HEAVY METAL DIFFUSION AND RETENTION IN A MINERAL BARRIER OF COMPACTED LATERITIC SOIL

DIFUSÃO E RETENÇÃO DE METAIS PESADOS EM BARREIRA MINERAL DE SOLO LATERÍTICO COMPACTADO

SANDRA GARCIA GABAS
Federal University of Mato Grosso do Sul, Campo Grande, MS, Brazil
E-MAIL: sandra.gabas@ufms.br

JORGE EDUARDO SOUZA SARKIS
Energy and Nuclear Research Institute, São Paulo, SP, Brazil
E-MAIL: jesarkis@ipen.br

MARIA EUGENIA GIMENEZ BOSCOV
Polytechnic School, University of Sao Paulo, São Paulo, SP, Brazil
E-MAIL: meboscov@usp.br

ABSTRACT

In tropical countries, lateritic soils are natural candidates for the construction of compacted clay liners (CCL) in waste disposal sites because of their availability and good engineering properties when compacted. Furthermore, a significant pollutant retention capacity has been observed in these soils, what may be reported to the expressive amount of constituent iron and aluminum oxides and hydroxides. This investigation aimed to assess diffusion and retention of toxic metals in a lateritic clay. Compacted specimens were submitted to diffusion tests carried out with solutions of Cd and Pb at pH values of 3.0, 5.5 and 8.0. Diffusion tests indicated diffusion coefficient of about 1×10^{-10} m²/s for both metals and showed relevant retention of both metals, lead more significantly (99% for pH 3.0, 5.5 and 8.0) than cadmium (96% for pH 3.0, 91% for pH 5.5 and 90% for pH 8.0), expressed by distribution coefficients K_d ranging from 100-300 mL/g for Cd and 500-2000 mL/g for Pb. A simplified mathematical simulation carried out with code POLLUTE indicates that, even in the case of accumulation of leachate in the bottom drainage system, a CCL constructed with this soil would perform adequately, reducing the concentration of diffused metals through the liner to less than 0.2% of the concentration in the leachate.

Keywords: diffusion, adsorption, toxic metals, tropical soils, liners

RESUMO

Os solos lateríticos são normalmente utilizados para a construção de camadas de impermeabilização de aterros (*liners*) em países tropicais devido à sua disponibilidade e às suas propriedades geotécnicas quando compactados. Além disso, há estudos que observaram sua significativa capacidade de retenção de poluentes, a qual deve estar relacionada à presença de óxidos e hidróxidos e de ferro e de alumínio. Este estudo teve como objetivo avaliar a difusão e a retenção de metais tóxicos em solo laterítico argiloso compactado. Amostras de solo compactado foram submetidas a ensaio de difusão com soluções de Cd e Pb em valores de pH de 3,0, 5,0 e 8,0. Os ensaios de difusão indicaram valores de coeficiente de difusão em torno de 10^{-10} m²/s para os dois metais e revelaram retenção elevada de cádmio e chumbo, com maior retenção de chumbo (99% para pH 3,0, 5,5 e 8,0) do que cádmio (96% para pH 3,0, 91% para pH 5,5 e 90% para pH 8,0), expressa por coeficientes de distribuição K_d variando entre 100-300 mL/g para Cd e 500-2000 mL/g para Pb. Uma simulação matemática simplificada, realizada com o programa POLLUTE, mostrou que *liners* construídos com esse solo apresentariam bom desempenho: mesmo no caso de acúmulo de lixiviado no sistema de drenagem de fundo, a concentração dos metais após difusão através da camada seriam significativamente reduzidas, a até menos do que 0,2% da concentração no lixiviado.

Palavras-chave: difusão, adsorção, metais tóxicos, solos tropicais, "liners"

1 INTRODUCTION

In the last decades, geotechnical researches directed to environmental problems have been carried out all over the world, most of them related to pollutant transport and retention, either to understand transport mechanisms in soils (Rowe 1988; Rowe et al. 1988, Shackelford 1989 e 1993, Shackelford & Daniel 1991) or to apply this knowledge in order to prevent or remediate contamination (Boscov 1997; Kersten et al. 1997; Roehl & Czurda 1998; Kugler et al. 2002; Varank et al., 2011; Syafalni et al. 2012).

The most frequent contaminants in the environment are pathogenic microorganisms, inorganic chemicals, organic chemicals and radioactive nuclides; amongst the inorganic chemicals, some toxic metals (Hg, Cd, Pb, Cr, Ni) have been continually accumulated in the environment for centuries (Aswathanarayana 1995; Lollar 2005).

Mobility of inert dissolved contaminants in porous media can be described by the advective/dispersive transport equation, which in one-dimensional form may be represented as:

$$\frac{\partial C}{\partial t} = D_l \frac{\partial^2 C}{\partial l^2} - u_l \frac{\partial C}{\partial l} \tag{1}$$

where l is the curve directional coordinate in the flow direction, u_l is the solute interstitial velocity ($u = v/n$, n is the soil porosity and v is the Darcy velocity), D_l is the hydrodynamic dispersion coefficient in the flow direction and C is the solute concentration. When interactions between contaminants and soil grains take place during the migration process, equation (1) may be expressed as:

$$R_d \frac{\partial C}{\partial t} = D_l \frac{\partial^2 C}{\partial l^2} - u_l \frac{\partial C}{\partial l} \tag{2}$$

where R_d is the retardation factor, which represents all sorption processes that lead to metal ions immobilization.

Most studies of soil contamination by metals focus on the evaluation of transport equation parameters such as diffusion coefficient and retardation factor. Metal sorption by soil particles is controlled by interactions between solid and liquid phases and involves different processes, i.e. adsorption (non-specific and specific), ion exchange reactions, complexation reactions and precipitation (Sposito 1989; McBride 1994). These differ by the type of chemical bonding and the strength of fixation of the metal to the soil particle, both important regarding long-term immobilization and remobilization.

Non-specific adsorption is considered a reversible reaction and normally takes place in permanent-charge minerals, i.e. some clay minerals, such as smectites and vermiculite. Specific adsorption is considered stronger than non-specific adsorption and occurs at variable-charge minerals surfaces, like the borders of kaolinite platelets, iron and manganese oxides and aluminum hydroxide, and organic matter. These minerals have electric charges which are pH dependent, resultant from the dissociation of hydroxyl ions (OH^-) and water molecules (H_2O^-) attached to their surfaces.

According to present environmental regulations in most countries, design of waste disposal sites include bottom liner and cover systems to prevent emissions of pollutants to the atmosphere, subsoil, surface water and groundwater. Since composite bottom liners have been recommended as the best technical solution (ISSMGE 2006), compacted clay liners (CCL) play an important role providing a smooth and resistant base for the installation of the geomembrane and a second defence line in the case of geomembrane failure; furthermore, CCLs may be the only impermeable layer in the bottom liner for specific climatic and hydro-geological conditions.

Fine-grained materials are used for CCLs to insure a sufficiently low hydraulic conductivity. In this situation the migration of contaminants occurs mainly by diffusion (Freeze & Cherry 1979; Rowe 1988; Shackelford 1989) and the transport equation can be simplified to a pure diffusion equation (no advective transport) with the form of Fick´s second law:

$$\frac{\partial C}{\partial t} = \frac{D^*}{R_d} \frac{\partial^2 C}{\partial l^2} \tag{3}$$

where D* is the diffusion coefficient. In this situation, the retardation factor and the retention mechanisms it encloses are cardinal to preview pollutant migration and to assess related risks.

The goals of this study were to evaluate lead and cadmium adsorption capacity of a compacted tropical residual clay, and to assess long-time behaviour of this soil as a bottom liner for waste disposal sites relative to metal adsorption.

## 2 MATERIAL AND EXPERIMENTAL METHODS

2.1 Soil

A residual lateritic clay of diabase from the State of Sao Paulo (southeast of Brazil) was investigated. Lateritic clays cover large extensions of the country and are often used as clay liners for landfill sites as well as in dam and pavement construction due to their good geotechnical properties when compacted, such as high shear strength, low compressibility, low expansibility in the presence of water, and low hydraulic conductivity. Samples were collected in the top 50 cm of a homogeneous, deep and extensive layer in the outskirts of the densely populated and highly industrialized city of Campinas.

The physicochemical characterization of the soil was determined by the following standard methods: ASTM D4972 for pH determination, Walkley-Black Method (Walkey and Black, 1934) for organic matter, Ion exchange resin (Rodella & Alcarde, 1994) for cation exchange capacity, point of zero charge from zero charge from titulometric method as exposed by Hemsi (2001), ASTM D422 for grain-size distribution, ASTM D4318 for liquid limit, plasticity limit and plasticity index, ASTM D698 for optimum water content and maximum dry unit weight.

2.2 Diffusion tests

Diffusion tests were carried out with compacted soil specimens in order to evaluate metal migration through a CCL and to provide controlled contamination for the posterior investigation of metal speciation in soil minerals. Samples collected in the field were homogenized, compacted at optimum water content with Proctor energy inside the diffusion cells, saturated with distilled water by capillarity, and finally topped with metal solutions. A schematic representation of the diffusion test is presented in Figure 1.
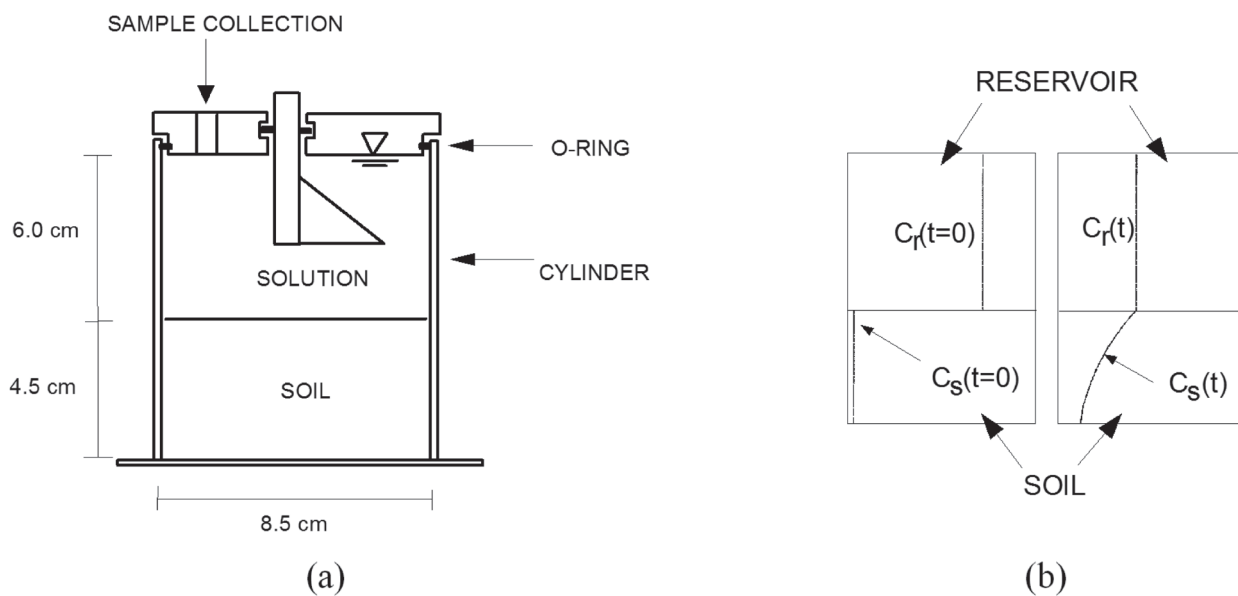
Figure 1 - Diffusion test: (a) assemblage; (b) initial and final concentrations in the reservoir and in the soil pore water.

Tests were carried out in a temperature controlled $(22\pm2)^{\circ}$C room. During diffusion (90 days), samples of the reservoir solution were collected for element determination. In the end of the tests, cells were dissembled, and soil specimens were extruded and cut into five slices of equal thickness. The soil slices were squeezed under a pressure of 25MPa in order to obtain the interstitial solution for chemical analysis. Metal adsorption in the top slice of the soil specimens was also evaluated using a sequential extraction procedure.

Cadmium (Cd) and lead (Pb) were chosen for the investigation because of their different mobility in soils. Concentration of test solutions was 3 mg.L^{-1} for Cd and 100 mg.L^{-1} for Pb, values based on the limits prescribed by the environmental protection agency of the State of Sao Paulo. For each metal, three solutions were prepared at pH values of 3.0, 5.5 and 8.0, which were adjusted by the addition of nitric acid and ammonia hydroxide.

Diffusion blank tests were also carried out with solutions at the same pH values without metal concentration in order to evaluate metal migration from the soil to the reservoir solution. Additional blank tests were executed to monitor pH along diffusion time.

Natural soil samples spiked with pollutants were used in this investigation to guarantee higher control of boundary conditions and mass balance; furthermore, the process of contaminating the soil by means of diffusion tests also provides the evaluation of transport parameters in structured specimens similar to in-situ conditions of compacted clay liners.

2.3 Evaluation of techniques for the study of adsorption in soil particles

Mineral separation and analytic techniques that allow punctual chemical microanalysis of soil particles were investigated in order to assess the possibility of a direct verification of metal retention.

The feasibility of X-ray spectrometry by energy dispersion (EDX) associated to scanning electron microscopy was evaluated to study metal fixation in soil particles. Both techniques consist, generically, of the incidence of an electron beam in a point of the material to be analyzed, exciting its electrons which then emit characteristic X-rays. These X-rays can be measured by a detector by means of energy dispersion or wave length; therefore, it is possible to identify the elements present in the material and under some circumstances even carry out punctual quantitative analyses. Electron microscopes allow visualization at micrometric to nanometric scales (McLaren 1991, Goldstein et al. 2003).

Application of at least 20kV-voltage is necessary for the chemical microanalyses. The incidence of the electron beam generates in the material an excitation energy bulb of approximately 2 μm (Goldstein et al. 2003). The large range of soil particles sizes, from more than 4.8 mm to less than 0.002 mm, may limit the utilization of these techniques. On one hand, electron microscopy allows the individualized visualization of soil particles; on the other hand, the size of the excitation energy bulb may exceed the dimension of a soil particle. The reply to the incidence of the electron beam in the microanalysis spectrum may not necessarily correspond to the surface of a clay mineral, but to the whole grain or even to more than one particle. As adsorption involves superficial electrical charges and ions, the observation scale ranges from a few nanometres to angstroms (e.g. ionic radii of Cd^{2+} and Pb^{2+} are, respectively, 0.075 and 0,119 nm), therefore not compatible to the investigated techniques. This was confirmed by Gonçalves et al. (2004), which identified Cu, Zn and Pb in contaminated sediments by means of chemical analysis and sequential extraction, but detected only Cu and Zn by EDX spectrometry without recognizing the location of these metals, whether adsorved by clay or metallic particles.

Separation techniques based on the physical-chemical properties of the mineral constituents were investigated as well. According to the soil composition (Tables 1 and 2), densimetric and electromagnetic separations were selected. For the densimetric separation, an aliquot of the soil clay fraction was suspended in water in order to separate particles of equal size and different densities (2.6 for kaolinite and from 4.4 to 5.2 for iron oxides, according to Parfenoff et al. (1970). Mineralogical analysis of the separated phases showed quantities of clay minerals in the "oxide" phase and of oxides in the "clay" phase.

For the electromagnetic separation, two aliquots, bulk sample and clay fraction, were submitted to an electromagnetic separator Frantz at 0.5 A, in order to separate the magnetic fraction (oxides) from the non-magnetic fraction (clay minerals). X-ray difractometry of both fractions showed the presence of oxides in the clay fraction.

These preliminary tests and the literature indicated that the sequential extraction procedure is the best technique, however indirect, to evaluate metal retention in the different mineralogical fractions of the soil.

2.4 Chemical analyses

Chemical analyses of cadmium, lead, iron and aluminum were carried out by induced coupled plasma atomic emission spectrometry (ICP-AES) (Atom Comp Series 800, Jarrel-Ash Division, USA) and induced coupled plasma mass spectrometry (ICP-MS) (Element, Finnigan MAT, Bremen, Germany).

Reservoir samples collected in the first fifteen days of diffusion were analyzed by ICP-AES due to the initial high concentrations, and henceforth by ICP-MS.

2.5 X-ray difractometry and microscopy

Mineralogical analysis of specimens of diffusion tests were carried out by means of X-ray difractometry and scanning electron microscopy with punctual spectrometry by energy dispersion in order to identity possible alterations in the soil structure and metal precipitation as crystals not present in the original mineralogy of the soil. The equipments utilized for these analyses were a Philips difractometer model MPD 1880 with copper tube, operated at 2.5 to 70°, and a Leo Microscope model Stereoscan 440, operating at 20kV and 5nA, silicon detector with coupled EDX X-ray spectrometer.

2.6 Simulation of pollutant transport through a landfill bottom liner

Simulations of metal migration through a CCL were carried out by means of program POLLUTE v.6.3.3 (Rowe et al. 1994) in order to assess the maximum concentration at the base of the CCL and the time necessary to achieve it, as presented by Rowe & Booker (1985). Two values of CCL thickness (0.6 m and 1.0 m) and permeability coefficient (1×10^{-9} m/s and 1×10^{-8} m/s) were investigated. CCL thickness values correspond to those requested by the environmental protection agency of the State of Sao Paulo for bottom liners of, respectively, municipal solid wastes (MSW) and hazardous wastes landfills. Hydraulic conductivity equal or lower than 1×10^{-9} m/s is mandatory in most environmental prescriptions and regulations; however, a value tenfold higher was also investigated to account for field variability in the compaction process and for the characteristic high sensibility of dry unit weight and permeability of compacted lateritic soils to small deviations in the moulding water content (Nogami & Villibor 1995, Boscov 1997).

Diffusion and distribution coefficients were determined by means of the diffusion tests. Additional input parameters for the numerical analyses e.g. porosity and dry unit weight were obtained from geotechnical laboratory tests.

3 RESULTS

3.1 Soil characterization

Physicochemical characterization (i.e. Atterberg limits, grain size distribution and mineralogy) and chemical properties and of the studied soil are summarized in Tables 1 and 2, respectively.

Table 1 - Physicochemical characterization of the soil.

Property		Result	Method
pH (Distilled water)		5.3	ASTM D4972
Organic matter content (%)		2.5	Walkley-Black Method (CAES, 2006)
Cation exchange capacity ($mmol_c$/kg)		56.6	Ion exchange resin (Rodella and Alcarde, 1994)
Point of zero charge (Hemsi 2001)		4.3	Titulometric method
Grain-size distribution (%)	Coarse sand	0	ASTM D422
	Medium sand	6	
	Fine sand	27	
	Silt (%)	14	
	Clay (%)	59	
Liquid limit, w_L (%)		45	ASTM D4318
Plasticity limit, w_P (%)		30	ASTM D4318
Plasticity index, PI (%)		15	ASTM D4318
Skempton activity, A		0.25	A = PI/C (C = clay content)
Optimum water content (%)		26.3	ASTM D698
Maximum dry unit weight (kN/m^3)		15.7	ASTM D698
Compacted hydraulic conductivity (m/s) (optimum water content, Proctor energy)		1×10^{-9}	Permanent-head flexible-wall permeability test
Mineralogical composition		Quartz, ilmenite, hematite, magnetite, gibbsite and traces of vermiculite	X-ray difractometry Magnetic treatment 0.5A Glycol+heating at 490oC+ FeO treatment

Table 2 - Chemical characterization of the soil by X-ray fluorescence.

Element	(%)	DL	Element	($\mu g.g^{-1}$)	DL
SiO_2	31.1	0.01	Ba	74	17
Al_2O_3	23.9	0.01	Ce	71	18
MnO	0.141	0.002	Cl	<15	15
MgO	0.17	0.01	Co	98	3
CaO	0.09	0.01	Cr	285	2
Na_2O	< 0.02	0.02	Cu	187	2
K_2O	0.03	0.01	F	< 223	223
TiO_2	5.21	0.007	Ga	35	1
P_2O_5	0.14	0.003	La	18	14
Fe_2O_3	24.4	0.01	Nb	33	1
		0.01	Nd	32	22
			Ni	104	2
			Pb	25	5
			Rb	3	2
			S	267	32
			Sc	50	4
			Sr	10	1
			Th	13	3
			U	4	3
			V	900	3
			Y	16	1
Loss of ignition	14.2		Zn	88	1
Total	99.5		Zr	383	3

L – detection limit

3.2 Diffusion tests

Blank diffusion tests showed that pH solution does not remain constant during the experiment (Figure 2), tending to the natural soil pH with time, and that there is metal diffusion from the soil to the reservoir solution. Maximum concentrations of cadmium and lead in the reservoir after 90 days of reverse diffusion are shown in Table 3. Cadmium was not detected in the natural soil by X-ray fluorescence, i.e. its concentration in the soil grains was bellow the detection limit of the analytical technique; however, the quantity of cadmium that migrated from the soil by diffusion was enough to significantly increase the concentration in the reservoir, tenfold at pH 5.5 and 8.0, and ~100 times at pH 3.0. On the other

hand, lead was detected in the soil, however reverse diffusion resulted in a slight increase in the reservoir concentration.
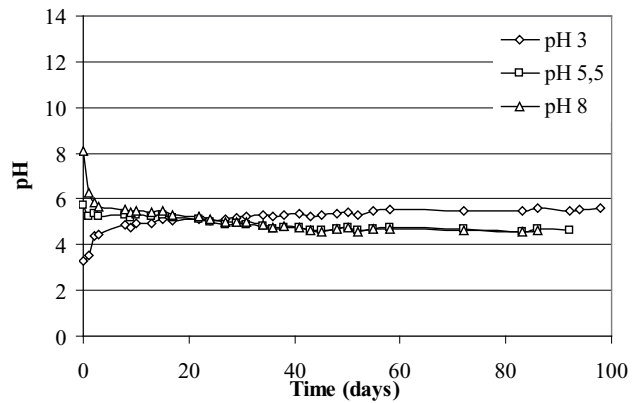
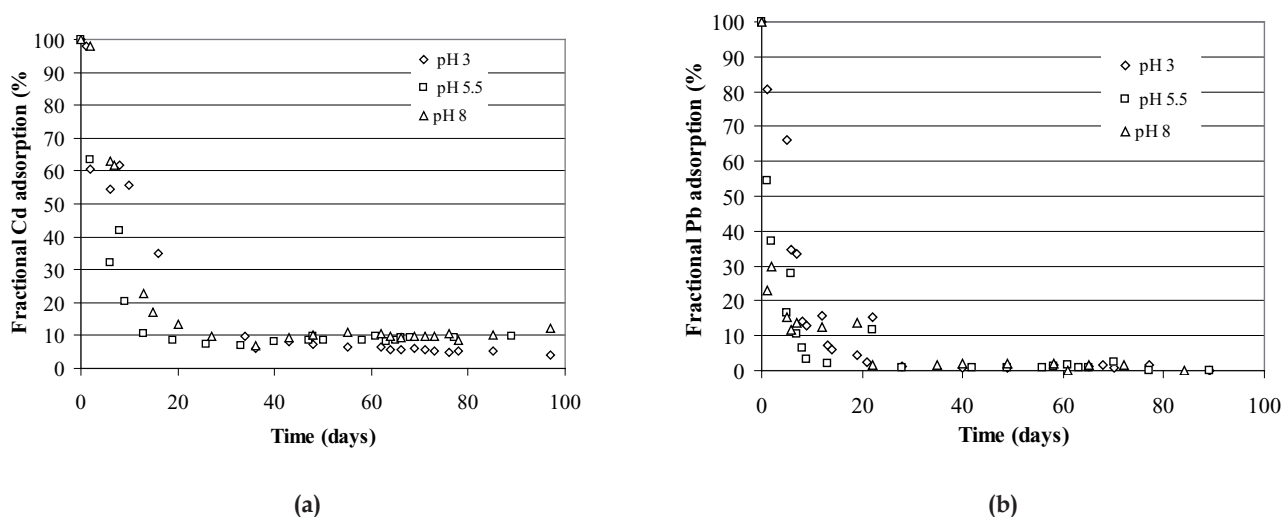**Figure 2** - pH of reservoir solutions along diffusion tests.

Table 3 - Metal concentrations in the reverse diffusion tests ($\mu g.L^{-1}$): reservoir solution and soil pore water.

Metal	Cadmium			Lead		
pH	3.26	5.75	8.09	3.26	5.75	8.09
Reservoir - Initial	0.002	0.001	0.001	0.024	0.002	0.003
Reservoir - Final	0.22	0.013	0.010	0.77	0.024	0.004
Layer 1 (Top)	0.01	0.00	-	0.03	0.17	0.11
Layer 2	0.03	0.01	0.19	0.06	0.27	0.04
Layer 3	0.02	0.03	0.06	0.09	0.19	0.17
Layer 4	0.03	0.04	0.09	0.03	0.04	0.13
Layer 5 (Bottom)	0.06	0.03	0.07	0.07	0.20	0.08

In the diffusion tests with metallic solutions, final reservoir concentrations were very low, indicating an expressive diffusion process occurring during the 90 days of the tests (Figure 3). This mass transfer mechanism is clearly pH dependent for cadmium: the more acidic the solution, the more significant was the diffusion (Figure 3a). This trend is not so clear for lead diffusion (Figure 3b).

Metal concentrations in the reservoir along time and in the soil pore water in the end of the tests are indicated in Table 4. Initial cadmium concentrations in the reservoir were approximately 3 $\mu g.L^{-1}$, whereas after 90 days of diffusion values of 0.12, 0.29 and 0.37 $\mu g.L^{-1}$ were measured,

respectively, for pH 3.0, 5.5 and 8.0. Cadmium that migrated from the reservoir to the soil specimen was partly retained by the soil grains and partly diffused downwards through the specimen. Concentrations in the soil pore water indicate the pollutant front entering the soil, with higher values in the top layer and a background value of approximately 0.10 $\mu g.L^{-1}$ at the bottom layer. Initial lead concentrations in the reservoir were about 100 $\mu g.L^{-1}$, decreasing to near 0.14 $\mu g.L^{-1}$ in the end of the test. The pollutant migration front in the soil specimens is also evident by the observation of the concentrations in the soil pore water of each layer.

(a)

(b)

Figure 3 - Diffusion from the reservoir to the soil at different pH values: a) cadmium; b) lead.

Table 4 - Metal concentrations in the direct diffusion tests ($\mu g.L^{-1}$): reservoir solution and soil pore water.

Metal	Cadmium			Lead		
pH	2.83	5.65	8.02	2.98	5.73	7.90
Reservoir - Initial	3.12	2.95	3.37	112	101	98.2
Reservoir - Final	0.12	0.29	0.37	0.14	0.14	0.13
Layer 1 (Top)	23.1	33.2	134	0.61	9.30	5.60
Layer 2	1.29	1.46	4.44	0.27	1.00	1.22
Layer 3	0.26	0.22	0.95	0.23	0.60	0.78
Layer 4	0.04	0.11	0.50	0.16	0.68	0.60
Layer 5 (Bottom)	0.10	0.15	0.09	0.10	0.24	0.27

Data were analyzed by means of software POLLUTE v.6 (Rowe et al. 1994). Adopted boundary conditions were: (i) finite solute mass over the top surface of the soil, corresponding to a volume of solution applied to the reservoir in the beginning of the test without posterior refilling; and (ii) zero solute flux at the bottom, representing the impermeable base of the diffusion cell.

Several values of the parameters D and K_d were tested to fit simultaneously the two curves generated by the experimental data in each test (i.e. concentration in the reservoir along time and concentration in the soil pore water along depth in the end of the test). The free solution diffusion coefficient at 25°C was adopted as the superior limit value for D, which corresponds to 7.17×10^{-10} m²/s and 9.45×10^{-10} m²/s for cadmium and lead, respectively. In all tests, a good fitting of both curves with the same parameters was not achievable; therefore, suitable ranges of values were adopted for each test and are presented in Table 5. A possible explanation could be that kinetics of metal adsorption is not negligible in tropical soils, since specific adsorption may be time dependent, whereas the model considers instantaneous non-specific adsorption. This possibility has already been mentioned by Stuermer et al. (2008). Another cause may be the occurrence of membrane behavior, which has already been observed in lateritic clays (Musso 2008). Nevertheless, some experimental data from adsorption batch tests by iron hydroxides were well fitted by a double-layer surface complexation model (Meng et al. 2014); the hydroxides in this case were not collected from a natural environment, but prepared in the laboratory under controlled conditions.

Table 5 - Parameters obtained from diffusion tests.

pH	Element	D^* $(m^2.s^{-1})$	K_d $(mg.L^{-1})$	R_d
3.0	Cd	1×10^{-10}	200-300	684-1025
5.5	Cd	1×10^{-10}	250	854
8.0	Cd	1×10^{-10}	100-200	342-684
3.0	Pb	1×10^{-10}	2000	6827
5.5	Pb	1×10^{-10}	700	2390
8.0	Pb	1×10^{-10}	500	1708

Diffusion coefficients resulted in approximately 1×10^{-10} m^2/s for both metals. These values are comparable with values in literature of 3×10^{-10} m^2/s to 9×10^{-10} m^2/s presented by Shackelford and Daniel (1991) for the diffusion of Cd in a kaolinitic clayey soil. Tsugawa (2004) obtained a diffusion coefficient for Cd of 0.7×10^{-10} m^2/s for a lateritic clay. Stuermer et al. (2008) determined values between 0.5×10^{-10} m^2/s and 4×10^{-10} m^2/s for Cd and Pb in a saprolitic silty soil.

Distribution coefficient (K_d) and retardation factor (R_d) values for both metals (Table 5) indicate that cadmium was less adsorbed than lead. Lower values of K_d and R_d for both metals at pH 8.0 could be explained by the fact that a significant amount of the metals may be hydrolyzed, therefore were not available for diffusion and adsorption.

The ratio of retardation factors for cadmium and lead at 3.0 pH in this research resulted in 1:10, whereas a ratio of 1:5 was obtained by Charlemyanont et al. (2009) for a lateritic soil through batch tests.

3.3 X-ray diffraction and microscopy tests

SEM techniques were applied to the natural soil and samples contaminated by lead and cadmium in the diffusion tests. Backscattering electron images from these samples do not show any evidence of cadmium and lead adsorbed to the surface particle (Figure 4); furthermore, EDX spectra do not reveal the presence of these metals (Figure 5). Cadmium and lead were not detected possibly due to the low concentrations of metal adsorption (Table 3) and detection limits of this technique.
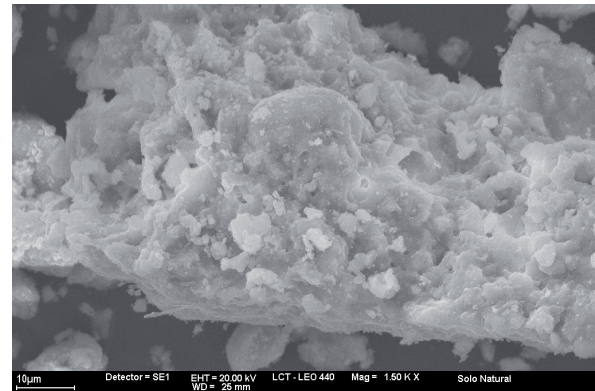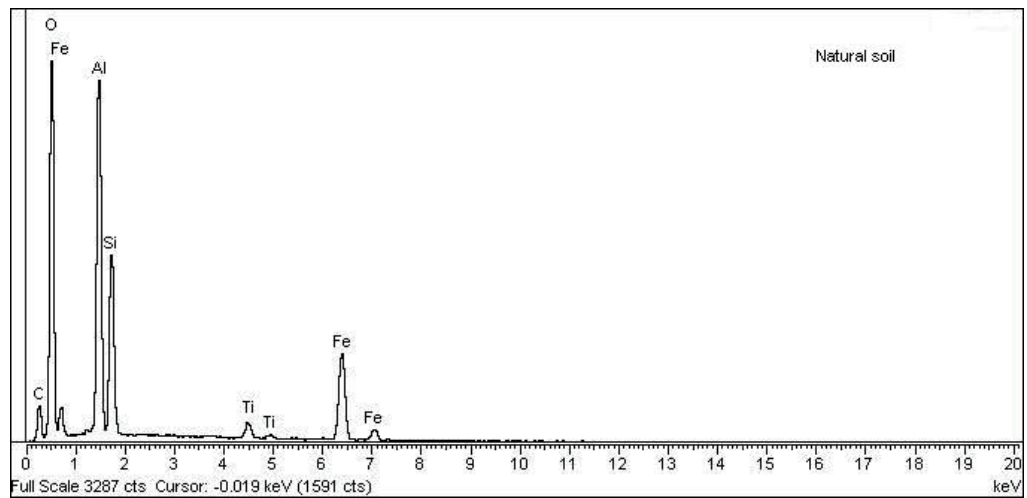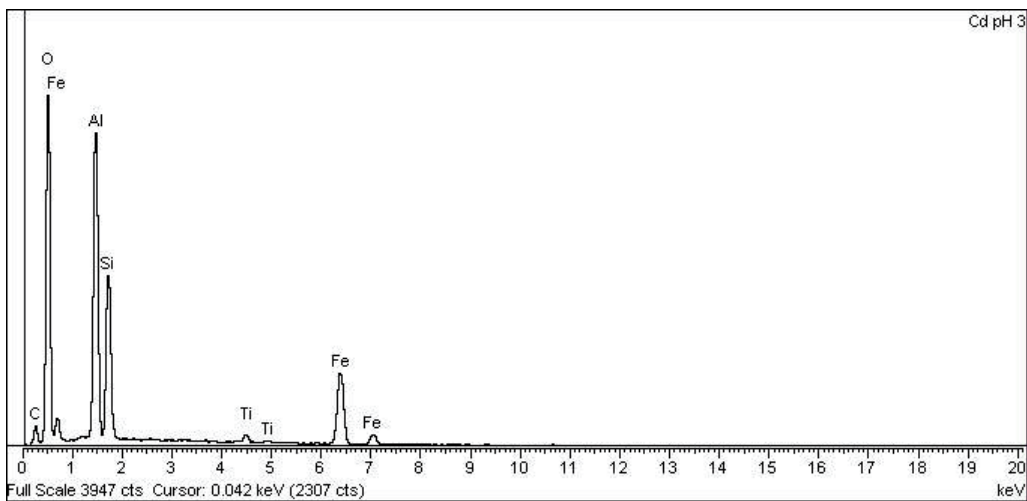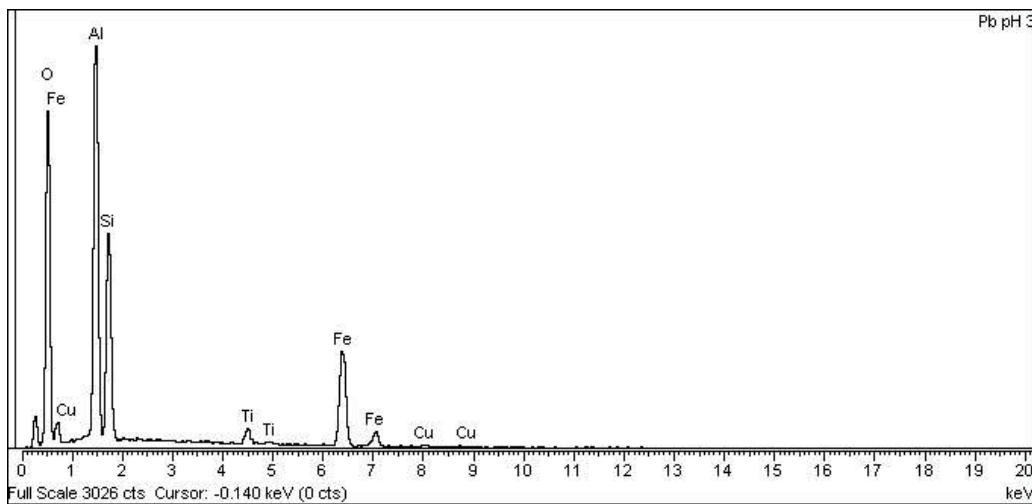

(a)
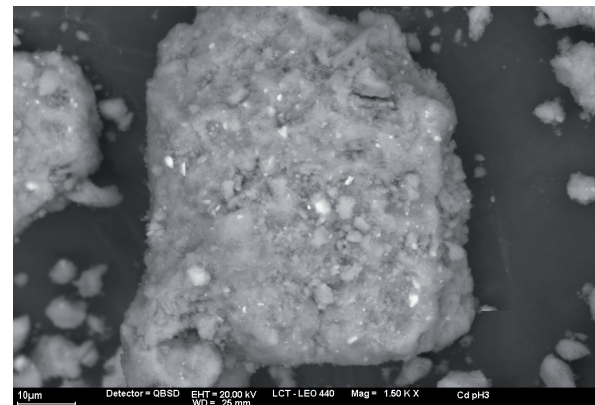
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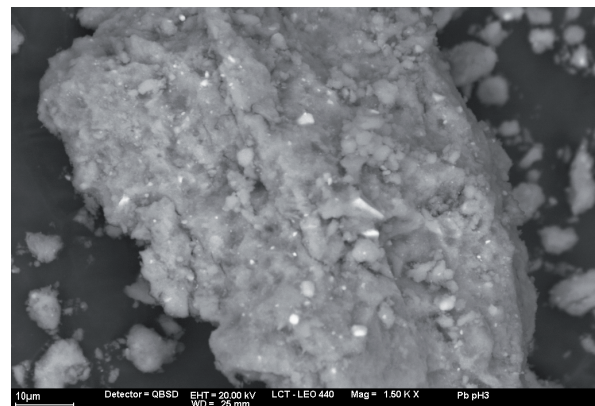
(c)

Figure 4 - Scanning electron images (SEM) from samples: a) natural soil (secondary electron image); b) cadmium diffusion test and c) lead diffusion test (backscattering electron images).

(a)

(b)

(c)

Figure 5 - Electron X-ray diffraction (EDX) spectrum from samples: a) natural soil; b) cadmium diffusion test and c) lead diffusion test.

3.4 Simulation of pollutant transport through a landfill bottom liner

Simulation results indicate that the permeability coefficient is the most significant parameter in the maximum pollutant concentration at the bottom of the CCL (Figure 6) when compared to diffusion (D*) and distribution (K_d) coefficients for the adopted range of parameter values. The maximum base concentration, considering zero adsorption and liner thickness of 0.6-1.0 m, varied from 2-2,5%

and 6.4-6.8% of the initial concentration applied at the top of the liner, respectively, for permeability coefficients of 1.10^{-9} and 1.10^{-8} m/s. For K_d and D* equal to, respectively, 10 mL/g and 1.10^{-10} m²/s, metal concentration at the bottom of CCL is 0.2% of the applied top concentration for a permeability coefficient of 1.10^{-9} m/s and 1.8% for 1.10^{-8} m/s. For the range of K_d values determined by the experiments, the maximum base concentration would be lower than 0.2% of the top concentration even for a 0.6m-thick CCL with a permeability coefficient of 1.10^{-8} m/s (Figure 7).
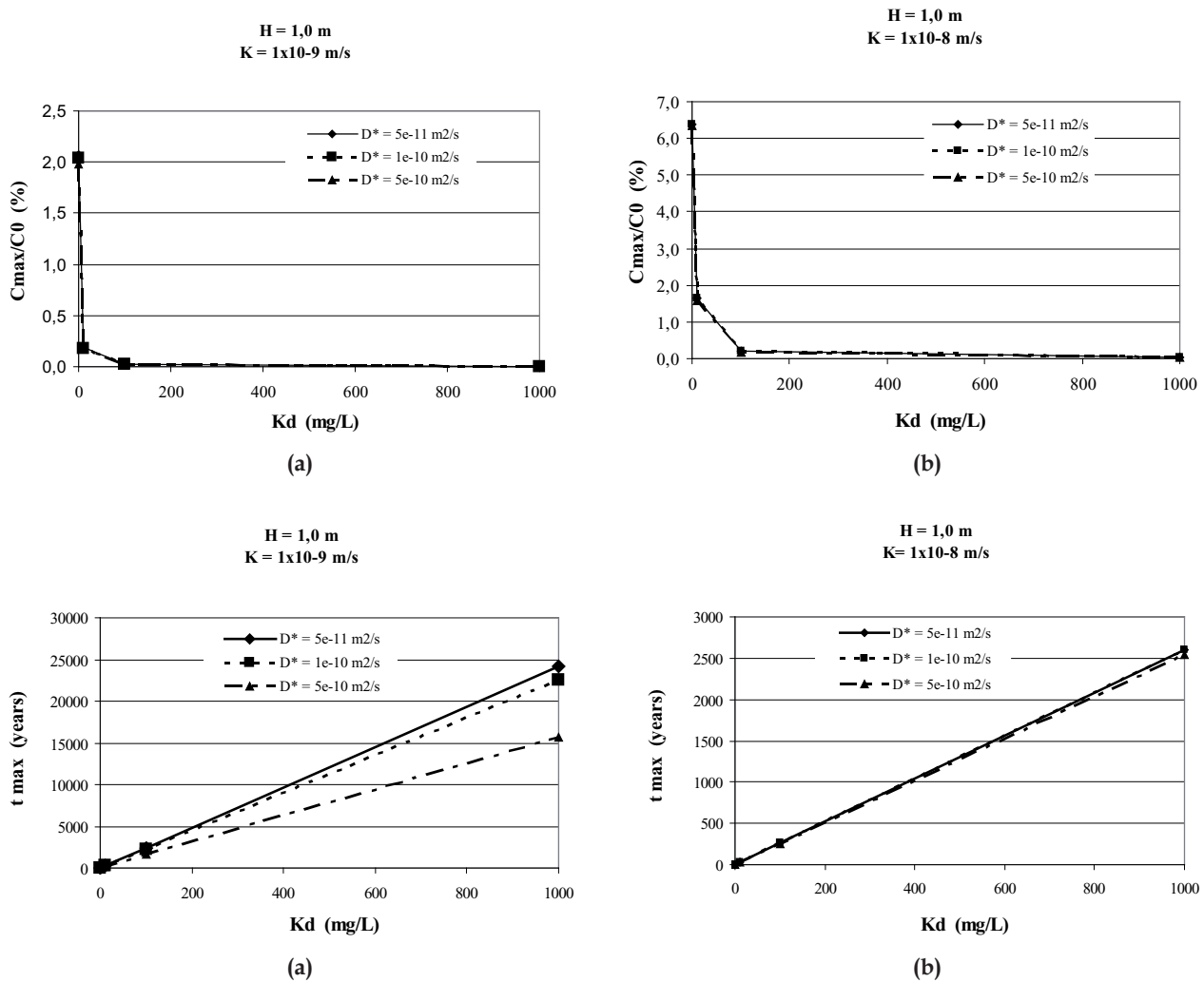

Figure 6 - Simulation of pollutant transport through a landfill bottom liner with 1.0 m thickness.

The distribution coefficient (K_d) is the main parameter involved in the time necessary to achieve the maximum pollutant concentration at the bottom of the CCL (Figure 15). Long periods are necessary to achieve the maximum concentration

at the bottom of a CCL; for instance, for a 0.6 m thickness almost 10,000 years would be necessary, indicating the effective heavy metal retention by a liner constructed with the lateritic soil.
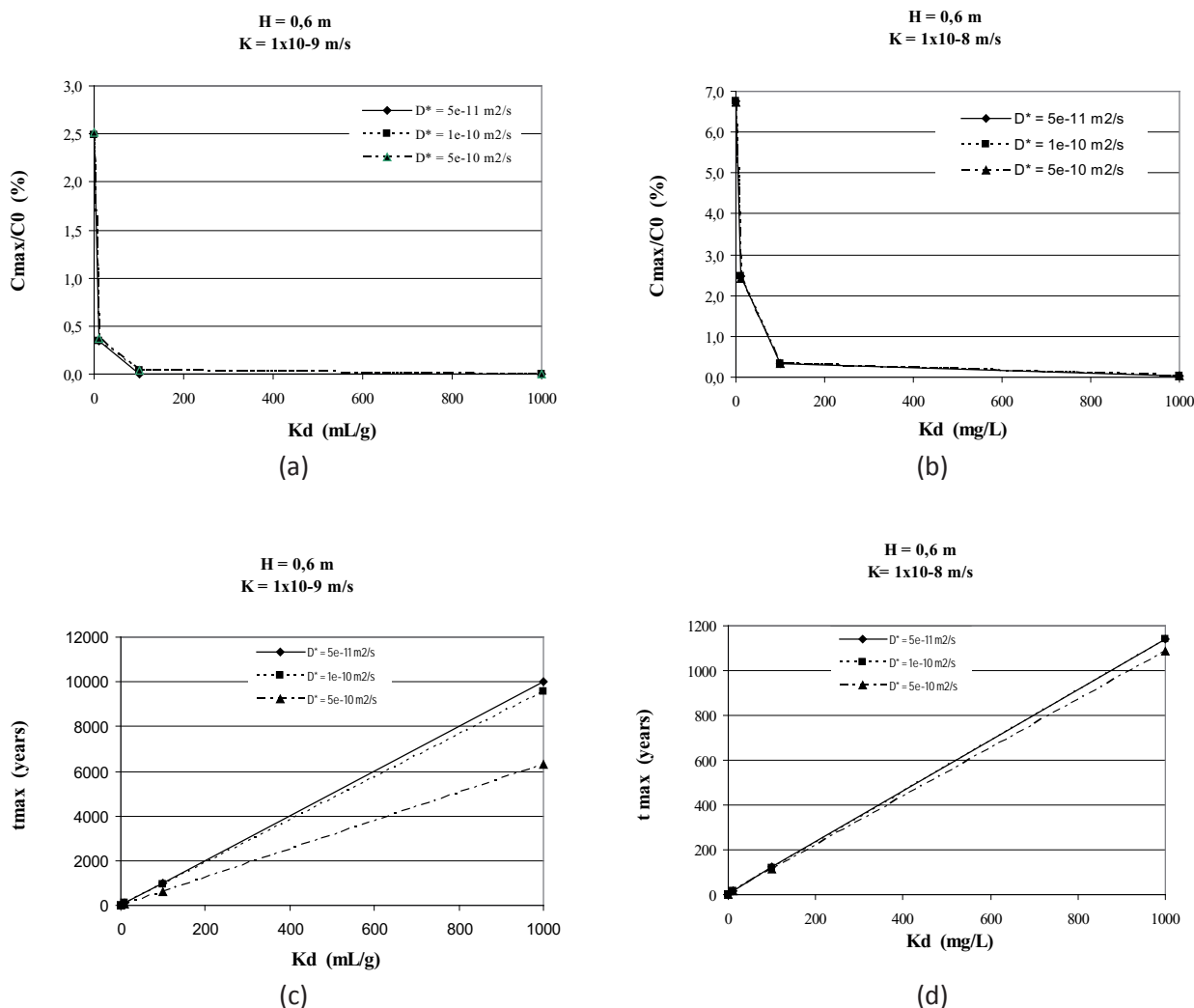
H = 0,6 m
K = 1x10-9 m/s

(a)

H = 0,6 m
K = 1x10-8 m/s

(b)

H = 0,6 m
K = 1x10-9 m/s

(c)

H = 0,6 m
K= 1x10-8 m/s

(d)

Figure 7 - Simulation of pollutant transport through a landfill bottom liner with 0.6 m thickness.

Syafalni et al. (2012) performed the treatment of landfill leachate via a coagulation-flocculation purification methodology using lateritic soil and noted its efficiency to remove 65.7% of the chemical oxygen demand (COD) and 81.8% of the turbidity. On the other hand, Varank et al. (2011), comparing the performance of four alternative liner systems in the migration of various organic and inorganic contaminants (heavy metals), including a non lateritic clay liner, concluded that leachate contaminants may reach the groundwater in trace concentrations.

4 CONCLUSIONS

Metal migration and retention through the soil is pH dependent and pH does not remain constant along the diffusion test. For pH 8.0,

the effect of hydrolysis limited the possibility of diffusion and adsorption, probably resulting in the formation and precipitation of metal salts and bases which however could not be detected by the EDX spectrometer.

Diffusion coefficients were estimated as $1x10^{-10}$ m²/s for cadmium and lead at the tested pH values.

Cadmium and lead were intensively adsorved in the lateritic soil, resulting in high retardation factors ($R_d > 100$).

Simulations of metal transport through 0.6-m clay liners constructed with this soil indicate that both metals would be practically immobile inside the soil layer, as maximum concentrations lower than 0.2% of the concentration applied at the top of the soil layer will reach the bottom of the layer after 2,000 years. However, the pollutant migration

model did not consider different bonding strengths and the effect of time on adsorption, so simulations must still be regarded as qualitative for lateritic soils, which are constituted by a high content of variable-charge minerals.

According to the results, we consider that compacted lateritic clay liners could, therefore, act as satisfactory mineral barriers for this type of contaminants, presenting different efficiency for different pollutants.

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