The 3rd International Conference on Water Resources and Arid Environments (2008)
and the 1st Arab Water Forum

Advanced Anaerobic Membrane Bioreactor Technology for Wastewater Treatment and Effluent Reclamation in Tunisia

Sami Sayadi
Bioprocesses Laboratory, Centre of Biotechnology at Sfax,
Sfax, Tunisia

Abstract

Anaerobic treatment is a mature technology at full scale for the treatment of municipal wastewater in warm climates where the wastewater temperature is 20°C or higher. Anaerobic treatment can provide savings in operation costs (no aeration and methane production) and a dramatically reduced production of biosolids. Coupled to membrane ultrafiltration, anaerobic membrane bioreactor (AMBR) offers the possibility of operating the system at high mixed liquor suspended solids concentration. The treatment of raw domestic wastewater originated from Ksour Essef (centre of Tunisia) and Sfax (south of Tunisia) was operated using an AMBR pilot plant. In both cases, the treatment led to a total removal of all tested pathogens. The quality of treated wastewater fits largely with WHO guidelines for unrestricted irrigation. However, the treatment was more technically feasible in the case of Ksour Essaf wastewater than for Sfax wastewater due to the frequent contamination of Sfax wastewater by industrial discharges. On the other hand, the AMBR process showed its robustness for converting high strength wastewater such as landfill leachate from Tunis (Jebel Chakir) solid waste discharge in biogas and water for reuse.

Key words: Domestic wastewater; landfill leachates, Anaerobic MBR; biogas, reuse

Introduction

The Mediterranean basin (and particularly North African countries) is one of the poorest region in the world in terms of water resources. An increased water consumption rate for irrigation purposes along with a high urban population growth, have had an adverse effect on water resources. Thus, most groundwater resources in the Mediterranean areas are at risk of being exhausted through overexploitation. In Tunisia, treated municipal wastewater is becoming one of the main alternative sources of water. Indeed, in 2007, 99 municipal wastewater treatment plants (WWTP) has treated a quantity of 215 millions of m^3 from which more than 30% are reused. The treated volume in

2011 is expected to be 266 millions m^3, whereas the reused wastewaters should reach more than 50%. However, especially in the eastern and northern Mediterranean regions, wastewaters are inefficiently treated and re-used for irrigation or sanitary purposes, serving as carrier of pathogens such as faecal coliforms, Salmonella, Helminths and Viruses or causing water pollution when discharged to water bodies. In general, municipal and industrial wastewaters are treated biologically, i.e. by activated sludge process or anaerobic process, using micro-organisms for degradation of organic pollutants.

Anaerobic digestion was described as a successful treatment technology for high strength industrial effluents [1]. Besides, over the last decade, the potential of the anaerobic processes as a treatment technology for low strength domestic wastewater has been evaluated. Nevertheless, domestic wastewater is quite complex due to the presence of fatty compounds, proteins, detergents, heavy metals and other toxic compounds. These characteristics impose limitations to the anaerobic process in respect to COD removal efficiency and also in terms of maximum organic and hydraulic loading rates to be applied. These limitations together with the slow net growth rate of anaerobic bacteria, increasingly stringent legislation of treated wastewaters and the opportunity of water reuse/recycle, increased the interest in membrane technology which is presented now as the potential technology for municipal wastewater treatment. Membrane–coupled anaerobic bioreactors have been applied as one alternative to the conventional anaerobic digestion process because they retain all micro-organisms in the reactor [2]. The membrane bioreactor (MBR) is an effective treatment technology for wastewater treatment and recycling. It has several advantages over conventional treatments such as reliability, compactness and optimal treated water quality [3]. In our Laboratory, we worked on anaerobic membrane bioreactor for the treatment and reuse of low and high strength wastewaters such as municipal, landfill leachates and industrial wastewaters. Our focus was on the development of AMBR technology for wastewater treatment with emphasis on the microbiological and toxicity characterizations of the treated water.

Objectives

The objective of this project is to study the performance of an AMBR for the treatment of low strength wastewater like municipal wastewater. The focus was to transform the municipal wastewater into different valuable streams such as biogas (energy) and water for irrigation containing large amounts of fertilizers. This study includes firstly a comparison of the treatment of raw urban wastewater originated from an industrial city: Sfax with that of Ksour Essef city, producing mostly domestic wastewater. Secondly, this research focuses on the conversion potential of landfill leachates in biogas using anaerobic membrane bioreactor technology.

Methodology

Wastewater sampling

Raw domestic wastewater was sampled from Sfax and Ksour-Essef wastewater treatment plants (WTPs). Sfax is an industrial region 270 km to the south of Tunis, Tunisia. However, Ksour Essef is a non industrial region 120 km to the north of Sfax, Tunisia. Wastewaters were collected and stored at 4°C until use. The physico-chemical characteristics of three samples of Sfax wastewater (SW) and one sample of Ksour Essef wastewater (KW) are shown in Table 1.
Landfill leachate (LFL) was collected at summer time from the controlled discharge of Jebel Chakir. The characteristics and average composition of Jebel Chakir- LFL were given in table 1. The feed solution was diluted to reach a reasonable value of COD. The HRT was kept constant (HRT = 7 d) during all the treatment and the OLR was increased by the decrease of the dilution of the feed solution.

Experimental apparatus

The experimental set-up was constructed within the frame of the Inco-med project “MBR recycling” and it was installed in Centre de Biotechnologie de Sfax, Tunisia. The schematic diagram of the experimental set-up is shown in Figure 1. The jet flow anaerobic bioreactor (3) was constructed of Plexiglas and having a working volume of 50 litres. The temperature was maintained constant at 37°C by circulating water through the water jacket of the reactor. Bioreactor is fed via peristaltic pump (2) from the wastewater storage tank (1). The influent was supplied through the nozzle (14) into the jet flow module. Nozzle is co-axially located at the top of an inner tube (15), this created a dawn flow in the inner tube and an up flow between the inner tube and the reactor wall. This circulation of the liquid allows a perfect homogenization of the medium. The reactor was coupled via a multistage centrifugal pump Lowara SV805 (2-3 kW, Qmax = 10-12 m^3 at 5-6 bars, and frequency controlled by a Stöber FBS/FDS) to a TECHNOCON GmbH ultrafiltration system composed by a membrane module Stork (Friesland BV) 10-Feet long. The membrane, which was Stork WFFX 0281, had 1 m^2 area, and 100 kDa cut-off. The cross-flow velocity was fixed at a value of 3 m s^{-1} and the trans-membrane pressure was varied from 1 to 2 bars. An inductive volumetric flow meter IFC090 was used for measuring the membrane inflow rate and the flow rate in the nozzle (5). A gas meter (Ritter) was used for measuring the biogas production (12). The pH was automatically adjusted at 7 by a pH regulation pump and a pH electrode (Dulcometer, Fa Prominent) using a solution of NaOH.

Analytical methods

COD was determined according to [4] standard method. BOD_5 was determined by the manometric method with a respirometer (BSB-Controller Model 620 T (WTW)). Total phosphorous was determined by [5] method. Total nitrogen was determined by [6] method. Total suspended solids (TSS) and volatile suspended solids (VSS) were determined according to the standard methods [7]. Heavy metals concentrations were measured using an atomic

absorption heavy metals analyzer (Perkin Elmer 1101B) with a hollow cathode lamp. 80% air/20% (v/v) acetylene was used as oxidizing fuel flame.

To get the gas composition, gas samples were taken with a syringe from the tank of biogas and analysed by a gas chromatograph (Model: IGC11 of DELSI.) equipped with a thermal conductivity detector. Volatile fatty acids (VFA) were analysed by a gas chromatograph (SHIMADZU GC-9A) equipped with a flame ionisation detector (SHIMADZU CR 6A). The turbidity was determined using a turbidimeter (WTW, turb 551 IR). The conductivity and the pH were determined using a conductivimeter model CONSORT C 831 and a pH meter model Metrohm 744, respectively.

GC-MS analysis

GC-MS was carried out to identify hydrocarbons and phenols present in LFL. An aliquot of 1 ml of ethylacetate-extractable products of the sample was injected splitless into the GC/MS (5975B inert MSD Agilent). The data were obtained on a DB-5MS column, 30 m length, 0.25 mm i.d. and 0.25 mm thickness (Agilent Technologies, J&W Scientific Products, U.S.A.). Carrier gas was helium. GC oven temperature started at 100°C and holding for 1 min to 260°C and holding for 10 min with program rate 4°C min^{-1}. The injector and detector temperatures were set at 250°C and 230°C, respectively. The mass range was scanned from 50 to 550 amu. The control of the GC/MS system and the data peak processing were carried out by means of the MSDCHEM Software.

Microbial estimation

Total Coliforms (TC), Faecal Coliforms (FC); Faecal *Streptococci (FS)* were estimated according to [8] and [9] water standard methods. MPN determination of *Salmonella* (S) was carried out by modified [10] method. Helminths (H) ova were extracted from wastewater by sedimentation-floatation according to [11] method adapted to wastewaters. Protozoan (P) cysts numeration was determined by the same protocol of helminths ova.

Phytotoxicity

Phytotoxicity was estimated by the determination of the GI according to [12] method using *Lepidium sativum* seeds.

Microtoxicity

The microtoxicity was determined according to [13] using the luminescent bacterium *Vibrio fischeri* LCK 480.

Results

Treatment of domestic low strength wastewater by AMBR: effect of industrial discharges

The treatment of Ksour Essef domestic wastewater by the AMBR was successful. Indeed, the quality of the permeate effluent was acceptable to be reused for irrigation. For the Tunisian wastewater standards for reuse in the

agricultural sector, the COD, BOD_5 and SS concentrations are 90, 30 and 30 mg/l, respectively. Indeed, the membrane bioreactor yielded an average COD removal rate of more than 76 % and an average BOD_5 removal rate higher than 84 % at a volumetric loading rate varying from 0.23 to 2 g COD $l^{-1}d^{-1}$. Permeate quality indicated that suspended solids were completely removed. The conductivity and the turbidity were monitored in the permeate and in the raw domestic wastewater effluent. The values of conductivity were of the same order in the raw wastewater as well as in the permeate (ranged from 2.3 to 3.6 mS cm^{-1}). Also, the turbidity of raw effluent ranged from 95 to 148 NTU. However, the permeate turbidity was less than 3 NTU, with a removal percentage of more than 98.4 %. Figure (2) shows that the rate of the biogas produced in the reactor increased with increasing volumetric loading rate (VLR). The methane yield expressed as the volume of methane produced per g of COD in the effluent ranged from 0.05 to 0.31 l CH_4 g^{-1} COD, which was close to the maximum value. The volatile fatty acids (VFA) concentration was monitored in the bioreactor and in the permeate and results showed that VFA production was insignificant in the reactor (data not shown). It was below the inhibitory limits permitting the stability of the methanogenic process. In the permeate, the VFA concentration was less than 0.25 g l^{-1}.

However, the use of AMBR for the treatment of Sfax (industrial city) wastewater resulted in low process efficiency (data not shown). The anaerobic process exhibited low adaptation of the consortium with drastic decrease in biogas productivity. This could be due to the considerable fluctuations in the wastewater composition and the possible contamination by industrial discharges. For this reason, 3 samples S1, S2 and S3 of wastewaters (Table 1) were collected and analyzed. The chemical composition of these samples demonstrated the low biodegradability of S1 and S2 (high COD/BOD_5 ratio) which confirmed a possible toxic contamination of these samples.

Phytotoxicity test using *Lepidium sativum* seeds was carried out for monitoring the toxicity of untreated and treated Sfax wastewater (SW) and Ksour Essef wastewaters. *Lepidium sativum* germination index (GI) is described as the most sensitive test used to evaluate the toxicity of wastewaters [14]. The GI determination of both untreated and treated SW revealed a strong phytotoxic character. Indeed, the germination index was less than 15% for untreated SW and did not exceed 50% for treated SW. However, phytotoxicity of treated KW was significantly reduced (Fig. 3). For example, GI of KW reached 80%. Several SW samples were analysed for their inhibitory potential of the well-known strain *Vibrio fischeri* (data not shown). Microtoxicity analysis of untreated SW showed that the mean value of microtoxicity was 51% noting that most of samples presented microtoxicity values more than 80%. Even after treatment, samples were still toxic. By contrast to SW, untreated KW showed a low toxicity. For the treated KW, since the inhibition percentage is lower than 20%, they are presumed no toxic (data not shown).

Thus, taking into account toxicity results, we can assume that treated KW are more appealing from a point of view of agricultural reuse.

The microbiological quality of MBR permeates fits with WHO guidelines for unrestricted irrigation. Indeed, TC, FC, FS, S, H ova and P cysts were removed to levels below the detection limit for both SW and KW permeates

(Table 2). Filtration parameters were optimized in this study and during the operation time, the cross flow velocity was fixed to a relatively high value of 3 m s^{-1} in order to avoid fouling and the transmembrane pressure was varied from 1 to 2 bar. The permeate flux was maintained at 9 $l/h.m^2$.

Treatment of high strength landfill leachate wastewater by AMBR

Landfill leachates (LFL) collected from Djebel Chakir (Tunisia) discharge area were found to be highly loaded with organic matter, ammonia, salts, heavy metals, phenols and hydrocarbons (Table 1). Despite the possibility of their biodegradability, they represent a threat to the environment and show some resistance to conventional wastewater treatment processes [15]. For these reasons, this study attempted to develop the AMBR for the treatment of LFL. LFL was treated without any physical or chemical pretreatment. However, attention was paid to optimize its state of acidification/stabilization. The organic loading rate (OLR) in the AMBR was gradually increased from 1 g COD $l^{-1}d^{-1}$ to an average of 6.27 g COD $l^{-1}d^{-1}$. At the highest OLR, the biogas production was more than 3 volumes of biogas per volume of the bioreactor. The mixed liquor volatile suspended solids reached a value of approximately 4 g/l in the bioreactor. At stable conditions, the treatment efficiency was high with an average COD reduction of 90% (Table 3, Figure 4) and biogas yield of 0.46 l biogas per g COD removed (Table 3). The hydrodynamic operation conditions of the ultrafiltration membrane were adjusted to have a permeate flux of 2.5 $l\ h^{-1}m^{-2}$.

Fig. 5a shows that LFL was highly loaded with hydrocarbons and phenols. These substances were reported to be toxic for biological growth. The challenge of this treatment was to investigate if acclimatized anaerobic bacteria under process intensification such as MBR are able to degrade such compounds. The GC-MS analysis (Fig. 5b) showed a complete removal of hydrocarbons and phenols having the retention time below 9 min, which correspond to compounds with molecular mass under 224. Up to this value, compounds seem to be toxic to anaerobic bacteria. The same substances are found in the permeate but at lower concentrations.

Conclusions

The use of MBR for the treatment of Sfax wastewater showed low process efficiency. This is believed to be due to the considerable fluctuations in the domestic wastewater composition and the possible presence of toxic compounds which inhibited both *Lepidium sativum* germination and *Vibrio fischeri* luminescence. However, the MBR proved to be efficient for the treatment and conversion into biogas (energy) of low strength Ksour Essef wastewater. Treated wastewaters were of good quality and fit with WHO guidelines for agricultural reuse. More over, AMBR technology showed its robustness during the bioconversion into methane of the problematic organic matter found in landfill leachates and exhibited 90% COD removal.

References

1. Arsov, R., Ribarova, I., Nikolov, N., Mihailov, G., Topalova, Y., and Khoudary, E., Two-phase anaerobic technology for domestic wastewater treatment at ambient temperature, *Wat. Sci. Tech.* 39, 115-122 (1999).

2. Brindle, K., and Stephenson, T., The application of membrane biological reactors for the treatment of wastewaters, *Biotechnol. Bioeng.* 49, 601-610 (1996).

3. Stephenson, T., Judd, S., Jefferson, B., and Brindle, K., *Membrane Bioreactors for wastewater treatment*, London, UK: IWA publishing (2000).

4. Knechtel, R.J., A more economical method for the determination of chemical oxygen demand, *J. Water Pollut. Control Fed.* May/June, 25–29 (1978).

5. Dabin, B., Application des dosages automatiques à l'analyse des sols, *Cah. Orstom., sér. Pédol.* III, 335-366 (1965).

6. Kjeldahl, J., A new method for the determination of nitrogen in organic matter, *Z. Anal. Chem.* 22, 366 (1883).

7. APHA, *Standard methods for Water and Wastewater Examination*, 18th ed. Amer. Public Health Assoc., Washington, D.C. (1992).

8. ISO 4832, Microbiology -- General guidance for the enumeration of coliforms -- Colony count technique (1991).

9. AFNOR, NF T90–411, Recherche et dénombrement des streptocoques du groupe D, *Méthode générale par ensemencement en milieu liquide (NPP)* (1989).

10. Yanko, W.A., Walker, A.S., Jackson, J.L., Libao, L.L., and Gracia, A.L., Enumerating Salmonella in biosolids for compliance with pathogens regulations, *Wat. Environ. Res.,* 67, 364-370 (1995).

11. EPA, *Environmental Regulations and Technology Control of Pathogens and Vectors in Sewage Sludge*, EPA-625/R-92/013, U.S. Environmental Protection Agency, Cincinnati, Ohio (1992).

12. Zucconi, F., Forte, M., Monaco, A., and de Beriotodi, M., Biological evaluation of compost maturity, *Biocycle*, 22, 27-29 (1981).

13. ISO 11348-2.Water quality-Determination of the inhibitory effect of water samples on the light emission of *Vibrio fischeri* (Luminescent bacteria test). Part 2. *Method using liquid-dried bacteria* (1998).

14. Wong, J.W.C., Mak, K.F., Chan, N.W., Lam, A., Fang, M., Zhou, L.X., Wu, Q.T., and Liao, X.D., Co-composting of soybean residues and leaves in Hong Kong, *Bioresour. Technol.,* 76, 99-106 (2001).

15. Ellouze M., Aloui F. and Sayadi S, Detoxification of Tunisian landfill leachates by selected fungi. 2008. *Journal of Hazardous Materials*, 150, 642–648.

Table 1: Physico-chemical composition of raw domestic wastewater originated from Sfax (S1, S2, S3), Ksour Essef (K1) and landfill leachate from Jebel Chakir (LFL).

	S1	S2	S3	K1	LFL
pH	7.62	7.7	7.8	7.23	7.31
EC (mS cm^{-1})	6.57	6.94	5.3	2.96	46
TSS (g l^{-1})	0.288	0.54	0.22	0.377	1.97
VSS (g l^{-1})	0.18	0.13	0.2	0.286	1.46
COD (g l^{-1})	0.670	0.90	0.419	0.786	85
BOD_5 (g l^{-1})	0.180	0.280	0.160	0.315	49
COD/BOD_5	3.72	3.21	2.61	2.49	1.73
N_t (mg l^{-1})	49.35	57	51.47	166	3177
P_t (mg l^{-1})	10.4	16	52.5	11.79	1600
Cu (mg l^{-1})	0.001	0.0016	0.25	0.02	553
Pb (mg l^{-1})	0.02	0.053	0.03	<0.041	1.58
Cr (mg l^{-1})	0.017	0.033	0.015	<0.015	0.75
Cd (mg l^{-1})	0.004	0.0025	0.0033	<0.004	<0.2

Table 2: Microbiological characteristics of untreated and treated SW and KW used to feed the AMBR, permeate and the microbial removal efficiency of the system.

	Untreated SW	Untreated KW	Permeate	RR (%)
T C (CFU 100 ml^{-1})	84 10^5	26 10^3	ND (in 1 ml)	100
F C (CFU 100 ml^{-1})	42 10^5	12 10^3	ND (in 1 ml)	100
FS (CFU 100 ml^{-1})	4.5 10^5	21 10^5	ND (in 1 ml)	100
S (MPN l^{-1})	940	940	ND (in 100 ml) 100	
H (Ova l^{-1})	9.5	11	ND (in 1 l)	100
P (10^2 Cysts l^{-1})	1680	1621	ND (in 1 l)	100

Values have been calculated as arithmetic means of samples appropriated on a period of 2.5 months; ND: not detected; RR: removal rate;

Table 3: Summary of the AMBR performance at stable conditions

Operation period	Hydraulic retention time HRT (d)	Organic loading rate OLR (g COD $l^{-1}d^{-1}$)	COD_{feed} (g l^{-1})	$COD_{permeate}$(g l^{-1})	COD removal (%)	Biogas yield
OLR 1	7	2.24	14.87	1.17	92.0	0.45
OLR 2	7	4.66	30.8	2.96	88.8	0.37
OLR 3	7	6.27	41	3.77	90.7	0.48

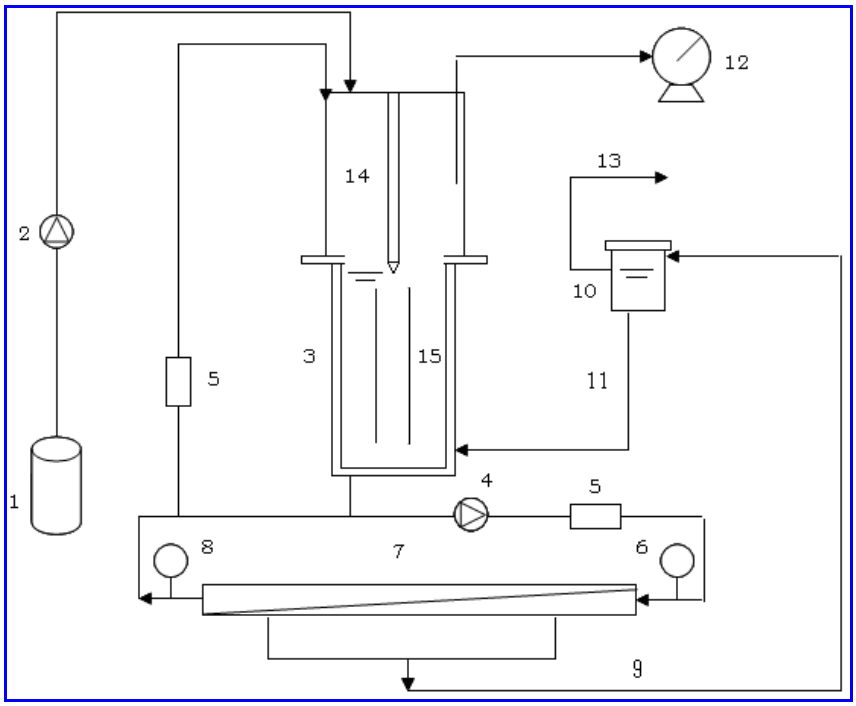

Figure 1: Schematic diagram of experimental process installed in Sfax, Tunisia: (1: raw domestic wastewater reservoir, 2: Peristaltic pump, 3: Jet Flow Anaerobic Reactor, 4: Circulation pump, 5: Flow meter, 6: manometer 1, 7: Ultrafiltration membrane, 8: manometer 2, 9: permeate, 10: Permeate tank, 11: Permeate recycling, 12: gas flow meter, 13: permeate discharged in the sewage system, 14: nozzle, 15: tube)

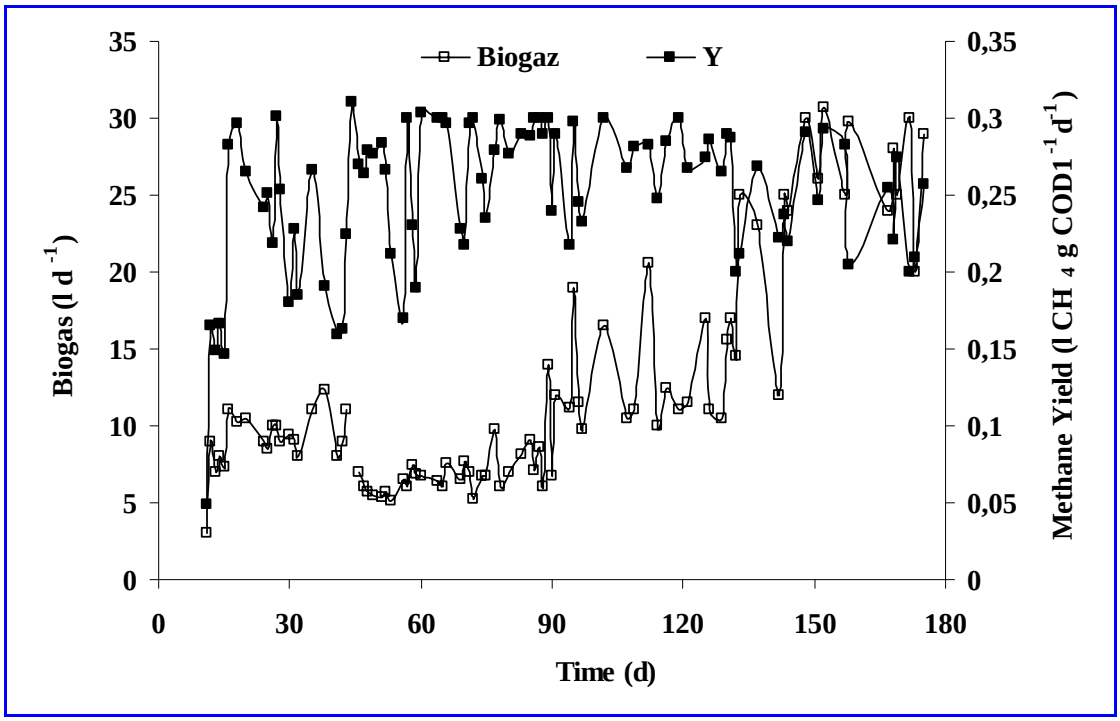

Figure 2: Biogas production and methane yield variation during Ksour Essef wastewater methanisation

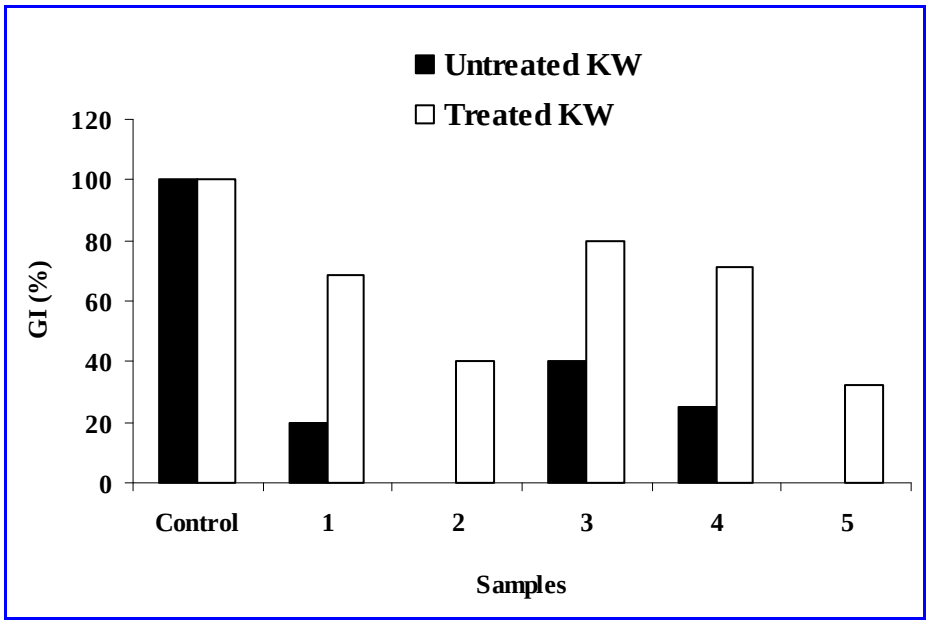

Figure 3: Germination Index (GI) of untreated and treated Ksour Essef wastewater

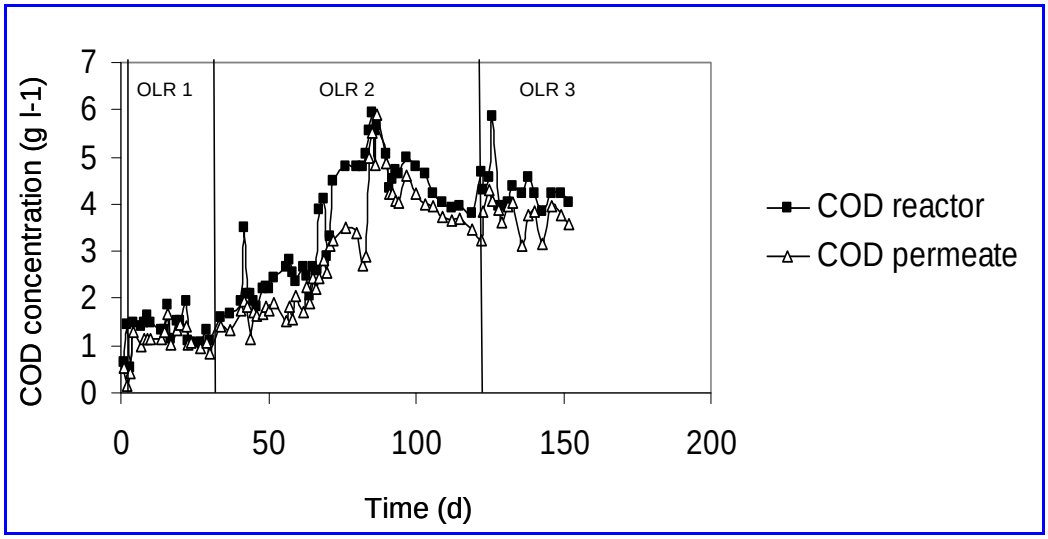

Figure 4: Evolution of COD in the reactor and in the permeate during the treatment of LFL in the AMBR

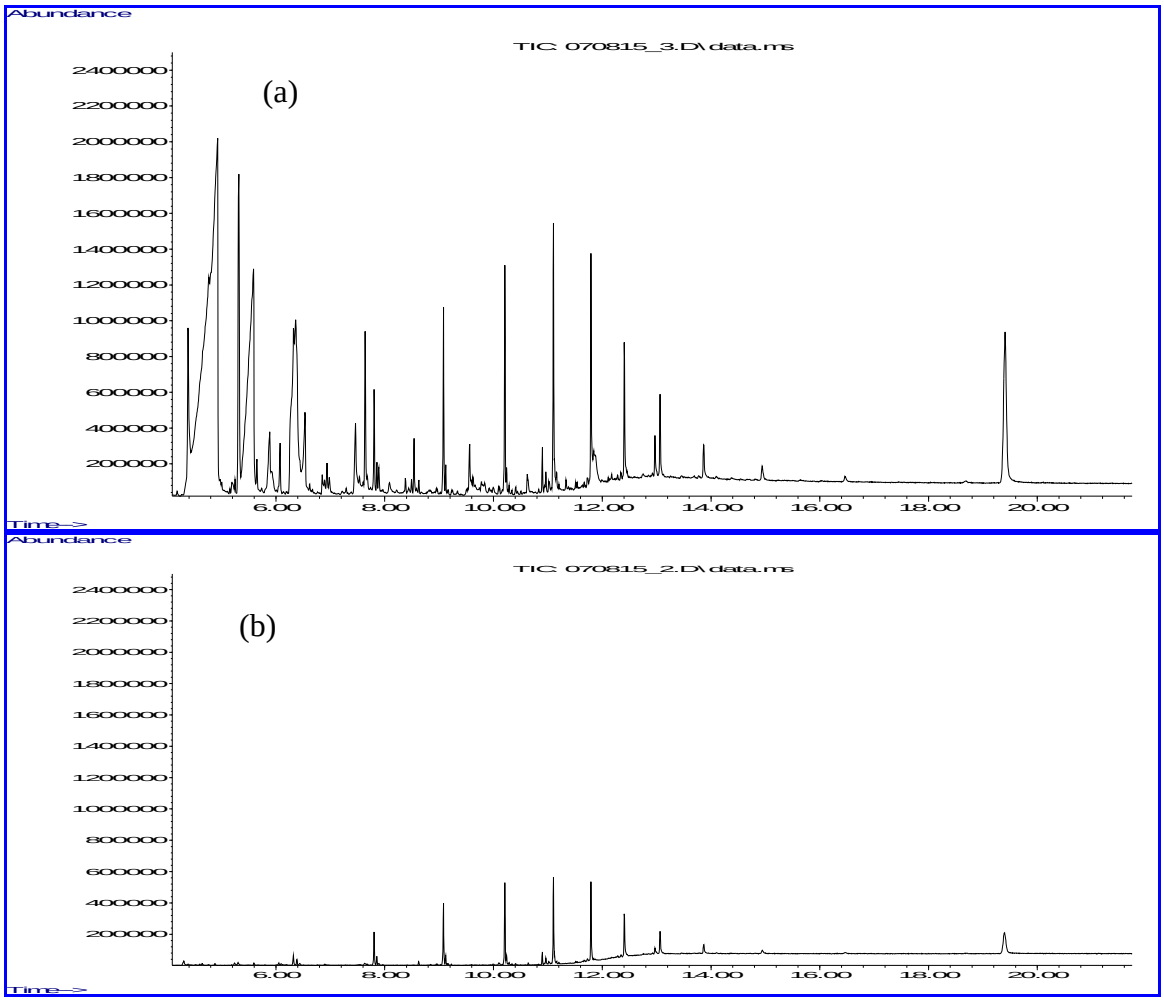

Figure 5: GC-MS chromatogram of ethylacetate-extractable products. The MS-identified compounds with respect to their retention time are the following:

(a) Untreated LFL: 4.387: Phenol; 4.769: Hexanoic acid; 5.587: Heptanoic acid; 5.887: Cyclohexanecarboxylic acid; 6.075: Hexanamide; 6.328: Octanoic Acid; 6.540: 2-Piperidinone; 7.463: Benzenepropanoic acid; 7.640: Pyridine; 7.810: 1-Tetradecene; 7.857: Tetradecane; 7.893: 7-Methylindole; 9.081: 1-Hexadecene; 9.122: Hexadecane; 10.210: E-15-Heptadecenal; 10.245: Octadecane; 10.628: Caffeine; 10.898: 1-Docosene; 10.969: Methyl-3-(3,5-diterbutyl-4-hydroxyphenyl) propionate; 10.022: Cyclohexadecane; 11.104: Cycloeicosane; 11.798: 7,9-di-tert-butyl-1-oxaspiro[4.5]deca-6,9-diene-2,8-dione

(b) Permeate: 9.081: 1-Hexadecene; 9.122: Hexadecane; 10.210: E-15-Heptadecenal; 10.898: 1-Docosene; 11.104: 1-Octadecene; 11.798: 7,9-Di-tert-butyl-1-oxaspiro(4,5)deca-6,9-diene-2,8-dione