

Assessment of the Intensity of Soil Pollution of Open-Air landfills by Heavy Metals (Cr, Hg, Pb and Zn) in the City of Brazzaville, Republic of Congo

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Abstract—In the Republic of Congo, solid waste from households and small industries of all kinds is dumped without prior treatment in public landfills, often in the open. The soil in these landfills is not protected and is therefore at risk of contamination by heavy metals. As a result, the surface water and groundwater in the surrounding area are also exposed to the same risks through various phenomena, since the storage of solid waste is the cause of large quantities of heavy metals being buried in the soil. Against this background, this study was carried out to assess the environmental impact of waste deposited in open dumps in the city of Brazzaville. Soil samples were taken from open dumps at four sites (Kinsoundi, Diata, Moukondo and Tsiémé). Physicochemical analyses such as pH, water content (TE), Organic Matter content (MO) and heavy metal content were carried out on soil samples from landfills at these four sites. Soil organic matter content was determined using the Walkley and Black method, while heavy metal content was measured using ICP-OES. The results showed that the investigated landfill soils are slightly alkaline (6.93–7.29) with low water content (12.53–18%) and rich in organic matter (1.38–5.45%). The soils also contain high levels of heavy metals: Cr (1724.67–6664.64 mg/kg), Hg (0.78–1.53 mg/kg), Pb (120–173.33 mg/kg) and Zn (920–5500 mg/kg). The values of the geo-accumulation index (1.72–5.63) and the pollution index (5.10–16.40) show a significant contamination of these soils by the above-mentioned heavy metals. Statistical processing using the correlation matrix was used to establish a series of relationships between the different parameters studied.

Keywords—heavy metals, contamination, pollution, landfills, environment, geo-accumulation index

I. INTRODUCTION

The protection of soil is of paramount importance for the maintenance of environmental, ecological and agricultural resources, and thus for the achievement of sustainable development [1]. In addition to its role as the medium on which plants grow, soil provides a habitat for animals and other micro-organisms [2]. The combination of high population growth in developing countries and challenging economic circumstances is driving uncontrolled urbanization [3].

This demographic expansion is occurring concurrently with the generation of considerable quantities of household and industrial waste, which is frequently discharged without

undergoing any form of preliminary treatment. This has resulted in the contamination of soil with metallic substances several studies have been conducted on soil contamination by heavy metals due to their non-biodegradability, toxicity, persistence and prevalence. For instance, Leopold Ekengele Nga and colleagues have demonstrated that all the heavy metals identified at the car tyre incineration site in the town of Ngaoundéré are closely associated with the constituents of the tyres. These metals were found to be concentrated in the first twenty centimeters of the surface [3]. The findings of studies conducted by Isabelle Vranken indicate that the copper refinery established in Lubumbashi at the beginning of the 20th century exerted considerable pressure on the surrounding environment, resulting in soil contamination and the destruction of vegetation [4]. These changes have had a significant impact on living conditions, human health and the health of local ecosystems.

The transfer of heavy metals into the subsoil depends not only on their physicochemical characteristics, but also on the properties of the underground environment [5]. The soil is a complex matrix in which the activity of the metal ion depends on its chemical speciation. Parameters such as organic matter content and pH are decisive in the chemical processes of precipitation, sorption and complexation of heavy metals in soils [5–7]. Indeed, several heavy metals can be very dangerous for human health and for other living beings when present in the environment at high concentrations [8, 9]. The aim of this study is therefore to present the impact of heavy metal contamination of open-air landfill soils. For this, four open-air landfills in Brazzaville were chosen. Some landfills concerned by this study are located in the watersheds of the rivers which cross the city of Brazzaville. Slopes can cause rainwater to move and encourage its penetration into waste, then contribute to the formation of leachate which can infiltrate and contaminate surface and groundwater.

II. MATERIALS AND METHODS

A. Location and Description of Landfill Sites

Brazzaville, the capital of the Republic of Congo, is situated between latitudes 4°6' and 4°23' South and

longitudes 15°5' and 15°25' East [10]. The city's climate is characterised by two distinct seasons: a rainy season from October to May, with a slight decrease in precipitation from January to February, and a dry season from June to September [11]. Brazzaville is mainly watered by numerous rivers, all of which flow into the Congo River to the south and east. These are the Djiri and Kélékélé in the North; Tsiémé, Mfoa and Madukutsékélé in the center; the Djoué and the Mfilou to the West. The soil types found in Brazzaville are podzols or podzolized soils; hydromorphic soils and ferrallitic soils. In general, the soils of Brazzaville are characterized by a sandy texture [12, 13].

B. Characteristics of Landfills and Typology of Waste in the Four Landfills

1) Kinsoundi landfills

The Kinsoundi landfill is located in district n° 1, Makélékélé, with a latitude and longitude of 04.28635° and 15.21398° respectively, and an area of approximately 2693.373 m², with ferrallitic soil type. This landfill is located in the watershed of the Djoué River. The main waste that constitutes this landfill is broken glass, tin cans, plastics, clothing waste, cardboard, and electronic devices.

2) Diata landfill

Diata Landfill is located in district n°1, Makélékélé, with an area of approximately 1678.46 m², latitude and longitude respectively: 04.27211° and 015.24971° and podzolized soil. The waste that constitutes this landfill is essentially: plant matter, cardboard, plastic bags and bottles, cans, pieces of sheet metal, batteries, electronic devices and clothing waste.

3) Moukondo landfill

With an area of approximately 4,295,794 m² and geographical coordinates: latitude (04.22756) and longitude (015.26872°), the Moukondo landfill is located in district no. 4 Mougali, in the watershed of the Tsiémé River, characterized by podzolized soil type. The waste that constitutes the Moukondo landfill is that coming from the market and the carcasses of car plates.

4) Tsiémé landfill

With an area of 1,398.936 m², latitude (04.22994°) and longitude (015.28378°), this landfill is located in district n° 6 Talangaï, in the watershed of the Tsiémé River and it has a soil amorphous. Waste from the Tsiémé landfill consists only of household waste such as: electronic devices, plastics, broken glass, sheet metal from car wrecks, etc.

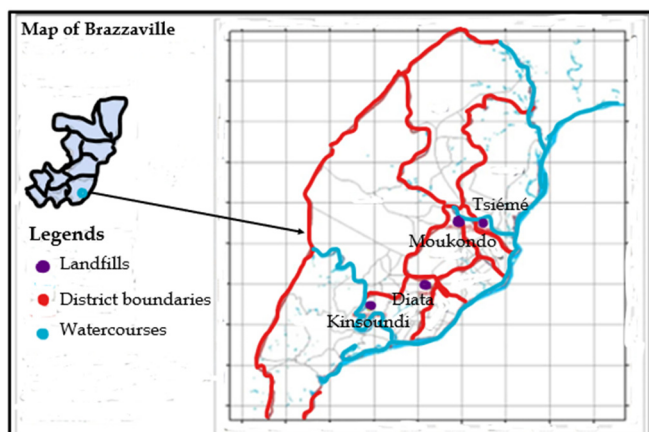


Fig. 1. City map and landfill sites.



Fig. 2. Photos of the four landfills studied: Kinsoundi landfill (a), Diata landfill (b), Moukondo landfill (c) and Tsiémé landfill (d).

Figs. 1 and 2 respectively represent the map of the city (landfill sites in yellow dots) and photos of four landfills.

C. Collection and Processing of Samples

In January, June and November of 2022, three soil sampling campaigns (P1, P2 and P3) were conducted at the landfill sites. An auger was used to extract five individual samples from each landfill site at a depth of 25 cm. The samples were placed in clean plastic bags and sent to the laboratory for analysis. In order to obtain a composite sample for each landfill, 2 kg of each individual sample was mixed and then dried for a period of ten days at room temperature. Following the drying process, the samples were subjected to a 2 mm mesh sieving procedure and subsequently ground using an agate mortar.

D. Measurement of Physicochemical Parameters

To ascertain the water content (TE) of the soil samples, 20 g of each soil sample was weighed with precision in a crucible and then placed in an oven at 105 °C for 24 hours. Following a period of cooling, the samples were weighed again. The water content was calculated using the formula in Eq. (1).

$$TE (\%) = (M1 - M2)/M1.100 = Mw/M1 \times 100 \quad (1)$$

With $M1$: mass of the sample before entering the oven; $M2$: mass of the sample at the exit from the oven; Mw : mass of water in the sample.

The pH was measured in the supernatant of a soil solution consisting of 50 g soil and 50 ml distilled water using a multiparameter. The organic matter content was determined according to Walkley *et al.*, 1934. This method is based on the oxidation of organic carbon by an excess of potassium dichromate ($K_2Cr_2O_7$) in an acid medium, followed by the determination of this excess with Mohr's salt [10].

E. Determination of Heavy Metal Concentrations

Prior to the determination of heavy metal concentrations, the soil samples were subjected to mineralisation by the action of aqua regia. This was achieved by dissolving 0.5 g of the composite soil sample in a solution containing 9 mL hydrochloric acid (HCl) and 3 mL nitric acid (HNO_3), at 95 °C for 75 mins on a heating block. The solution is increased to 50 mL with distilled water [11].

Concentrations of dissolved heavy metals in the soil samples were measured using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES).

F. Assessment of Soil Contamination by Heavy Metals

1) Geo-Accumulation Index (Igeo)

The geo-accumulation index is employed for the assessment of the intensity of metal pollution [12]. This empirical index compares a given concentration with a value considered to be a geochemical background [13]. The geo-accumulation index is calculated using the formula in Eq. (2) [12].

$$I_{geo} = C_n / (1.5 \times B_n) \quad (2)$$

where I_{geo} = Geo-accumulation Index; n = element considered; C_n = concentration measured in the sample; B_n = geochemical background.

Müller [12] defined a scale of values according to the intensity of the pollution, which specifies the following: $I_{geo} < 0$ (class 0), contaminated soil; 0~1, slightly contaminated soil; 1~2, moderately contaminated soil; 2~3, moderately to heavily contaminated soil; 3~4, heavily contaminated soil; 4~5, heavily to extremely contaminated soil; and $I_{geo} > 5$, extremely contaminated soil [14].

2) Pollution index

The pollution index provides an overall indication of the pollution levels at each study site. In fact, it is the average of the ratios of heavy metal concentrations in the soil samples in relation to the threshold limits of the heavy metal standard. In this study, four heavy metals and their respective limits set by the AFNOR NF U44-041 standard were considered. The pollution index was calculated using the formula presented in Eq. (3) [15].

$$IP = \frac{\frac{[Pb]}{100} + \frac{[Cr]}{150} + \frac{[Zn]}{300} + \frac{[Hg]}{1}}{4}} \quad (3)$$

A pollution index exceeding one (1) indicates the presence of multiple metals in the soil [16].

III. RESULTS AND DISCUSSION

A. Physico-chemical Parameters

1) Water content

The variation in water contents recorded in landfill soils over the three seasons is not uniform. Average values range from a minimum of 9.2% to a maximum of 18%. Fig. 3, which shows these variations indicates that the highest water content values are recorded during the third sampling campaign (P3) while the lowest values are recorded in the first and second sampling campaigns (P1 and P2). This observation can be explained by the fact that the third campaign was carried out during the rainy season when the soils are wetter.

2) Hydrogen Potential (pH)

The average pH values recorded in the soil samples indicate that these soils are slightly basic (Fig. 4). Indeed, the average pH of landfill soils is all between 7.0 and 7.5; which could indicate that these soils are less rich in carbonate. Le tacon F. [17] showed that at atmospheric pressure, the pH of soil solutions increases with the presence of an excess of

carbonate. Increasing pH can induce the formation of species which can precipitate and limit the solubility, and the bioavailability of all ionic species in the soil. The results on the average pH values obtained in the landfill soils studied are similar to those reported by Chaer *et al.* [18] for soil from the Tangier wild landfill in Morocco. However, they differ from the pH values founded by Baghdadi *et al.* [19] in 2015 for soil solutions from the landfill in the town of Béni-Mellal in Morocco. The pH results obtained in our study at the different sites are acceptable and meet the standards.

3) Soil organic matter content

The organic matter contents obtained in landfill soils are not homogeneous, they vary from one landfill to another, and from one sampling campaign to another. Average organic matter values range from a minimum of 1.38% to a maximum of 5.45% (Fig. 5). These soils have a relatively high average content of organic matter in comparison with the organic matter contents observed by Ogouvidé [20] during a study on the soils of the household waste landfill in Lomé in Togo. The low organic matter values of Kinsoundi ($P1 = 0.13$ and $P3 = 0.15$), Diata and Moukondo ($P1 = 0.13$ and $P2 = 0.134$) can be justified by the presence of waste that is difficult to degrade, such as plastics.

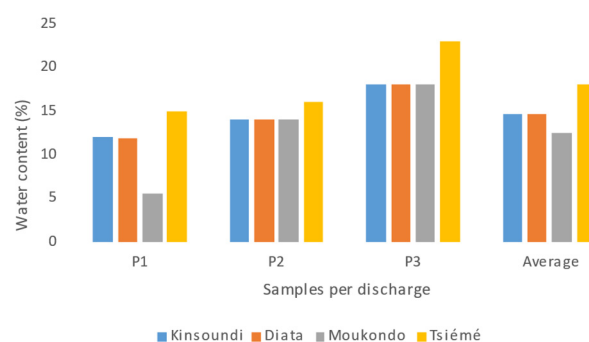


Fig. 3. Water content.

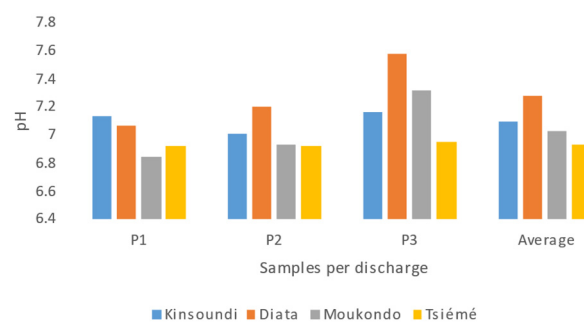


Fig. 4. Hydrogen Potential (pH).

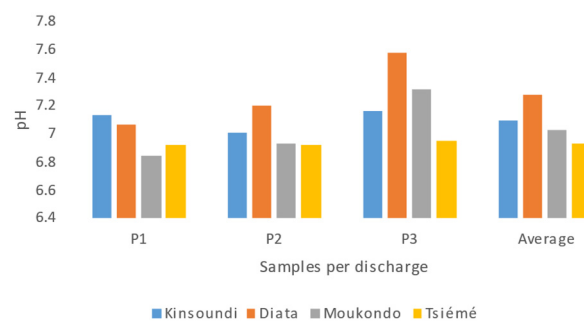


Fig. 5. Organic Matter content (OM).

B. Concentrations of Heavy Metals

1) Chromium concentration

The results obtained on the chromium concentration in the soils of these landfills are presented in the graphs of Fig. 6. These results show that the average concentration of chromium in the soils of these landfills is higher than the standard which is set at 150 mg/kg. These concentrations are not homogeneous in the soils of these landfills; this may be due to the heterogeneous nature of the waste. The average chromium concentrations in these soils vary as follows: 1724.67 mg/kg in the soil of the Kinsoundi landfill, 6664.64 mg/kg in the soil of the Diata landfill, 4036.33 mg/kg in the soil of the Moukondo landfill and 6477.67 mg/kg in the soil of the Tsiémé landfill.

These chromium concentrations obtained in the soils of these landfills are much higher than those observed in the soils of landfills in France (150 mg/kg), India (127.90 mg/kg) and Canada (64 mg/kg) [18]. They are also significantly higher than the concentration detected in the Akreuch landfill [21]. The high chromium content in targeted landfills can be explained by the presence not only of household waste but also by waste from small industries existing around these landfills and many others such as paper, cardboard and wood [22].

2) Mercury concentration

Fig. 7 shows the variation in average mercury concentrations in soil samples from these landfills. The results show 0.78 mg/kg in the soils of the Kinsoundi landfill, 1.15 mg/kg in the soils of the Diata landfill, 0.78 mg/kg in the soils of the Moukondo landfill and 1.53 mg/kg in the soils of the Tsiémé landfill. The average mercury levels recorded in the soils of the Diata and Tsiémé landfills far exceed the standard recommended by AFNOR NFU44-041 (1 mg/kg). This indicates significant contamination of the soils of these landfills by this metal. The high levels of mercury in these landfills could be explained by the presence of waste containing sources of mercury. Taking into account toxicological reference values for total mercury present in humans [23], it would be possible to observe health effects such as disruption of protein synthesis, damage to nerve centers, etc. among populations living around these landfills. The work carried out by Choe et al. 2003 Testify that soils and sediments can constitute a reservoir of mercury when it adsorbs on iron or manganese oxyhydroxides or even on organic matter present [24]. This reservoir can release mercury into different compartments of the environment for long periods of time [25].

3) Lead concentration

The results obtained are presented in Fig. 8 and indicate that the average values of lead concentrations in the soils of these landfills vary between 120 mg/kg and 173.33 mg/kg and are higher than the AFNOR NFU44-041 which sets the lead value at 100 mg/kg. The high presence of lead in the soils of these landfill sites indicates that these soils are contaminated and this could be explained by the presence for a very long time of waste such as used (lead-based) batteries and accumulators, car engine drainage products and residues of other used products, metals, and waste from household appliances, particularly television screens, all containing lead [22]. The work carried out by Pacyna and al. shows that

lead is one of the least mobile metallic elements in the soil which can accumulate on the surface; it is not leached too deeply by leaching and undergoes few transformations [23].

Table 1 below shows the comparison between the metal contents (expressed in mg/kg) in the soils of the landfills studied in Brazzaville and those of the landfills of some other countries with the standards in force.

4) Zinc concentration

Fig. 9 shows that the average zinc contents in the soil samples from these landfills vary as follows: 5500 mg/kg in the soils of the Diata landfill, 2076.66 mg/kg in the soils of the Kinsoundi landfill, 2070 mg/kg in the soils of the Tsiémé landfill and 920 mg/kg in the soils of the Moukondo landfill. These average zinc contents obtained in this study are much higher than the AFNOR NF U 44-041 standard which limits this value to 300 mg/kg. Zinc is considered relatively non-toxic, as its toxicity to humans is low [24]. However, at high concentrations, zinc can become toxic [25].

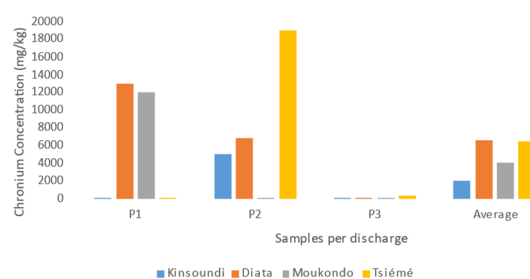


Fig. 6. Concentration of Cr.

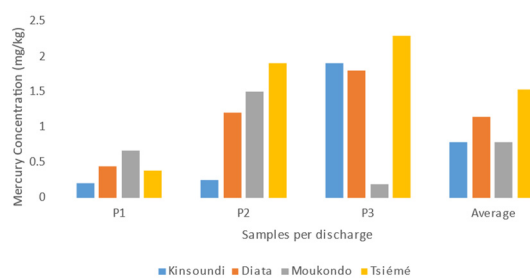


Fig. 7. Hg concentration.

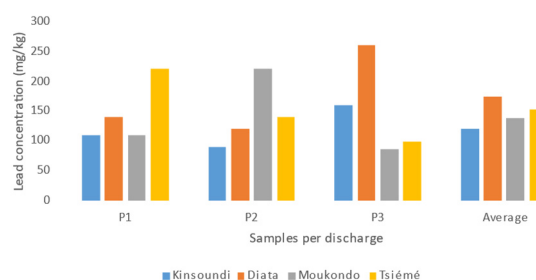


Fig. 8. Concentration of Pb.

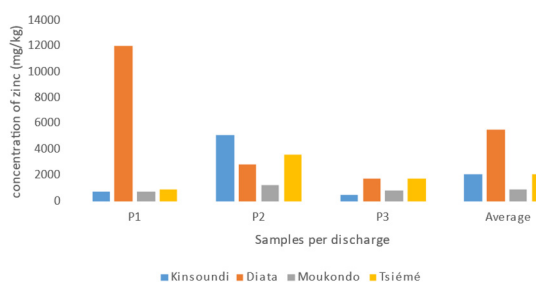


Fig. 9. Zn concentration.

Table 1. Comparison of average heavy metal contents in the soils of some landfills

Landfill names or countries	Cr	Hg	Pb	Zn
Akouébé, Abidjan (Ivory Coast) [26]	73.5	4.82	933.37	1250.55
Ahfir-Saidia (Morocco) [27]	75.74	----	656.56	62.87
India [27]	127.9	----	206.4	122.3
France [27]	150	----	100	300
AlAin (United Arab Emirates) [28]	19.1	----	13.7	117
Kinsoundi, Brazzaville (Congo)	1724.67	0.78	120	546.66
Diata, Brazzaville (Congo)	6664.64	1.15	140	5500
Moukondo, Brazzaville (Congo)	4036.33	0.78	238.67	920
Tsiémé, Brazzaville (Congo)	6477.67	1.53	153	2070
Afnor standard 44-041	150	1	100	300

(expressed in mg/kg)

C. Statistical Processing

Table 2 below presents the correlation matrix between the various parameters of this study.

Table 2. Physicochemical and chemical inter-element correlation matrix of soil samples from the landfills studied

Variables	YOU	pH	MO	Cr	Hg	Pb	Zn
YOU	1						
pH	-0.373	1					
MO	0.610	-0.956	1				
Cr	0.446	0.089	-0.032	1			
Hg	0.897	-0.323	0.822	0.779	1		
Pb	-0.552	-0.379	0.101	0.040	-0.265	1	
Zn	0.129	0.709	-0.620	0.761	0.374	-0.283	1

Principal component analysis applied to soil samples makes it possible to highlight the geochemical mechanisms that govern the vertical migration of heavy metals and to verify the similarity of the source of the metals in landfill soils. The correlation matrix shows correlation values, which measure the degree of linear relationship between each pair of variables. The closer the correlation coefficient between two elements is (+1), the more they tend to be the same, and vice versa. The results then indicate on the one hand a positive relationship between water content (TE) and Organic Matter content (MO), TE and Hg and MO and Hg which means an interdependence between these pairs of variables, and on the other hand a positive correlation between Cr and Hg and between Cr and Zn this could indicate that these metals are all of the same origin which is the anthropogenic origin.

D. Parameters for Assessment of Metal Contamination in Soils

1) Geo-accumulation index

a) Chromium geoaccumulation index: Igeo (Cr)

The average values of the chromium geoaccumulation indices in the soils of these landfills are presented in Fig. 10 and vary as follows: 1.72 in the soil of the Kinsoundi landfill, 5.27 in the soil of the Diata landfill, 2.42 in the soil of the Moukondo landfill and 3.86 in the soil of the Tsiémé landfill. The recorded chromium pollution intensities indicate that the soils of these landfills are characteristic of polluted soils and soils heavily polluted by chromium.

b) Mercury geoaccumulation index: Igeo (Hg)

The average values of the mercury geoaccumulation indices in the soils of these landfills are presented in Table 2 and follow the trend observed in Fig. 11: 2.44 at the Kinsoundi landfill, 3.55 at the Diata landfill, 3.87 at the Moukondo landfill, 3.81 at the Tsiémé landfill. The average

values of the mercury geoaccumulation indices are not uniform in the soils of these landfills and they are all greater than 3. The soils of these landfills are therefore class 4, that is to say heavily polluted by mercury.

c) Lead geo-accumulation index: Igeo (Pb)

Fig. 12 presents the variation in the geoaccumulation index of lead in the soils of the landfills studied. In the soil of the Kinsoundi landfill, the geo-accumulation index of lead is between 3 and 4 while in the soils of the Diata, Moukondo and Tsiémé landfills, the geoaccumulation index is between 2 and 3 (Table 2). These values indicate that these soils are class 2, therefore polluted by lead.

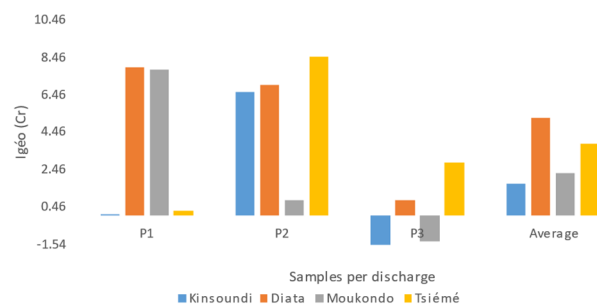


Fig. 10. Cr geo-accumulation index.

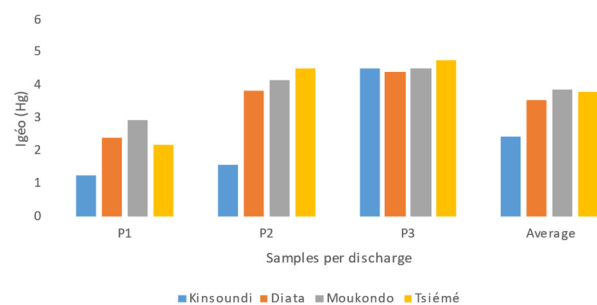


Fig. 11. Hg geo-accumulation index.

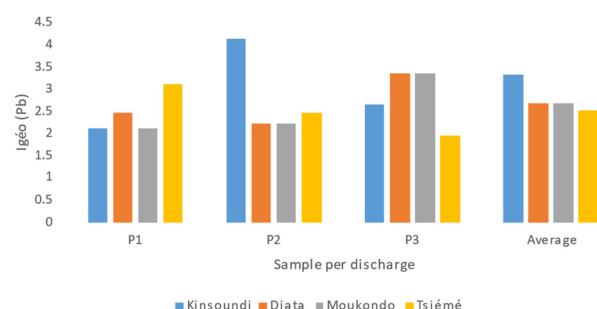


Fig. 12. Pb geo-accumulation index.

d) Zinc geoaccumulation index: Igeo (Zn)

The average values of the geoaccumulation index of zinc in the soils of the landfills studied are presented in Table 2 and follow the trend observed in Fig. 13. In the soil of the Kinsoundi landfill Igeo (Zn) is 3.89, in the soil of the Diata landfill Igeo (Zn) is equal to 5.63, in the soil of the Moukondo landfill Igeo (Zn) is 3.52 and in the soil of the Tsiémé landfill Igeo (Zn) obtained is of 4.5.

These values show that there is a large accumulation of zinc in these soils and give them the characteristics of moderately polluted soils and heavily polluted soils. The high accumulations of zinc in landfills show that the zinc pollution

in the soils of these landfills is of anthropogenic origin.

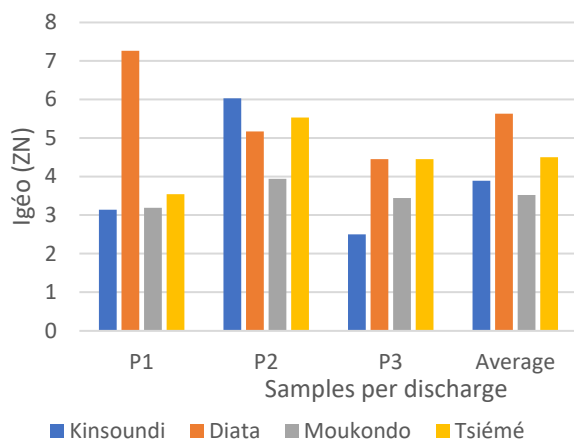


Fig. 13. Zn geo-accumulation index.

2) Heavy metal Pollution Index in landfill soils (PI)

The average values of the pollution index recorded in the soils of these landfills vary from one landfill to another and this variation follows the following trend: 5.10 in the soils of the Kinsoundi landfill, 16.4 in the soils of the Diata landfill, 8.04 in the soils of the Moukondo landfill, and 13.28 in the soils of the Tsiémé landfill (Table 3). The average Pollution Index (PI) values recorded are all above the threshold value (PI = 1), which leads to the conclusion that the soils of all these landfills are polluted by the heavy metals studied.

Table 3. Parameters for evaluating soil contamination by heavy metals

Settings	Samples	Kinsoundi	Diata	Moukondo	Tsiémé
Igeo(Cr)	P1	0.1	7.94	7.84	0.26
	P2	6.60	7.04	0.80	8.50
	P3	-1.54	0.84	-0.39	2.82
	Average	1.72	5.27	2.42	3.86
Igeo(Hg)	P1	1.25	2.39	2.94	2.18
	P2	1.57	3.84	4.16	4.50
	P3	4.50	4.42	4.50	4.77
	Average	2.44	3.55	3.87	3.81
Igeo(Pb)	P1	2.11	2.46	2.11	3.11
	P2	5.14	2.23	2.23	2.46
	P3	2.65	3.35	1.75	1.96
	Average	3.33	2.68	2.03	2.51
Igeo(Zn)	P1	3.14	7.26	3.19	3.54
	P2	6.03	5.17	3.94	5.53
	P3	2.50	4.45	3.44	4.45
	Average	3.89	5.63	3.52	4.50
IP	Average	5.10	16.40	8.04	13.28

IV. CONCLUSION

The aim of this study was to evaluate the intensity of heavy metal pollution Cr, Hg, Pb and Zn in the soils of four open-air landfills in the city of Brazzaville. Soil samples were taken, treated and then analyzed to determine certain physicochemical parameters and the concentrations of these heavy metals in order to assess their contamination. The determination of the physicochemical parameters of landfill soils showed that they are subject to anthropogenic influence. These soils are not very rich in water with values varying between 5.6 and 23%, which can facilitate the vertical migration of rainwater and transport heavy metals deep until reaching the aquifers.

The pH recorded in these soils indicates that they are slightly basic with values between 6.85 and 7.32. The soils of

these landfills are rich in organic matter with contents between 0.15 and 9.9%. The concentrations of all heavy metals determined in these landfills' soils are higher than those of the AFNOR 44-041 standard. The average concentrations are between 1724.7 and 6477.67 mg/kg for chromium, 0.78 and 1.53 mg/kg for mercury, 120 and 173.33 mg/kg for lead and finally 920 and 5500 mg/kg for zinc. The disposal of waste in open landfills without prior treatment is the cause of the accumulation of heavy metals in the soil. Furthermore, the contamination of soil at these landfill sites is a matter of significant concern. The average values for the geo-accumulation index and the pollution index exceed the threshold values of soil unpolluted by heavy metals, indicating a considerable degree of contamination. The present study did not verify the impact of this heavy metal pollution in the rivers near these landfills and on the health of the populations living nearby. Hence a study would be necessary to check the aquatic environment and the health of the populations living around these landfills.

CONFLICT OF INTEREST

The authors declare no conflicts of interest in relation to this work.

AUTHOR CONTRIBUTIONS

E.C.B is the principal investigator of this article and contributed to the collection of samples; K.B.M.L and B.M.J.C contributed to the analysis of the results as well as their interpretation; K.O.A.C and M.N.P participated in the Reading; MT provided general supervision of the work; all authors had approved the final version.

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