



RESEARCH ARTICLE

Aluminum accumulation in treatment using submerged membrane electro-bioreactor of young landfill leachate: Statistical analysis

Gülizar Kurtoğlu Akkaya^{1,*}, Nur Ayvaz Çavdaroğlu², Mehmet Sinan Bilgili³

¹Necmettin Erbakan University, Department of Environmental Engineering, Meram, 42140, Konya, TURKEY

²Kadir Has University, Business Administration Department, Fatih, 34083, İstanbul, TURKEY

³Yildiz Technical University, Department of Environmental Engineering, Esenler, 34220 İstanbul, TURKEY

ABSTRACT

Herein, landfill leachate containing high amount of organic matter, which is quite difficult to treat, was first treated using the new submerged membrane electro-bioreactor (SMEBR) system. Aluminum (Al) electrode was used for the treatment of leachate in the SMEBR and Al accumulation was detected. This study aims to examine Al accumulation in the treatment of leachate with high organic content in the SMEBR system. The Al values obtained were plotted on a graph using MS Excel, and Mann-Whitney U test was used to determine whether there is a statistical difference between the observed Al values. Also, correlations between Al accumulations and conductivity and TOC in SMEBR and SMBR were evaluated. Resultantly, it was found that relationship between Al and conductivity is very weak, correlation between Al and TOC% is a weak-moderate, the Al accumulation in the SEMBR has a linear relationship with time and there is a very strong correlation between the two variables ($R^2= 0.7591$). Its correlation with time in the SMBR is moderate ($R^2= 0.3316$). MS Excel 2016 and Minitab 16.0 programs were utilized in the statistical analyses.

Keywords: Aluminum accumulation, landfill leachate, membrane bioreactor, statistical analysis, submerged membrane electro-bioreactor

1. INTRODUCTION

In landfill, leachate production is an inevitable consequence of water leaking through degradation and separation of waste. Depending on factors such as landfill age, precipitation, seasonal weather changes and waste composition, leachate characteristics vary and pollution load is higher than other wastewaters [1-3]. The leachate contains toxic components, recalcitrant structures and heavy metals that can damage humans and ecosystems. This hardly treatable wastewater can be effectively treated by electrochemical methods such as biological treatment, chemical precipitation, chemical oxidation, coagulation-flocculation. In addition, leachate can be treated by membrane bioreactors (MBR) which are integrated into biological treatment processes or formed by combination of membrane filtration systems such as ultrafiltration and nanofiltration externally [4]. MBRs are also reported to be effective in the treatment of leachate. In recent years, electrocoagulation (EC), which provides

removal of pollutants by the introduction of electric current to metal electrodes, has been used successfully in the treatment of leachate and wastewater of many different characteristics [5-11]. There is no need to add any coagulants or chemicals in the EC process and the cost is quite low compared to conventional systems. Several chemical and physical mechanisms occur during EC. The metal electrode used is dissolved by electrolysis. Coagulant species and hydroxides depending on ambient pH are formed which can coagulate and desaturate suspended particles or remove contaminants by flotation, adsorb contaminants or assist their precipitation. By EC treatment, a high rate of pollution removal is provided. However, after the EC process is carried out, large amounts of EC sludge are produced [12-13]. Formed sludge contains a high number of hydroxides ($M(OH)_3$) of metal electrodes and also shows dependence on the characteristics of the treated wastewater.

Recently, hybrid system submerged electromembrane bioreactor (SMEBR) has been prominent in

Corresponding Author: ka.gulizar@gmail.com (Gulizar Kurtoglu Akkaya)

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wastewater treatment. The SMEBR covers the simultaneous operation of the SMBR system and the EC system in a single reactor. The metal electrodes are immersed in the SMBR for EC to occur in the SMBR. Literature studies showed that by applying the electric field to the electrodes, a high rate of waste water treatment can be achieved by both MBR and EC working together [14-17], membrane clogging can be significantly reduced [18-20] and active sludge properties can be improved much more comparing with conventional MBR systems [21-22]. In the SMEBR, metal hydroxide sludge is produced both by biological treatment and by dissolving the metal electrode in EC systems. Accumulation of Al was observed in a study of Bani Melhem and Elektorowicz [23] MBR also allows coagulant to be added to improve activated sludge properties. These coagulants form metal containing precipitated sludge as in EC. It was stated that if the sludge remains in MBR systems for a long time, it can also accumulate in bacteria and inhibit nitrification [23].

In the literature, there is no study about the accumulation of the soluble metal electrode in the SMEBR or coagulant added externally to the MBR in the bioreactor. The aim of this study is to find the daily amount of Al accumulated in the SMEBR system and to determine the amount of Al that can be accumulated by statistical interpretation. For the first time, young leachate was treated with SMEBR and Al was determined by ICP-OES by sampling the waste activated sludge almost every day throughout the operation. The data obtained were modeled by appropriate tests.

2. MATERIALS AND METHODS

2.1. Experimental set up

Two reactors, SMEBR and SMBR, were installed (Fig 1) in Yildiz Technical University Research Laboratory. The reactors were made of plexiglass. Both reactors are 19.5 cm in diameter and 63 cm in height. The working volume of the reactors is 5 L. Operation conditions in reactors, hydraulic retention time (HRT) and organic loading rates (OLR) were 5 days and 13.2 kg COD m⁻³ day, respectively. In order to provide filtration, hollow fiber membranes were used. Hollow fiber membranes were obtained from the National Research Center on Membrane Technologies (MEM-TEK). The membrane pore diameter and the effective surface area are 0.4 μm and 0.05 m², respectively.

In the SMEBR, cylindrical aluminum (Al) anode and cathode electrodes are placed in the bioreactor. Perforated electrodes were used to ensure homogeneous mixing in the reactor. Perforation ratio of perforated anode and cathode electrodes were 73.3 and 15.6%, respectively and both electrodes were 1 mm thick. The distance between the electrodes was 5.4 cm. The Al electrodes were immersed into activated sludge in the SMEBR approximately 18 cm from the bottom. The electrodes were connected to the Direct Current (DC) power supply and the timer was used to deliver intermittent DC. The current density applied to the SMEBR was determined as 24 mA cm⁻² [24]. At the earliest stage of SMEBR lasted for 25 days, 180 s

electrical field per day during 5 days of HRT was applied to the activated sludge of young leachate [25]. Consequently, the applied daily electrical field was increased since no satisfactory difference was observed between SMEBR and SMBR in terms of treatment performance [25]. An electrical field of 360 s day⁻¹ was applied for another 25 days in the same current density with 12-h intervals during 5 days of HRT. Accordingly, the first 25-day period was named as Stage I and the second 25-day period was named as Stage II.

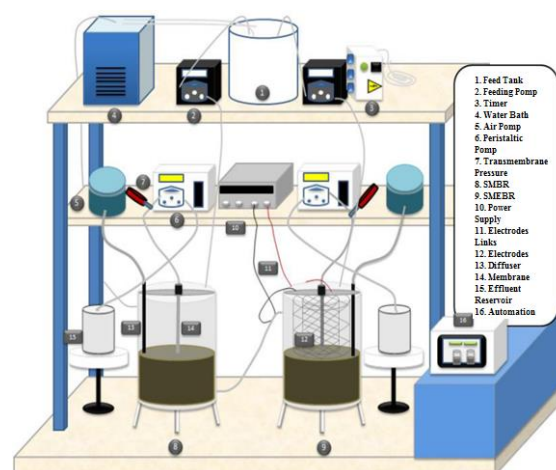


Fig 1. Schematic shown of SMEBR and SMBR [25]

2.2. Analytical procedure

Leachate was obtained from Leachate Treatment Plant in Odayeri Solid Waste Landfill Site located on the European side of Istanbul. The conductivity, TOC and Al values of the young leachate used were between 20.2-28.3 mS cm⁻¹, 17,000-23,000 mg L⁻¹ and 2.50-2.58, respectively. Conductivity in SMEBR and SMBR was measured using Termoscientific Orion 5 star. In both reactors, inlet and outlet wastewater samples were taken daily. The TOC concentrations of the reactors were analyzed using the HACH IL 550 TOC-TN instrument. TOC removal efficiencies were calculated using the degradation rate equation: Degradation Rate (%) = $(A_0 - A_1) / A_0 \times 100$ where A_0 and A_1 represent the initial initial and final percent of the parameter, respectively.

2.3. Statistical analysis and computational procedure

Statistical models were used for modeling the amount of Al accumulated in SMEBR and SMBR mathematically and to find out whether the accumulation amounts in both bioreactors differ from each other. For this purpose, MS Excel 2016 and Minitab Version 16.0 software were used on Windows 10 operating system. In all analyzes, the hypotheses were tested at 95% statistical significance level.

Firstly, univariate linear regression analysis was applied for mathematically modeling the amount of Al accumulation in both bioreactors. The relationship between two variables (Al accumulation and time) was measured by this analysis conducted using MS Excel

2016 Data Analysis module. In this relationship, time (measurement day) is an independent/descriptive variable where Al accumulation is dependent/explained variable. Following the simple linear regression, the relationship between the two variables was also visualized using the Graphing module of MS Excel 2016 (Fig 4 and Fig 5).

Mann-Whitney U test, one of the non-parametric statistical analysis methods, was used to see whether the amounts of Al accumulated in SMEBR and SMBR were significantly different from each other. The main reason for using this analysis method was that it was determined that the data obtained due to the insufficient sample numbers ($N = 25$) did not have a normal distribution according to the Shapiro-Wilk W test. Mann-Whitney U test (found by Henry Berthold Mann and Donald Ramson Whitney) is used to determine whether the mean values of the two data are equal. In this test, after obtaining two data sets, $x_i = \{x_1, x_2, \dots, x_n\}$ and $y_i = \{y_1, y_2, \dots, y_n\}$, the test statistics (U_x , U_y , and U) are calculated as:

$$U = \min(U_x, U_y) = \begin{cases} U_x = n_x n_y + \frac{n_x(n_x+1)}{2} - R_x \\ U_y = n_x n_y + \frac{n_y(n_y+1)}{2} - R_y \end{cases} \quad (1)$$

In the above equation, n_x represents the magnitude of the x data set and n_y represents the magnitude of the y data set, and R_x represents the corrected sum of the order data of x data set and R_y represents the corrected sum of the order data of y data set. The test is significant at the determined level of significance, if the observation is as $U \leq U_{critical}$; that is, the Zero Hypothesis (H_0 : There is no difference between the order of the two samples) is rejected and the Alternative Hypothesis (H_1 : There is a difference between the order of the two samples) is accepted.

In large samples (ie, $n_x > 20$ and $n_y > 20$), the U statistic has an almost Normal distribution, $N(\mu_U, \sigma_U)$, which can be represented as (mean and standard deviation values of μ_U and σ_U , U). In this case, the standardized z statistic ($z_{critical} = \pm 2.58$ at $\alpha = 0.01$ significance level for two-tailed test) can be calculated and interpreted as follows:

$$Z = \frac{U - \mu_U}{\sigma_U} = \frac{U - (n_x n_y)/2}{\sqrt{\frac{n_x n_y (n_x + n_y + 1)}{12}}} \rightarrow |z| = \begin{cases} |z| > z_{critical} \rightarrow p < \alpha \rightarrow H_1 \\ |z| < z_{critical} \rightarrow p > \alpha \rightarrow H_0 \end{cases} \quad (2)$$

In addition, Box-Plot graphs of Al values in two bioreactors were plotted to show descriptive statistics (eg minimum, first cartil (Q_1), medyn (Q_2), third cartil (Q_3), maximum) of the obtained data (Fig 7).

3. RESULTS AND DISCUSSION

3.1. Conductivity change

Conductivity is a numerical expression of the ability of an aqueous solution to conduct electricity. The conductivity of the water depends on the total and relative concentrations of ions, their mobility, their valence and the measurement temperature in the water. By measuring the conductivity of the water, the

number of ions in the water can be determined approximately. Leachate is a highly conductive wastewater. The active sludge conductivity value of the leachate in SMEBR and SMBR studies was determined to be less than 20 mS.

Fig 1 shows the conductivity values in SEMBR and SMBR. Conductivity in the SMEBR was measured at lower values than the SMBR system. Applying the electric field in the SMEBR reduces conductivity [26]. This reduction is due to charge neutralization. The ionic species consisting of total dissolved solids in the activated sludge coagulated with the positive ions obtained by dissolving the electrodes and the conductivity decreased with the reduction of the ionic species. Conductivity depends on many factors. pH in SMEBR and SMBR was 8.26-9.07, 8.54-9.0 and 8.5-9.00, 8.44-9.00; and the temperature varied between 21-27 °C, 21-27 °C and 21-30 °C, 22-29 °C in Stage I and Stage II, respectively. In Stage II, the amount of Al given to the water was higher than that of Stage I. As a result, the conductivity values in Stage II were lower than those in Stage I (Fig 2). The contribution of the electric current is evident here. Ilhan et al. [27] found similar situation in his study. Wang et al. [28] stated that the conductivity values decreased with the introduction of electrical current into the aqueous medium. Conductivity was affected by both temperature and pH changes, so no stable increase or decrease was recorded. The polarization of the electrodes occurred on days 8, 22 and 46 in the SMEBR, so the current could not be fully transmitted to the activated sludge as 26 mA cm⁻². Therefore, conductivity in SEMBR increased in the following days compared to SMBR. In addition, there are high amounts of Ca and Na salts in the leachate, and their accumulation over time can increase the conductivity in the SMEBR.

3.2. TOC change

Electrochemical treatment is carried out together with biological treatment for SMEBR treatment of leachate. When electric current is applied in the SMEBR, Al³⁺ is produced by electrooxidation of the aluminum anode. The electrolytic dissolution of the aluminum anode is first converted to Al(OH)₃ at convenient pH levels, Al³⁺ and Al(OH)₂ at low pH and finally to polymerized Al_n(OH)_{3n} [29]. Other components such as Al(OH)²⁺ and Al₂(OH)₂⁴⁺ are formed depending on the pH of the aqueous medium. While water oxidation produces hydrogen and oxygen gas in the anode, hydrogen gas and hydroxide in the cathode are produced by reducing water. In addition, oxidation produces a strong oxidizing hydroxyl radicals and forms of dehydrogenated and hydroxylated species can react with organic pollutants [30-33]. Accordingly, when leachate enters the anode and cathode from the region between the reactor wall and the anode, it is subjected to electrokinetic conditions and organic oxidation occurs. Thus, it contributes to TOC removal.

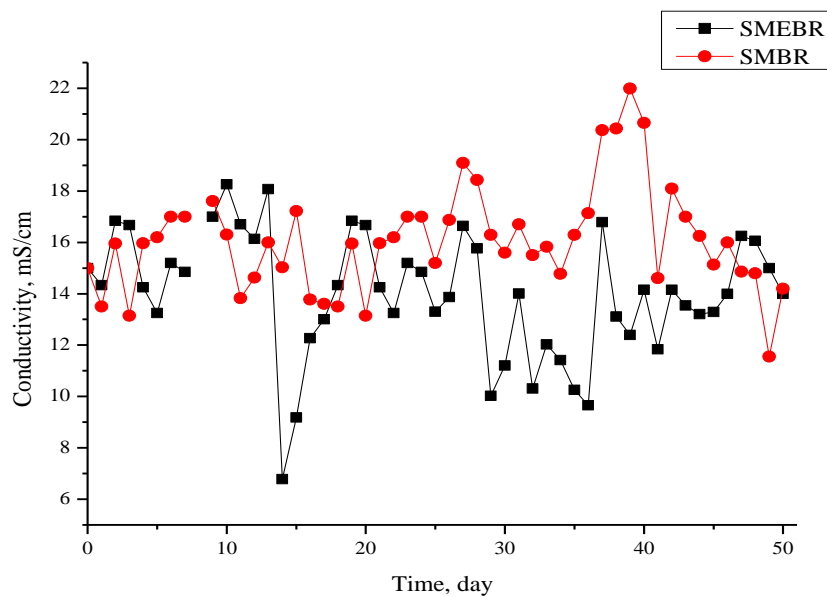


Fig 2. Conductivity change in SMEBR and SMBR

TOC removal efficiency is presented in Fig 3. Average TOC removal in Periot I was 89, 88% for SMEBR and SMBR. In Stage II, TOC removal efficiency in SMEBR was increased 7% with increasing electric current compared to SMBR. Feng et al. [34] found that the application of electric current increases TOC removal. There was a reduction in TOC removal efficiency from 86% to 81% in 20-22nd days of Stage I. This was because the electrical current cannot be transmitted as 24 mA cm⁻². Electrodes were taken out of the reactor and checked and it was observed that the electrodes were passivated. Physical washing was performed

under the tap water with the help of sponge, but the biofilms on the electrodes could not be removed. The electrodes were then soaked in 1% HCl acid for 1 hour followed by physical washing. Electrical current could not be introduced to the SMEBR for 3 days. In order to obtain more treatment efficiency by SMEBR compared to SMBR in the treatment of young leachate, the applied electric field exposure time has been doubled and the reactors were operated for a further 25 days (Stage II). On day 25, the electrodes were reintroduced into the SMEBR. Passivation in SEMBR on days 31, 32 and 42 in Stage II affected TOC removal.

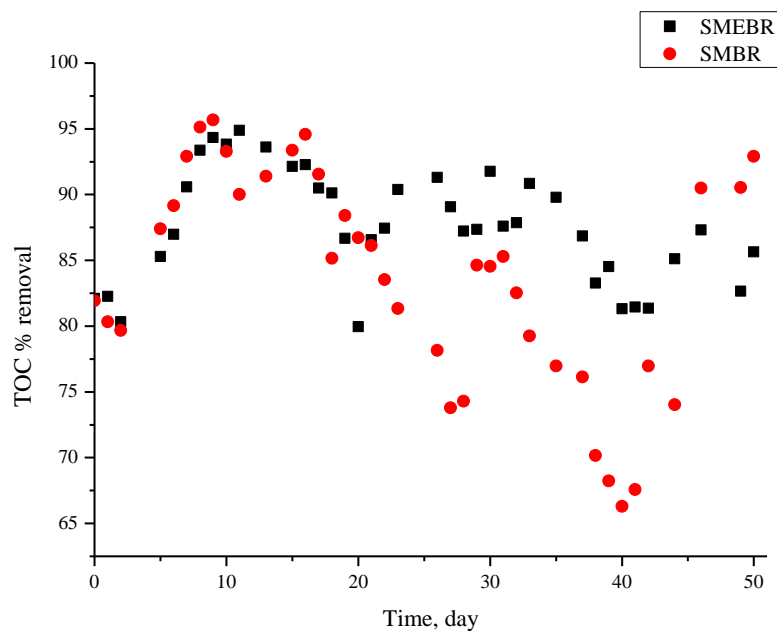


Fig 3. TOC change in SMEBR and SMBR

3.3. Statistical analysis

3.3.1. Modelling of Al accumulation in SMEBR and SMBR

SMEBR is an effective treatment method for wastewater treatment. With the dissolution of metal electrodes in the SMEBR, the pollution removal efficiency is higher than in other treatment processes. However, dissolving electrodes in the SMEBR accumulate in the activated sludge as well. There is uncertainty as to how much electrodes will accumulate

during the operation of reactors. This study was conducted to find out how much Al accumulates daily and to estimate how much it can accumulate in longer operations. Firstly, waste activated sludge samples were taken from SMEBR and SMBR and Al values were plotted using MS Excel. These drawings are given in Fig 4. As young leachate is a difficult wastewater, occasional operational problems were observed; sludge samples could not be taken because of sludge loss due to the sludge swelling and overflowing and Al determination could not be conducted.

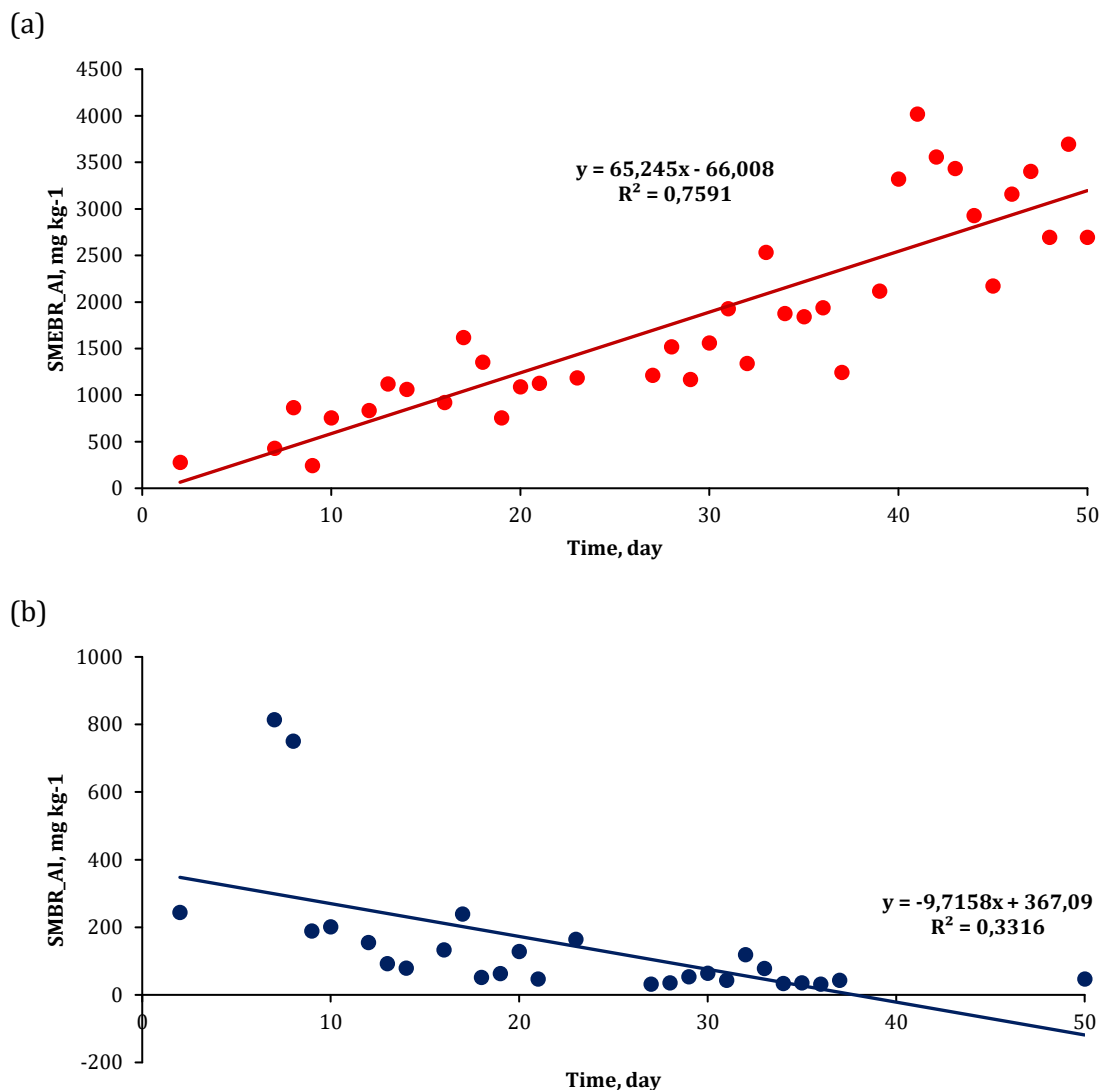


Fig 4. Change of Al over time in waste activated sludge in SMEBR (a) and SMBR (b)

In the SMEBR, coagulant agents Al ions are produced by electrooxidation of the Al electrode in situ in the activated sludge media when the DC electric field is applied to the leachate. Activated sludge pH varied between 8.01-9.02 in SMEBR. It can be said that Al(OH)₃ and polymerized Al(OH)_{3n} precipitated in this pH range. The phosphorus value in the leachate is TP=10.2-12.0 mg L⁻¹. SMEBR and SMBR effluent TP values were less than 0.1 mg L⁻¹. Therefore, Al reacts

with phosphate ions to form AlPO₄ in the reactors and precipitation occurs.

In the SMBR, Al influent in activated sludge is only possible through leachate. Al reactions occurring in the SMEBR also occur in the SMBR. However, Al was found in the waste activated sludge in the range of 0.03-0.82 g kg⁻¹ since there was no external Al introduction.

The sludge retention time in SMEBR and SMBR is approximately 15 days. The sludge retention time was

checked regularly. Operating problems such as sludge swelling or overflow occurred in the SMBR. The sludge retention time and daily discarded sludge amount could not be kept stable. Therefore, Al increased on days 6, 15, 19, 22 and 31. On days 40 and 50, the amount of Al was less than 0.0 g kg⁻¹. In SMEBR, sludge retention time remained stable compared to SMBR. The SMEBR had less sludge swelling/overflow problems comparing to SMBR. In a study, it was stated that Al accumulation prevents sludge swelling [35]. Al accumulation in the SMEBR turned out as an advantage over SMBR in the treatment of young leachate. Therefore, Al accumulation is related to sludge retention time. In the SMEBR, Al amount increased by accumulation in the reactor since more Al dissolved in Stage II over Stage I and all Al could not be discarded in the daily sludge.

Al accumulation in waste activated sludge was observed in SMEBR and SMBR for 50 days. In order to estimate how much Al will accumulate in SMEBR for more than 50 days operated processes, time-dependent change of Al values is modeled by regression analysis and the following equations are obtained:

$$Al \text{ (SMEBR)} = -66.008 + 65.245 \cdot (\text{day}) \tag{3}$$

$$Al \text{ (SMBR)} = 367.09 - 9.7158 \cdot (\text{day}) \tag{4}$$

As can be seen in Fig 4 and the above equations, an increasing Al accumulation occurred over time in the SMEBR, whereas the opposite is depicted in the SMBR, which decreased over time. Al accumulation in the SMEBR bioreactor indicated by (3) is statistically significant at 95% significance level. In addition, the R² value of simple linear regression is 0.75; there is also a strong correlation between the two variables (R² = 0.7591).

On the other hand, the accumulation of Al in the SMBR reactor does not give a linear appearance as in the SMEBR. For this reason, different functional transformations (eg logarithmic, exponential, etc.) have been tried and the most suitable transformation is found to be 1/x. When the change of 1/Al value over time is modeled, the following graph and equation are found:

$$1/Al \text{ (SMBR)} = 0.0008 + 0.0006 \cdot (\text{day}) \tag{5}$$

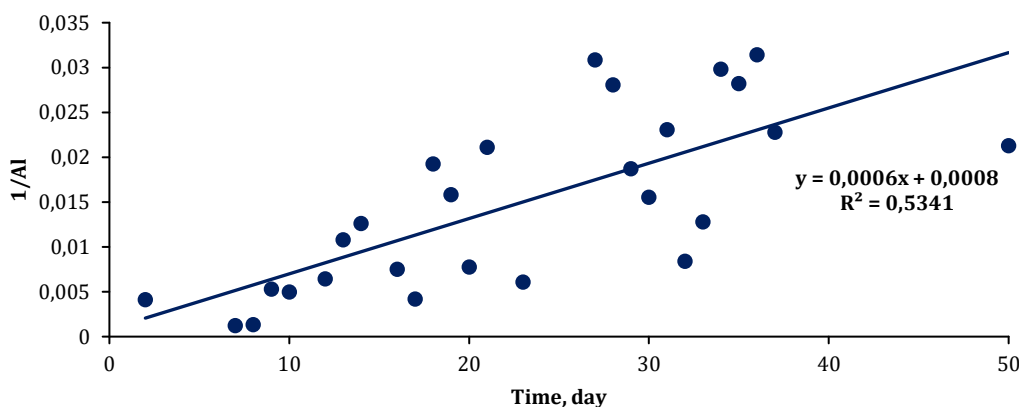


Fig 5. Variation of 1/Al value over time in SMBR

The correlation of 1/Al value over time is moderate (neither strong nor weak) (R² = 0.5341) and equation (5) explains Al accumulation in SMBR more accurately than linear equation (4) (R² value of linear equation is only 0.3316). Therefore, if it is desired to estimate the accumulation of Al values in SMBR over time, using equation (5) will provide more accurate results. This time, it can be deduced that the accumulation of Al in the SMBR decreases with time since not the Al value but the value of 1/Al will increase in direct proportion over the day variable.

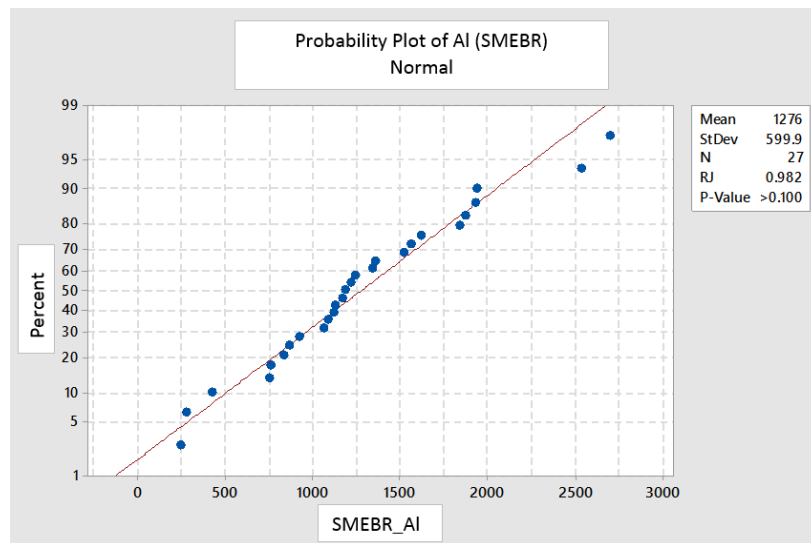
3.3.2. Statistical Comparison of SMBR and SMEBR

Not only were the relation of Al accumulations in SMEBR and SMBR over time measured but also statistical differences were also measured. For this purpose, Al data obtained from both reactors on the same days were used. Since the size of this data set is only 27, the normal distribution assumption would not be appropriate. As a matter of fact, when the Shaphiro-Wilk test was applied, Al (SMBR) values do not have normal distribution (p < 0.1) (Fig 6).

Since one of the data sets does not have a normal distribution, it would be more appropriate to perform non-parametric tests. Thus, Mann-Whitney U test was applied to Al values data sets obtained from two reactors using Minitab software. The result is as follows:

| | N(sample size) | Median |
|---|----------------|--------|
| SMBR | 27 | 78.1 |
| SMEBR | 27 | 1183.3 |
| Point estimate for η1 - η2 is -1083.5 | | |
| 95.1 Percent CI for η1 - η2 is (-1285.0,-887.7) | | |
| W = 387.0 | | |
| Test of η1 = η2 vs η1 ≠ η2 is significant at 0.0000 | | |

(a)



(b)

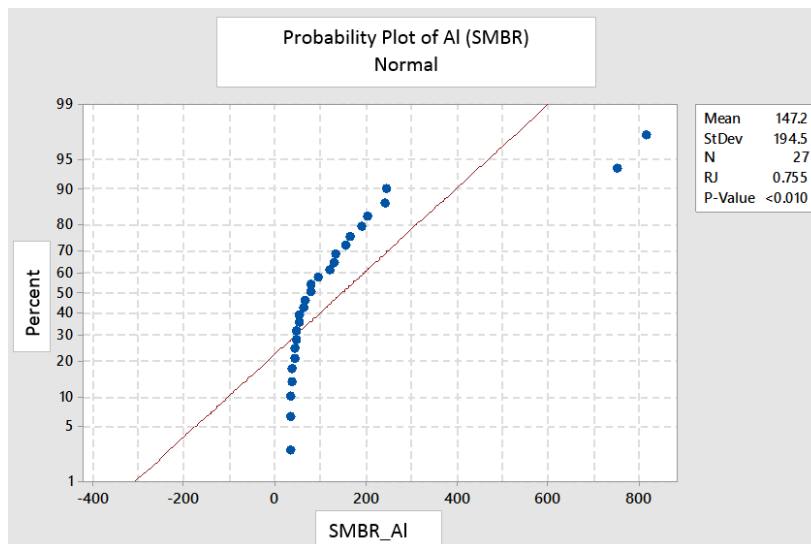


Fig 6. Probability plot representation of SMEBR (a) and SMBR (b) AI values

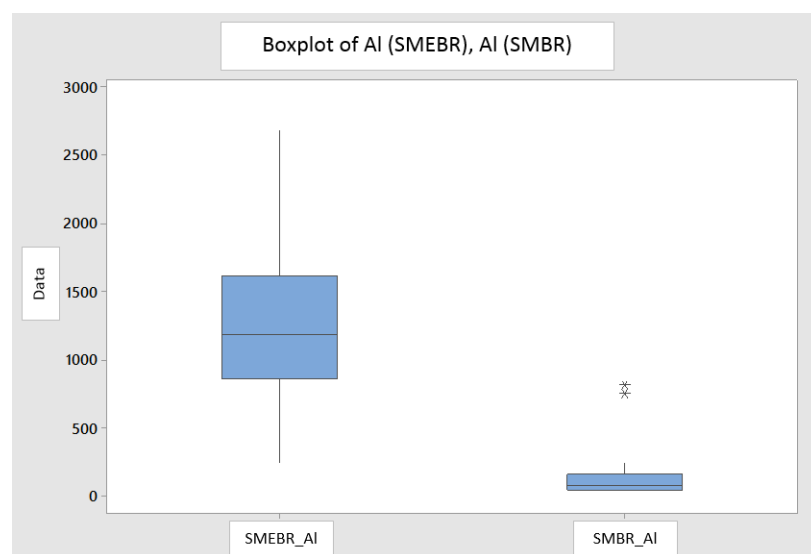


Fig 7. Display of SMEBR and SMBR AI values in Boxplot

In summary, the two data sets are significantly different (99% significance level). Al accumulation rate in SMEBR (median value = 1183.3) is significantly higher than Al deposition rate in SMBR (median value = 78.1). The distribution of data from both reactors is also summarized in the boxplot graph below. As can be seen, both the median and the maximum values of the Al accumulation in the SMBR are much lower than the Al accumulation in the SEMBR.

3.3.3. Relation of Al accumulation with conductivity and TOC values in SMEBR and SMBR

The relationship between accumulated Al amounts in SMEBR and SMBR, and conductivity and TOC% values was examined. The relationship between the data from the SMEBR reactor is as in the following Table 1.

As is known, the correlation coefficient r is between -1 and 1. It can be said that if the value of r approaches -1, there is a stronger negative correlation, and if it approaches +1, there is a stronger positive correlation between the two variables. At values close to 0, the correlation between the two variables is also small. In this case, a negative but very poor relationship between Al accumulation and conductivity is observed according to Table 1. There is also a negative, moderate relationship between Al and TOC; it is neither too strong nor weak.

Table 1. Correlation coefficients between Al accumulation and other parameters in SMEBR

| Parameter | Al |
|--------------|----------|
| Conductivity | -0.17712 |
| TOC% | -0.54366 |

There is also a negative, moderate relationship between Al and TOC; it is neither too strong nor weak.

According to the data obtained from SMBR reactor, the correlation coefficients between these variables are as follows (Table 2):

Table 2. Correlation coefficients between Al accumulation and other parameters in SMBR

| Parameter | Al |
|--------------|----------|
| Conductivity | 0.148134 |
| TOC% | 0.375518 |

According to Table 2, there is a positive correlation between Al and other variables. However, the relationship between Al and conductivity is very weak. There is a weak-moderate correlation between Al and TOC%. It can be said that Al accumulation negatively affects the variables, even though it has a weak correlation with these variables. Because it is stated that Al accumulation in biological systems affects nitrification bacteria. With this effect, conductivity can lead to a decrease in TOC% values.

3.4. Impact of Al Accumulation on SMEBR and Environment

Electrocoagulation, chemical coagulation or the addition of appropriate coagulant to any point of the treatment process are the processes performed to improve the quality of treated water. However, metal-containing sludges are formed in these processes. Also, for the SMEBR, the amount of Al increases day by day with the electric current applied to the activated sludge. Bani Melhem and Maria [23] stated that nitrification bacteria were affected by the accumulation of Fe electrode used in SMEBR system. Al coagulants added to the activated sludge system to improve sludge properties have been observed to affect nitrified bacteria [23, 36]. In the treatment of young leachate by SMEBR, Al accumulated in the reactor had no significant effect on nitrification. On the contrary, it reduced sludge swelling/foaming in the SMEBR. No studies have been found in the literature on the assessment of Al accumulated in waste SMEBR sludge. However, in one study, aluminum sulfate was added to MBR. Al sludge was removed from MBR by adsorption [37]. In addition, studies were carried out for the elimination of metal-containing sludges obtained from drinking water treatment, electrocoagulation/coagulation wastewater treatment and coagulant added treatment processes. Okuda et al. [38] used sludge with metal content in plant growing in their study. For this purpose, Al amount was reduced by applying extraction pre-treatment on Al sludge.

Many scientists produced adsorbents by reusing metal sludges [39-41]. They used them again in wastewater treatment and removal of heavy metals (Hg, Pb, As, Cr, Cu) [40, 42, 43]. Recycling of sludge reduces the cost of coagulant metals used in wastewater treatment plants [39]. Al is also a very common metal in the crust. Therefore, it is not possible to adjust the limits in the soil. Al-containing sludge has also been used as soil conditioner or fertilizer [44], [45]. The most important parameter in these applications is soil pH. Depending on the pH of the soil, Al may cause toxic effects on the soil. This negatively affects plant growth [46-48].

As a result, the waste sludge obtained from the SMEBR can be easily applied to the soil by controlling the soil pH and can be used as an adsorbent. It is therefore that the Al-containing sludge to be obtained in the SMEBR will not pose a problem in the treatment of leachate.

4. CONCLUSIONS

In this article, the Al accumulation in treatment of leachate with high organic content by SMEBR was evaluated by statistical studies such as regression analysis and Mann Whitney U test. Mathematical modeling has been performed to estimate how much Al accumulation will occur if the SMEBR is operated for more than 50 days. Al accumulation had no significant effect on the SMEBR. Therefore, the use of the SMEBR system in leachate treatment is advantageous over SMBR. This superiority of SMEBR over SMBR has not been discussed in the literature before, and our study makes an important contribution in this respect.

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