



Innovative device to assay leachate production in non-sanitary landfills

Natalia de Souza Pelinson¹ · Marjolly Priscila Bais Shinzato^{1,2} · Alice Kimie Martins Morita¹ · Leandro Guimarães Bais Martins^{1,2} · Edson Cezar Wendland¹

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Abstract

Improper landfills (waste dumps) are common waste disposal systems in developing countries and represent sources of environmental pollution. These sites defy researchers and managers because they lack structures to collect liquid, solid and gaseous samples, which make it challenging to monitor local environmental quality. In this work, we show one device for sample collection to monitor leachate quality in a closed waste dump in Brazil. During the installation of this device (Leachate Monitoring Station, LMS), interesting facts about the structural, physical, and chemical composition of an old dump could be visualized. Two different kinds of leachate were found: the accumulated leachate (AL), a thick dark fluid entrapped above non-degraded material, and the mobile leachate (ML), a lighter liquid which flowed into the LMS, and thus was not stagnant like AL. In the AL, the chemical oxygen demand and total ammoniacal nitrogen average concentrations were about 21,500 mg/L and 1000 mg/L, respectively, which were considerably higher than the ML concentrations, of about 1100 mg/L and 200 mg/L, respectively, for the same parameters. Thus, despite the lower concentrations of hazardous substances in the ML, the waste body stores pockets of leachate (AL) with significant concentrations of hazardous compounds, even after 15 years of the dumpsite closure. Moreover, waste solubilization assays showed that the solid material could not be considered inert according to the Brazilian Standard Norm NBR 10004/2004. The installation of the LMS enabled a new understanding about pollutant accumulation inside waste deposits and provided an effective, low-cost tool to monitor leachate production in non-sanitary landfills. The results warn about the risks that old dumpsites still pose to the environment and reinforce the need for a post-care action plan for managing uncontrolled waste deposits.

Keywords Dumpsites · Old leachate · Accumulated leachate · Leachate reservoirs

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✉ Natalia de Souza Pelinson
natalia.pelinson@gmail.com

Marjolly Priscila Bais Shinzato
marjollyps@gmail.com

Alice Kimie Martins Morita
akmmorita@gmail.com

Leandro Guimarães Bais Martins
leandrogbm@gmail.com

Edson Cezar Wendland
ew@sc.usp.br

¹ São Carlos School of Engineering (EESC), University of São Paulo (USP), 400 Trabalhador São Carlense Avenue, São Carlos, SP 13566-590, Brazil

² Federal University of Mato Grosso Do Sul (UFMS), Costa e Silva Avenue, Campo Grande, MS 79070-900, Brazil

Introduction

Waste disposal on the land is an old practice of municipal solid waste (MSW) destination. Although sanitary landfills are considered to be environmentally correct methods of final disposal, open dumps are still commonly used in developing countries, mainly due to the lack of environmental inspection and the availability of land [1–5]. Some examples of currently active open dumps can be cited¹ in Africa (e.g., Mbeubeuss dumpsite in Senegal; Lapite, Awotan, Solous 2 and Eneka in Nigeria; Arlington and Luipaardsvlei in South Africa; and Lagoon in South Sudan), Asia (e.g., Al-Akaider in Jordan; Ghazipur, Okhla and Deonar in India; Jam Chakro in Pakistan; Bishkek in Kyrgyzstan; and Payatas in the Philippines) and Latin America (e.g., La Duquesa in the Dominican Republic; Trutier in Haiti; La Chureca in

¹ <https://www.atlas.d-waste.com/>

Nicaragua; El Trebol in Guatemala; Tegucigalpa in Honduras, Reque and El Milagro in Peru; K'ara K'ara in Bolivia; Bariloche in Argentina; Carpina, Camacan, Divinópolis and Jau in Brazil).

In Brazil, 3331 cities disposed of more than 29.7 million tons of waste in dumpsites and controlled landfills in 2016 [6]. A great quantity of waste was still placed in improper sites, representing more than 40% of the total MSW collected all over the country in that year (in numbers, more than 81,260 t/days of the 195,450 t/days collected did not receive adequate final destination). Unfortunately, the surveys also estimated that about 7 million tons of daily waste or 10% of the waste produced were not even collected in 2016.

These data are worrisome considering that the Brazilian Waste Policy approved in 2010 [7] established a deadline for closing the dumpsites until 2014 and that the bill PL2289/2015, which establishes new deadlines for the closure of dumpsites (between 2018 and 2021), is still in process (in 2019) for being approved in the National Congress.

With the creation of the National Waste Policy, many sites stopped the disposal activities, but did not adopt mitigation techniques or perform environmental monitoring, and were considered closed. Many others did not even close and are now acting informally [8, 9]. By the end of 2017, only 44% of Brazilian states completed their waste management plans, the proportion of sanitary landfills did not significantly increase, and most Brazilian municipalities used unsuitable sites for final disposal [9, 10].

Therefore, similarly to other developing countries, the existence of legal apparatus did not guarantee its effective implementation [9], and solid waste is still not properly disposed of in Brazil. Additionally, it is important to note that only deadlines for the deposits' closures have been discussed in the Brazilian Waste Policy, with no detailed instructions of site-specific aftercare criteria. Therefore, the problem is not only related to the existence of dumpsites but to their effective closure.

Hence, the presented scenario may trigger significant impacts, since the fluids generated by waste landfills have been extensively studied mainly because of its potential for polluting air [11–13], soil [14, 15], and water [16, 17]. In this context, the contamination of groundwater resources by such deposits can be considered a public health problem [18–20] and can generate long-lasting environmental impacts, since the produced leachate affects the aquifers for decades or even centuries after the ending of disposal activities, independently of the deposits' sizes [16, 21].

Several authors have monitored the leachate quality of different waste deposits and created graphs and tables of their characteristics [22–29]. Nonetheless, few types of research have analyzed either the quality of the deposited waste or the liquid produced inside dumpsites, since there

is no adequate drainage network in these deposits, and thus the collection of the material has been impracticable inside the waste mass.

Apart from the lack of data concerning dumpsite leachate characterization, there is also an insufficient understanding regarding the heterogeneities existent in waste deposits. Some studies have recently evaluated the existence of hotspots in landfills, in terms of leachate characteristics, waste composition [5, 30] and/or methane production [12, 13]. Based on the mentioned studies, the existence of high concentrations of contaminants has not yet been totally explained but has been associated with waste ages and composition, as well as with the presence of bulky or coarse-grained materials.

It is important to highlight that the heterogeneities existent in waste deposits are especially relevant in improper landfills since their buried content varies from municipal solid waste to industrial waste, construction and demolition debris, and even hazardous substances. The variability in the deposit content and the different landfilling methodologies in these sites lead to the generation of plumes with distinct characteristics, adding complexity to the understanding of areas impacted by these waste deposits [31].

It is important to highlight that non-sanitary landfills do not have specific structures for collecting and monitoring the fluids. The financial reality of many countries that still have many dump units in operation is not compatible with more sophisticated tools as presented by [5] inserting equipment for real-time collection and monitoring, especially due to high installation and maintenance costs. To collect samples in these contexts, simple solutions could be adopted for monitoring potentially contaminated sites.

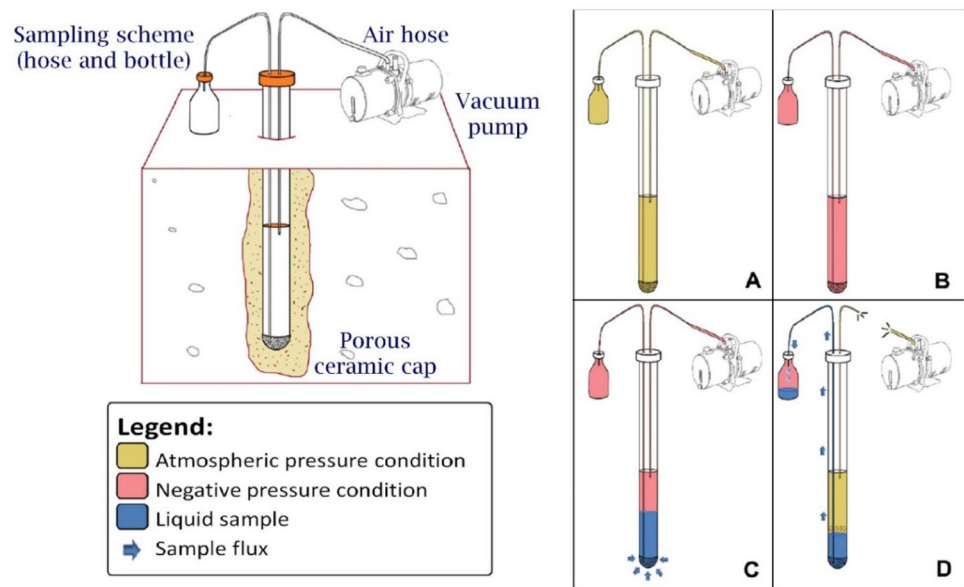
The primary purposes of this manuscript were to show one device for leachate sampling in a closed waste dump in Brazil and present data of the landfilled content and the leachate stored in waste layers with different deposition ages. The goal was to investigate the aspects and heterogeneities of the waste mass composition (solid material and leachate) and contribute to the discussions about the contamination generated by irregular deposits.

Methodology

Study area and location of the dump site

The study area was a dumpsite placed in São Carlos city, São Paulo state, in southeastern Brazil. São Carlos climatic conditions are considered humid subtropical (*Cfa*) according to Köppen and Geiger Classification. The average temperature in this site is 19.7 °C, and the average annual precipitation is about 1440 mm.

Fig. 1 Illustrated diagram of the operation of the lysimeter for liquid samples collection. In **a** it is possible to observe the lysimeter subjected to atmospheric pressure, before collection; in **b**, there is negative pressure applied throughout the vacuum system; in **c**, the negative pressure transports the liquid sample into the lysimeter through the porous capsule; And finally, **d** the air hose is disconnected from the vacuum pump, allowing the atmospheric pressure to enter the system, which forces the sample through the collection hose to the collecting flask. Source: Modified from [60]



The studied waste dump was established without any bottom liners, fluid drainage, or isolation system. This deposit covered an approximated area of 48,400 m² and received an amount of mixed waste of 440,000 m³, or about 390,000 tons, considering an average waste destination of about 2000 tons/month during 16 years [32]. The waste included MSW, construction and demolition waste, health services waste, and industrial waste [32–34].

In the region where the dump is installed, there are sediments of the Botucatu Formation—outcrop portion of the Guarani Aquifer System (GAS), which is an important groundwater reservoir [34–37]. The groundwater represents about 50% of the drinking water of São Carlos, and there is a water spring downstream the dumpsite that contributes to the Ribeirão do Feijão watershed, which also provides about 50% of the São Carlos water supply [34]. It is important to highlight that the waste body itself is placed below the water table: the landfill depth varies from about 6 to 12 m [33], whereas the leachate pockets varies from 2 to 7 m inside the waste mass.

Therefore, this site is part of the list of contaminated areas of the State of São Paulo [38], and its last official data collection and monitoring were carried out by the municipal government in 2011 [34].

Waste and leachate samples collection and analysis

Provided that simple and low-cost solutions are required to monitor leachate production in dumpsites and non-sanitary landfills in developing countries, a designed Leachate Monitoring Station (LMS) of 7.0 m deep was installed (see Fig. 3SM in the supplementary material) in the studied dumpsite based on hydrogeochemical studies performed in

contaminated areas [39, 40]. The LMS, regarded as a hand-dug well with six lysimeters installed at different depths, was projected to propitiate the collection of leachate samples from distinct waste layers in future monitoring studies (Fig. 1).

The hand-dug wells design is known worldwide, facilitating standardization and it does not require mechanization. Additionally, the porous medium allows the retention of moisture, so devices such as vacuum lysimeters, commonly adapted to management in agriculture, can be installed for collection along a profile, allowing the observation of seasonal variations that occur in depth.

The collection of leachate samples in the lysimeters can be proceeded using a vacuum pump, applying suction times which are a function of the moisture content in the layer. In other words, the vacuum is applied until the system is saturated, during periods of time which can vary from 3 min up to 40 min. After leachate collection, it is highly recommended to filter the collected liquid using 0.45 nm membranes, to permit the standardization of samples and geochemical analysis.

The LMS did not reach the bottom of the waste deposit (~ 10 m), mainly because it could contribute to increasing the local contamination, which was unreasonable. We emphasize that individual protection equipment was used in all stages of drilling, collection, transport, and analysis of samples.

During the construction of the LMS, systematic sampling of solid and liquid samples was performed, enabling the characterization of the waste mass and the collection of preserved material with legible writing (see Figs. 1SM and 2SM in the supplementary material), which guided the dating of the material from the layers of the waste body.

Table 1 Characteristics of the samples collected from Santa Madalena Dumpsite

Samples	Classification	Description	Quantity	Parameters analyzed	
Solid	SW-D	Solid waste samples for dating	Solid waste samples with legible writing (packaging, magazines, and newspapers)	24	Dating
	SW-A	Solid waste samples for analysis	Waste and soil samples	15	Moisture and solid contents
Liquid	AL	Accumulated Leachate	Retained liquid collected while accumulated inside the waste mass	14	Electrical conductivity (EC), pH, temperature, alkalinity, nitrogen compounds, major ions, metals, DOC, COD, BOD ₅ ;
	ML	Mobile Liquid	Mobile (flowing) liquid collected after flowing inside the Leachate Monitoring Station	2	EC, pH, temperature, alkalinity, SiO ₂ , nitrogen compounds, major ions, metals, DOC, COD, BOD ₅ ;
	EXT	Soil and waste extract	Solubilization extract from the soil and waste material (SW-A)	15	Nitrogen compounds, pH, major ions, metals, COD

Different kinds of samples were collected: solid samples for dating (SW-D), solid samples for physicochemical analysis (SW-A), soil samples for physical characterization (Soil), and liquid samples from the accumulated leachate (AL) and mobile leachate (ML).

It is important to emphasize that the accumulated leachate (AL) was regarded as the concentrated liquid retained above non-degraded material, which only started to appear below 0.9 m deep since there was no liquid retention above this depth. AL samples from the depths of 2.1 m, 2.7 m, 3.8 m, 4.3 m, 4.6 m, and 5.8 m were collected above soil covering layers or non-degraded waste mixed with soil.

On the other hand, the term mobile leachate (ML) was given to a light-yellow liquid that permeated through the bottom of the LMS whenever the excavation stopped. The ML was not stagnant as the AL but could flow. Its presence was not associated with physical barriers, but possibly with a higher pressure inside the waste mass which maintained this leachate in it. Hence, since the excavation reduced the pressure in the region, it is supposed that this ML found conditions to flow into the LMS. ML samples were collected at 0.8 m and 2.5 m deep.

Afterward, 15 additional samples (EXT) were obtained from the solubilization of the solid samples (SW-A) to evaluate the classification of the waste deposited. Table 1 shows the different samples collected and the corresponding analysis.

Finally, the disturbed soil sample (soil) collection was carried out along the excavation progress, to obtain homogeneous samples from each depth and different deposited layers.

The samples collected for physicochemical analysis (AL and ML) were sampled, stored, transported to the laboratory [41], and analyzed in accordance with the criteria recommended in Standard Methods for the Examination of Water and Wastewater [42]. The leachate samples (AL and ML) were analyzed as raw samples to quantify the total content

of metals in that old leachate so that the analysis covers suspended, colloidal, and dissolved fractions. This choice has been made because the studied dump presented significant leachate pockets, which could enhance metals migration.

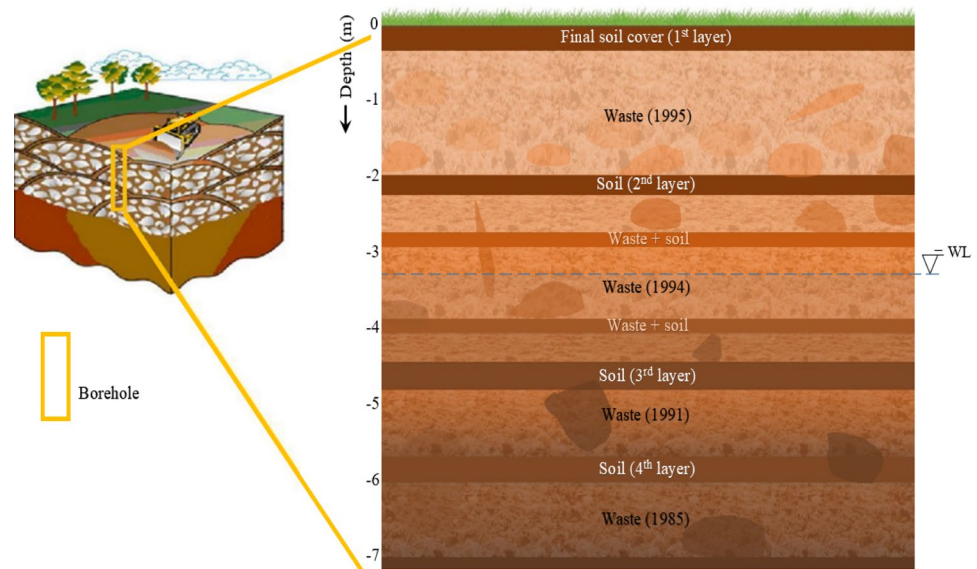
The solid material collected in different layers and depths (SW-A) was solubilized according to specific standard techniques described in NBR 10,006, “Procedure for obtaining solubilized solid waste extract” [43]. Following this norm, the samples were dried until 42 °C, 250 g were weighed and triturated, 1000 mL of distilled water were added to the solid samples (so that a liquid/solid ratio of 4 was obtained), and the solution was stirred for 5 min. The recipient was covered and remained without mixture for seven days at 25 °C. Finally, the solution was filtered in a 0.45 µm membrane, and the obtained sample was called solubilized extract (EXT).

The liquid samples (AL, ML, and EXT) were characterized considering alkalinity (titration method), dissolved organic carbon (DOC), chemical oxygen demand (COD), biological oxygen demand (BOD), total Kjeldahl nitrogen (TKN), major ions (concentrations of sulfate, nitrate, nitrite, phosphate, chlorides, ammoniacal nitrogen), and metals. Additionally, the moisture content of the solid material (SW-A) was determined following NBR 10,644, Water—Determination of residues (solids)—Gravimetric method [44]. All samples were analyzed for different parameters and purposes (see Table 1).

Statistical analysis

Statistical analysis was performed to: (a) identify the most important parameters which characterize AL samples; (b) verify differences between the two recognized kinds of leachate, AL and ML, and (c) verify differences among leachates originated from different deposits. For achieving a and c purposes, principal component analysis (PCA) was performed using a XLSTAT software package [45], considering factor loadings > 0.6 acceptable for a rational

Fig. 2 Schematic profile of the deposit in Santa Madalena Farm



interpretation. To analyze the differences between AL and ML (purpose b), Fisher's test was conducted.

It is important to emphasize that, for conducting PCA with AL samples (purpose a), each collection was treated separately, to show the AL's variability. Thus, the samples were separated according to the layers from which they were collected (1985, 1988, 1991, 1994, and 1995).

Finally, for comparing leachates from different landfills (purpose c), data from [5, 20, 25, 46, 47, 55], and the present manuscript were also evaluated using PCA [46]. was used as a great historical reference for the main leachate characteristics; [47] conducted research at 19 landfills in the United States and concluded that older landfills tend to generate large amounts of leachate; [5] carried out monitoring in the vadose zone of an old unlined landfill (dump) in Israel; [20] presented data from a young landfill leachate; [55] and [25] were regarded as Brazilian references.

Results and discussion

Schematizing the waste body profile of Santa Madalena Dumpsite

In the waste layers of Santa Madalena dump, a mix of materials was visually identified, including health services objects, pruning and sweeping street debris, oils, construction and demolition waste, and tannery industrial waste, even though the most significant proportion was the municipal solid waste (MSW).

The waste composition was assessed by [48] after analyzing 100 kg of waste from 2 layers. From the 1995-layer, the gravimetric fractions were: 53.1% of organic matter, 18.5% of plastics, 7.5% of metal, 2.8% of paper, 3.4% of glass, and

14.7% of others, while from the 1985 layer, the gravimetric values were 44% of organic matter, 20% of plastics, 10% of metal, 7% of paper, 5% of glass, and 14% of others. The gravimetry was compatible with other studies in the same site [49] and with the average Brazilian waste composition [6]; additionally, it showed a reduction in the organic matter content from the 1995-layer (~53%) to the 1985 layer (~44%).

This tendency was even more evident when these data were compared to the gravimetry obtained during the landfill's operation [49], showing a decrease from 56 to 44% in the percentage of organic matter. This diminution of organic matter content accompanied an increase in the percentage of plastic, metals, glass, and others, which varied from 44 to 56%. This behavior was attributed to the degradation of organic matter, as well as to the leaching of smaller particles, and the resilience of plastic, metals, and glass.

The evaluation of 7 m of the waste deposit profile provided a conceptual model of the dump layout merely extrapolating the investigated site. Four slight soil cover layers and four waste layers deposited in different years (1985, 1991, 1994, and 1995) were recognized. The dimensions and ages of the different layers are shown in Fig. 2. Note that the 1985 layer is supposedly the first one to be deposited in this profile, since the 1988 waste materials were settled above the 1983 layer, following a similar method of horizontal deposition until 1992 [36].

From the systematic collection in the waste body, it was observed that the frequency of the soil cover was not daily and it was more noticeable after 1988. The waste layers' thicknesses were quite irregular due to the arrangement of the waste in the gully, which had a non-uniform terrain base with different depths and slope gradients. In the end, waste and soil layers filled the gully, so that the landfill has a final

topography of 15(H):1(V), and presents soft contour lines to prevent erosion problems. Although the deposit presents a final soil cover with fine sand and vegetation, this topsoil is not proper as a containment layer because it has a low clay content. In this sense, the waste dump has inefficient hydraulic devices which allow the input, output, and accumulation of water within the waste body.

Waste layers' physical parameters

The soil covers consistently presented less moisture than the waste layers, and the water content values varied throughout the investigated profile (See Table 1SM in the supplementary material). The moisture content of the waste body in the first half meter from the surface was 12%. After this depth, these solid samples (SW-A) indicated that the dump was more humid, with an average of 50% of moisture content.

It is important to emphasize that the high values of moisture content could be associated with the presence of non-degraded plastic inside the dump, which behaved as reservoirs storing significant volumes of liquids (the accumulated leachate, AL), generating moisture heterogeneity in the waste mass. Thus, in the case of irregular waste deposits, the plastic wastes may assume function as a non-projected barrier, minimizing the discharge of high loads of pollutants through containing and storing great leachate volumes and contaminated sediments inside the waste body. This process was considered especially relevant in Brazil, where the use of plastic bags has been abundant; moreover, the degradation of organic matter leading to the existence of higher proportions of non-degraded material, as previously discussed, may increase the relative importance of these barriers.

Nonetheless, it is important to emphasize that these reservoirs can release the pollutants after some time, collaborating to the leaching through the waste after the infiltration of water through the dumped material. Climate changes, for example, may increase the out-wash of contaminants from the waste dump, which is a hypothesis of [50]. Additionally, it is important to emphasize that the increase of moisture content provided by these pockets may considerably collaborate to leachate generation [51, 52]. This is worrisome since no retention structure nor mitigation procedures have been planned for this deposit.

Regarding the waste temperature, the deeper the position in the waste mass was, the lower the temperature variations were. Thus, the current average temperature during the excavation was in the range of 20–21 °C (See Table 1SM in the supplementary material), whereas it ranged from 35 to 37 °C during the Santa Madalena dumpsite operation, from 1980 to 1996 [49]. Since the initial stages of decomposition can increase the waste mass temperature to values higher than 30 °C [51], this first result indicated that the Santa Madalena waste deposit is in an advanced stage of decomposition.

Finally, concerning the total volatile solids (TVS), the values obtained from the samples collected in the drilling conducted in the present research (SW-A) are presented in Table 1SM of the supplementary material. It is important to highlight that TVS include degradable materials (e.g., cellulose and hemicellulose), and recalcitrant compounds (e.g., lignin and plastics) [53], and that high fraction of TVS percentages reveal the presence of non-degraded organic matter.

From Table 1 SM, it is also possible to note that the TVS in the 1994 waste layer was about 40%, which is relatively high for an advanced stage of decomposition. This result can be related to the period during which the waste layer was exposed to atmospheric conditions: while the 1985 layer was exposed for six years (being covered by a thin layer of soil during the last three years) and the 1991 layer was exposed for three years, the 1994 layer was exposed for less than one year. Hence, it is possible to assume that the lower period of exposure to oxygen anticipated anaerobic conditions in the 1994 layer, which promoted slower decomposition compared to the one under aerobic conditions.

Solubilization extracts characteristics

Concerning the analysis of the solubilization liquid (EXT, the liquid obtained from the solubilization assays), the values for pH, COD, ions, and metals show a vertical quality variation along the waste profile (Fig. 3). The limit values (LV) were plotted on the Na, Mn, Al, Fe, Cd, Pb, and Cr graphs, which had results above the limiting concentrations for the residue to be classified as inert considering the Brazilian Standard Norm NBR 10004 [54]. For other metals like zinc, copper, silver, and barium, the values were below the limit values.

Thus, although the remained leaching potential was not analyzed, the results considering the Brazilian Standard state that the material is not inert yet. Therefore, the solubilization results indicated that grounded waste could still behave as a source of water and soil contamination.

In relation to the presence of the water level inside the deposit, it could be noticed that some parameters of the solubilized samples (EXT) showed a decrease below the water level (phosphates, COD, Co, Sr, Ca, and Ba), which can be associated with their easier transport in the water medium. Nonetheless, all other parameters were still high with the presence of leachate pockets, not only for the solubilized sample (EXT) but also for the accumulated (AL) and mobile (ML) leachates. This observation shows that even being in contact with water, which could lead to the contaminants more natural dissolution and transport, the residues did not behave much differently from the studies where the water was not in contact with the waste.

Moreover, it is interesting to note that the evaluated parameters in the studied waste dump had similar ranges to

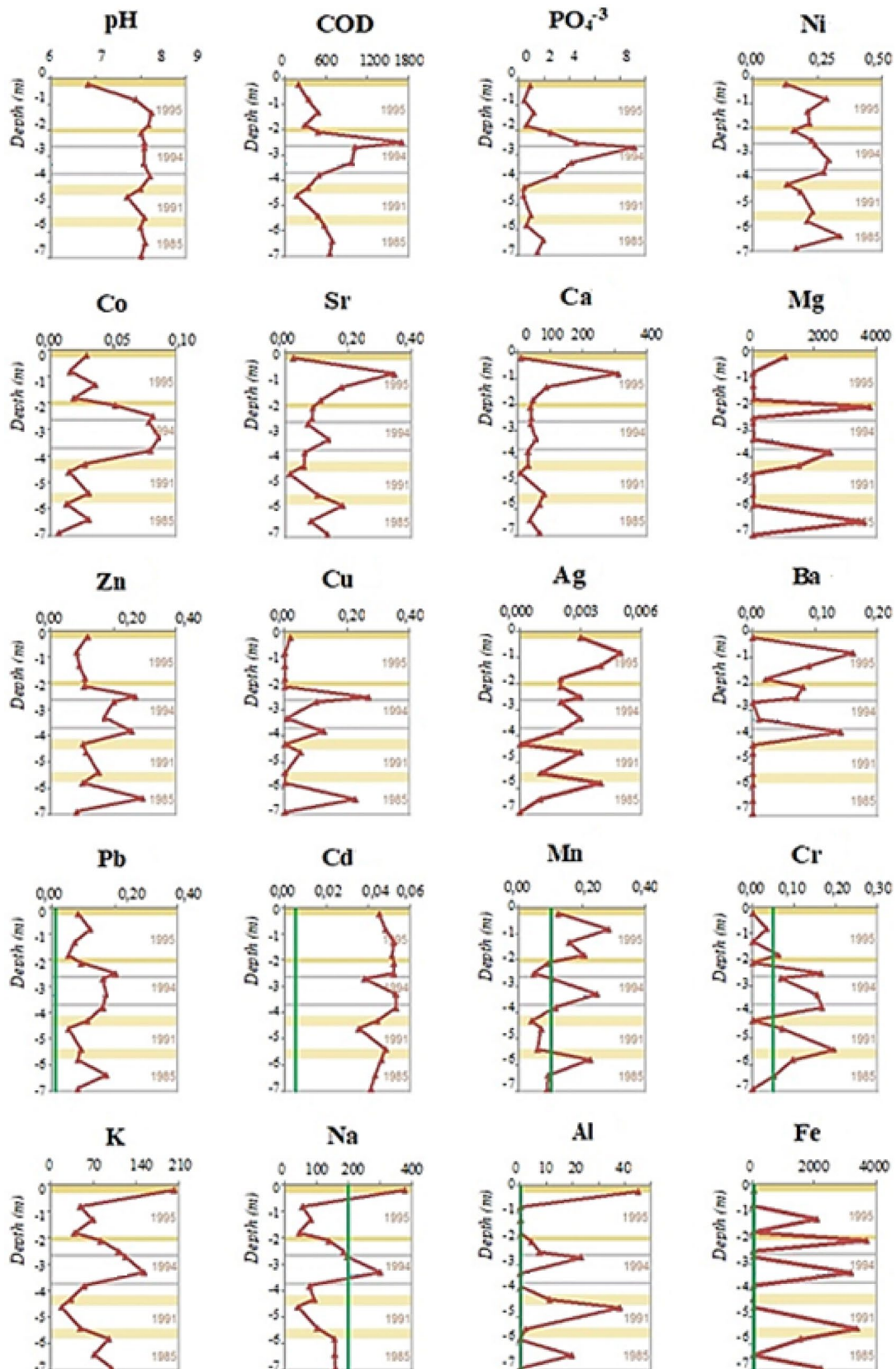


Fig. 3 Parameters analyzed in the solubilized waste samples (EXT). The green vertical lines represent the limiting values for the residue to be considered inert, and the blue horizontal line represents the water level inside the waste mass

the ones obtained for landfills, even though a dumpsite does not have any isolation system, which could contribute to the pollutants leaching and consequent concentrations decrease inside the waste mass with time.

This similarity could be related to the heterogeneities existent inside the waste deposit, with the existence of soil layers, non-degraded material and plastic barriers that may reduce leaching and/or promote a delay in the pollutants' release. Likewise, it is interesting to note that, in many cases, higher metals concentrations were found above non-degraded material, showing that it behaved as a physical barrier and did not permit the compounds to leach to deeper layers.

Leachate characteristics

The results of the parameters analyzed for the collected leachate samples (AL and ML, previously described) are shown in Fig. 4. Note that this preliminary study did not intend to present detailed geochemical research, but rather promote a better understanding of irregular deposits structures and their contamination potential after years of closure.

The ML samples had low concentrations of organic and inorganic compounds, as expected for old leachate, and their values were close to the minimum ones found for the accumulated leachate (AL), except for COD, zinc, iron, and aluminum. These samples were collected only twice during the excavation; the different collections presented similar concentration values.

Regarding the AL samples, it was possible to note the visual similarities between the concentration distribution curves of PO_4^{-3} and some metals (Pb, Zn, Ca, Ni, Sr, Ba, Cd, and Cu), including the increase associated with the 1985 waste layer. This could be explained by the fact that phosphates can contribute to metal precipitation, especially for the divalent species [56].

Sodium and potassium present similar solubilities and are more significant contributors to electrical conductivity; thus, it is interesting to notice that their concentrations in the AL increased with depth, providing evidence of vertical flow in consonance with the electrical conductivity, alkalinity, TKN, TAN, and COD, which are more expressively concentrated in the 2.8 m and 4.3 m deep samples.

The parameters Sr, Cd, Ni, TKN, PO_4^{-3} , Zn, Mn, and Ag, as well as the conductivity, indicated more uniform concentrations. The lowest concentrations of most of the evaluated parameters (except Mg, Sr, PO_4^{-3} , and BOD) were detected in the first samples, collected from 0.9 to 1.3 m deep, in the most active region of the unsaturated zone [17]. Although the region immediately close to the surface could be especially active, there was not enough liquid to be collected in the superficial portion.

On the other hand, the highest concentrations of the evaluated parameters (except Ba, Cr, BOD, COD, TKN, NO_2^- , and NO_3^-) were detected below 3.8 m, which coincided with the "saturated zone" of the waste dump, or the region with more leachate pockets. The low concentrations in the first layers and the higher concentrations in the layers below are a further indicator of vertical contaminant transport.

This behavior shows that the presence of the leachate pockets inside the waste body provided higher dissolution of substances, but did not enhance the contaminants leaching and/or washing. The contaminants were somewhat retained inside the waste body, which was partially explained by the existence of heterogeneities generated by non-degraded material.

The TKN and TAN curves are similar in depth, mainly due to $\text{NH}_4\text{-N}$, which is counted in TKN. The profile presented a mean of about 65% of the total nitrogen in the form of ammoniacal nitrogen and 35% of organic nitrogen (N_{org} referring to the non-hydrolyzed protein and microorganisms). The average of the TAN results below the level of the mobile leachate (3.5 m) was 30% higher than the average of the results above this depth; in samples from 1.8 m, 2.1 m, 2.5 m, 4.3 m, 4.6 m depths, the organic N content was higher than 40%. In this way, it corroborates the statement that ammonia is the most persistent material in MSW deposits [52].

The pH remained alkaline (between 7.7 and 8.9) along the whole profile of Santa Madalena dumpsite, a typical characteristic of the methanogenic phase of the decomposition [57]. Additionally, the AL samples presented high alkalinity, factors that decrease the inorganic mobilization [51], which is also indicated by the low electrical conductivity.

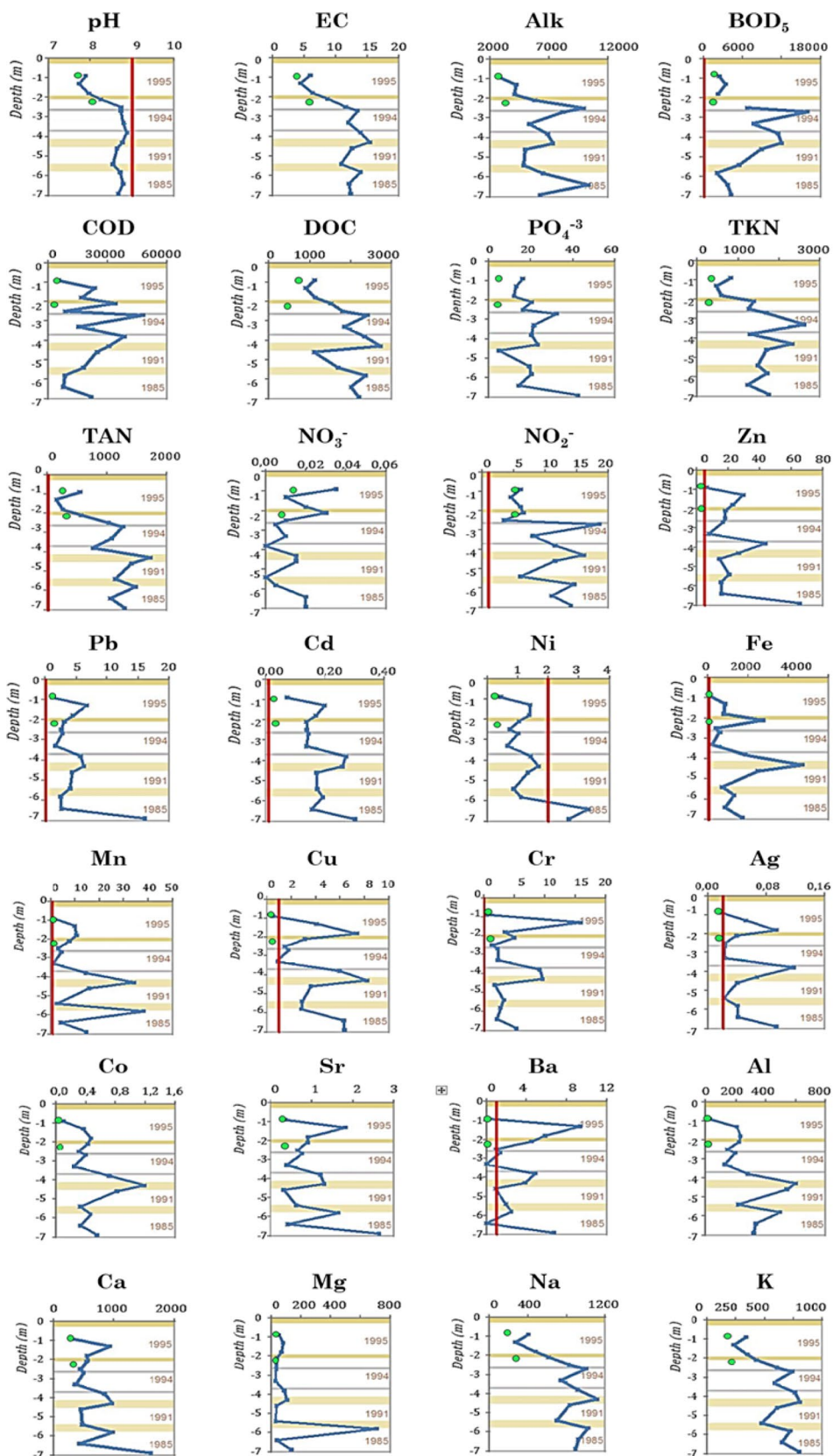
In almost all AL samples, the concentrations of metals (Pb, Cu, Cr, Fe, Mn, $\text{NH}_4\text{-N}$, Cd, Ni, Ba, Ag, and Zn) were higher than the permitted values for releasing effluent into the environment system by the Brazilian guidelines [58].

The concentrations of some metals in the solid sample (EXT) were similar to the concentrations of these metals in the leachate (AL and ML), indicating a significant mobilization and equilibrium through the natural leaching within the waste mass. On the other hand, the concentrations of chromium and copper in the leachate are lower than the concentrations in the solid content, indicating a low mobilization of these elements in natural conditions.

Substances variations between different layers and landfills: a statistical analysis

Principal Component Analysis (PCA) for AL is presented in Fig. 5. For the conducted PCA, only F1 and F2 were presented since the cumulative variability was about 65%. Note that Pearson's matrices of correlations are shown in the supplementary material.

Fig. 4 Parameters analyzed in the accumulated leachate (AL); the green dots represent the mobile leachate (ML), the blue horizontal line represents the water level inside the waste mass, whereas the red vertical lines represent the limiting values for effluent emissions in Brazil



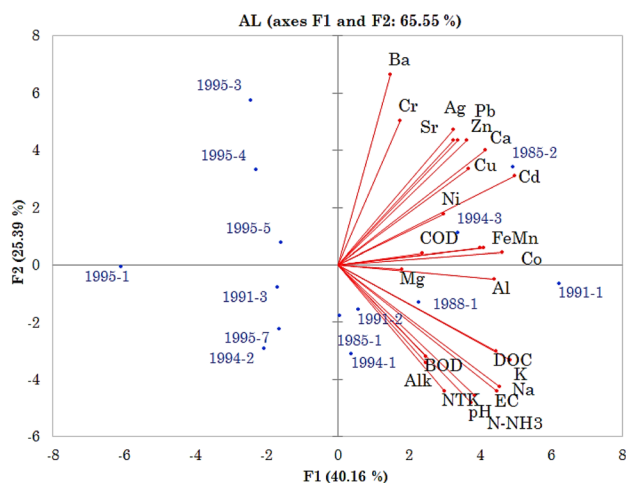


Fig. 5 Plots of variables and observations with PCA for accumulated leachate (AL)

Figure 4 showed that F1 was dominantly represented by 14 variables (pH, EC, DOC, N-NH₃, Al, Mn, Fe, Na, Ca, Cd, Co, K, Cr, Cu, and Zn) and F2 by 5 variables (Ag, Ba, Cr, Pb, and Sr).

The similarities verified between the different samples could be mainly explained by the content found in the different layers and were not dependent on the existence of soil layers, and/or the waste age. Thus, samples collected from layers 1991–2, 1985–1, 1994–1, 1988–1, and 1991–1 presented a dark color mass (regarded as materials in a high stage of decomposition) which can have contributed to parameters associated with organic matter (DOC and BOD), nitrogen, and electrical conductivity. Contrary to these parameter vectors, diagonally, the observations from the less degraded layers can be observed, which had a greater volume of non-deformed material (1995–3, 1995–4, and 1995–5). Finally, the layers 1985–2 and 1994–3 contained considerable volumes of metallic materials, which agree well with the higher concentrations of metals.

This analysis shows that the leachate accumulated above non-degraded material (AL) has considerable variability, depending on the characteristics of the surrounding material. Thus, their characteristics are local and do not follow a pattern within the waste deposit. Additionally, its occurrence and characteristics are not dependent only on the existence of intermediate soil layers, which strengthens the hypothesis of non-degraded material functioning as barriers for the accumulation of this kind of leachate.

Regarding the comparison of the AL with the ML, the Fisher's Test ($\alpha = 0.05$, 95% confidence interval on the ratio of variances) identified that AL and ML are statistically different (p value < 0.0001), showing evidence that the leachate trapped in pockets within the deposit (AL) has characteristics different from the surrounding leachate (ML).

Furthermore, as it was previously mentioned, different AL samples also have distinct characteristics, showing the enormous heterogeneity existent in such waste deposits.

Finally, aiming to evaluate whether the studied leachate had characteristics similar to other landfills leachates, data (Table 2) from [5, 25, 46, 47, 55], and the present manuscript (considering both AL and ML) were treated by a PCA (Fig. 6). This analysis could group [25, 46, 55] with our data, showing a similarity between leachates generated by sanitary landfills and dumps (landfills without liners, leachate collection or drainage control). Note that [20] was also used for statistical analysis, but because it presents very recent data (from a deposit under 5 years of operation), it was not reported in the comparison table.

It is important to highlight that, even if only ML is to be considered in the analysis, the evaluated concentrations are still in the range obtained by [55, 59]. Thus, even not having liners and being situated below the water table, the studied deposit produces leachates similar to the ones produced by sanitary landfills. This result corroborates the hypothesis that irregular waste deposits may present heterogeneities (non-degraded material and soil layers) which behave as barriers to the leaching of contaminants, keeping high concentrations of them even years after the deposits' closure. Thus, even being in the methanogenic phase and having possibly suffered a dilution of contaminants due to the existence of the leachate pockets inside the waste mass, the studied deposit is still a considerable source of contaminants.

The presented results show the importance of monitoring abandoned waste deposits, due to the existence of high levels of contaminants even years after the ending of disposal activities.

Conclusions

The studied abandoned waste dump was listed on the contaminated areas list of São Paulo state [38] and the contamination risk has been associated with the storage of old solid waste and leachate without any environmental control devices. To allow leachate sampling and monitoring, a Leachate Monitoring Station (LMS) was installed within the waste mass. This approach was innovative especially in Brazil, where dumpsites leachate monitoring is not performed due to the lack of collection devices, and where irregular deposits are still abundant.

During the construction of the LMS, interesting characteristics of the waste dump have been noticed and analyzed and were presented in this paper. Two different kinds of leachate were collected inside the waste mass: the mobile leachate (ML), presenting much lower concentrations of metals and other components, and the accumulated leachate (AL), storing high concentrations of organic matter and metals.

Table 2 Leachate characteristics obtained in the present investigation and other authors' research

References	Present manuscript	Ehrig [46]	Souto and Povinelli [25]	Masoner et al. [47]	Aharoni et al. [5]	Pauli et al. [55]
Landfill age (years or phase)	15–30	Various	Various	Various	40–60	0–15
Deposit classification	Dump	Landfill	Landfills	Landfills	Dump	Landfill
Location	Brazil	Germany	Brazil	USA	Israel	Brazil
pH (–)	7.7–8.9	7.5–9.0	5.9–9.2	6.0–7.7	5.9–9.7	7.33–7.73
Alkalinity (mg L ⁻¹ CaCO ₃)	2000–10,300	300–11,500	125–20,000	600–12,200	400–40,600	6200–500
Biochemical oxygen demand (mg L ⁻¹)	400–16,000	20–550	3–17,200	–	40–54,000	2780–3030
Chemical oxygen demand (mg L ⁻¹)	1000–47,800	500–4500	20–35,000	–	570–11,4000	5990–7120
Chlorides (mg L ⁻¹)	0–1200	0–5000	200–6900	160–3040	1000–6000	–
Electrical conductivity (mS cm ⁻¹)	4–15	–	0.1–45	1.7–16.5	–	13–15
Ammoniacal nitrogen (mg L ⁻¹ N-NH ₃)	150–1700	30–3000	0.3–3000	10–1790	0–3,840	1200–1240
Nitrite (mg L ⁻¹ N-NO ₂)	3–18.5	0–25	0–70	–	–	665–675
Nitrate (mg L ⁻¹ N-NO ₃)	0–0.04	–	0–270	0–5	0–75	–
Total nitrogen (mg L ⁻¹)	310–2600	0.1–50	0.6–5000	–	–	1365–1370
Phosphate (mg L ⁻¹)	3.0–42.0	–	0–80	0–25	–	10–2
Ag (mg L ⁻¹)	0.001–0.12	–	–	0–0.005	–	–
Al (mg L ⁻¹)	6–600	–	–	0.080–1.180	0.001–1 (*)	–
Ca (mg L ⁻¹)	290–1600	10–2500	–	50–915	10–8200	110–130
Cd (mg L ⁻¹)	0.1–0.3	0.0005–0.14	0–0.6	0–0.0100	–	–
Co (mg L ⁻¹)	0.1–1.2	0.004–1.0	–	0.001–0.085	–	0.08–0.10
Cr (mg L ⁻¹)	0.2–15	0.03–1.6	0.006–1.0	0.007–0.350	0.001–1.0 (*)	0.25–0.30
Cu (mg L ⁻¹)	0.1–8.2	0.004–1.4	0–3.0	0.020–0.730	–	0.02–0.04
Fe (mg L ⁻¹)	60–4200	3.0–280	0.01–720	0–80	0–2400	55–57
K (mg L ⁻¹)	210–800	40–350	–	7–725	10–2300	700–850
Mg (mg L ⁻¹)	28.9–707	50–4000	–	40–1130	10–1300	–
Mn (mg L ⁻¹)	0.4–40	0.03–45	0.1–30	0.045–9.090	0–1.0	0.9–1.0
Na (mg L ⁻¹)	250–1100	–	–	165–2550	100–3450	–
Ni (mg L ⁻¹)	0.3–3.3	0.02–2.0	0–1.4	0.015–0.575	0.01–10 (*)	0.13–0.17
Pb (mg L ⁻¹)	0.3–16	0.008–1.0	0–7.0	0.001–0.110	0.001–1.0 (*)	0.25–0.35
Sr (mg L ⁻¹)	0.3–2.6	0.3–7.0	–	0.675–7.370	–	0.50–0.65
Zn (mg L ⁻¹)	0.6–65	0.03–4.0	0.01–35	0.070–3.520	–	1.57–170

*Some values from Aharoni et al. [5] were extracted directly from their graphs once the tables do not present all the concentration values

Non-degraded materials—especially plastic—and soil layers stored the AL, forming trapped pools that allowed the concentration of pollutants and may have contributed to a delay in the pollutants' release into the environment, working as temporary physical barriers within the waste body. This variation in both liquids is a significant evidence that abandoned dumpsites still contain retained liquids which may pose a risk for the surrounding environment. This corroborates the research of [5], which identified hotspots inside the waste mass.

The statistical comparison (PCA) between the data measured in the present work and the prediction data in the literature showed that the values of [25, 46, 55] could also be used to characterize leachate in Brazilian dumpsites.

The statistical analysis also showed that leachates differ not only among different countries—mainly because of the MSW composition, landfill structure, and climate [51, 61]—but also within the same waste body, due to the different composition and ages, and to the formation of barriers that accumulate highly concentrated leachate.

It is important to highlight that this containment of leachate by non-projected barriers may have made the contaminant concentrations be within the range expected for landfills. This result is especially relevant considering the existence of the leachate pockets inside the waste mass in the studied case, indicating that this condition did not necessarily accelerate leaching and contaminant transport.

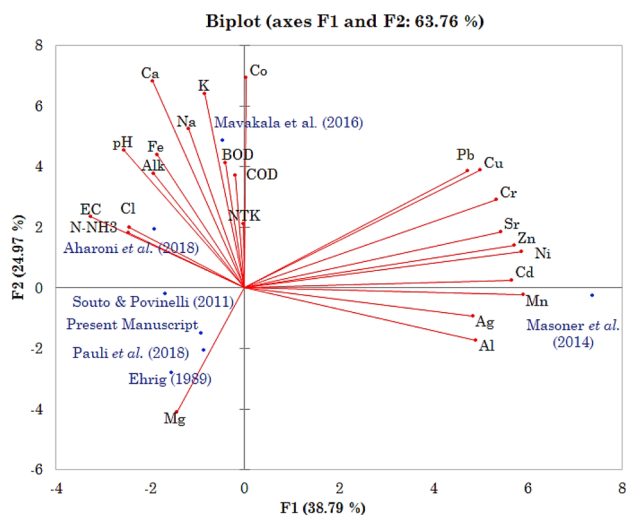


Fig. 6 PCA from different authors' data

The waste deposit age (between 15 and 30 years considering the different layers' ages) and the composition of its leachate indicated that it is in an advanced stage of decomposition (methanogenic phase), when low concentrations of organic and inorganic contents are expected to be found, suggesting a reduction, or even the absence, of pollution risks. Nonetheless, this research indicated that 30 years—the minimum period of post-care closure required by American regulations [62]—might not be a sufficient monitoring period for an abandoned dumpsite under subtropical climatic conditions and with improper groundwater sampling (abandoned sites), since it has been shown that old leachates may present high concentrations of pollutants, including ammonia and heavy metals. This information agrees with [52], who questioned the absence of risks associated with old landfills. Moreover, based on solubilization assays, it could be concluded that the surveyed residues cannot be considered inert yet.

Therefore, this research showed the urgency to identify the potential risks that dumpsites impose on the surrounding communities and ecosystems not only during their operation but also after its closure, mainly due to the large number of these abandoned deposits around the world. Provided that local assessments are necessary to better manage such risks, one effective and low-cost device for leachate collection is presented, supporting its adoption in other abandoned dumpsites and non-sanitary landfills.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest.

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