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Application of a membrane sequencing batch reactor for landfill leachate treatment

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Abstract

A bench-scale membrane sequencing batch reactor (MSBR) was used for the treatment of "mature" landfill leachate, originated in the municipal solid waste landfill of Thessaloniki (North Greece). A hollow fiber ultrafiltration membrane module was used for the separation of the biomass from the treated effluent. The average quality of the raw leachate (feed) was 2456 mg/L COD, 375 mg/L total nitrogen (TN) and 8.2 mg/L PO_4-P. Hydraulic retention time (HRT) was kept constant at 10 days for most experimental runs whereas the solids retention time (SRT) was practically infinite, as almost no waste sludge removal took place during an overall MSBR operational period of 4 months. The initial concentration of suspended solids in the mixed liquor (MLSS) was 7000 mg/L and it was increased up to 15,300 mg/L. Several operational patterns were investigated in order to obtain maximum organic carbon and nutrients removal. Specific attention was given to the denitrification step. COD removal efficiency was as low as 40% and it was always below 60%. The poor MSBR performance in terms of COD removal was attributed to the high SRT, which had an apparent effect on the activity of the system's drastic biomass. On the other hand, TN removal efficiency was very satisfactory during the most stages of the MSBR operation, reaching a maximum value of 88%. Finally, PO_4-P removal efficiency varied between 35 and 45% in the first 50 days of the MSBR operation. The subsequent PO_4-P accumulation in the treated effluent was due to the direct addition of KH_2PO_4/K_2HPO_4 solutions in the feed leachate, aiming to improve the C:N:P ratio.

Keywords: Landfill leachate treatment; Membrane sequencing batch reactor (MSBR)

1. Introduction

Sanitary landfilling still remains an important method, used for the disposal of domestic wastes, especially among the Mediterranean countries. By definition, a landfill site is a large area of the ground, normally lined, that is used for tipping/ disposal of waste material. As long as the rainfall exceeds the rate of water evaporation, the liquid

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level (leachate) within the landfill area will tend to rise [1]. The chemical composition of leachate is dependent upon the age and maturity of the landfill site. In "fresh" leachate from young landfills (the acid-phase landfills), the concentrations of organic compounds (BOD_5 and COD) are very high, whereas in "mature" leachate from old landfills (the methanogenic-phase landfills), the levels of organic matter are substantially lower [2].

The correct site management can substantially reduce the amount and strength of the produced leachate, although it cannot eliminate it. The major environmental problem usually associated with landfill leachate production is the heavy contamination of nearby existing underground and surface waters [2]. Thereby, it is imperative that the generated leachate must be collected and efficiently treated, before being discharged back to the environment.

The treatment of landfill leachate includes both biological methods and physicochemical processes. The latter are considered by far more costly and of lower effectiveness and reliability. On the other hand, biological treatment schemes based on suspended-growth biomass, such as conventional activated sludge (CAS) processes, were proved to exhibit satisfactory and consistent performance in terms of organic carbon and nutrients removal, providing enough hydraulic residence time [3]. These traditional treatment schemes use an aerobic/biological tank followed by a settlement chamber for sludge separation.

An important variation of the CAS process, which is commonly applied for the treatment of leachates is the sequencing batch reactor (SBR). The main feature of SBR technology is that both biological oxidation and sludge separation are carried out in the same tank. Furthermore, its cycle operation can be easily modified at any time to offset possible changes in process conditions, influent characteristics, or effluent objectives. However, the SBR process presents certain drawbacks, such as the problem of inadequate sludge settling, which results in poor clarification and a turbid effluent [4].

The aforementioned disadvantages can be overcome by using a membrane-coupled sequencing batch reactor (MSBR). Sludge separation through membrane filtration leads to complete biomass retention and the development of high suspended solids concentrations in the mixed liquor. Typical MLSS concentrations in MSBRs are in the range of 15,000–20,000 mg/L, though some authors suggest that MLSS should not exceed the value of 12,000 mg/L. Another key feature of these systems is the complete decoupling of sludge age (SRT) and hydraulic residence time (HRT), which in combination with the high MLSS content can result in almost an order of magnitude intensification of the biological process, which is directly translated into a substantial reduction of the reactor volume [5].

The basic disadvantage of membrane bioreactors, including MSBRs, is the high capital and operating cost, which is basically determined by the cost of the membrane itself. Membrane costs are approximately proportional to plant size as opposed to traditional plants, which show a scale-economy. Additionally, common to all membrane systems, is the inevitable fouling of membranes that rather often takes place. This limits the maximum achievable permeate flux and leads to substantial cleaning requirements and, in some cases, even to membrane replacement [5].

The aim of the present study was to assess the performance of a bench-scale MSBR fed with "mature" landfill leachate, containing rather recalcitrant organic compounds, not easily amenable to biological treatment, as well as relatively high levels of ammonium nitrogen. Several operational patterns were tested in order to obtain maximum COD and total nitrogen removal. Specific attention was given to the denitrification step. Finally, the gradual development of transmembrane pressure (TMP) was recorded and correlated with the system's overall operational parameters.

2. Materials and methods

2.1. Leachate feed

Several sampling events were performed within an overall period of 4 months. Leachate samples were received from the main municipal landfill site of Thessaloniki metropolitan area (North Greece), which accepts 1000–1200 t/day of municipal solid wastes. The produced leachate was collected in an artificial pond, located at the lowest side of the landfill [3].

The main physicochemical characteristics of the landfill leachate used in this study are shown in Table 1. The high pH values and the low COD concentrations clearly indicate a "mature" (aged) landfill leachate, which is (partially) stabilized in the open pond.

2.2. Experimental setup

The experimental system consisted of a completely mixed bioreactor in which a hollow fiber ultra-filtration membrane module was directly submerged. The bioreactor was made of a cylindrical Plexiglas vessel (14 × 50 cm), with a working volume of 5 L. Raw leachate was fed to the MSBR with a small variable speed peristaltic pump (SEKO PR1). A similar peristaltic pump was used for the addition of methanol solution in the reactor during the denitrification periods. The treated leachate/permeate was removed by means of a variable flow suction pump (WATSON MARLOW 503U) connected to the top header of the membrane module. The TMP values were read on the screen of an analogue manometer (Kindmen), located at the permeate line. A simplified flow sheet of the experimental MSBR unit is shown in Fig. 1.

The membrane used in this study was the ZW-1 bench-scale module, manufactured by Zenon Environmental Inc. Its main characteristics are presented in Table 2. The ZW-1 module is equipped with a central aeration tube, which was directly connected to the air transmission line. This tube supplied air close to the bottom header of the module, where orifices are located. The air introduced in the MSBR was used for both scouring the accumulated material away from the membrane surface, as well as for providing the biomass with the necessary oxygen concentration. It was also used for complementary mixing of the mixed liquor. Mixing of the MSBR content during the aerobic and anoxic periods was obtained by means of magnetic stirrer and flat-blade impeller.

Table 1
Physicochemical characteristics of "mature" landfill leachate

Parameter	Minimum value	Maximum value	Mean value[a]
pH	8.3	8.8	8.4 (36)
Alkalinity, mg/L $CaCO_3$	1474	2848	2014 (33)
Turbidity, NTU	98	154	103 (20)
COD, mg/L	1391	3977	2456 (21)
TN, mg/L	310	509	375 (14)
NH_4-N, mg/L	207	279	238 (13)
NO_3-N, mg/L	7.7	25.4	16.5 (13)
PO_4-P, mg/L	5.2	13.7	8.2 (15)

[a]Numbers in parentheses refer to total number of measurements.

2.3. Analytical methods

For the evaluation of the MSBR performance samples were collected from the mixed liquor, the raw leachate and the treated effluent/permeate. Mixed liquor samples were analyzed for suspended solids (MLSS, MLVSS and MLNVSS), dissolved oxygen (DO) and pH in accordance to standard methods [6]. Influent and effluent samples were photometrically tested for COD, TN, NH_4-N, NO_3-N and PO_4-P, according to standard measurement procedures (Lasa 100, Dr. Lange). The latter samples were in addition analyzed for alkalinity, turbidity and pH, according to standard methods [6].

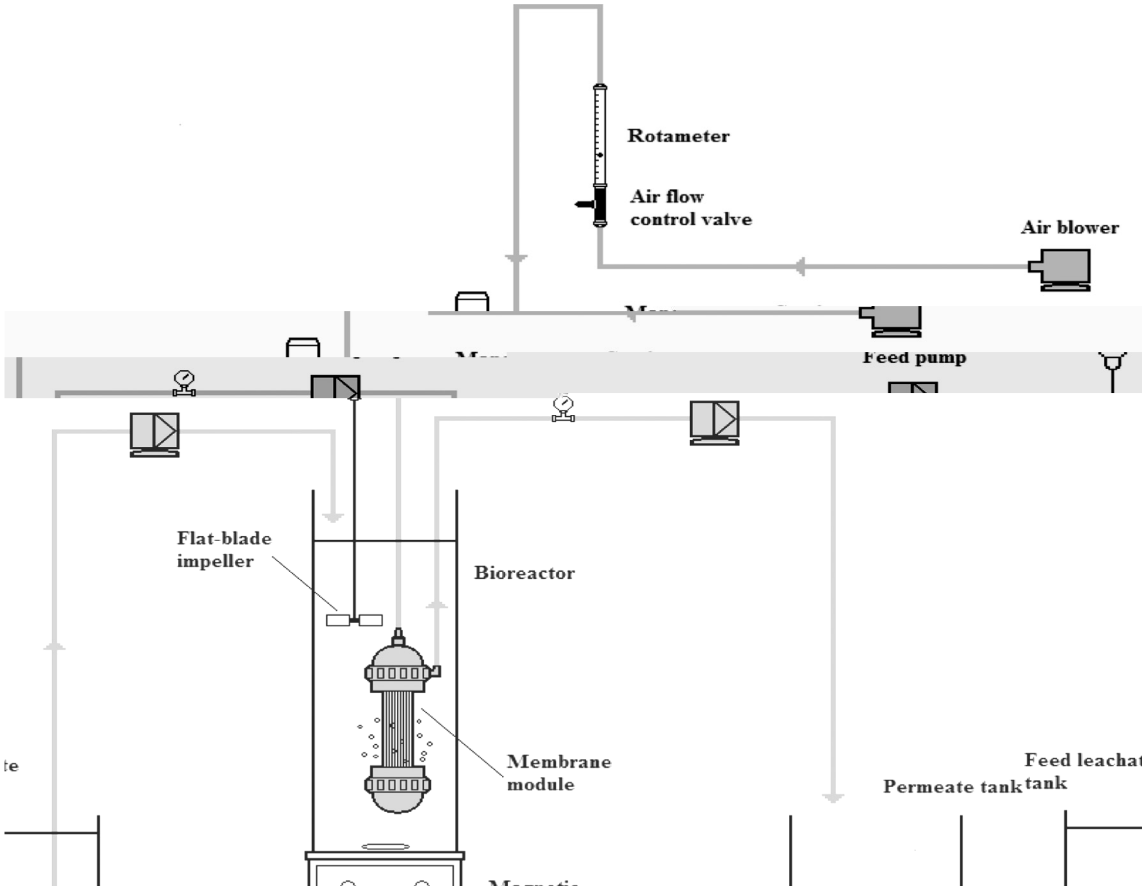

Fig. 1. Schematic diagram of the experimental MSBR unit.

3. Results and discussion

3.1. System start-up and operational patterns

Prior to the landfill leachate treatment runs, the MSBR unit was fed for over 3 months with synthetic wastewater of gradually increasing strength in terms of COD and TN concentrations (COD varied from 465 mg/L up to 4800 mg/L, whereas TN varied from 65 mg/L up to 570 mg/L), in order to examine the effect of influent characteristics on the operation performance of the unit. The bioreactor was initially inoculated by the addition of sufficient quantity of activated sludge, collected from the recirculation channel of the conventional biological municipal wastewater treatment plant of Thessaloniki (located in the nearby city of Sindos). After the completion of these experiments, the MSBR content was gradually replaced with landfill leachate. The replacement period lasted for about 20 days, aiming to the acclimatization of the system's biomass to the new influent characteristics.

The subsequent MSBR operation with landfill leachate influent lasted for 4 months. MSBRs are designed to operate under non-steady state flow conditions, similar to SBRs. The inherent flexibility degree of MSBR systems is due to the fact that these systems work in a time, rather than in a space sequence [7]. Several operational patterns/cycles were examined during the

Table 2
Characteristics of the membrane module used in the MSBR unit

Model	ZW-1, submersible ultra-filtration membrane
Configuration	Hollow fibers, outside-in filtration
Material	PVDF
Nominal pore size	0.04 µm
Absolute rejection	0.1 µm
Nominal surface area	0.047 m^2
Maximum allowable TMP	62 kPa (0.62 bar)
Typical operating TMP range	10–50 kPa (0.10–0.50 bar)
Operating pH range	5–9
Maximum OCl^- exposure	1000 mg/L

4-month experimentation period. All examined patterns consisted of four steps in common, which were achieved by programmable time-switches in sequence as follows: (1) fill, (2) react (alternation of aerobic and anoxic periods), (3) draw/removal of effluent (membrane filtration), and (4) idle. The basic characteristics of two representative patterns are shown in Tables 3 and 4, respectively. During the fill step, 250 mL of raw leachate was fed to the MSBR. The same volume of treated leachate/permeate was produced by membrane filtration during the draw step. The HRT was kept constant at 10 days for most experiments and so did the total cycle time, which lasted for 12 h (2 cycles/day).

3.2. Suspended solids

The variation of total (MLSS), volatile (MLVSS) and non-volatile (MLNVSS) suspended solids of the mixed liquor during the overall MSBR operational period is presented in Fig. 2. The initial MLSS concentration was 7000 mg/L and the fraction of volatile solids (active biomass) was 77%.

As shown in Fig. 2, during the initial 2 months of the MSBR operation, the MLSS content varied between 6000 and 10,000 mg/L (fraction of volatile solids between 55 and 75%). In order to increase the biomass concentration, certain sludge volumes, obtained from a pilot-scale MSBR treating a 1:1 mixture of high-strength synthetic wastewater and landfill leachate, were added directly to the bench-scale MSBR. Sludge addition was performed on selected days over an operational period of 40 days (shown as steep biomass increases in Fig. 2). As a consequence, the MLSS content increased up to 15,300 mg/L (fraction of volatile solids 76%) at the end of 100th day of the MSBR operation. No sludge wastage took place during the whole operational period, thereby resulting in a high (i.e. almost infinite) SRT.

Table 3
Characteristics of a 12-h operational cycle with single aerobic and anoxic period (applied with several variations from day 1 to day 84)

Operational step	Mixing	Aeration	Time (h)	% of cycle time
Fill	Yes	No	1.25	10.4
React-aerobic	Yes	Yes	7.00 (6.00)	58.3 (50.0)
React-anoxic	Yes	No	1.00 (2.00)	8.3 (16.6)
Draw	Yes	Yes	2.42	20.2
Idle	Yes	No	0.33	2.8
Total cycle time			12.00	100.0

Table 4
Characteristics of a 12-h operational cycle with alternation of aerobic and anoxic periods (applied with several variations from day 85 to day 120)

Operational step	Mixing	Aeration	Time (h)	% of cycle time
Fill	Yes	No	1.25	10.4
React-aerobic 1	Yes	Yes	3.00	25.0
React-anoxic 1	Yes	No	1.00	8.3
React-aerobic 2	Yes	Yes	3.00	25.0
React-anoxic 2	Yes	No	1.00	8.3
React-aerobic 3	Yes	Yes	1.00	8.3
Draw	Yes	Yes	1.33	11.1
Idle	Yes	No	0.42	3.5
Total cycle time			12.00	99.9

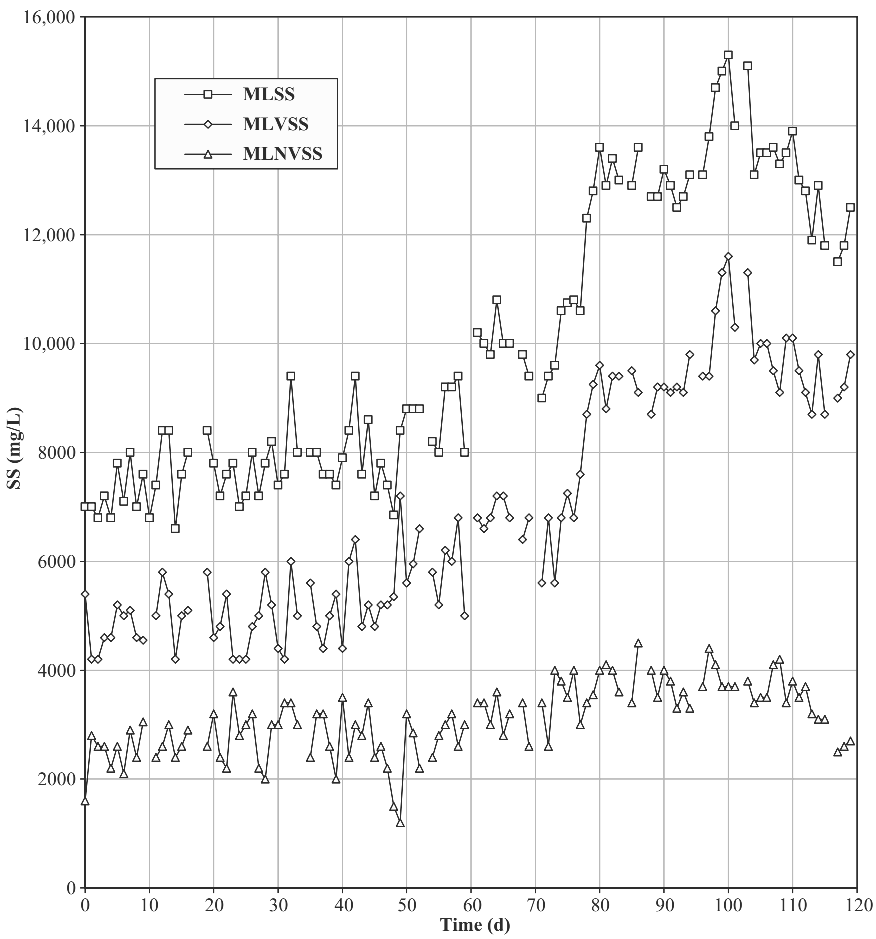

Fig. 2. Suspended solids variations during the MSBR operation.

Finally, during the last 20 days of the MSBR operation, 200 mL of mixed liquor were removed from the bioreactor every other day, aiming to decrease the biomass concentration, as it was found that the system performance was not improved at such high MLSS content. As a result of sludge removal, the MLSS concentration decreased to 12,500 mg/L (fraction of volatile solids: 78%) at the end of the 4-month experimentation period.

3.3. Organic compounds and nutrients removal

The corresponding treatment results in terms of COD, TN, NH_4-N, NO_3-N and PO_4-P concentrations of both influent and treated effluent are shown in Figs. 3–7 respectively.

According to the experimental data presented in Fig. 3, COD removal efficiencies varied between 40 and 60% and were not affected by the applied biomass concentration and by the operational patterns. In addition to the increase of the reactor's MLSS content, certain other measures were taken in order to substantially improve the biological oxidation of organic compounds. Such measures included the direct addition of pure methanol, as well as K_2HPO_4 or KH_2PO_4 in the raw leachate in order to optimize the C:N:P ratio. However, these measures proved to be insufficient for the enhancement of COD removal rate. The apparent inability of the MSBR to obtain higher COD removal efficiencies can be attributed to the low biodegradability of certain leachate compounds (e.g. humic or fulvic acids), as well as to the high sludge age. The practically infinite SRT probably

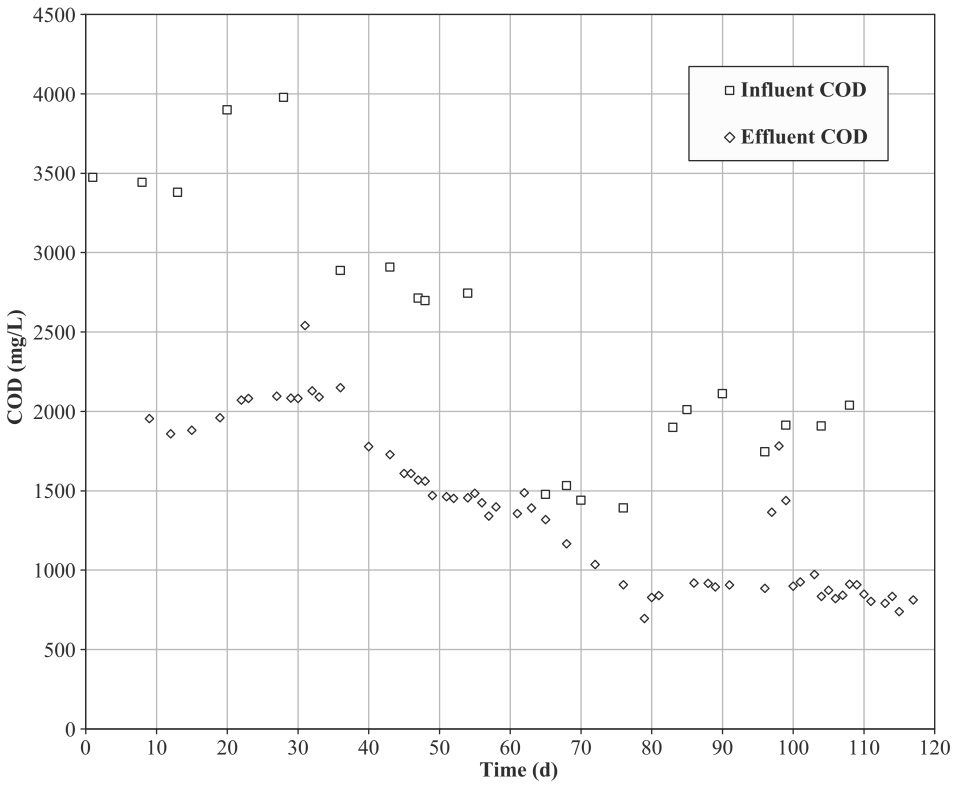

Fig. 3. Influent and effluent COD concentrations during the MSBR operation.

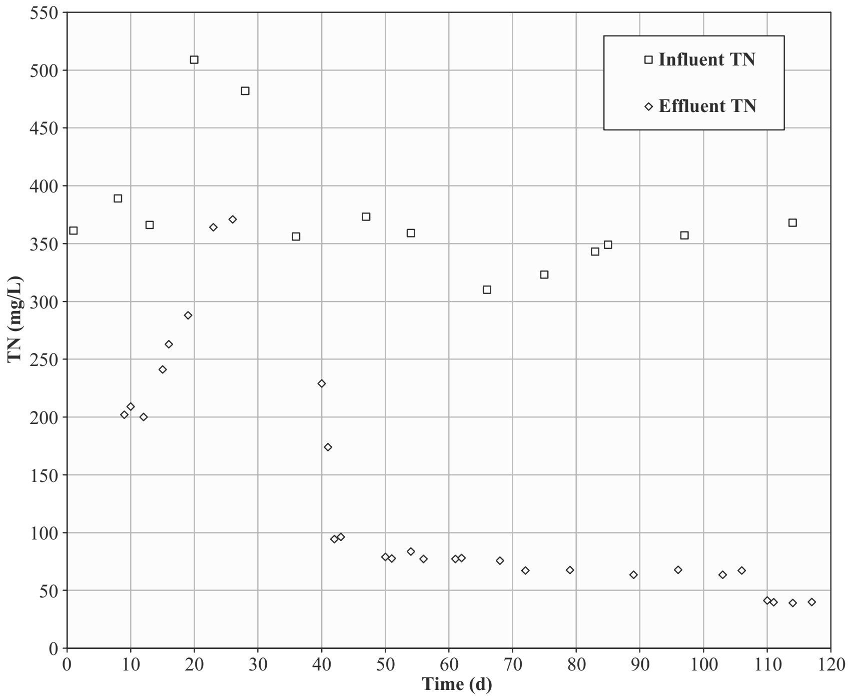

Fig. 4. Influent and effluent TN concentrations during the MSBR operation.

lead to gradual approach to the system's bioxidation capacity. Therefore, the selection of an appropriate SRT value by the appropriate sludge removal is important, in order to maintain the activity of drastic biomass population.

According to the experimental data presented in Fig. 4, high total nitrogen (TN) concentrations were observed during the 1st month of the MSBR operation, possibly due to the nitrogen accumulation in the mixed liquor. This accumulation was attributed probably to the inability of the system's denitrifying bacteria to further utilize the residual COD in the reactor after the aerobic period. This was further verified by the high effluent NO_3-N concentrations (Fig. 6), which showed a similar behavior to that depicted in Fig. 4, whereas the corresponding NH_4-N concentrations (Fig. 5) were negligible. In order to enhance the system denitrifying capacity, 2 mL of pure methanol were directly added in the feed leachate and were readily consumed by the denitrifying microbial population during the anoxic reaction step (see the Table 3 operational cycle). Between 50th and 105th days of the MSBR operation, TN removal efficiency was increased to about 78% (TN < 80 mg/L, NO_3-N < 20 mg/L). During the subsequent days, TN removal efficiency was further increased up to 88% (TN < 50 mg/L, NO_3-N < 10 mg/L), by adding appropriate volumes of a 1:5 methanol solution at the beginning of each anoxic reaction step by a peristaltic pump (Table 4 operational cycle). The effluent NH_4-N concentrations were very low during the whole MSBR operation, indicating the system's excellent nitrifying capacity (almost 100% removal of influent NH_4-N concentrations).

According to the experimental data presented in Fig. 7, PO_4-P removal efficiencies varied between 35 and 45% during the first 50 days of the MSBR operation. The gradually increasing

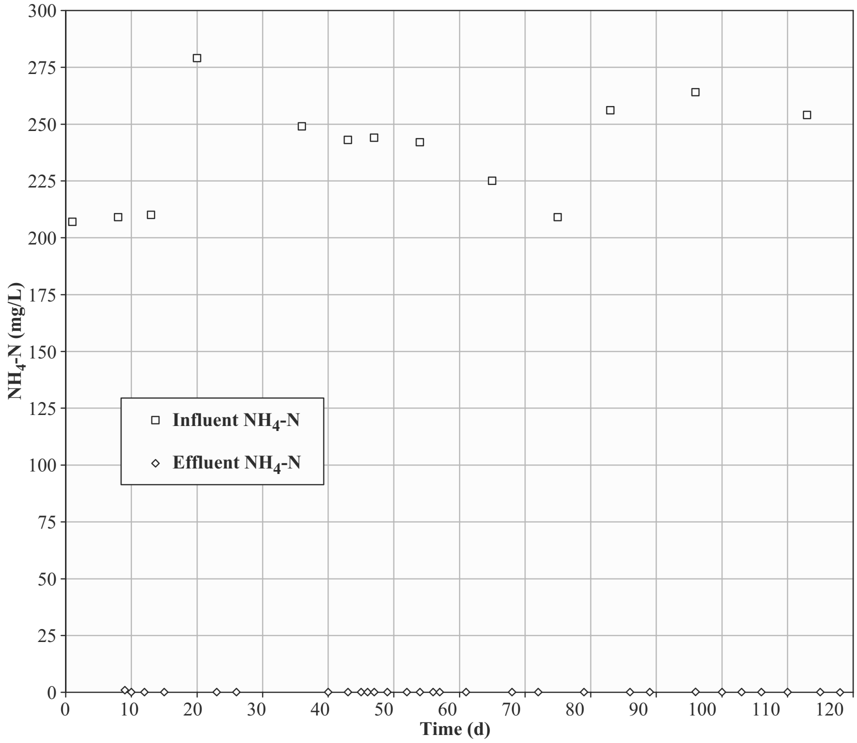

Fig. 5. Influent and effluent NH_4-N concentrations during the MSBR operation.

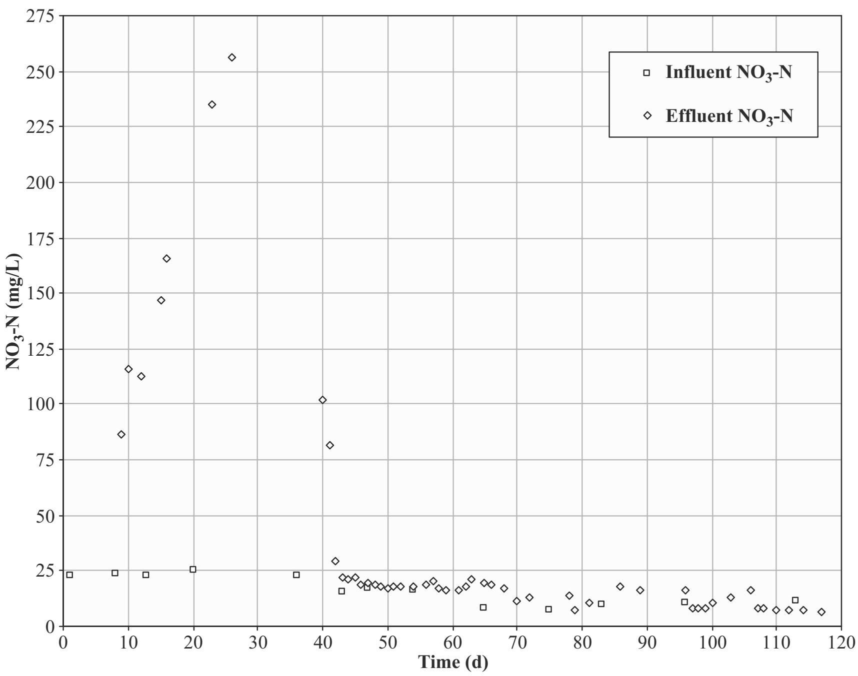

Fig. 6. Influent and effluent NO_3-N concentrations during the MSBR operation.

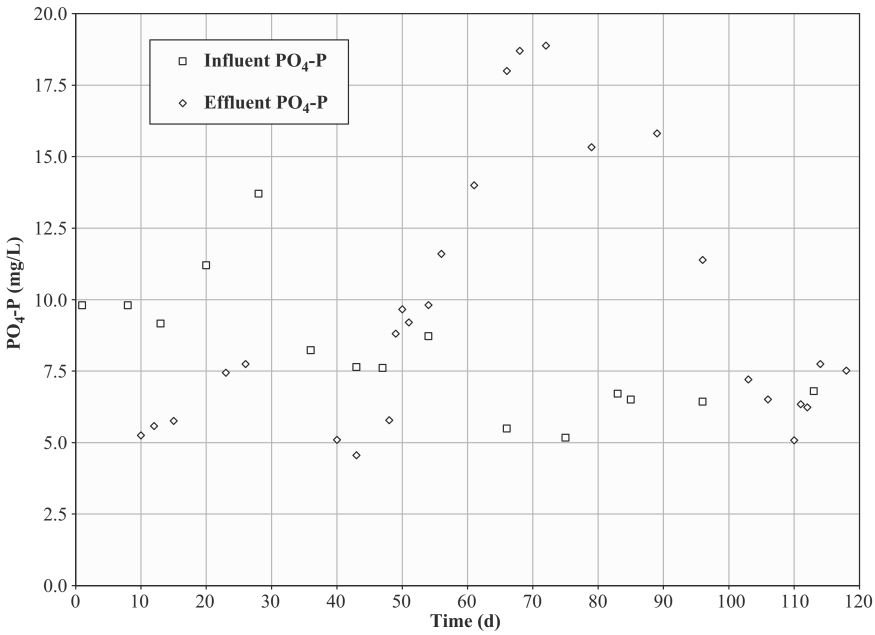

Fig. 7. Influent and effluent PO_4-P concentrations during the MSBR operation.

effluent PO_4-P concentrations during the next 20 days were due to the daily addition of 0.080 g of KH_2PO_4 in the feed leachate in order to improve the C:N:P ratio. After the addition of KH_2PO_4 was stopped, the effluent PO_4-P concentrations gradually returned to lower values. Greater PO_4-P assimilation in the activated sludge could be achieved in combination to the simultaneous bioxidation/utilization of the recalcitrant influent COD. Due to its high SRT, the system's biomass contributed to the effluent PO_4-P concentrations through lysis phenomena.

4. Conclusions

MSBR technology is a cost-effective method for landfill leachate treatment. By combining intense biological oxidation with complete rejection of suspended solids, MSBRs can produce a treated effluent with desired characteristics. A bench-scale MSBR reactor was examined in this work for the treatment of a high strength landfill leachate, under various operational stages, using an MLSS concentration varied between 7000 and 15,000 mg/L.

Low COD removal rate was observed, ranging from 40 to 60% and that was attributed to the high SRT values; therefore, the frequent sludge removal is a key parameter for preserving the activity of the drastic biomass. However, a high nitrification and denitrification capacity was achieved, resulting in an effluent with negligible ammonia–nitrogen concentrations and low nitrate–nitrogen content. Nevertheless, phosphorous removal reached up to 45% and was affected by the addition of KH_2PO_4/K_2HPO_4 solutions in the influent, aiming to improve the C:N:P ratio.

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