

ISBN 0 9521673 0 1



International Conference on

ENVIRONMENTAL POLLUTION

Science • Policy • Engineering

PROCEEDINGS: VOLUME ONE

Edited by

B. Nath

L. Candela

L. Hens

J. P. Robinson

EUROPEAN CENTRE FOR POLLUTION RESEARCH

Proceedings of ICEP.2, Volume 1

Published by the *European Centre for Pollution Research*, Queen Mary & Westfield College, University of London, London E1 4NS, UK

© 1993 European Centre for Pollution Research

ISBN 0 9521673 0 1

All rights reserved; no part of this publication may be reproduced, stored in any retrieval system, or transmitted by any means, electronic, mechanical, photocopying, recording or otherwise without the prior written permission of the publisher.

Printed and bound in Budapest by the AKAPRINT Press of the Hungarian Academy of Sciences.

The Editors and the Publisher wish to make it clear that any statements or views expressed in the papers contained in this Volume are those of the authors, and no responsibility is taken for the accuracy of such statements or views. Occasional reference to manufacturers, trade names and proprietary products may be inevitable in order to achieve factual reporting. However, no endorsement of named products is intended nor is any criticism implied of similar products which are not mentioned.

ASPECTS OF CONTAMINATION PRODUCED BY DOMESTIC WASTE LANDFILLS ON RECEIVING WATERS IN MADRID PROVINCE

**J. Pastor*, A. Urcelay*, M.J. Adarve†, A.J. Hernández†
and A. Sánchez***

*Centro de Ciencias Medioambientales, CSIC, Madrid, Spain

†Dpto. Geología, Universidad de Alcalá de Henares, Madrid, Spain

ABSTRACT

This study describes some aspects of the anion content in surface waters and ground waters as well as in the soils affected by three landfills in the Province of Madrid. The anions concerned are chlorides, fluorides, sulfates, phosphates and nitrates. The pH and conductivity were also determined. These parameters may constitute abiotic indicators to observe the alterations produced in the water and soil by the leachates from the landfills. The results show that the concentrations of the parameters analyzed increased considerably in some water and soil samples from the surroundings of the landfill. It is important to determine the effects of these anions on the water and soil and to create predictable patterns in order to protect or restore them.

KEYWORDS: landfill, leachate, surface water, ground water, soils, contamination.

1. INTRODUCTION

Rivers and groundwater are the main water supply sources for human beings and consequently the quality of such water needs to be maintained. In recent decades most rivers and aquifers have been polluted as a result of excessive silting due to deforestation, urbanization, cultivation, industrialization and various other human activities. Municipal and domestic wastes, industrial effluents and agricultural run-off are the main factors contributing to water pollution.

When water percolates through solid municipal wastes in a landfill, it dissolves inorganic and organic components which result from the degradation processes. They produce contaminated leachates which may constitute a large potential health risk for both the surrounding ecosystems and the population. The factors influencing the affection in water and soil include leachate composition and volume, regional climate, hydrogeological characteristics such as substratum porosity, permeability and composition, landfill proximity to surface waters and the depth of the water-table, annual rainfall, refuse quantity and quality, exploitation characteristics and age of the landfill site.

The chemical contaminants of the landfill sites near a stream are likely to be released into the stream. This may influence aquatic life and also constitute a potential health hazard for the public at large. Through leaching the released contaminants are eventually dispersed in the stream, but some of them may also be adsorbed by the sediment. The contamination of surface water, the underlying groundwater and bioaccumulation in the surrounding biota are possible routes by which these landfill leachates affect both environment and human population.

There is consistent information about the composition of landfill leachates^{(1),(2)}. Robinson and Maris (1983)⁽³⁾ studied the chemical composition of the adjacent ground- or surface waters, Murray et al.

(1981)⁽⁴⁾ tried to evaluate the degree of groundwater contamination through landfill leachates, and Cyr et al (1987)⁽⁵⁾ described physico-chemical data of the leachates and their inorganic contents, particularly heavy metals.

Among landfill pollutants, inorganic soluble anions have acquired growing importance because they control the entire ionic balance of the ecosystems. Their pollutant effects are in the process of being well understood^{(6),(7),(8)}. Several authors note that some inorganic contaminants from a site, such as anions continue to leach for decades. The anion components include chlorides, fluorides, sulphates, phosphates and nitrates. Some of these anions are key elements for plant growth (nitrates, phosphates and sulphates). Some are important for the nutrition of soil organisms and others are elements that can be harmful or toxic for animals and plants, e.g. fluorides⁽⁹⁾. It is therefore relevant to determine the release of chemical contaminants from municipal landfills.

This study continues previous research^{(7),(10),(11),(12)}. The overall aim of the project is to predict local trends of landfill evolution as regards the soil, behaviour of rainfall leachates from landfills and the surface and groundwater affected by them.

2. MATERIAL AND METHODS

2.1. Landfill characterization.

The landfills studied were sealed between September 1986 and April 1988. They have an undulating profile with slopes reaching a height of between 5 and 15 metres and store municipal waste. These landfills were not controlled because prior to their exploitation and sealing no environmental controls were carried out to prevent contamination by leachates of the area outside the landfill. The surface soil of the landfills is clearly of a sandy loam texture with outstanding coarse sand and rather abounding in stones. The range of texture, however, varies from sandy loam to clay. The waste is generally found at about 30 cm from the surface although in some cases it lies at no more than 10 cm. The points with the greatest depth reach about 80 cm.

The landfills lie in the north of the Province of Madrid (Figure 1). Their geological features are quite uniform, and consist of Paleozoic gneisses as well as granite rocks of the Guadarrama Mountains at an altitude of between 850 and 1100 m. The soils are oligotrophic, have an acid pH and low nutrient content. It is grassland used for grazing cattle and for timber.

The hydrological conditions of the landfills do not differ very much. The annual rainfall in the area ranges from about 750 to 800 mm. The monthly mean temperature lies between 4^o C in January and 23.5^o C in July. The streams affected by the landfills carry an irregular volume of water due to the unequal distribution of the rain and to the characteristics of the rocky substratum. This means that most streams are dry in the summer.

The material is not very permeable and makes it difficult or impossible for leachates to get into the "aquifers". As a result most of the leachates are scattered around the surface and affect the streams near the landfills. However, the leachates on the surface or in the substratum ("hard rocks") move fast, especially when there are many fractures and fissures, and can contaminate the springs in the vicinity of the landfills. The groundwater in the hard rocks move along the fractures or fissures of the studied area, following the direction of the slopes with the steepest gradient, a fact described by González (1991)⁽¹³⁾. The direction of ground- and surface water in the landfill areas is illustrated in Fig. 1 and the arrows mark the flow of the groundwater.

The hard rocks consist of minerals which do not break down easily. This means that the ground- and surface water is of good quality. Water conductivity from the springs at the top of the mountain lies between 10 and 20 $\mu\text{S}/\text{cm}$ and that from the springs at lower contours between 50 and 100 $\mu\text{S}/\text{cm}$ ⁽¹⁴⁾.

2.2 Analytical procedures

The water and leachate samples from three landfills were collected in plastic-lined drums after winter and spring rains in 1989 and 1990. The samples were stored at 4 + 1^o C. The leachates are understood to

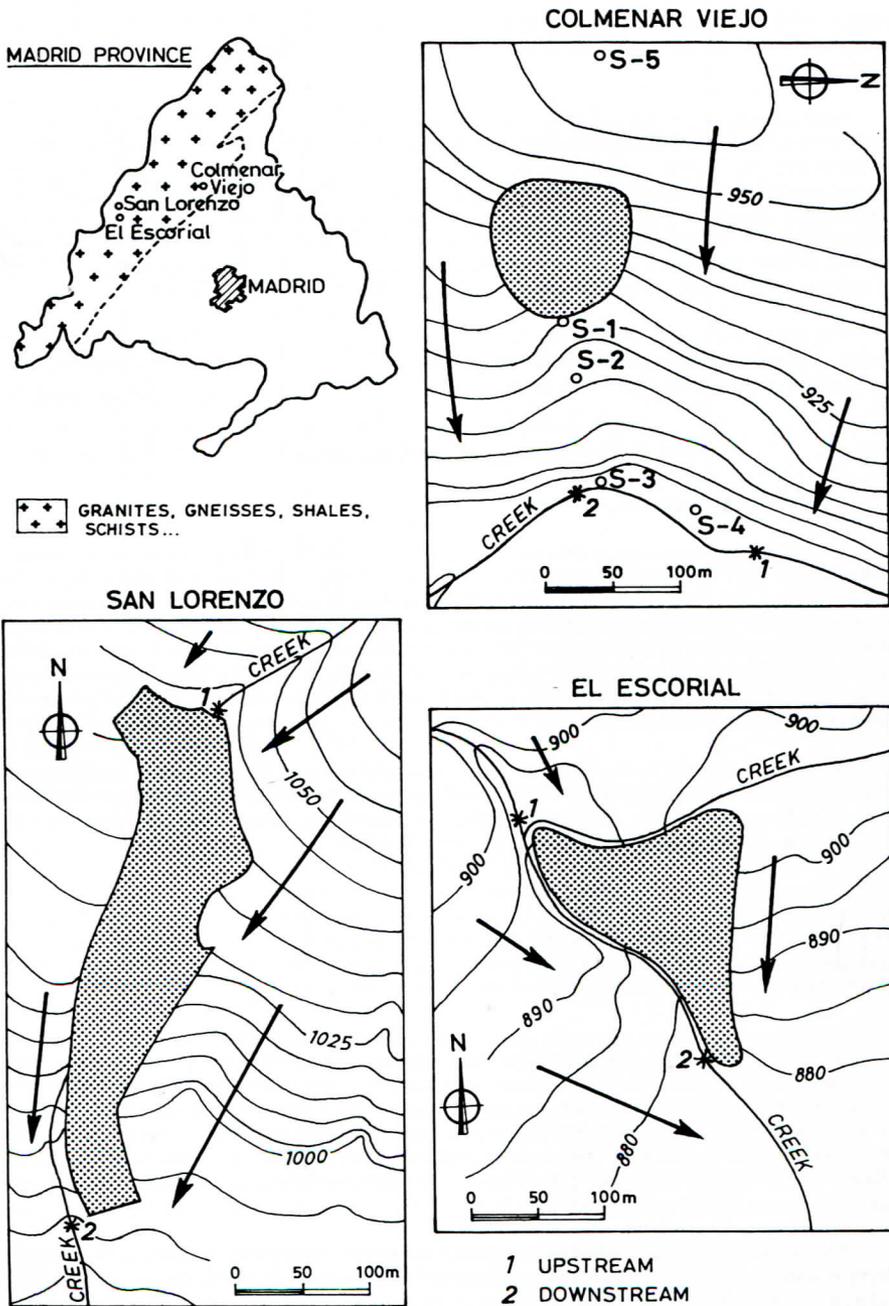


FIGURE 1. Topographic map and location of the landfills.

be rainfall leachates since they are a combination of the water running off the surface of the landfills and of filtered water (in contact with the waste products), which flows into the groundwater or the surface streams after percolating through the wastes. More samples were collected from surface waters, ie, from streams or creeks, and from springs.

Soil samples were collected from the Colmenar Viejo landfill area in June 1991. These samples were dried at room temperature and then centrifuged to extract the aqueous content. All samples were filtered adequately when necessary. The anion concentration of the water, leachates and soil samples was analysed by means of ion chromatography, following the method described by Urcelay and Pastor⁽¹⁴⁾. The equipment used was a Dionex Model 10 Chromatograph, with an AG4A precolum, an AS4A separator column and an AHMS suppressor column, connected to a register and a Hewlett-Packard 3390A integrator. The anions were measured immediately after sample collection.

3. RESULTS AND DISCUSSION

Tables 1,2 and 3 summarize the analytical results of studying the soluble anion content in the different water sample (expressed in mg/l) from the landfills, the surrounding streams, springs and creeks. Nitrites and phosphates are practically non-existent. The lowest anion concentrations always correspond to F⁻, since fluorides and phosphates do not break down easily. The content of other, more soluble anions is greater.

Table 1 shows the chemical data obtained from characterizing the leachates of the waste landfills. The leachates are scattered more widely in the environment as the degradation period progresses. Rainfall leachates from landfills are acid and have a high inorganic anions content (F⁻, Cl⁻, NO₃⁻ and SO₄²⁻). This shows that the landfills are a source of pollution for the nearest ecosystems, even if the leachates are diluted by rainwater, especially in the rainy season. Even within the same landfill the quality of the leachates changes gradually, not only because of the amount it rains, but because some waste components break down quickly, while others do so slowly. After comparing these values with those given by various authors from different countries^{(15),(16),(17),(18),(19),(20),(21)}, it was found that in the landfills of this study the leachate pH remained in the lower ranges (4.7-5.9 vs 4.0-9.0). The same as in the bibliography, the NO₂⁻ and PO₄³⁻ content was practically non-existent and is therefore not given in the Table. The Cl⁻ and SO₄²⁻ content coincides with that of the authors mentioned above, while the F⁻ and NO₃⁻ content is clearly higher in the present study.

TABLE 1 pH, conductivity and anion content in landfill rain leachates.

	pH	CONDUCT (μ s/cm)	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
			----- mg/l -----			
Dec-January	5.0	2180.0	7.3	590.5	178.7	350.8
	5.2	2100.0	6.7	535.5	203.7	262.5
	4.7	4440.0	7.1	849.0	416.8	510.8
	5.9	3560.0	7.0	1433.0	111.7	348.3
February	5.5	4770.0	7.4	1820.0	377.3	455.3
	5.6	4700.0	7.2	1847.0	395.0	476.5
	5.6	4800.0	7.7	1829.0	380.0	462.5
April	5.3	14110.0	8.1	7160.0	1290.0	580.0

The results show that nitrate leaching is significantly higher than the values reported for another landfill in the same area, ie, the Collado-Villalba landfill studied by Fernandez-Serrano et al. (1992)⁽²²⁾. The reason for that is the fact that the landfills in this study lie in heavily grazed areas. The pH levels, conductivity and anion content increased in the course of the study.

It was also observed that the conductivity and the anion content of the leachates are in agreement with the temperature and rainfall conditions during the time of the study. Thus, the analysis of the samples corresponding to December-January are more diluted because of the high rainfall in that period

(250-350 mm). The samples from February, when the rainfall was very low (10 mm), clearly show a greater concentration. The spring samples, finally, when rainfall was sufficient, but not high (80 mm), already show a greater microbial activity typical for springtime (mean temperature 10.7°C) and a high anion content together with very high conductivity.

The parameters studied at the springs and creeks thought to be affected by the landfills are shown in Table 2. Except those for nitrate, the values of all the physico-chemical parameters measured in these waters were found to be below the maximum permissible level and lower than those of landfill leachates. The ability of groundwater to reduce these leachate pollutants is usually neglected in the assessment of groundwater pollution risks. Contamination may be the consequence of surface filtrations due to the thin layer of soil on the landfill or to the accidental introduction of leachates in the aquifers. Sampling points may also be affected by the groundwater following the fractures in the rock. In fact the water samples collected at the springs close to the landfill had much higher Cl⁻, NO₃⁻ and SO₄²⁻ concentrations than those found by González (1991)⁽¹³⁾ at springs of the natural mountain environment not affected by landfills. The Cl⁻ values of the last samples range from 1.4 to 3.5 mg/l; those of SO₄²⁻ from 1 to 9 mg/l and those of NO₃⁻ from 0 to 0.06 mg/l. As was pointed out earlier, the NO₃⁻ content cannot entirely be attributed to the landfills since there are other sources of pollution, such as the cattle grazing activity of the region. The conductivity range of these "natural" springs varies from 43 to 79 µs/cm, which is also much lower than the values found at the springs near the landfill.

The pH, Cl⁻ and SO₄²⁻ values, however, found at the springs in the present study clearly have much lower levels than those of the groundwater samples from the Collado Villalba landfill⁽²²⁾ near El Escorial. The F⁻ and NO₃⁻ levels, on the other hand, are much higher in the samples of this study. That is due to the more intense grazing activity in the areas studied if compared with the important second residence developments abounding in the surroundings of the Collado-Villalba landfill.

The main differences between the affected springs and creeks as regards the chemistry of their water the Cl⁻ and NO₃⁻ levels, which were clearly lower in the creeks, while the F⁻, pH, conductivity and SO₄²⁻ values, on the other hand, were rather similar.

TABLE 2 pH, Conductivity and anion content in the springs and creeks near the landfills.

	pH	CONDUCT (µs/cm)	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻
			----- mg/l -----			
SPRINGS						
Dec-January	5.8	103.2	0.2	5.7	21.4	25.7
	6.6	167.1	0.7	20.2	0.2	33.7
	6.8	87.2	0.6	5.0	0.2	8.4
	6.6	418.0	0.7	19.8	51.3	84.3
	7.0	368.0	0.7	44.9	27.8	80.6
CREEKS						
Dec-January	5.9	66.1	0.8	3.5	16.6	13.2
	7.3	194.7	0.6	4.1	15.5	35.1
	7.0	291.0	0.2	5.4	9.2	74.4

In the samples from closer to the landfills, higher anion concentrations were observed than in the streams affected by the landfills (Table 3). As the distance from the landfills grew, the anion concentration became lower, ranging near that of the stream.

Table 3 shows the results obtained for the upstreams and downstreams samples of the streams directly affected by the landfills. In physico-chemical terms, the streams were found to be below permissible limits except for nitrates. On several occasions the nitrate concentration exceeded 10 mg NO₃⁻/l in affected waters. The ionic concentrations of chloride, sulphate and fluoride was always below the maximum limit⁽²²⁾. As regards the concentrations there were differences between the upstream and downstream samples of the landfill, with the higher concentrations logically in the latter. However, these downstream concentrations are much lower than those of the rainfall leachates, which shows how much the contaminating power of these leachates is reduced by their passing through the soil. The nature and

volume of the receiving water and its diluting capacity certainly has a significant effect on the damage potential of the discharge of leachates⁽¹⁶⁾.

TABLE 3 pH, conductivity and anion content in the streams (upstream and downstream) surrounding the landfills.

	pH	COND ($\mu\text{s}/\text{cm}$)	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ⁻²
			----- mg/l -----			
UPSTREAM						
Dec-January	6.5	73.0	0.7	3.4	4.1	11.2
	6.9	29.1	0.4	1.9	0.3	2.8
	6.9	66.6	1.0	15.4	6.7	15.8
	6.7	72.8	0.8	3.4	2.3	11.3
	7.5	25.3	0.2	1.4	0.4	2.3
February	6.7	75.6	0.8	3.8	2.1	11.1
	6.8	26.6	0.6	1.2	12.5	2.5
	6.7	336.0	0.6	9.0	23.1	99.5
April	6.8	25.5	0.2	1.2	0.7	2.1
	6.4	100.8	0.5	4.2	0.4	8.9
DOWNSTREAM						
Dec-January	6.6	112.6	0.2	6.1	4.0	21.7
	6.8	167.2	0.0	11.9	10.6	40.1
	6.7	148.0	0.8	10.3	5.2	13.5
	6.5	90.4	0.7	5.0	2.3	9.8
	7.1	227.0	0.6	16.7	16.7	44.4
February	6.3	560.0	0.9	7.3	25.7	37.0
	6.8	194.7	0.7	8.9	9.6	26.6
	6.9	359.0	0.6	9.8	18.9	101.6
April	6.6	182.7	0.3	4.2	7.8	27.5
	6.7	103.6	1.2	5.2	0.5	8.7

The analytical values obtained from the soil samples (Table 4) are very varied, but they decrease significantly as the distance from the landfills increases. Figure 1 shows where the sample points were situated. Soil sample S-1 was only affected by the run-off of surface water. Soil sample S-2 has the highest levels because it comes from the area with the rainfall leachates. The high amounts of nitrates in the soil are not all a consequence of the landfill, but are also due to heavy cattle grazing. The chemical parameters of soil samples S-3 and S-4 were lower than those of sample S-2, because of the soil filtration effect and in view of the distance between the sampling points (100 and 150 metres).

TABLE 4 pH, conductivity and anion content in the soil samples affected by the Colmenar Viejo landfill. (June-1991)

LOCATION	SAMPLE Nº	pH	CONDUCT ($\mu\text{s}/\text{cm}$)	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ⁻²
				----- mg/kg -----			
Landfill cover	S-1	5.80	1230	*	238	9	25
Landfill bottom	S-2	4.80	17000	*	2133	388	645
Landfill bottom	S-3	5.40	4190	*	355	220	90
Landfill bottom	S-4	5.60	790	*	238	135	83
Surrounding landfill	S-5	7.10	1640	*	205	98	175

* traces

4. CONCLUSIONS

The results described above suggest that the higher levels of contamination are found in the immediate vicinity of the landfills nearest to the bottom edge. The surrounding springs influenced by landfill leachates have high anion concentrations. It was also observed that these concentrations decreased significantly as the distance from the landfills increased. The landfill impact on the water which is most directly affected is summarized as follows: the pH is reduced by an average one tenth, Cl⁻ increases by about 4 ppm, NO₃⁻ by 5 ppm, SO₄⁻ by 16 ppm and conductivity by about 131 μs/cm. These are significant figures view of the important volume of water in the streams at sampling time. The differences between the up- and downstream samples of the affected streams also showed that the landfills had a greater environmental impact on the water in February.

The power of the leachate is diluted by other surface water and/or reduced in its passage through the edaphic materials. Due to the dilution effect it was found that the concentration of these contaminants was reduced considerably especially in the affected surface streams.

From the point of view of landfill management a compromise must be reached by monitoring these sites in order to assess the abiotic state of the landfill environment. For this purpose the use of sensitive plant species or abiotic indicators appear to be advisable.

It may be concluded that anionic contaminants are released in siliceous landfills. The levels in nearby areas are high, whereas, for the moment, they remain within an acceptable range in areas that are further away. The anion levels in neighbouring streams affected by landfills are in general also within an acceptable range. However, periodic control of the sites for contaminant levels is recommended to assure environmental protection of the land.

ACKNOWLEDGEMENT

This research project was financed by the Autonomous Communities of Madrid and Castilla-La Mancha.

REFERENCES

1. Ho, S, Boyle, W.C. and Ham, R.K, 'Chemical treatment of leachates from sanitary landfills', J. Water Pollut. Control Fed. 76, pp 1776-1791 (1974)
2. Apgar, M.A. and Langmuir, D, 'Ground water pollution potential of a landfill above the water table', Groundwater 9, pp 76-81 (1971)
3. Robinson, H.D. and Maris, P.J, 'The treatment of leachates from domestic wastes in landfills', Water Res. 17, pp 1537-1548 (1983)
4. Murray, J.P, Rouse, J.V. and Carpenter, A.B, 'Ground water contamination by sanitary landfills leachates and domestic water in carbonate terrain' Water Res. 15, pp 745-757 (1981)
5. Cyr, F, Mehra, M.C, and Mallet, V.N, 'Leaching of chemical contaminants from a municipal landfill site' Bull. Environ. Contam. Toxicol. 38, pp 775-782 (1987)
6. Pastor, J, Alia, M, Hernández, A.J, Gil, A. and Urcelay, A, 'Inorganic contaminants from disturbed soils and municipal landfill sites in central Spain', ISWA. Proc. 6th International Solid Wastes Congress. ATEGRUS, Madrid (1992)
7. Alia, M, Urcelay, A, Gil, A, Bello, A, and Pastor, J, 'Toxic effects of leachates from urban landfill. The 1991' Eurotox. Congress. Maastricht. The Netherlands. pp 224 (1991a)
8. Alia, M, Pastor, J, Urcelay, A, Gil, A y Bello, A, 'Estudio de la toxicidad de los lixiviados procedentes de un vertedero clausurado en un municipio de la Sierra de Madrid'. En Medio Ambiente: V Encontro Galego-Portugues de Química, pp 273-274 (1991b)

9. Alía, M, Urcelay, A, Gil, A, Bello, A. and Pastor, J, 'Estudio ecotoxicológico de fluoruro procedentes de vertederos de residuos sólidos urbanos' Rev. Toxicol. (en press.)
10. Pastor, J, Adarve, M.J, Urcelay, A. y Hernández, A.J, 'Aproximación al estudio del impacto ambiental de los residuos urbanos sobre el medio natural y social'. En Residuos sólidos y líquidos: su mejor destino. Ed. ANQUE, Madrid, pp 55-67 (1991)
11. Pastor, J, Hernández, A. J, Adarve, M. J, and Urcelay, A, 'Some chemical characteristics of sedimentary soils in the mediterranean environment: a comparison of undisturbed and disturbed soils' Appl. Geochem. Suppl. Issue 2, pp 195-198. Great Britain, (1993)
12. Adarve, M.J. and Rebollo, L.F. 'Incidencia ambiental de un vertedero sellado de residuos sólidos localizado en Móstoles (Madrid) sobre la composición natural de las aguas subterráneas', Geogaceta, No. 13, pp 3-6 (1993)
13. González, J. Hidrogeología de las rocas plutónicas y metamórficas en la vertiente meridional de la Sierra de Guadarrama, Tesis Doctoral, Universidad Autónoma de Madrid, (1991)
14. Urcelay, A. y Pastor, J. 'La cromatografía iónica y los aniones solubles de suelos sobre sustratos sedimentarios de la meseta sur. Henares Rev. Geol. (en prensa)
15. Freeze, R.A. and Cherry, J.A. Groundwater Prentice Hall. Inc. New Jersey, pp 604 (1979)
16. Cameron, R.D. and Koch, F.A, 'Toxicity of landfill leachates' J. W. P. C. F. 52, pp 760-769 (1987)
17. López Vera, F. Contaminación de las aguas subterráneas. Ed. MOPU. Madrid. pp 78 (1990)
18. Kettern, J.T, 'Biological and Physico-Chemical treatment of landfill leachate' ISWA. Proc. 6th International Solid Wastes Congress. ATEGRUS, Madrid (1992)
19. Gavasci, R, Misiti, A, Rolle, E. and Sirini, P. 'The capacity of natural materials to immobilize toxic compounds contained in landfill leachates' ISWA. Proc. 6th International Solid Wastes Congress. ATEGRUS, Madrid (1992)
20. Christensen, T.H, Lyngkilde, J. and Kjeldsen, P. 'Fate of landfill leachate pollutants in groundwater: A field study at the old Vejen landfill (Denmark)'. ISWA. Proc. 6th International Solid Wastes Congress. ATEGRUS, Madrid (1992)
21. Fernández-Serrano, M.E, Herréz, I, Carreras, N. y Quejido, A. 'Environmental degradation by a municipal solid wastes disposal in Collado-Villalba landfill' ISWA. Proc. 6th International Solid Wastes Congress. ATEGRUS, Madrid (1992)
22. Who, International Standard for Drinking Water. 3rd. Edition, World Health Organisation. Geneva, (1971)