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## Trace elements in Ultisol irrigated with landfill leachate and public-supply water

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### Key words:

liquid waste  
agricultural use  
heavy metals

### ABSTRACT

Landfill leachates have considerable amounts of organic matter and mineral nutrients and, when applied in environmentally adequate amounts, show fertilizing potential for the soil-plant system. This study aimed to analyze the effect of landfill leachate (LFL) and the public-supply water (PSW) depths on the contents of trace elements in an Ultisol. This work was carried out from April 21 to August 21, 2012, in split-split-plot scheme; LFL and PSW depths (T1 - 0 mm of LFL and 1067 mm of PSW; T2 - 60 mm of LFL and 1067 mm of PSW; T3 - 120 mm of LFL and 1067 of PSW, T4 - 180 mm of LFL and 1067 of PSW, and T5 - 240 mm of LFL and 1067 mm of PSW) in the plots, the analyzed layers (0-0.2 m and 0.2-0.4 m) in the subplots and the sampling times (0, 30, 60, 90 and 120 days) in the sub-subplots. Soil samples were collected every 30 days for 120 days to characterize the trace elements Fe, Mn, Pb, Cd, Zn, Cu and Ni. There was leaching of Pb, Cd and Ni in the Ultisol, while the T2 treatment was the most recommended to prevent contamination of groundwater.

### Palavras-chave:

resíduo líquido  
aproveitamento agrícola  
metais pesados

## Elemento traços em um argissolo irrigado com percolado de aterro e água de abastecimento público

### RESUMO

Os percolados de aterros sanitários possuem aportes apreciáveis de matéria orgânica e nutrientes minerais e, quando aplicados em quantidades ambientalmente adequadas, revelam fertilizante potencial para o sistema solo-planta. Objetivou-se analisar o efeito de lâminas de percolado de aterro sanitário (LFL) e água de abastecimento público (PSW) nos teores de elementos traços em um argissolo. O experimento foi montado no período de 21/04 de 2012 a 21/08 de 2012, em esquema de parcelas subdivididas tendo, nas parcelas, as lâminas de LFL e PSW (T1 - 0 mm de LFL e 1067 mm de PSW; T2 - 60 mm de LFL e 1067 mm de PSW; T3 - 120 mm de LFL e 1067 de PSW; T4 - 180 mm de LFL e 1067 de PSW; e T5 - 240 mm de LFL e 1067 mm de PSW); nas subparcelas as profundidades analisadas (0 a 0,2 m e 0,2 a 0,4 m) e nas subsubparcelas os tempos de amostragem (0, 30, 60, 90 e 120 dias). Amostras de solo foram coletadas a cada 30 dias durante 120 dias para caracterização dos elementos traço Fe, Mn, Pb, Cd, Zn, Cu e Ni. Houve lixiviação de Pb, Cd e Ni no argissolo enquanto o tratamento T2 foi o mais recomendado para evitar a contaminação do lençol freático.



## INTRODUCTION

Landfill leachate is a liquid waste that is difficult to be treated and contains various pollutants classified as inorganic macro-components, heavy metals, priority pollutants, xenobiotic organic compounds and microorganisms (França & Ruaro, 2009; Castilhos Júnior et al., 2010; Dacanal & Beal, 2010; Queiroz et al., 2011; Zhang et al., 2013).

According to Zou et al. (2013), the heavy metals Cu, Pb, Cd, Cr, Zn, Mn and As are detected in leachates from urban solid waste at worrying concentrations. Gonçalves Júnior et al. (2015) warn about the risks of accumulation, at levels toxic to plants, of the heavy metals Cu, Zn, Fe, Mn, Cd, Pb and Cr present in solid wastes.

In the survey conducted by Silva et al. (2005) about the types of soil in the Brazilian semiarid region, there was a predominance of Latosols, Argisols, Quartzarenic and Litholic Neosols, Plansols and Vertisols. In this context, Oliveira et al. (2010) analyzed the sorption of Cu, Cr, Zn, Cd, Pb and Ni in five soil classes and observed that Argisol promoted the highest retention of the heavy metals and, consequently, lower contamination of groundwater.

Water scarcity, the pollutant potential of the leachate and the predominance of Argisol in the Brazilian semiarid region are aspects that stimulate the production of grasses in landfill cells, aiming the production of biomass for energetic purposes and the minimization of deforestations in the CPSWtinga biome, as reported by Coelho et al. (2015a, b).

Given the above, this study aimed to analyze the effect of the application of landfill leachate and public-supply water depths on the contents of trace elements of an Argisol in the semiarid region.

## MATERIAL E METHODS

This study was carried out from April 21 to August 21, 2012, at the Experimental Unit of Water Reuse (UERA) of the Federal Rural University of the Semiarid Region (UFERSA), in Mossoró, RN, Brazil (5° 11' 31" S; 37° 20' 40" W). According to Köppen's classification, the climate of the region is BSw<sub>h</sub> (Alvares et al., 2013).

The soil in the experimental area was classified as eutrophic Red Yellow Argisol (EMBRAPA, 2013), which has an impermeable layer from the depth of 0.4 m. Tables 1 and 2 show the physical and chemical characteristics of the soil in the experimental area, before the application of landfill leachate (LFL), determined using the methodology of Donagema et al. (2011).

Table 1. Initial physical characteristics of the soil used in the experiment

Layer (m)	Soil density kg dm <sup>-3</sup>	Sand	Silt	Clay	Moisture (%) - MPa	
					0.01	1.5
0-0.20	1.61	0.86	0.10	0.05	11.43	3.67
0.20-0.40	1.65	0.71	0.07	0.23	21.47	14.17

The leachate used in the experimental tests of the present study was collected in a Class II landfill (non-inert wastes) of Mossoró, RN. This landfill receives, every day, 150 tons of urban solid waste, generating a mean of 50 L h<sup>-1</sup> of leachate. The leachate was transported from the landfill to the UERA/UFERSA using a 1.0 m<sup>3</sup> impermeable tank, while the water used in the experiment came from the public supply system of the Water and Sewerage Company of Rio Grande do Norte State (CAERN).

The chemical characteristics of the LFL and PSW were monthly determined, from May to August 2012 (Table 3), according to the norms of the Standard Methods for the Examination of Water and Wastewater.

LFL applications were performed every two days, before irrigation with PSW, in order to favor the leaching of the trace elements along the soil profile. The LFL was measured in a 1 L graduated cylinder and applied using a watering can, homogeneously, on the soil surface of each experimental plot.

For the application of the PSW, an experimental bench was built, which consisted of the following components: a) 16-m<sup>3</sup> concrete tank; b) 0.25-hp motor pump; c) 130-µm disc filter; d) main line of PVC tube, with length of 24 m and nominal diameter of 32 mm; e) derivation line of PVC, with length of 12 m and nominal diameter of 50 mm; and f) 24 lateral lines spaced by 0.50 m, with pressure-compensating emitters with normal flow rate of 1.6 L h<sup>-1</sup> and spacing of 0.30 m between emitters.

The drip irrigation set occupied an area of 38.44 m<sup>2</sup>, which had 25 experimental plots. Each experimental plot had dimensions of 1.0 x 1.0 x 0.30 m between plots, where elephant grass (*Pennisetum purpureum* Schum.) was cultivated. Water demand was estimated based on reference evapotranspiration (ET<sub>o</sub>), through the Penman-Monteith equation.

The daily depths of the treatments used in the experimental tests were as follows: a) T1 – Only PSW; b) T2 – 1 mm of LFL and PSW; c) T3 – 2 mm of LFL and PSW; T4 – 3 mm of LFL and PSW; and T5 – 4 mm of LFL and PSW, adapted from the study of Matos et al. (2013). The sums of the LFL and PSW depths, along the experimental period, are shown in Table 4.

The five plots subjected to the treatment T<sub>1</sub> did not receive any basal fertilization, while the others received only the supply of nutrients present in the LFL, especially N, P and K,

Table 2. Initial chemical characteristics of the soil used in the experiment

Layer (m)	pH	EC dS m <sup>-1</sup>	OM dag kg <sup>-1</sup>	N g kg <sup>-1</sup>	P	K <sup>+</sup>	Na <sup>+</sup>	Ca <sup>2+</sup>	Mg <sup>2+</sup>	Al <sup>3+</sup>	H + Al	SB	CEC	V	M	ESP
0-0.20	6.6	0.12	0.68	0.54	328.24	104.45	13.64	2.09	0.79	0.00	0.28	3.21	3.49	92.67	0.00	1.77
0.20-0.40	6.6	0.13	0.57	0.62	275.48	121.88	9.28	2.46	1.17	0.00	0.30	3.98	4.28	93.95	0.00	0.90
			Fe	Mn	Cu	Zn	Ni	Pb	Cd							
0-0.20			97.58	88.21	0.52	6.13	0.07	0.27	0.01							
0.20-0.40			126.72	90.56	0.46	3.77	0.13	0.29	0.01							

EC – Electrical conductivity of the soil saturation extract; OM – Organic matter; H + Al – Potential acidity; SB – Sum of bases; CEC – Cation exchange capacity; V – Effective cation exchange capacity; M – Aluminum saturation; ESP – Exchangeable sodium percentage

Table 3. Mean values and standard deviations of pH and trace elements of the landfill leachate (LFL) and public-supply water (PSW) of four samplings performed from May to August 2012

Fluids	pH	Fe	Mn	Pb	Cd	Zn	Cu	Ni
		(mg L <sup>-1</sup> )						
LFL	7.45 ± 0.45	5.073 ± 0.790	3.655 ± 6.803	2.806 ± 1.071	0.133 ± 0.097	1.131 ± 0.880	0.100 ± 0.112	2.076 ± 0.709
PSW	7.47 ± 0.36	0.265 ± 0.018	0.009 ± 0.005	0.186 ± 0.085	0.003 ± 0.001	0.052 ± 0.003	0.012 ± 0.011	0.118 ± 0.027

Table 4. Depths of the treatments with landfill leachate (LFL) and public-supply water (PSW) used in the experimental tests

Treatment	Depth (mm)		
	LFL	PSW	LFL + PSW
T <sub>1</sub>	0	1067	1067
T <sub>2</sub>	60	1067	1127
T <sub>3</sub>	120	1067	1187
T <sub>4</sub>	180	1067	1247
T <sub>5</sub>	240	1067	1307

with mean values of 592 and 4.90 mg L<sup>-1</sup> and 58.55 mmol<sub>c</sub> L<sup>-1</sup>, respectively.

For the evaluation of trace elements in the soil, samples were collected every 30 days during 120 days, in the layers of 0.0-0.2 and 0.2-0.4 m, based on the study of Alcântara et al. (2011), who performed soil sampling in a common landfill. This sampling used a Dutch auger, and two single samples were collected to form a composite sample, in each layer and for each experimental plot, always close to the dripper and the elephant grass.

Later, the samples were stored in sterile plastic bags, which were labeled and sent to the Laboratory of Analysis of Soil, Water and Plant (LASAP) of the UFERSA, where the contents of iron (Fe), manganese (Mn), lead (Pb), cadmium (Cd), zinc (Zn), copper (Cu) and nickel (Ni) were quantified through atomic absorption spectrophotometry, as proposed by Donagema et al. (2011).

The experiment was set in a randomized block design with five replicates, in a split-split-plot scheme, evaluating LFL and PSW depths (T<sub>1</sub>, T<sub>2</sub>, T<sub>3</sub>, T<sub>4</sub> and T<sub>5</sub>) in the plots, soil layers (0.0-0.2 m and 0.2-0.4 m) in the subplots and sampling periods (0, 30, 60, 90 and 120 days) in the sub-subplots, following the methodology proposed by Silva et al. (2011).

The contents of trace elements were subjected to analysis of variance by F test at 0.05 probability level. Statistical analyses were performed using the computational program Sisvar 5.1 Build 72 (Ferreira, 2011).

Table 5. Summary of the analysis of variance for the variables Fe, Mn, Pb, Cd, Zn, Cu and Ni

Source of variation	Degrees of freedom	Mean square						
		Fe	Mn	Pb	Cd	Zn	Cu	Ni
Blocks	4	1434.51	7627.99	0.79	0.003	44.52	0.58	0.006
Treatments (T)	4	147.42 <sup>ns</sup>	1454.39 <sup>ns</sup>	0.02*	0.00007*	27.05 <sup>ns</sup>	0.32 <sup>ns</sup>	0.0007 <sup>ns</sup>
Residuals (a)	16	509.30	632.73	0.006	0.00002	68.40	0.32	0.0007
Layer (L)	1	2251.14*	78400.40**	0.03 <sup>ns</sup>	0.00005 <sup>ns</sup>	1379.31**	0.69 <sup>ns</sup>	0.04**
L x T	4	173.84 <sup>ns</sup>	409.01 <sup>ns</sup>	0.006 <sup>ns</sup>	0.00004 <sup>ns</sup>	16.66 <sup>ns</sup>	0.24 <sup>ns</sup>	0.0008 <sup>ns</sup>
Residual (b)	16	376.54	288.12	0.01	0.00006	12.97	0.30	0.0007
Sampling period (SP)	4	63842.58**	12233.05**	4.43**	0.06**	33.11 <sup>ns</sup>	2.28**	0.47**
SP x T	16	193.16 <sup>ns</sup>	481.38 <sup>ns</sup>	0.01 <sup>ns</sup>	0.00003 <sup>ns</sup>	26.57*	0.31 <sup>ns</sup>	0.0010 <sup>ns</sup>
SP x L	4	2109.73**	5611.98*	0.02 <sup>ns</sup>	0.0001 <sup>ns</sup>	25.90 <sup>ns</sup>	0.25 <sup>ns</sup>	0.009**
SP x L x T	16	99.29 <sup>ns</sup>	430.52 <sup>ns</sup>	0.006 <sup>ns</sup>	0.00004 <sup>ns</sup>	9.57 <sup>ns</sup>	0.33 <sup>ns</sup>	0.001 <sup>ns</sup>
Residual (c)	164	493.76	2160.69	0.03	0.0001	15.94	0.32	0.001
CV plot (%)		46	31	15	14	182	159	21
CV subplot (%)		40	21	21	24	79	152	21
CV sub-subplot (%)		46	57	30	39	88	159	29

Note: \*\*, \* and <sup>ns</sup>, F significant at 0.01 and 0.05 probability levels and not significant at 0.05 probability level

## RESULTS AND DISCUSSION

There was no statistical difference of the interaction sampling period (SP) x layer (L) x treatment (T) for any of the studied variables; however, T and L showed isolated significant effects on the contents of Pb and Cd and of Fe, Mn, Zn and Ni, respectively (Table 5). These statistical differences are related to the leaching of trace elements, as well as to the values of organic matter, cation exchange capacity, hydrogen potential and clay percentage, which are different along the soil profile (Tables 1 and 2), interfering with the sorption and desorption of the trace elements.

As to the behavior of Fe along the soil profile (Figure 1A), the highest values were observed in the layer of 0.0-0.2 m, for all treatments, among which T<sub>5</sub> showed the highest values. The increment of this element in the soil layers of the treatments T<sub>2</sub> and T<sub>5</sub> is due to the presence of Fe in the LFL (Table 3); however, its sorption in the surface layer is associated with the greater content of organic matter (Table 2).

These results are much lower than the range from 1350 to 6728618 mg dm<sup>-3</sup> presented by Fortunato et al. (2012) for mangrove areas intended for disposal of residues from port activities for more than 50 years.

According to Figure 1B, the contents of Mn decreased along the soil profile for the applied treatments. In all treatments, there were similar behaviors and the highest contents of this element were observed in the layer of 0.0-0.2 m, which can be justified by the increment of Mn in the soil due to LFL application and Mn sorption due to the greater content of organic matter of the surface layer (Table 2).

These results corroborate those found by Oliveira et al. (2014), who evidenced slight increase in Mn contents in the surface layer of a Cambisol irrigated with proportions of domestic wastewater and well water, besides the tendency of reduction of these values along the soil depth.

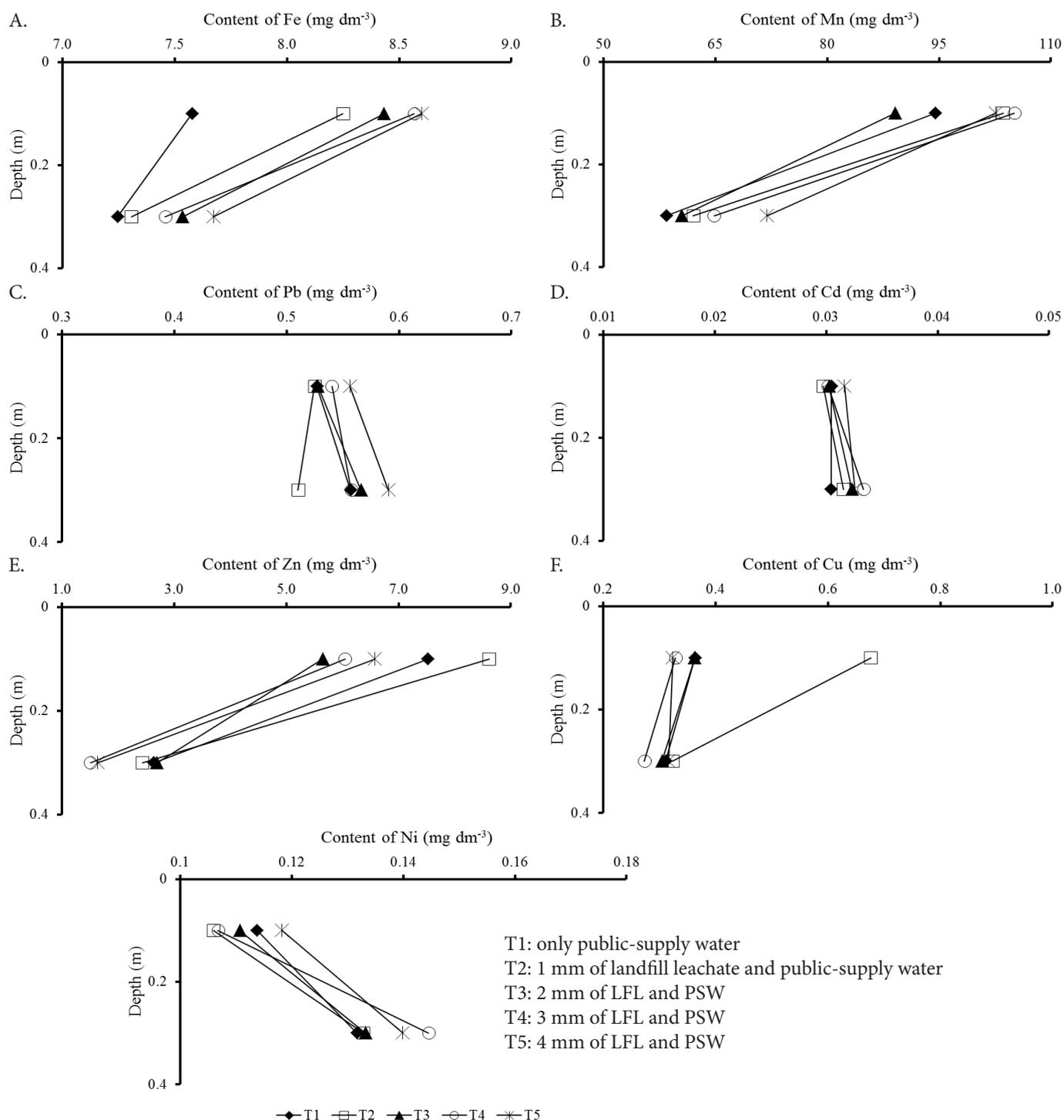


Figure 1. Behavior of Fe, Mn, Pb, Cd, Zn, Cu and Ni as a function of soil depth and treatments

There was effect of the treatments on the increase of the contents of Pb, along the soil profile (Figure 1C), except in T2, in which the contents of Pb slightly decreased, indicating that there was leaching of this element. This fact is attributed to the decrease in the values of cation exchange capacity and clay percentage, along the soil profile. Since the values of these characteristics are lower in the surface layer, the exchange complex of the clays is saturated by the Pb from the LFL, thus occurring desorption.

Although the treatments have similar behaviors, T5 showed higher Pb contents in the soil profile, while T2 stood out from the others in the minimization of the environmental risks. Such results were much lower than the values of 5.76 and 6.53 mg dm<sup>-3</sup> obtained by Alcântara et al. (2011) in the

layers of 0-0.2 and 0.2-0.4 m, respectively, of a dystrophic Yellow Argisol used as common landfill of urban solid waste. According to the CONAMA resolution n° 420/2009 (Brasil, 2009), the contents of Pb observed in the studied soil after LFL application are not worrying, because they are much lower than the threshold value of 72 mg dm<sup>-3</sup> established for the prevention of toxicity.

There was significant effect of the treatments on the increments of the contents of Cd (Figure 1D); however, there was higher leaching of this element in T4, due to the decrease in the values of cation exchange capacity and clay percentage along the soil profile (Tables 1 and 2). These results do not corroborate those reported by Alcântara et al. (2011) for a

dystrophic Yellow Argisol used as common landfill of urban solid waste, because Cd was not detected in the layers of 0-0.2 and 0.2-0.4 m. According to the CONAMA resolution n° 420/2009 (Brasil, 2009), the contents of Cd observed in the studied soil after LFL application do not represent environmental risk, because they are below the level of 1.3 mg dm<sup>-3</sup>, established for the prevention of toxicity.

The contents of Zn reduced along the soil profile for the applied treatments (Figure 1E). All treatments showed similar behaviors and the highest contents of this element were occurred in the layer of 0.0-0.2 m, due to the sorption performed by the greater content of organic matter (Table 2). Always when the highest LFL depths were applied to the soil, lower contents of Zn were observed in both studied layers.

These values were much lower than the range from 34.71 to 650.90 mg dm<sup>-3</sup> reported by Fortunato et al. (2012), who studied mangrove areas used as areas of disposal of port activity residues. In addition, the values of Zn observed in the studied soil after the application of LFL are not worrying, since they are inferior to the limit of 300 mg dm<sup>-3</sup> for the prevention of toxicity (Brasil, 2009).

Cu contents were significantly altered by the time of application of the depths; however, there is a tendency of accumulation of this element in the surface layer, especially in T2 (Figure 1F), due to the sorption of the organic matter, which is present in larger amounts in the surface layer (Table 2). Comparing the results of the present study with those of Fortunato et al. (2012), Cu contents were much lower than the range of 12.89 to 828.67 mg dm<sup>-3</sup> observed in mangroves contaminated with port wastes. According to the CONAMA resolution n° 420/2009 (Brasil, 2009), the Cu contents observed in the studied soil after the application of LFL do not represent environmental risk, because they are much lower than the value of 60 mg dm<sup>-3</sup>, established for the prevention of toxicity.

Ni contents increased along the soil profile for all applied treatments (Figure 1G), demonstrating that there was leaching of this element. Therefore, attention must be paid, because it may cause pollution of groundwaters.

Ni leaching was favored by the lower values of cation exchange capacity and clay percentage of the surface layer. The Ni present in the LFL saturated the exchange complex of the clays, promoting higher desorption. On the other hand, the higher values of cation exchange capacity and clay percentage in the layer of 0.2-0.4 m resulted in greater sorption of this element. Although the treatments showed similar behaviors, higher mean Ni content in T4 in the layer of 0.2-0.4 m.

These results differ from those presented by Alcântara et al. (2011) in an Argisol of common landfill, who observed Ni contents of 1.62 and 2.33 mg dm<sup>-3</sup> in the layers of 0-0.2 and 0.2-0.4 m, respectively. According to the CONAMA resolution n° 420/2009 (Brasil, 2009), the Ni contents observed in the studied soil after LFL application are not worrying, because bare lower than the limit of 30 mg dm<sup>-3</sup>, recommended for the prevention of toxicity.

## CONCLUSIONS

1. Due to the higher sorption, there was accumulation of Fe, Mn, Zn and Cu in the surface layer of the Argisol, while

the decrease in the values of cation exchange capacity and clay percentage along the soil profile favored the desorption and, consequently, leaching of Pb, Cd and Ni.

2. There was no significant effect of the interaction of leachate and public-supply water depths, sampling layer and sampling period on the alteration of the contents of the studied trace elements.

3. The depth corresponding to 60 mm of leachate and 1067 mm of public-supply water was the most recommended for preventing the accumulation of trace elements in the soil and contamination of the water table.

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