

# Assessment of Contaminated Soils by Heavy Metals in Municipal Solid Waste Landfills in southern Brazil

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*Abstract* – The paper presents an evaluation of the environmental impact of inadequate solid waste disposal in Municipal Solid Waste landfills in southern Brazil. It was found that inorganic contamination can reach critical levels according to the reference values recommended by the Brazilian environmental protection agencies. The physical-chemical parameters (granulometric analysis, pH, CEC, TOC and heavy metal concentrations) and mineralogical analyses were assessed for the potential mechanisms of retention and migration of contaminants in surface soil and water in the studied area. The relative abundance of the reactive mineral phases are the primary controllers of sorption processes in soils, and plays a critical role in the regulation of contaminant fate and transport. Furthermore, phytoremediation experiments were conducted with *Impatiens walleriana* Hook. f., a flower species well adapted to the Brazilian climate. *I. walleriana* was effective in removing metals from contaminated soils and showed bioaccumulation of Cu, Zn, Cr and Ni. The remediation program proposed here is inexpensive as this species adapts itself easily.

*Key -words*: Municipal solid waste landfills, surface soil, heavy metals, contamination, phytoremediation.

## 1 Introduction

In Brazil, most solid wastes are disposed in municipal landfills. Due to the inavailability of appropriate areas and the difficulties associated with technologies and management strategies, both related to financial problems, many of the municipal leaders have neglected environmental and social problems related to solid wastes. Like many others, the Municipal Solid Waste landfill in the city of Novo Hamburgo in southern Brazil, has an environmental passive related to the inadequate disposal of solid wastes. Solutions are not simple due to scarce financial resources for implementing well-known and effective techniques for remediation. The inadequate disposal of solid waste is recognized as one of the most serious problems of our times. The effects of dumping wastes into the environment are incalculable and include water scarcity, water contamination, and an increase in environmental pollution related diseases [1,2,3,4,5].

This paper presents a preliminary evaluation of the environmental impact of inadequate solid waste disposal at the MSW landfills, and shows that the inorganic contamination in soils and water by some heavy metals may be critical when considering the references values recommended by the Brazilian environmental protection agencies.

The bulk mineralogical assemblage of the soil in the landfill area was evaluated for an approach to

predict natural attenuation process and contaminants behavior in surface and subsurface environments. Natural attenuation phenomena related to transport of contaminants involve physical, chemical and biological processes able to reduce the contaminant concentrations and could be an effective remediation strategy [6]. The transport and fate of contaminants in soils and groundwater are highly coupled to the nature and relative abundance of the reactive mineral phases that are the primary controllers of sorption processes in soils, thus serving as important regulators of contaminant transport. Major challenges in understanding the processes controlling contaminant behavior in the environment include the complexity of the soil and aquifer matrix [7].

Furthermore, the implementation of a phytoremediation program is being considered. A study with the *Impatiens walleriana* Hook. f., a flower species well adapted to the Brazilian climate, was carried out. Despite being a Balsaminaceae of African origin, it is easily grown and is present in urban gardens and natural forests. The remediation process is usually chosen according to the best mechanism suited to improve the environment quality. Phytoremediation, botanical-bioremediation or green-remediation are used in cases where plant species represent the main remediation mechanisms or are required to start the process [8]. Phytoremediation can be used to remedy contaminated waters and soils in sites with dispersed

contamination and low contaminant concentration [9,10,11,12]. The contaminant concentrations must be within the tolerance limits of the species used. For the bioremediation process previously chosen species are planted *in situ* to promote phytoextraction process. After the process is complete the plants are removed resulting in an environment free from toxic substances or less contaminated. The final disposal of the remaining biomass depends on the cultivated species. In the phytoremediation mechanism the plants that accumulate high metal concentrations usually have a slow growth, low biomass content and shallow radicular system [8]. Phytoremediation has usually been used for removal of heavy metals in contaminated sites using several varieties of species, for example, *Echinochloa crysgalli* and *Abutilon avicennae*, [13], but can also be used on soils with high concentration of soluble salts and chloride, sodium, magnesium, calcium and potassium ions [14] and nitrates through biomass production [15].

The remediation program proposed here is very inexpensive as this plant species adapts itself easily.

## 2 Characterization of the landfill area

The studied area is located in the city of Novo Hamburgo, 80 km from Porto Alegre (Fig. 1a), with approximately 250,000 people and a generation of 180 tons/day of municipal solid wastes. The MSW landfill is located in an area of 10 hectares, and comprises of a central sorting unit, a composting unit, a final disposal system and an effluent treatment unit (Fig. 1b).

The surrounding area has well-developed vegetation of the Atlantic Forest ecosystem, as well as cultivated species, e.g. *Eucalyptus* sp. Geologically, correspond to the contact areas of first tholeiitic basaltic floods of the volcanic pile of the Paraná-Etendeka Province (Early Cretaceous) that overlies unconformably the eolian sandstone rocks of the Botucatu Formation (Late Jurassic) [16]. The *in situ* soil developed from the volcanic parent material corresponds to red-yellow Latosols relating a gently wavy landscape.

In the landfill area, the most of untreated effluents flow into small drainage streams, particularly in the rainy season, flowing then into the Guaíba River basin, which providing the water for the metropolitan region of Porto Alegre.

The activities of the final disposal unit were halted in 2005 due to problems of structural and geotechnical instability in the central part and intense erosion on the final coerture system. Since

then, the MSW landfill has been used as a transfer station, where a small fraction of the waste is sorted and the remainder is disposed temporarily on the ground surface (Fig. 1b). This site is inappropriate for the disposal of MSW because there is no protection for the waterproofing basal layer, which allows the free infiltration of contaminants into the soil and subsoil, mainly during rainstorms.

## 3 Materials and Methods

Samples were collected throughout the soil profiles at the solid waste disposal site (Fig. 2), up to approximately one (01) meter deep. Liquid samples corresponding to the shallow groundwater table were collected at the bottom of the profile and leachate samples were collected at the edges of the waste pile. The size of the soil particles was analysed to determinate the sand, silt and clay content. Mineral assemblage was examined by X-ray diffraction (XRD) on random powder and with oriented preparation. The data were obtained using a SIEMENS D5000 diffractometer equipped with Fe-filtered Co K $\alpha$  radiation, 40 kV and 40nA (UFRGS). Bulk soil samples were first gently ground and separated into <2 $\mu$ m fractions by sedimentation and centrifugation. Clay minerals were identified from the <2 $\mu$ m clay fractions. Air drying (AD), ethylene glycol solvates (EG) and heat treatments (500°C) were carried out for each sample.

Chemical analyses of the bulk soil were performed in the Soil and Water Laboratory of the School of Agronomy (UFRGS). The heavy metal (Cu, Cr, Zn, Cd, Ni, Pb) content were determined by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), using the USEPA 3050b method for environmentally available metals [17]. Hg was determined by EPA 7471; Total Organic Carbon (TOC) by wet combustion/Walkey Black/ 0.01%; total phosphorus Mehlich Methods/ 1 mg/dm<sup>3</sup>, total nitrogen (TKN) Kjeldahl / 0.01 % and chlorides by molecular absorption spectrometry. Duplicates of all samples were used. Analyses of liquid samples were performed according to the Standard Methods for Examination of Water and Wastewater (APHA, 1995) [18].

The experimental study for the phytoremediation program consisted in growing *I. walleriana* for 75 days in two different soil substrata: one corresponding to the top soil (S1) and the other 30 cm deep (S2). *I. walleriana* was the chosen species because of its wide occurrence

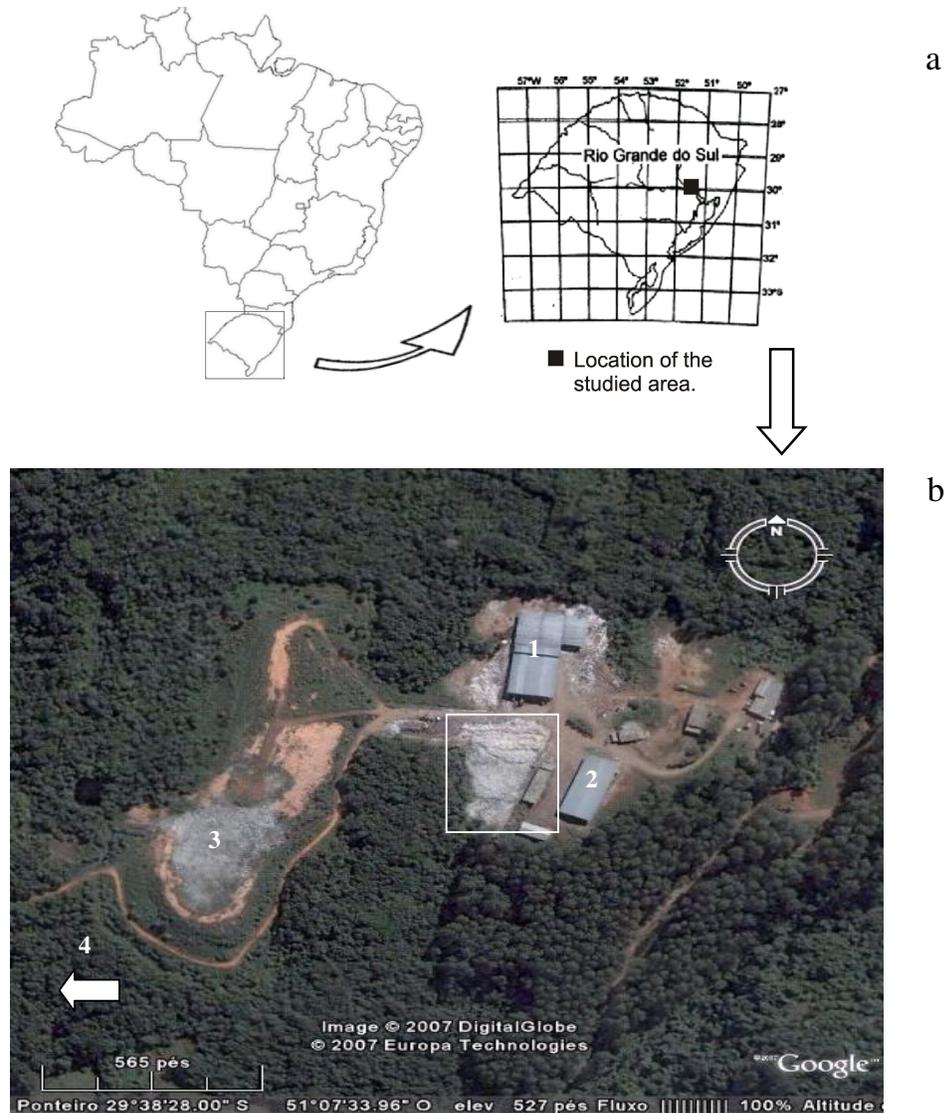


Figure 1. a) Map of the location of the studied area, Rio Grande do Sul State, Brazil; b) MSW landfill composed of 1. Central sorting unit, 2. Composting unit, 3. Final disposal system and, 4. Effluent treatment unit. Square = inadequate solid waste disposal site.

around the MSW landfill. The metal content and the physical-chemical parameters of *I. walleriana* biomass were also determined before and after the experimental procedures.

## 4 Results

### 2.1 Characterization of the surface soils

Tables 1 and 2 show the values of metal concentrations, total organic carbon (TOC), cation exchange capacity (CEC), fine sediment content (< 63 $\mu$ m) and pH of the analyzed samples. The soils

present typical texture, from sandy loam to loam, with 42% to 54% clay and silt, and low content of organic carbon (maximum 1%), which varies with depth. The reduced percentage of organic carbon may be the result of the sandy texture. The pH values for the samples vary little, between 6.1 and 6.3, thus establishing moderately acidic pH conditions.



Figure 2. MSW disposed directly on surface soils.

Table 1. Fine sediment content (&lt; 63µm), cation exchange capacity – CEC (cmolc/kg) and pH.

Parameters	Sample 02 (0 cm)	Sample 04 (30 cm)	Sample 05 (60 cm)	Sample 06 (100 cm)
< 63 µm (%)	46.05	42.07	48.88	54.10
CEC	20	21	20	22
CEC's pH	5.9	6.1	6.1	6.2

Table 2. pH, Total Organic Carbon (T.O.C %) and heavy metal concentrations (mg/kg) in soil samples.

Parameters	Sample 01 (0cm)	Sample 02 (10 cm)	Sample 03 (30 cm)	Sample 04 (40 cm)	Sample 05 (60 cm)	Sample 06 (100 cm)	Prevention limits*
pH	6.1	6.1	6.2	6.3	6.3	6.3	--
T.O.C.	0.56	0.2	1.1	0.46	0.18	0.3	--
Pb	15	20	12	15	18	17	72
Cu	116	120	127	132	126	109	60
Cr total	116	113	120	105	147	111	75
Cd	0.2	0.2	0.2	0.2	0.2	0.2	1.3
Ni	87	97	88	91	104	81	30
Zn	91	72	85	85	71	69	300
Hg	0.07	0.02	0.02	0.01	0.02	0.02	0.5

\* Maximum Values of Reference (prevention limits) recommended for soils [19].

The lowest pH values are found in the top of the soil profile. The total mineralogical constitution of the soil determined through XRD is feldspar, quartz,

iron oxy-hydroxide (hematite) and clay minerals. The clay minerals, determined at the <2µm fraction, are predominantly kaolinite (Fig. 3).

Concerning the distribution of metals throughout

the soil, a slightly decreased concentration of Zn, Ni, Cr and Cu was observed from the top to the bottom of the profile, while Pb shows little variation (Fig. 4). Cu and Zn showed greater concentration at the intermediate levels of the profile, between 30 cm and 60 cm, and Ni and Cr at around 60 cm, precisely at the levels where the soil presents an increased content of fine particles. The metal content in the soil, compared to the reference values recommended by the Brazilian environmental protection agencies [14], show that Cu, Cr and mainly Ni are above the *prevention* limits (Fig. 5). Concentrations of a particular substance above the prevention values may be harmful to the quality of the soil. These values must be used to regulate the release of substances into the soil and, if surpassed, the impacts resulting from the introduction of pollutants must be monitored [19].

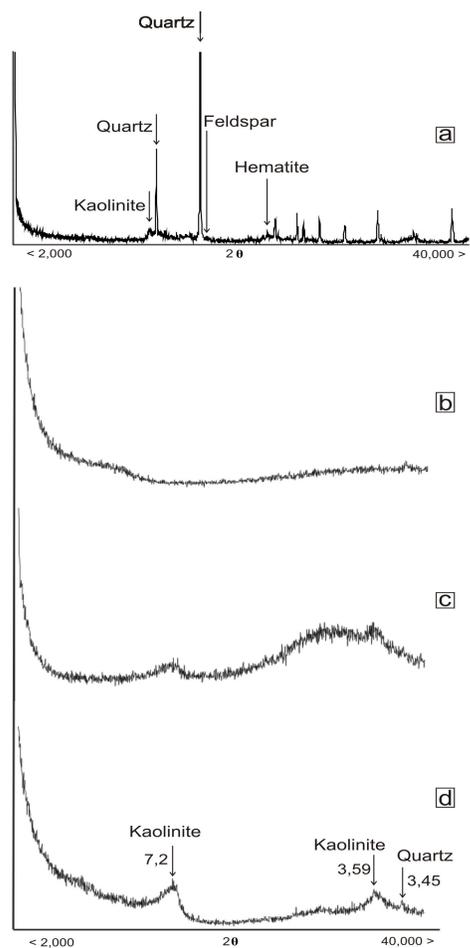


Figure 3. XRD patterns of mineral contents and clay minerals (<2 $\mu$ m fraction). a) bulk sample, b) heat treatments (500°C), c) ethylene glycol solvates, d) air drying.

Soil and sediment are the mediums in which the metals appear in larger concentrations, indicating retention of these trace elements at the site. The soil properties that affect the retention and mobility of heavy metals are primarily pH, cation exchange capacity (CEC), organic matter and ion competition [20,21,22,23,24]. Depending on the pH, some metals show a greater or lesser solubility, which may lead to precipitation or not. Thus the moderately acidic pH conditions of the studied soil would not favor the formation and precipitation of heavy metals in the form of hydroxycations, which usually occur at pH values above 7.0 [25,26,27].

The low CEC presented by the soil (~20 cmolc/kg) indicate the presence of mineral phases of low activity, i.e., with a predominance of minerals, such as kaolinite, Fe and Al oxy-hydroxides [25,28]. Therefore, the total CEC values of soils reflect the variable contributions of organic matter, clay minerals and Fe, Al and Mn oxy-hydroxides. The CEC is highest in soils where clay minerals with a 2:1 layer type such as smectites (80-150 meq/100g) are predominant but lower with clay minerals with a 1:1 layer type such as kaolinite (3-15 meq/100g) [28, 29]. The CEC of oxy-hydroxides is low (2-6 meq/100g).

According to their physical-chemical and mineralogical characteristics, the soils can have a great capacity to retain ions and compounds through adsorption and complexation reactions on particles surfaces. Organic matter, Fe and Mn oxides and clay minerals are able to form complexes and adsorb several metals, due to the surface charge of these materials [25,28]. Surface reactive phases of soils and aquifers, comprised of phyllosilicate and metal oxohydroxide minerals along with humic substances, play a critical role in the regulation of contaminant fate and transport [7].

The soil particles can present variable negative and positive surface charges, depending on the pH conditions. The kaolinite presents strong pH dependences for sorption and indicates that the surface charge results from reactions in the sorbent-solution interface [28]. The surface reactive functional groups (amphoteric sites) having acid/base properties could explain the metal adsorption on reactive edge sites aluminol (Al-OH) and silanol (Si-OH) functional groups [7].

In the studied soil, pH is higher than the point of zero net charge (p<sub>znc</sub>) of kaolinite (P<sub>ZNC</sub> = 4.6), indicating the predominance of negative charges on the particle surfaces and probable cation retention. Individual metals, however, have a narrow range of pH values over which the percentage of sorption increases. In order of the selectivity sequences, Cr

and Cu present a higher affinity sorbing in a lower pH range than Ni [29]. Thus, despite the low total CEC, the surface reactive phases of the studied soil are potentially important for the effects of selectivity adsorption and reduction of mobility of the metals in the soil profile and their percolation into the groundwater.

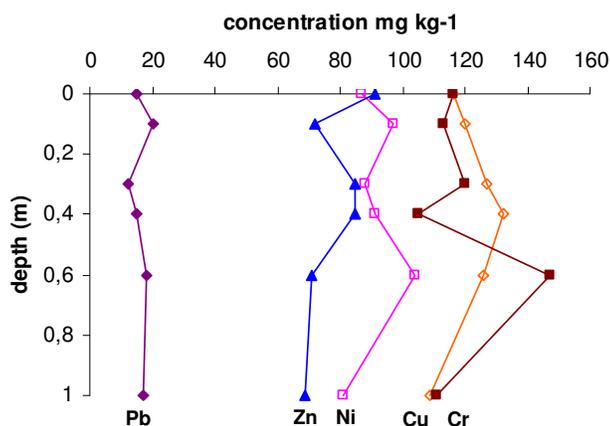


Figure 4. Vertical distributions of metals throughout the soil profile.

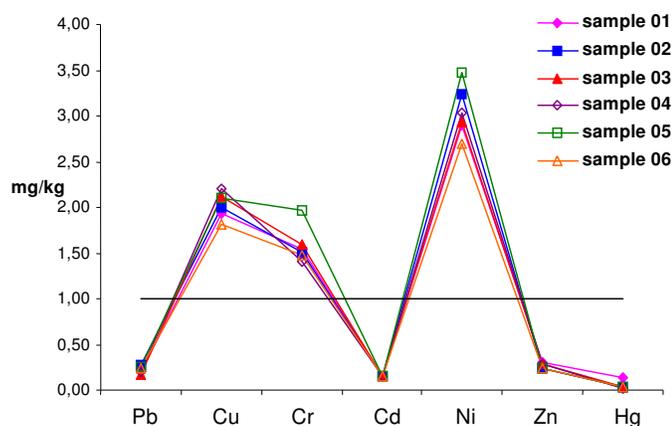


Figure 5. Heavy metal concentrations compared to the references values. Analyses normalized to prevention values.

## 2.2 Characterization of the leachate and water from the landfill

Table 3 shows the values of the physical-chemical parameters determined in the collected samples of leachate and shallow groundwater table. The high values of DQO in the leachate, above 11.000 mg/L, indicate a high pollution potential for

the percolating liquid; in addition, the high values of DBO<sub>5</sub> and DQO indicate a high amount of transported organic compounds [30]. The measured pH of the groundwater was 6.5, while the pH of leachate was 7.9, indicating a predominance of leachate from garbage at an advanced stage of decomposition. 4.600 mg/L of CaCO<sub>3</sub> reveals that the liquid is alkaline and at an advanced stage of decomposition. The high DQO and DBO<sub>5</sub> values suggest that the leachate is composed of a mixture of garbage at different stages of decomposition [30, 31]. The DQO value, well above the DBO<sub>5</sub> value (5.900 mg/L), may be related to the presence of materials with low biodegradability [32]. The elevated content of chlorides, around 2.000 mg/L of Cl, is probably related to the high solubility of most salts, facilitating the process of solubilization and/or lixiviation.

Table 3. Physical-chemical parameters of the leachate and water samples

Parameters (mg l <sup>-1</sup> )	Leachate	Water	Maximum values*
pH	7.9	6.5	--
D.B.O. <sub>5</sub>	5.920	253	--
D.Q.O.	11.252	586	--
Chlorides	2.079	721	--
Nitrogen (Total)	785	13.8	--
Phosphorus (Total)	9	1.34	--
Alkalinity (mg/l CaCO <sub>3</sub> )	4.643	444	--
Pb	0.281	0.138	0.01
Cu	0.35	0.047	2.00
Cr (Total)	2.34	n.d.	0.05
Cd	0.036	n.d.	0.005
Ni	0.52	0.036	0.02
Zn	1.74	0.21	5.00

\*Maximum Values of Reference recommended for groundwater [19,35]. n.d. = not determined

In general, concentrations of DQO, DBO<sub>5</sub>, chlorides, nitrogen, phosphorus, alkalinity and heavy metals in the leachate are significantly less in water (Fig.6), indicating the efficiency of the soil in removing or decreasing the concentration of these elements. The soil acts as an effective filter in removing the dissolved and colloidal organic matter present in the leachate, through the activities of the microorganisms, which convert it in mineralized matter and enable adsorption reactions [33,34]. As for heavy metals, there was an evident reduction in

the content of all analyzed metals in the water, especially Cr and Zn. This decrease is probably caused by the mineralogical characteristics of soil in the studied area, which is capable of selectively promoting mechanisms of attenuation, taking into account the adsorption and complexation factors of these elements.

Table 3 shows the metal concentrations and the respective maximum values recommended for groundwaters according to the Brazilian environmental agencies [19,35]. Despite the evident reduction in the concentration of metals in the water, some metals such as Pb and Ni are above the maximum values allowed (Fig. 7), while Cr and Cd are below the detection limit of the methods used. These norms regulate the potability values of groundwater according to the World Health Organization standards [36]. Nevertheless, it must be mentioned that the analysis of water in the study area corresponds to the shallow groundwater (non-confined aquiferous) therefore it is more vulnerable to contamination. However, this water may leak into the deeper aquifers, carrying the contaminants, and lead to alterations in the potability standards of the groundwater in the region. In general, the decreased concentrations of metal ions found in the water can be attributed to the low solubility of several of these metals in the prevalent pH conditions – 6.5; to the retention mechanisms by the soil and the material in suspension, and the complexation of metals with organic matter [37].

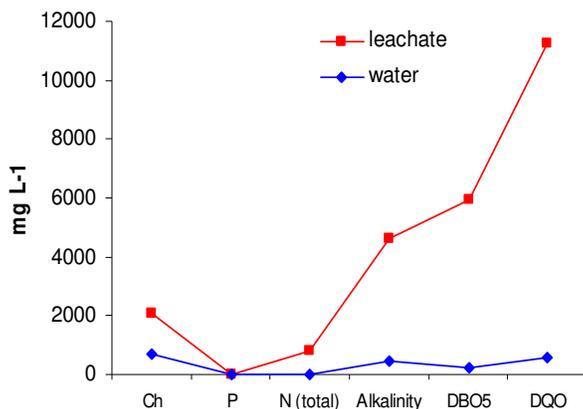


Figure 6. Chlorides (Ch), phosphorus (P), nitrogen (N total), alkalinity, DBO<sub>5</sub> and DQO content of the leachate and water samples.

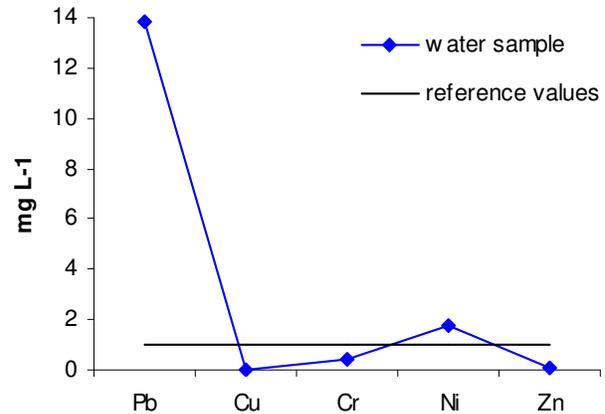


Figure 7. Metal content in water compared to the references values. Analyses normalized to *maximum* potability values for groundwater.

### 2.3 Phytoremediation experiments

The preliminary results show that phytoremediation with *I. walleriana* was effective in removing metals from the soils contaminated by MSW. Table 4 shows the metal concentrations in the biomass plants, as well the concentration of metals in the two studied substrata (S1 and S2) before and after the experiment.

The results indicated an increase in the concentration of metals such as Cu, Zn, Cr, Ni and Pb in the biomass plant after 75 days of experimentation in the tested substrata. Figure 8 shows the variation of metal concentrations observed in the biomass and at the different substrata, before and after the experiment. An increase in the content of Cu, Zn, Cr and Ni was observed in the biomass at the end of the experiment. The Cd and Pb levels remained constant. The concentrations of Cu, Zn and Ni appeared to have decreased at the end of the experiment in both substrata (S1 e S2), with a marked decrease for Cr. The increase in the metal levels in the biomass shows this species' ability to retain and remove these metals, particularly chromium. Despite the retention and bioaccumulation of metals by *I. walleriana*, the final levels of Cu, Cr and Ni present in these soils were still much higher than the *prevention* values for soils determined by the Brazilian environmental agencies [19].

Chemical parameters such as pH and organic carbon were analyzed to understand the availability

of metals in experimental conditions. A decrease in the pH to a range of 5.0 to 5.5 was observed with the increase of organic carbon, at the end of the experiment, which may have increased the mobility (or solubility) of the metals, making them more available to the plant. The retention of metals on soil organic matter is weaker at low pH, resulting in more available metal in the soil solution for root absorption [38]. The pH is perhaps the soil factor that most affects the availability of heavy metals for plant nutrition [39]. At pH values above 7, heavy metals tend to form hydroxy cations, which precipitate on the surface of the oxides, making them more unavailable for the plants as the pH rises. Increases in the solubility of Cd, Cr, Co, and Zn in the soil were observed in the acidic pH that occurs in agricultural soils (5.0 to 6.0) [26]. Chromium was the metal with the highest concentration in the biomass of *I. walleriana*. The mobility and bioavailability of chromium are influenced by the pH and the oxidation state, with the most stable forms in natural environments being trivalent Cr (III) and hexavalent Cr (VI). In experimental conditions, the removal of Cr by *I. walleriana* can be attributed to a greater availability of this element, with the reduction of pH. In general, the solubility of Cr (III) is maximal at a pH <5.0 [27,40].

The fact that the pH and the organic carbon content have varied during the experiment may thus have increased the bioavailability of metals for the plant, particularly Cr. On the other hand, the greater solubility/mobility of metals can permit in natural environments a greater migration of these elements into the physical environment, altering the characteristics of the water and soil.

## 5 Conclusions

The preliminary evaluation of the environmental impact caused by the inadequate final waste disposal at the MSW landfills shows that the inorganic contamination in the soil and water has reached critical levels for some heavy metals, according to the reference values recommended by the Brazilian environmental protection agencies. The concentrations of Cu, Cr (total) and Ni in the soil are higher the *prevention* limits.

According to the physical-chemical and mineralogical characteristics the soils show an ability to retain ions. The mineralogical analysis and the low CEC (~20 cmolc/kg) indicate the presence of mineral phases of low activity, such as kaolinite and Fe oxy-hydroxides, for the sorption processes.

However, the variable charges on surface reactive mineralogical phases play a critical role in the metal adsorption and complexation reactions. The average pH of the studied soil is higher than the point of zero net charge (pzc) of kaolinite (PZNC = 4.6), suggesting a potential effect for the mechanisms of adsorption and reduction of metal mobility through the soil profile and percolation into the groundwater.

The high values of the physical-chemical parameters and heavy metals in the leachate were significantly reduced in the water, indicating the relative efficiency of the soil in the landfill area to promote mechanisms of attenuation of these elements. Nevertheless, the concentrations of Pb and Ni in the shallow groundwater are above the maximum values established for the potability of groundwater.

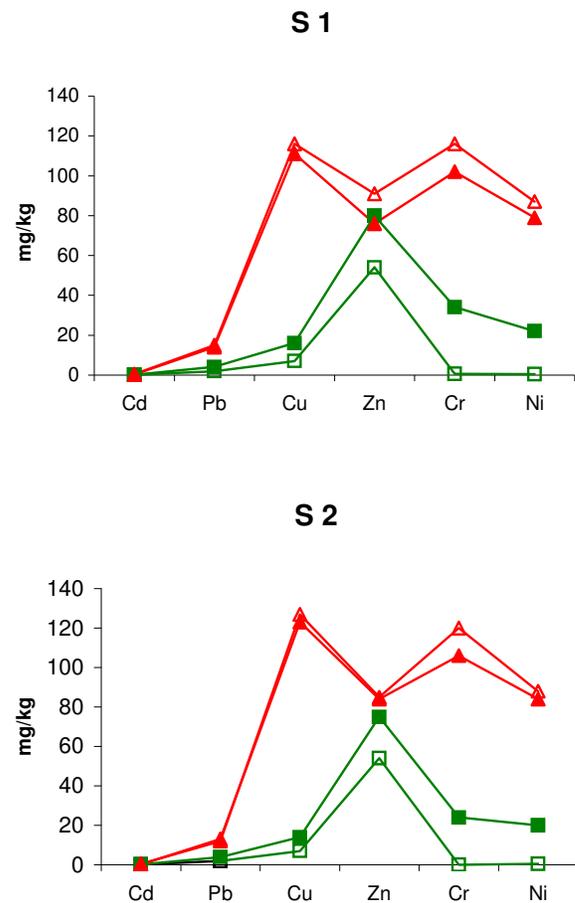


Figure 8. Metals content in the *I. walleriana* biomass and soils (S1 e S2), before and after the experiments. Biomass before  $\square$  and biomass after  $\blacksquare$  experiments; soils before  $\triangle$  and after  $\blacktriangle$  experiments.

Table 4: The metal content (mg L<sup>-1</sup>) and Total Organic Carbon (T.O.C) (%) measured in the soils (S1 and S2) and the biomass of *I. walleriana*, before and after 75 days of the experiment.

Parameters	S1 (1)	S1 (2)	S2 (1)	S2 (2)	Biomass (1)	Biomass from S1 (2)	Biomass from S2 (2)	Prevention values*
Cu	116	111	127	123	7	16	14	60
Zn	91	76	85	84	54	80	75	300
Cd	<0.4	0.3	0.4	0.2	0.2	0.2	0.2	1.3
Cr <sub>Total</sub>	116	102	120	106	0.6	34	24	75
Ni	87	79	88	84	0.4	22	20	30
Pb	15	14	12	13	2	4	4	72
Hg	0.07	0.03	0.06	0.02	n.d	n.d.	n.d.	0.5
pH	6.3	5.6	6.2	5.5	--	--	--	
T.O. C.	0.56	1.5	1.1	1.09	--	--	--	

(1) before experiment; (2) after experiment. Reference values (prevention limits) recommended for soils [19,35].

Phytoremediation with the *I. walleriana* species proved to be effective in removing metals from the soils contaminated by solid wastes. The vegetable biomass presented bioaccumulation of metals, such as Cu, Zn, Cr and Ni, after a period of 75 days. The decreased pH (~5.5) and the increased organic carbon observed after the experiment may have contributed to the increase in the solubility of the metals, making them more available for the plant. However, these variations may permit a greater mobility of these metals through the physical environment, leading to harmful alterations in the quality of the soil and water. Future studies should focus on the separate parts of the plant (root, stalk, leaves and flowers) in order to check for the potential of phytoextraction/bioaccumulation.

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