soil collected on this site and its surroundings, we can deduce the environmental risk incurred by the population living in the neighbourhood of this site and suggest some perspectives to improve the actual situation.

2 Materials and Methods

2.1 Study's Area And Sampling Sites

Buterere site is a former landfill located North of Bujumbura, and next to the wastewater treatment plant (Fig.1). This site has been the dumping ground of MSW from all sources, since 1983, and was formally closed in 2009. At the beginning, the surface of the site was outside of Bujumbura but the gradual enlargement of this city meant that the site first became surrounded by urban housing areas, then gradually transformed into living space, starting from its ends. The total surface area of Buterere site is around 20ha occupied for 2/5 by residential houses, 1/5 by crops and 2/5 by pastures and stagnant swamps.

2.2 Sampling

The sampling sites identified by GPS are noted B1, B2, B3, B4, B5, B6, B7, B8, B9, B10 and CBmix (Fig. 1). To obtain representative samples, soil coring is carried out at a depth varying from 0 to 50cm and at different points that are sufficiently close together. These samples were packed in polyethylene bags for transport to chemical and environmental analysis laboratories. After drying in the open air for one week, a representative sample is constituted by the quartering method to obtain, in the end, eleven soil samples codified as follows: SB1, SB2, SB3, SB4, SB5, SB6, SB7, SB8, SB9, SB10. The sample SBmix results from the homogeneous mixture of several samples collected approximately 20 m away from the external contour of the landfill at points CB12, CB14, CB15.

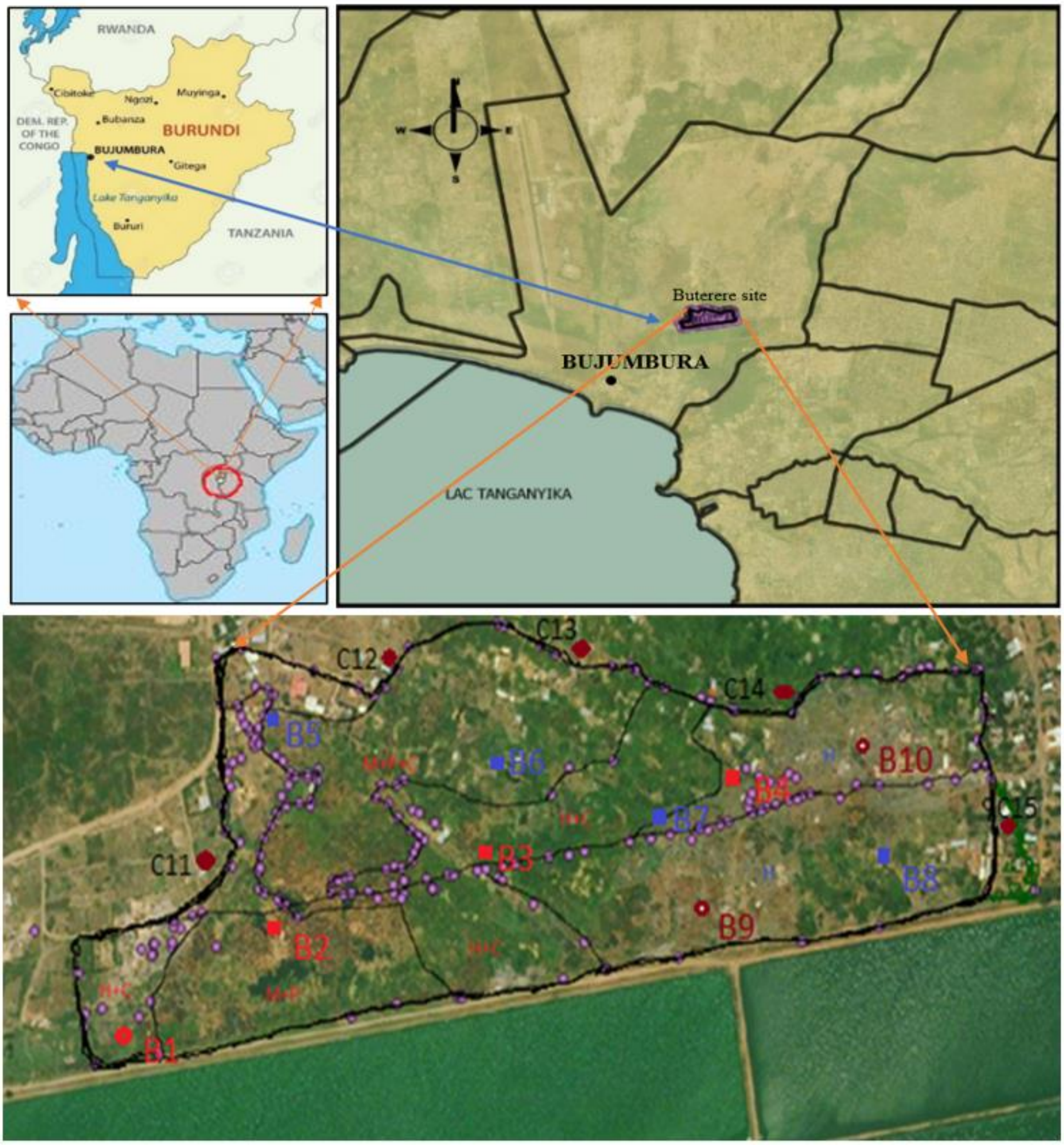

Fig. 1. Study's area and sampling site

2.3 Samples' Analysis

Soil samples from the 15 points on site were previously dried in an oven at 105 ° C and then crushed for 30 seconds. A portion of the powder obtained was subjected to physicochemical characterization, respectively by X-ray fluorescence (XRF) and by thermogravimetry (ATG). The analysis of heavy metals (Al, Cr, Cu, Fe, Mn Ni, Pb and Zn) was performed by X-Rays Fluorescence (XRF) S4 PIONEER type.

For heavy metals such as Hg, As and Cd, with low content in soils, additional measurements have been made by ICP-OES. The wet digestion method described by Binning and Baird [14] was used to prepare the samples for analysis. A sample of about 1 g of the soil was digested with a mixture of concentrated HCl (6.0 ml) and HNO3 (0.3 ml) to almost dry and allowed to cool before adding 20 ml of a solution of 5M HNO3. These digested sample solutions remained for 12 hours before being filtered. Each metal-loaded filtrate was transferred to a 100 ml volumetric flask and brought to line with a solution of 0.5M HNO3. The analysis of this solution by ICP-OES of VISTA MPX type was carried out for the measurement of heavy metal concentrations.

These experimental values were compared with the reference limit values. Descriptive statistics were produced by SPSS 22 and in addition, the Pearson correlation was determined between the metal pairs in order to assess the tendency for the simultaneous presence of the different heavy metals identified in this study. The carcinogenic and non-carcinogenic risk indices were also estimated on the basis of these experimental results, with reference to the risk assessment guides and the manual of exposure factors [15]; [16].

2.4 HEALTH RISK ASSESSMENT

Health risk assessment associated with non-carcinogenic contaminants are typically expressed in terms of the ratio of the determined dose of a contaminant to the reference dose (RfD) below which such contaminants might likely pose any appreciable health risk. This non cancer risk assessment method is the target hazard quotient (THQ) and was determined in this study as described by US EPA [15]; [16].

A person can be exposed to metals by three main routes: ingestion, inhalation and skin contact. If the individual is located near a contaminated site, the approach consists of a set of equations which make it possible to estimate a concentration of a soil contaminant for which exposure is potential via the absorption routes and is equivalent to the concentration of a risk of cancer [16]. This method can be applied to the case of Buterere, where the site is not isolated, the population being exposed to it continuously: from childhood to adulthood because the population erected houses there, practice agriculture and livestock. It should also be noted that a similar case has been identified in Tunisia where part of an old landfill has been transformed into an urban leisure area [5]. The heavy metals content (median values) of soils collected on the Buterere site and analyzed by X-RF and ICP-OES had been then used for this approach, taking into account the admissible values [18]; [19] [20].; [21].

We are therefore inspired by the pollution indices that are commonly used, to assess the health risks linked to exposure to a site contaminated with heavy metals [16]; [17]; [21]; [20]. Thus, for the Buterere site, the daily exposure estimated by oral ingestion and by skin contact was estimated respectively using Eqs. 1, 2, 3,4 and 5:

$$\mathrm{CDI}_{\mathrm{ingest-oral}} = \frac{\mathrm{CSxIRSxEFxED}}{\mathrm{BW\ x\ AT}} \mathrm{xCF} \quad (1)$$

$$\mathrm{CDI}_{\mathrm{dermal}} = \frac{\mathrm{CS\ x\ SA\ x\ AFx\ ABS\ x\ EF\ x\ ED}}{\mathrm{BW\ x\ AT}} \mathrm{\ x\ CF} \quad (2)$$

$$\mathrm{CDI}_{\mathrm{inhale-soil}} = \frac{\mathrm{CS\ x\ ET\ x\ EF\ x\ ED}}{\mathrm{PEFx24\ x\ AT}} \mathrm{\ x\ CF} \quad (3)$$

$$\text{Total cancer risk (CR)} = \sum_k^n \mathrm{CDI}_k \mathrm{\ x\ SF}_k \quad (4)$$

$$\text{Chronic Hazard Index (HI)} = \sum_k^n \mathrm{CDI}_k \,/\, \mathrm{RFD}_k \quad (5)$$

With: CDI: Chronic daily uptake; CS: Exposure-point concentration EF: Exposure Frequency (350days/year); ED: Exposure duration (30 years for adults et 6 years for children); AF: Adherence factor 0.007mg/cm2; BW: body weight: 70 kg for adults and 15kg for children; AT: Averaging time for carcinogens: 365x70days; ABS: dermal absorption fraction: 0,001; CF: units conversion factor: 10−6 kg mg−1 PEF: particle emission factor: 1.36 × 109 m3 kg−1; IRS: ingestion rate: 100 mg·d-1; SA: exposure skin area: 2800 cm2 for children and 5700cm2. SFk is the slope factor for substance k (kg d-1 mg-1); RFDk is the chronic reference dose for the heavy metal k. Acceptable or tolerable risk for regulatory purposes is between 10-6 and 10−4. The values of HI> 1 show that there is a non-carcinogenic risk, and when HI <1 the reverse is applied [16]; [22].

3 RESULTS AND DISCUSSION

3.1 SOIL CHARACTERISATION

Analysis of soil samples from the former Buterere landfill, presently closed, showed a fire loss ranging from 5.6% to 18.8%. The thermogravimetric analysis showed a mass loss of around 1% at 100°C of water and around 7% at 500°C corresponding to the release of CO_2, NO_2 and H_2O with evidence of the presence of decomposing organic matter and around 4% at 750°C corresponding to CO_2, CO and bound H_2O, due to the decomposition of carbonated compounds (Fig.2). The XRD revealed the presence of usual soil compounds such as quartz, feldspaths (albite, microcline), muscovite (or illite), kaolinite (clay), calcite and hematite (Fig.3).

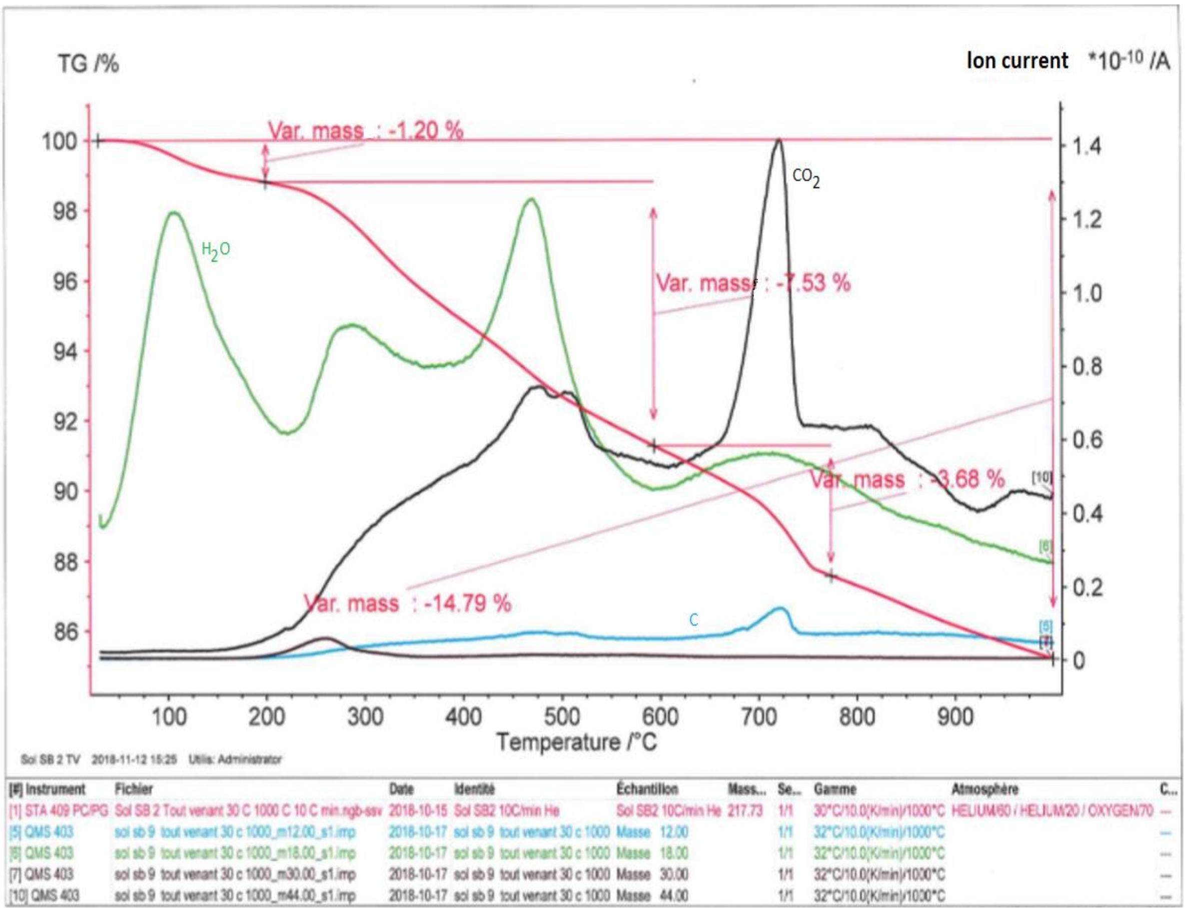

Fig. 2. Thermogravimetric characterization of Buterere soil samples

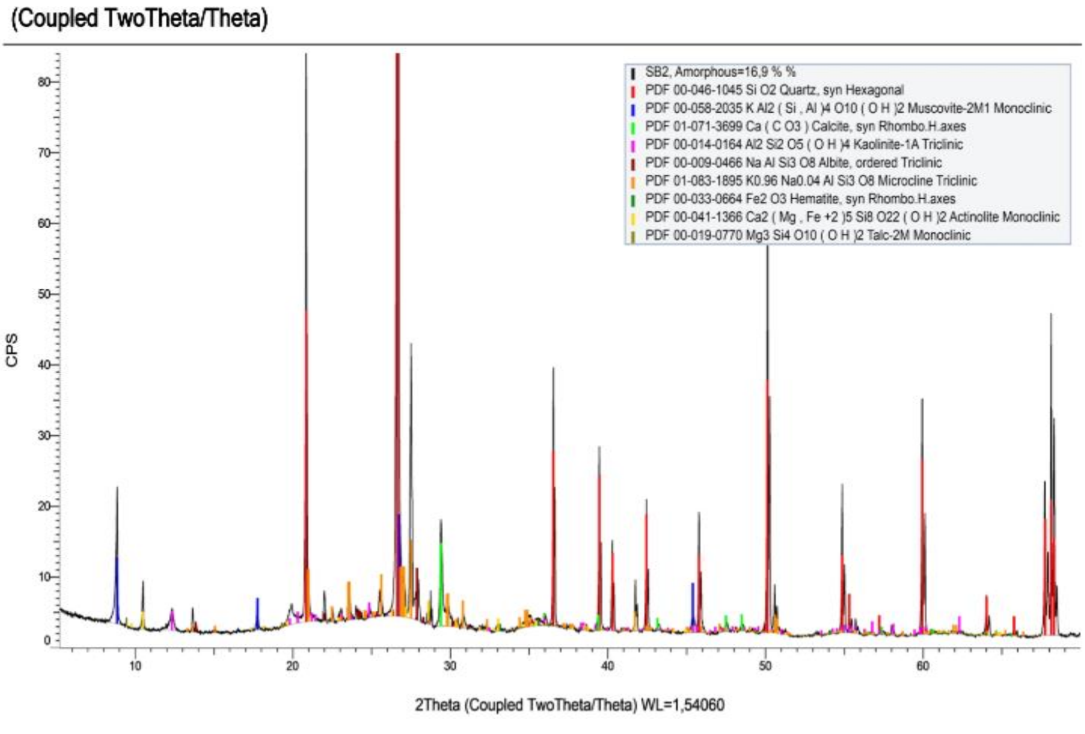

Fig. 3. X-Ray diffraction of soils from Buterere site

The results of XRF and ICP-OES analysis highlight the presence of heavy metals in the soils of the Buterere site. Results of the heavy metal contents are summarized in Table 1, ranging from 48900 to 102300 mg/kg in Al, 0.8 to 4.0mg/kg for Cd, 17800 to 46800mg/kg for Fe, 277 to 1563mg for Mn; 40 to 2503mg/kg for Pb; 686 to 1533mg/kg for Ni; 57 to 236mg/kg for Cu; 84 to 1277mg/kg in Zn, and 300 to 1903mg/kg in Cr. Concentration in As and Hg were below the detection limit.

Table 1. Heavy metals contents in soil samples on Buterere site

Heavy metal	Soils of Buterere (mg/kg)				WHO (1996)	EU (2015)	MEF (2007)
	Minima	Median	Mean	Maxima			
Al	47900	62200	65800±13100	102300	-	-	-
Cd*	0.8	1.6	2.0±0.9	4.0	0.8	3	1
Cr	300	393	520±250	1,903	100	100	100
Cu	57	109	124±56	236	-	100	100
Fe	17800	29400	29400±6200	45800	7000	-	-
Mn	277	904	880±380	1563	-	-	-
Ni	686	1207	170±220	1533	35	90	50
Pb	40	90	350±430*	2503	85	150	60
Zn	84	445	500±310	1277	50	300	200

**Cadmium was analysed by ICP-OES.*

Generally, findings showed that the heavy metals' contents are not homogeneously dispersed on the Buterere site but the concentration levels do generally exceed the limit values [18]; [19]; [20]. The authorized values for the heavy metals content in soils are respectively 7000mg/kg in Fe, 60-150 mg/kg in Pb, 35-90mg in Ni, 50-300 in Zn, 100mg/kg in Cr and 1-3mg/kg in Cd. This study showed that high levels of concentrations of several heavy metals, identified on site and listed, were confirmed and most of them exceeded the reference values (Table 1). For the specific cases of the elements Pb and Cr, it should be pointed out that their high average concentration is due to the fact that high concentrations were measured in two precise places of sampling (in B4 and B9) while the values in all other places of sampling on site were significantly lower. This finding is explained by the fact that the landfill site received various waste, some of which may contaminate a small parcel of the landfill with a specific element (example: Pb batteries). In order to better assess the contamination of the site by heavy metals in relation to WHO requirements, a Student test was carried out which allows a statistical analysis by comparing the average contents with the reference values [20]. The results of this Student test are presented in Table 2.

Table 2. Heavy metals values in the soils compared to reference value of WHO [18]

Heavy metal	t	Bilateral significance (p-value)	Average difference	Confidence interval about a difference at 95%	
				Inferior	Superior
Cd	3.609	0.005	1.2000	0.459	1.941
Cr	3.009	0.013	419.182	108.81	729.55
Cu	1.236	0.245	24.000	-19.26	67.26
Fe	8.758	0.000	22400.091	16700.93	28099.25
Ni	13.415	0.000	1134.727	946.26	1323.19
Pb	1.310	0.220	67.727	-47.50	182.96
Zn	3.844	0.003	446.182	187.54	704.83

It appears that the contents of Cd, Cr, Fe, Ni and Zn at the site significantly exceed the reference values ($p < 0.05$) unlike the average values of Pb and Cu which do not prove that they are in excess ($p > 0.05$). As for the Al and Mn contents, the lack of limit values in the literature has not made it possible to rule on the quality of this site with regard to these elements.

The origin of these various pollutants was probably due to the continuous degradation and decomposition of unsorted municipal solid waste in a wet environment (rainy season) at variable temperatures. The result consisted in some infiltration into the ground and spatial dispersion due to runoff and wind. In addition, the Buterere site is not closed. Cows, goats, chickens,... feed on the premises and the population has already started to occupy the premises as well. Agriculture is practiced on this soil and houses made of sustainable materials are gradually erected there since the site no longer receives municipal solid waste. In view of the results of this study, it is shown that this site has contamination with heavy metals which induces risks for biodiversity in general and humans in particular.

As the sampling has been carried out randomly, it is possible to assess whether there is a correlation between the heavy metals identified. The method which can be useful is that of the Pearson correlation test, carried out using the SPSS 22 software. By introducing the contents obtained in the micropollutants present on the different sampling locations, it is possible to determine the bivariate correlation of pairs of heavy metals, which generates the correlation matrix summarized in Table 3. The significance level of the correlation coefficient ($p < 0.05$) allows us to characterize the validity of the correlation.

Table 3. Correlation matrix for heavy metals concentrations in the soils of Buterere

	Al	**Cd**	**Cr**	**Cu**	**Fe**	**Mn**	**Ni**	**Pb**	**Zn**
Al	1								
Cd	-0.408	1							
Cr	-0.016	-0.083	1						
Cu	-0.660*	0.853**	0.232	1					
Fe	-0.156	0.717*	0.462	0.727*	1				
Mn	-0.629*	0.493	0.346	0.800**	0.604*	1			
Ni	-0.809**	0.522	-0.495	0.513	0.013	0.300	1		
Pb	-0.359	0.649*	-0.031	0.505	0.694*	0.278	0.457	1	
Zn	-0.613*	0.665*	0.398	0.848**	0.829**	0.804**	0.395	0.724*	1

** The correlation is significate at the level 0.05 (bilateral) ** The correlation is significate at the level 0.01 (bilateral)*

Examination of Table 3 shows a significant correlation for the simultaneous presence of certain metals on the entire Buterere site. It is found that the presence of Cd is positively correlated with that of Cu, Fe and Pb ($p < 0.05$). Likewise, the presence of Fe is positively linked to that of Mn and Zn ($p < 0.05$). The significant correlation between the values of heavy metal concentrations reflects their simultaneous release, the origin of which is that of an identical source of MSW deposits on the old landfill, as pointed out in UYO [23]. In other words, these correlations reveal heavy metal contents linked to heterogeneous waste dumped in the past on the old landfill which induced a significant contribution in pollutants on the site. It goes without saying that certain end-of-life objects can contain variable charges of heavy metals which unfortunately end up in the ground at the landfill sites.

3.2 Risk Assessment Of Health And Environment

The average heavy metal contents (Table 1) obtained for the site under study were used to assess the indices of carcinogenic and non-carcinogenic risks run by children and adults. Table 4 summarizes all the results obtained using these values on the basis of the methodology proposed in similar recent studies [17]; [22].

For children, the results show that, for ingestion, the estimated value of the chronic non-carcinogenic index (HI) is 2.94. It should be noted that the admissible value of HI must be less than the unit (Table 4). So, the value obtained is then greater than the guide value of 1. It should also be noted that this index of 2.94 is greater than that of the UYO site in Nigeria17 whose value was 1.04 x10-2. This high value of 2.94 means that children 0 to 6 years old, exposed to this site, can develop chronic non-carcinogenic diseases during their growth and this result shows the vulnerability of their state of health.

Table 4. Non carcinogenic index (HI) and carcinogenic index (CR) by ingestion, contact and inhalation on Buterere site

Heavy metal	Ingestion			skin contact			Inhalation		
	HQi Children	HQi Adults	CRi	HQi Children	HQi Adultes	CRi	HQi Children	HQi Adults	CRi
Al	8,41E-01	9,01E-02	-	8,24E-04	3,59E-04	-	6,85E-06	1,37E-05	-
Cd	2,56E-02	2,74E-03	1,04E-06	2,51E-03	1,09E-03	4,15E-09	2,08E-05	4,16E-05	3,96E-11
Cr	4,43E-03	4,75E-04	3,56E-08	2,17E-03	9,47E-04	1,42E-10	1,80E-05	3,61E-05	1,35E-12
Cu	3,96E-02	4,25E-03	-	3,88E-05	1,69E-05	-	3,23E-07	6,45E-07	-
Fe	5,37E-01	5,75E-02	-	5,26E-04	2,30E-04	-	4,37E-06	8,74E-06	-
Mn	8,04E-02	8,61E-03	-	6,13E-03	2,67E-03	-	5,09E-05	1,02E-04	-
Ni	1,09E-01	1,16E-02	3,96E-04	1,07E-04	4,65E-05	1,58E-06	8,85E-07	1,77E-06	1,50E-08
Pb	1,28E+00	1,37E-01	4,08E-08	8,35E-03	3,64E-03	1,62E-10	6,94E-05	1,39E-04	1,55E-12
Zn	2,13E-02	2,28E-03	-	1,04E-04	4,55E-05	-	8,67E-07	1,73E-06	-
HI=∑HQi CR =∑CRi	2,94E+00	3,15E-01	3,97E-04	2,08E-02	9,05E-03	1,58E-06	1,72E-04	3,45E-04	1,51E-08

This risk is proportionally linked to exposure to Pb (43%), to Al (28%); Fe (18%) and Ni (3.7%). For adults, the chronic non-carcinogenic index (HI) related to oral soil exposure is 3.15x10-1; this value is below the reference value from which exposed adults could not develop chronic diseases. Thus children are the most at risk category for chronic pathologies compared to adults and this corroborates the results of the reference [24]. However, the HI indices for children and adults for the Buterere site are greater than 1.04x10-2, value obtained for the UYO site in Nigeria [23]

In addition, the average carcinogenic risk index (CR) for the Buterere site is 3.97x10-4 and exceeds approximately 4 times the required value (in the range from 10-6 to 10-4). The population living closely on the site under study could develop carcinogenic pathologies during their life (current life expectancy from 0 to 60 years in Burundi). This carcinogenic risk is mainly associated with exposure to Ni present in the soils of the Buterere site. Moreover, other elements such as Cd, Pb and Cr do contribute by increasing this risk. Compared to that found in Nigeria [17] which was 5.71x10-3, the value found in this study for oral ingestion is higher showing a higher risk for children to develop a cancer.

As for skin contact and soil inhalation, the values obtained from these indices remain within the range of admissible values, as shown in Table 4. Indeed, for skin contact, the HI index rises to 2.08x10-2 and 9.05x10-3 respectively for children and adults. These values are low if compared to the reference values equal to the unit. The same observation is highlighted for exposure to inhalation where the average value of this chronic non-carcinogenic index HI rises to 1.72x10-4 and 3.45x10-4 respectively for children and adults. These values remain below the acceptable limit of 1.00, which shows that, near this site, the inhalation of particles does contribute less than oral ingestion and chronic nuisance for children and adults. The value found for the carcinogenic risk is 1.51x10-8, lower than the reference value of 1.00x10-4 which means that the carcinogenic risk due to inhalation on site remains negligible.

To summarize, the values obtained for chronic non-carcinogenic and carcinogenic risks prove that the Buterere site presents a danger for the population living in its vicinity. Exposed individuals are at risk of developing chronic non-carcinogenic diseases and cancer, generally linked to the ingestion of soil particles contaminated by heavy metals. Remedial measures are necessary to prevent long-term nuisance for the population concerned.

Prospects for rehabilitating the soil at the Buterere site, contaminated with heavy metals, are being considered and phytostabilization, under study, will be proposed to reduce the spread of this pollution. On the other hand, sorting MSW to recover material is another way of remediation for the future to be proposed to the Bujumbura inhabitants [25]. MSW sorted by category helps to promote recycling of some fractions (paper, glass, metal, etc.) and valorisation for some other fractions (fermentable, biomass waste, etc.) to produce compost or briquettes fuel [26]. Finally, it is important to separate some harmful waste such as batteries, medicines, etc. and avoid to dump mixed contaminated waste on uncontrolled landfills that generate nuisances presented in this work.

4 Conclusion and Perspectives

The study shows that the site, in addition to the current soil composition, is contaminated by heavy metal (Cd, Cr, Cu, Fe, Fe, Mn, Ni, Pb and Zn), at levels that generally exceed the limit values. The levels obtained experimentally show that this site poses an environmental risk to biodiversity in general and to the health safety of the people who live there specifically.

The health risk assessment showed that children exposed to this site are at risk to develop chronic diseases, mainly due to the involuntary ingestion of soil particles. The possibility of developing cancer pathologies has also been proven for children and adults exposed during their lifetime to the Buterere site. Nevertheless, the assessment of the health risks linked to exposure through the skin and inhalation has shown that they are negligible. Remediation is necessary to prevent long-term nuisance for the population. Phytostabilisation is proposed to be used for contaminated soils by heavy metals (under study) and sorting MSW as a preventive solution to avoid dumping mixed contaminated waste on uncontrolled landfills.

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Conflict of interest

The authors state there is no conflict of interest.

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