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Spatio-Temporal Behavior of Physico-Chemical Parameters of Fez Controlled Landfill Leachate

Imane El Machrafi[1], Abdennasser Baali[1], Ibrahim Touzani[2], Youssra Ahouach[1], Jamal Naoura[3], Kawtar Fikri-Benbrahim[2*]

[1] Laboratory of Engineering, Electrochemistry, Modelling and Environment, Faculty of Sciences Dhar El Mahraz, Sidi Mohamed Ben Abdellah University, Fez 30050, Morocco

[2] Laboratory of Microbial Biotechnology and Bioactive Molecules, Sciences and Technologies Faculty, Sidi Mohamed Ben Abdellah University, Fez, P. O. Box 2202, Morocco

[3] Laboratory of Natural Resources and Environment, Polydisciplinary Faculty of Taza, University of Sidi Mohammed Ben Abdellah, Taza 35000, Morocco

* Corresponding author's e-mail: kawtar.fikribenbrahim@usmba.ac.ma

ABSTRACT

Aiming to preserve and protect the environment, physical and chemical quality of young and mature leachate samples from Fez controlled landfill were studied and assessed. Therefore, leachate samples were taken and analyzed during the period of January–September 2022. The physico-chemical characterization of both young and mature leachates revealed a significant load of organic matter (biological and chemical oxygen demands: BOD_5 and COD) and minerals (NTK and PO_4^{3-}). In addition, the metallic trace elements (MTEs) concentrations recorded for Cr, Fe, Al and Ni exceeded the threshold levels required in national standards, and their quantitative distribution followed the same pattern at both sites studied: Ni > Fe > Cr > Al > Zn > Pb. The Principal Component Analysis (PCA) enabled to reduce the database to a smaller dimension by gathering as much information on their trends and correlations. Two components were selected which account for the sum of 64.06% of the factors.

Keywords: leachate, physico-chemical parameters, metallic trace elements, controlled landfill, Fez.

INTRODUCTION

Today's environmental problems are wide-ranging and arise across a diverse territory. In this regard, the industrial expansion and population explosion are leading to a considerable rise in waste of all kinds in both industrialized and emerging countries. Moreover, household waste is one of society's major challenges [Zoubi et al., 2010]. The most common form of solid waste management is through controlled and/or uncontrolled open landfill. Landfills and other waste accumulation sites represent a threat to the public's health and the environment [Siddiqi et al., 2022]. Proliferation of rodents and insects, toxic gas emissions as well as pollution of water resources are just some of the health risks that can be caused and the nuisance related to such sites.

The degradation processes of waste in landfill sites are linked to biological and physico-chemical reactions, which subsequently generate soluble substances that are leached and infiltrated later by meteoric waters [El Mrabet et al., 2022]. These transformation mechanisms lead to the production of a liquid effluent known as leachate [Zhang et al., 2019]. Leachate is a significant vector of pollutant load [Cheng et al., 2021] and a major environmental threat due to the complexity as well as variety of its pollutants. Leachate can contain various micro-organisms and substances in diffused or solidified form that are approximately biodegradable, such as MTEs that are extremely toxic even at low concentrations because they are non-biodegradable [Parvin and Tareq, 202; Enitan et al., 2018], as well as chemicals and minerals

[Merzouki et al., 2015]. Releasing such effluents in their untreated state can contaminate soil, surface water and even groundwater, thereby threatening human health [Ahouach et al., 2023; Chiguer et al., 2016; Baun and Christensen, 2004]. Thus, different approaches to leachate treatment were examined, highlighting many economic, sustainable and environmental benefits, namely biological processes: aerobic or anaerobic [Mishra et al., 2023; El-Gohary and Kamel, 2016], chemical and physical processes: adsorption, advanced oxidation processes, chemical precipitation, coagulation/flocculation and membrane technologies [Abdel-Shafy et al., 2024; Mojiri et al., 2021].

In Morocco, socio-economic activities linked to population growth and to consumption patterns changes are generating a large amount of solid household waste, currently reaching almost 18.000 tons per day. As a result, Morocco has established a whole set of laws and programs to protect the environment and to promote sustainable development. Foremost among these is the National Household Waste Program (PNMD), which involves setting up controlled landfill sites as well as investing in ecologically sustainable development.

Nowadays, it is well known that human activities produce many substances that are harmful to humans and the environment, particularly in urban areas where the combination of these activities conducted to a progressive deterioration of the qualities of water, soil and air. For this purpose, the present study concerns the quality assessment of young and mature leachate from Fez's controlled landfill, in terms of physicochemical parameters and metallic elements. The correlations between these elements were studied in order to detect potential links between them.

MATERIALS AND METHODS

Study area

The controlled public landfill situated in the south-east of Fez city, on the Ouled Mhamed to Sidi Harazem Road, covers 120 hectares and receives more than 800 tons per day of waste of all kinds (Figure 1). The landfill receives solid waste, such as household waste from domestic activities, similar household waste from local markets, green waste, tannery waste and debris from demolition activities. ECOMED is the group delegated to manage Fez's controlled landfill, and is responsible for planning, building, financing, equipping and operating the landfill.

Sampling

Sampling campaigns were carried out at two sites (A1: characterized by the young leachate, and A2: characterized by the mature leachate) over nine months, from January 2022 to September 2022, at a rate of 9 samples per site (Figure 2). Leachate samples designed for physico-chemical and heavy metal analysis were taken in one-liter polyethylene bottles. The latter were labelled and then transported to the laboratory in a cooler at a temperature ± 4 °C.

Physico-chemicals parameters measurements

During sampling, in-situ measurements of electrical conductivity (EC), pH and temperature were taken using a Consort C561 multiparameter analyzer. Suspended solids (SS) were determined using the 0.45 µm membrane filtration method. Biological oxygen demand (BOD_5) was determined using a WTW OxiTop IS 6. The chemical oxygen demand (COD) was determined by oxidation in an

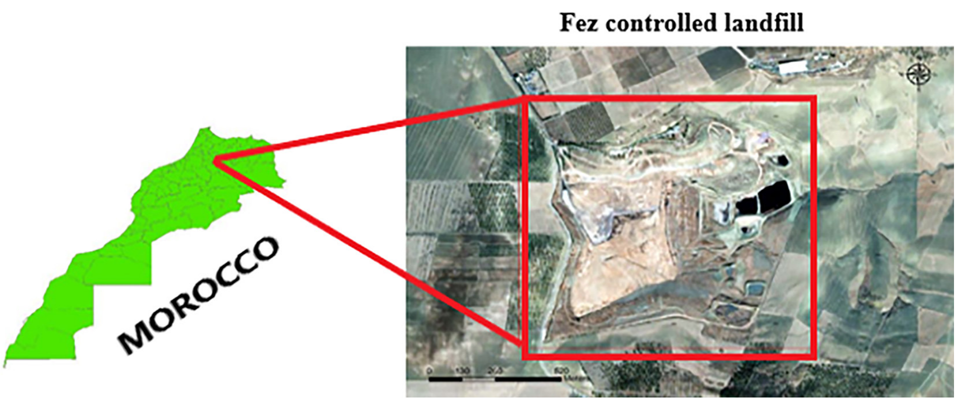

Figure 1. Geographical location of Fez controlled landfill

Figure 2. A1 – young leachate basin; A2 – mature leachate basin from Fez controlled landfill

acid solution by excess potassium dichromate at a temperature of 148 °C of the oxidizable matter under the test conditions, in the presence of silver sulfate as a catalyst. Total Kjeldahl nitrogen (TKN) was determined by mineralizing the sample with concentrated sulfuric acid at high temperature in the presence of a catalyst (selenium). Ortho-phosphate ions (PO_4^{3-}) were determined by ammonium molybdate spectrometry. Nitrates (NO^{3-}) and nitrites (NO^{2-}) were measured by molecular spectroscopy. To quantify heavy metals, the samples were digested into concentrated nitric acid (HNO_3). Afterwards, they were filtrated using 0.45 µm pore size filters. The concentration of Metallic Trace Elements (Al, Cr, Fe, Ni, Pb and Zn) was determined by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES), type J.P. SELECTA. S.A, at Sidi Mohamed Ben Abdellah University's (USMBA) Innovation Centre in Fez. The parameters analysis was completed in compliance with the standards recommended by Rodier et al. [2009].

RESULTS AND DISCUSSION

Table 1 presents the average values obtained for the physico-chemical parameters during characterization and monitoring for the two studied

Table 1. Average values of physico-chemical parameters of young (A1) and mature (A2) leachates from the Fez controlled landfill

Parameters	Unit	A1	A2	MLDS*
Temperature	°C	26.05	19.02	30
pH	–	6.65	8.28	5.5-9.5
EC	mS/cm	25.95	44.04	2.7
SS	mg/l	1336.75	2374.29	100
COD	mg/l	12577.31	9879.90	500
BOD_5	mg/l	5673.15	1067.30	100
NTK	mg/l	1245.51	1017.56	40
NO^{3-}	mg/l	9.73	12.21	NA
NO^{2-}	mg/l	2.28	3.32	NA
PO_4^{3-}	mg/l	2.83	4.52	15
Al	mg/l	3.51	2.15	10
Cr	mg/l	5.80	8.97	2
Fe	mg/l	30.50	17.08	5
Ni	mg/l	34.77	36.54	5
Pb	mg/l	0.33	0.36	1
Zn	mg/l	1.61	1.45	5

Note: NA: not available; MDLS*: Moroccan Liquid Discharges Standards (MLDS, 2018).

sites, A1 and A2, containing respectively young and mature leachates. Figure 3 represents the spatio-temporal evolution of the studied parameters (T°, pH and EC) at both sites.

Temperature

The temperatures of young leachate A1 and mature leachate A2 fluctuate around 24 °C and 16 °C, respectively, during the cold season. However, over the summer, temperatures rise remarkably, reaching 29 °C and 21 °C for young leachate A1 and mature leachate A2, respectively. It should be highlighted that the temperature of the young leachate is always higher than that of the mature leachate due to chemical reactions and micro-organism development. However, these values meet the direct and indirect discharge limit standards. Furthermore, these values remain lower than those found for the leachates from the Akouédo landfill in Abidjan, Ivory Coast [Adjiri et al., 2008] and higher than the value recorded for the leachate from the Tamelast landfill, Greater Agadir (Morocco) [Asouam et al., 2021].

pH

During the conducted sampling campaigns, the mean pH values were 6.65 and 8.28 at sites A1 and A2 respectively. These values show that the pH of young leachate is slightly acidic, whereas the pH of mature one is alkaline. The high pH values recorded in the mature leachate may be explained by the low concentrations of volatile organic compounds due to the anaerobic degradation of the waste. In addition, the pH of young leachates tends to increase gradually over time as a result of the decreasing rate of partially ionized free volatile fatty acids. However, these values are within national standards. Moreover, the recorded pH values are similar to those found at the Meknes landfill leachate (Morocco) [Mejraoua and Zine, 2017].

Electrical conductivity (EC)

The average of recorded electrical conductivities at sites A1 and A2 were 25.95 ms/cm and 44.04 ms/cm, respectively. Such very high electrical conductivity values indicate that the leachate from the sites analyzed is marked by high mineralization activity and a high salt load. It is thought that this high level of mineralization is mainly due to the solid domestic and industrial waste received by the landfill, which has a high salt and chloride load. However, these results remain similar to certain studies [Abd El-Salam and Abu-Zuid, 2015; Abdelwaheb et al., 2012].

Suspended solids (SS)

The mean values recorded for SS are about 1336.75 mg/l in the young leachate and 2374.29 mg/l in the mature leachate. The high concentrations of suspended solids in the mature leachate might be due to the advanced state of decomposition of the

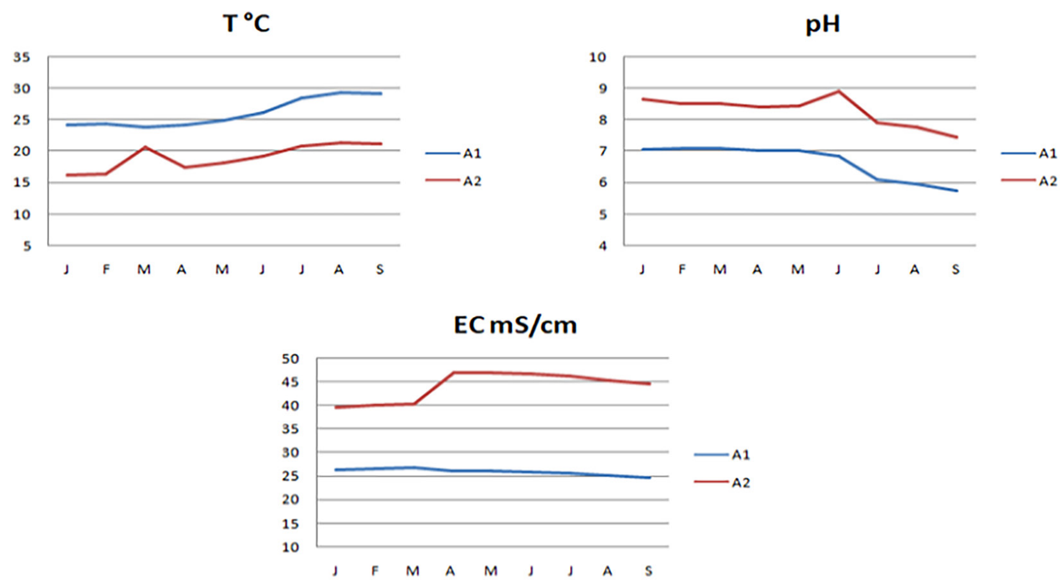

Figure 3. Comparison of the temporal evolution of parameters (T°, pH and EC) of young (A1) and mature (A2) leachates from Fez controlled landfill

waste to produce very contaminated and polluted leachates. The values are significantly higher than national standards for direct or indirect discharges. However, they remain lower than those found in the leachates from the Mohammedia-Benslimane landfill [Hamri et al., 2021] and higher than those found in the leachates from the controlled landfill in Essaouira [Chiguer et al., 2016] and Rabat [El Atmani et al., 2021].

COD and BOD_5

The average COD values for sites A1 and A2 are 12577.31 mg/l and 9879.90 mg/l, respectively. In turn, those registered for BOD_5 were 5673.15 mg/l at A1 and 1067.30 mg/l at A2. These results greatly exceed Moroccan standards, indicating that the organic content of young leachates is significantly higher than that of mature leachates, which means that young leachates may remain completely non-degradable and therefore have a high organic load, probably due to strong degradation of organic acid [Hussein et al. 2019; Erses et al, 2008]. Instead, leachates in both sampling sites showed the characteristics of the methanogenic phase, caused by pH values increasing from an average of 6.65 to 8.27.

Evaluation of the BOD_5/COD biodegradability index

The mean values of the BOD_5/COD ratio were around 0.45 for the site A1 and 0.1 for the site A2. These values reveal that the leachate of site A2 has exceeded the mature phase as the site has been operating since 2004, which means that it can be classified as an old and aged landfill and may contain a considerable quantity of biologically inert materials. In addition, the low values of the BOD_5/COD ratio in the leachate indicates its richness in non-biodegradable organic compounds, for which physicochemical treatment may be more appropriate than biological processes.

Total Kjeldahl nitrogen and phosphate (PO_4^{3-})

Mean values for total Kjeldahl nitrogen measured at sites A1 and A2 were 1245.51 mg/l and 1017.56 mg/lm respectively. Phosphate contents were 2.83 mg/l at A1 and 4.52 mg/l at A2. Those results show that total nitrogen (NTK) is very abundant in both leachate sites. This could be attributed to the compaction of waste in the landfill. Typically, in mature leachate the ammonia NH_3/total Kjeldahl nitrogen ratio is usually above 70% [Elmaghnougi et al., 2018]. Results are concordant with those of some works that record high NTK values in other Moroccan landfills [Benaddi et al., 2022; Merzouki et al., 2015]. The low levels of orthophosphates recorded are generally due to their consumption by microorganisms.

Nitrates (NO^{3-}) and nitrites (NO^{2-})

Nitrates constitute the final phase of nitrogen oxidation and the most oxidized form of nitrogen in the leachate. Average nitrate concentrations were 9.73 mg/l for site A1 and 12.21 mg/l for site A2. These values are well below the maximum level permitted by the WHO, corresponding to 50 mg/L. Nitrites are produced either by nitrate reduction or ammonium incomplete oxidation [Rodier et al., 2009]. The average nitrite concentrations measured are very low (2.28 mg/l in A1 and 3.32 mg/l in A2). Consequently, these two components do not pose any pollution risk to the leachates.

Metallic trace elements

Comparing the concentrations of metallic trace elements (MTEs) measured in the samples analyzed reveals a relatively high metal load in the young leachate compared with the mature leachate. Similarly, the distribution of metals follows the same order for both sites: Ni > Fe > Cr > Al > Zn > Pb. Ni is the most abundant metal, with respective values of 34.77 and 36.54 mg/l in A1 and A2. Fe and Cr also have high average concentrations, with values of 30.50 mg/l and 5.80 mg/l at A1 and 17.08 mg/l and 8.97 mg/l at A2, respectively. Nevertheless, the values recorded for Ni, Fe and Cr exceed the national guidelines for surface water and water designed for irrigation. The results remained similar to works that have reported high MTEs levels in leachates [Chiguer et al., 2016] and higher than others [Razak et al., 2020; Talbi et al. 2020; Tahiri et al., 2017]. The mean concentrations of Al and Zn are 3.51 and 1.61 mg/l in A1; and 2.15 and 1.45 mg/l in site A2, respectively. As for Pb, the concentrations measured in the leachates are extremely low (0.33 mg/l in A1 and 0.36 mg/l in A2). The values noted for Al, Zn and Pb remain below the values permitted by Moroccan norms. Although the concentrations of these metals do not exceed the authorized limits, they can constitute a health threat and have a significant impact on groundwater [Mor et al., 2018]. The presence of MTEs in leachate is typically generated by the collection of a wide range of wastes, such as industrial waste, medical waste, batteries, lead-containing paints, plastics, paper, cardboard and cigarette butts. Indeed, [Torkashvand et al. 2021] reported that increased MTEs concentrations in leachate are proportionally related to the increase of cigarette butts in waste.

Principal component analysis (PCA)

PCA is a statistical method that summarizes quantitative data in an individual/variable table by creating a small number of new variables, which are the main components [Liu et al., 2022; Touzani et al., 2020]. A data matrix containing 16 variables (T, pH, EC, SS, COD, BOD_5, NTK, PO_4^{3-}, NO^{3-}, NO^{2-}, Al, Cr, Fe, Ni, Pb, and Zn) from sites A1 and A2 was used for PCA.

Table 2 shows the correlation matrix for the 16 parameters measured, where significant Pearson correlation coefficients greater than 0.5 are interpreted. In this study, the determinant of the correlation matrix is relatively very low and not equal to zero ($1.774\ 10^{-13}$), which indicates that one or more variables have a different correlation with one or more other variables. The correlation matrix shows the existence of high correlation degrees between one or more variables. Several correlations are recorded between temperature and SS, BOD_5, NTK, NO^{3-}, Fe, Ni, and Pb, as well as between pH and conductivity, on the one hand, and SS, BOD_5 and MTEs (Fe, Ni and Pb) on the other hand. These results also highlight another set of variables that are negatively correlated with each other, namely T-pH, T-EC, pH-BOD_5, pH-nitrogenous elements and metals. Similarly, this analysis highlighted a group of variables made up of descriptors that are positively correlated with each other, including orthophosphates, nitrogenous elements and organic pollution load indicators. In addition, significant positive correlations exist between Cr-Al, Fe-Ni, Fe-Pb and Ni-Pb. The positive correlations between the different variables studied in this study are generally comparable with other studies [Saha et al., 2023; Rhouat et al., 2019]

In this study, Bartlett's sphericity test showed a Chi-square value of 359.342, with a degree of freedom of 120 and a significance level of 0.000, confirming that the variables are significantly correlated. In addition, the KMO test had a validity of 0.587 (Table 3), which is a good indicator since it is relatively higher than 0.5 [Chen et al., 2022], confirming the satisfactory adequacy of the data set useful for the PCA.

Both components selected hold an eigenvalue above or equal to 1 and together hold 64.06% of the total information (Table 4). The first axis reports 48.35% of the overall information, through the following ten variables: temperature, SS,

Table 2. Pearson correlation matrix for the studied variables and MTEs

Parameter	T	pH	EC	SS	COD	BOD_5	NTK	PO_4^{3-}	NO^{3-}	NO^{2-}	Al	Cr	Fe	Ni	Pb	Zn
T	1															
pH	-.730	1														
EC	-.371	.718	1													
SS	.900	-.761	-.544	1												
COD	.289	.058	.075	.438	1											
BOD_5	.941	-.812	-.484	.935	.324	1										
NTK	.696	-.422	.050	.435	.051	.592	1									
PO_4^{3-}	.592	-.517	-.045	.295	-.146	.477	.845	1								
NO^{3-}	.604	-.509	-.305	.377	-.208	.536	.838	.756	1							
NO^{2-}	.660	-.519	-.056	.444	-.016	.600	.659	.756	.506	1						
Al	-.038	.548	.462	-.096	.517	-.186	-.147	-.197	-.302	-.025	1					
Cr	-.423	.743	.383	-.321	.392	-.443	-.509	-.661	-.516	-.423	.700	1				
Fe	.732	-.566	-.442	.596	.087	.664	.703	.513	.803	.323	-.217	-.361	1			
Ni	.691	-.420	-.264	.528	.032	.635	.635	.358	.642	.219	-.182	-.367	.823	1		
Pb	.679	-.300	-.129	.507	.261	.627	.637	.341	.588	.221	.003	-.204	.741	.885	1	
Zn	.341	-.103	.117	.391	.224	.254	.149	-.068	-.121	-.005	-.028	-.077	.133	.248	.177	1
a. Determinant = $1,774\ 10^{-13}$																

Table 3. KMO index and Bartlett test

Precision measurement of Kaiser-Meyer-Olkin sampling		0.587
Bartlett's sphericity test	Approximate Chi-square	359.342
	Ddl	120
	Significance of Bartlett	0.000

Table 4. Total variance explained

Component	Initial eigenvalues			Extraction Sum of squares of selected factors		
	Total	Variance percentage	Accumulated percent	Total	Variance percentage	Accumulated percent
1	7.737	48.358	48.358	7.737	48.358	48.358
2	2.513	15.709	64.067	2.513	15.709	64.067

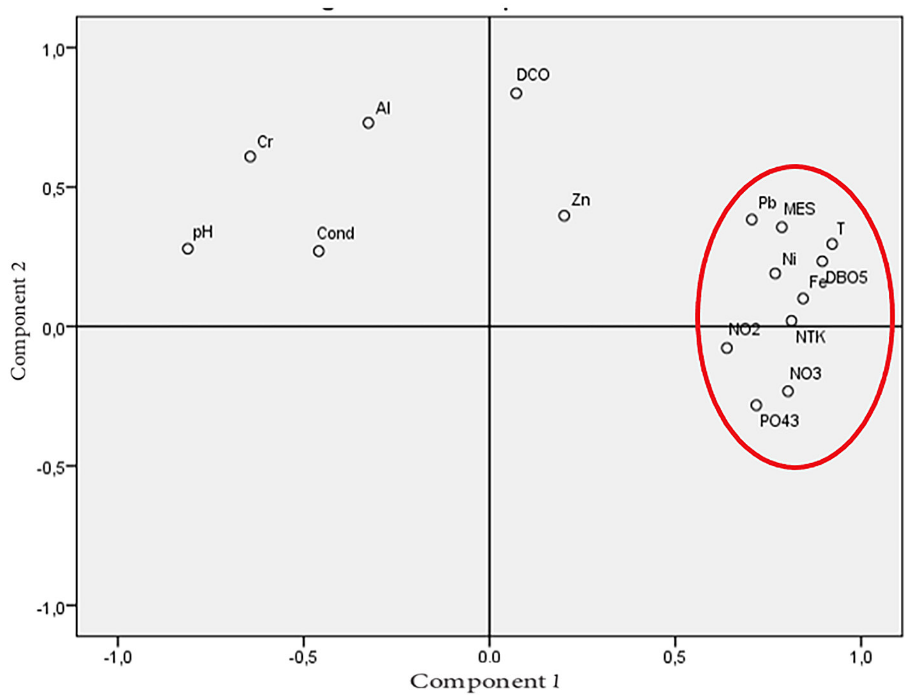

Figure 4. Principal component analysis results: Variables projection on the two factorials plane

BOD_5, NTK, PO_4^{3-}, NO^{3-}, NO^{2-}, Fe, Ni and Pb, which include most of the chemical parameters indicative of organic pollution and Metallic Trace Elements. The second component, representing 15.70%, is defined positively by the following variables: pH, COD, Al, Cr and Zn. The first two factorial axis of the PCA for the variables studied are depicted in Figure 4.

CONCLUSIONS

On the basis of the analysis of young and mature leachate from the Fez controlled landfill during this study, most of the chemical parameters show high pollution levels, surpassing the national standards, particularly indicator parameters of organic load (SS, COD and BOD_5) and total nitrogen.

Furthermore, this study confirmed the attendance of MTEs in the leachates analyzed, which appears relatively linked to the human pressure applied on the study area, generating a significant impact on groundwater contamination and subsequently constitutes a major environmental problem. Therefore, a physical treatment appropriate to the existing environmental and socio-economic conditions should be anticipated, for the purpose of improving their quality to meet the actual standards and saving Fez city's resources. The statistical analysis of the data revealed strong positive correlations between the different variables. Moreover, the PCA enabled to decrease the database size by gathering all possible information on their trends and to distinguish the characteristics of leachate quality at the sites studied. For this purpose, the model initially developed from 16 variables has been simplified to a relatively simple model based on two factorial axes.

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