

MOLECULAR WEIGHT DISTRIBUTION OF POLLUTANTS IN LEACHATE FROM FULL SCALE LANDFILL SITE

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ABSTRACT

The main objective of this study was to investigate the differences between the main pollutants in young and intermediate age landfill leachate in terms of molecular weight distribution (MWD). Parameters of chemical oxygen demand (COD), five day - biochemical oxygen demand (BOD₅), ultraviolet-visible spectrophotometry (UV-VIS spectrum), total kjehldahl nitrogen (TKN), ammonia nitrogen (NH₄-N) and colour were fractioned by membranes with the nominal pore size of 1µm, 0.05 µm, 100 kDa, 50 kDa, 10 kDa, 1 kDa and 0.5 kDa. According to the MWD results appropriate treatment technology could be choosen easily. According to the MWD results the ratio of soluble fractions (< 0.5 kDa) to total COD was 34% in young leachate (YL), whereas low molecular weight (MW) fractions were dominant in intermediate leachate (IL) having this ratio of 71%. Presence of lower MW compounds in IL was also confirmed with UV-VIS absorbance spectra and its spectrum was higher than the YL. According to the specific ultraviolet absorption (SUVA) values, the organic contents of all of these processes were hydrophilic. TKN and NH₄-N analysis showed that in IL all the nitrogen present is in ammoniacal form, instead in YL there is still the presence of organic nitrogen. Furthermore more than half of the TKN was less than 0.5 kDa while 6 % and 16 % TKN found in wastewater from IL and YL was higher than 1 µm respectively. The observations of this study may provide useful criteria to choose a suitable landfill leachate treatment processes.

Keywords: Leachate age, landfill, molecular weight.

1. INTRODUCTION

Landfill is one of the major disposal methods in the solid waste management system and landfilling remains nowadays the most widely used municipal solid waste (MSW) treatment in most countries (Berthe *et al.*, 2008; Campagna *et al.*, 2013; Yaman *et al.*, 2015). Landfills release a wide range of chemical compounds due to the waste degradation in the entire life cycle. The release of indeterminated volumes of leachate would introduce the risks to public health and the surrounding environment (Liu *et al.*, 2015; Christensen *et al.* 2001).

Nowadays, although landfill engineering practice and regulations have been applied in protecting the surrounding environment by the use of leachate treatment systems and the controlling of leachate removal (Zhang *et al.*, 2007; Oloibiri *et al.*, 2015; Nivala *et al.*, 2007; Renou *et al.*, 2008). Treatment efficiency has not been satisfied yet. Moreover, in some developing countries, there is a considerable

number of closed landfill areas which were operated without any engineering protection, and leachate from these sites is may be arise over the next several decades.

High concentration of organic matter and inorganic components, heavy metals, xenobiotic organic compounds and microorganisms can be found in the landfill leachate. (Zhang *et al.*, 2007). Thus, a detailed characterization of landfill leachate is very important to choose appropriate treatment technologies and to evaluate its effects on the environment. Determination of MWD of pollutants in leachate also helps to evaluate appropriate treatment technologies and performance of the leachate treatment systems by providing information on the physical characteristic of pollutants in the leachate. However, MWD may affect the behavior and reactivity of pollutants and this information may be very important to comment the fate of the pollutants (Dulekgurgen *et al.*, 2006).

Numerous studies indicated that leachate properties varied significantly as the landfill becomes older (Renou et al., 2008; Buck and Bruland 2007; Campagna et al., 2013; Shehzad et al., 2015). As a landfill moves through each phases of its life cycle, the leachate compositions change dramatically, which might introduce additional challenges for the development of environmental protection technologies. For example, the concentration of organic matter in YL is generally higher than the concentration in IL (ISTAC 2003; Campagna et al. 2013; Amor et al., 2015) The organic matter in water bodies is of great importance due to its effect on fate and transport of contaminants, speciation of metals (Magda et al., 2015; Regadío et al., 2015; Christensen et al., 2011) and the overall ecology and dynamics of water bodies (Regadío et al., 2015; Christensen et al., 2011). Before taking any decision about the treatment plant processes, the concentration of organic matter and other pollutants should be determined. COD is one of the most used conventional parameters to evaluate the organic matter concentration. The characterization of the MWD of COD is a valuable tool when assessing the effect of this material on the performance of water treatment systems as well as its geochemical origin. For example, the size as defined by the MWD has been identified to influence both microbial populations (ISTAC 2003; Buck and Bruland 2007; Amor et al., 2015; Magda et al., 2015; Regadío et al., 2015) and the removal of COD in water treatment systems. However, it is known that components of the COD vary widely due to the complexity of landfill leachates (Steinberg, 2004; Buck and Bruland 2007; Zhang et al., 2012; Amor et al., 2015) Therefore, it is necessary to investigate the pollutant composition in landfill leachate, which can improve the understanding about the distribution, environmental behaviors and fates of individual components. As a general rule, leachate is characterized by high values of COD, pH, NH₄-N and heavy metals, as well as strong colour and bad odour.

Landill leachate has a wide range of NH₄-N concentration (Kulikowska, 2012). The treatment of NH₄-N concentration in the leachate is very vital step because NH₄-N causes to toxicity. As a result determining of MWD of NH₄-N helps to evaluate proper treatment processes. For example, biological methods are only effective to treat low molecular weight (LMW) compounds, for high molecular weight (HMW) NH₄-N can be treated by combination of physical–chemical and biological methods (Zhang *et al.*, 2013).

UV-Vis and colour are other important parameters for the leachate treatment. UV-Vis absorption is known to be a useful indicator for aromaticity and MW of organic matter in the water (Williams, 1998). For example, HMW substances can be easily treated than low molecular substances with coagulation (Yaman *et al.*, 2015).

Landfill leachate is a very dark coloured liquid formed matters and the decomposition of organic matter such as humic acid may cause the water to be yellow, brown or black (Wiley *et al.*, 2014). Landfill leachate could be a major contamination source of ground and surface water. Therefore, it is important to find sustainable option to treat this colour effectively before being discharged into the environment (Wiley *et al.*, 2014).

This study is focused on determination of MWD of YL and IL from a full-scale landfill in order to recognize their characteristics and differences, which could help evaluate appropriate treatment technologies and performance of the leachate treatment systems.

2. MATERIALS AND METHODS

2.1 Site and leachate samples

Istanbul is the biggest metropolitan city of Turkey with a population over 15 million and it has to manage 10,000 tons of solid waste every day. Municipal solid waste (MSW) compositions are averagely 42.97% organics, 7.76% paper, 4.8% plastic, 9.37% nylon, 6.2% glass, 5.8% metal, and 9.28% others (ISTAC, 2003). The samples of landfill leachate used in this study were all collected from a full-scale Odayeri landfill site in Istanbul, and stored in plastic holders at 4 °C. The samples belong to leachate of 1-2 years old (YL), and nearly 10 years old (IL). All leachate samples were collected from drainage pipe-line of the landfill site. The characterizations of YL and IL are shown in Table 1.

Parameters —	Leachate types	
	YL	IL
Colour, PtCo	8100	3100
рН	6,5	7,7
COD, mg l ⁻¹	31700	3850
BOD ₅ , mg l ⁻¹	28700	1700
BOD ₅ /COD	0,91	0,44
DOC, mg l ⁻¹	11180	1440
UV ₂₅₄	42,7	27,8
UV ₂₈₀	32,7	21
UV ₃₂₀	21,7	10,7
TKN, mg l ⁻¹	2640	3550
NH ₄ -N, mg l ⁻¹	2325	3530
Conductivity, mS cm ⁻¹	32,6	37,8
TDS, g l ⁻¹	21,8	32,6

Table 1 Characterization of YL and IL

2.2 Membrane characteristics

In this study, microfiltration membranes of 1 μ m and 0.05 μ m pore sizes and ultrafiltration membranes (from Microdyn Nadir Co.) with a nominal molecular weight (NMWL) of 100 kDa, 50 kDa, 10kDa, 1 kDa (from Pall Co.) and 0.5 kDa (from Spectrum laboratories, Inc.) were used for the sequential filtration process. Moreover 100 kDa, 50 kDa, 10kDa, 1 kDa and 0.5 kDa filters nominal pore sizes are; 7.9 nm, 6.3 nm, 3.5 nm, and 2 nm respectively. At the present, there is a difficult task to reach to a consensus one single definition of size fractions. This difficulty was pointed out in a study (Williams, 1998) in which the variations among the exact cut off sizes given in the studies using categorization based on particle size segregation are highlighted. It was considered the Millipore's definition of NMWL: a UF membrane with stated NMWL will retain at least 90 % of a globular solute of that molecular weight in Daltons (Millipore Ultrafiltration Guide, 2007). UF membranes have a distribution of pore diameters, not just pores of one diameter (Leiviska *et al.*, 2008).

2.3 Sequential filtration

Microfiltrations were performed using a lab-scale filtration system with flat sheet membranes. Ultrafiltrations were performed with a stirred cell (Amicon, Model 8400), it consists of a methylmethacrylate glass holder with a Volume of 350 mL. After all filtration, almost 100 % volume of water was filtered successfully. Effective membrane filter diameter and filtration area were 76 mm and 41.8 cm², respectively. The samples were filtered by a pressure applied with nitrogen gas, the operation pressure was 2 bar, except for the last 0.5 kDa filtration, where a pressure of 4 bar was used because 0.5 kDa filter pore size was very small and 2 bar was not strong enough for filtration. The filters were first washed with 70% (7+3) ethanol for 20 min., thereafter washed in deionized water to remove the ethanol in apparatus and membrane. The sequential filtration was carried out with the following steps shown in the Figure 1. In the first step 800 mL of sample filter 1 μm after 100 ml sample was taken and 700 mL of water filter with 0.05 μm , and these steps were continued sequentially.



Figure 1 Sequential filtration of MWD and the key parameters measured

2.4 Analytical

All the analyses were carried out following the Standards Methods for Water and Wastewater of American Public Health Association (APHA), American Water Work Association (AWWA) and Water Environment Federation (WEF). The COD analysis was executed by open reflux method with the method number of 5220-B in standard method of APHA-AWWA-WEF. For UV-VIS spectrum, the absorptions at 254, 280, 285 and 320 nm wavelengths were investigated.

The 5-day BOD₅ test was carried out following the Standard Procedure of 5210-B in standard methods of APHA-AWWA-WEF. TKN and NH4-N were analysed according to the SM 4500 N_{org} C and SM 4500 NH₃ C, respectively. Organic nitrogen was calculated by the differences of TKN and NH₄-N. The conductivity analyses were carried out using a multi-parameter of Thermo Scientific (model Orion 5 star).

3. RESULTS AND DISCUSSION

The first characterization indicates that BOD_5 and COD values are higher in the YL, because the degradation and humification process is still at the beginning in YL. BOD_5/COD ratio is a common way to evaluate the biodegradability of the leachate (Sophonsiri *et al.*, 1999; Amor *et al.*, 2015). It can be noticed that, the IL is less biodegradable than the young one, moreover, the BOD_5/COD ratio in IL is still nearly 0.4, which indicates relatively low biodegrability (Leiviska *et al.*, 2008). This also means that the leachate is not properly "old" because it is still biodegradable, so this sample is not a real old and stabilized leachate, but rather an "intermediate" leachate (Leiviska *et al.*, 2008).

Normally, old landfill leachates (> 10 years), present a low BOD₅/COD ratio (<0.1), indicating low biodegradability due to the release of HMW recalcitrant organic molecules (Jensen *et al.*,1999; Vilar *et al.*, 2011; Amor *et al.*,2015). The YL has a high BOD₅/COD ratio, therefore a biological process is strongly recommended, because there is high amount of organic substrate available for bio-degradation. The higher UV absorbance in YL is probably due to the fact that in this sample the NOM (like humic and fulvic acid) has a higher concentration than in the IL (Kang *et al.*,2002).

3.1 Organic contents

It can be seen from Figure 2, the particulate COD fractions (>0.05 μ m) of YL and IL were 16% and 14%, respectively. These ratios indicated that the COD of both leachates were mainly comprised of dissolved organics. COD fractions for 100 kDa-50 kDa, 50 kDa-10 kDa, 10 kDa-1 kDa, 1kDa–0.5 kDa and < 0.5kDa were 14%, 1%, 4%, 31% and 32% in YL and 9%, 4%, 1%, 2% and 70 % in IL, respectively. More than half of the COD in YL and IL had medium molecular weight (MMW) (<0.05 μm and >0.5kDa). According to these results, a biological process is strictly recommended for the treatment of YL and IL because the microorganisms can use these medium and LMW substrates, for the reasons that part of it is the readily biodegradable COD taken into account the BOD₅ concentration of the YL (Table 2). Zhao et al., (2012), also reported that LMW of COD can be easily used by microorganisms. In accordance with microbial degradation kinetics, the soluble readily biodegradable COD fraction consists of nearly small biodegradable particles, which are easily passesd across cell membrane and then metabolized in a short time (Zhao et al., 2012). On the contrary, using of the particular biodegradable COD and the soluble slowly biodegradable COD fractions takes longer since these component comprise larger particle and need extra cellular breakdown prior to their transport into the cell for biodegradation. COD fractionation is quite important due to the response of biomass in activated sludge varies with particle size of these components.As a result of these, the huge range of biodegradation rates defined for different COD fractions is likely to have a correlation with physical categorization in terms of particle size and physical state in solution (Dulekgurgen et al., 2006).



Figure 2. MWD of COD concentration

Therefore, a traditional biological process, or, better, an MBR process can be very effective for COD removal for YL (Zhao *et al.*, 2012). With leachate aging, there is a shift of COD fractions towards HMW. The IL, nevertheless, seems to have a higher COD fraction of LMW (< 0.5kDa). The results indicate that significant parts of COD in IL have very LMW. This can be due to the fact that the COD of IL is in soluble inert form (Bijan and Mohensi 2005). For this kind of IL an advanced pre-oxidation (O₃, fenton process, etc.) may be necessary to break down the inert compounds and make them ready for biodegradation. Besides, the low (<0.5kDa) and high molecular (>0.05 µm) weight distributions were 71% (2773 mg COD/L) and 14%, respectively, in IL and Kang *et al.* (Kang *et al.*, 2002) also observed similar results.

On the one hand, RO and NF membranes which have lower molecular weight cut off (MWCO) than 0.5 kDa can not seen feasible due to LMW of COD in YL and IL (Cakmakci *et al.*, 2012; Ozyaka *et al.*, 2015)

However, flocculation and precipitation processes can be effective HMW than 0.1 kDa (Uner *et al.*,2006). As seen in Table 2, BOD₅ concentration in YL is very high and most of its LMW of COD can be utilized by

microorganisms. Thus, biological treatment should be applied to treat YL before membrane processes (Li et al., 2009).

After biological treatment of organic matter, membrane processes can be used to enhance treated water quality.





3.2 UV-VIS spectra

The total spectra is used to understand the MWD of the samples. The UV-VIS spectra of samples containing high concentrations of organic matters (humic acids, fulvic acids) are the result of overlapping absorbances of numerous different chromophores caused by the very complicated structures of humic substances (Edzwald and Tobiason, 1999).

UV-Vis absorbances at different wavelengths (254, 280 and 320) were studied to determine the organic matter contents in each MWD. It is known that high UV-Vis absorbance between 190 nm and 800 nm shows the existing of humic substances, whereas aromatic content of organic matter can be observed at low UV-Vis wavelengths (Chen and Tobiason, 1999). Aromatic organics are best detected using the 254 nm wavelength, because of the strong absorption properties of these double bond organics. In addition, saturated ketones and some aromatic organics detected at 280 nm wavelength and 285 nm wavelengths is an indicator of DOC, and 320 nm is also an indicator of COD in the landfill leachate (Gu *et al.*, 2002).

In this study, the results of the UV-Vis absorptions were shown in Figure 3. The absorbance value at 254 nm was almost same for both YL and IL. The absorbance values at 254 nm was high for both leachates and it means that aromatic organics may be dominants organics in the leachates. A strong decline of UV-Vis absorbance was observed after 0.5 kDa filtration for both YL and IL.

As expected at 280 nm, theabsorbance values was higher with IL, because of the presence of high molecular weight and high aromaticity organic matter. 285 nm is an indicator of DOC, and it can be noticed that the absorbance values is higher in the IL than in the YL.

Most part of UV-Vis absorbances were removed by 100 kDa. Thus, aromatic contents of organic matter can be removed by ultrafiltration membranes which have 100 kDa or lower MWCO.

3.3 SUVA

If SUVA value is higher than 4, it indicates HMW organic matter which has hydrophobic character and if SUVA is lower than 2, it indicates LMW organic matter which has hydrophilic character (Bijan and Mohensi 2005, Li *et al.*, 2009; Zhao *et al.*, 2012; Cakmakci *et al.*, 2012; Ozyaka *et al.*, 2015). Figure 4 shows the distribution in SUVA values of YL and IL. SUVA values was lower than 2 l/mg.m which indicate that these waters are in hydrophobic characteristics. Hence conventional coagulation–flocculation is not a proper method for removing highly hydrophilic organic (Edzwald and Tobiason *et al.*, 1999.).

SUVA with MW of $0.1 \,\mu$ m was 38% and 40% in YL and IL, respectively. According to these values, it can be concluded that SUVA is mostly composed of high molecular organics in both leachates. It was obvious that pretreatment with or over 0.1 μ m can successfully remove major part of SUVA.



Figure 4 MWD of SUVA₂₅₄ values

3.3 Nitrogen

Concentrations of TKN and NH₄-N are shown in Figure 5. As seen from Figure 5, there is a bit diffences between TKN and NH₄-N concentrations. It means a part of organic nitrogen is still available in YL. Unlike

YL, organic nitrogen is almost converted into NH_4 -N due to the nearly same concentrations of TKN and NH_4 -N. These results are in accordance with Kang *et al.*, (2002), and Renou *et al.*, (2008), studies.

The main source of NH₄-N in leachate results from the anaerobic hydrolysis of proteins in the solid wastes. NH₄-N slowly releases from the solids because of the slow anaerobic hydrolysis. Thus, NH₄-N concentration increases day by day in landfill leachate. If TKN is almost equal to NH₄-N, it can be assumed that the landfill is stabilized (Hautala *et al.*, 2000).

Most of TKN and NH₄-N (66.40% and 66.70 respectively) passed from 1 kDa (Figure 5) from the both leachates was very low. These values show that when precipitation is added to flocculation, MF, UF and NF membranes are not sufficient to remove TKN. Zaho *et al.* (2012) found that LMWorganic nitrogen was effectively removed by means of biological processes. In the study, TKN and NH₄-N were mostly less than 1 kDa MWD; this means the TKN and NH₄-N can also be reduced by biological treatment processes such as combination of anoxic and aerobic.



Figure 5 MWD of TKN and NH₄⁺-N

3.4 Colour

Colour is an important water parameter due to the astehtic and the live life in water. The leachate colour dependences on the landfill ages as shown in Figure 6. All samples in YL and IL are of brownish colour, due to the aromatic structures of humic and fulvic acids (ISTAC, 2003).

The colour of the YL is darker than the IL. Most of colour of both leachates can be removed by 1 μ m and 0.05 μ m according to the MWD. This means that the largest part of colour is produced by colloidal suspended solids and HMW of dissolved matters. In YL, removal efficiencies of colour by 1 μ m and 0.05 μ m is higher than IL. These results are nearly similar with Leiviska *et al.*, (2008), studies. They obtained the highest turbidity and colour removal in correspondence of the 0.22 μ m. Moreover, after 10 kDa MWD, the colour of the YL and IL can be reduced up to 93 % and 67%, respectively. Thus, it is assumed that most parts of colour in YL and IL can be removed by microfiltration or ultrafiltration membrannes. When an NF with an MWCO of 1 kDa is applied, the colour can be decreased by 96 % and 78% from the YL and IL. In addition to membrane filtration, ozonation and biological treatment can also be applied in order to remove LMW organics before being filtered through the NF or RO membranes (Campagna *et al.*, 2013). Moreover, low-molecular-weight organics with high biodegradability are easily biodegraded by anaerobic treatment (Leiviska *et al.*, 2008).



Figure 6. MWD of colour

4. CONCLUSIONS

In this study, YL and IL were analyzed and evaluated according to the pollution concentration and MWD. The following conclusions can be drawn:

- COD in YL is constituted from LMW compounds, which can easily be used by microorganisms. Traditional biological process, or, better, an MBR process can be very effective for removal of LMW COD. Unlike YL, IL is constituted from soluble and non-biodegradable compounds, so the biological treatment may not be very effective to remove COD. Some solutions to treat this kind of leachate could be mixing it with YL, or an advanced oxidation to break down the nonbiodegradable compounds and make them ready to biodegradation.
- 100 kDa or lower MWCO ultrafiltration membranes can be removed most part of UV–Vis absorbances.
- SUVA values show that YL and IL organic matters have hydrophilic characters. Thus, biological and physical (membrane processes) prosesses can be used to remove organic matter in YL and IL.
- Most part of nitrogen is NH4⁺-N in YL while almost all of it is NH4⁺-N in IL. In both cases the membrane treatment system was not effective for nitrogen removal according to the MWD analyses. Bological treatment processes such as combination of anoxic and aerobic can be used to treat TKN and NH4-N.

- Most part of colour may occurred due to colloidal suspended solids and HMW matters according to the MWD analyses. The colour can be removed by 100 kDa and lower MWCO of ultrafiltration membranes.
- This study only shows the evaluations of YL and IL detailed characterizations. Thus, treatability studies may be applied to understand the effects of MWD on treatment performance.

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