



Heavy Metals Leaching of Cementitious Matrices Containing Waste Ash from Brazzaville Landfills (Republic of Congo)

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Authors' contributions

This work was carried out in close collaboration with all authors. Author KMM defined the subject and wrote the first draft of the manuscript. Author MBML performed the analysis. Author JMO wrote the protocol and supervised all the study. All authors read and approved the final manuscript.

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ABSTRACT

Inadequate management of ash generated by household waste burned in open air in Republic of Congo, generates pollution sites especially through heavy metal contamination (Pb, Ni, Cr, Cu, Zn). The aim of this study is to evaluate the chemical durability of cementitious matrices containing waste ashes and their ability to retain in their structure heavy metals contained in these ashes. To do this, we collected 40 kg of waste coming from 4 landfills (A, B, C, D) in Brazzaville city. Waste was then burned and turned into ash. 6 cementitious matrix formulations (P1, P2, P3, P4, P5, P6) based on Dolisie Portland cement were made by varying the cement/ash/lime ratios. We carried out static leaching tests with raw ashes and cementitious matrices at pH = 7 and at 25°C in distilled water during 30 days. This study shows that cementitious matrices containing ash have good potentiality for retention of heavy metals (more than 75%). But, cementitious matrices containing ash and lime give better results with retention rates between 90-99%.

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1. INTRODUCTION

In many developing countries, solid and liquid waste management infrastructures are almost non-existent. Population growth, diversification of economic activities as well as new lifestyles and consumption patterns led to an increase waste deposits. Thus, we are witnessing an uncontrolled development of "unregulated" waste dumps called open landfills in many African countries due to the lack of regulation and appropriate infrastructure. The proliferation of these wild dumps, which have no technical facilities, seriously affect population health and environment [1]. It has been established that 70% of waste produced in Congo is biodegradable (household and similar waste), and the remaining 30% is divided between industrial and hospital waste [2]. These wastes are all mixed in the landfills. Solid waste in the two main cities of Congo, Brazzaville and Pointe-Noire, comes from household waste, craft, hospital and hotel activities. Wastewater is collected and discharged, without prior treatment, into Congo river in Brazzaville and into Tchinouka river, Songolo river and sea at Pointe-Noire [2]. Indeed, these landfills are real risks of pollution of the air, soil and water: landfill waste ferments and produces biogas mainly rich in CH₄, a greenhouse gas that contributes to global warming [3]. The fermentation juice from these landfills flows through soil by infiltration and may contaminate drinking water resources [4,5]. Toxic components of these wastes such as heavy metals (mercury, chromium, lead and many others) can leach into groundwater as well as into surface water [6]. In Congo, solid waste is commonly burned on the ground in open air. Outdoor burning leaves the operator with no control of the combustion process. This produces pollutants and ashes that can be dangerous and that will likely impact on the surrounding soil and water. In addition, foods that are not fully burned attract wild animals. Recent studies showed that ash from burning and accumulated in soil can contribute significantly to soil sorption and reduction of organic contaminants leaching [7,8], as well as heavy metals [9]. These studies showed that burning ash can influence soil persistence and mobility of organic contaminants and heavy metals to various compartments of environment. Several alternatives can be considered for ash management, including solidification/ stabilization. Indeed, the solidification makes it possible to transform

waste ash into a massive solid little porous and little permeable in order to limit as much as possible the possibility of dispersion in environment [10]. Stabilization consists of a physical and / or chemical retention of polluting species in a solid matrix by ionic absorption or exchange mechanisms [11] in order to make them poorly soluble and not very mobilizable. The product of these two processes thus makes it possible to limit the dispersion of pollutants in natural environment. Immobilization of ultimate waste by cement is the most widely worldwide used technique for the final treatment of hazardous waste because of its advantages: low economic cost and easy implementation [12]. Waste immobilized by the cement can represent a source of pollution because they are influenced not only by the mechanical loadings but also by environment (freeze-thaw, acid rain, saline and aggressive waters ... etc.) [13]. Characterization by leaching tests is therefore necessary to predict the immobilization of this ultimate waste and the durability of these materials in long term. The aim of this study is to evaluate chemical durability of materials resulting from the stabilization / solidification by hydraulic binders of waste ashes from Brazzaville landfills. This assessment consists on static leaching tests based on standardized tests applied for porous materials obtained by stabilization/ solidification of waste ashes. The objective is to quantify the release of heavy metals contained in porous materials obtained by stabilization/ solidification of waste ashes.

2. MATERIALS AND METHODS

2.1 Study Site

The present study was carried out in Brazzaville, Republic of Congo. The city of Brazzaville has a varied relief of plain at 280 m altitude, plateaus in the west at an altitude of 310 - 325 m and hills all around the northern rim at an altitude of 400 to 475 m. The plateaus and hills are composed of sandy-clay soils on a sandstone base. As for the plain consisting of sandy soils, it occupies the center of the city of Brazzaville extending along the Congo River. Its base is chalky sandy clay [14]. The city of Brazzaville has a humid tropical climate still called "Congolese low" climate which is characterized by abundant rainfall (1,273.9 mm rainfall / year) with an average temperature of 25°C with amplitudes ranging from 15 to 32°C [15]. All domestic solid waste generated in

Brazzaville is dumped in uncontrolled landfills. For a population estimated at 1,411,015 inhabitants, the city of Brazzaville produced in 2008 about 36,121,986 tons of household waste; which corresponds to 256 kg / person / year and 1,403 wild landfills were identified [16]. Waste is deposited on the ground. No provision is made to collect leachate and to preserve the soil by minimizing its infiltration into the subsurface. This creates a scenario where contaminant concentrations increase with time. Close to these wild landfills live populations that use well water for their food taken from the groundwater. So, there is a high risk of groundwater contamination from the landfill. Fig. 1 shows a map of Brazzaville and the 4 landfills chosen for this study:

Landfill A: This wild dump is called “*the poudrière*” and is located in the district N°7 Mfilou. It has a flat geomorphology near a stream. It has been existing for 7 years.

Landfill B: This wild dump is called “*Moukondo Market*”. It is located in district N°4 Mougali. It has an inclined geomorphology and has been existing for 20 years.

Landfill C: This wild dump is called “*City 17*”. It is located in district N°7 Mfilou. It has an inclined geomorphology with a depth of about 20 m. It has been existing for 16 years.

Landfill D: This wild dump is called “*Emeraude Mikalou*” is located in district N°6 Talangai. It has an inclined geomorphology with a depth of about 10 m. It has been existing for 9 years.

2.2 Landfills Waste Sampling

Waste was collected in 4 landfills, located in Brazzaville in several districts: Landfill A, Landfill B, Landfill C and Landfill D. The collection of household waste in landfills was done manually

using a shovel. Quantity of waste taken was 10 kg per landfill, i.e. a total of 40 kg. Waste was then sun-dried in open air for seven (7) days. Dried waste was then burned in a garden incinerator for 30 minutes. Burned residues (ashes) were then recovered from the bottom of the garden incinerator and stored in glass jars.

2.3 Mobility of Heavy Metals in Ash

To simulate mobility of heavy metals in ash, we carried out static leaching tests whose experimental procedure is defined by the AFNOR X31-240 standard. The leaching solution used is distilled water. The test consists in putting 5 g of ash in contact with 50 g of distilled water (liquid/solid ratio = 10) with constant stirring at room temperature (25°C) for 16 hours. After 16 hours of contacting, leachate is filtered on filter paper and solid (hydrated ash) is recovered and dried in an oven at 100°C to allow the evaporation of residual water.

2.4 Materials Studied

In order to propose a methodology for evaluating pollutants (heavy metals) transfer from waste ash included in materials resulting from the stabilization/ solidification by hydraulic binders, we developed several formulations of materials. Formulations studied in this work are based on Dolisie Portland cement, lime and waste ash. Cement composition is given in Table 1. Immobilization of waste ash was carried out by the cement with a water / cement ratio equal to 0.5 to promote the release of chemical elements [17]. Different formulations were made by varying the ash / cement / lime ratios. These formulations are presented in Table 2. Stabilized / solidified materials were synthesized in UC2V laboratory (Plant and Life Chemistry Unit) at Faculty of Science and Technology of Marien N'GOUABI University.

Table 1. Composition of Portland cement of Dolisie (% weight)

CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	Na ₂ O	K ₂ O	NiO	PbO	Cr ₂ O ₃
64,8	17	6,5	4,6	3,3	2	1,8	/	/	/

Table 2. Composition of studied materials (% weight)

Cementitious matrices	P1	P2	P3	P4	P5	P6
Ashes (% weight)	50	70	50	50	70	70
Hydrated cement (% weight)	50	30	40	30	20	10
Lime-CaO (% weight)	0	0	10	20	10	20



Fig. 1. Map of Brazzaville indicating the location of the 4 landfills chosen for this study. The landfills are indicated by red, purple, black and blue dots

2.5 Static Leaching Tests on Cementitious Matrices

The aim of static leaching tests is to monitor the release of pollutants (heavy metals) from waste ash included in monolithic materials resulting from stabilization/ solidification by hydraulic binders. Materials samples of 1 cm × 1 cm × 0.5cm in size are put in contact with distilled water. The ratio of material surface / volume of water is 0.1, so as never to reach the saturation of species followed. Material samples are placed at the bottom of glass vials to allow the lixiviant to bathe all faces of monoliths. Vials are then carefully closed to prevent the admission of air in interior. These leaching tests were carried out over a period of thirty (30) days at 25°C. These experiments carried out on short times, still far from saturation, will make it possible to quantify the relaxation in solution of the elements contained in cementitious matrices and to promote development of an alteration film on the surface of materials. Leaching tests were carried out at UC2V laboratory (Plant and Life Chemistry Unit) at Faculty of Science and Technology of Marien N'GOUABI University.

2.6 Leachate Analysis Methods

Leachate analyzes were carried out in UC2V laboratory (Plant and Life Chemistry Unit) at the Faculty of Science and Technology of Marien N'GOUABI University and in National Institute of Research in Exact and Natural Sciences (IRSEN). The Lovibond 330-900 nm spectrophotometer was used for the analysis of cement and heavy metals in leachates. Heavy metals analyzed in leachates are: copper, aluminum, lead, nickel, chromium and zinc.

3. RESULTS AND DISCUSSION

3.1 Leaching of Heavy Metals from Ashes

Results of leachate heavy metal analyzes coming from household waste ashes of four (4) landfills are reported in Table 3. These results were compared to landfill leachate discharge requirements [18]. Results recorded in Table 3 show a great variability in leachate composition. Heavy metals analysis of leachates shows the almost total absence of aluminum in leachates from landfills A, B, C and D. Average concentrations of other heavy metals in leachates vary between 1 and 6.5 mg/l for copper, 3.2 and 21.8 mg/l for lead, 5 and 12.5 mg/l for nickel, 3.4 and 6.5 mg/l for chromium

and 0.7 and 1.4 mg/l for zinc. Lead, nickel, chromium and copper concentrations are very important. These values far exceed landfill leachate discharge requirements. Only zinc remains below these requirements. This chemical characterization show that these ashes are classified as hazardous and toxic wastes. Especially since some contaminants can be leached from ashes and migrate to a permeable medium. Soil is a heavy metal retention factor because they can be trapped in superficial part of ground [19]. For this purpose, the ashes must be immobilized with cement to reduce their pollutant potential.

3.2 Leaching of Heavy Metals from Cementitious Matrices

Lead, nickel and chromium are three heavy metals that present environmental hazards. We will therefore focus our study on the leaching of these three elements. Monolithic leaching tests were performed on cementitious matrices for 30 days to track percentage of heavy metals (lead, nickel and chromium) immobilized by cement. The results obtained are represented in Fig. 2, Fig. 3, Fig. 4 and Fig. 5. These figures show an overview of Pb^{2+} , Ni^{2+} and Cr^{3+} concentrations resulting from waste burning landfills (1, 2, 3 4) and cementitious formulations P1, P2, P3, P4, P5 and P6 leachates: in these figures, the curve trends is the same for all landfill waste ashes. It is observed that raw ash solubilize significant quantities of heavy metals: between 3 and 22 mg/l for lead, 5 and 13 mg/l for nickel and 0.01 and 0.36 mg/l for chromium. Heavy metals concentrations in stabilized/ solidified cementitious matrices P1, P2, P3, P4, P5 and P6 leachates were greatly reduced compared to concentrations of Pb^{2+} , Ni^{2+} and Cr^{3+} ions released from raw ash. This can be explained by the fact that during hydration of cement in presence of ash, two phenomena occur: On the one hand, heavy metals can be trapped in cement structure, which reduces their mobility, on the other hand, heavy metals can react with another species present in cement during hydration reactions, which can change chemical speciation and make it less soluble and less mobile [20]. Compared with the landfill leachate discharge requirements [18] which sets discharge limit values at 0.5 mg/l for lead and 2 mg/l for nickel and chromium. The curves indicate that the addition of lime influences the results of leaching. In fact, in cementitious matrices P3, P4, P5 and P6 leachates, Pb, Ni and Cr are weakly solubilized compared to P1

and P2. Lead concentrations in P1 and P2 cementitious matrices are still very much above the norm regardless of type of ash, while the nickel and chromium contents are below standard in both formulations. It appears that lead remains above the norm in CaO-free cementitious matrices P1 and P2. Fig. 6, Fig. 7, Fig. 8 and Fig. 9 show the behavior of heavy metals in leachates of cementitious matrices P3, P4, P5 and P6 in which CaO was added. The maximum concentrations in P3, P4, P5 and P6 cementitious matrices leachates are of the order of 0.7 mg/l for lead, 0.9 mg/l for nickel and the chromium concentrations are close to detection limit. This can be explained by the fact that addition of lime improves the physical properties of cementitious matrices, in particular physical durability and dimensional stability [21]. This then leads to better retention of pollutants inside P3, P6 cementitious matrices, despite the presence of lime, can be explained by the fact that during the hydration of the cement in a basic medium, P4, P5 and P6 cementitious matrices compared to P1 and P2 that do not contain lime. The low leaching of lead and nickel in the P3, P4, P5 and P6 cementitious matrices, despite the presence of lime, can be explained by the fact that during the hydration of the cement in a basic medium, lead precipitates mainly in form of $Pb(OH)_2$ hydroxide and $Pb(OH)_3$ complex ion [22] and nickel predominantly precipitates as hydroxide $Ni(OH)_2$ [23]. However, pH of leaching solutions is close to pH dissolution of $Pb(OH)_2$, $Pb(OH)_3$ and $Ni(OH)_2$ phases, hence the weak passage in solution of Pb^{2+} and Ni^{2+} ions despite the

presence of lime in the cementitious matrices. The very weak release of chromium in the water is due to the fact that during cement hydration, Cr^{3+} transforms into Cr^{6+} [24]. This justifies its very low presence in leachates. Thus, only the cementitious matrices P3, P4 and P6 have lead, nickel and chromium concentrations below the limit values set by the landfill leachate discharge requirements. Lead, nickel and chromium concentrations in the P5 cementitious matrix remain below the limit values set by the leachate discharge standard except for one containing ash from the landfill 4.

3.3 Heavy Metal Retention in Monoliths

From the results of Table 4, it is observed that concentrations of heavy metals from the cementitious matrix leachates decrease compared to concentrations of the same elements released from raw ash. Formulations enriched with lime CaO, P3, P4, P5 and P6, have best retention rates of heavy metals, greater than 90% compared to raw ash. P3 and P4 formulations have the best retention rates of heavy metals. Retention rates are between 97-98% and 98-99% for lead respectively for P3 and P4, 98-99% for nickel and greater than 99% for chromium for both formulations. To this end, we can say that process of immobilization of heavy metals by Dolisie Portland cement applied to raw ash considerably reduced the polluting character and the release of toxic heavy metals in surrounding environment.

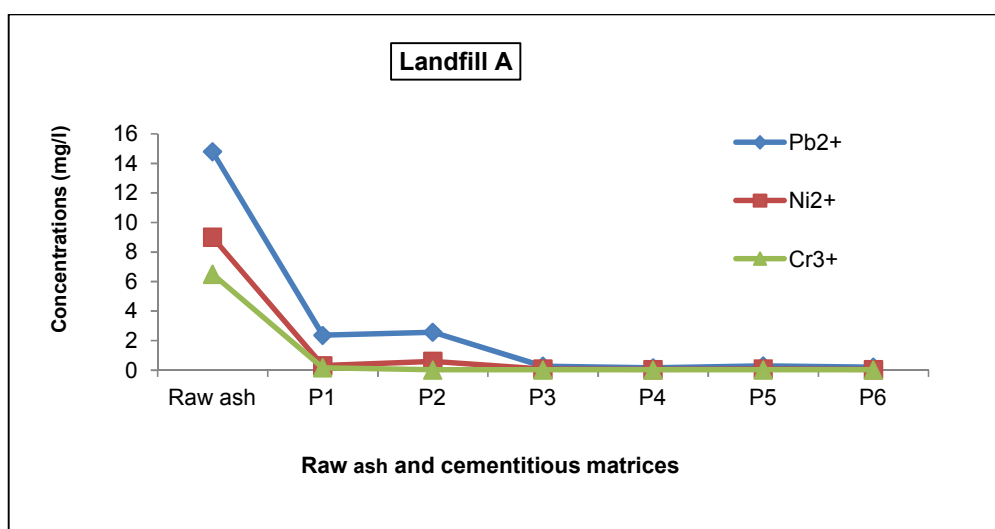


Fig. 2. Heavy metal leaching of raw ash and cementitious matrices P1, P2, P3, P4, P5 and P6 containing ashes from landfill A

Table 3. pH and heavy metals concentrations (mg/l) in ash leachates

	pH	Cu	Al	Pb	Ni	Cr	Zn
Landfill A (Cité des 17-Moukondo)	11,4	1	<0,01	14,8	9	6,5	1,2
Landfill B (Emeraude-Mikalou)	11,5	1,5	0,2	17,5	11	4,2	1,4
Landfill C (Poudrière-Mfilou)	11,3	3,2	<0,01	3,2	5	3,5	1
Landfill D (Marché-Moukondo)	12,5	6,5	0,08	21,8	12,4	3,4	0,7
Landfill leachate discharge requirements [18]	/	0,5	/	0,5	2	2	2

Table 4. Retention rate of heavy metals in cementitious matrices (%)

Cementitious matrices	Cement	Ash	Cao	Retention rate	Retention rate	Retention rate
				Pb ₂₊	Ni ₂₊	Cr ₃₊
P1	50	50	0	80-85	90-95	98-99
P2	30	70	0	75-80	88-90	94-96
P3	40	50	10	97-98	98-99	> 99
P4	30	50	20	98-99	98-99	> 99
P5	20	70	10	90-95	98-99	> 99
P6	10	70	20	95-97	98-99	> 99

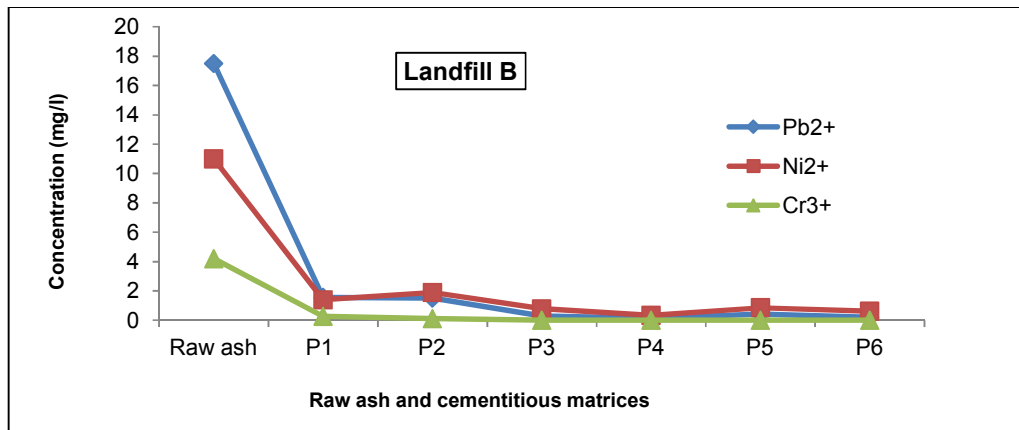


Fig. 3. Heavy metal leaching of raw ash and cementitious matrices P1, P2, P3, P4, P5 and P6 containing ashes from landfill B

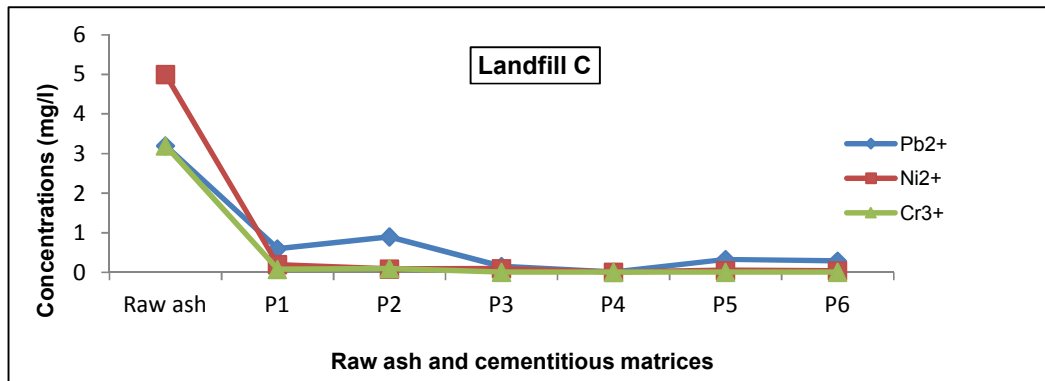


Fig. 4. Heavy metal leaching of raw ash and cementitious matrices P1, P2, P3, P4, P5 and P6 containing ashes from landfill C

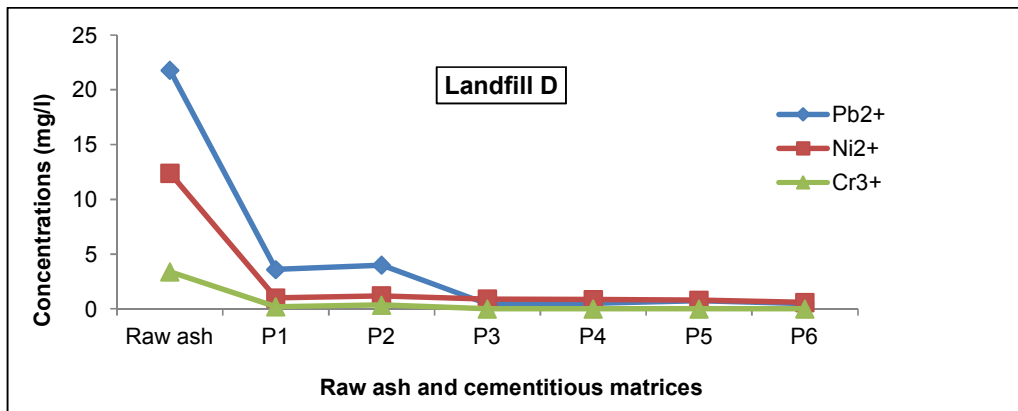


Fig. 5. Heavy metal leaching of raw ash and cementitious matrices p1, p2, p3, p4, p5 and p6 containing ashes from landfill D

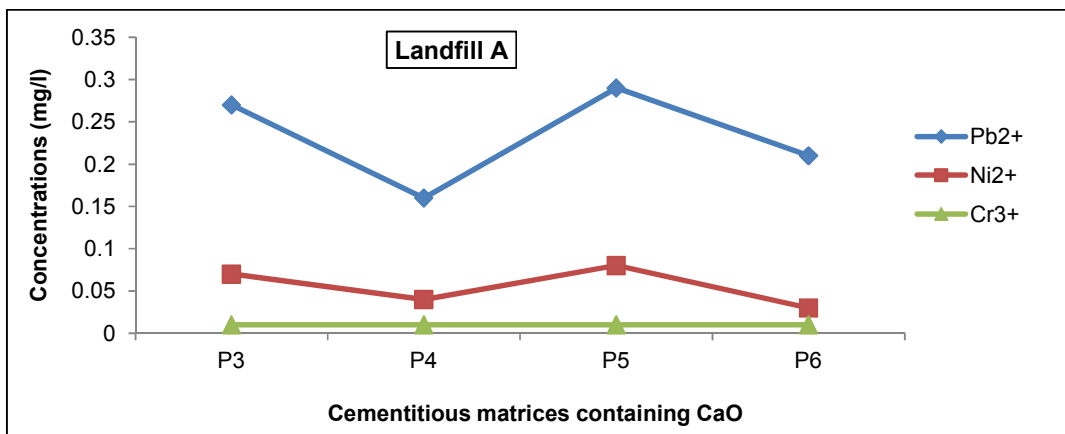


Fig. 6. Heavy metal leaching of cementitious matrices P3, P4, P5 and P6 containing ash from landfill A and lime CaO

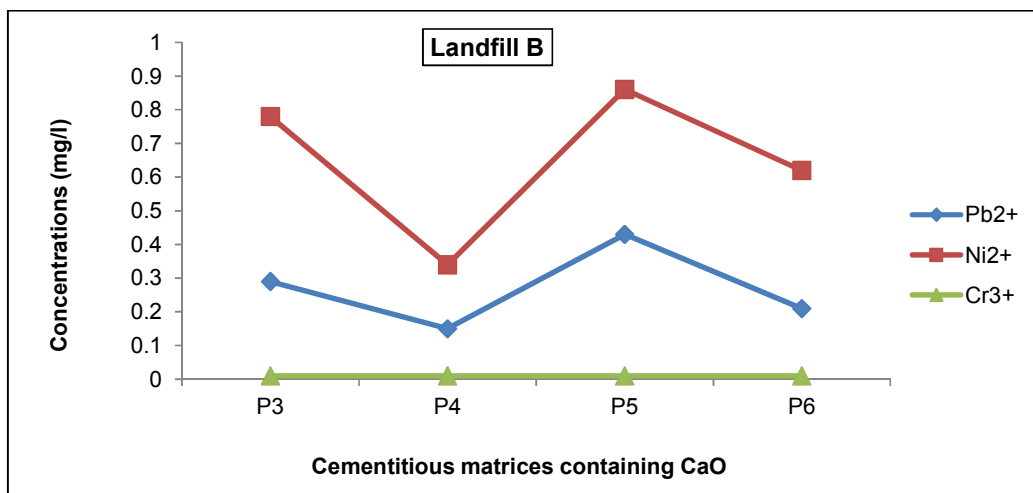


Fig. 7. Heavy metal leaching of cementitious matrices P3, P4, P5 and P6 containing ash from landfill B and lime CaO

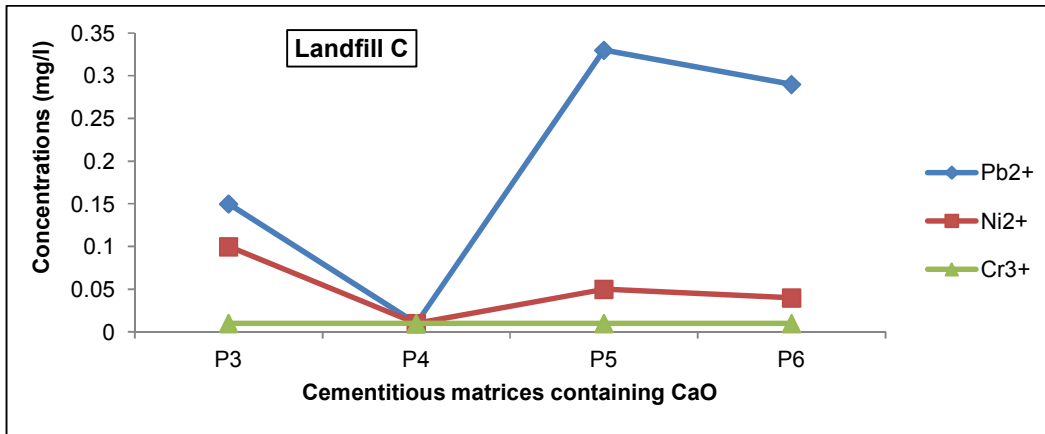


Fig. 8. Heavy metal leaching of cementitious matrices P3, P4, P5 and P6 containing ash from landfill C and lime CaO

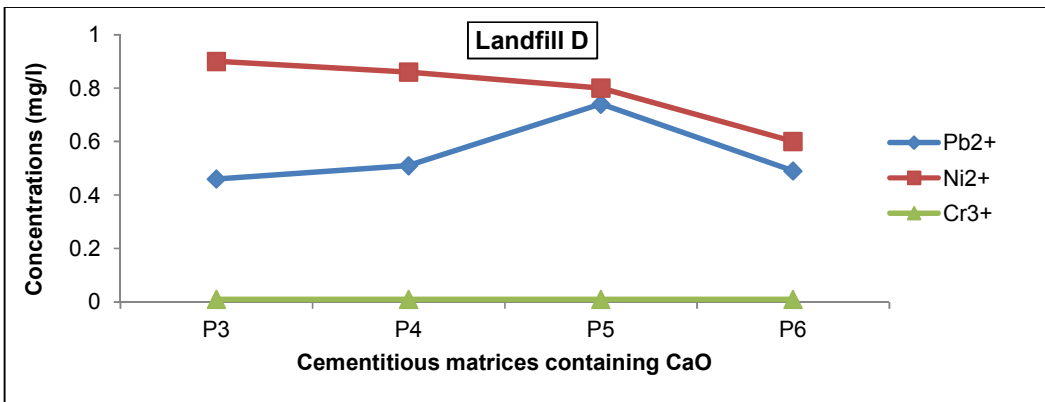


Fig. 9. Heavy metal leaching of cementitious matrices P3, P4, P5 and P6 containing ash from landfill D and lime CaO

4. CONCLUSIONS

In summary, the results of leaching tests applied to waste ashes and cementitious matrices allow us to conclude that: On the one hand, characterization of waste ashes by dissolution in solution shows that these ashes contain significant concentrations of lead, nickel and chromium. As a result, raw ashes are classified as hazardous waste or pollutants and must undergo immobilization treatment by cement to reduce their pollutant character. On the other hand cementitious matrix leachates presented small amounts of release of heavy metals versus raw ash. However, cementitious matrices containing CaO lime have the best heavy metals retention rates because their concentrations in leachate remain below the requirements of the landfill standard leachates. Thus, results obtained in terms of quantities of heavy metals

retained for 30 days compared with those of ashes in the raw state, demonstrate the efficiency of process adopted for immobilization of heavy metals contained in ash by Portland cement of Dolisie.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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