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Treatment of mature landfill leachate in tropical climate using membrane bioreactors with different configurations

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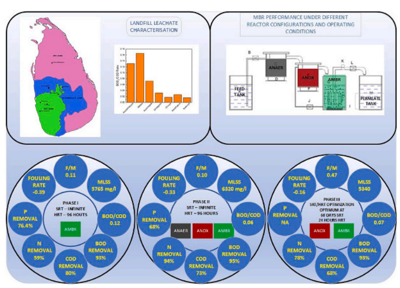
HIGHLIGHTS

- Leachate collected from seven landfill sites covering 3 climatic zones in Sri Lanka.
- They were characterized as mature leachate based on the analyses.
- A laboratory scale MBR system was optimised for treating the actual leachate.
- SRT of 60 days and HRT of 24 h were found to be the optimum.
- MLSS and membrane fouling rate were related to SRT.
- Slowly biodegradable substances and nitrogenous compounds can be removed by the MBR.

GRAPHICAL ABSTRACT



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ABSTRACT

This study describes the collection of landfill leachate from seven sites in different climatic zones of Sri Lanka and characterizes the landfills through the analyses of leachate quality. Membrane bioreactors (MBRs) with different configurations were employed to treat some of those leachates. An aerobic MBR (AMBR) system was operated in three Phases. In the first Phase, an AMBR alone, in the second Phase an anaerobic reactor followed by an anoxic reactor and an AMBR and in the third Phase an anoxic reactor followed by an AMBR were operated. In Phases I and II, the sludge retention time (SRT) and the hydraulic retention time (HRT) were kept at infinite (as no intentional wasting of sludge was made) and 96 h; in Phase III, the SRT was varied from 60, 30, 20 to 10 days and under each SRT, the HRT was varied from 96, 48, 24 and 12 h. The optimum operating conditions for the configuration used in Phase III was established through extensive experiments which had a SRT. The three MBR configurations removed more than 93%, 64.8% and 59% of BOD_5, COD and total nitrogen respectively. They also removed large amounts of slowly biodegradable substances and nitrogenous compounds other than NH_4^+, NO_3^- and NO_2^-. Relationships between SRT and MLSS as well as SRT and fouling rate of membrane have been found. The study illustrates the capabilities of MBR in treating landfill leachate.

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1. Introduction

Municipal solid waste management is identified as one of the main concerns in modern world. Several methods are utilized for solid waste management and solid waste landfilling remains a common method (Mandal, 2019). These facilities can be identified as open dump sites and sanitary landfills (Morita et al., 2021). Most of the developing and under developed countries are yet to be transformed from using open dumps to sanitary landfills. However the landfilling is major threat to the environment as it could generate toxic landfill leachate and pollute surrounding water sources (Nazia et al., 2021), both groundwater and surface water along with emitting methane to the atmosphere (when there are no recovery systems installed) and contributing to the increase in greenhouse gas emission (Chaudhary et al., 2021).

The liquid formed by the decomposition of waste and the intrusion of rainwater into the landfill is known as landfill leachate. Moisture enters the waste in a landfill, dissolves the pollutants into a liquid phase, and becomes sufficient to trigger a liquid flow, resulting in the formation of leachate (Kamaruddin et al., 2021). The quantity and quality of leachate generated vary from one landfill to the next, with short- and long-term fluctuations due to age, climate, hydrogeology, and waste composition changes (Vaccari et al., 2019). A landfill goes through four phases throughout the course of its life: aerobic, acetogenic, methanogenic, and stabilization phases (Tałałaj et al., 2019). During these times, leachate characteristics such as pH, Biological Oxygen Demand (BOD_5), Chemical Oxygen Demand (COD), (Ammonium-Nitrogen) NH_4^+-N, heavy metals content, and biodegradability change. Additionally, the leachate production is divided into three stages: young leachate (landfill is being operated for less than 5 years), intermediate leachate (between 5 and 10 years), and old leachate (more than 10 years). Leachate produced in new landfills has a high BOD_5/COD ratio (greater than 0.5), which indicates better biodegradability of the leachate (Bhalla et al., 2013).

Conducting a literature review, it was identified that a comprehensive leachate quality characterizing study has not performed in Sri Lanka. Sewwandi et al. (2012) has conducted leachate quality analysis covering 12 waste disposal sites, where the samples had collected only once from each sampling site. This study indicated the leachate quality from landfills in Sri Lanka exceeded the maximum tolerance limits in Sri Lankan Standards. Apart from this study, leachate quality analysis covering all the climatic zones of the country is not currently available. Few other studies reported leachate quality from Gohagoda dumpsite located in Kandy district in Sri Lanka (Wijesekara et al., 2014; Dharmarathne and Gunatilake, 2013; Kumarathilaka et al., 2016; Vithanage et al., 2014). These studies did not report leachate quality from any other sites in Sri Lanka. Considering this, aiming at identifying the leachate quality from dumping facilities Sri Lanka, leachate characterizing study was conducted covering seven dumping sites from three climatic zones in Sri Lanka. It is important to understand the characteristics of leachates generated to identify proper treatment options (Fan et al., 2006; Mukherjee et al., 2015). Various biological and physical/chemical treatments have been developed to treat the leachate under the settings mentioned above (Gao et al., 2015). Biological technologies are frequently used to treat the majority of the biodegradable fraction in the leachate, while physical/chemical approaches used as pre/post treatment to eliminate specific persistent pollutants (Costa et al., 2019). Biological treatment of landfill leachate, particularly matured landfill leachate, is extremely challenging (Ahmed and Lan, 2012). The development of combined biological and physical/chemical separation processes to protect water resources has led to the recognition of membrane bioreactor (MBR) system as the process of choice for the treatment of high-strength wastewater characterized by a high content of complex and recalcitrant compounds (Hashisho and El-Fadel, 2016).

MBR can be thought of as a conventional activated sludge system with effective membrane filtration. It takes the place in the conventional wastewater treatment's secondary sedimentation stage (Hao et al., 2018). MBRs have been used to treat wastewater from a variety of sources, including food and meat (Abyar and Nowrouzi, 2020), pharmaceuticals (Kaya et al., 2016), paper and pulp, textiles (Jegatheesan et al., 2016), wineries, and oil (Asif et al., 2019). The MBR consists of two parts: a bioreactor and a membrane module that separates the treated and cleaner effluent from residual suspended solids (Hashisho and El-Fadel, 2016).

MBRs have become a choice for leachate treatment after being widely used in the treatment of both municipal and industrial wastewater (Judd, 2010). Zhang et al. (2020) reviewed full-scale MBR usage in China to treat leachate. Most of the reported studies on usage of MBR for leachate treatment focus on treatment of young leachate (Chiemchaisri et al., 2011; Li et al., 2010; Hasar et al., 2009a; Yiping et al., 2008). Reported literature lacks studies on attempts treat real landfill leachate in matured conditions from tropical countries.

In this study, MBR technology was tested in treating landfill leachate obtained from two landfill sites. Different reactor configurations (aerobic MBR, Anaerobic reactor – anoxic reactor – aerobic MBR and, anoxic reactor – aerobic MBR) with varying sludge retention times (SRT) and hydraulic retention times (HRT) were used to identify the optimum conditions for treating the leachates used in the study.

The main objectives of the study are:

1. Characterise and analyze the variations of landfill leachate emanating from solid waste dumping facilities situated in different climatic zones of Sri Lanka
2. Setup a laboratory scale membrane bioreactor with different configurations and analyze the performance of those systems in treating landfill leachate under different operating conditions
3. Find the optimum operating conditions of a MBR to treat landfill leachate.

2. Methodology

2.1. Characterization of landfill leachate in Sri Lanka

2.1.1. Identification of landfills

Six open dumping sites and an engineered landfill were selected as the study locations for the leachate characterization. The engineered landfill located at Dompe and the open dumping sites located at Gohagoda, Kelaniya, Karadiyaana, Kurunegala, Anuradhapura and Matale are shown in Figure A1 in the supplementary materials. The locations of the dumpsites are also provided in Figure A1. Kelaniya open dumpsite is a closed dump and other six dumping facilities are active during the sampling period.

The island is traditionally divided into three climatic zones viz. 'dry', 'intermediate' and the 'wet zone' as shown in Figure A1 based on seasonal rainfall (Pathmeswaran et al., 2018). The wet zone receives relatively high mean annual rainfall of over 2500 mm, particularly from the south-west monsoons (from April to June) and does not have any pronounced dry periods. The dry zone receives a mean annual rainfall of less than 1750 mm, mostly through the north-east monsoons, which extends from October to January and has a distinct dry season from May to September. The intermediate zone receives a mean annual rainfall between 1750 and 2500 mm with a short and less prominent dry season. The type of vegetation differs between these zones with south-western lowlands marked by the presence of dense rain forests, while tropical dry forests prevail in the dry zone (Karunaweera et al., 2014). The landfills and dumpsites located at all three climatic zones (Dry, Intermediate and Wet) were selected for the leachate characterization. Figure A1 shows the classification of landfills and dump sites according to climatic zones.

To analyze the correlation of the leachate quality with rainfall, rainfalls recorded in rain gauges operated by the Department of Meteorology of Sri Lanka in the close proximities to the solid waste disposal sites were collected. The temporal variation of rainfall values (mm/

week) is given in the supplementary data.

2.1.2. Sampling and characterization

Leachate samples were collected from each location from September 2019 to February 2020 at monthly frequency. One liter of leachate was collected from each landfill/dump site in plastic containers and transported to the laboratory. Containers were stored at 4 °C prior to the characterization. In-situ measurements were done for the pH, Dissolved Oxygen (DO), Electrical Conductivity (EC) and temperature. For the pH measurements, a Thermo Scientific Orion Star A325 pH meter and for the DO, EC and temperature, a multi probe meter (Orion Star A325, USA) were used. COD, BOD_5, Total Organic Carbon (TOC), Total Nitrogen (TN), PO_4^{3-}, SO_4^{2-}, Nitrate-Nitrogen (NO_3^--N) and Nitrite-Nitrogen (NO_2^--N) were measured following the Standard Methods for the Examination of water and wastewater (American Public Health Association, 2005).

2.2. Leachate treatment using laboratory scale MBR

2.2.1. Experimental set-up

The laboratory scale experimental set-up was composed of two 4.5 L acrylic sheet tanks for anaerobic and anoxic reactors as well as one 7.5 L acrylic tank for aerobic reactor. A hollow fiber hydrophilic Polyethersulfone (H-PES) microfiltration membrane module with pore size of 0.1 μm was submerged in the aerobic reactor to separate the biomass from the permeate. Aeration for the scouring of membrane surface and for the biological degradation of pollutants present in the leachate was supplied by a compressed air pump. The reactor was seeded with waste activated sludge obtained from the sewage treatment plant of the temple of tooth relic in Kandy (a sequential batch reactor). Magnetic stirrer was used for the agitation of leachate in the anaerobic and anoxic tanks as shown in Fig. 1. Recirculation from the aerobic tank to anoxic tank was provided by means of a peristaltic pump. The recirculation ratio was kept at 1:1 with respect to the influent flow rate. Peristaltic pumps were used to feed the anaerobic reactor with desired flow rates and to pump permeate out from the membrane module submerged in the aerobic reactor. Automatic backwashing was provided for 1 min after every 14 min of permeating so that 15 min cycles were continued during the experiments. A vacuum gauge was fixed between the membrane module and permeating pump to measure the *trans*-membrane pressure over time.

2.2.2. Operating conditions

The reactor system was run under different conditions for 351 days with two interruptions at the end of 60 days and 183 days. Table 1 summarizes the operating conditions during different phases of reactor runs. During Phase I, only the aerobic reactor was used along with the membrane module which is referred to as AMBR. This experimental design was chosen to start the treatment process with a simple system. Sludge was not removed intentionally during the initial phase thus had infinite SRT and the HRT was maintained at 4 days. For the first phase of the study, the landfill leachate was obtained from Karadiyana dumpsite. The anaerobic reactor and anoxic reactors were introduced during Phase II before the AMBR and SRT and HRT in AMBR were kept as in Phase I. This system is referred to as A2O-MBR. The reason for selecting A2O-MBR was to investigate the optimum performance of an MBR configuration. During Phase III, only the anoxic and aerobic reactors were used with the membrane module submerged in the aerobic reactor. This system is referred to as Anx-AMBR. The third phase consisted of sub phases where the SRT and HRT were changed to optimize the conditions in the system. The results from Phase II of the experiment indicated that higher percentage of the pollutant removal was found to be from the aerobic reactor. The contribution of the anaerobic reactor was minimal for the lower strength leachate observed from sites in Sri Lanka. Considering the pollutant removal observed from the reactors during the experiments, Phase III was designed as Anx-AMBR system. HRTs of 96 h, 48 h, 24 h and 12 h were used with SRTs of 60 days, 30 days, 20 days and 10 days of operations. The influent landfill leachate was obtained from the Gohagoda dumpsite for Phases II and III. Further, the change from one phase to another coincided with the two interruptions. At the beginning of each phase, new sludge was introduced with acclimation periods of 20 days for aerobic sludge and 28 days for anaerobic and anoxic sludge. Table 1 does not include these acclimation periods.

2.3. Sampling and analysis

During Phase I of the experiments, influent and effluent samples were collected weekly. Samples from influent, outlet of the anaerobic reactor, outlet of the anoxic reactor and final effluent were collected during the second phase and samples from the influent, anoxic reactor outlet and final effluent were collected during the third phase. The samples were tested for COD, BOD_5, TOC, TN, NH_3–N, NO_3^--N, NO_2^--N and PO_4^{3-} following the standard methods (American Public Health Association, 2005). For the measurements of MLSS, samples were taken from the aerobic MBR and concentration was analyzed according to the Standard Method 2540 D (American Public Health Association, 2005). We measured the initial biomass concentrations in the anaerobic and anoxic reactors. However, both due to time constraints and also observing that the major removal of various pollutants was occurring in the AMBR, we were measuring the MLSS only in the AMBR afterwards.

2.3.1. Fouling analysis

Trans-membrane Pressure (TMP) and permeate flow rates were measured during all the phases of the study. The flux was calculated using equation (1) (Sagbo et al., 2008).

$$Flux\ (LMH) = \frac{Permeate\ flow\ rate\ \left(\frac{L}{h}\right)}{Membrane\ surface\ area\ (m^2)} \tag{1}$$

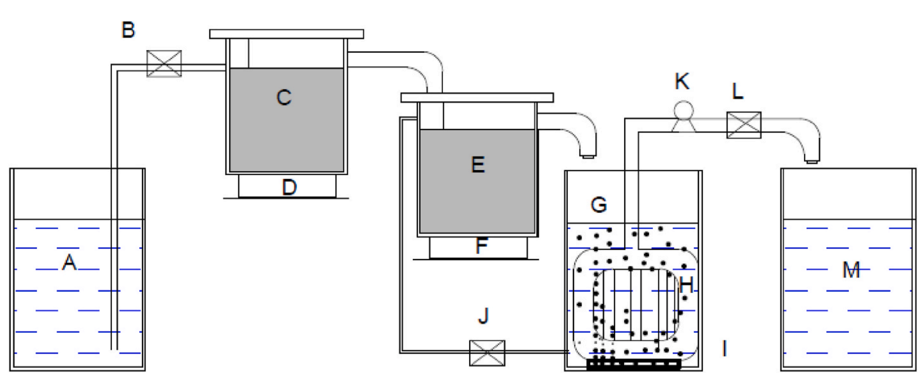


Fig. 1. Schematic diagram of the experimental setup (A – Feed tank, B – Feed pump, C – Anaerobic reactor, D – Magnetic stirrer, E – Anoxic reactor, F – Magnetic stirrer, G – Aerobic reactor, H – Membrane module, I – Air diffuser, J – Recirculation pump, K – Vacuum gauge, L – Permeate pump, M – permeate tank).

Table 1
Operating conditions for different phases of experiments.

Phase	Phase I	Phase II	Phase III			
Reactor Configuration	AMBR	A2O-MBR	Anx-AMBR			
SRT (days)	Infinite SRT	Infinite SRT	60	30	20	10
HRT (hours)	96	96	96,48,24,12	96,48,24,12	96,48,24,12	96,48,24,12
Number of days of operation (days)	60	105	77	35	28	28

AMBR – Aerobic MBR.
A2O-MBR – Anaerobic Reactor, Anoxic Reactor, Aerobic MBR.
Anx-AMBR- Anoxic Reactor, Aerobic MBR.
Infinite SRT – No intentional sludge removal was made.

Using the flux and TMP, specific flux is calculated in (LMH/bar) using equation (2).

$$Specific\ flux\left(\frac{LMH}{bar}\right)=\frac{Flux\ (LMH)}{TMP\ (bar)} \tag{2}$$

2.3.2. Membrane cleaning

The TMP and flux were monitored regularly and when the TMP increased to more than 70 kPa or flux decreased below 4 LMH, the physical cleaning of the membrane module by means of a soft brush and distilled water was performed. Due to operational issues during the experiments new membrane modules were introduced for each phase of the experiment, so that each phase was started with new membrane module with similar properties. The membrane modules were chemically cleaned by dipping in 10% sodium hypochlorite (NaOCl) solution for 1 h once the permeate flux dropped below 3 MLH.

3. Results and discussion

3.1. Characteristics of leachate obtained from various parts of Sri Lanka

The range and average values for pH, EC, Temperature, BOD_5, COD, TOC, TN, PO_4^{3-}, SO_4^{2-}, NO_3^--N and NO_2^--N of each landfill are shown in Table 2. The pH of leachate obtained from all the sites were alkaline, ranging from 7.1 to 8.3. The alkaline composition of leachates implies that the dumping sites have reached maturity (Vahabian et al., 2019). The pH of leachate becomes alkaline in nature when the concentration of partially ionized free volatile fatty acids taken up by methane-producing bacteria falls (Li et al., 2020). Furthermore, as a site becomes older and more established, pH of the leachate tends to rise progressively from slightly acidic to alkaline levels (Lee et al., 2022). The evidence of higher pH values of leachate (>7.5) for historic landfills (Aziz et al., 2010), where they are also capable of carrying a greater load of dissolved compounds, strongly agrees with the long years of operation of these landfills (Naveen et al., 2017). Because the ratio of old and stabilized garbage to freshly deposited waste was high, as was the alkalinity, acidogenic leachates were not seen even when new waste was deposited at these landfill sites even if they are active (Hussein et al., 2019). When investigating the relationship of pH variation with rainfall, a definite relationship couldn't be observed.

When comparing the EC values of leachate, Gohagoda and Kelaniya dumpsites and Dompe landfill had EC values under 5 mS/cm. All the other dumpsites had EC values between 5 and 25 mS/cm. Except for Karadiyana dumpsite, the EC values of the dry and intermediate zones are considerably greater than the wet zone leachate.

The amounts of BOD_5 and COD in open dumpsites were found to be greater than those detected in leachate samples taken from the Dompe sanitary landfill. The organic content of sanitary landfills is much lower than that of open dumpsites, according to this finding. When analyzing the correlation of rainfall with COD variation, all the locations showed a negative correlation except for the Kurunegala dumpsite. This indicated that the COD concentration has decreased with the increase in rainfall. When comparing the mean BOD_5 values, Dompe landfill showed the lowest value of 43.03 ($\pm$14.17) mg/L. Anuradhapura dumpsite showed the highest BOD_5 value of 508.45 ($\pm$187.49) mg/L. It can be seen that the leachate from dry and intermediate zones showed a higher mean BOD_5 value compared to Wet zone leachate. Except for Matale and Karadiyana dumpsites, all the other sites showed a negative correlation with rainfall which indicates that increasing of rainfall results in a decrease in the BOD_5.

The BOD_5/COD ratios of the leachate from selected locations are shown in Fig. 2. Highest BOD_5/COD value was recorded from Matale dumpsite and the lowest from Kelaniya dumpsite. It can be seen clearly that high BOD_5/COD values were recorded for leachate from dry and intermediate zones and the lower values from wet zone. Leachate from a young landfill (<5 years) is typically characterized by high BOD_5 and COD concentrations, and pH value below 6.5 (Lindamulla et al., 2022, Tałałaj et al., 2021). In contrast, leachate from a mature or stabilized landfill (>10 years) usually moderately high strength of COD, and BOD_5/COD ratio lower than 0.1 (Adhikari et al., 2014). Hence, all the locations except for Anuradhapura and Matale dumpsites can be identified as matured landfills.

The BOD_5/COD ratio is used to describe the proportions of biodegradable organic elements in landfill leachate. The BOD_5/COD ratio also reflects the maturity of a landfill. A ratio more than 0.4 indicates that the leachate is in the acid phase, while a ratio less than 0.1 indicates that the organic compounds in the leachate are less biodegradable (Corsino et al., 2020). The BOD_5 and COD values recorded during the study were below 400 mg/L and 4000 mg/L respectively for Gohagoda and Dompe sites, indicating they have achieved the matured state. This occurs when biological processes may quickly remove much of the biodegradable organic material during the early stages of landfilling. Because of the age of the landfills (>20 years), which can be termed as old landfills and may contain a significant amount of biologically inert materials, where lower BOD_5/COD ratios (0.1) were recorded, indicating that they have attained a stable phase (Sewwandi et al., 2012).

Anuradhapura dumpsite has the highest Sulphate concentration of 101.3 mg/L compared to the lowest value of 14.2 mg/L in Kurunegala. The relationship between Sulphate variation and rainfall is summarized in Table 2. Except for Gohagoda dumpsite, all of the other locations revealed a negative correlation, indicating that as rainfall increases, the concentration of Sulphate in leachate decreases. The Sulphate change could be due to the dilution of leachate by rain. Karadiyana dumpsite, in particular, received a -0.999 correlation coefficient, showing a high negative association.

Gohagoda dumpsite had the highest mean value of 125.0 mg/L for nitrite. Kurunegala dumpsite has the lowest mean value. Dry and Intermediate zone leachate had lower mean values than Wet zone leachate, as can be seen in Table 2. When examining the link between Nitrite variation and rainfall, it can be seen that, with the exception of Matale, all other sites have a negative relationship. Karadiyana, in particular, displayed a substantial negative correlation. The correlation between the other places was moderate. Thus, considering the characteristics, the leachate from landfills in Sri Lanka studied during the study can be classified as matured leachate.

The average pH values from all the three zones were observed as 7.8.

Table 2
Characteristics of leachate from selected dumpsites in Sri Lanka (ODS=Open Dump Site, LF = Landfill).

	Location	Anuradhapura	Matale	Kurunegala	Gohagoda	Dompe	Karadiyana	Kelaniya
Region		Dry	Intermediate	Intermediate	Wet	Wet	Wet	Wet
Status		Active	Active	Active	Active	Active	Active	Closed
Type		ODS	ODS	ODS	ODS	LF	ODS	ODS
pH	Range	7.5–8.0	7.9–8.3	7.3–8.1	7.5–7.9	7.1–8.2	7.5–8.2	7.3–7.9
	Mean (Standard Deviation)	7.8 (±0.140)	8.0 (±0.171)	7.8 (±0.259)	7.7 (±0.096)	7.9 (±0.399)	7.9 (±0.180)	7.7 (±0.179)
	Correlation with Rainfall	−0.586	−0.444	0.659	0.249	0.509	−0.854	−0.895
Temperature (°C)	Range	31.2–37.9	27.0–33.0	28.8–31.2	28.4–30.1	29.8–34.9	28.6–38.2	31.7–33.8
	Mean (Standard Deviation)	34.8 (±2.78)	28.9 (±2.25)	29.9 (±0.87)	29.1 (±0.51)	32.1 (±1.81)	33.7 (±1.77)	32.8 (±0.86)
EC (mS/cm)	Range	7.68–17.54	11.21–16.75	6.35–14.01	0.87–13.63	1.39–3.01	1.05–23.15	1.25–7.25
	Mean (Standard Deviation)	12.78 (±2.61)	13.20 (±1.54)	10.28 (±2.54)	3.78 (±4.52)	1.97 (±0.58)	11.90 (±5.70)	3.78 (±2.00)
	Correlation with Rainfall	0.770	−0.700	0.849	−0.655	−0.545	−0.923	−0.880
COD (mg/L)	Range	790–13,480	430–3080	1250–8605	1520–5900	1250–5320	1110–21,500	2420–7930
	Mean (Standard Deviation)	3875 (±4339.49)	1720 (±757.89)	4655 (±3028.67)	3523 (±1402.49)	2443 (±1289.78)	8182 (±5665.41)	4496 (±1770.12)
	Correlation with Rainfall	−0.937	−0.872	0.494	−0.959	−0.497	−0.969	−0.564
BOD_5 (mg/L)	Range	300.0–960.0	32.4–414.0	74.4–837.0	30.8–365.2	22.1–68.1	16.2–452	33.2–98.1
	Mean (Standard Deviation)	508.45 (±187.49)	284.50 (±132.62)	337.49 (±214.22)	111.71 (±104.64)	43.03 (±14.17)	213.77 (±162.81)	72.74 (±20.12)
	Correlation with Rainfall	−0.324	0.500	−0.998	−0.558	−0.786	0.408	−0.968
Sulphate (mg/L)	Range	15–250	0–80	0–24	0–180	0–40	0–450	0–100
	Mean (Standard Deviation)	101.3 (±96.91)	29.8 (±28.02)	14.2 (±9.09)	52.0 (±55.64)	21.4 (±16.38)	87.3 (±100.44)	24.5 (±27.96)
	Correlation with Rainfall	−0.518	−0.309	−0.223	0.938	−0.477	−0.999	−0.157
Nitrite (mg/L)	Range	0–100	20–90	0.15–60	20–400	30–300	0–250	5–250
	Mean (Standard Deviation)	36.0 (±27.16)	46.0 (±24.58)	25.6 (±19.46)	125.0 (±138.72)	88.0 (±89.29)	57.1 (±68.01)	69.6 (±90.70)
	Correlation with Rainfall	−0.367	0.056	−0.223	−0.307	−0.673	−0.996	−0.733
Nitrate (mg/L)	Range	10–1400	30–150	5–54	12–1300	0–31	0–100	23–90
	Mean (Standard Deviation)	488.0 (±599.64)	73.0 (±39.10)	30.7 (±15.20)	244.3 (±437.29)	16.6 (±10.81)	23.1 (±20.38)	39.2 (±23.07)
	Correlation with Rainfall	−0.504	−0.566	0.994	−0.394	−0.026	−0452	−0.373
Phosphate (mg/L)	Range	6–500	5–28	24–49	11.7–62	2.8–8.5	0–90	5.3–50
	Mean (Standard Deviation)	137.7 (±152.03)	22.9 (±6.64)	35.72 (±7.34)	28.5 (±14.78)	5.8 (±1.84)	26.5 (±16.14)	11.9 (±12.86)
	Correlation with Rainfall	−0.512	−0.996	0.914	−0.676	−0.698	−0.997	−0.660
TOC (mg/L)	Range	1119–12,290	421–1250	773–1364	556–987	30–95	141–3009	10–1052
	Mean (Standard Deviation)	6139 (±4493.06)	696 (±289.13)	978 (±157.51)	808 (±128.80)	57 (±19.09)	1275 (±822.89)	619 (±382.78)
	Correlation with Rainfall	−0.511	−0.295	−0.168	0.271	0.713	0.413	−0.874
TN (mg/L)	Range	465–15,390	838–1404	677–1323	239–1620	21–187	400–2687	124–366
	Mean (Standard Deviation)	3469 (±5899.12)	1081 (±191.04)	945 (±154.33)	855 (±557.09)	62 (±53.15)	1400 (±631.65)	199 (±66.25)
	Correlation with Rainfall	−0.519	−0.391	−0.5561	−0.994	−0.418	−0.422	−0.423
BOD_5/COD	Range	0.046–0.273	0.017–0.757	0.011–0.201	0.01–0.058	0.008–0.036	0.017–0.044	0.009–0.028
	Mean (Standard Deviation)	0.158 (±0.091)	0.262 (±0.255)	0.119 (±0.075)	0.030 (±0.017)	0.020 (±0.010)	0.028 (±0.010)	0.018 (±0.006)
	Correlation with Rainfall	0.999	0.937	−0.878	−0.015	0.042	0.880	−0.074

The strength of leachate could be seen reducing from dry zone to intermediate zone to wet zone. The values of EC, BOD_5, Sulphate, Phosphate, TOC and TN were found to reduce from dry zone to wet zone (characteristics of leachate obtained from the three climatic zones of Sri Lanka are given in the Supplementary Materials). This could be attributed to the dilution effect that can occur due to higher amount of rainfall recorded in wet zone compared to the dry zone. The BOD_5/COD ratio in dry and intermediate zones indicate that leachate from intermediate and dry zones are slightly more biodegradable compared to those from the wet zone.

3.2. *Performance of MBR in treating matured landfill leachate under different operating conditions*

3.2.1. *Influent characteristics*

The leachate obtained from the Karadiyana dumpsite was used for the Phase I of the study. Average COD, BOD_5, TN and PO_4^{3-} concentrations in the feed for Phase I were 2482.6 mg/L 292.6 mg/L, 475.3 mg/L and 23.7 mg/L, respectively. During the Phase II and Phase III, the feed leachate was obtained from Gohagoda dumpsite. During the Phase II, the average influent COD, BOD_5, TN, NH_3–N, NO_3^--N, NO_2^--N and PO_4^{3-}

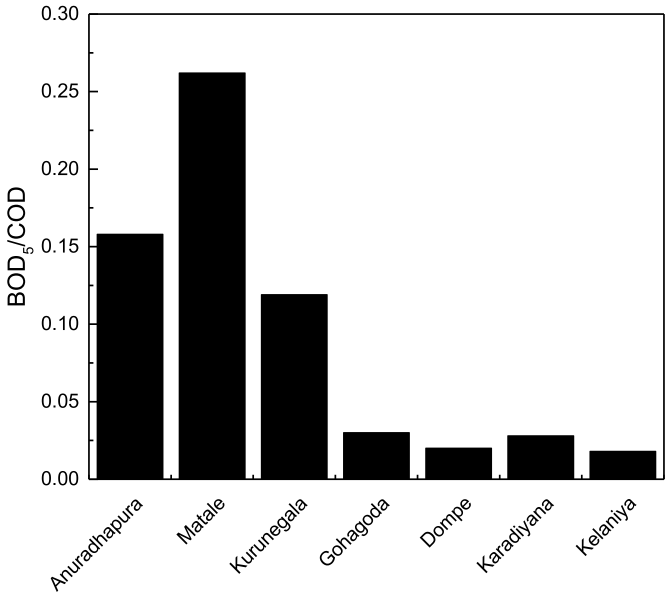


Fig. 2. Variation in BOD_5/COD ratio with landfill locations.

were 1978.1 mg/L, 113 mg/L, 784.7 mg/L, 98.7 mg/L, 32.1 mg/L 2.0 mg/L and 14.2 mg/L, respectively. The influent conditions for different conditions used in the Phase III are shown in Table 3.

3.2.2. *MLSS in the MBR systems*

The capability of biological treatment and the stability of the MBR system are both dependent on the growth of sludge concentration (Chen et al., 2012). During the Phase I, the MLSS in the AMBR increased from 4700 mg/L to 6690 mg/L within a period of 36 days. Initially, sludge taken from the sewage treatment plant of Temple of Tooth relic was introduced to the system. With the introduction of landfill leachate to the system, gradual growth of MLSS was observed. As there was no intentional sludge removal during the Phase I of the study, the MLSS grew at an average rate of 55.3 mg/L/day. This is a considerable increase of MLSS in the system and would have caused 5500 mg/L rise per every 100 days. This would have changed due to factors such as food to microorganisms (F/M). The increase in MLSS would reduce the growth rate. MLSS in the aerobic MBR was measured during the Phase II as well. An increase of MLSS from 5400 mg/L to 7020 mg/L was observed within 105 days. MLSS increased at rate of 15.4 mg/L/d. Comparatively low level of COD and BOD_5 observed in the feed of Phase II and the presence of anaerobic and anoxic reactors prior to aerobic MBR reduces the food for microorganisms making them grow at a slow rate. During the third phase with periodic sludge removal, a decrease in MLSS over time was observed. Fig. 3 (a) illustrates these changes in MLSS over the 3 phases of the study. The initial MLSS concentration of both the anaerobic and anoxic reactors during Phase II was 5000 mg/L. The initial MLSS concentration in the anoxic reactor during the Phase III was 5500 mg/L.

The average MLSS over different SRTs tested during the Phase III of the study are shown in Fig. 3 (b). According to the study, the relationship given by equation (3) was identified between MLSS (mg/L) in aerobic MBR and SRT (days) of MBR with a reliability (R^2) of 0.97.

$$MLSS = = 855.27 \ln(SRT) + 2117.6 \quad (3)$$

3.2.3. *Water quality parameters*

The average values of water quality parameters in the operations of all three phases are shown in Table 3 which is used in the subsequent discussions. It should be note that in Phase III, the SRT varied from 60 days to 10 days and the HRT was changed from 96 h to 12 h under each

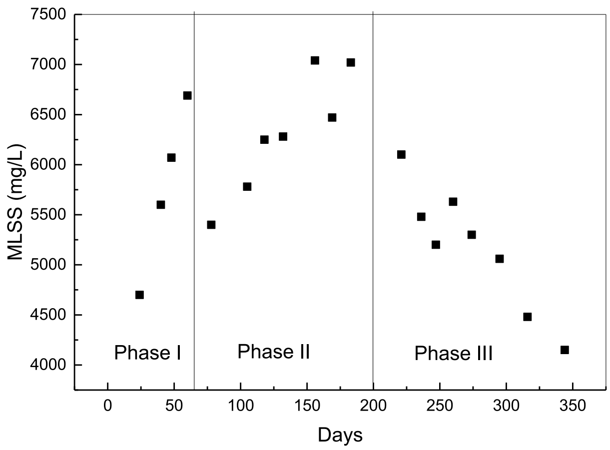
Table 3
Influent and treated leachate characteristics of anaerobic effluent, anoxic effluent and AMBR effluent during the three phases of the experiments.

			COD (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	Overall COD removal (%)	
I	2482.6	AB	AB	479.6	80.7	
II	1978.1	1750.6	1660.6	539.2	72.7	
III	2430.0	AB	NA	856.3	64.8	
			BOD_5 (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	Overall BOD_5 removal (%)	
I	292.6	AB	AB	13.8	95.3	
II	113	94	120	8	93.2	
III	167.7	AB	NA	11.1	93.4	
			TN (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	N removal (%)	Utilized COD:N
I	475.3	AB	AB	194.8	59.0	7.1
II	784.7	701.6	627.6	207.7	73.5	2.5
III	NA	AB	NA	NA	NA	NA
			NH_3–N (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	N removal (%)	Utilized COD:N
I	NA	AB	AB	NA	NA	NA
II	98.7	81.6	56.3	5.5	94.5	*
III	106.0	AB	NA	22.9	78.4	NA
			NO_3^--N (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	N removal (%)	Utilized COD:N
I	NA	AB	AB	NA	NA	NA
II	32.1	37.1	37.4	43.4	−35.2	*
III	NA	AB	NA	NA	NA	NA
			NO_2^--N (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	N removal (%)	Utilized COD:N
I	NA	AB	AB	NA	NA	NA
II	2.0	1.5	4.6	20.0	−882.2	*
III	NA	AB	NA	NA	NA	NA
			Other-N (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	N removal (%)	Utilized COD:N
I	NA	AB	AB	NA	NA	NA
II	651.9	581.3	529.3	138.8	78.7	*
III	NA	AB	NA	NA	NA	NA
			PO_4^{3-} (mg/L)			
Phase	Influent	Anaerobic Effluent	Anoxic Effluent	AMBR Effluent	P removal (%)	Utilized COD:P
I	23.7	AB	AB	5.6	76.4	110.7
II	14.2	36.5	83.5	26.4	68.3	35.1
III	17.0	AB	NA	21.4	NA	NA

NA – Not Available, AB – Reactor not used during the experiments.

HRT in the aerobic MBR. The removals of BOD_5 and COD under each of those conditions are shown in Table 4. It can be seen from Table 4, increasing the HRT at a given SRT did not improve the removal of COD, BOD_5 and NH_3–N significantly. Sadri et al. (2008) reported that the impact of HRT on COD, BOD_5, NH_3–N and metals removal is not considerable. Hashisho and El-Fadel (2016) also reported that the effect of HRT on performance of MBR is less evident. But increasing the SRT improved the removal of the above water quality parameters significantly. A longer SRT allows for the development of slow-growing bacteria as well as the establishment of specific microbial species required for the breakdown of slowly biodegradable substances (Hasar et al., 2009b, Hashisho and El-Fadel, 2016). A 24 h Of HRT and a 60-day SRT

(a)



(b)

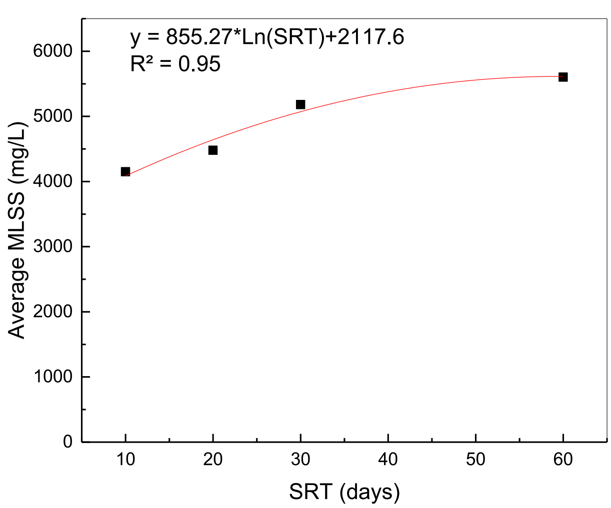


Fig. 3. (a) Variation of MLSS in AMBR in different phases of the study, (b) Variation of average MLSS vs SRT during Phase III.

gave 66%, 93% and 81% removal of COD, BOD_5 and NH_3–N removal respectively. This condition could be considered as the optimum operating conditions of the Anx-AMBR system to treat the landfill leachate considered in this study. Further, larger the SRT, lesser the amount of sludge wasted and therefore the lesser the costs associated with sludge treatment. Further increase in SRT as in the Phase I and the Phase II would have yield better removal of the pollutants with increased fouling rate making the operational issues as discussed in a later section. A 24 h Of HRT and a 30-day SRT gave 52%, 90% and 80% removal of COD BOD_5 and NH_3–N removal respectively. This is second best but significant drop in COD removal efficiency was observed. Therefore, in Table 3, the results for Phase III indicate a SRT of 60 days and HRT of 24 h.

Long term variations of the parameters considered are given in the in the Supplementary Materials. The data from Phase I and Phase II of the experiments indicated improvements in the performance of the reactor systems over time. In Phase III, where the operating conditions were changed during the experiments, the variations were depending on the SRT as discussed above.

Table 4
COD, BOD_5 and NH_3–N removal efficiencies in Phase III for different SRTs and HRTs.

		COD				
			HRT (hours)			
		Influent (mg/L)	12	24	48	96
SRT (days)	10	2370	30%	29%	22%	27%
	20	2455	32%	28%	38%	44%
	30	2485	50%	52%	50%	51%
	60	2477	62%	66%	61%	67%
		BOD_5				
			HRT (hours)			
			12	24	48	96
SRT (days)	10	144.5	89%	91%	91%	93%
	20	125.0	87%	90%	90%	90%
	30	130.8	91%	90%	91%	90%
	60	819.0	94%	93%	89%	92%
		NH_3–N				
			HRT (hours)			
			12	24	48	96
SRT (days)	10	84.0	65%	59%	61%	65%
	20	94.8	65%	61%	64%	77%
	30	118.6	79%	80%	77%	78%
	60	110.0	75%	81%	82%	81%

3.2.3.1. COD removal. All the indicators showed a drop in removal efficiencies with decrease in SRT. This can be due to the decrease in the biomass at shorter SRTs. COD is commonly utilized in the field of wastewater treatment as a surrogate metric for total organic material concentration (Chen et al., 2012). For LFL treatment using MBRs, a broad range of COD removal efficiencies have been observed, ranging from as low as 23% (Brasil et al., 2021) to over 90% (Chen and Liu, 2006). The MBR has used to treat matured landfill leachate and removal efficiencies more than 75% has been reported (Sadri et al., 2008; Aloui et al., 2009; Robinson, 2007). The mean COD concentration in the feed landfill leachate during the Phase I of the experiments was 2482.6 mg/L with a standard deviation of 165.8 mg/L. The mean permeate COD concentration during the corresponding period was 479.6 mg/L with a standard deviation of 86.5 mg/L. This gave an average removal efficiency of 80%. For infinite SRT aerobic MBR systems, treating landfill leachate COD removal of 60%–97% is reported (Tsilogeorgis et al., 2008; Sang et al., 2007). During the Phase II, the average COD concentration of the influent leachate was 1978 mg/L with a standard deviation of 342.6 mg/L. The corresponding permeate COD concentration was 539 mg/L giving mean removal efficiency of 73%. The anaerobic and anoxic reactors used during the Phase II did not have considerable effect on COD removal from the landfill leachate. The COD removal efficiencies of the two reactors were 11% and 5% respectively. The Aerobic reactor with MBR had 68% of COD removal which was 93% of the total COD removal efficiency of the system.

In the Phase III when the SRT and HRT changed, the COD removal efficiencies changed considerably. Table 4 shows the mean removal efficiencies of parameters considered for different HRTs and SRTs considered. COD removal efficiencies show that the SRT has higher influence on COD removal efficiency in MBR treating landfill leachate. For all the HRTs considered, the COD removal efficiencies for SRT of 10 days were below 30%. The COD removal efficiency dropped from 67.46% for SRT of 60 days to 22.36% for SRT of 10 days. The reduction of the COD removal in lower SRTs could be due to the lower contact time between the biomass and substrate. Setiadi and Fairus (2003) reported that SRTs less than 32 days (16 and 24 days) did not offer enough contact time between the biomass and the substrate, resulting in higher permeate COD concentrations over time. Hasar et al. (2009a) also stated that the COD removal efficiencies are decreasing with the decrease in SRT.

3.2.3.2. BOD_5 removal. Similar to COD, the mean BOD_5 concentration in the feed (raw landfill leachate) during the Phase I of the experiments was 292.6 mg/L with a standard deviation of 19 mg/L. The mean permeate BOD_5 concentration during the corresponding period was 13.8 mg/L with a standard deviation of 3.3 mg/L. This gave an average removal efficiency of 95%. During Phase II, the average BOD_5 concentration of the influent leachate was 113 mg/L. The corresponding permeates BOD_5 concentration was 8 mg/L giving a mean removal efficiency of 93%. BOD_5 removal efficiencies show that they were influenced by both the SRT and the HRT. Higher HRTs and higher SRTs removed more BOD_5 from the landfill leachate. The high elimination of biodegradable organic matter is typical of most biological LFL treatments (Ahmed and Lan, 2012).

The average COD of the leachate used as feed for the MBR systems in Phases I, II and III were 2483 mg/L, 1978 mg/L and 2430 mg/L respectively. Similarly, the BOD_5 values were 293 mg/L, 113 mg/L and 168 mg/L respectively. Therefore, the BOD_5/COD ratios in the three Phases were 0.118, 0.057 and 0.070 respectively. Thus, when the BOD_5/COD changed in the range between 0.057 and 0.118, and the removal percentage of BOD_5 was in between 93% and 95% although different configurations of MBR were used to treat the leachate in different Phases. This indicates the BOD_5 removal was not affected much by the BOD_5/COD ratio as the absolute BOD_5 values in the influent ranged from 113 mg/L to 293 mg/L which are sufficient for microbial growth under suitable conditions. However, COD removal reduced from Phase I to Phase III. Large SRT and HRT used in Phase I provided better COD removal supported by higher BOD_5/COD ratio. During the Phase II lower BOD_5/COD ratio was observed compared to the Phase I and thus the COD removal were also lower. This could have resulted by lower F/M also. In Phase III, increased BOD_5/COD compared to that was in Phase II assisted in a COD removal; however, the removal was slightly less than that in Phase II. It should be noted that in Phase III, the SRT and HRT considered for the analysis were 60 days and 24 h respectively, which were considered as the optimum operating conditions of the Anx-AMBR (this is discussed in section 3.2.3).

3.2.3.3. Removal of non-biodegradable/slowly biodegradable composition of the leachate. The non-biodegradable/slowly biodegradable composition of the leachate can be represented by the difference between the COD and the BOD_5 of the leachate. Fig. 4 shows the ratio between BOD_5 and the non-biodegradable/slowly-biodegradable components in the feed and the permeate from the aerobic MBR in Phase I; the ratio between BOD_5 and the non-biodegradable/slowly-biodegradable components in the feed, the effluent from anaerobic and anoxic tanks as well as the permeate from the aerobic MBR in Phase II and the ratio between BOD_5 and the non-biodegradable/slowly-biodegradable components in the feed and the permeate from the aerobic MBR in Phase III.

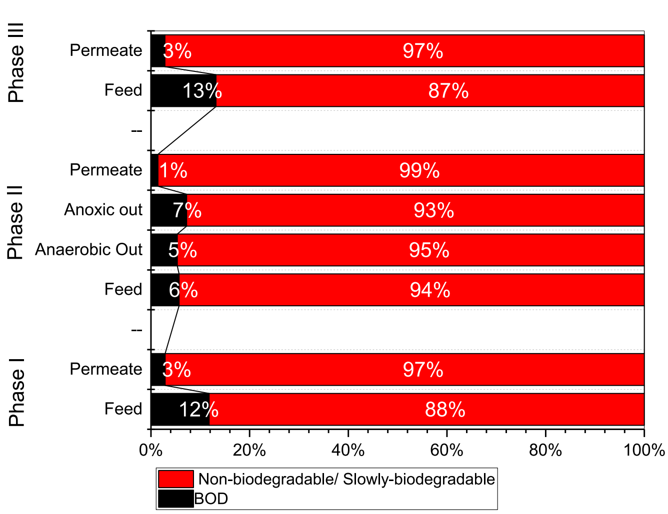


Fig. 4. Biodegradable and non-biodegradable/slowly biodegradable fractions in feed and effluents during the 3 Phases of the study.

The ratio of food to microorganisms (F/M) is also computed to evaluate its influence on the removal performance of the MBR reactors. The average F/M in Phases I, II and III were 0.11, 0.10 and 0.47 respectively, if F is considered as the influent concentrations of COD in those respective Phases. The F/M ratio rapidly dropped in response to changes in MLSS concentration, eventually approaching 0.07 kgCOD/kgMLSS/d during the phase II. Because of the low loading, the majority of microorganisms in MBR had a restricted supply of substrate and were forced to enter the endogenous respiration state rather than the physiological growth stage. As a result of the restricted nutrition, microbes would enter the "stable phase" (Chen et al., 2012). However, if the F is considered as the BOD_5 of the influent concentration, then the F/M values in those Phases were 0.05, 0.01 and 0.03, respectively showing a drastic drop compared to the F/M values computed using COD values. However, since the MBR systems removed slowly-biodegradable components in the leachate as discussed above, it is appropriate to consider the F/M values as the values computed using COD values.

3.2.3.4. Nitrogen removal. Total Nitrogen: As mentioned previously, the leachate used for Phase I was obtained from Karadiyana dumpsite and for Phases II and III it was obtained from Gohagoda dumpsite. Thus, drastic increase in the influent TN concentration was found in Phase II (784.7 mg/L) compared to that of in Phase 1 (475.3 mg/L). However, the TN removal increased from 59% to 73.5% indicating the influence of the anaerobic and anoxic reactors used in Phase II an addition to the AMBR that was used (as in Phase I). The COD:N utilized in Phase I was 7.1 which was closer to the COD:N utilized in conventional activated sludge process (10:1). However, COD:N utilized in Phase II was significantly low (2.5). Decrease in the COD from Phase I to Phase II (from 2482.6 mg/L to 1978.1 mg/L) along with the utilization of anaerobic and anoxic reactors could be the reason for this. Detailed study on the utilization of various nitrogenous species was conducted during Phase II to understand the use of nitrogenous species in the A2O-MBR. In Phase III, TN was not measured instead the removal of NH_3–N was measured for the analysis.

NH_3–N: During Phase II, the average NH_3–N concentration of the influent leachate was 98.7 mg/L. The corresponding permeate NH_3–N concentration was 5.5 mg/L giving an average removal efficiency of 95%. During Phase III (60 day SRT and 24 h HRT), however, the removal of NH_3–N reduced to 78.4% although the average influent concentration (106.0 mg/L) was similar to that of in Phase II. The capacity of MBRs to cope with changes in feed and loading circumstances has been demonstrated by the lack of large oscillations in NH_3–N removal efficiency and effluent NH_3–N concentrations (Ahmed and Lan, 2012).

Other nitrogenous species and nitrogen mass balance: In Phase II, NO_3^--N and NO_2^--N concentrations along the treatment train were measured. The NO_3^--N concentrations in the influent and the effluents from anaerobic reactor, anoxic reactor and AMBR were 32.1, 37.1, 37.4 and 43.4 mg/L respectively. Similarly, The NO_2^--N concentrations in the influent and the effluents from anaerobic reactor, anoxic reactor and AMBR were 2.0, 1.5, 4.6 and 20.0 mg/L respectively. The above measurements allow to calculate other nitrogenous species such as organic nitrogen along the treatment train. The concentrations of other nitrogenous species in the influent and the effluents from anaerobic reactor, anoxic reactor and AMBR were 651.9, 581.3, 529.3 and 138.8 mg/L respectively. This shows an excellent removal (78.7%) of nitrogenous species other than NH_3–N, NO_3^--N and NO_2^--N.

The following chemical reactions capturing nitrification along with the cell synthesis of *Nitrosomonas* and *Nitrobacter* (Mccarty and Haug, 1971) can be used to compute the amount of NH_4^+-N required to produce the concentrations of NO_3^--N and NO_2^--N that are produced while the influent was passing through the entire A2O-MBR (those concentrations were 11.3 mg/L NO_3^--N and 18 mg/L NO_2^--N as given in Table 3):

Nitrosomonas:

$$55\ NH_4^+ + 76\ O_2 + 109\ HCO_3^- \rightarrow C_5H_7NO_2 + 54\ NO_2^- + 57H_2O + 104H_2CO_3 \quad (4)$$

Nitrobacter:

$$400\ NO_2^- + NH_4^+ + 4H_2CO_3 + HCO_3^- + 195\ O_2 \rightarrow C_5H_7NO_2 + 400\ NO_3^- + 3H_2O \quad (5)$$

Overall:

$$NH_4^+ + 1.83\ O_2 + 1.98\ HCO_3^- \rightarrow 0.021C_5H_7NO_2 + 1.04H_2O + 0.98NO_3^- + 1.88H_2CO_3 \quad (6)$$

Thus, to produce 11.3 mg/L of NO_3^--N, 11.3 mg/L of NO_2^--N would have been required; to produce this NO_2^--N along with the amount NO_2^--N produced and unutilized (18 mg/L) would have required oxidation of 29.84 mg/L of NH_4^+-N. However, the total NH_4^+-N removed by A2O-MBR was 93.23 mg/L. Thus 63.39 mg/L of NH_4^+-N and 513.1 mg/L of N from other nitrogenous species must have been utilized by heterotrophic bacterial biomass as nitrogen source to degrade the BOD_5 and other slowly degradable substances. The sum of NH_4^+-N and the N from other nitrogenous substances that is equal to 576.1 mg/L is very close the TN utilized by A2O-MBR (577 mg/L). Further, A reduction in NO_2^--N in the anaerobic reactor indicates the presence of denitrifiers; however, they do not seem be present in sufficient concentration for complete denitrification to occur.

3.2.3.5. Phosphorus removal. Table 3 shows the utilization of phosphorus (measured in terms of PO_4^{3-} and converted to PO_4^{3-}-P when calculating the utilization). It can be seen that the AMBR used in Phase I has utilized COD:P of 339.3:1. It indicates that large amount of P is utilized when degrading the COD that is present in the leachate and 76.4% of P was removed from the influent. It should be noted that the conventional COD:P of 100:1 has been well exceeded when treating landfill leachate. When A2O-MBR is used in the second phase, as expected P is released from the microbial biomass present in the anaerobic reactor by phosphorus accumulating organisms (PAO) present in the reactor. This has continued in the anoxic reactor as well. The P has been taken up by the PAO again leading to 69% removal of P. However, the utilization ratio of COD:P in the AMBR was not as high as it was in Phase I. When Anx-AMBR was used in Phase III, the influent and effluent concentrations of P was similar to that of in Phase II, where A2O-MBR was used indicating similar performance of A2O-MBR and Anx-MBR with respect to the overall utilization of P.

3.2.3.6. Other water quality parameters. Total organic carbon (TOC): Average TOC removal efficiency of 87% was observed during the Phase I of the study. The influent TOC was 0.178 times the COD of the influent and 1.517 times the influent BOD_5. This indicates that the oxidisable organic carbon is a small fraction in the landfill leachate even though the TOC is more than the biologically oxidisable organic carbon. During Phase II, only slight reduction of TOC could be observed after anaerobic and anoxic reactors. Aerobic MBR removed considerable amount of TOC with average removal efficiency of 57%. The overall TOC removal efficiency of the entire system was 63%.

pH: During Phase II where anaerobic reactor, anoxic reactor and aerobic reactor were used, the pH was measured in effluent from each of the reactors and the that of the influent. If the alkalinity in the inflow to the AMBR is not maintained, denitrification in the anoxic tank should raise the pH and nitrification in the AMBR should lower it (Moazzem et al., 2020). In the current study, denitrification did not occur and thus the increase in pH in anoxic effluent could not be observed. The pH in the AMBR effluent could have been maintained by the sufficient alkalinity in the feed leachate.

3.3. Fouling analysis

Variations of flux and TMP over the study period for all the three phases are given in Figure A3 under supplementary data ((a) variation of flux with time, (b) Variation of TMP with time). During the experiments it could observed that the flux was dropping and TMP was increasing when membrane fouling occurred. When the flux drooped below 4 LMH or TMP increased over 70 kPa, the membranes were cleaned with soft brush using distilled water. After cleaning a significant drop in TMP and recovery of the flux could be observed as in Figure A3.

The fouling rates were determined at each phase of the study (Fig. 5). For each condition, the specific flux was determined using Equation (2). Slope of the specific flux, determined by plotting specific flux against number of days, for the phase III, was found to be −0.304 LMH/bar/day,-0.258 LMH/bar/day, −0.221 LMH/bar/day and −0.153 LMH/bar/day for SRTs of 10 days, 20 days, 30 days and 60 days respectively (Fig. 5 (c)). The slope of specific flux is identified as the specific fouling rate (El-Fadel et al., 2018). The fouling rate dropped by 0.068 LMH/bar/day moving from 30 days SRT to 60 days SRT. Reduction of fouling rate by 0.037 LMH/bar/day was observed on increasing 20 days SRT to 30 days SRT. When moving from 20 days SRT to 10 days an increase of fouling by 0.046 LMH/bar/day could be observed. El-Fadel et al. (2018) reported similar results on changes in fouling with SRT for high strength landfill leachate treatment using MBR. Extracellular Polymeric Substances (EPS), the main contributor towards fouling of membranes in MBR, is higher at lower SRTs whereas at higher SRTs, the EPS concentration is lower. The decrease in fouling with increase of SRT could be due to the fall in EPS in the system in moderate MLSS concentrations. At higher (or infinite) SRTs an increase in fouling rate can be observed. The higher MLSS concentrations despite the lower EPS could have blocked the pores of the membrane and caused the higher fouling rate. The fouling rates over different SRTs tested during the Phase III of the study are shown in Figure A4. According to the study, the relationship given by equation (7) was identified between fouling rate (LMH/bar/day) in and SRT (days) of MBR with a reliability (R^2) of 0.9992.

$$Fouling\ rate = -4 \times 10^{-5}(SRT)^2 + 0.0057(SRT) + 0.3568 \quad (7)$$

3.4. Recommendations and practical applications of this study

3.4.1. Recommendations

Currently there are no particular discharge standards for landfill leachate in Sri Lanka. The Central Environmental Authority of Sri Lanka has issued guidelines for industrial wastewater discharge into the inland water bodies, ocean and public sewers (Authority, 2008). The parameters considered in the study were compared with the guidelines for industrial wastewater discharge to inland water bodies. The guidelines have focused only on parameters such as BOD_5, COD, TSS, pH, NH_3–N and heavy metals. The leachate samples collected for characterization shows higher concentrations when compared to the values given in the guidelines. The treatment of the leachate with MBR reduces the pollutant concentrations below the discharge guidelines, except for the COD. The discharge guidelines for industrial wastewater requires COD of 250 mg/L or below whereas the effluent from the MBR system consisted of a minimum COD of 480 mg/L. Observing, that COD has been unable to reach the specified limit a tertiary treatment unit such as an adsorption column could be proposed to integrate into the MBR to polish the effluent to meet the guidelines. Further the absence of landfill leachate discharge standard in Sri Lanka is an important point to highlight. It is recommended to develop a landfill leachate discharge standards as landfills are the main method of solid waste management that is being practiced in Sri Lanka.

3.4.2. Practical applications

Currently, pre-treatment followed by biological treatment and polishing treatment is the most widely used combination for the treatment of landfill leachate and MBRs can be used as the biological treatment component. With removal efficiencies of BOD, COD, and NH_3–N

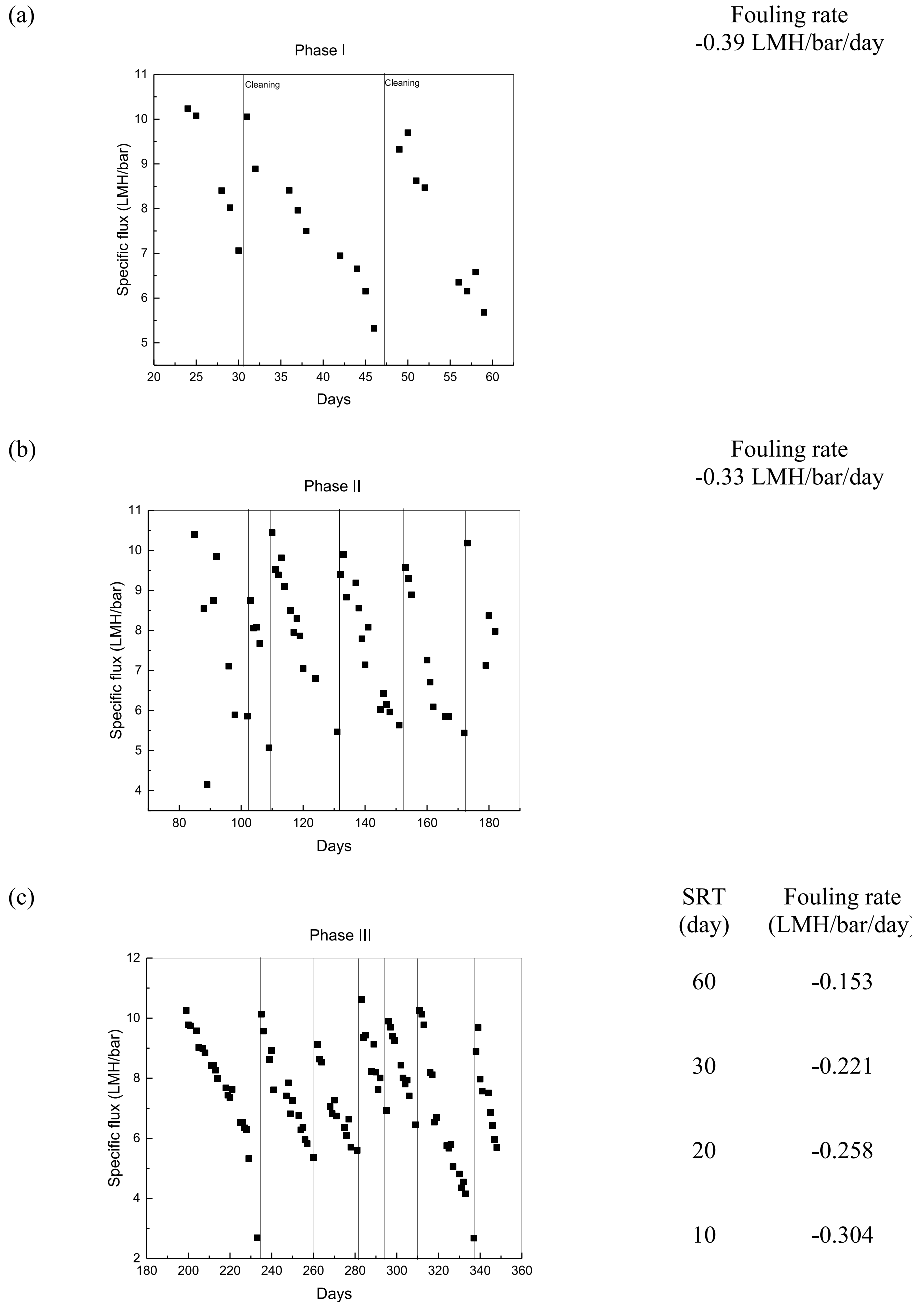


Fig. 5. Variation of specific flux (LMH/bar) through the membrane (a), Phase I, (b) Phase II, (c) Phase III.

exceeding 90%, they are competitive in the treatment of leachate when compared to many other existing biological treatments. The total capital cost of MBRs has been generally in the range of USD 7700–17000/(m^3/d of wastewater treated). Laeachate treatments utilizing full-scale MBRs typically have footprints between 5 and 25 m^2/(m^3/d of wastewater treated) (Zhang et al., 2020). Full-scale MBRs have an average operating cost of USD 3.5–5/m3 of wastewater treated, of which 43% was for energy costs, 19% was for chemicals, and 17% was for replacing membrane elements (Zhang et al., 2020). Thus, optimizing the performance of MBR is essential in reducing both capital and operating costs. This study shows that MBRs can be used to treat landfill leachate in tropical climates and when they are operated optimally, they will have (i) high removal efficiencies of pollutants present in the leachate, (ii) lower footprints, (iii) lower sludge generation and (iv) lower rate of fouling of membranes.

4. Conclusions

Leachate was collected from seven solid waste dumping facilities covering 3 climatic zones namely wet zone, intermediate zone and dry zone of Sri Lanka. The pH of leachate obtained from all the sites were alkaline in nature, ranging from 7.1 to 8.3. The alkaline composition of leachates implies that the dumping sites have reached maturity. A laboratory scale MBR system was operated in 3 phases to treat matured landfill leachate obtained from dumpsites in Sri Lanka. SRT and HRT were varied during the Phase III to identify the optimum operating condition to treat leachate. A SRT of 60 days and a HRT of 24 h was identified to be the best combination to operate the system in Anx. AMBR configuration.

Credit author statement

Lahiru Lindamulla: Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original Draft, Visualization. Ramod Jayewardene: Methodology, Formal analysis, Investigation. Samanthi Wijerathne: Methodology, Investigation. Maazuza Othman: Methodology, Writing - Review & Editing, Supervision. Nadeeshani Nanayakkara: Methodology, Resources, Writing - Review & Editing, Supervision. Shameen Jinadasa: Conceptualization, Methodology, Resources, Writing - Review & Editing, Supervision, Project administration. Gemunu Herath: Supervision, Writing - Review & Editing, Supervision. Veeriah Jegatheesan: Conceptualization, Methodology, Formal analysis, Writing - Review & Editing, Visualization, Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2022.136013.

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