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Landfill Leachate Treatment in Jet-Loop Membrane Bioreactor Operated Under Different Organic Loading Rates

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ABSTRACT: Since treatment of landfill leachate is quite complicated, there is a need to develop a system that is capable of providing high treatment efficiencies. In this study, the treatment performance of a jet - loop membrane bioreactor (JLMB) operated at different organic loading rates was investigated by observing the changes in Chemical Oxygen Demand (COD), Total Kjehldahl Nitrogen (TKN) and Total Ammonia (NH_3) concentrations. The same COD removal rates (83%) were observed at all the studied loading rates, and it should be noted that the biodegradable part of the leachate was removed completely. It was also observed that the NH_4 – N / TKN ratios, which were found to be approximately 0.9, did not change throughout the study. However, further treatment technologies, such as reverse osmosis, nanofiltration or ion - exchange, should be employed for the complete removal of inert COD and NH_3 in order to meet related discharge limits.

Key words: Landfill leachate, Jet-loop, Membrane bioreactor, Organic loading rate

INTRODUCTION

Due to its economic advantages, landfill is one of the most widely used methods for the ultimate disposal of municipal solid wastes (Renou *et al.*, 2008; Castrillón *et al.*, 2010). Landfill leachate is generated by percolating rain water on to the active part of the landfill site and by a series of physical, hydrolytic and fermentative degradation of organic matter, inorganic ions and heavy metals present in solid wastes. Leachate characteristics vary from one landfill to another, and over time, depending on many factors, such as the nature of the solid waste, the filling method, the level of compaction, the rainfall characteristics of the region, and the stage of decomposition of the waste (Di Laconi *et al.*, 2006; Kheradmand *et al.*, 2010; Schiopu *et al.*, 2010). The contaminant-laden concentrated leachate makes the treatment of these types of wastewaters quite complicated. COD removal is challenging, because of leachate characteristics (origin and age), treatment process type and operational factors. It was reported that MBR-based treatment technologies achieve greater COD removal rates for leachate having less biodegradable BOD:COD rates (0.03–0.16), compared to conventional systems which achieve COD removals of around 63% treating leachate with BOD:COD ratios of 0.21–0.3 (Alvarez-Vazquez *et al.*, 2004). Considering these above mentioned factors, it is necessary to develop a novel system that can yield high treatment efficiencies throughout the landfill life.

As the name implies, in jet-loop reactors, dispersion is achieved by a liquid jet drive (Dutta *et al.*, 1987; Dirix and Wiele, 1990; Velan and Ramanujam, 1991; Farizoglu and Keskinler, 2006). Liquid is injected into the reactor with a high velocity, which causes a fine dispersion of liquid and gaseous phases (Salehi *et al.*, 2005). The liquid and gas inside the draft tube flow downwards and after reflection at the bottom of the reactor, the mixture rises in the annulus between the wall of the reactor and the draft tube. At the upper end of the draft tube, a part of the fluid is recycled into the draft tube by momentum of the liquid jet (Farizoglu *et al.*, 2004). The buoyancy force of the bubbles formed also aids the loop to be continuous. The size of the bubbles is dependent on the liquid velocities and the resulting turbulence in the jet (Behr *et al.*, 2009). More information on the bubble size and their distributions can be found elsewhere (Behr *et al.*, 2009).

There are two kinds of gas dispersion in liquid phase. The first takes place through the nozzle or at the end of the nozzle, depending on the nozzle type, and the second during the dispersion of liquid within

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the reactor (Dirix and Wiele, 1990). High mass transfer properties of these types of reactors arise from both of these dispersions. The circulation of liquid for several times with the help of the draft tube increases the retention time of the gas phase in the liquid. (Wachsmann *et al.*, 1984; Velan and Ramanujam, 1991; Gaddis and Vogelpohl, 1992). In jet-loop reactors, the bubbles formed in the reactor increase the oxygen transfer to the microorganisms, and provide homogeneous dispersion of the biomass. In these types of reactors, a soluble gas, usually oxygen is transferred from a source into the liquid phase containing microorganisms. Gas, here oxygen, passes through a series of barriers created by a number of parameters related to bubble hydrodynamics, such as, temperature, cellular activity and density, solution composition, interfacial phenomena, and other factors (Bailey and Ollis, 1986).

In jet-loop reactors, due to the use of nozzle, microorganisms appear to be present in dispersed form as individuals, and not as flocs. Therefore, one of the important barriers of mass transfer, namely the diffusive transport into cellular floc does not exist (Bailey and Ollis, 1986). Some microorganisms may gather at the vicinity of the gas bubble-liquid interface, resulting in faster transportation of oxygen (Bailey and Ollis, 1986). Consequently, compared to classical treatment systems, these compact systems require less space and present a flexible approach with high treatment efficiencies in treating wastewaters having high organic loads. However, poor sludge settleability, and consequently, cloudy effluent is one of the most serious problems with jet-loop reactors (Bloor *et al.*, 1995). Therefore, the use of membrane filtration together with a jet-loop reactor not only overcomes this problem, but also increases sludge concentration in the reactor. Since leachate is quite difficult to treat, the selection of a reliable treatment process is a very significant initial step. The factors affecting the design of treatment systems are mainly the effluent discharge standards, technological alternatives and cost. During this study, an effective jet-loop membrane bioreactor (JLMB) was designed, constructed and used for the treatment of leachate.

MATERIALS & METHODS

Leachate used in this study was taken from a waste landfill site, which has been in operation since 1995, in Istanbul. As it is well-known, leachate characteristics are important in reflecting biodegradation properties (Tatsi and Zouboulis, 2002), and they might vary dramatically in time. Therefore, in this study, the physical and chemical characteristics of the leachate were monitored throughout the study and presented in Table 1.

The cylindrical reactor (outer tube) and the draft channel (inner tube) of the jet-loop membrane bioreactor were made of plexiglass having a conical bottom (height 125.5 cm, inner diameter 15 cm) with a height to diameter ratio of about 8.4:1, as depicted in Fig. 1a. As the name implies, the jet was formed by the jet head where the liquid and air were introduced through a nozzle at various ratios (Dutta and Raghavan, 1987; Dirix and Wiele, 1990; Velan and Ramanujam, 1991; Velan and Ramanujam, 1992; Salehi *et al.*, 2005; Ozkaya *et al.*, 2006; Fan *et al.*, 2007). A detailed schematic presentation of the nozzle used and the upper part of the reactor can be seen in Fig. 1b and Fig. 1c, respectively.

The temperature of the reactor was kept at 25 ± 2ºC by a cooling unit placed in the degassing tank. Cooling was carried out using tap water. Leachate was fed to the degassing tank with a peristaltic pump (Heidolph 5201). Both air and liquid flows were measured using related flowmeters (ifm and Krohne). Key operational parameters, such as dissolved oxygen, temperature and pH were measured continuously with

Table 1. Characteristics of the landfill leachate used

Parameter	mg/L[a]	Parameter	mg/L
pH	7.4–8.1	Al	1.5-2.5
COD	12000-15000	Pb	1.0-1.2
Inert COD	2250-2400	Zn	0.75-0.85
BOD_5	4000-7000	Ni	0.40-0.50
TSS	1300-1800	Fe	10.00-17.00
TKN	2400-2800	Ag	0.03-0.04
Org N	200-300	Cu	0.040-0.050
Total NH_3-N	2200-2500	As	0.040-0.060
Total P	8.5-10.2	Sb	0.055-0.070
Mg	340-360	Cd	0.022

[a]Concentration unit, except pH

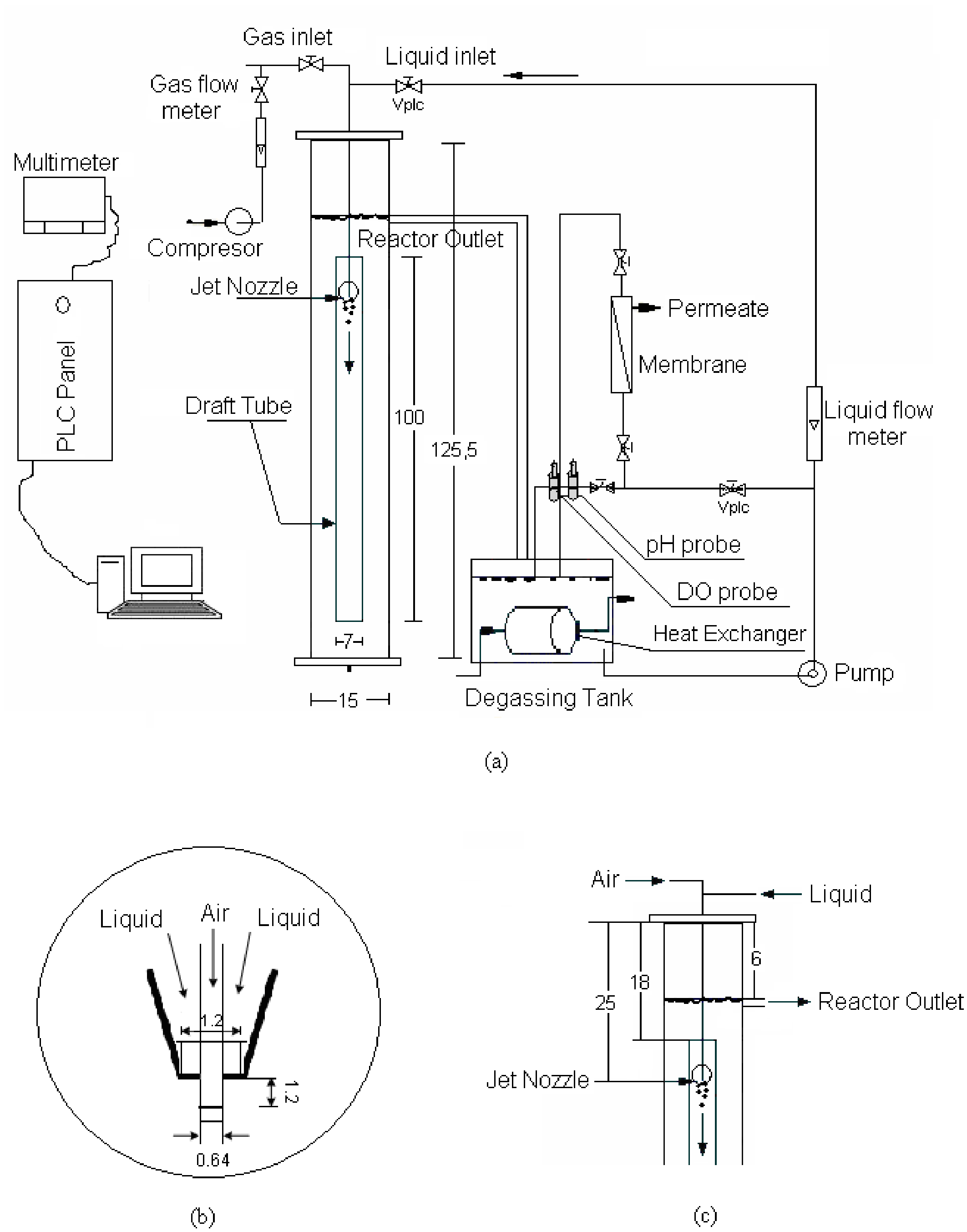

Fig. 1. (a) Schematic view of reactor, (b) detailed view of the nozzle and (c) the upper part of the reactor (V_{plc}= PLC controlled valve, the sizes are in cm)

a multi-parameter measurement device (Hach-Lange). The data recorded were transferred to a computer by the SCADA system (Siemens WinCC). A Programmable Logic Controller (Phoenix Contact ILC 350 IB) was used for the purpose of system control and monitoring. 28 L of inoculum taken from activated sludge tank of a domestic wastewater treatment plant was introduced to the JLMB. Initially, sucrose, as substrate, was added to the system in order to enable easier adaptation of microorganisms to the turbulent medium. Low volumes of leachate were introduced to the system two days after the start-up and the microorganisms were gradually adapted to the complex leachate with high concentration of pollutant. MLSS concentration reached 4200 mg/L after 58 days of operation. Slow increase of MLSS concentration was thought to be due to loss of biomass by foaming. The kinetic parameters of the system were determined respirometrically, as detailed elsewhere (Ince *et al.,* 2008).

During the course of the study, the COD, TKN, total NH_3 and MLSS analyses were carried out on a daily basis. The heavy metal analyses for the wastewater characterisation were performed using Atomic Absorption Spectrometer (AAS) with a graphite furnace (Pinel-Raffaitin *et al.,* 2006). All the chemicals used were of analytical reagent grade and water used during the experiments was laboratory distilled water. The COD analyses were carried out according to the STM 5220 C (APHA, 2005). The TKN, total NH_3, TP analyses were also performed using the STM 4500-Norg B Macro-Kjeldahl, STM 4500-NH_3 C and STM 4500-P D methods, respectively (APHA, 2005). For the determination of inert COD, wastewater fraction analysis method was used (Park *et al.,* 1997).

An external microfiltration membrane unit (Microdyn-Nadir – MD 063 TP 2N) with a pore size of 0.2 µm was placed at the outlet of the reactor. In case of high-strength / low-volume wastewaters, the tubular side-stream membrane bioreactors (MBRs) are commonly preferred (Robinson, 2005). Additionally, the tubular membranes ensuring high level of agitation were recommended for wastewaters, such as leachate, which is rich in inorganic dissolved solids (Robinson, 2005). The membrane, made of polypropylene, has an efficient filtration inner surface area of 0.2 m^2 and was formed of 19 tubes, each having an inner diameter of 5.5 mm. According to the manufacturer's recommendations, membrane chemical back-washing was carried out every seven days using NaOH (5%) for 60 min under a pressure of 100 kPa. After each chemical back-washing, the membrane module was rinsed with distilled water until neutral pH values were attained. The physical back-washing was carried out every 24 hours using tap water for 3 minutes at a pressure of 200 kPa.

RESULTS & DISCUSSION

The mean values of COD and BOD_5 of leachate were 13225 and 5789 mg/L, respectively. As known, BOD_5:COD ratio represents the proportion of biodegradable organics in leachate. In case of young leachate, large portion of the organic matter consists of volatile fatty acids which are easily biodegradable. Therefore, the BOD5:COD ratio during this phase is generally 0.4–0.5 or even higher (Ozkaya *et al.,* 2006). As the landfill gets older, the BOD5:COD ratio decreases to reach almost zero (Fan *et al.,* 2007). This is due to the decomposition of most of the organics present in leachate over time. In this study, the mean BOD5:COD ratio of landfill leachate was found to be 0.44, which indicated that the leachate can be treated using biological treatment. A lower BOD5:COD ratio was expected for the leachate collected from an intermediate-old landfill site. The high values obtained indicated that the leachate from newer cells might have reached to the leachate from old cells. The reason for significant difference between biodegradable COD and BOD5 results was thought to originate from the toxic substances and refractory materials present in leachate (Marttinen *et al.,* 2002).

The JLMB system was operated continuously for approximately 11 weeks. As can be seen from Fig. 2a, during the first 10 days of the run, the COD removal efficiencies were quite low as a result of low MLSS concentrations at the start-up period. After the initial few days, COD removal efficiencies of around 80% were obtained throughout the study, in which the organic loading rate varied between 4.46 and 9.72 kg COD/m3·d. It should be noted that due to the low flux values, during the course of the study, the desired organic loading rates (OLR) and hydraulic retention times (HRT) values could not be applied. At each OLR study, the system was run until the steady-state conditions were reached, and after that, the system was run for at least another 7 days.

As can be seen from Fig. 2a, the OLR changes did not affect the COD removal efficiencies which stayed around 80-85%. Here, the COD removed was the biodegradable part of the leachate. The effluent COD value was around 2000 mg COD/L, which was non-biodegradable portion of the total influent COD. However, it's worth noting that the inert COD values of influent were higher than that of effluent. This could be explained in three ways: (i) the refractory substances present in leachate were broken down into smaller pieces due to high turbulent medium. Therefore, these substances have the chance of more contact with microorganisms in jet-loop bioreactors; (ii) the refractory substances could stay longer in the activated sludge due to the loop formed (Farizoglu and Keskinler, 2006), and (iii) the membrane unit integrated to the system and selective action of the dynamic layer formed during the filtration both have effects on the removal of the refractory substances.

A short-term foaming was observed initially at each applied loading rate. It is believed that the organic loading rate is crucial for optimal conditions of microorganisms. When the loading rate was changed, more extracellular polymeric substances (EPS) (Meng *et al.,* 2009) were released (Nakajima and Mishima, 2005), which cause foaming (Judd, 2006). As soon as, the steady-state conditions were reached, the foaming problem was over.

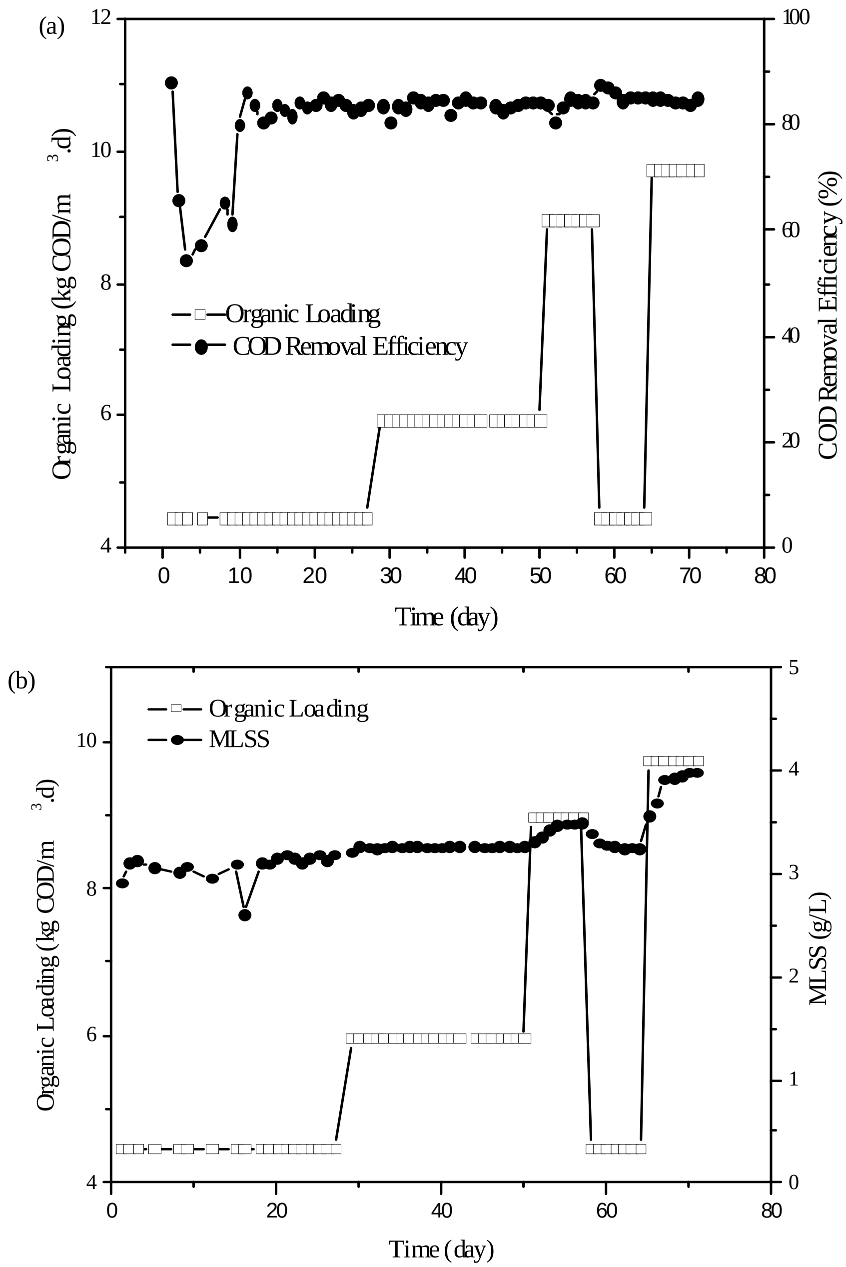

Fig. 2. (a) The COD removal efficiencies and (b) MLSS concentration change in relation to the OLR

Contrary to the literature (Yildiz *et al.,* 2005; Farizoglu and Keskinler, 2006), the biofilm formation was observed on the inner surface of the reactor immediately after the reactor was started up. The different biofilm formations could be due to the characteristics of substrate and the activated sludge used. It was also observed that the biofilm formed did not detach from the surface in time, since biofilm was not thick enough to prevent oxygen transfer to the attached microorganisms.

The HRT values were adjusted to 1.35 and 2.93 days and the SRT values were adjusted to 2.16 and 5.33 days, by a peristaltic pump. Here, the advantage of JLMB, which treats highly polluted wastewaters at low HRT values and high OLR values in one unit, comes forward, especially compared to classical biological treatment systems (Kurniawan *et al.,* 2006) treating such wastewaters with more than one unit. Direct comparison of the results, obtained in this study, with the other literature studies is difficult. Changing characteristics of leachate from one landfill to another is one of the major reasons. In literature, the HRT values for an MBR treating leachate ranged from 1 to 4 d (Setiadi and Fairus, 2003; Alvarez-Vazquez *et al.*, 2004; Vasel *et al.*, 2004; Chaturapruek *et al.*, 2005). With regard to SRT, the literature values ranged from 10 to 100 d (Yao *et al.*, 2008; Hasar *et al.*, 2009; Svojitka *et al.*, 2009). It should be noted that higher SRT values could not be obtained in the system due to the combined effect of two factors, which are high OLR and low membrane flux values.

In another treatment study on leachate with low BOD:COD ratios (0.03–0.16) using an MBR, it was reported that the COD removal rate was 80% with OLR values of 1–3 kg COD/m3·d and HRT values of 2-3 day (Alvarez-Vazquez *et al.*, 2004). In this study, the COD removal rate was around 80% and the effluent COD values were lower than that of influent inert COD values. Furthermore, the BOD:COD ratio and OLR applied were higher and the HRT value was almost the same compared with the study carried out by Alvarez-Vazquez *et al.*, 2004.

The relationship between OLR and MLSS concentration is given in Fig. 2b. It was observed that the MLSS concentration increased in relation to the OLR increase. When a new loading rate was applied, the MLSS concentration showed small fluctuations initially, however, in a few days a constant value was attained. It's worth noting that although a change was observed in the MLSS concentrations with different loading rates, this effect was not the same at each loading rate due to the biofilm formation on the inner surface of the reactor.

The effluent TKN and total NH3 concentrations, which were monitored from the tenth day onwards, changed in parallel to the loading rate applied (Fig. 3). The NH4-N:TKN ratio, which is a parameter used for system stability (Gharsallah *et al.*, 2002), can also be used as an indicator for nitrification. Nitrosomonas can only oxidize NH4-N to NO2-N, while Nitrobacter is limited to the oxidation of NO2-N to NO3-N (Cheremisinoff, 1996). In cases where nitrification occurs, the effluent NH4-N:TKN ratio is expected to be lower than the influent NH4-N:TKN ratio. The NH4-N:TKN ratios, which were approximately 0.9, did not change throughout the study, therefore, it was thought that nitrification did not occur in the JLMB because of high concentrations of total NH3 and short SRT values. As known, high concentrations of total NH3 have an inhibition effect on nitrobacteria and nitrosomonas species (Vadivelu *et al.*, 2007). It was also reported previously that an MBR can be operated efficiently at long SRTs with high carbonaceous and nitrogenous matters removal (Stephenson *et al.*, 2000).

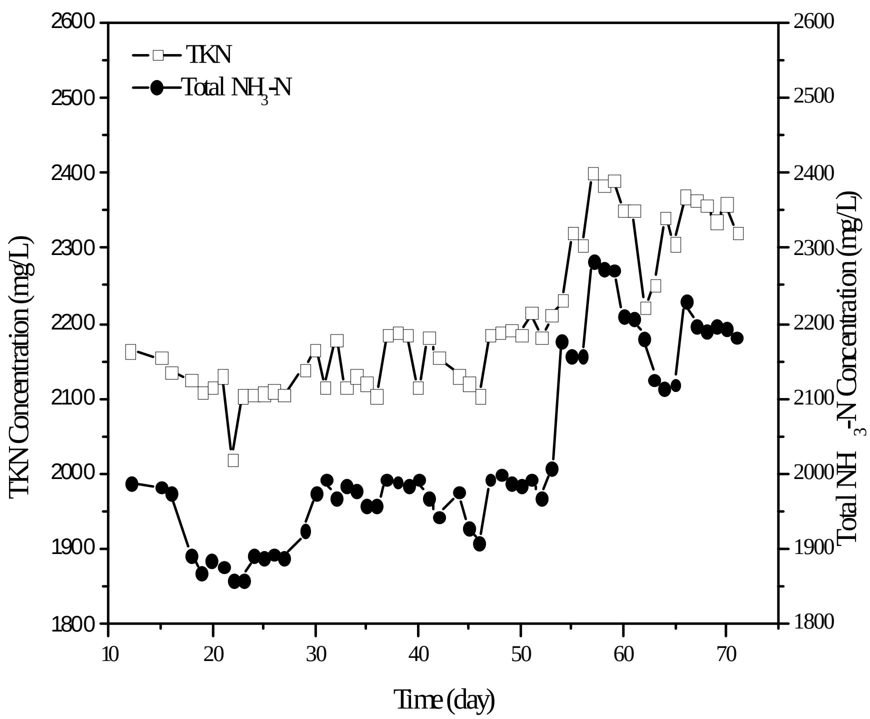

Fig. 3. The effluent TKN and total NH_3 concentrations

CONCLUSION

Compared to the classical systems, JLMB is quite advantageous in treating biodegradable part of highly polluted wastewaters in one-stage with lower total retention times and higher organic loading rates. In this study, at all the studied loading rates, the same COD removal rates were attained and it was observed that the biodegradable part of the leachate was completely removed. However, it should be noted that the effluent of the system needs subsequent (post-) treatment, since the discharge limits could not be met. Thus, in conclusion, advanced treatment technologies such as reverse osmosis, nanofiltration or ion-exchange could be employed for the complete removal of inert COD and NH3 from the effluent of JLMB.

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