

HEAVY METAL BEHAVIOR ASSOCIATED WITH COVER SOILS OF TWO SANITARY LANDFILLS IN SÃO PAULO METROPOLITAN REGION



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ABSTRACT

Urban solid waste (residential, commercial, municipal, industrial) disposed in Brazilian sanitary landfills, contain in small quantities, potentially toxic and hazardous components, which include the group of heavy metals. The aim of this research was to study the behavior of these ions when associated with soil, wastes and leachates of two sanitary landfills in Metropolitan Region of São Paulo, São Paulo State. Heavy metals enrichment was detected in the cover soil of both landfills, showing low concentrations of interstitial ions and high levels of adsorbed metallic ions. Columns experiments containing soil and artificial waste (bananas) layers reproduced efficiently phenomena that occur in real sanitary cells. Different stabilization phases (aerobic, aerobic/anaerobic, anaerobic) were monitored by systematic collection of leachates, whereas ionic behavior related to soils, was accompanied by solution injections with known concentrations of Pb, Cu, Cr and Ni. Soils answers to percolations were distinct, showing in Landfill I soil higher adsorptions due to its texture, which dominated mineralogy and cationic exchange capacity of Landfill II soil. During the initial decomposition phases, the production of organic acids (complexes) in leachate, the raise of ionic force and pH drop, favored ionic availability. The decrease of heavy metal ions in leachate during later phases was a consequence of pH elevation and chemical transformations, which culminated in the methanogenic phase. Ionic fixation in soils, especially Pb and Cu, occurred due to unsolvable compounds formation (carbonates, oxo-hydroxides) whereas Ni retention was essentially related to adsorption phenomenon. Chromium suffered influence of pH and Eh, and precipitated as chromium (III) during methanogenic phase. The efficiency of sanitary phenomena reproduction in columns is an evidence that soil used to cover compacted solid waste at landfills must be selected.

1. INTRODUCTION

Urban solid waste (residential, commercial, municipal, industrial) disposed in Brazilian sanitary landfills contains, even in small quantities, hazardous components, within which may be included the group of the heavy metals. After refuse is disposed, no control exists of its distribution inside the landfill. Thus, environmental basal protection depends on the efficiency of the landfill construction and operation, which includes waterproofing, daily and final compacted soil cover, leachate and gas collection systems etc.

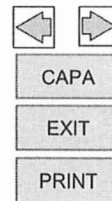
The aim of this research was to monitorate the geochemical behavior of the heavy metal ions lead (Pb), copper (Cu), chromium (Cr) and nickel (Ni), in relation with soil, wastes and leachates of two sanitary landfills in the Metropolitan Region of São Paulo, São Paulo State, Brazil. Landfill I (São Paulo District) receives domestic and industrial wastes and sewage sludge. In Landfill II (Mauá District) domestic refuse predominates.

In studied landfills, the sanitary cells are composed by a compacted garbage layer (5 m high), which are daily covered by a compacted soil layer (10 – 50 cm). The soil cover usually comes from areas nearby landfill but are also provided by extracted material obtained during the landfill construction.

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The soil from Landfill I is characterised by a clay-loam texture, low cationic exchange capacity-CEC ($13,7 \text{ mmolc.dm}^{-3}$) and kaolinite dominant mineralogy, having also muscovite, illite and oxi-hidroxides. The soil from Landfill II is more sandy, shows a similar mineralogy as that present in Landfill I, differing because of the presence of vermiculite, which is responsible for a higher CEC (38 mmolc.dm^{-3}). The pH values measured in both soils varied from 4 to 5, and displayed positive ΔpH ($\text{pH}_{\text{CaCl}_2} - \text{pH}_{\text{KCl}}$) obtained for most samples demonstrating cation adsorption capacity.

Column tests that simulated sanitary cells of each landfill were used to study the mechanisms of liberation, transport and fixation of metals within landfills. Some construction aspects of both landfills were considered for an adequate reproduction of site conditions. The same background soils present in the landfills were used in the experiments.

2. EXPERIMENTS

Laboratory simulations of a soil/waste/water/heavy-metal system allowed to control waste decomposition phases and associated leachate characteristics.

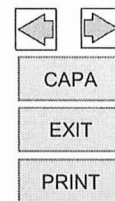
Three experiments were carried out using PVC columns containing soil – artificial waste – soil layers. The width of these layers was based on refuse *versus* soil proportion of landfills, which corresponds to approximately 8:1. The proportion was reduced to 4:1 in order to accelerate decomposition process. Minced bananas were used as artificial waste; this fruit was chosen due to its easier decomposition and for being seedless.

Experiment 1 – Leachate generation and compositions were monitored through two 6" diameter and 50 cm high columns, corresponding A to a background column and B to a column contaminated by heavy metals. Percolated liquids were collected using perforated 1" tubes that simulate wells. Representative soil (< 2 mm size fraction) of Landfill I was used in this experiment.

Experiment 2 – Two consecutive cells were simulated inside one column (C). Leachate of upper cell and lower cell were collected separately. Soil of Landfill I was used.

Experiment 3 – As in experiment 1, a background column (D) and a column impacted with heavy metals (E) were also used. Thus, columns diameter were smaller (3"), improving more decomposition acceleration. Soil layers were composed of Landfill II soil.

Columns A and D were leached with distilled water, whereas the other columns were leached with a solution containing 1 mg dm^{-3} of Pb, Cu, Cr and Ni. The ionic solutions were introduced once a week in the columns (200 mL in columns A, B and C; 100 mL in D and E), and leachate samples were collected after 12 hours in order to measure pH, oxidation-reduction potential (Eh), electrical conductivity (EC), and establish Na^+ and K^+ concentration. In the sequence, samples were treated with $\text{H}_2\text{O}_2/\text{HNO}_3$ and were analysed by atomic absorption spectrometry (AA700BC/CG) in able to determinate metals content.



3. RESULTS AND DISCUSSION

Experiments 1 and 2 were monitored for 12 months, and the third experiment for 9 months. Leachate produced in all columns presented similar behavior when considering analytical parameters, as showed in the representative diagrams of Figure 1.

Four different phases, aerobic, aerobic/anaerobic (transitional), anaerobic and methanic, could be associated to the decomposition processes (Figure 1), which were respectively compared with other studies and experiments (Pohland and Harper, 1985; Bozkurt et al., 2000; Kjeldsen et al., 2002; among others).

In the aerobic and the transitional phases, pH drop was due to organic acid formation due to the continuous CO₂ development, which was responsible for material solubilization, high concentrations of Na and K in leachate and, consequently, EC elevation. During subsequent phases, electrolyte concentration decreased gradually and pH increased progressively as a consequence of chemical transformations that took place in the refuse.

Eh oscillated during most part of the time and became more reduced at the end, when the methanic phase was achieved. This methanogenic phase occurred only in Experiment 3 due to the column diameter. As expected, in column C (Experiment 2) the mass transference from the upper cell to the lower cell was responsible for reaction intensification in the lower cell, causing a delay of the aerobic/anaerobic phase.

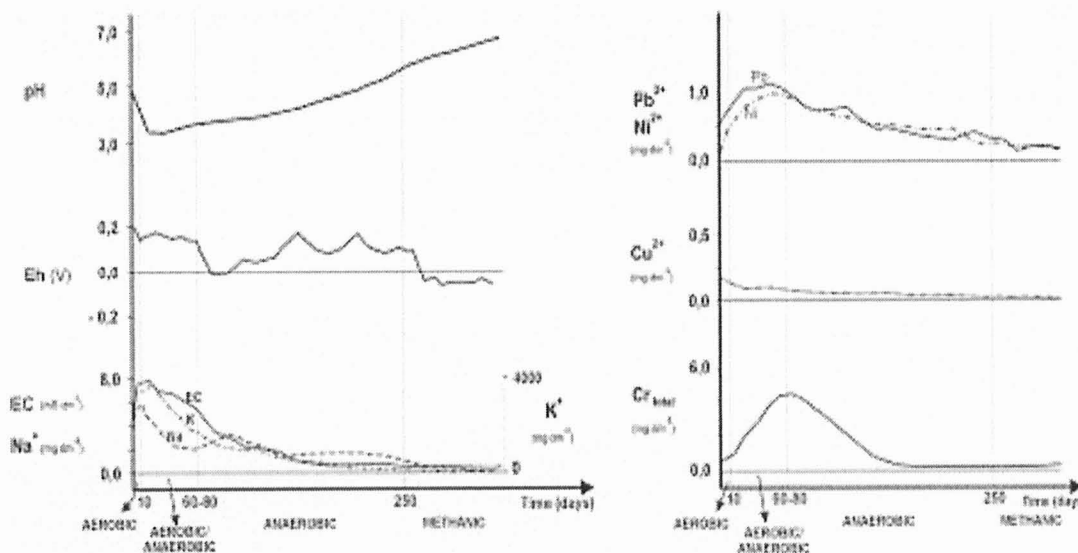
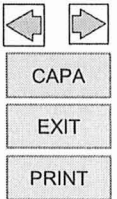


Figure 1. Leachate characteristics development along degradation phases in columns.



During the experimental time a continuous heavy metal adsorption was verified. The ionic availability was conditioned by pH of environment.

In the initial phases, material solubilization due to the decrease of pH and the raise of the ionic force favored the displacement of adsorbed metals and its ionic availability as free ions (Figure 1). Thus, the decrease of heavy metal ions in leachate during later phases was a consequence of pH elevation and chemical transformations. Conditions with pH higher than 5 favored adsorption of Pb and Cu in soils and their precipitation as unsolvable carbonates and oxi-hydroxides, having CO_2 derived from the anaerobic phase. Ni concentration didn't varied as a consequence of pH and Eh variation and continued in solution as water-complexes.

Chromium suffered influence of pH and specially of Eh, precipitating as chromium (III) during the methanic phase, due to the reduction conditions that prevailed associated to alkaline pH values.

After finalizing the experiments, all the materials of the impacted columns were separated and submitted to nitric extraction and AAS determination for available metals. Analyses indicated that heavy metals were more adsorbed in the upper soil layer within columns B and C. Metals were less fixed in organic matter due to the formation of soluble complex ions that favored ionic mobility. The intermittent addition of electrolytes in the system and the presence of organic matter worked against the total occupation of adsorption sites.

The behavior in column E was different because of the sandy texture of Landfill II soil, which propitiated more velocity of leaching flow. Consequently, residence time and adsorption of ions in the soil was smaller. In this column the accumulation of heavy metals in the organic matter layer was due to higher pH values that favored precipitation of them as oxi-hydroxides.

4. CONCLUSION

Heavy metal behavior in a soil/waste/leachate system depends of the chemical and physical-chemical conditions imposed by the waste degradation phases in the initial (metal availability) and final stages (decrease of heavy metal ions).

The equilibrium pH value of experiments was arround 8,0, being similar to that measured in landfill leachates, which demonstrated that during the stababilization phases heavy metals are mainly retained and fixed due to adsorption or precipitation.

Ionic retention was more efficient in the soil from Landfill I due to its texture. This is an evidence that soil used to cover compacted solid waste at sanitary landfills must be selected.



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