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STATISTICAL APPROACH OF ASSESSING HORIZONTAL MOBILITY OF HEAVY METALS IN THE SOIL OF AKOUEDO LANDFILL NEARBY EBRIE LAGOON (ABIDJAN-COTE D'IVOIRE)

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Abstract

The present study aim to quantify heavy metals in the Akouedo landfill soil and evaluate the extent of their contamination, to better understand the horizontal migration of these pollutants towards the Ebrie lagoon located downstream. The Kruskal-Wallis test was used to evaluate the heavy metals concentrations levels according to the upstream downstream disposition of soil sample sites. Then the Mann-Whitney test was applied to find if variances between parameters concentrations in the soil sample sites were homogenous. The results show that the soil is rich in organic matter (organic carbon and organic nitrogen) with alkaline pH (7.94 < pH < 8.7), characteristic of the landfill leachate. Metals such as lead (Pb) and zinc (Zn) are retained by the organic matter while the other metals (Cd, Cr, Cu, Fe) migrate relatively downstream. Thus, the flow of infiltration water into the soil of the Akouedo landfill drains more easily iron, cadmium, copper, chromium towards the lagoon Ebrie located downstream, increasing the risk of pollution by heavy metals.

Keywords: *landfill; heavy metals; soil; horizontal mobility; Ebrie lagoon; Abidjan; Côte d'Ivoire*

Introduction

Landfilling is the most common method for disposing of solid wastes. One of the major problems associated with landfilling is the generation of large quantities of heavily polluted leachate. The contaminants are released from the landfill leachate passing through the soils and water and polluting streams, creeks and water wells with organic and inorganic matters [1-7]. Among the different pollutants occurring in leachates, heavy metals constitute one of the pollutant groups that are kept under surveillance because they can be particularly dangerous when they are able to contaminate drinking water. Their movement through soils is mainly related to the fluid dynamics and to the sorption during the solid phase, the latter being in turn controlled by the composition of both solid and liquid phases [8]. The water resource pollution by heavy metals in the vicinity of landfill is often discussed in the literature [9 -12]. So, an accurate risk assessment on contaminated landfills should consider the risk of heavy

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metal transfer to the groundwater and surface water. That thus creates the need for knowing the pollution state of the landfill soils and especially for following the migration of heavy metals in these soils.

In Côte d'Ivoire, singularly in Abidjan town, the increase of the waste production is correlated with the demographical developpement. This situation is accompanied by an increase of solid waste production with 2500 to 3000 tons/day [13]. The waste generated in the town is dumped in Akouédo landfill which has been in operation for more than 30 years. This one is a non controlled landfill which constitutes a persistent threat for the population and proximity water [14]. There is a study already announced a vertical mobility of the heavy metal from the landfill to the soil [13]. This vertical mobility can cause the contamination of the groundwater. The geological context primarily made up of sand and clay could offer a priori conditions favourable to the migration of the pollutants towards groundwater. Nevertheless, some authors [15] showed that the quality of water from the upstream groundwater collecting field is not yet reached by the pollutants of the landfill and is of good quality for the domestic needs. The soil is generally regarded as a buffering zone that prevents or reduces groundwater and surface water pollution by retardation and dispersion [16, 17]. However, several investigations indicated that the migration of heavy metals in the soil was very low during the first decades after deposition, compared to the accumulated amount [8, 18]. So, the vadose zone can be considered as a source of nearby water pollution since heavy metal seep to unsaturated zone over a long period of time [19]. Therefore, continued attention to heavy metals from the landfill leachate in the soil is necessary. Unfortunately, no study has yet shown whether the leachate from the Akouedo landfill migrates into the soil with heavy metals to the Ebrie lagoon downstream in the direction of water flow.

This study aims to quantify heavy metals in the Akouedo landfill soil and evaluate the extent of their contamination, to better understand the horizontal migration of these pollutants towards the Ebrie lagoon.

Materials and methods

a. Study area

Akouedo landfill is located at the east part of Abidjan, 18 km from the center between 390,000 and 399,000 m East longitude and between 587,000 and 600,000 m North latitude of UTM referential Zone 30 (Fig. 1). It extends on a 153 ha and the area is not prepared before the use for waste disposal. Urban solid residues were simply deposited on the land since 1965 [13]. No proper compaction of solid waste is carried out at the site and the underground drainage system; liner cover system and leachate collection system were found absent. Consequently, the leachate is drained in an anarchistic way under the heaps of garbage. Akouedo landfill receives approximately 550 000 tons of waste per year, of which two thirds are composed of domestic waste and the third remaining of industrial and some dangerous waste [14].

The predominant geology in the area is the sedimentary formations made from up to bottom of clayey sands, medium sands and coarse sands resting on a granitic and schistous base (Fig. 2).

The climate is humid subtropical with two rainy seasons (april-july and october-november) and two dry seasons (august-september and december-march). The rains are important during rainy seasons (150-450 mm per month) and low during the dry seasons (32-100 mm per month). Temperatures are generally located between 22°C and 29°C.

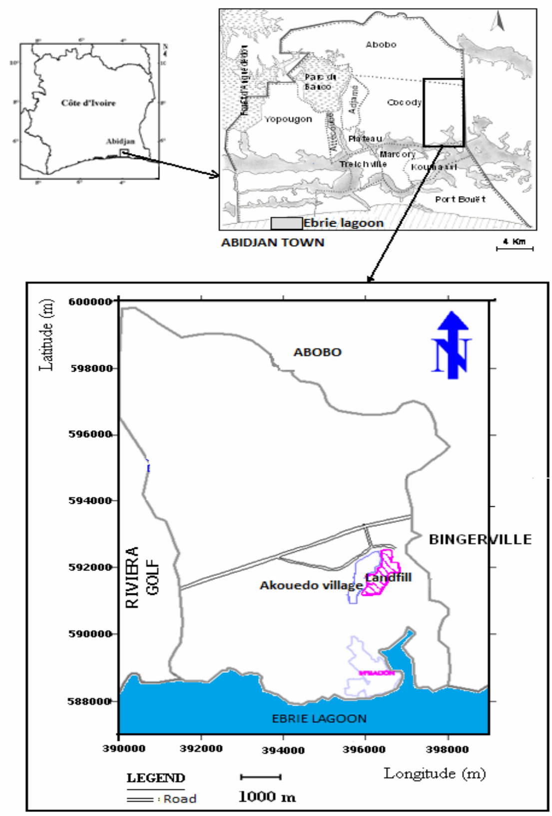


Fig. 1. Location of Akouedo landfill, Abidjan-Cote d'Ivoire

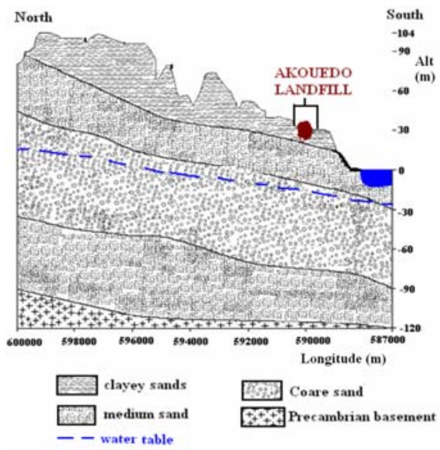


Fig. 2. Geological profile of the Akouedo area

b. Soil samples collection

Soil samples were taken from three stations, in February 2004. The sample site (A) was positioned on the landfill. The flow of the leachate being made North towards the South, two other sample sites (B and C) were positioned downstream of the landfill. The site B is to 150 m of the landfill and the site C to approximately 350 m downstream (Fig. 3).

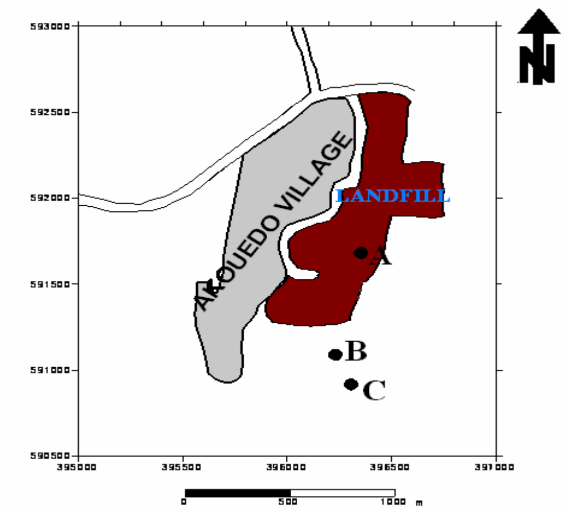


Fig. 3. Location of the soil sampling sites: A, B and C.

The collection of samples was done with an auger. This material made it possible to sample up to 5 m of depth. The procedure consists in putting pressure on the auger by rotation in the direction of clockwise. Tool gradually sinking into the soil column and recovered sample is collected in a labeled bag. The tools make samples every 20 cm in the soil column. On the landfill area, sampling stopped with 1.2 m for profile A because of solid waste which prevented the tool from being inserted beyond these depths. As for the two profiles located downstream from the landfill, samplings reached the maximum depths of 5m. A total of 56 samples of soil were collected.

c. Analytical methods

Heavy metals of the soil samples were extracted using an ammonium acetate solution 1N with pH 4.8 with a 1:5 soil/solution ratio. Analysis sample was composed of 10 g of fine grained soil(2 mm), weighed and put in an Erlenmeyer with 50 ml of extraction solution acetate ammonium-acid acetic. The mixture was mechanically agitated during 30 min and filtrated in a dry flask on a washed filter with the acetic acid. Total concentrations of Zn, Pb, Cu, Fe, Cr and Cd were measured by the Atomic Absorption Spectrophotometer Varian AA 200 after calibration with standards prepared in the acetate ammonium solution. Organic matter content and pH of the soil sample were also determined. The organic carbon analysis was performed by the method of Walkley and Black [20] based on the oxidation of organic carbon by potassium dichromate ($K_2Cr_2O_7$) in acid medium. The excess potassium dichromate was used to determine the amount of organic carbon. The NTK (Kjeldahl nitrogen) was determined by the Kjeldhal method. In this method, the nitrogenous organic matter is mineralized by 98% concentrated sulphuric acid (d = 1.84), hot. The carbon and hydrogen are released to the state of dioxide carbon and water. The nitrogen transformed into ammonium is fixed by the sulphuric acid as sulphate of ammonium.

The active acidity (pH_w) and reserve acidity (pH_{KCl}) of the soil were measured respectively from deionized water and KCl solution with a 1:2.5 soil/solution ratio after equilibration for 24 hours [20].

d. Statistical analysis

Data were analyzed using NCSS.6 software. Horizontal mobility of heavy metals in soil was searched with the Kruskal-Wallis test [21] which was used to compare the concentrations of heavy metals according to the upstream downstream disposition of soil profiles. Then the Mann-Whitney test [22, 23] was applied to find if variances between upgradient and downgradient parameters concentrations were homogenous. A probability of $p < 0.05$ was used to determine the statistical significance in all analyses.

Results and Discussion

Physical and chemical characterization of the landfill soil

The physical and chemical characterization of the landfill soil in the collected samples from the three sample sites (A,B,C) is presented in Table1. The organic matter represented by the organic carbon (CO) and organic nitrogen (NTK) is very important at all three sample sites. However, the concentrations are higher on the landfill (sample site A) than the two other sample sites (sample sites B and C). Also, the soil of Akouedo landfill is a reducer medium with a pH (pH_w and pH_{KCl}) ranging generally between 7.24 and 8.70. This abundance of organic matter is the consequence of the presence at the landfill of biodegradable domestic solid waste and of a high microbiological activity for the mineralization of nitrogen and carbon forms. According to some authors [24], high values of pH are favorable for mesophilic microorganisms colony activity that grow at temperatures between 20°C and 40°C and degrade the organic matter.

Heavy metals (Cu, Fe, Zn, Pb, Cd, Cr) have a heterogeneous spatial distribution. The average concentrations are more important at site A and decreased gradually from site A to site C. Indeed, the presence of old lead paint, tires, batteries, plastics, rubber, are the basis of the high concentration of heavy metals in soils [13]. However, certain metals such as iron have a maximum concentration higher at site B (12950 ppm) compared to the site A (10750 ppm) whose average concentration was calculated on a small number of samples. So, there is heterogeneous distribution of concentration for each parameter around its average concentration. This phenomenon is supported by standard deviation and coefficient of variation (between 27 and 149%), high (>15%) for most studied parameters (Cu, Fe, Zn, Pb, Cd, Cr, CO, NTK).

Table 1. Physical and chemical characteristics of Akouedo soil sample sites

Parameters	Average	Min	Max	SD	CV (%)
Sample site A					
Cu (ppm)	132.38	43.10	351.40	125.42	94.74
Fe (ppm)	5626.67	850.00	10750.00	4696.42	83.47
Zn (ppm)	534.53	53.90	1143.22	408.67	76.45
Pb (ppm)	426.30	24.70	1450.00	583.81	136.95
Cd (ppm)	3.40	1.00	7.10	2.36	69.38
Cr (ppm)	71.82	63.50	88.90	10.10	14.07
CO (ppm)	9441.51	2377.36	34641.51	14107.94	149.42
NTK(ppm)	10752.00	3920.00	28000.00	9902.66	92.10
pHw	8.70	8.00	9.30	0.49	5.69
pH_{KCl}	8.14	7.10	8.60	0.59	7.30
Sample site B					
Cu (ppm)	72.32	48.80	114.40	17.99	24.88
Fe (ppm)	2741.33	1266.67	12950.00	2274.38	82.97
Zn (ppm)	199.00	18.60	650.90	150.40	75.58
Pb (ppm)	96.68	18.40	293.60	67.51	69.83
Cd (ppm)	7.28	3.10	9.70	1.61	22.08
Cr (ppm)	85.49	46.20	119.60	17.83	20.85
CO (ppm)	6004.53	3226.42	12226.42	2190.54	36.48
NTK(ppm)	5174.40	3360.00	11200.00	1735.70	33.54
pHw	7.96	7.30	9.00	0.44	5.53
pH_{KCl}	7.24	6.50	8.40	0.49	6.74
Sample site C					
Cu (ppm)	46.98	37.20	64.00	6.20	13.20
Fe (ppm)	2060.67	1050.00	5750.00	1038.35	50.39
Zn (ppm)	200.77	47.50	845.27	179.89	89.60
Pb (ppm)	48.32	10.30	140.50	32.91	68.11
Cd (ppm)	3.76	0.70	8.90	2.22	59.11
Cr (ppm)	57.37	27.70	87.80	16.01	27.91
CO (ppm)	4550.94	1018.87	14773.58	2776.47	61.01
NTK(ppm)	4435.20	3360.00	8400.00	1198.02	27.01
pHw	7.94	7.30	8.70	0.33	4.11
pH_{KCl}	6.88	6.30	7.90	0.38	5.57

SD - standard deviation, CV (%) - coefficient of variation, CO – organic carbon,
NTK – total Nitrogen determined by Kjeldhal method,
pH_w – pH of soil matrix water, pH_{KCl} - pH of soil matrix

b. Horizontal mobility of heavy metals

The mobility of heavy metals is analyzed from a distribution of heavy metals at the sample sites and the relationship between these sites (Fig. 4 and Table 2).

There is heterogeneous distribution of copper at three sample sites (Fig. 4a). The upstream site located at the landfill (site A) has 50% of samples with a copper concentration between 55 ppm and 350 ppm. However, the two other sites (sample sites A and B) have a distribution of concentrations less spread with lower medians. The Kruskal-Wallis test shows that there is a significant difference ($p = 1055\ e^{-7}$) between the three sample sites. The Mann-Whitney test applied to sites (Table 2) confirms this significant difference between sites A and C ($p = 0.008043$) and between sites B and C ($p = 1.83e^{-8}$). But, this test shows also that there is no significant difference ($p= 0.1614$) between sites A and B. This similarity between the two sites could be attributed to horizontal mobility of copper from site A to B.

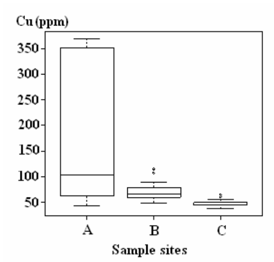
For iron, the median concentration is 5500 ppm. This concentration is greater than those of sites A (median = 2000 ppm) and B (median = 1780 ppm). By Kruskal Wallis test, there is a significant difference ($p = 0.03756$) between the three sample sites (Fig. 4b). However, the Mann-Whitney test allows establishing direct relationship between iron concentration of site A and site B because it seen relatively no significant difference ($p = 0.1769$) between the two sites. Iron can move from site A to B.

It's also noted that 50 percent of samples have a concentration between 300 ppm and 1150 with a median of 600 ppm for zinc (Fig. 4c). These concentrations are higher than those obtained at sites B and C with respective values of 148 and 125 ppm. Despite this difference in concentrations, the three sampling sites were not statistically different ($p = 0.06162$) according to the Kruskal-Wallis test. So the three sites are similar regarding Zinc concentrations. However, the Mann-Whitney test applied to these three sites reveals that the difference is not significant ($p = 0.6862$) only between sites B and C since the difference is significant between sites A and B ($p = 0.02679$). It is clear from this analysis that the site A has a different behavior than the two other sites. So Zn does not migrate from site A to the other two sites downstream. This result confirms those of somes authors [13] who found that zinc was not very mobile because it is strongly adsorbed by organic matter.

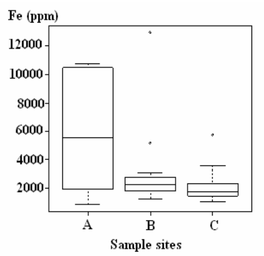
The Pb concentration is heterogeneous at the site A. Median concentration of this site is 280 ppm and 25 percent of these concentrations are between 280 and 980 ppm. These concentrations are very high compared to the other two sites. These sites are significantly different ($p = 0.0001436$) according to the Kruskal-Wallis test (Fig. 4d). Thus, there's no exchange of lead between the sample sites. The Mann-Whitney test confirms this hypothesis with the significant differences observed between sites A and B ($p = 0.03548$), between sites B and C ($p = 0.0002187$) and, between sites A and C ($p = 0.007304$). So, Pb is almost immobile on the different sites. Indeed, the authors [13, 24] have shown that this metal is strongly retained on organic matter, a situation similar to the zinc one.

For Cd, the average concentration is higher (C = 7.5 ppm) at site B and it is law and almost identical (C = 3 ppm) at sites A and C (Fig. 4e). According to the Kruskal-Wallis test, there is a significant difference ($p = 3.99e^{-5}$) between all the sampling sites. It is therefore impossible to establish a relationship between these sites on the migration of Cd. However, the Mann-Whitney test is more accurate since it pointed out that the difference was not significant between sites A and B ($p = 0.06066$) and, A and C ($p = 0.8219$). The similarity between sites A and C can then be attributed to the level of concentration of Cd in the two sites, as they have the same median. However, the similarity between sites A and B can highlight the cadmium migration from site A to site B. This migration may be a cause of increased concentration of cadmium observed at site B located downstream of site A.

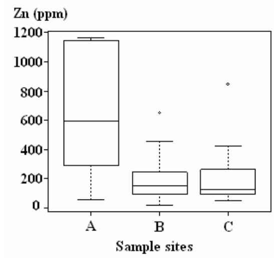
Distribution of chromium at the sampling sites is broadly similar to that of cadmium (Fig. 4f). Median concentration (C = 85ppm) at site B is higher than those found in A (C = 56 ppm) and B (C = 70 ppm). The Kruskal-Wallis test makes no relationship between the concentrations of this metal at three sample sites ($p < 0.05$). But the Mann-Whitney test indicates homogeneity ($p = 0.06415$) in the distribution of Cr between site A and B. These results highlight a mobility of Cr from upstream (Site A) to downstream (Site B). The high mobility of chromium has also been observed on the same landfill during a study on the vertical migration of heavy metals in the soil [13].



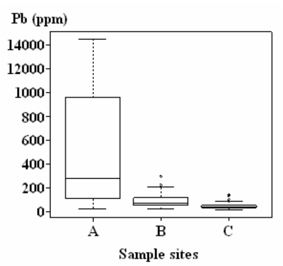
a – Cooper (p-value = $1.055e^{-07}$)



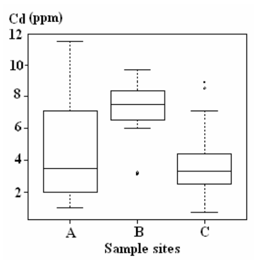
b – Iron (p-value = 0.03756)



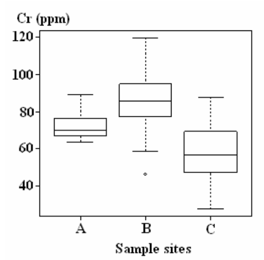
c – Zinc (p-value = 0.06162)



d – Lead (p-value = 0.0001436)



e – Cadmium (p-value = $3.995e^{-05}$)



f – Chromium (p-value = $5.526e^{-06}$)

Fig. 4. Distribution of heavy metals at sample sites according to Kruskal Wallis test ($p<0.05$)

Table 2. Statistically correlated sample sites compared to the concentrations of heavy metals (Mann-Whitney test, $p < 0.05$)

Parameters	Samples sites	B	C
Cu	**A**	0.1614	**0.00804***
	B	-	**$1.830e^{-08}$***
Fe	**A**	0.1769	**0.04283***
	B	-	0.05468
Zn	**A**	**0.02679***	**0.02679***
	B	-	0.6862
Pb	**A**	**0.03548***	**0.007304***
	B	-	**0.0002187***
Cd	**A**	0.06066	0.8219
	B	-	**5.846e-^{06}***
Cr	**A**	0.06415	**0.03547***
	B	-	**2.172e-^{06}***

** significant difference*

c. Influence of environmental parameters on the heavy metals horizontal mobility

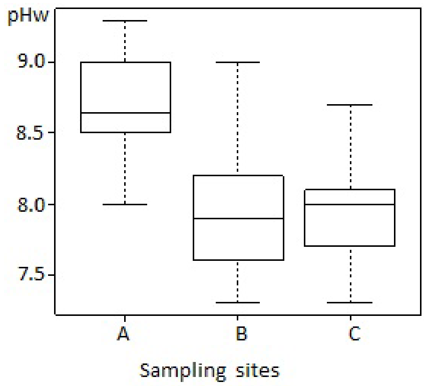
The Kruskal Wallis and Mann Whitney tests with a significant level of $p < 0.05$, allowed to highlight the mechanisms that govern the horizontal migration of these metals (Fig. 5 and Table 3).

pH_W and pH_{KCl} have a similar evolution (Figure 5a and 5b). Indeed, it was shown that the pHw is always above of 0.2 to 1.5 units, as appropriate, to that of pH_{KCl} [25]. The findings indicate that pHs are more alkaline at the site of the landfill (site A) with respective medians of 8.7 and 8.4. However, the medians are lower at site B (pH_W = 7.8; pH_{KCl} = 7.2) and site C (pH_W = 8.0; pH_{KCl} = 7.8). The landfill under these pH, produces alkaline leachates that contaminate the soil below [13]. Kruskal Wallis and Mann Whitney tests with significant levels of $p < 0.05$, allows to note that, generally all the sites are significantly different at the pH_W and pH_{KCl}.

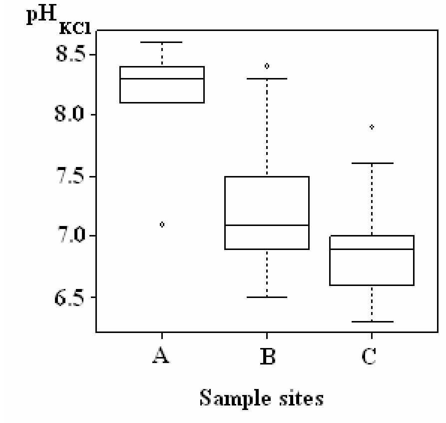
For organic matter, concentration is high on site A with a very heterogeneous distribution. Thus, although the organic carbon reaches values of 4000 ppm (figure 5c), the median of the distribution (C = 2800 ppm) is less than those of site B (C = 5400 ppm) and site C (C = 3700 ppm). This heterogeneous distribution is observed even for organic nitrogen (figure 5d). The heterogeneous distribution is the fact that the wastes are compacted and covered with layers of soil. So, the part of the soil in contact with the waste has high concentrations of organic matter and the other one in surface has low concentrations [13]. Findings of Mann-Whitney test also confirm the heterogenity of organic matter on the site of the landfill and around because it seems difficult to establish a logical in its spatial distribution. However, it must be mentioned that organic matter is abundant because of the presence of a significant amount of biodegradable waste in the landfill.

Heavy metals are mostly very dangerous when contaminate groundwater or surface water. In Akouedo landfill soil, it is possible to establish a direct relationship between immobility of Pb and Zn and their strong affinity with organic matter. Indeed, some authors [1, 26] have reported that heavy metals have always tended to accumulate in surface horizons rich in organic matter in acid medium (pH < 6). Carboxylic functional groups (COOH), phenolic and/or alcohol (OH), carbonyl (C = O) and amino (NH_2) are heavily involved in the complexation of metal cations [27]. The pH variation is one that easily modifies the behavior of metals. Thus, the pH of the soil and its solution has a very strong influence on the soil's capacity to retain pollutants but also to release them. The pH_{KCl} seems to have a well strong influence on the metals compared to pH_w. Therefore, the pH_{KCl} must be regarded as the "pH" of the soil as it takes into account all its physical and chemical characteristics [21]. But similarly, mobile metals (Cu, Cd, Fe, Cr) were observed between the upstream and downstream sample sites despite the alkaline pH which should favor adsorption of the metals. The same observation was done on the Londrina landfill in Brazil where mobile fractions of Co, Mn and Zn were observed in alkaline pH medium [1]. According to some authors [1, 15, 28] this behavior of metals is due to the influence of leachates from landfill that migrates into the soil according to the slope of the area and degree of soil saturation in some metals.

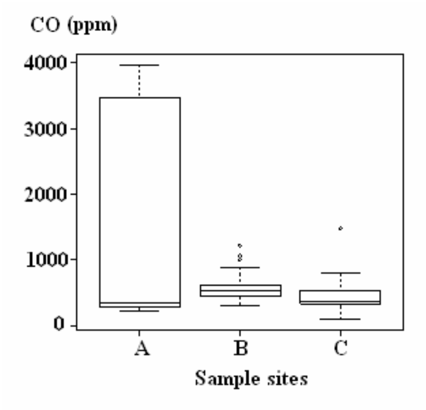
Therefore, the findings obtained on the soil of Akouedo landfill indicate that environmental parameters and the evolution of the pollution plume can affect the horizontal mobility of heavy metals.



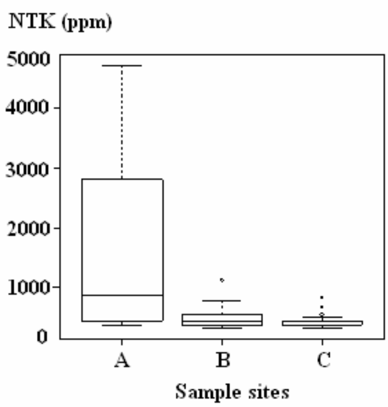
a-pH$_w$ (p-value = 0.0089)



b-pH$_{KCl}$ (p-value = 0.0001337)



c-organic carbon (p-value = 0.03098)



d-Kjeldahl nitrogen (p-value = 0.01054)

Fig. 5. Distribution of environmental parameters at sample sites according to Kruskal Wallis test ($p < 0.05$)

Table 3. Significance level of environmental parameters of sample sites (Mann-Whitney test, $p < 0.05$)

Parameters	Sample sites	B	C
pH$_w$	**A**	**0.005804***	**0.002361***
	B	-	0.9223
pH$_{KCl}$	**A**	**0.004973***	**0.0003861***
	B	-	**0.006623***
CO	**A**	0.2925	0.8409
	B	-	**0.007708***
NTK	**A**	**0.04571***	**0.009278***
	B	-	0.06007

**significant difference*

Conclusion

This study showed how heavy metals from the soil of the Akouedo landfill could migrate horizontally and pollute the waters of the Ebrie lagoon located downstream. Three sites were positioned in the same direction as the flow of water and soil samples were collected at depths of 1.2 m (site A), 5 m (site B) and 5 m (site C). The findings of physical and chemical analysis showed that heavy metals and organic matter are very concentrated in the soil whose pH is alkaline. Statistical tests of Kruskal Wallis and Mann Whitney realized with the significant level of 0.05 have revealed the heterogenity of the distribution of heavy metals at each sample site, the adsorption of Pb and Zn on the soil rich in organic matter and horizontal mobility of other metals (Cd, Cr, Cu and Fe). Mobility of these metals found at site B was located 150 m from the landfill. Although these metals have a slow migration, they threaten the Ebrie lagoon located downstream.

Therefore, further studies, including analysis of soil in closer profiles are necessary to elucidate the extent and mechanism of metal transport.

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