THE LEACHATE COMPOSITION OF AN OLD WASTE DUMP CONNECTED TO GROUNDWATER: INFLUENCE OF THE RECLAMATION WORKS

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Abstract. This study has been carried out in an uncontrolled dump in the suburbs of Buenos Aires, Argentina. This dump still contains solid wastes of different types in direct contact with the human consumption aquifer. After 18 years of overturning the dump was closed in 1992, and the reclamation works started.

Three leachate samplings were carried out in three different zones of the dump, selected according to the topography, the age of residues and the degree of water contact. These samplings were done before the reclamation works, at the end, and four months after they were finished. Several chemical parameters, including heavy metals in solid and dissolved fractions, were analyzed.

Multivariate analysis (PCA) was performed to help to understand the effects of the labors on leachate composition. The works could be responsible for a temporal increase of dissolved heavy metals and several chemical variables in the leachate. At the sites of the dump where the modifications were carried out, the levels of some elements analyzed recovered their former values a few months after the reclamations works. At the farthest site, the impact and recovery effects of the works on leachate composition, were delayed.

1. Introduction

Leachates that originate due to the disposal of domestic and industrial solid wastes are an aquifer contamination source. Their chemical characterization is the first step for a meaningful environmental impact analysis (Pohland and Harber, 1986). Most of the studies have been performed on landfills containing an unsaturated zone between the wastes and the groundwater, where the contaminants are attenuated by diverse processes (Kjeldsen, 1993; Mirecki and Parks, 1994; Bielsa *et al.*, 1978; Matthess, 1980). The risk of aquifer contamination is increased if solid wastes are directly connected with groundwater (Zanoni, 1972).

The chemical composition of the leachates is very variable, according to the waste types, method of disposal, dump depth (UNESCO, 1980; Pohland and Harber, 1986), age of residues, degree of wetness (Zanoni, 1972; Canter *et al.*, 1987) and the landfill management. The climatic factors, such as temperature and precipitation, also have an influence on the leachate volume (UNESCO, 1980; Pohland

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and Harber, 1986; Kjeldsen, 1993) and concentration levels (Pohland and Harber, 1986).

Heavy metals are commonly detected in leachates (Meyer, 1973; UNESCO, 1980; Pohland and Harber, 1986). The risk of the groundwater contamination will depend on the physical and chemical conditions that influence their mobility, such as pH, redox potential (Kargbo, 1994), dissolved oxygen (Förstner and Carstens, 1989), salinity (Warren and Zimmerman, 1994), organic matter level (Kargbo, 1994; Förstner and Carstens, 1989) and temperature (Warren and Zimmerman, 1994).

This study, the first of this type for the region, has been carried out in one of the dumps situated in the Buenos Aires suburbs. The dump is characterized by the accumulation of residues of diverse origin with no kind of control, and by the disposal of solid wastes in direct contact with the semiconfined aquifer. This aquifer supplies drinking water by means of domiciliary wells to a population of about 20 000 dwellers residing around the dump. The objectives of this study are:
  (a) to evaluate the chemical composition of the leachate;
  (b) to analyze the influence of the reclamation works on the evolution of the leachate composition.

2. Materials and Methods

2.1. STUDY AREA

2.1.1. *General description*
The study area is an unmanaged dump which extends to 4 ha with a maximum depth of ca. 25 m, which has been active from 1974 to 1992. It is located in the district of Florencio Varela, Buenos Aires, Argentina (Figure 1). Its Northeastern border limits with Las Piedras stream. This stream is shallow (depending on precipitation), mean flow of 0.37 m^3 s^{-1}, biologically contaminated (unpublished data); and has no hydrologic relation with the subjacent aquifers (Girardi, 1992).

2.1.2. *History*
The dump originated from an excavation for the extraction of calcrete. This excavation was flooded when the Pampeano aquifer was perforated. When the excavation was abandoned, the dumping of high amounts of residues from domiciles, industries, hospitals and veterinarians began. In February of 1992, the dump was closed, and the municipal authorities began the works of attempting to recover the area. Operations started when the domiciliary garbage which had accumulated in the borders was dragged into the central zone. The residues were then compacted and covered with earth.

Historic information was obtained from interviews with neighbors and scavengers of the dump. Nevertheless, to verify this data, photographs of the site were obtained from various conferences and newspapers.
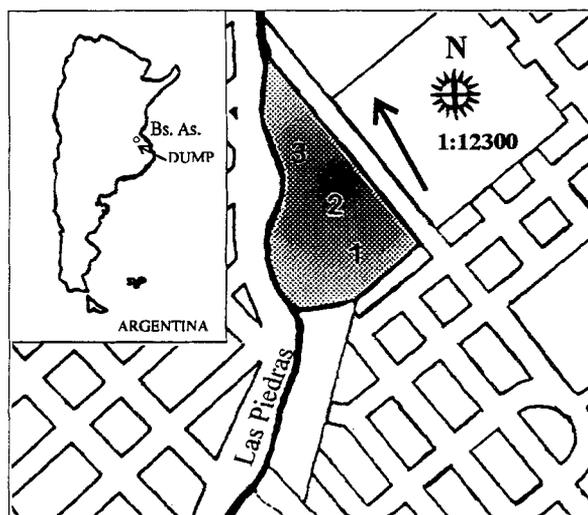
Fig. 1. Location of the dump. The shadow area corresponds to the dump. The numbers indicate the sites of sampling; the arrow indicates the groundwater flow direction.

2.1.3. *Dump sites*

At the beginning of this study, three zones could be distinguished. They were named Sites 1, 2 and 3, with numbers increasing with the groundwater flow direction.

Site 1 consisted of recent domiciliary garbage variably disposed and wastes contacting only with precipitation water.

Site 2, located in the middle of the dump, was the lowest topographic zone, receiving rain and runoff waters. It was in direct contact with the waters of the Pampeano aquifer. This site, coincident with the maximum depth of the excavation, comprised wastes of intermediate time. The field was not consolidated, being like a 'floating island' which gave way when it was treaded on. The solid residues were watersaturated throughout the year.

Site 3 contained the oldest waste dumps. It was covered with earth before the beginning of this study. The works did not affect the surface of this zone.

2.2. ENVIRONMENTAL CHARACTERISTICS

2.2.1. *Climate*

According to Thornthwaite's classification and using local records during the period 1961–1980, the climate is sub-humid-humid, mesothermal (B2), with a mean annual precipitation of 1029 mm, and little or absent water deficit (C2). March is the month of heaviest rainfalls and June is the lowest. The mean annual temperature is 16 °C, with minimum and maximum means monthly of 10 °C and 22.7 °C respectively. The mean real evapotranspiration is 803 mm, with a mean annual hydric

excess of 226 mm. The month of greatest evapotranspiration is March, which, with January and February, comprise the period of least useful water reserve. Local data of temperature and precipitation during the study period are not significantly different from the values of mean annual data.

2.2.2. *Hydrogeology*

The near-surface lithologic units beneath the dump include the Puelches Formation and the 'Pampean Silts'. The Puelches Formation dates from the Pliocene age and has, principally, pluvial origin. It is a sandy unit, and its thickness in the area is between 30 and 54 m. The 'Pampean Silts', which are found in the upper part of the Puelches Formation, are loessoides calcareous silts intercalated with sandy silts, heavily clayey in the base. This base, 5 m thick, contains a high proportion of clays, mainly illite (González Bonorino, 1965). Its vertical permeability is 0.034 m day^{-1} (Girardi, 1992).

The Puelches Formation contains the confined aquifer (Puelche), which is the principal supplier of drinking water for the zone. At the study area, the intense withdrawal of groundwater by a petrochemical plant produces a depression cone. The 'Pampean Silts' contain the unconfined aquifer (8–9 m) and the semiconfined Pampeano aquifer (18–24 m).

2.3. METHODS

2.3.1. *Sampling plan*

During June 1991, a preliminary sampling was performed in the three dump sites, to evaluate the presence of heavy metals and their variations among zones. The presence of water along the whole section of the dump could produce, on one hand, an interconnection of leachate between zones and, on the other hand, a greater homogeneity within each zone than in a sanitary landfill. Two samples were taken at different depths (5 and 9 m) in the central site to evaluate differences related to depth. According to the results obtained and the characteristics of the system, a sampling plan including three samplings of leachate from each site was performed. The samplings were carried out in November 1991, before the beginning of the reclamation works (BWS), in April 1992, near the end of the works (EWS), and in August, four months after the works were finished (AWS). At least one sample was collected from each site. It was only during the EWS and AWS that the superficial leachate, produced as a result of the works of covering and compaction, was sampled. A total of 15 samples was obtained including the preliminary sampling.

The samples were extracted at the point of maximum possible depth by means of perforation with a trial-pit. The extraction involved sinking a PVC pipe and manually pumping with pipes of polypropylene with a retaining valve. The samples were extracted at different depths ranging from 1–9 m. Temperature and conductivity were recorded in the field. Leachate samples were filtered and transported

TABLE I

Mean and ranges of the analyzed variables in the leachate samples

Parameters	n^a	Mean	Min.	Max.
Depth (m)	11	3.2	O	9
Temp ($^{\circ}$C)	11	23	18	31
pH	11	7.4	6.9	8.32
Conductivity (μS cm^{-1})	11	3749	1350	8500
Total Solids (mg l^{-1})	8	22041	1263	119285
Alkalinity (mg l^{-1})	11	1769	180	6074
Hardness (mg l^{-1})	11	819	335	1880
Chloride (mg l^{-1})	10	727	116	1901
Sodium (mg l^{-1})	7	180	99	516
Potassium (mg l^{-1})	7	245	118	668
Calcium (mg l^{-1})	11	196	54	490
Magnesium (mg l^{-1})	11	68	21	160
Sulphate (mg l^{-1})	10	14	7	33
Nitrate (mg l^{-1})	7	2	n.d.	15
Ammonium (mg l^{-1})	8	58	10	121
TNK (mg l^{-1})^b	8	399	107	1142
COD (mg l^{-1})^c	8	6675	398	27555
Chromium (μg l^{-1})	11	57	n.d.	170
Lead (μg l^{-1})	11	41	n.d.	120
Mercury (μg l^{-1})	11	0.2	n.d.	2
Copper (μg l^{-1})	10	42	n.d.	140
Iron (μg l^{-1})	11	5760	50	35000
Zinc (μg l^{-1})	11	130	n.d.	600

^{a} Samples number.
^{b} Total nitrogen by Kjeldahl.
^{c} Chemical oxygen demand.
n.d.: not detectable.

to the laboratory in plastic bottles. Nitric acid, 1/1000, was added to an aliquot of the supernatant solution containing the dissolved heavy metals in order to avoid their adsorption on the bottle walls and preserve their contents up to the moment of analysis (Inland Waters Directorate, 1979).

2.3.2. *Sample analysis*

Chromium, lead, mercury and some physical-chemical variables were determined in the preliminary sampling. Others parameters included in Table I were analyzed subsequently. Physical and chemical variables were determined according to standard methods (APHA, 1981). Heavy metals were determined in suspended solids as well as in the liquid fraction of the leachate samples. Suspended particles were removed by centrifugation and filtration through 0.45μ Millipore

filters in the laboratory. The suspended solids were digested in closed recipients with a $HF/H_2SO_4/HClO_4$ mixture. An atomic absorption spectrophotometer with different sources of atomic production was used for metals determination. Lead, chromium, iron, zinc and copper were analyzed by electrothermal atomization with graphite furnace (GFA) at levels below 50 ng ml^{-1} and atomic absorption spectrometry with an air-acetylene flame was used for the determination of higher levels. Mercury was determined using the technique of cold atomic vapor generation (Fuller, 1977; Minoia and Caroli, 1989). Analyses were performed with an atomic absorption spectrometer (Shimadzu 6501) coupled to the device for electrothermal atomization Shimadzu GFA 2000 and autosampler ASC 2000 when it was necessary.

Determination limits for each element in solution were: Cr: 0.02 mg l^{-1} (ETA-AAS); Pb: 0.01 mg l^{-1} (ETA-AAS); Fe: 0.02 mg l^{-1} (FAAS); Cu: 0.01 mg l^{-1} (FAAS); Zn: 0.02 mg l^{-1} (FAAS); and Hg: 0,002 mg l^{-1}.

2.3.3. *Statistical analysis*

Ordination of samples based on chemical data was performed using principal component analysis (PCA) to simplify the description of the set of interrelated variables without losing much of the information (Afifi and Clark, 1990). PCA was considered an exploratory technique useful in gaining a better understanding of the interrelationships among the variables and the changes produced by the reclamation works.

Data were standardized to reduce the variables to a common scale (Pielou, 1984). All samples, except those of the preliminary sampling, were used. The variables in Table I selected for PCA were those determined in 10 or more samples. Mercury was rejected because all data (except one) were below the determination limit. Calcium and magnesium variations were not considered because hardness was included.

3. Results

The average concentrations and the ranges of the variables analyzed in the leachate samples are shown in Table I. With respect to the temporal variation of the chemical variables at Sites 1 and 2 (Table II), the levels of conductivity, alkalinity, hardness, and chloride were increased during the EWS, returning approximately to their initial levels during the AWS except for chloride at Site 2. Total solids, COD and TNK declined during the AWS in respect of EWS. The concentration of different nitrogen compounds could give a comparative idea of the redox environment. Sites 2 and 3 appeared to have the most and the least reduction conditions respectively. The analysis of Site 2 and superficial samples (puddles) at Site 1 revealed a decrease of total nitrogen during the AWS in respect of EWS together with a proportionate increase of ammonium. COD, total solids, and the ammonium proportion at Site

TABLE II

Chemical composition of leachate from the three sites of the dump during study period

	Site 1			Site 2			Site 3		
	BWS[a]	EWS[b]	AWS[c]	BWS	EWS	AWS	BWS	EWS	AWS
pH	7.2	7	7	8.3	7.6	6.9	8.1	7.2	7.1
Conductivity ($\mu S\ cm^{-1}$)	2600	6500	1850	6000	8500	1850	3600	2700	1638
Alkalinity ($mg\ l^{-1}$)	1090	5064	1121	180	6074	974	180	1127	1140
Hardness ($mg\ l^{-1}$)	783	1880	485	577	1184	486	653	482	335
Chloride ($mg\ l^{-1}$)	196	1285	116	848	1332	1901	892	193	–
Total Solids ($mg\ l^{-1}$)	–	119285	3730	–	10986	1263	–	1570	34955
COD ($mg\ l^{-1}$)[d]	–	27555	1422	–	12782	1550	–	398	6552
TNK ($mg\ l^{-1}$)[e]	–	245[f]	107[f]	–	1142	109	–	150	868
NH3/TNK.1000	–	40[f]	963[f]	–	0.2	8.8	–	65.3	102.5
NO3/TNK.1000	–	0.5[f]	1.7[f]	–	0.01	0.04	–	100	0.21

[a] Before works sampling.
[b] Ending works sampling.
[c] After works sampling.
[d] Chemical oxygen demand.
[e] Total nitrogen by Kjeldahl.
[f] Superficial samples.

TABLE III

Heavy metals in solid fraction of leachate samples

Sample depth (m)	n[a]	Site 1			Site 2			Site 3		
		Mean	Min.	Max.	Mean	Min.	Max.	Mean	Min.	Max.
		5	2	9	5	4	8	6	5	7
Chromium	4	70	43	94	85	32	132	307	103	649
Lead	4	125	60	187	117	104	123	203	75	290
Mercury	4	1.6	0.6	2.7	1.2	0.8	2.1	1.9	0.9	4.0
Iron	3	186	124	237	161	148	172	259	177	391
Copper	3	93	58	127	91	33	135	103	63	133
Zinc	3	329	195	462	219	150	346	266	209	308

[a] Samples number.
All units are in mg kg^{-1}, except iron in mg kg^{-2}.

3 showed similar increments. However, conductivity, alkalinity and hardness did not present such behaviour.

Metals presented higher concentrations in solid fraction than in solution. Adsorbed metal concentrations also showed relatively lesser variations than dissolved ones between the samplings. Site 3 presented the greatest levels of adsorbed metals. Table III shows these values, including the samples of preliminary sampling. The concentrations for each dissolved and solid fraction in the samples of all the sites are shown in Figures 2 and 3 respectively.

At Site 1, the levels of dissolved metals such as Cr, Pb and Cu, increased during the EWS (Figure 2). After the reclamation works the levels of these dissolved metals diminished. An outstanding decrease in all metal levels in the solid fraction was observed during the EWS (Figure 3).

At Site 2, all metals in the solution showed a strong increase in concentration during the EWS. Traces of Hg, always detected in the solid fraction, were determined in solution only at this site at this time. The effect of recuperation during the AWS was observed to a different extent for all metals, except Zn, perhaps due to its higher possibility of hydroxyl complex formation (Figure 2).

The behaviour of metals at Site 3 is shown in the same figure. Dissolved Pb, Cu and Zn, increased in a delay time in respect of Sites 1 and 2. Iron, which was the only metal that increased in the former sampling, diminished at both its dissolved and absorbed levels. The temporal evolution of the sites is well evidenced in the PCA of samples performed on standardized values of the 12 chemical variables mentioned above (Figure 4). The temporal sequence for each site has been marked with solid lines in the figure. The first three factors explain over 77% of the total variance in the data. Factor 1 (44% variance explained) accounted for the temporal evolution at Sites 1 and 2, correlated with conductivity, alkalinity, Pb, Cu and hardness. At both Sites 1 and 2 the levels of these variables increased during the
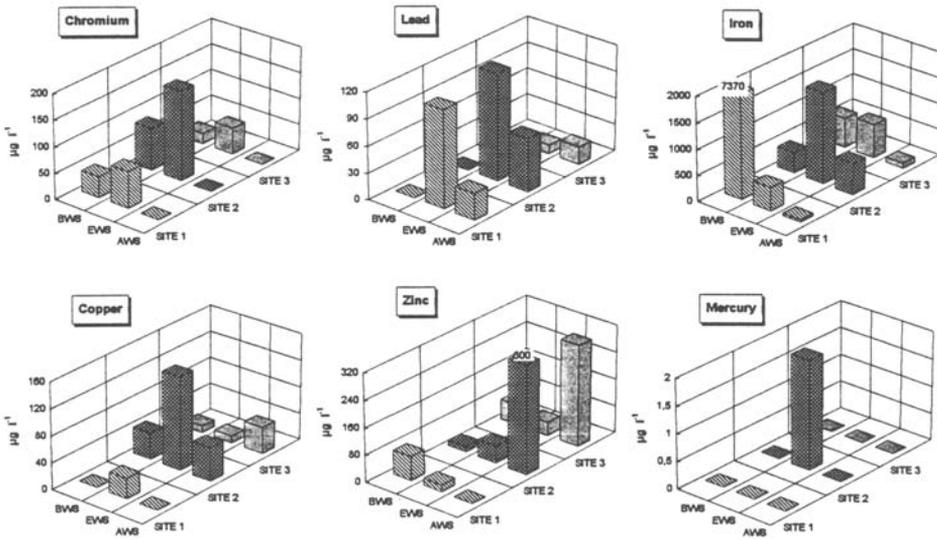
Fig. 2. Temporal variation of dissolved metals in the three sampling sites. BWS: before works sampling; EWS: ending works sampling; AWS: after works sampling.
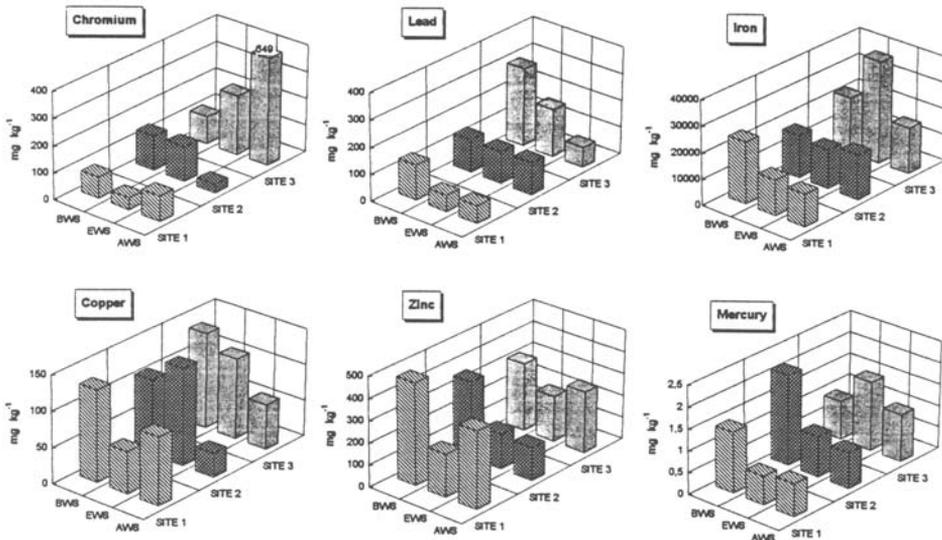
Fig. 3. Temporal variation of adsorbed metals in the three sampling sites. BWS: before works sampling; EWS: ending works sampling; AWS: after works sampling.
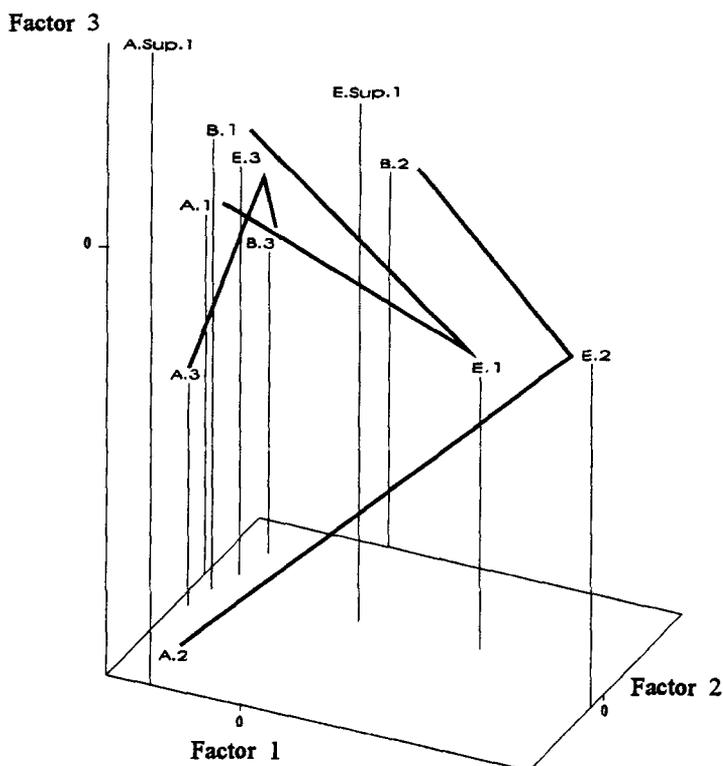
Fig. 4. PCA ordination of samples. B: BWS, E: EWS, A: AWS, Sup: superficial samples, 1, 2, 3: sampling sites.

EWS (samples displayed on the right of the axis) recuperating their initial levels after reclamation works. Factor 2 explains 19% of the total variance and represents changes in chemical parameters for Sites 2, 3 and to a lesser extent Site 1, that occurred during the reclamation works. This component is correlated with Zn and pH, which, in turn, are inversely related. The BWS samples are displayed on the upper part of the axis and the AWS samples on the lower portion with increased concentration of Zn and lesser values of pH. Factor 3 accounted for 14% of the total variation, related to chloride and Fe, which in turn, were inversely related. Thus, superficial samples with a high concentration of Fe and low concentration of chloride are displayed on the top. In contrast, the AWS of Sites 2 and 3 are displayed on the bottom part of the axis coincident with the decrease in Fe concentration.

4. Discussion and Conclusions

The ranges of the variables determined are similar to those found by other authors for different kinds of waste dumps (Canter *et al.*, 1987; UNESCO, 1980; Pohland

and Harber, 1986; Förstner and Carstens, 1989; Monoharan *et al.*, 1989; Gourdon *et al.*, 1989). Considering that this system is permanently saturated with rain and underground waters which dilute the leachate, this similarity was not expected. The continuous contact of wastes with aquifer-oxidized water could probably have increased decomposition and chemical processes involving the dissolution of substances, and thus would compensate for the dilution effects of the leachate.

Our results show that the reclamation works affected the leachate composition. The effects were evidenced principally by the increase in concentration of several parameters in the leachate samples of Sites 1 and 2. The PCA summarizes the relevant chemical parameters affected by the works at these sites. The chemical parameters which increased during the EWS could reflect changes in factors such as buffer capacity (alkalinity), ionic strength/activity (conductivity), migration potential (chlorides), and redox environment (nitrates and sulfates) (Pohland and Harber, 1986). For instance, the global increase of the ammonium/total nitrogen ratio during the AWS at Sites 2 and 3, and the superficial samples at Site 1, could reflect the increase of the reduction power due to oxygen diminution, related to the residues compaction.

With regard to heavy metals, pH has been cited as the principal factor influencing its mobility (Warren and Zimmerman, 1994; Kargbo, 1994; Basta and Tabatabai, 1992). But pH variations (6.9–8.3) in this system were not enough to explain the increase of dissolved heavy metals during the EWS. These increments could have been produced by other changes in the chemical environment, favoring the passage of heavy metals to solution. Consequently, variables such as conductivity, chloride, bicarbonates and COD would play the main role in determining the process of heavy metals partitioning in this system. COD and chloride, for instance, promote the formation of soluble complexes of heavy metals (Förstner and Carstens, 1989; Knox and Jones, 1979).

At Site 3, the changes in leachate composition were reflected later (AWS), by the increase of COD, TNK, dissolved Pb, Cu, Zn and adsorbed Cr. The distinct behavior of Fe involving increases at Sites 2 and 3 during the EWS, and diminution at AWS in all sites, could be related to their great mobility.

PCA analysis indicates that Fe is the main component in superficial samples. These data are relevant considering that the leachate from the puddles flowed into the waters of the Las Piedras stream. Some samples of the stream (unpublished data) showed a greater concentration of Fe in the waters located downstream of the dump than those upstream. Other metals could also dissolve in the leachate and migrate to the Las Piedras stream.

The results for adsorbed metals show, on one hand, a decrease since the works, because of the diluting effect of the covering earth in Sites 1 and 2. On the other hand, Site 3 would reflect the historical contamination of the system, due to its location and characteristics. The maximum levels of adsorbed metals were registered in this site.

Many studies performed under laboratory conditions have explained successful-ly the influence of leachate characteristics on metal mobility (Fuller, 1980; Förstner and Carstens, 1989; Lietz and Galling, 1989; Basta and Tabatabai, 1992). Our study has attemped to present information about global changes in leachate composition dealing with chemical details, which are very difficult to control in this system.

Some authors argue that the high precipitation and temperature increase the rate of leachate production, attenuating the contaminant levels by dilution (Pohland and Harber, 1986; UNESCO, 1980). Others, such as Förstner and Carstens (1989), argue that 'oxidation of sulphidic minerals by intruding rainwater could mobilize trace metals, involving dissolution and reprecipitation during the passage of oxidized water through the deposit, which would concentrate critical elements'. In our case, the EWS coincided with the period of lowest accumulated water as well as with the month posterior to the period of greater precipitation and temperature of the year. But we consider that if weather factors were the cause of the changes observed in the leachate, they would have affected the sites to the same extent, which was not the case in this study. Moreover, the permanent saturation of residues due to the contact with aquifer reduces the importance of rainwater effects.

Our results confirm that the reclamation works were the determining factor causing the changes detected in the leachate composition during this study. Some reclamation works in different systems may mobilize the adsorbed heavy metals, such as dragging works in rivers. In the case of dumps, several reclamation works are used by minimizing the amount of percolated rainwater through the deposited residues such as the waste compaction and impervious earth covers. But these operations 'would be highly ineffective in situations where the moisture source originate from below the dump' (Zanoni, 1972). Also, the earth cover used in the actual dump was not made of impervious material. Our results show that the changes produced by the earth cover and compaction in the leachate, could produce aquifer contamination by heavy metals. The recovery of some parameters to initial levels may be explained by the migration of certain chemical species from Sites 1 and 2 to Site 3, and/or its percolation to deeper zones, as in the Puelche aquifer. In the future, the increase of pollution measured during the EWS could affect domiciliary wells. Further studies are necessary to determine the contaminants migration to the aquifers. Data obtained from wells of this area are being analyzed to reveal further aspects of this problem.

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