

VOL. 28, 2012

Guest Editors: Carlo Merli Copyright © 2012, AIDIC Servizi S.r.l., **ISBN** 978-88-95608-19-8; **ISSN** 1974-9791



DOI: 10.3303/CET1228004

Variations of the Physical and Chemical Characteristics of Sanitary Landfill Leachate after Percolation into Tropical Soils

Miriam G. Miguel^{*a}, Leandro Belinassi^a, Ligia M. Domingues^a, Edson A. A. Nour^a, Sueli Y. Pereira^b

^aFaculty of Civil Engineering, Architecture and Urbanism, State University of Campinas, Avenida Albert Einstein, 951, Campinas, São Paulo, Brazil

^bGeoscience Institute, State University of Campinas, Rua João Pandiá Calógeras, 51, Campinas, São Paulo, Brazil miriammiguel@uol.com.br

Tropical soils are common in Brazil and present very peculiar geotechnical properties, such as fine particle aggregation; natural cementation; and high porosity, which often turn away from the classical models of the Soil Mechanics. Due to this, they are now the focus of recent studies for use as alternative materials for several applications in geotechnical site. This research studies two tropical soils from the Southeastern Brazil to evaluate the variations of the physical and chemical characteristics of sanitary landfill leachate after percolation into these soils. Soil samples were submitted to geotechnical, physical, chemical and mineralogical tests. After these tests, soil samples were compacted with 3 % above their optimum moisture content and placed in polyvinyl chloride columns. The leachate was collected from a regional sanitary landfill and characterized by physical and chemical tests with relation to volatile fatty acid (VFA), chemical oxygen demand (COD), pH, alkalinity, and metals (Cr, Mn, Fe, Co, Ni, Cu, Zn and Pb), before and after percolation into the soils by means of column tests. Curves of concentration *versus* percolation time for COD, VFA, and metals were obtained for the two soils. COD concentration increased with percolation time for the two soils, while the adsorption/desorption behaviours were different for the two soil, mainly for VFA, Pb, Ni and Co.

1. Introduction

Domestic waste treatment has been a major challenge for Brazil and others countries. Sanitary landfill for domestic waste has been an environmental and technical solution, however, it is not yet economically feasible, due to constructive demands. The sanitary landfill construction requires a waterproof foundation to protect the natural soil and groundwater against leachate flow. Compacted clay liner (CCL) and/or polyethylene synthetic membranes have been used for this purpose (Daniel, 1993). Studies about the permeability and the mechanisms that influence the contaminant transport, such as advection, diffusion, dispersion and sorption, are necessary for the evaluation of liners (Shackelford, 1993).

Bentonite is an expansive clay and presents low hydraulic conductivity when saturated. These properties make the bentonite as good raw material for construction of liners on the sanitary landfill; however, there are only a few bentonite deposits in Brazil, what makes its application very costly. In tropical areas, however, there is dominance of lateritic soils, which are the results of the pedological alterations caused by physical and mainly chemical weathering. The laterization process is the

Please cite this article as: Goncalves Miguel M, Belinassi L., Domingues L.M., Nour E.A.A. and Pereira S.Y., 2012, Variations of the physical and chemical characteristics of sanitary landfill leachate after percolation into tropical soils, Chemical Engineering Transactions, 28, 19-24 DOI: 10.3303/CET1228004

transport of particles under the action of infiltration and evaporation, forming a porous superficial horizon with the more stable minerals remaining, such as quartz, kaolinite, gibbsite, hematite, and goethite. The mineralogical composition of the lateritic soils allows the adsorption of the heavy metals due to the presence of the oxides and hydroxides of iron and aluminium. Studies have shown that the use of compacted tropical clays (Paraguassú et al., 2002; Boscov, 2010; Nascentes et al., 2011; and others) can be a technical and economic alternative for the use as liners.

Focusing on this context, this research studies the variations in the physical and chemical characteristics of the effluent from the leachate sanitary landfill before and after the percolation in two compacted tropical soils found in the Southeastern São Paulo State, Brazil, with the purpose of usage as liners for a sanitary landfill foundation.

2. Material and methods

2.1 Soil and leachate samples

The Experimental Field of Soil Mechanics and Foundations (EFSMF) of the State University of Campinas (Unicamp), at Campinas city, presents a colluvial tropical soil profile with about 6.5m of thickness, typical of Serra Geral Formation, representing about 14 % of the region of Campinas city. Laranjal Paulista city presents typical residual tropical soils, from Passa Dois and Tubarão Geological Formation. Both cities are located in the southeastern state of São Paulo, Brazil.

Undisturbed soil samples studied were originated from the superficial layer of EFSMF (Soil 1) and the Laranjal Paulista city (Soil 2). The soil sample from EFSMF was collected by excavation of the deep inspection pit shaft with 1.0 m of diameter, between 1.5 and 2.5 m depth. The soil sample from Laranjal Paulista was collected at 0.5 m depth from the clay deposit of the city's ceramic industries.

The soil samples were submitted to firstly geotechnical tests: grain-size distribution, with and without dispersant use in the sedimentation phase (ASTM, 1998); consistency limits; compaction on Normal Proctor Energy; permeability to water; and secondly physical, chemical and mineralogical tests such as cation exchange capacity (CEC); pH; and x-ray diffraction.

The leachate samples were collected directly from a leachate lake from the Delta A Municipal Waste Landfill, in Campinas city. The leachate flow rate of the sanitary landfill has been 80m³ by day.

2.2 Testing procedures

The two tropical soils were compacted by the standard Proctor method (ASTM, 1995) and their values of the optimum moisture content for the maximum compaction efficiency were obtained. In the permeability tests of the rigid wall, iron permeameters were used. The soils were compacted 3 % above their optimum moisture contents and were submitted to saturation with distilled water by backpressure, which took about a week. After the saturation, the permeability tests of the variable load initiated, following the recommendations of the Brazilian Standard (ABNT, 1995).

The column tests with leachate percolation were performed with PVC column and connections, acrylic plates and metallic rods with endless screws. The soils were also compacted 3 % above their optimum moisture contents and submitted to saturation with distilled water by backpressure during about 72 h. After that, the percolation with raw leachate was initiated.

The raw leachate and the effluent solutions from the columns were submitted to the physical-chemical tests as follows: concentration of volatile fatty acid (VFA), according to DiLallo and Albertson (1961), chemical oxygen demand (COD), hydrogen potential (pH), and alkalinity, according to APHA (1998). The metal concentrations were determined by ICP-MS XseriesII (Thermo) with CCT (Collision Cell Technology) that is inductively coupled plasma mass spectrometry.

3. Results and discussion

3.1 Geotechnical, physical and chemical parameters of the soils

The geotechnical, physical and chemical parameters of both soil samples are presented in Table 1. High values of the grain unit weight for Soil 1 are highlighted in Table 1, indicating the presence of iron oxides and hydroxides. Nonetheless, with respect to plasticity properties, both samples can be classified as CL-ML, according to USCS Classification. However, the values of CEC are greater for Soil 2, pointing its higher mineral activity.

Table 1: Geotechnical, physical and chemical parameters of the soils

Parameters	Soi	11	Soil 2		
Grain Unit Weight (kN/m ³)	30	.2	26.8		
Liquid Limit	4	5	47		
Plasticity Index	1	7	18		
Cation Exchange Capacity (CEC) (cmol/kg)	6,	8	19,4		
pH (CaCl ₂)	5.	2	3.9		
pH (H ₂ 0)	5.	9	4.3		
Organic Matter (%)	1.	7	1.0		
Maximum dry density (g/cm ³)	1.52		1.49		
Optimum moisture content (%)	28.5		25.1		
Saturated permeability coefficient to 20 °C (m/s)	1.2 x	10 ⁻⁹	6.4 x 10 ⁻¹⁰		
Grain size fractions		Percent by m	by mass (%)		
Grain-size fractions	WD	WD ND		ND	
Coarse Sand	1	1	0.5	0.5	
Medium Sand	7	14	1	1.5	
Fine Sand	14	33	12	12	
Silt	29	50	56	60	
Clay	49	2	30.5	26	
Textural Classification	Silty clay	Sandy silt	Clayey silt		

WD: with dispersant; ND: without dispersant

According to the MCT classification (Cozzolino and Nogami, 1993), specific for tropical soils, Soil 1 was classified as LG' (lateritic clayey soil), and Soil 2, as NA' (non lateritic sandy soil).

By observing Table 1, it is possible to notice that the Soil 1 showed higher micro aggregation than Soil 2. This fact shows that Soil 1 suffered more intense pedogenetic processes, such as leaching, and resulted in the more porous soil and with iron oxides and hydroxides acting as cement among the grains and with micro aggregation. The grain-size distributions allowed classifying Soil 1 as silty clay with dispersant and as sandy silt without dispersant. Nonetheless, the Soil 2 can be classified as clayey silt using or not using the dispersant.

In the x-ray diffraction tests were found mineral-clay kaolinite, iron oxides hematite, quartz and plagioclase. However, differences between the chemical compositions of the samples were detected by the presence of swelling and chemically active minerals: Soil 1 presented mineral-clay montmorrilonite and Soil 2, hydromica. In addition, Soil 1 presented gibbsite, which indicates longer aging in relation to Soil 2, leaning towards a lower chemical activity, agreeing with the CEC results (Table 1). As far as pH values are concerned Soil 2 presented lower values than Soil 1, however, both presented acid pH values, typical of tropical soils.

The values of the saturated permeability coefficient were obtained for the compacted soils with 3 % above the optimum moisture content (Table 1). Notice that the Soils 1 and 2 presented permeability coefficients relatively low. These results show the potential of these soils to be used as liners of sanitary landfills. It is also possible to notice that for the same compaction energy, the Soil 1 presented higher values than optimum moisture content and maximum dry density comparing to the Soil 2 curve. The distinct behavior of these soils when compacted can be associated to their different textures and mineralogical compositions, given by the x-ray diffraction tests.

3.2 Column tests

The results obtained from the chemical analysis of the raw leachate and effluent solutions, from the determining of the pH, alkalinity, volatile fatty acid (VFA), and chemical oxygen demand (COD), are presented in Table 2.

The pH obtained from the raw leachate was 7.9 and its alkalinity was 8064 mg/L (Table 2), what characterizes it as basic. The pH value found is the result of a biological stabilization of the organic

matter present in an old sanitary landfill, in which such behavior was verified by the evaluated characteristics.

In Table 2, it is possible to notice that the pH values from effluent solutions were lower than the raw leachate value. The effluent solutions from Soil 1 presented pH values around neutrality, while Soil 2 presented values which characterize the environment as acid. The pH values found in the effluent solutions are in accordance with the alkalinity values obtained.

Figure 1 was elaborated with the relations between VFA and COD concentration from effluent solutions (C) and initial concentrations from raw leachate (C_0), in relation to percolation time defined by Table 2.

Time (days)	рН (-)	VFA (mg/L)	COD (mg/L)	Total Alkalinity (mg/L)	Time (days)	рН (-)	VFA (mg/L)	COD (mg/L)	Total Alkalinity (mg/L)
Soil 1				Soil 2					
0*	7.9*	221.4*	1920*	8064*	0*	7.9*	221.4*	1920*	8064*
130	7.0	-	223	30	30	4.4	121.8	93	0
220	6.7	14.8	348	9.6	75	4.3	341.5	544	0
420	7.4	30.0	2138	41.8	115	4.1	693.6	615	0
* raw leachate (C ₀ values)					205	3.5	1208.4	980	0

Table 2. Concentrations in the percolation versus time



Figure 1. Leachate ratio concentration versus percolation time for COD and VFA

The COD concentrations increased slightly in the effluent solutions of both soils, while the VFA concentrations decreased in the effluent solutions of the Soil 1 and increased in the effluent solutions of the Soil 2 (Figure 1). The increased VFA concentration is due to degradation of the organic matter. Consequently, the COD concentration is decreased. Results indicated that the Soil 1 was able to adsorb the organic matter presented in the raw leachate, because VFA and COD concentrations decreased over time and the organic matter was not degraded. On the other hand, there were favorable conditions for the organic matter degradation in the Soil 2, because the increased VFA concentration (Figure 1) occurred in the effluent solutions.

The metal concentrations in the raw leachate and effluent solutions in relation to the times of column tests are shown in Table 3, for Soils 1 and 2. Cr, Co, Ni and Pb metals were totally adsorbed by Soil 1, not being detected by the chemical analysis from the first effluent solution after 130 days (Table 3). Fe was also adsorbed by Soil 1, however it maintained very low concentrations in the effluents since the first sampling. Mn presented higher concentrations in the effluent solutions than the initial concentration in the raw leachate, indicating that the soil desorbed this metal from its own chemical composition. Zn

was decreasingly eliminated in the two first effluent solutions until it could not be detected by the equipment in the last one. Nonetheless, Cu was totally eliminated; its concentration value was not detected in effluent solution for 220 days.

Soil 2 presented a very distinctive behavior from Soil 1 with relation to Co, Ni and Pb as seen in Table 3. The concentration of these elements increased in the effluent solutions with time, indicating that they were being desorbed from the chemical composition of the Soil 2. As far as the other metals such as Mn, Fe, Cr, Cu and Zn, the Soil 2 behavior was very close to the Soil 1, except for the magnitude of the concentrations of these metals, which were distinct.

Chamical	Time (days)				Time (days)				
Chemical	0*	130	220	420	0*	30	75	115	205
Element	Soil 1 (mg/L)				Soil 2 (mg/L)				
Cr	0,3*	<dl< td=""><td><dl< td=""><td><dl< td=""><td>0,3*</td><td>0.3</td><td>ND</td><td>ND</td><td>ND</td></dl<></td></dl<></td></dl<>	<dl< td=""><td><dl< td=""><td>0,3*</td><td>0.3</td><td>ND</td><td>ND</td><td>ND</td></dl<></td></dl<>	<dl< td=""><td>0,3*</td><td>0.3</td><td>ND</td><td>ND</td><td>ND</td></dl<>	0,3*	0.3	ND	ND	ND
Mn	0.3*	11.3	13.4	11.4	0.3*	25.0	60.3	40.8	46.5
Fe	48.8*	0.1	0.4	0.1	48.8*	17.3	0.2	0.4	0.6
Co	0.1*	ND	ND	ND	0.1*	0.1	0.8	0.4	0.4
Ni	0.1*	ND	ND	ND	0.1*	0.1	0.6	0.4	0.4
Cu	0.1*	0.1	ND	0.1	0.1*	ND	0.2	0.1	0.1
Zn	0.1*	2.3	0.2	ND	0.1*	14.1	11.3	7.0	6.0
Pb	0.3*	ND	ND	ND	0.3*	0.1	0.8	0.7	1.4

Table 3 – Concentration in relation to the time of percolation (one decimal)

* Raw leachate (C_o values); < DL = < Determination limit; ND = Not detected

The Cu originated from the raw leachate was totally freed by the effluent solutions. Even though the formation of the Fe and Mn hydroxides can control the immobilization of the Cu, it can form complexes with the organic matter present in the soil and become more mobile. In Figure 1, it is observed increased concentrations of COD in the effluent solutions, indicating probable removal of organic matter and Cu.

As observed in Table 1, the pH values obtained in the effluent solutions indicated acidity after the leachate percolation in Soil 2. However, in Soil 1, the effluent solutions indicated pH about the neutrality. This fact can be determined by the distinct behaviors of the soils in relation to Co, Ni and Pb. The relations between Cu, Ni, Pb, Fe, Co and Cr concentrations from effluent solutions (C) and initial concentrations from raw leachate (C_0), in relation to percolation time (Table 3) for Soils 1 and 2, are presented in Figure 2 for comparison purposes. The same relations for Zn and Mn concentrations are shown in Figure 3.





Figure 2. Leachate ratio concentration versus percolation time for Cu, Ni, Pb, Fe, Co and Cr.

Figure 3. Leachate ratio concentration versus percolation time for Zn and Mn.

There is similar behavior of desorption among Pb, Ni and Co metals up to 75 days, when Pb begins to be eliminated more intensely by Soil 2 and the Ni and Co begin to be desorbed, but under stabilization (Figure 2). Mn and Zn are desorbed by both soils (Figure 3).

The adsorption potential of Cr by the soil was also shown by Nascentes (2006) and Korf et al. (2008). According to Nascentes (2006), the Cu, Pb and Cr metals are absorved in the soil because they have a high affinity with the iron oxides present in it. Even though Soil 2 presented iron oxides in its chemical composition, according to the x-ray diffraction test, this soil was not capable of absorbing Pb (Figure 2). The Mn desorption, observed from both studied soils (Table 3), was also verified by Nascentes (2006), which highlights the importance of evaluation the mobility of the Mn in liners of compacted soils, due to its great mobility. The adsorption of Zn by Soil 1 in the last effluent solution (Table 3 and Figure 3) can be associated to the desorption of the Mn, in which sites were occupied by metals of higher affinity, like Cr, Cu, Ni and Fe and after by Zn.

Conclusion

This study shows the potential use of both tropical soils as liners on sanitary landfills for their relatively low values of permeability coefficient to water, in the orders of 10^{-9} and 10^{-10} m/s. However, with the percolation of raw leachate in both soils, increasing concentrations of COD and VFA in the effluent solutions were noticed, which makes their application in liners for sanitary landfills in need of greater studies and analysis, because their occurrence is an index of potential water course pollution. The lateritic soil was able to adsorb more metals (Pb, Zn, Ni and Co) than the non-lateritic tropical soil. Both soils adsorbed Cr and Fe and desorbed Mn and Cu.

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