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Post-treatment of anaerobically-treated compost leachate by membrane systems: emphasis on molecular weight distribution

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ABSTRACT

Compost leachate contains high concentrations of organic matter, sulphate and ammonia which requires combined treatment systems. In case of the use of membrane containing combined systems, the effect of pretreatment on molecular weight distribution (MWD) is important in terms of appropriate membrane selection. In this study the leachate from Istanbul full-scale composting plant was firstly treated in an anaerobic fluidized bed reactor (AFBR). Performance of the reactor was low due to the inhibiton by high ammonia content while treatment efficiencies of COD and SO_4^{2-} were around 41% with 50% ammonia removal. During the anaerobic treatment high molecular weight materials were mostly converted to low molecular fractions. However, changes in the distribution of molecular fractions differed in each pollutant parameters. Subsequent membrane treatment scheme was determined according to the molecular weight distribution analyses. Particular and collodial materials from AFBR effluent was effectively treated by MF and UF membranes. Post-treatment studies were performed using four different NF and RO membranes and performance comparison was made based on removal efficiency and flux changes. BW30 membrane provided the lowest treatment efficiency while other TXN45, NF90 and XLE membranes had similar effluent quality. Effluent from all membrane systems met discharge limits and optimum treatment scheme has been suggested as AFBR+MF+UF+TXN45 based on operational flux values.

Keywords: Compost leachate; AFBR; Molecular weight distribution; NF; RO membrane

1. Introduction

In recent decades, composting has gained significiant interest for the disposal of municipal solid waste (MSW) with high organic matter content. Comparison to other disposal methods, MSW composting has advantages of higher organic stabilization, minimum waste amount to be landfilled, pathogen destruction, production of agricultural additives and less emission of greenhouse gases [1]. During the composting, heterogeneous organic materials are converted to more homogenous and humus-like matters under controlled biological processes. Although composting provides many benefits, high-efficient and cost-effective treatment of leachate remains the main challenge in sustainable MSW management. One of the main problems associated with the treatment of

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compost leachate is the variability in the quantity and chemistry. Amount and characteristics of compost leachate are influenced by some factors such as the composting technology employed, composition of MSW, maturity of compost, the degree of cover and climatic conditions [2,3]. Compost leachate may contain high concentration of dissolved and suspended matter, organics, phenol, heavy metals, ammonia, phosphorous and other contaminants [4–6]. If the leachate is discharged without proper treatment, many harmful affects will arise in aquatic environments including the depletion of dissolved oxygen by the biodegradation of organic matters and eutrophication by rich nutrients. Although treatment of landfill leachate has been extensively documented, treatibility of compost leachate has been rarely studied.

Researchers have applied some physical, chemical and biological treatment technologies for the removal of pollutants from compost leachate such as adsorption [7], fenton process [8] and membrane bioreactor [9]. Anaerobic treatment technologies have been successfully applied for the treatment of high-strength industrial wastewaters along with sanitary landfill and compost leachates [10–13]. Up to now, various anaerobic bioreactors have been proven for efficient removal of organics and energy production in the form of methane. Anaerobic fluidized bed reactor (AFBR) can be operated at high organic loading rates and short hydraulic retention times and provide considerable treatment efficiencies along with better mass transfer, high amount of attached biomas and long-term stabile operation [14]. Moreover, researchers reported that ultrafiltration and nanofiltration membrane systems are feasible for the removal of organics, heavy metals and other contaminants from leachates [2,15,16]. Although separately applications of anerobic treatment and membrane separation are efficient for the removal of pollutants, combination of those systems with other treatment technologies have been recommended to enhance effluent quality and meet discharge limits [17,18]. Furthermore, promising treatment performances have been documented when membrane processes are applied as polishing step for the effluents from various reactors [19,20].

It the evaluation of treatment alternatives and monitoring the pollution removal, conventional concentration measurements is inadequate and molecular weight distribution (MWD) provides a deep analysis [21–23]. MWD analysis has been widely used in membrane studies to characterize pollutant fractions and to evaluate the changes in pollutants after treatment [24,25]. Researchers reported that raw leachate contains molecules with a wide range of molecular weights, from 40,000 Da to <100 Da while 37% of the COD in landfill leachate consists particulate or colloidal (>100 kDa) and 63% is soluble (<100 kDa) [26,16]. Trebouet et al.[27] found that majority of organics in stabilized leachate is lower than 1000 Da. In this study combined treatment of AFBR and membrane filtration have applied on compost leachate and changes in the fractions of the pollutants were determined by molecular weight distribution analyses.

2. Experimental

2.1. Composting leachate

Istanbul full-scale composting plant has been operated since 2001 for compost production from organic content of mixed MSW. The plant is the second biggest facility in Europe with its production capacity and it receives 15000 tons of mixed MSW on daily basis. The composting technologies and process properties have been well documented elsewhere [1,28]. Fresh and stabilized leachate are collected together in a tank and sent to the leachate treatment facility in Odayeri Sanitary Landfill Area [29]. Leachate samples were taken from the tank once a week and kept at 4°C until use in lab-scale experiments.

2.2. Anaerobic treatment set-up

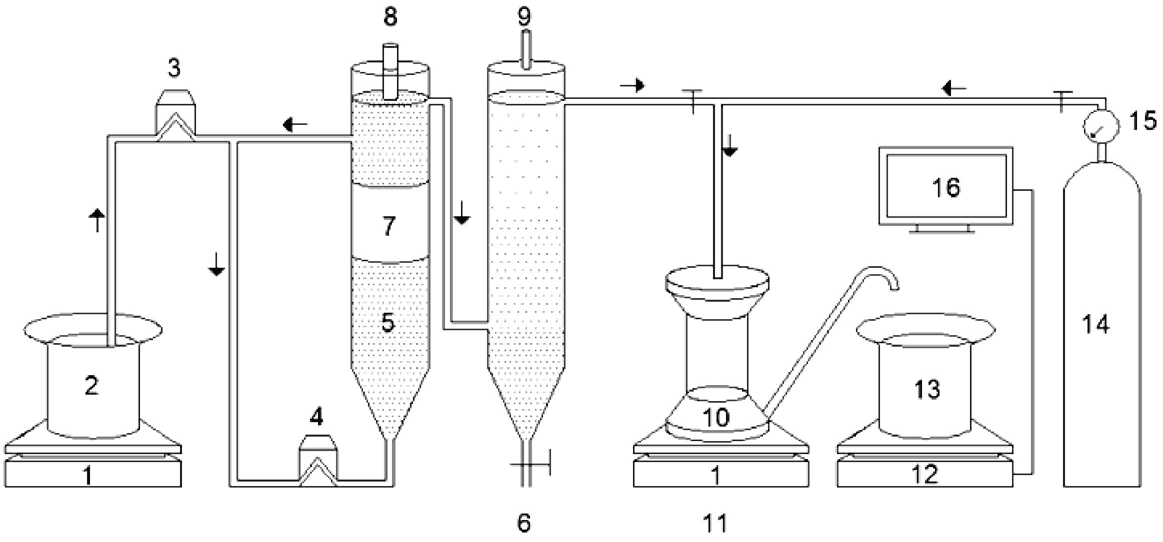
The schematic diagram of combined treatment system is given in Fig. 1. Anaerobic treatment experiments were performed in a fluidized bed reactor (AFBR). Fine particle pumice (NMP 16) was used as supporting material which was placed at the bottom of the reactor and it was fluidized at 100%. The total volume of the AFBR was 670 mL. The effluent has been recycled in order to prevent sedimentation and to accelerate the granulation of the sludge. Recycling helped the pumice forms a fluidized bed. The gas generated in the reactor was collected by the gas collection system located at the upper part of the outlet channel. The reactor was operated at the temperature of 35± 2°C using an electrical heating blanket. Reactor was operated at constant hydraulic retention time (HRT) of 5 din fluidized bed and volume of biogas collection chamber was not included. Raw leachate was stirred with a magnetic stirrer before feeding into AFBR. During the operation no sludge was removed or recycled and AFBR was operated at infinity SRT. The pH of compost leachate in the reactor was kept in the range of 6.5 and 7.5 using H_2SO_4 and NaOH.

Development of microbial community and acclimation were provided by feeding synthetic wastewater. The inoculum was anaerobic granular sludge from a full-scale UASB reactor treating wastewater from food industry at 37°C. Total solid content of inoculum was 326.6 g/L and 36% of total solid was in the form of volatile solid [30].

COD concentration of synthetic wastewater was gradually increased as 5000 mg/L and 8000 mg/L for easily adaptation of anaerobic microorganisms. Following 28 d of operation, AFBR was fed by the mixed of synthetic and raw leachate at the ratio of 2:3 based on COD content (10000 mg/L) for 14 d. The changes in COD content at start-up period was as shown in Fig. 2. Subsequent experiments were performed with real raw compost leachate only.

2.3. Molecular weight distribution with membrane

Molecular weight distributions of pollutants were determined by the filtration of anaerobically treated effluent through the membranes with nominal molecular weight between 0.1 kDa and 0.22 µm. The flow diagram of filtration procedures was as shown in Fig. 3. Experiments were performed with a stirred cell (Amicon Model 8400) in a methyl-methacrylate glass holder having a volume of 350 mL. Effective filtration area and membrane filter diameter were 41.8 cm^2 and 76 mm, respectively. Filtrations were performed in cross-flow mode under nitrogen gas pressure of 7 bar except for the 100 kDa filtration of 3 bar. The filters were firstly washed with ethanol 70% for 20 min then washed



1	Mixer	5	AFBR	9	Biogas collection	13	Permeate
2	Raw leachate	6	Sludge removal	10	Membrane bed	14	N tube
3	Feed lump	7	Heating	11	Filtration apparatus	15	Pressure gauge
4	Recirc pump	8	Thermometer	12	Weighting	16	Computer

Fig. 1. Schematic diagram of experimental setup.

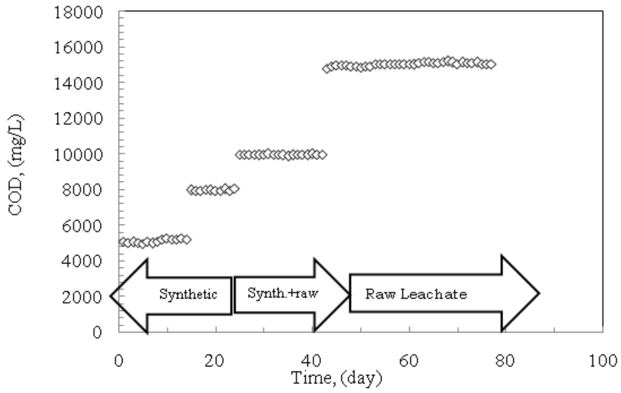


Fig. 2. COD changes in feed solution during start-up period.

with deionized water to remove the ethanol. All filtration experiments were conducted by using 100 ml of samples in offline mode.

2.4. Membrane treatment set-up

Effluent from AFBR was first filtrated by MF (0.45 µm) and UF (UP150) membranes. Suspended solids were removed by MF while UF membrane was used to remove high molecular weight colloids to prevent pore blocking of the subsequent membranes. NF (TSXN45, NF90) and RO (XLE, BW30) membranes were separately operated for the final treatment of leachate. The specifications of membranes used in the experiments are given in Table 1. Membranes

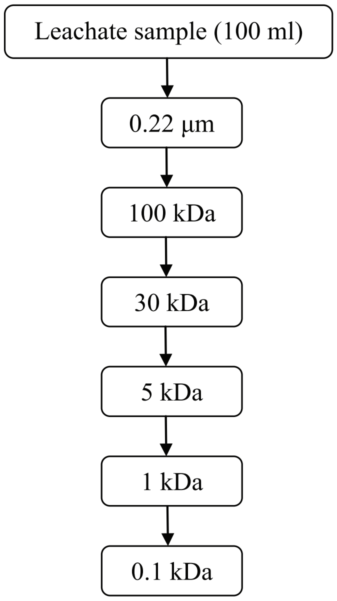


Fig. 3. Procedures of particle size distribution analysis.

were compared in terms of flux behavior and removal performance. Membrane experiments were performed using a stirred membrane cell (Amicon, Model 8400) with the diameter of 76 mm and effective membrane area of 41.8 cm^2. It consists of a methyl-methacrylate glass holder with a volume of 350 mL. The operational pressure was 7 bar for XLE, BW30, TSXN45 and NF90 membranes, while operational

Table 1
Membrane specifications

Membrane	UP150	TS XN45	NF90	XLE	BW$_{30}$
Initiative	Low press.	TRİSEP XN$_{45}$	NF90	ELE*	B.W. (LE)**
Class	UF	NF	NF	RO	RO
Type	PES***	PA TFC***	PA TFC***	PA TFC***	PA TFC***
Max. temp (°C)	95	–	45	45	45
Max. pres (Bar)	–	41.1	41	41	41
pH range	1–14	4–11	2–11	2–11	2–11

*Extra low energy, **Brackish water (Low energy), ***Polyethersulfone ***Polyamide thin film composite

pressure of 3 bar were performed for the UP 150 filtration. Membrane cell was placed on a magnetic stirrer to maintain cross-flow filtration beside hinder the settlement. Two different pumps were used in the system for feeding and recirculation. Recirculation pump was operated continuously (24 h/d; 7 d/wk). Feed pump was operated at 45 min/d to feed the batch reactor and controlled by a time clock. Influent and effluent samples were analyzed for COD, BOD$_5$, NH$_4^+$, SO$_4^{2-}$ and UV$_{254}$ to evaluate the removal performance of NF and RO membranes. Filtered water was weighed and the data was collected using a personal computer to calculate flux. Weight was recorded at 60 s intervals by computer to observe of flux variation during filtration.

Table 2
Composition of raw leachate

Parameter	Value		
COD (mg/L)	15 025	±	100
Dissolved COD (mg/L)	11 000	±	100
BOD$_5$ (mg/L)	3 600	±	60
pH	8.5	±	0.5
ORP (mV)	–110.5	±	10
Ammonia (mg/L)	1 577	±	30
Total solids (mg/L)	17 700	±	100
Volatile solids (mg/L)	5 200	±	100
Phosphorus (mg/L)	4.10	±	0,10
Sulfate (mg/L)	3 300	±	25
Alkalinity (mg CaCO$_3$/L)	3 000	±	100
UV$_{254}$	191	±	5

2.5. Chemical analyses

All analyses were performed by following the procedures in Standard Methods [31]. Changes in organic matters were characterized by the analysis of chemical oxygen demand (COD) and 5-d biochemical oxygen demand (BOD$_5$). COD was determined by the method of the closed reflux colorimetric method (5220D) while BOD$_5$ tests were performed according to the method of SM-5210B. Dissolved COD was determined by the separation of samples through a 0.05 µm pore-sized filter while the filtrated organics was defined dissolved matters. Ammonia was analyzed by using a thermal digester (Velp Scientifica). UV$_{254}$ was measured by a double-beam UV-VIS spectrophotometer (Shimadzu UV-1800) with 1 nm of resolution. pH and ORP were measured using a multimeter (Thermo Orion 5 star). All the analyses were done in triplicate by using analytical grade chemicals.

3. Results and discussion

3.1. Leachate characteristics

The characteristics of the raw compost leachate used in experiments were as given in Table 2. Compared to previously reported compost leachate characteristics, COD and solid concentrations are comparatively low while ammonia and pH level are significantly higher [13,32–34]. The variations were probably due to the difference in organic matter content and composting technology. BOD$_5$/COD ratio was 0.24 which indicates that biodegradability of leachate organics is lower compared to fresh leachates. Especially in fresh leachates, acidic pH is related with the high quantity of volatile fatty acids thus those result also elevated COD levels. The low COD and high pH in this study indicates that most of volatile fatty acids were converted to final products. NH$_3$-N concentration was high as 1300 mg/L and this elevated ammonia level could be associated with the complete degradation amount of protein organics. Total NH$_3$-N is sum of free (unionized) and ionized ammonia while free ammonia level increases at elevated pH. Researchers indicated that inhibition of methanogens in anaerobic reactors is due to the mainly by free ammonia [35]. In the study of Rajagopal et al. (2013), they reported that free ammonia level increases eight fold when the pH changes from 7 to 8. According to pH level of raw leachate, it can be concluded that most of the ammonia was in the form of free ammonia [36]. On the one hand, Liu et al. [32] stated that leachates with NH$_3$-N less than 1500 mg/l could be efficiently treated in anaerobical systems.

3.2. AFBR performance

Mean removal efficiencies of AFBR were as given in Table 3. Organic matter treatment efficiencies were low with 50% BOD$_5$ and 41% COD removal levels. The highest treatment efficiency was 58% for ammonia and removals of sulfate and UV$_{254}$ were around 30%. Low COD removal by AFBR could be attributed to the high concentration of ammonia and sulfate in compost leachate. At operational pH value of AFBR, free ammonia level is high and causes toxic effect on methane producing bacteria (MPB). In anaerobical conditions, sulfate reduction is accomplished by sulfate-reducing bacteria (SRB) and they compete with MPB for the consuming of organics [37]. The competition is mainly dependent on both SO_4^{2-} concentration and COD/SO_4^{2-} ratio. Studies on sulfate reduction by different carbon sources have revealed that SRB has low contribution on COD removal when the COD/SO_4^{2-} ratio in wastewater was more than 2 [38,39]. Furthermore, Dinkel et al. (2010) indicated that sulfate reduction is inhibited at ammonia levels over 840 mg/L and their contribution on COD removal was at low level [40].

In this study, COD/SO_4^{2-} ratio was 4.3 and the ammonia concentration was two fold higher than inhibition level. On the other hand, it is known well that 1 mol (96 g) SO_4^{-2} reduction consumes 2 mol COD (64 g) under anaerobical conditions [41]. In AFBR, reduction of 1050 mg/L SO_4^{-2} equals to 700 mg COD/L which indicates that about 17% of total COD was consumed by SRB. Therefore, high amount of COD was used for CH_4 production in AFBR. Additionally, incomplete removal of sulphate was assumed due to the high ammonia concentration.

3.3. Changes in molecular weight distribution

3.3.1. Organics

Molecular weight distributions of raw and anaerobically-treated COD are shown in Fig. 4. In raw leachate, organics having MW > 0.22 µm were dominant while 44.2% of the COD was as in the form of particulate or colloidal and the remaining was in soluble form (<100 kDa). During anaerobic treatment, high molecular weight fractions of COD and BOD_5 were converted to low molecular fractions. In terms of COD, percentage of particles with MW < 100 kDa increased to 83% after the anaerobic treatment. Similarly, Zhao et al. [42] reported reduction in MW less than 100 kDa fraction of leachate after activated sludge treatment. It is thought that MW of 5kDa was the crucial point for COD parameter in present study. Particles with MW less than 5 kDa were 31.8% in raw water and it is increased to 73.6% after the anaerobic treatment. On the other hand, fractions higher than 5 kD decreased from 68.2% to 26.12% after the anaerobic treatment. Most of the reduction was obtained for the 5 kDa–100 kDa fraction and the >0.22 mm fraction. In contrast, all BOD_5 fractions decreased except 100 kDa–0.22 µm which was slightly increased from 7% to 23%. In comparison to raw leachate, COD fraction less than 0.1 kDa in treated water increased while same fraction of BOD decreased. The increase in COD was associated with the degradation of higher organic molecules into smaller fractions whereas reduction in BOD was due to consumption in anaerobic conditions. Similar changes in organic fractions during anaerobic treatment was reported by other researchers [23,43]

Table 3
Concentrations of influent and effluent leachate of AFBR

Parameter	Influent (mg/L)	Effluent (mg/L)	Removal (%)
COD	15 025±100	8 900±50	31
BOD_5	3 600±60	1 800±35	50
SO_4^{-2}	3 300±25	2 250±15	32
NH_4^+	1 577±30	653±10	58
UV_{254}	191±4	126±2	34

3.3.2. Ammonium, sulphate and UV_{254}

A significant fraction of the ammonium was <0.1 kDa in raw compost leachate, which is different from the COD fractionation. It is shown in Fig. 5 that all ammonium fractions decreased but high molecular weight fraction (100 kDa–0.22 µm) slightly increased. These results indicated that high molecular ammonium particulates converted to low molecular weight fractions which then were removed by anaerobic processes. In the case of SO_4^{2-}, low and high molecular weight fractions were almost equally distributed. Percentages of the whole of

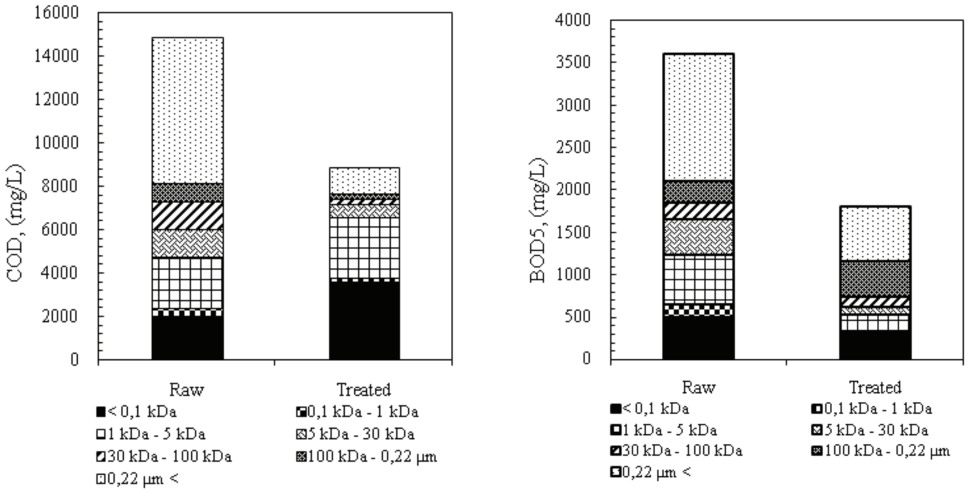


Fig 4. Molecular fractions of COD and BOD.

the SO_4^{2-} fractions decreased, while molecular weights higher than 0.22 µm fraction increased. This transformation was due to partial degradation of larger molecules. Majority of the aromatic organics were >0.22 µm in raw compost leachate and the most of the reduction of UV_{254} absorbance obtained for this fraction. The percentage of the high molecular aromatic organics (UV_{254}) decreased, while relatively low molecular weight fractions (<5 kDa) significantly increased after the anaerobic treatment. Majority of aromatics in anerobic effluent was in the range of 0.1–30 kDa and those fractions could be effectively removed by membrane systems.

3.4. Performance comparison of membranes

AFBR effluent was filtrated through the combination of microfiltration (0.45 µm) and ultrafiltration membrane (UP150) to remove suspended and colloid materials. Characteristics of influent leachate for membrane system were as given in Table 4.

As can be seen from Table 4, removals of COD and BOD_5 were mainly achieved by MF while UP150 provided twice higher ammonium removal than MF. On the other hand, there was no UV_{254} removal in MF and it was decreased about 42% after UP150 filtration. During the operation of UP150 membrane, flux decreased from initial value of 20 L/m^2·h below 10 L/m^2·h almost within 70 min. Effluent from UP150 was separately filtrated by four different membranes and comparison of removal efficiencies were as shown in Fig. 6.

As can be seen in Fig. 6, XLE membrane provided the highest removal efficiencies in all parameters except ammonium. In all membrane systems, the highest removal efficiencies of COD, BOD_5, SO_4^{-2} and UV_{254} were 97.2%, 99.6%, 96.3% and 99.5% respectively. Except BW30, all membranes provided the similar NH_4^+ treatment efficiencies around 93%. In comparison to other membranes, BW_{30} had the lowest treatment performances with 89.3% COD, 98.8% BOD_5, 85.8% SO_4^{-2} and 92% NH_4^+. On the other hand, BW_{30} achieved higher decrease in UV_{254} compared to TSXN45 and NF90

Flux trends of the membranes were as shown in Fig. 7. On the one hand, BW_{30} and TSXN45 membranes showed similar sharp decrease in operational flux. Initial flux of BW_{30} was 4 L/m^2·h declined below 2 L/m^2·h within 50 min and it was almost stable until the end of the filtration period of 180 min. Operational flux of TSXN45 decreased from the initial value of 25 L/m^2·h to below 20 L/m^2·h within 70 min. Reduction in flux increased until the end of filtration with the final flux of 13 L/m^2·h. The initial flux of 2.75 L/m^2·h declined below 2 L/m^2·h within 25 min for NF90 membrane and it was 1.5 L/m^2·h at the end of operation. For XLE membrane, flux declined from 8 L/m^2·h below 3 L/m^2·h within 27 min and the operation was completed with 2 L/m^2·h. In this study, no flux recovery strategy was applied since membrane filtration was applied for the advanced treatment of AFBR effluent in batch mode. However, in order to recover flux and reduce membrane fouling in continuous operation, in situ or ex situ chemical cleaning by using acids, bases, chelates or surfactants can be applied [44].

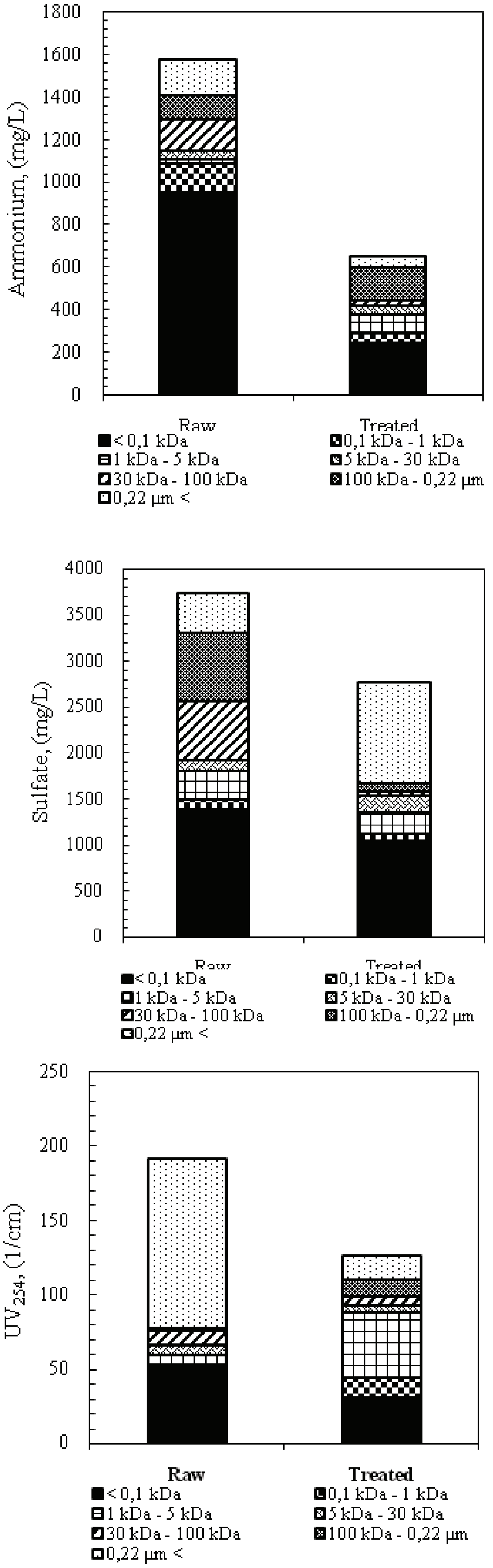


Fig 5. Molecular fractions of NH_4^+, SO_4^{2-} and UV_{254}.

Table 4
Influent characteristics for membrane systems

Membranes	COD (mg/L)	BOD_5 (mg/L)	SO_4^{-2} (mg/L)	NH_4^+ (mg/L)	UV_{254} (1/cm)
MF	8 900±50	1 800±35	2 250±15	653±10	126±2
UP150	6 550±20	575±12	2 227±15	605±10	126±2
TSXN45,NF90,XLE,BW$_{30}$	4 550±15	504±10	1 889±10	511±5	73±2

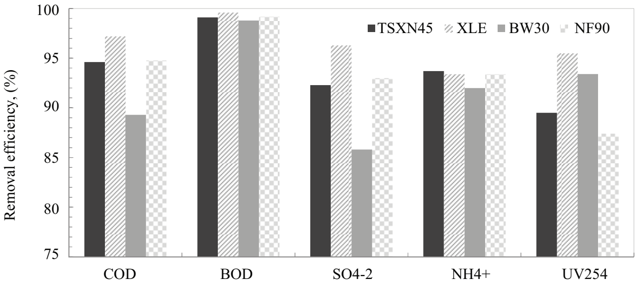


Fig. 6. Pollutant removal efficiencies of membranes.

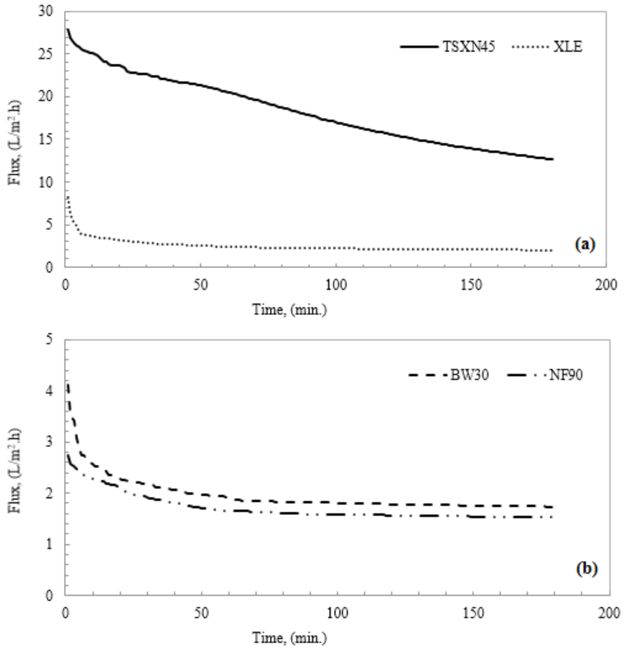


Fig. 7. Changes in operational flux of membranes a) TSXN45 and XLE; b)BW30 and NF90.

5. Conclusions

Leachate from a full-scale composting plant was treated in the combined system of an anaerobic fluidized bed reactor and membrane system. Configurations of membrane systems were determined according to the molecular weight distribution analysis of anaerobic effluent. Evaluation of membrane systems was conducted based on flux changes and the removal efficiencies of pollutants from the effluent of anaerobic fluidized bed reactor.

The results of this study may be drawn as follows:

- Low removal efficiencies of COD, BOD_5 and UV_{254} in AFBR indicated the inhibition of anaerobic process by high ammonia.
- Similar to organic matter, anaerobic sulphate reduction was negatively affected by ammonia and low level of COD was used for sulfate removal.
- During the anaerobic treatment high molecular weight fractions of all pollutants were significantly decreased while percentages of low molecular weights are different in each parameter.
- MF and UF membranes successfully removes particular and colloidal matter and significant amounts of organics and sulfate are removed in subsequent NF and RO membranes.
- TXN45, NF90 and XLE membranes provide closer effluent quality while BW30 membrane has the lowest treatment performance.
- Effluent quality from all membrane systems meet with discharge limits.
- The highest flux is obtained with TXN45 membrane. Therefore, combination of AFBR+MF+UF+TXN45 could be suggested for the treatment of compost leachate.

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