Nitrogen removal during the treatment of wastewater and landfill leachate in a biological aerated filter

G.D. Jojoa-Unigarro, S. González-Martínez

Institute of Engineering, National University of Mexico (Universidad Nacional Autónoma de México), Ciudad Universitaria, 04510 Mexico City, Mexico

Abstract

Several efforts have been found to treat landfill leachate together with municipal wastewater. Treatment of leachate and municipal wastewater has extended worldwide using already existing wastewater treatment plants. A biological aerated filter (BAF) was used to treat leachate from Mexico City's largest landfill together with synthetic wastewater. After stable operation of the BAF under organic loading rates of 8.1, 5.3, and 3.1 $gCOD/m^2 \cdot d$, landfill leachate was added to the wastewater in volume proportions of 1, 2, and 3 %. Without leachate addition, total COD removal decreases with increasing organic load reaching a maximum value of 94 % with the lowest organic load. With leachate addition, COD removal decreases with increasing leachate concentration. Without leachate, full nitrification was achieved with the two lower organic loads and 93 % NH_4-N removal was achieved with the highest organic load. With 1 % leachate addition, full nitrification was achieved; NH_4-N removal was 71 and 39 % with 2 and 3 % leachate added, respectively. The oxygen uptake rates decrease with increasing leachate concentration. With 3 % leachate, O_2 uptake rates for endogenous respiration are higher than for substrate respiration. Substrate respiration rates tend to a straight line with intercept near zero.

Keywords

Leachate; oxygen uptake rates; biological aerated filter; BAF; nitrification

INTRODUCTION

Leachate from landfills are complex mixtures of inorganic substances, heavy metals, ammonia nitrogen, biodegradable and recalcitrant organic compounds, which are a product of precipitation, percolation, compaction and degradation of solid wastes (Kjeldsen et al., 2002). Leachate can infiltrate, pollute groundwater and travel long distances contaminating water sources (Maqbool et al., 2011). Leachate treatment can be classified into biological and physicochemical. Biological processes are generally used for the treatment of young leachates, because they contain biodegradable organic substances. Physicochemical processes are used for the treatment of old leachates containing high concentrations of ammonia nitrogen and recalcitrant organic compounds (Renou et al., 2008; Cassano et al. 2011).

The limitations of biological treatments to treat old leachates can be overcome by performing a combined treatment of leachate and municipal wastewater. The treatment of leachate and municipal wastewater has extended worldwide and has an economic advantage when using already existing wastewater treatment plants when the leachate to wastewater mixture does not exceed certain values, for example 10 % (Renou et al., 2008). Most articles found report combined treatment of leachate and wastewater using aerobic systems. For leachate/wastewater ratios of 0.2 to 0.5 COD removals higher than 80% have been achieved (Del Borghi et al., 2003; Çeçen and Aktas, 2004, Qiu et al., 2007). Ferraz et al. (2014) show that a biological aerated filter (BAF) can achieve high removal rates of carbonaceous material and also oxidize recalcitrant organic matter from leachates when a pretreatment of leachate is implemented to remove excess ammonia nitrogen.

This work evaluates the combined treatment of old leachate from Mexico City's largest landfill with synthetic wastewater, without pretreatment of the leachate, using a pilot biological aerated filter.

MATERIALS AND METHODS

Pilot filter and filter media

The pilot filter consists of a PVC pipe filled with lava stones. The effluent was placed 10 cm above the upper stone layers to allow treated wastewater to flow freely through the effluent pipe. Two other effluent pipes were placed at 20 and 30 cm above the upper filter media layer; the first one to collect backwash water and the other as emergency effluent. A diffuser was placed at the bottom of the filter to supply both low-pressure air for oxygen supply and high-pressure air for backwashing (figure 1). The main characteristics of pilot filter and filter media are shown in table 1. Sampling valves were placed at 20 cm intervals.

Table 1. Pilot and filter media characteristics

Pilot filter characteristics		Filter media characteristics	
Total height	1.90 m	Average particle size	9.0 mm
Internal diameter	0.15 m	Specific area	427 m^2/m^3
Effective volume	9.02 L	Apparent porosity	51 %
Filter bed depth	1.0 m	Total surface area	7.7 m^2
Total volume	16.7 L	Lava stones density	1,540 kg/m^3

Filter media

The material selected as filter media is of volcanic nature (*tezontle* is the common name in Mexico) and has high porosity, roughness, structural strength, resistance to attack by chemicals and exposes adequate surface area for biofilm formation. Important is availability and low cost (table 1 and figure 2).

Synthetic wastewater

In order to reduce operation variables, synthetic wastewater was used. The carbon source is a mixture of maltodextrin (62 mg/L, 105 mgCOD/L) and hydrolyzed protein from sorghum (310 mg/L, 245 mgCOD/L). Also added were $(NH_4)_2SO_4$ (140 mg/L, 30 mg/L as nitrogen), $MgSO_4·7H_2O$ (25 mg/L), K_2HPO_4 (56 mg/L, 10 mg/L as phosphorus), $FeSO_4·7H_2O$ (20 mg/L), $NaHCO_3$ (105 mg/L), and a solution containing the following micronutrients H_3BO_3, ZnCl, $CuCl_2$, $MnSO_4·H_2O$, $(NH_4)_6Mo_7O_2·4H_2O$, $AlCl_3$, $CoCl_2·6H_2O$, $NiCl_2$ to result in corresponding concentrations of 0.05 mg/L in the wastewater. This composition has been tested in several investigations with success (Cobos-Becerra and González-Martínez, 2014).

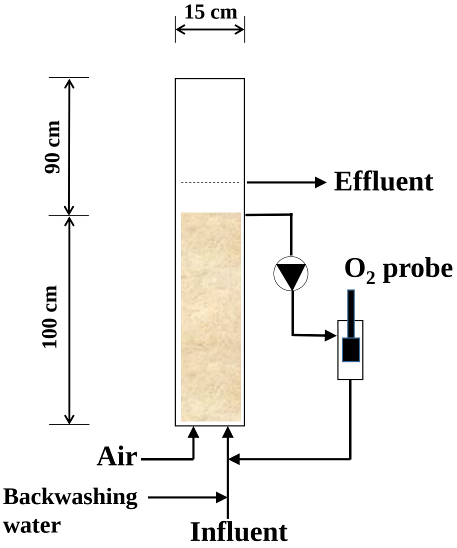

Figure 1. The pilot filter.

Figure 2. Lava stones with 9.0 mm average size.

Landfill leachate

The leachate used in this study was from the landfill *Bordo Poniente* third stage, located in Mexico City. The landfill was in operation for 30 years and was closed in 2012 after receiving approximately 12,500 ton/d urban wastes, mostly unsorted. The leachate analysis resulted in 1408 mg/L COD, 150 mg/L BOD, 818 mg/L Kjeldahl nitrogen (KN), 731 mg/L ammonia nitrogen (NH_4-N), 163 mg/L phosphorus from phosphate (PO_4-N) and pH 8.5. The leachate was classified as old, mainly due to high contents of ammonia nitrogen and a BOD/COD ratio of 0.1.

Filter operation

During the start-up the reactor was inoculated with sludge from an activated sludge municipal wastewater treatment plant and the system was allowed to run under an organic loading rate (OLR) of 3.1 $gCOD/m^2·d$ until stable operation could be observed. After that, three organic loading rates were sequentially adjusted to 8.1, 5.3, and 3.1 $gCOD/m^2·d$. The organic loading rate was increased by increasing the influent COD. After the operation under each OLR reached stability (constant COD removal and at least 3 cellular retention times (CRT) were achieved) the new OLR was adjusted.

Considering that microbial systems are more stable under lower OLR, the leachate was added when operating under an OLR of 3.1 $gCOD/m^2·d$. At different times, leachate volume rates of 1, 2, and 3 % were added to the wastewater. The pilot filter operated under a regime of continuous wastewater flow of 66 L/d, which resulted in constant hydraulic retention time (HRT) of 3.7 h. The air flow was maintained at 2 L/min.

Wastewater analysis

In order to evaluate the filter performance, the following parameters were determined: COD, Kjeldahl nitrogen, ammonia and nitrate nitrogen, phosphorus from phosphates, total and volatile solids. All determinations were performed according to Standard Methods (APHA, 2012).

Oxygen uptake determinations

Respirometry is the measurement and interpretation of the biological oxygen consumption rate under well-defined experimental conditions. Because oxygen consumption is directly associated with both biomass growth and substrate removal, respirometry is a useful technique for monitoring, modelling and control of the biological processes (Vanrolleghem, 2002).

The arrangement is shown in figure 1. Effluent water was taken and pumped through a device where a dissolved oxygen probe was placed and then the water was allowed to return to the reactor through the lowest sampling port to close the circuit. While the influent wastewater pump was turned off, in order not to modify the hydraulic conditions inside the filter, the pump acting on this device was allowed to run with the same flow as the influent wastewater flow. With the device running all the time, alternately, the air supply to the filter was cut during 15 min and then allowed again another 15 min during one whole HRT (3.7 h). Dissolved oxygen concentration was continuously recorded during the duration of the experiment.

This procedure was repeated several times for every organic loading rate without leachate and, later, with leachate. The day after each experiment, during 5 HRT, the filter was fed with wastewater without carbonaceous matter and ammonia nitrogen. The objective was to create a condition when the dissolved oxygen uptake could be considered endogenous (without exogenous substrates). Then, the procedure above described was repeated to obtain oxygen uptake rates without exogenous substrate and evaluate the effects of leachate on the performance of the microorganisms.

RESULTS

COD removal

During the start-up period the reactor was operated under an organic loading rate of 3.1 $gCOD/m^2·d$ during 26 days and, after this point, the water analysis began. From day 1 to 78 the backwashing procedure was performed every third day and the solids in the reactor tended to accumulate at the bottom. After day 78, backwashing was performed every day.

Figure 3 shows that at the influent soluble and total COD presented differences caused by concentrate precipitation in the container. The wastewater feeding procedure was changed to prepare fresh synthetic wastewater every day after day 78. Figure 3 also presents the overall performance of the reactor according to COD analysis. From day 26 to 93 the OLR was set to 8.1 $gCOD/m^2 \cdot d$ After day 80 the results were stable: COD_T and COD_D removal rates were 90 and 92 %, respectively.

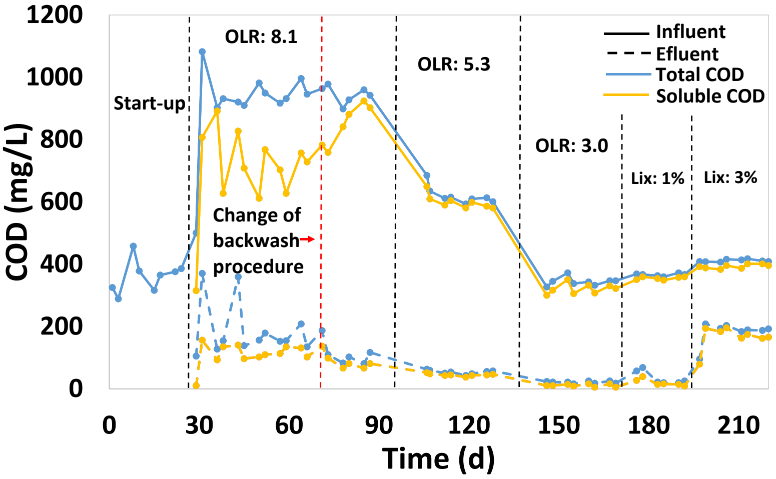

Figure 3. Total and dissolved COD along the whole experimentation.

Table 2 presents values for influent, effluent and removal for total and dissolved COD during the whole experimentation. In this table total suspended solids (TSS) in the effluent is also shown. From day 93 to 135 the OLR was 5.3 $gCOD/m^2 \cdot d$ and the COD removal rates were 91 and 93 % for COD_T and COD_D, respectively. From day 135 to 170 the OLR was adjusted to 3.0 $gCOD/m^2 \cdot d$ and the cod removal was 94 and 97 % for total and dissolved COD, respectively. The las two stages correspond to operation under an OLR of 3.0 $gCOD/m^2 \cdot d$ with the addition of leachate at 1 and 3 % in volume in the wastewater. Total COD removal slightly decreased (from 94 to 90 %) when leachate in 1 % volume was added and soluble COD decreased from 97 to 94 %. When leachate at 3 % volume was added, an important decrease was observed in total COD from 90 to 60 % and, for soluble COD, from 94 to 58 %.

Table 2. Average results for COD and solids for all experimentation stages.

Organic load	**Total COD**			**Dissolved COD**			**TSS**
	Infl	**Efl**	**Rem**	**Infl**	**Efl**	**Rem**	**Efl**
$gCOD/m^2 \cdot d$	mg/L	mg/L	%	mg/L	mg/L	%	mg/L
8.1*	922	181	80	708	111	84	53
8.1	932	95	90	887	74	92	35
5.3	620	54	91	600	45	93	23
3.1	344	21	94	321	11	97	12
Lix 1%	366	35	90	355	20	94	13
Lix 2%	381	122	68	374	108	71	---
Lix 3%	411	165	60	393	165	58	20

* Backwashing procedure was performed every three days.
All other values were determined with daily backwashing.

Originally, the experimental proposition considered leachate addition to a maximum of 5 % in volume of the wastewater. But, as the important removal reduction was observed with 3 %, it was decided to try another 2 % to evaluate the possible removal reduction with a lower leachate concentration. Table 2 presents also COD removal rates for 2 % leachate addition indicating that there is proportional removal decrease with the leachate concentration.

Table 2 also present total suspended solids (TSS) in the effluent. As expected, TSS decrease with the organic load from 35 to 23 to 12 mg/L for OLR 8.1, 5.3, and 3.1 gCOD/m^2·d, respectively. When leachate with 1 % was added, the TSS did not change significantly when compared to 0 % leachate was added. For leachate addition 3 %, TSS increased to 20 mg/L indicating that the produced biomass did not remain as attached but as suspended solids transported with the water flow.

Ferraz et al (2014) obtained similar removal rates to this research. They operated a filter fed with a mixture of municipal wastewater and mature leachate at concentrations of 2 and 5 % with retention time of 24 h. The mixture was treated through stripping to remove part of the ammonia nitrogen. Removal rates were 98 % BOD, 80 % COD and 90 % TSS under an organic load of 0.4 kgCOD/m^3·d at 2 % volume leachate in municipal wastewater.

Figure 4 presents the total COD profiles along the filter height for the operation without and with leachate addition. With the exception of leachate addition 2 %, all other values were determined with one week difference (duplicate) to guarantee system stability. The two profiles for every condition are similar in all cases.

For the system without leachate addition the highest removal is achieved through the first 20 cm of the filter. The higher the COD, the higher the removal in the first 20 cm. For the lowest influent COD, after the first 20 cm, no significant removal can be observed. For higher COD values, the removal is low after the first 20 cm.

With leachate, although the removal during the first 20 cm is between 35 to 55 % for the three different leachate concentrations, the rest of the filter height removes only between 20 to 35 %. The COD removal increases with decreasing leachate concentration.

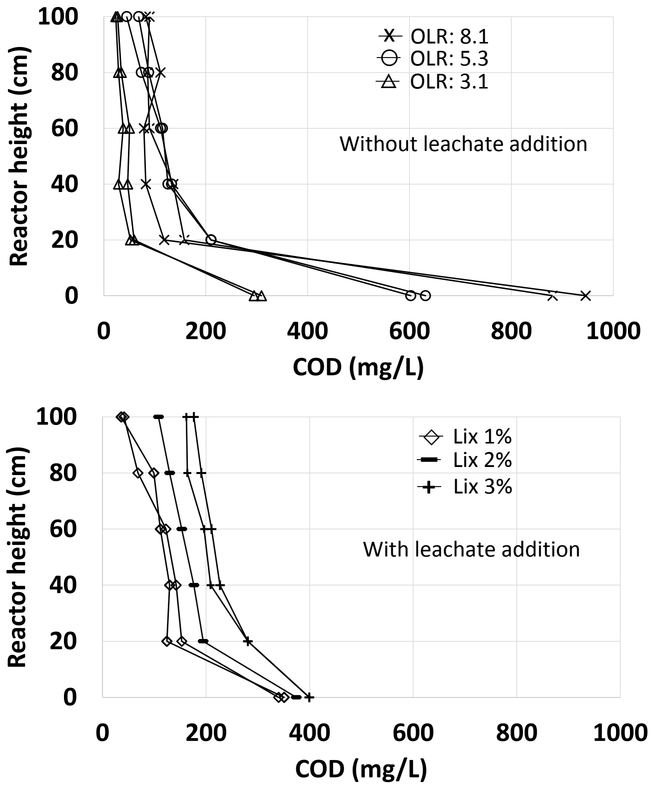

Figure 4. Total COD profiles for all experimental conditions without and with leachate addition.

Nitrogen removal

Figure 5 shows ammonia and nitrate nitrogen concentrations during the whole experimentation. Before the changing of the backwashing procedure from every 3 days to every day, nitrification was poor and, after the change, satisfactory nitrification was achieved. With the OLR of 8.1 gCOD/m^2·d, ammonia nitrogen removal was 93 %. Lowering the OLR improved nitrification to near 100 % for OLR of 5.3, 3.1 gCOD/m^2·d without leachate addition and with 1 % leachate addition. With leachate addition of 2 %, ammonia nitrogen removal decreased to 71 % and to 39 % with 3 % leachate (table 3). It is evident that leachate addition has a negative effect on nitrification. From figure 5, nitrate concentrations in the effluent correspond to nitrification indicating that ammonia nitrogen was mostly oxidized to nitrate and not used for microbial reproduction.

Çeçen and Çakiro (2001) worked with a leachate concentration of 5 % in municipal wastewater removing 84 % of COD and 95 % of ammonia nitrogen under 10 days of aeration. They observed that most of the ammonia nitrogen removal was ammonia volatilization when only 17 % of ammonia nitrogen was converted to oxidized nitrogen compounds.

Figure 6 shows the behaviour of nitrogen concentrations along the filter height. Independently of the OLR, for the cases without leachate addition, ammonia nitrogen profiles indicate that almost all nitrification was observed within the first 20 cm of the filter. Nitrate production was proportional to the ammonia nitrogen concentration in the influent. For the lowest OLR, within the first 20 cm, nitrate nitrogen reached a maximum value of 28 mg/L, slightly decreasing afterwards to reach approximately 20 mg/L at the effluent.

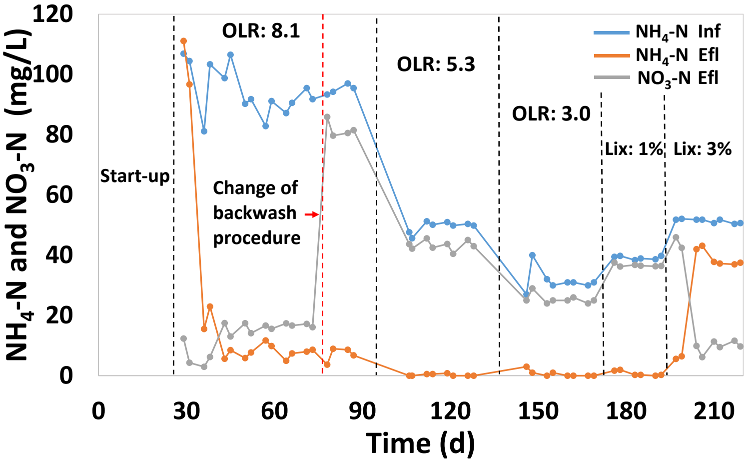

Figure 5. Ammonia nitrogen at influent and effluent and nitrate nitrogen at the effluent during the whole experimentation.

The highest OLR produced almost 70 mg/L nitrate nitrogen within the first 20 cm and then increasing the concentration to 85 mg/L at the effluent.

Table 3. Average results for ammonia nitrogen, nitrate, and phosphorus for all experimental stages.

Organic load	**NH_4-N**			**NO_3-N**	**PO_4-P**		
$g/m^2 \cdot d$	**Infl** mg/L	**Efl** **mg/L**	**Rem** %	**Efl** mg/L	**Infl** mg/L	**Efl** mg/L	**Rem** %
8.1*	94	9	90	13	41	34	17
8.1	95	7	93	82	42	28	33
5.3	49	ND	100	43	30	18	40
3.1	32	ND	100	25	16	12	25
Lix 1%	39	ND	100	37	17	12	29
Lix 2%	45	13	71	28	17	12	29
Lix 3%	51	31	39	18	17	14	18

* Backwashing procedure was performed every three days.
All other values were determined with daily backwashing.

Ammonia nitrogen concentrations for leachate addition of 1 are similar to those without leachate addition: The highest removal takes place within the first 20 cm, observing slight removal within the other 80 cm (figure 6). Slightly higher ammonia nitrogen concentrations can be observed for 2 % leachate addition indicating that leachate affects negatively nitrification. Only with the leachate addition of 3 % the behaviour is different: From influent to effluent, concentrations tend to a straight line indicating poor ammonia nitrogen removal. The nitrate nitrogen production behaves corresponding to ammonia nitrogen concentration: The higher the leachate concentration, the lower the nitrate nitrogen production. It can be concluded that 1 % in volume leachate concentration does not affect negatively the performance of the filter and nitrification and that 3 % leachate concentration is high presuming inhibition of nitrification.

The highest phosphorus removal (40 %) was observed with OLR of 5.3 g/m^2·d. With leachate addition 1 and 2 % the removal was 29 % and decreased with 3 % leachate to 18 % (table 3).

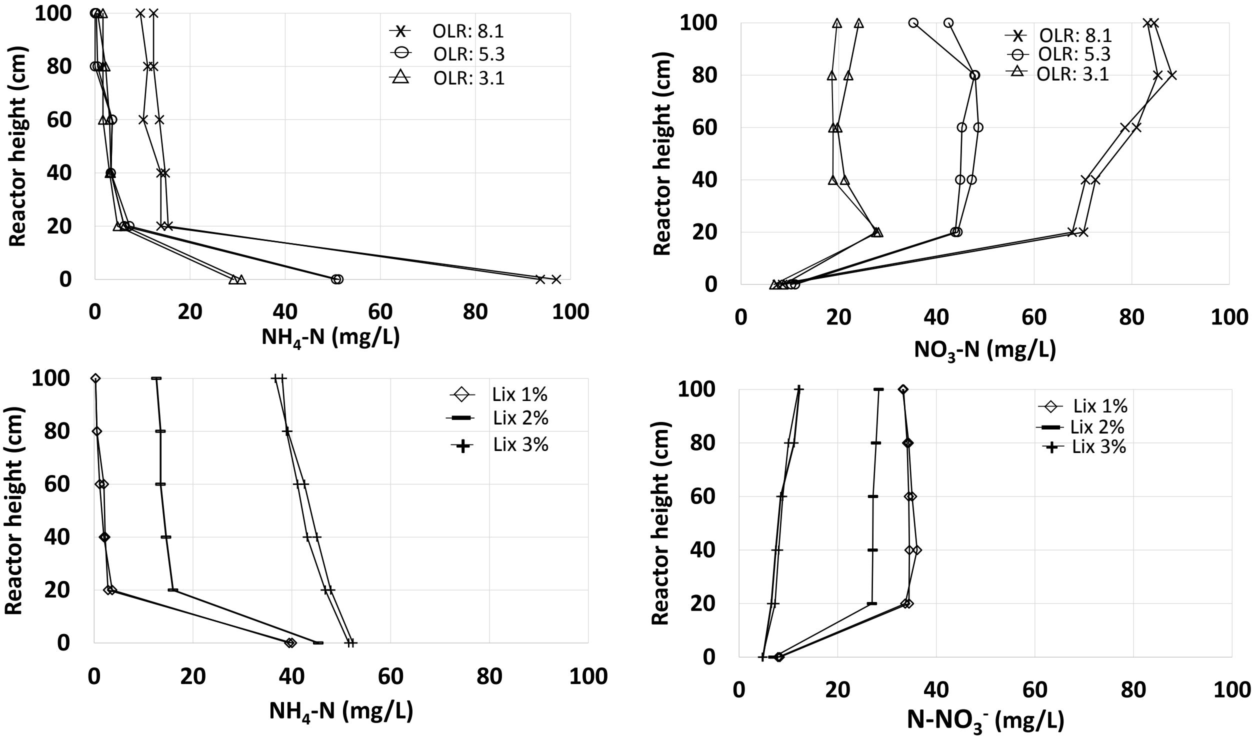

Figure 6. Ammonia and nitrate nitrogen concentrations as function of filter height (profiles).

Respirometry

As an example of the results, figure 7 shows the raw curves obtained during one HRT when aeration was provided every 15 minutes and the oxygen uptake curves (lines) from the first 3 minutes after aeration was shut-down every 15 minutes. The slopes are similar in all cases and the differences were caused by the oxygen concentration at the beginning of every determination. This procedure was made for every tested condition in the experiment.

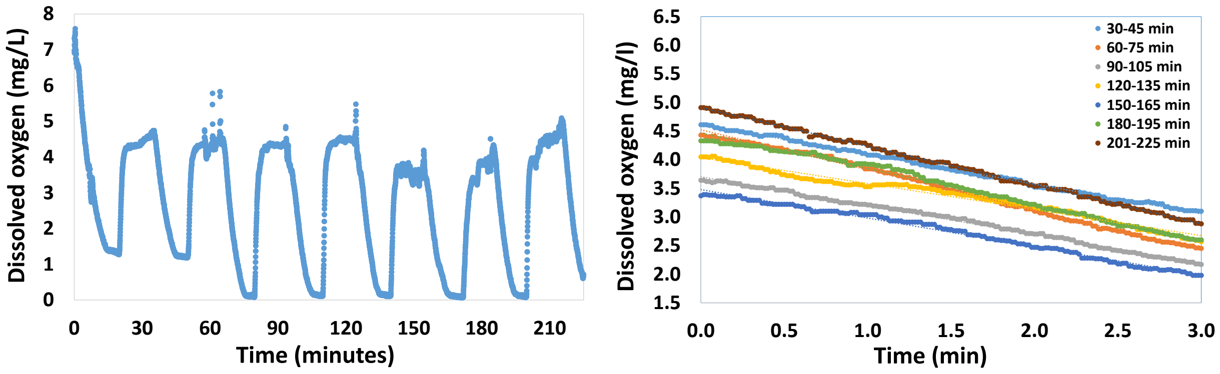

Figure 7. On the left is the dissolved oxygen concentration raw curve and on the right is the corresponding points to the first 3 minutes after air supply was suspended every 15 min. This example correspond to the OLR 8.1 g/m^2·d without leachate.

Derived from the oxygen uptake rate determinations, figure 8 presents the oxygen uptake rates for all the conditions applied during the experimentation. Figure 8 also shows the oxygen uptake rates for substrate and endogenous respiration. Oxygen uptake rates increase with increasing substrate concentration (COD).

For the lower organic loads (3.1 and 5.3 $gCOD/m^2 \cdot d$, corresponding to 350 and 600 mgCOD/L) exogenous and endogenous uptake rates increase almost parallel but, for the OLR 8.1 $gCOD/m^2 \cdot d$ (1,060 mgCOD/L) endogenous respiration rates increase slightly from 0.184 to 0.210 $mgO_2/L \cdot min$ indicating that endogenous uptake rates tend to a maximum: Microorganisms can accumulate a finite amount of reserve substances that, eventually, can be used as endogenous substrate. This behaviour affects the total oxygen uptake rate correspondingly. Important is to note that substrate (exogenous) respiration tends to a straight line and the intercept to zero.

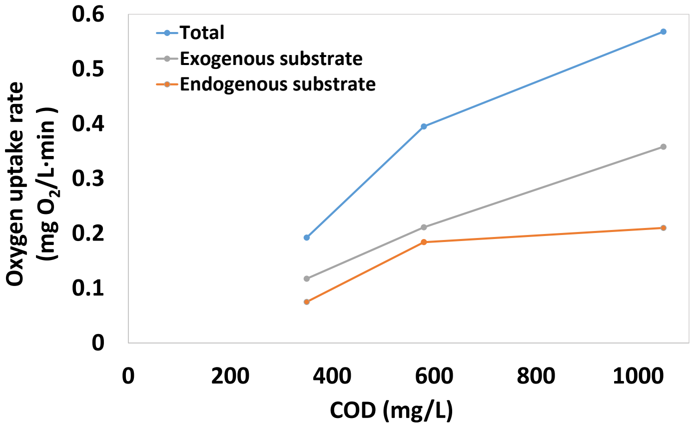

Figure 8. Oxygen uptake rates without leachate addition.

Figure 9 presents the oxygen uptake rates in relation to leachate concentration. With increasing leachate concentration, the substrate and total oxygen uptake rates decrease. The endogenous respiration rate tend to decrease for 1 % leachate, then it remains unchanged for 2 % and for 3 % the rate slightly increases.

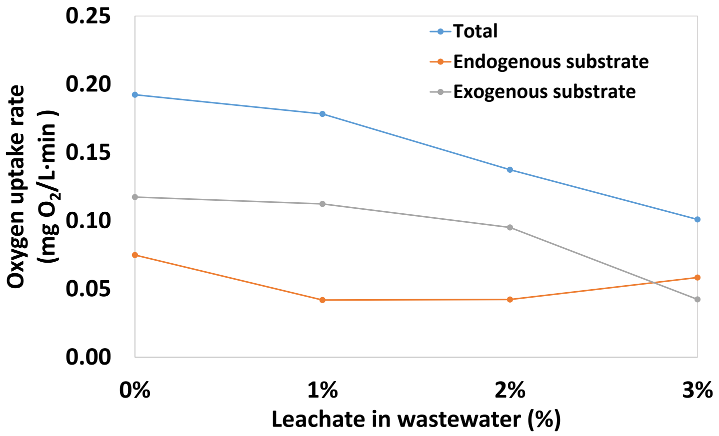

Figure 9. Oxygen uptake rates with leachate addition.

Leachate negatively affects respiration rates similarly to COD removal. The lowest respiration rate is observed with the highest leachate concentration of 3 %. At this concentration, the endogenous uptake rate is higher than the substrate respiration rate.

CONCLUSIONS

Without leachate addition, COD removal decreases with increasing organic load, reaching a maximum value of 94 % with the lowest organic load. With leachate addition, COD removal decreases with increasing leachate concentration from 94 % without leachate to 90 % with 1 % leachate and to 60 % with 3 % leachate.

Without leachate, approximately 80 % of COD removal takes place within the first 20 cm of the filter. Without leachate, full nitrification was achieved with the lower organic load and 93 % NH_4-N removal was achieved with the highest organic load. With 1 % leachate addition, full nitrification was achieved; NH_4-N removal was 71 % with 2 % leachate and 39 % with 3 % leachate.

Without leachate, oxygen uptake rates increase with increasing substrate (COD) concentration. Substrate respiration rates tend to a straight line with intercept near zero. With leachate the oxygen uptake rates decrease with increasing leachate concentration. With 3 % leachate, uptake rates for endogenous respiration are higher than for substrate respiration.

ACKNOWLEDGEMENT

This research was possible thanks to the support provided by the General Directorate for Academic Affairs (DGAPA) of the National University of Mexico, project IN110115, and to the scholarship provided by the Mexican National Council for Science and Technology (CONACyT).

REFERENCES

American Public Health Association (APHA), American Water Works Association (AWWA), Water Environment Federation (WEF), 2012. *Standard Methods for the Examination of Water and Wastewater*, 22th ed. APHA, AWWA, WEF, Washington.

Del Borghi, A. Binaghi, L., Converti, A., Del Borghi, M. 2003. Combined Treatment of Leachate from Sanitary Landfill and Municipal Wastewater by Activated Sludge. *Chem. Biochem. Eng. Q.*, 17 (4) 277– 283.

Cassano, D., Zapata, A., Brunetti, G., Del Moro, G., Di Iaconi, I., Oller, I., Malato, S., Mascolo, G. 2011. Comparison of Several Combined/integrated Biological-AOPs Setups for the Treatment of Municipal Landfill Leachate: Minimization of Operating Costs and Effluent Toxicity. *Chemical Engineering Journal*. 172 (1) 250-257.

Çeçen, F.; Çakiro, D. 2001. Impact of landfill leachate on the co-treatment of domestic wastewater. *Biotechnology Letters*, 23 (10) 821–826.

Çeçen, F., Aktas, Ö. 2004. Aerobic Co-Treatment of Landfill Leachate with Domestic Wastewater. *Environmental Engineering Science*. 21 (3) 303–312.

Cobos-Becerra, Y.L. and González-Martínez, S. 2014. Residence time distribution determination in a submerged filter with and without aeration using a tracer. *J Chem Technol Biotechnol*. 89 (2) 276-281

Ferraz, F.M., Povinelli, J., Pozzi, E., Vieira, E.M., Trofino, J.C., 2014. Co-treatment of landfill leachate and domestic wastewater using a submerged aerobic biofilter. *J. Environ. Manag*. 141, 9-15

Kjeldsen, P., Barlaz, M.A., Rooker, A.P., Baun, A., Ledin, A., Christensen, T.H., 2002. Present and long-term composition of MSW landfill leachate: a review. *Crit. Rev. Environ. Sci. Technol*. 32 (4) 297-336.

Maqbool, F., Bhatti, Z.A., Malik, A.H., Pervez, A., Mahmood, Q. 2011. Effect of landfill leachate on the stream water quality. *Int. J. Environ. Res.* 5 (2) 491-500.

Renou, S., Givaudan, J. G., Poulain, S., Dirassouyan, F., Moulin, P. 2008. Landfill Leachate Treatment: Review and Opportunity. *Journal of Hazardous Materials*. 150 (3) 468–93.

Qiu, L., Ma, J., Zhang, L. 2007. Characteristics and Utilization of Biologically Aerated Filter Backwashed Sludge. *Desalination*. 208 (1–3) 73–80.

Vanrolleghem, P. A. 2002. Principles of respirometry in activated sludge wastewater treatment. Universiteit Gent, Gent, Belgium.