



Research Article / Araştırma Makalesi

RESEARCH OF LEACHATE TREATABILITY WITH BIOLOGICAL, PHYSICAL, CHEMICAL AND MEMBRANE BIOREACTOR PROCESSES**Şahan DEDE*¹, Arzu ULUTAŞ¹, S. Kadri YİĞİT²**¹İZAYDAŞ, Hazardous Waste Incineration Plant, KOCAELİ²Department of Mechanical Engineering, University of Kocaeli, KOCAELİ

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ABSTRACT

One of the disposal methods of household wastes is landfill. In our country, 1 kg per person and totally 30 million tons household waste per day is produced. It is the responsibility of the municipalities to carry out collection and disposal of household wastes and there are 79 landfill areas present in our country. In terms of environment pollution, the most important subject is treatment of leachate at landfills. Leachate consists many of pollutes that arise from the contents of the household waste is formed with biological reactions when landfilling, water content of the waste and rain water coming from outside environment to the storage site. Collection and treatment of leachate is important due to its content of high amount organic substances, nitrogenous compounds, heavy metals, chlorinated organic and inorganic salts and its potential to cause pollution to soil and underground waters. The characteristics and quantity of leachate varies for every landfill and depend on content of household waste, climate conditions, structure and depth of landfill area, structure of surface cover and operation properties. Leachate is the most difficult waste water for treatment because of high contaminant properties. In this respect, generally, a combination of physical, chemical, biological methods and adsorption and membrane technologies from advanced treatment methods are used for leachate treatment. In this study, İZAYDAŞ Solid Waste Landfill Area where Kocaeli province's solid waste is stored, leachate treatability with biological-chemical and submerged MBR processes is utilized and compared with full scale MBR Treatment Plant treatment efficiency and all these have been evaluated. Prototypes are not laboratory scale, but in the size of a small treatment facility and this way, it is aimed to provide close to real results and in addition, operational problems are identified.

Keywords: Threatment, Leachate, landfilling, membrane bioreactor.**ÇÖP SIZINTI SULARININ BİYOLOJİK, FİZİKSEL, KİMYASAL VE MEMBRAN BİYOREAKTÖR PROSESİ İLE ARITILABİLİRLİĞİNİN İNCELENMESİ****ÖZ**

Evsel atıkların bertaraf yöntemlerinden birisi atıkların düzenli depolama sahalarına alınmasıdır. Ülkemizde kişi başı günde 1 kg, toplamda 30 milyon ton evsel çöp ortaya çıkmaktadır. Evsel çöplerin toplanması ve bertarafı belediyelerin sorumluluğunda olup ülkemizde halen 79 adet katı atık düzenli depolama sahası bulunmaktadır. Çöp depolama sahalarında çevre kirliliği açısından en önemli konu, oluşan çöp sızıntı sularının arıtılmasıdır. Evsel çöpün içeriğinden kaynaklanan çok sayıda kirlileti parametreyi bünyesinde bulunduran çöp sızıntı suları, çöpün depolanması ile oluşan biyolojik reaksiyonlar, çöpün içinde bulunan suyun ve dışarıdan depolama sahasına giren yağışla oluşmaktadır. Çöp sızıntı suları, içerdikleri yüksek miktardaki organik maddeler, azotlu bileşikler, ağır metaller, klorlanmış organik ve inorganik tuzlardan dolayı toprak ve yer altı sularının kirlenmesine neden olduğundan bu sularının toplanması ve arıtılması önem arz etmektedir. Çöp sızıntı sularının karakteri ve miktarı, her bir depolama sahası için farklılıklar göstermektedir ve evsel atığın içeriğine, iklim, depo sahasının yapısına, deponun derinliğine, yüzey örtüsünün yapısına ve işletme özelliklerine bağlıdır. Sızıntı suları, yüksek kirlileti özelliği nedeniyle, arıtımı en zor atık suların başında gelmektedir. Bunun için sızıntı sularının arıtımında genellikle fiziksel, kimyasal ve biyolojik metotların kombinasyonu, ileri arıtma metotlarında ise adsorpsiyon ve membran teknolojileri kullanılmaktadır. Bu çalışmada, Kocaeli ili evsel atıklarının depolandığı İZAYDAŞ Katı Atık Düzenli Depolama Alanlarından kaynaklanan çöp sızıntı sularının biyolojik-kimyasal ve MBR batık membran sistemi ile arıtılabilirliği değerlendirilmiş ve gerçek boyutta kurulan MBR Arıtma Tesisinin proses arıtma verimleri karşılaştırılmıştır. Prototipler laboratuvar ölçekli olmayıp küçük bir arıtma tesisi büyüklüğünde yapılarak sonuçların gerçeğe daha yakın alınması hedeflenmiş, ayrıca işletmesinde yaşanabilecek problemler tespit edilmiştir.

Anahtar Sözcükler: Arıtma, çöp sızıntı suyu, düzenli depolama, membran biyoreaktör.

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1. INTRODUCTION

Regular stacking is one of the most important methods for disposal of the household wastes. Until the 2000s, many countries used landfill stacking method for disposal of household wastes (Lema et al., 1988). However, recently, the methods other than landfill stacking have found implementation areas as well. When the current applications existing in the world are examined, in disposal of household wastes;

Recycling processes,
Regular Landfill,
Biological Processes,
and Thermal disposal methods are observed to be applied.

In our country, regular landfill disposal is the chosen method but in especially recent years, in especially Istanbul and Kocaeli, thermal disposal method have been considered. The climate of the city, geological structure, population and economic situation are main factors. However, it can be said that the most commonly used method is still regular landfill. The biggest disadvantage of this method is the need of space and treatment requirement of the waste leachate.

The leaking water at solid waste storage sites cause soil pollution as well as pollution to underground waters due to their dark colored heavy smelly content having high levels of organic materials (fatty acids and humic acids), macro inorganic substances (Ca, Mg, Na, K, NH_4^+ , Fe, Mn, Cl⁻, SO_4^{2-}), nitrogenous substances, heavy metals (Cd, Cr, Cu, Pb, Ni, Zn), chlorinated organic and inorganic salts and even at low concentrations, aromatic hydrocarbons, phenols and pesticides (Wang vd., 2002; Morawe vd., 1995, Wagner vd., 2013). The solid wastes in the regular landfill area undergoes a series of physical, chemical and biological decays from high molecule weight components to simpler components. The rainwater and the water in the content of the Landfill go through the solid wastes, passes through the filling in the direction of the hydraulic inclination, therefore cause leachate formation and movement. It is of great importance to carry out treatment since this leachate water cause pollution in case it reaches to the soil or receiving environment. The polychlorinated biphenyls (PCB) and heavy metals that may be available in the content of leachate are very dangerous to human health (Morawe et al., 1995).

Landfill leachate is evaluated in terms of parameters such as biological oxygen demand (BOD), chemical oxygen demand (COD), BOD/COD ratio, suspended solid material, pH, ammonia nitrogen ($\text{NH}_3\text{-N}$), total kjeldahl nitrogen (TKN) and heavy metals. Even though leachate water composition shows differences in stabilization phases, they are usually examined under three different categories; young, medium and old waste. (Renou et al., 2008). The leachate water contains many components and its quality varies. By checking the leachate water quality, important information can be learned regarding to stabilization situation and age of the waste.

The amount of solid waste regular landfill facility leachate water depends on permeability value of final top cover layer, climate conditions, solid waste composition, landfill age etc. factors. The composition of the leachate water composition varies depending on the content of the solid waste, pH, redox potential, climate conditions and landfill age. Aside from organic and inorganic ions and metals, the leachate waters may contain micro contaminants as well. The age of the deposit is one of the most important factors effecting the characteristics of the leachate water. With the increasing age of landfill, biological decomposition becomes completed and the ratio of easily biodegradable organic materials fall. For this reason, in young landfills, $\text{BOD/COD} < 0.5$ while in old landfill leachate waters, $\text{BOD/COD} < 0.2$ applies (Oztürk, 1999, 2010).

The challenges in biological treatment of Landfill leachate waters are high COD, high BOD/COD ratio, ammonium content, toxic chemical features, presence of toxic chemicals like heavy metal ions etc. (Park et al., 2001; Kargi and Pamukoglu, 2003). The methods developed for treatment of leachate water are physical, chemical, biological and advanced treatment methods. Biological methods are aerobic and anaerobic methods and they get rid of organic materials and

ammonia nitrogen. They work very efficiently for young storage areas especially. In case BOD/COD is high, this efficiency applies and when this BOD/COD ratio decreases, the efficiency decreases as well. It is very hard to achieve a high level of treatment and output water quality by using one of only any of these methods. For this reason, in treatment of leachate waters, generally a combination of physical, chemical and biological methods are used while in advanced treatment methods, adsorption and membrane technologies are used (Yalılı et al., 2006; Kargi et al., 2003; Rodrigues et al., 2004; Trebouet et al., 2001; Wiszniowski et al., 2006; Amokrane et al., 1997; Bohdziewicz et al., 2001; Vogelphl et al., 1995).

The chemical methods used in treatment of leachate waters are a combination of coagulation-flocculation (Amokrane et al., 1997), chemical deposition and chemical-electrochemical oxidations (Chiang et al., 2001); and on the other hand, biological methods are a combination of aerobic, anaerobic and anoxic processes. Physico-chemical methods are usually used with biological methods in order to remove the materials which cannot be removed with biological methods (Bohdziewicz et al., 2001; Ahn et al., 2002).

Table 1. Physical, chemical and biological processes used in treatment of leachate

	Purification Processes	Purpose
Physics	Deposition/flotation	Suspended Solid substance removal
	Filtration	
	Stripping with air	Amonnia and volatile organic compounds removal
	Adsorption	Organic matter removal
	Ion exchange	Dissolved inorganic matter removal
	Reverse Osmosis	Organic and inorganic matter removal
	Evaporation / incineration	Reverse osmosis concentrate disposal
Chemical	Neutralization	pH control
	Chemical precipitation	Treatment of heavy metal and some anions
	Coagulation / flocculation	Non-sedimenting suspended material removal
	Chemical oxidation	Organic matter removal, detoxification
Biological	Active mud	
	Sequencing batch reactors	
	Aerated lagoon / stabilization ponds	
	Biofilm systems (trickling filter, rotating biological discs)	Organic carbon removal
	Anaerobic lagoon and contact tanks	
	Anaerobic (upflow mud bed, filter or hybrid) reactors	
	Nitrification/denitrification	Nitrogen removal

Due to the organic structures not easily degrading, in old Landfill landfill areas, biological decomposition of leachate waters is hard. For this reason, it is deemed necessary to use physical and chemical methods together with biological treatment (Robinson and Grantham, 1998). Chemical coagulation and carbon adsorption are rather popular processes but their organic removal performance are not at desired high levels (Loizidou et al., 1992; Tchobanoglous et al., 1993). Treatment methods often preferred are summarized in Table 1.

Solid waste leachate water contains significant amounts of stubborn, resistant, hard to process organic contaminants such as humic acid and fulvic acid etc. (Cortez et al., 2010). These stubborn organic matters can not be dissolved using the conventional biological methods commonly used in household solid waste treatment facilities. For this reason, especially in recent years, ultrafiltration, nanofiltration and reverse osmosis membrane technologies are increasingly used.

Even though for household wastes, generally, in treatment of surface waters, usage of membrane is efficient but in Landfill leachate treatment, membrane usage is not applied before biological treatment is completed. In the Landfill leachate water with high contamination, first, biological treatment shall be provided, then, depending on the output water quality discharge standards, membrane usage shall be applied. For the discharge of these wastewaters, denitrification and phosphorous removal shall be applied. At the next stage, color removal, oxidification of heavy metal and organic microcontaminants, active carbon, coagulation or nanofiltration systems are required.

IZAYDAS Solaklar Municipal Landfill Facilities have been storing wastes since year 1997. At the end of year 2015, in the Household Waste Storage Areas, in total, 5.700.000 tons of waste storage have been carried out. For choosing the best process for the Landfill leachate water occurring in IZAYDAS storage site, studies have been carried out, chemical+biological treatment, reverse osmosis+evaporation, electrocoagulation and MBR processes have been considered and by establishing prototypes of some processes, trial studies have been carried out. Resulting from these studies, taking the process investment cost, operation cost, output waste water quality achievement, ease of operation and the area needed, process choosing have been carried out. It has been decided that for the treatment of Landfill leachate in IZAYDAS, the most suitable process is membrane bioreactor establishment and construction of this facility have been commenced and at the beginning of the year 2015, it has been put into operation.

2. TREATABILITY STUDIES IN PROCESS SELECTION

Regarding to treatment of waste leachate waters, prototypes of some processes have been established in the site and treatability studies have been carried out. These studies are biological + chemical treatment and submerged membrane system MBR process.

2.1. Treatability Studies with Biological-Chemical Methods

In biological-chemical treatment system, a metal tank with aerobic living conditions with aeration with 30 m³ volume and in chemical unit, each with 1 m³ volume; lime, HCl and polyelectrolyte dosing PVC based tanks are used. The prototype process have been designed with 5m³/h flow. The Landfill leachate water is taken into biological tank first, then to the chemical treatment unit. The biological part is equipped with oxygen analyzer, chemical unit tanks and pH measurement sensors. The amount of water pumped to the system is controlled with flowmeter. The process flow chart is shown in Figure 1. and prototype facility general view is shown in Figure 2.

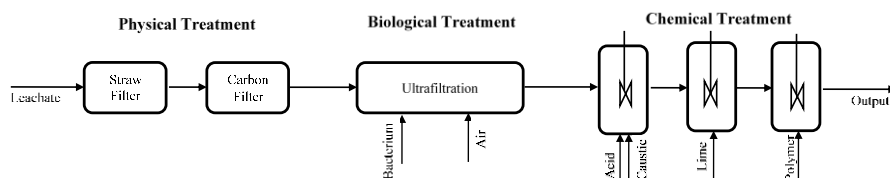


Figure 1. Biological-chemical treatment prototype facility flow chart



Figure 2. Biological-chemical treatment prototype facility appearance

The trial operation have been carried out within a 30 days time period. In the first 15 days, the biological part have been operated and after this, the chemical treatment was taken into operation. Landfill leachate water entry average COD value was about 7500 mg/l, SS entry value was approximately 500 mg/l. As it can be seen from the graph in Figure 3, after 15th day of the trial operation, the output values have been stabilized in an order and COD values dropped under 1000 mg/l but suspended solid values only fell below the desired value of 350 mg/l in 5 days.

With this method, the treatability of leachate water did not reach to the desired level. Resulting from the study, it has been understood that biological treatment is not sufficient for treating leachate water, that the output values can be taken to a specific level when used with chemical treatment, however, due to raise of pH values, it is required to lower the pH again, that suspended solid could be lowered only at the end of the studies.

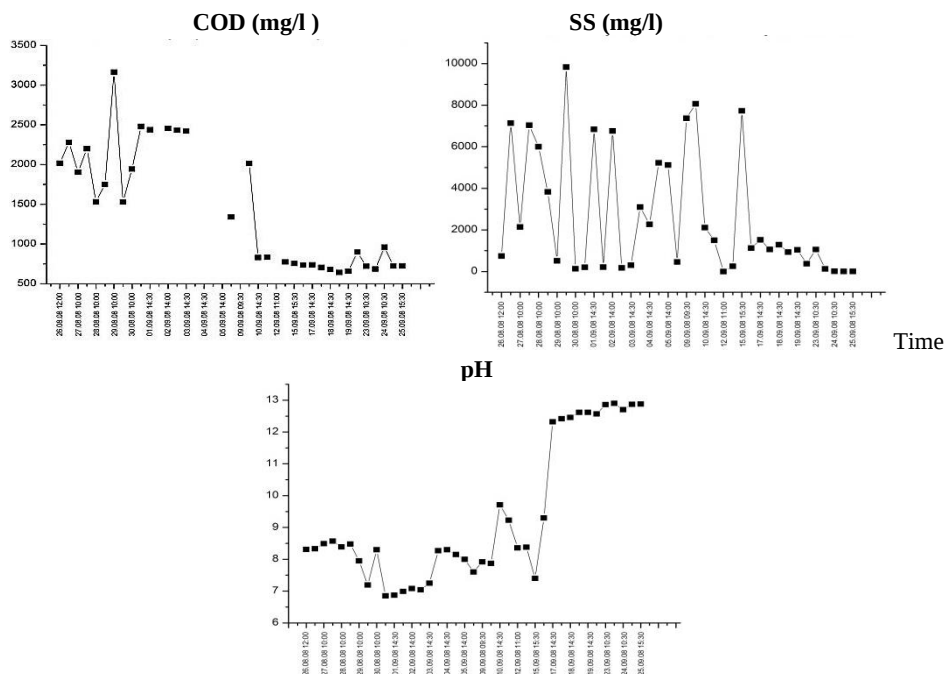


Figure 3. Biological-chemical treatment prototype facility performance graphs

2.2. Submerged Membrane Type Membrane Bioreactor Process Trial

In treatability of Landfill water, usage of other trial study of membrane bioreactor (MBR) process have been carried out. With the prototype established in the site, with 1m³/day flow, water treatment have been carried out and the results have been evaluated. In this study, a process with 5 m³ biological part active, in a tank with ventilation and submerged membrane used in it was chosen. The Landfill water was initially taken to an anoxic tank of 0.5 m³ volume, then from the biological section, flow over membrane have been carried out. The trial study have been carried out for 2.5 months, over 15 days and four different stage, the analyses were evaluated. In the first 15 days period, results below COD 500 mg/l and SS 10 mg/l have been reached. In the second period trials, COD values have been around 2000 mg/l and in this process, SS was not examined. In the third period, the membrane output values have been showing differences, SS values have been below 10 mg/l. In the last 15 days period, it has been observed that the MBR output values are at the levels desired. The process flow chart is shown in Figure 4. and prototype facility general view is shown in Figure 5.

When the graphs in Figure 6 are examined, observing different results in the processes and observing peak values in some days are due to clogging of the submerged membrane. When evaluating in general, it has been observed that after biological treatment, Landfill water can be treated with membrane process with this experimental study.

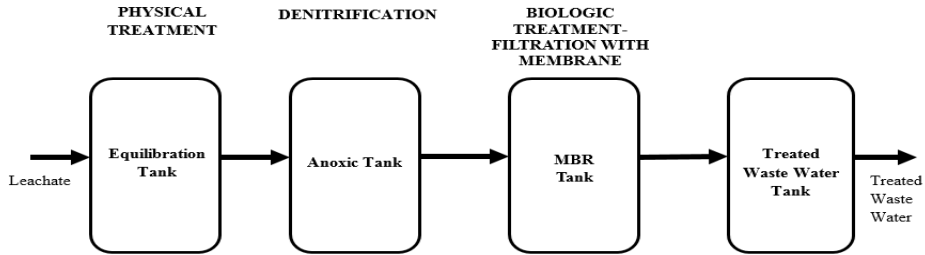


Figure 4. Membrane Bioreactor Prototype Facility flow chart



Figure 5. MBR Prototype Facility General Appearance

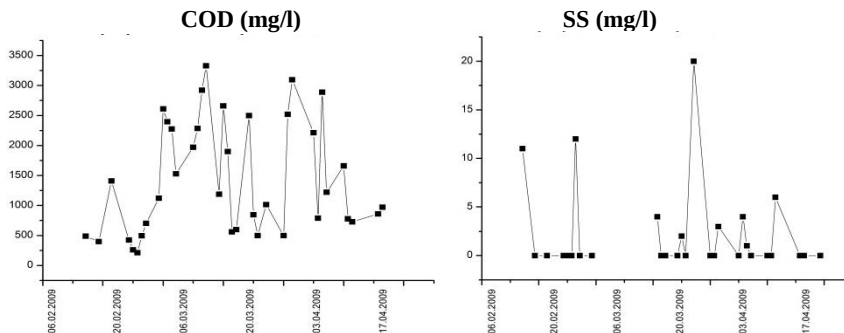


Figure 6. Analysis result graphs of MBR Prototype Facility

3. LANDFILL LEACHATE WATER TREATMENT FACILITY

When the prototype studies, literature evaluations and reference MBR applications are taken into consideration, it has been decided that for the treatment of Landfill leachate waters that will occur in storage of Kocaeli household wastes, the most suitable process is bioreactor (MBR) process. In 2014, MBR Landfill Leachate Water Treatment facility was taken into operation, the desired discharge values have been achieved and treatment of Landfill leachate water have been carried out ever since. The Landfill Water Treatment Facility have been projected with 500

m³/day capacity. The output water of the facility is given to Izmit Water and Sewerage Administration (ISU) sewer line and for this reason, the projecting have been carried out taking the parameters and limit values specified in ISU Wastewater Sewerage Discharge Regulation. As treatment process, the combination of Membrane Bioreactor (MBR) and Nanofiltration (NF) have been chosen. Resulting from the process, the mud and nanofiltration waste with biological characteristics are disposed in the concentrated regular landfill areas. The treated water coming from the facility is given to the ISU sewerage line via the collector line. The process main flowchart is shown in Figure 7 and the facility general appearance is shown in Figure 8.

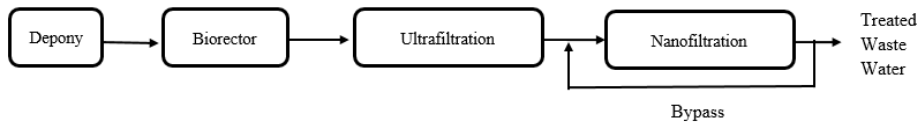


Figure 7.Facility flow chart



Figure 8. Landfill Leachate Water Treatment Facility General Appearance

2. The Landfill water entry parameters taken as basis in projecting the facility are given in Table

Table 2.Landfill leachate water entry parameters

Parameter	Unit	Low	Medium	High
Chemical Oxygen Need (COD)	mg/l		15000	
Biochemical Oxygen Demand (BOD)	mg/l	1000	7500	10000
Inert COD	mg/l		1500	
Suspended Solid (SS)	mg/l		1000	
Total Kjeldahl Nitrogen (TKN)	mg/l	800	1650	1800
Ammonia Nitrogen (NH ₃ -N)	mg/l	650	1500	1750
Total Phosphor	mg/l	0.21	10	24
Alkalinity (CaCO ₃)	mg/l	6800	8100	8500

The facility consists of; physical pre-treatment, balancing pool, anoxic-aerobic pool, ultrafiltration-nanofiltration and mud dewatering sections. The appearances of the facility main sections are shown in Figure 9.



Aerated sand-oil trapping balancing and biological pools UF membranes

Figure 9.Facility main sections

In the biological treatment pool, Chemical Oxygen Demand (COD) and nitrogen removal of leachate water are carried out. In the aerobic section, the air provided by blowers are distributed with diffusers and oxygen for the life of bacteria is being provided. In this section, with the help of the bacteria, with the nitrification of the ammonia nitrogen is converted to nitrate and nitrite. In the anoxic section, aeration is not applied, with denitrification, nitrate is converted to nitrogen gas.

For the separation of active mud- treated wastewater, at the biological treatment output, the ultrafiltration membranes are located instead of sedimentation pools. UF membranes are in hydrophilic pipe shaped, made of polyvinyl fluoride, in asymmetric structure and with 30 nm average pore size. It has been designed as two modules. According to treatment capacity, modules are operated and this way, when cleaning for the membranes is required, the other module is taken into operation and this way, the continuity of operation is provided. The flow rate of membranes are constantly monitored and in case it decreases, chemical washing and cleaning processes are applied.

Due to refractory COD, in order to provide the desired COD at the output, after UF, nanofiltration unit with 250 m³/day capacity have been established. The membranes used for nanofiltration are made of polypiperazineamide fine film composites. With nanofiltration unit, an important part of COD which is not treated in biological treatment is treated, with ultrafiltration at the discharge pool, ISU sewerage discharge quality standards are met. The concentrated waste in NF membranes is disposed in regular landfill areas. The water treated with nanofiltration is recycled by using in the cleaning of the roads within the facility.

At the Landfill leachate waters entering the treatment facility with COD 7500 mg/l and NH₄ 2000 mg/l the output results shown in Figure 11.

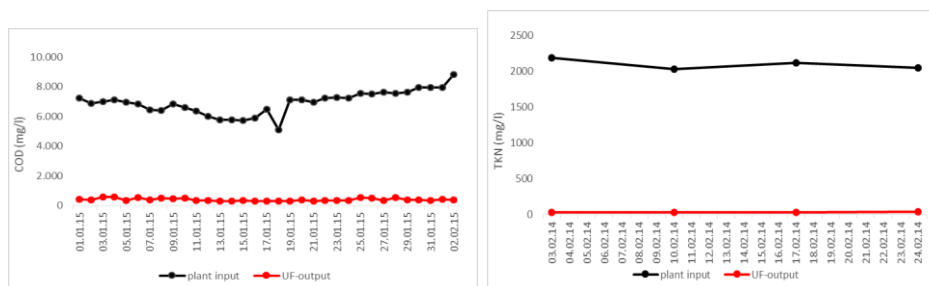


Figure 11. UF output COD and TKN performance graphs

In treatment of landfill leachate waters, the most suitable method among the methods specified in Table 1 is MBR treatment method. Membrane Bioreactor (MBR) systems are used as submerged and external membrane and provide very good efficiencies. With the usage of membrane, mud separation is achieved at very high levels and the leakages to output water is at a minimum level. MBR's operated with long mud age are systems with low mud production and high energy consumption.

The operating costs are high and operation conditions are delicate. Membrane clogging and foam formation are the most frequently encountered problems. However, when comparing to other systems, this is one of the processes with best treatment efficiency. With active carbon/ additional treatment stages, it is possible to achieve very high level of output quality. With the membrane treatment method applied to leachate water, very good efficiency is achieved. However, after membrane treatment, the concentrate formed is the problem encountered. Basically, membran technologies such as ultrafiltration, nanofiltration and reverse osmosis do not eliminate the pollution but just put them in smaller volumes and make them concentrated (Chaudhari and Murthy, 2010). For disposal of the concentrate with very high level of contaminant to the nature, especially Chemical Oxygen Demand (COD) value shall be reduced to below standard level values (Ayala et al., 2008).

Alvarez et al. (2004) carried out a study and examined the studies on leachate water characterization and treatment in the last 40 years and of the treatment efficiency of MBR system. In the leachate water, COD removal is determined by leachate water stability, treatment stages, HRT and organic loadings according to this study. 157 researches in the literature have been examined and their treatment processes and efficiencies have been demonstrated. In the treatment of leachate water, for young landfill areas (less than 5 years old) where leachate water characteristics are determined by the input characteristics, multi stage systems biological treatment and/or long periods of aeration, 99% COD removal can be achieved. For old leachate waters (>5 years), BOD/COD ratio is below 0.3 and with classical biological treatment, 60% COD removal can be achieved. In case nitrification is carried out with sufficient carbon resource, it will be at 95% level and in leachate waters with BOD/COD ratio of 0.1-0.2, in COD removal, a system with three stages, MBR/AC/NF is more efficient according to the study (Alvarez et al., 2004). Discharge output water of MBR treatment facility established in IZAYDAS facilities verifies this information.

4. CONCLUSION

In this study, regarding to treatability of landfill leachate waters of solid waste storage sites, evaluation over the parameters of discharge output of biological-physical-chemical, submerged membrane MBR prototype and constructed real size membrane bioreactor treatment facilities have been carried out. The results of experimental study showed that it is not going to be possible to reach to the required level on leachate waters with hard to treat characteristics only with biological-chemical treatment units and that with membrane bioreactor process, efficient treatment is achievable. It has been found out that in treatment of landfill leachate waters, MBR type treatment facilities can achieve a successful treatment which is proven in experimental and operating facilities.

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