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Review on Landfill Leachate Treatments

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Abstract: Problem Statement: Sanitary landfilling is the most common way to eliminate solid urban wastes. An important problem associated to landfills is the production of leachates. This study is a review of landfill leachate treatments. **Approach:** The advantages and disadvantages of the various existing leachate treatments discussed under the items: (i) Leachate channeling (combined treatment with domestic sewage, recycling) (ii) Biological processing (aerobic and anaerobic) (iii) Chemical/physical treatment (flotation, coagulation/flocculation, chemical precipitation, adsorption, ammonium stripping, chemical oxidation, ion exchange and electrochemical treatment) (iv) Membrane filtration (microfiltration, ultrafiltration, nanofiltration and reverse osmosis). **Conclusion:** The major fraction of old or biologically treated leachate was large recalcitrant organic molecules that are not easy removed during biological treatment. So that, in order to meet strict quality standards for direct discharge of leachate into the surface water, a development of integrated methods of treatment, a combination of biological, chemical, physical and membrane process steps, were required. Today, the use of membrane technologies, more especially Reverse Osmosis (RO), either as a main step in a landfill leachate treatment chain or as single post-treatment step had shown to be an indispensable means of achieving purification.

Key words: Landfill leachate, review, biological treatment, physical/chemical treatment, membrane filtration

INTRODUCTION

Landfilling of municipal waste is still a very important issue of the waste management system in the world. For instance, the biodegradable municipal waste is expected to be reduced to 75% by weight of 1995 levels by 2010.

Wastes cause two types of pollution, which correspond to the migration into the natural environment of:- [1]leachates, defined as water that has percolated through the wastes (rainwater or groundwater seepage), a source of soil and groundwater contamination and[2]biogas produced by the fermentation of organic matter, a source of air pollution. With regard to leachates, controlling the pollutant loading means reducing its quantity.

Some alternative methods such as recycling, composting and incineration are nowadays very much encouraged but even incinerations create residue of approximately 10-20% that must be ultimately landfilled. At present, modern landfills are highly engineered facilities designed to eliminate or minimize the adverse impact of the waste on the surrounding environment. However, the generation of contaminated leachate remains an inevitable consequence of the existing waste disposal practice and the future landfills.

Landfill effluents (leachate) need to be pre-treated on site to meet the standards for its discharge into the sewer or its direct disposal into surface water. In the world the problem of leachate treatment has been existed for sometime now, but a universal solution has not been found.

The aim of this study is to make a review on the state of art in landfill leachate treatment and provides a comparative evaluation of various treatment processes.

Leachate production and characteristics: Rainfall is the main contributor to generation of leachate. The precipitation percolates through the waste and gains dissolved and suspended components from the

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biodegrading waste through several physical and chemical reactions. Other contributors to leachate generation include groundwater inflow, surface water runoff and biological decomposition[59]. Liquid fractions in the waste will also add to the leachate as well as moisture in the cover material. Moisture can be removed from the landfill by water consumed in the formation of landfill gas, water vapor removed in the landfill gas and leachate leaking through the liner[69].

The quantity of Leachates are depend on rainwater percolation through wastes, biochemical processes in waste's cells, the inherent water content of wastes and its degree of compaction into the landfill tip. The production is generally greater whenever the waste is less compacted, since compaction reduces the filtration rate[37].

There are many factors affecting the quality of leachates, i.e., age, precipitation, seasonal weather variation, waste type and composition. In particular, the composition of landfill leachates varies greatly depending on the age of the landfill[63]. There are three types of leachates have been defined according to landfill age (Table 1). As landfill age increased, organics concentration (COD) in leachate decreased and increase of ammonia nitrogen concentration[35]. Landfill leachates from old sites are usually highly contaminated with ammonia resulting from the hydrolysis and fermentation of nitrogen containing fractions of biodegradable refuse substrates[11]. The existing relation between the age of the landfill and the organic matter composition may provide a useful criteria to choose a suited treatment process. In general, leachates may contain large amounts of organic matter (biodegradable, but also refractory to biodegradation), where humic-type constituents consist an important group, as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts.

The characteristics of the landfill leachate can usually be represented by the basic parameters COD, BOD, the ratio BOD/COD, pH, Suspended Solids (SS),

Table 1: Landfill leachate classification vs. age[19]

	Young	Medium	Old
Age (year)	<1	1-5	>5.0
pH	<6.5	6.5-7.5	>7.5
COD (g L^{-1})	>15	3.0-15	<3.0
BOD_5/COD	0.5-1	0.1-0.5	<0.1
TOC/COD	<0.3	0.3-0.5	>0.5
NH_3-N (mg L^{-1})	<400	400	>400
Heavy metals (mg L^{-1})	>2.0	<2.0	<2.0
Organic compound	80% VFA	5-30% VFA+ HA+FA	HA+FA

VFA = Volatile Fat acids. HA = Humic Acid. FA = Fulvic Acids

Ammonium nitrogen (NH_3-N), Total Kjeldahl Nitrogen (TKN) and heavy metals. Recirculation of leachate will produce stabilized leachates containing relatively low concentrations of degradable carbon compounds but high concentrations of ammonia[11] therefore, COD and BOD will be removed, but ammonia concentrations will climb.

Landfill leachate treatments:

Leachate channeling:

Combined treatment with domestic sewage: One common means of leachate disposal is piping into the sewer system for discharge into the sea or, preferably, for combined treatment with domestic sewage at conventional sewage plant. It was preferred for its easy maintenance and low operating costs[1]. However, this option has been increasingly questioned due to the presence in the leachate of organic inhibitory compounds with low biodegradability and heavy metals that may reduce treatment efficiency and increase the effluent concentrations[10]. An argument in favor of this alternative treatment is that nitrogen (brought by leachate) and phosphorus (brought by sewage) don't need to be added at the plant. Among the few studies published, authors tried to optimize the volumetric ratio of leachate in the total wastewater. Combined treatment was investigated by Diamadopoulos *et al.*[13] using a Sequencing Batch Reactor (SBR) consisting of filling, anoxic, toxic and settling phases. When the ratio of sewage to leachate was 9/1, nearly 95% BOD and 50% nitrogen removals were obtained at the end of the daily cycles. COD and NH_3-N reduction decreased with increasing landfill leachate/domestic wastewater ratio. Moreover, the effluent quality may be improved with Powdered Activated Carbon (PAC) addition, particularly if the leachate input exceeds 10%[9].

Recycling: A widespread technique used in many landfills consists in recycling leachate back through the tip because it was one of the least expensive options available[37]. Recently, authors showed benefits of this technique. Bae *et al.*[6] reported that leachate recirculation increased the moisture content in a controlled reactor system and provided the distribution of nutrients and enzymes between methanogens and solid/liquids. Significant lowering in methane production and COD was observed when the re-circulated leachate volume was 30% of the initial waste bed volume[12]. Also, Rodriguez *et al.*[61] reported a 63-70% COD lowering in an anaerobic pilot plant with recirculation. The leachate recycle not only improves the leachate quality, but also shortens the time required for stabilization from several decades to 2-3 years[58]. High recirculation rates may adversely affect anaerobic degradation of solid wastes. For instance, Ledakowicz and Kaczorek[36] observed that leachate recirculation can lead to the inhibition of methanogenesis as it may cause high concentrations of organic acids (pH<5) which are toxic for the methanogens. Furthermore, if the volume of leachate recirculated is very high, problems such as saturation, ponding and acidic conditions may occur[36].

Biological treatment: Biological purification processes are classified as aerobic or anaerobic depending or whether or not the biological processing medium requires an O_2 supply. In aerobic processing organic pollutants are mainly transformed into CO_2 and solid biological products (sludge) by using the atmospheric O_2 transferred to the wastewater. In anaerobic treatment organic matter is converted into biogas, a moisture comprising chiefly CO_2 and CH_4 and in a minor part into biological sludge. Biological processes have been shown to be very effective in removing organic and nitrogenous matter from immature leachates when the BOD/COD ratio has a high value (>0.5). With time, the major presence of refractory compounds (mainly humic and fulvic acids) tends to limit process's effectiveness[37].

Aerobic treatment: An aerobic treatment should allow a partial abatement of biodegradable organic pollutants and should also achieve the ammonium nitrogen nitrification. Aerobic biological processes based on suspended-growth biomass, such as aerated lagoons, conventional activated sludge processes and Sequencing Batch Reactors (SBR), have been widely studied and adopted[5,33,38]. Attached-growth systems have recently attracted major interest: the Moving-Bed Biofilm Reactor (MBBR) and biofilters. The combination of membrane separation technology and aerobic bioreactors, most commonly called membrane bioreactor, has also led to a new focus on leachate treatment.

Aerated lagoons have generally been viewed as an effective and low-cost method for removing pathogens, organic and inorganic matters. Their low operation and maintenance costs have made them a popular choice for wastewater treatment, particularly in developing countries since there is a little need for specialized skills to run the system[77]. Maehlum[47] used on-site anaerobic-aerobic lagoons and constructed wetlands for biological treatment of landfill leachate. Overall N, P and Fe removals obtained in this system were above 70% for diluted leachate. Orupold *et al.*[53] studied the feasibility of lagooning to treat phenolic compounds as well as organic matter. Abatement of 55-64% of COD and 80-88% of phenol was achieved. However, as stricter requirements are imposed, lagooning may not be a completely satisfactory treatment option for leachate in spite of its lower costs[77]. In particular, authors claimed that the temperature dependence of lagooning is a significant limitation because it mainly affects microbial activity.

Activated sludge processes are extensively applied for the treatment of domestic wastewater or for the co-treatment of leachate and sewage. However, this method has been shown in the more recent decades to be inadequate for handling landfill leachate treatment [40]. Even if processes were proved to be effective for the removal of organic carbon, nutrients and ammonia content, too much disadvantages tend to focus on others technologies:

- Inadequate sludge settleability and the need for longer aeration times[46]
- High energy demand and excess sludge production[21]
- Microbial inhibition due to high ammonium-nitrogen strength[37]

Consequently, only few works are recently available concerning landfill leachate treatment by activated sludge methods. Hoilijoki *et al.*[21] investigated nitrification of anaerobically pre-treated municipal landfill leachate in lab-scale activated sludge reactor, at different temperatures (5-10°C) and with the addition of plastic carrier material. Aerobic post-treatment produced effluent with 150-500 mg L^{-1} of COD, less than 7 mg L^{-1} of BOD and on an average, less than 13 mg L^{-1} of NH_3-N . Addition of PAC to activated sludge reactors enhanced nitrification efficiency on biological treatment of landfill leachate[21].

Sequencing Batch Reactor is ideally suited to nitrification-denitrification processes since it provides an operation regime compatible with concurrent organic carbon oxidation and nitrification[13]. Process characteristics, summarized by Diamadopoulos *et al.*[13] and Dollerer and Wilderer[14], resulted in a wide application for landfill leachate treatment[71,77]. Many authors[14,29] have reported COD removals up to 75%. Also, 99% NH_3-N removal has been observed by Lo[43] during the aerobic treatment of domestic leachates in a SBR with a 20-40 days residence time. The greater process flexibility of SBR is particularly important when considering landfill leachate treatment, which have a high degree of variability in quality and quantity[30].

Due to main problems of sludge bulking or inadequate separability[14] in conventional aerobic systems, a number of innovative aerobic processes, called attached-growth biomass systems, using biofilm, have been recently developed. These systems present the advantage of not suffer from loss of active biomass. Also nitrification is less affected by low temperatures[33] than in suspended-growth systems and by inhibition due to high nitrogen content.

Trickling Filters has been investigated for the biological nitrogen lowering from municipal landfill leachate. Biofilters remain an interesting and attractive option for nitrification due to low-cost filter media[27]. In a recent work, above 90% nitrification of leachate was achieved in laboratory and on-site pilot aerobic crushed brick filters with loading rates between 100 and 130 mg L^{-1} d^{-1} of NH_3-N at 25°C and 50 mg L^{-1} d^{-1} NH_3-N even at temperatures as low as 5-10°C, respectively[27]. In the last decade, maximum ammonia rejection of 97 and 75 % in a trickling filter were respectively claimed by[34] and Martienssen and Schops[49].

Moving-Bed Biofilm Reactor (MBBR)- process is based on the use of suspended porous polymeric carriers, kept in continuous movement in the aeration tank, while the active biomass grows as a biofilm on the surfaces of them. Mains advantages of this method compared to conventional suspended growth processes seems to be: higher biomass concentrations, no long sludge-settling periods, lower sensitivity to toxic compounds[46] and both organic and high ammonia removals in a single process[22]. For instance, Welander *et al*.[74] reported nearly 90% nitrogen removal while the COD was around 20%. In case of treating high strength ammonia leachate, no inhibition of nitrification is encountered[74]. Moreover, the use of Granular Activated Carbon (GAC) as porous material offers an appropriate surface to adsorb organic matter and optimized conditions for enhanced biodegradation[22]. Thus, a steady-state equilibrium is established between adsorption and biodegradation[22]. Imai *et al.*[24,25,26] developed an efficient biological activated carbon fluidized bed process. Nearly, 70% refractory organics were removed by coupling biological treatment and adsorption process[26]. After optimizing the reactor operating regime, Horan *et al*.[22,46] proved possible to reach 85-90% ammonia reduction and 60-81% COD reduction.

Anaerobic treatment: An anaerobic digestion treatment of leachates allows to end the process initiated in the tip, being thus particularly suitable for dealing with high strength organic effluents, such as leachate streams from young tips[8]. Contrary to aerobic processes, anaerobic digestion conserves energy and produces very few solids, but suffers from low reaction rates[66]. Moreover, it is possible to use the CH_4 produced to warm the digester that usually works at 35°C and, under favorable conditions, for external purposes.

Performances of conventional anaerobic suspended-growth digester has been studied by[8,66]. Typical values of 80-90% and nearly 55% COD removals were reached in anaerobic lab-scale tank at 35°C and ambient temperature, respectively

Some studies revealed good performances of anaerobic sequencing batch reactors. These systems are able to achieve solid capture and organic lowering in one vessel, eliminating the need for a clarifier. Recently, nutrient reduction from pre-treated leachate was carried out using a lab-scale SBR by[71]. Sequential anaerobic-aerobic operations resulted in COD, NH_3-N and $PO4^{3-}$-P removal of 62, 31 and 19%, respectively, at the end of cycle time (21h). Also, in the initial period of the landfill, sufficient organic abatement in the anaerobic reactor through methanogenesis and denitrification, can enhance better nitrification in the following aerobic reactor. Therefore, anaerobic-aerobic system is recommended to bring down simultaneously organic and nitrogen matter[49,74]. For instance, Kettunen and Rintala[31] showed that COD removal was 35% in the anaerobic stage while in the combined process the COD and BOD_7 removals were up to 75 and 99%.

In last decades, the performance improvement of the existing anaerobic process was believed to be a promising option and so, high rate reactors have been designed in order to reduce long digestion time[40].

Up-flow Anaerobic Sludge Blanket (UASB) process is a modern anaerobic treatment that can have high treatment efficiency and a short hydraulic retention time[40]. UASB reactors, when they are submitted to high volumetric organic loading rate values [16], have exhibited higher performances compared to other kinds of anaerobic reactors. The process temperatures reported have generally been 20-35°C for anaerobic treatment with UASB reactors. In these conditions, the average performance of COD decrease efficiency was always higher than 70% at ambient temperature (20-23°C) and 80% at 35°C. Up to 92% COD decreases were obtained by Kennedy and Lentz[30] at low and intermediate organic loading rates (between 6 and 19.7 g/L/d of COD). Only a few studies have been conducted at temperatures between 11-

23°C[16,32] although leachates may be cooler than that, especially in cold countries. Kettunen and Rintala[32] showed that leachate can be treated on-site UASB reactor at low temperature. A pilot-scale reactor was used to study municipal landfill leachate treatment (COD 1.5-3.2 g L^{-1}) at 13-23°C. COD (65-75%) and BOD_7 (up to 95%) removals were achieved at organic loading rates of 2-4 kg m^{-3} d^{-1} of COD. Garcia *et al.*[16] concluded that COD rejection efficiency was not affected by temperature between 15 and 35°C. These promising results show that high-rate treatment at low temperature may minimize the need for heating the leachate prior to treatment, which may thus provide an interesting cost-effective option [16]. The main disadvantages of such a treatment stay sensitivity to toxic substances[66].

The anaerobic filter is a high rate system that gathers the advantages of other anaerobic systems and that minimizes the disadvantages. In an up-flow anaerobic filter, biomass is retained as biofilms on support material, such as plastic rings[52]. For instance, Henry *et al.*[20] demonstrated that anaerobic filter could reduce the COD by 90%, at loading rates varying from 1.26 to 1.45 kg m^{-3} d^{-1} of COD and this for different ages of landfill. Total biogas production ranged between 400 and 500 L.gas/kg COD destroyed and methane content between 75 and 85%.

Hybrid bed filter consists on an up-flow sludge blanket at the bottom and an anaerobic filter on top. This device acts as a gas-solid separator and enhances solid's retention without causing channeling or short-circuiting[52]. Enhanced performances of such a process results from maximization of the biomass concentration in the reactor. Nedwell and Reynolds[52] reported steady state COD removal efficiencies of 81-97% under methanogenic digestion, depending upon organic loading rate. One drawback of hybrid reactor, as well as anaerobic filter, is the added cost of the support media.

Suidan *et al.*[65] and Imai *et al.*[24,25,26] reported studies on carbon-assisted fluidized beds. The combined biodegradation and adsorption process provide a means for removing a variety of organic compounds[65]. Imai *et al.*[24] found that the biological activated carbon fluidized bed process was much more effective for treating old landfill leachate than the conventional one such as activated sludge and fixed film processes.

Physical/chemical treatment: Physico-chemical methods are used along with the biological methods mainly to improve treatment efficiency or make them possible when the biological oxidation process is hampered by the presence of bio-refractory materials. The techniques are applied for removing non-biodegradable (humic, fulvic acid) and/or undesirable compounds (heavy metals, AOXs, PCBs...) from the leachate.

Flotation: For many years, flotation has been extensively used and focused on the decrease of colloids, ions, macromolecules, microorganisms and fibers[80]. However, until to date, very few studies have been devoted to the application of flotation for the treatment of landfill leachate. Recently, Zouboulis *et al.*[80] investigated the use of flotation in column, as a post-treatment step for removing residual humic acids (non-biodegradable compounds) from simulated landfill leachates. Under optimized conditions, almost 60% humic acids removal has been reached.

Coagulation-flocculation: Coagulation-flocculation may be used successfully in treating stabilized and old landfill leachates[63]. It is widely used as a pre-treatment[4,78], prior to biological or reverse osmosis step, or as a final polishing treatment step in order to remove non-biodegradable organic matter. Aluminum sulfate, ferrous sulfate, ferric chloride and ferric chloro-sulfate were commonly used as coagulants[4,79]. The application of bioflocculant, in comparison with traditional inorganic coagulants has been recently investigated by Zouboulis *et al.*[79], for the lowering of humic acids. It revealed as a viable alternative since 20 mg L^{-1} bioflocculant dosage was sufficient in providing more than 85% humic acid removal.

Several studies have been reported on the examination of coagulation-flocculation for the treatment of landfill leachates, aiming at process optimization, i.e. selection of the most appropriate coagulant, identification of optimum experimental conditions and assessment of pH effect[4]. Synthesis of recent works clearly reveal that iron salts are more efficient than aluminum ones, resulting in sufficient Chemical Oxygen Demand (COD) reductions (up to 50%), whereas the corresponding values in case of aluminum or lime addition were moderate (between 10 and 40%)[44]. Nevertheless, combination of coagulants[44] or addition of flocculants together with coagulants may enhance the floc-settling rate[4] and so the process performance (COD abatement up to 50%).

However, this treatment presents some disadvantages: consistent sludge volume is produced and an increase on the concentration of aluminum or iron, in the liquid phase, may be observed[63].

Chemical precipitation: Chemical precipitation is widely used as leachate pre-treatment in order to remove high strength of ammonium nitrogen (NH_3-N). In a study, Li *et al*.[39] confirmed that the performance of a conventional activated sludge process could be significantly affected by a high concentration of NH_3-N. The COD removal declined from 95 to 79%, when the NH_3-N concentration in wastewater increased from 50 to 800 mg L^{-1}. Li *et al*.[38,39] precipitated ammonium ions as Magnesium Ammonium Phosphate (MAP) with the addition of $MgCl_2.6H_2O$ and $Na_2HPO_4.12H_2O$ with a $Mg/NH_4/PO_4$ ratio of 1:1:1 at a pH of 8.5-9. Ammonium concentration was reduced from 5600 to 110 mg L^{-1} within 15 min by this method. Yangin *et al*.[76] and Altinbas *et al*.[3] studied MAP precipitation after anaerobic pre-treatment of domestic wastewater and landfill leachate mixture. Maximum ammonia lowering was obtained as 66% at a pH of 9.3 at the stochiometric ratio whereas ammonia lowering reached to 86% at the same pH above the stochiometric ratio. In MAP precipitation at the stochiometric ratio and above the stochiometric ratio, ammonia concentration, in the Upflow Anaerobic Sludge Blanket (UASB) reactor, was reduced to 31 mg L^{-1} and 13 mg L^{-1}, respectively. Recently, struvite precipitation (Mg: NH_4: PO_4 = 1:1:1) was applied to anaerobically pretreated effluents for ammonia removal[3]. Ammonium nitrogen depletion was observed as 85, 72 and 20% at pH of 9.2, 12 and 10-11, respectively.

Adsorption: The adsorption process is used as a stage of integrated chemical-physical-biological process for landfill leachate treatment[17], or simultaneously with a biological process[28]. The most frequently used adsorbent is granular or Powdered Activated Carbon (PAC). Carbon adsorption permits 50-70% removal of both COD and ammonia nitrogen[4]. Consequently, activated carbon adsorption aim is to (i) ensure final polishing level by removing toxic heavy metals or organics i.e., AOXs, PCB (ii) support microorganisms. Other materials, tested as adsorbents, have given treatment performances close to those obtained with activated carbon. These are zeolite, vermiculite, illite, keolinite, activated alumina and municipal waste incinerator bottom ash[4].

Ammonium stripping: Due to its effectiveness, ammonium stripping is the most widely employed treatment for the removal of NH_3-N from landfill leachate. High levels of ammonium nitrogen are usually found in landfill leachatesand stripping can be successful for eliminating this pollutant, which can increase wastewater toxicity[50]. If this method is to be efficient, high pH values must be used and the contaminated gas phase must be treated with either H_2SO_4 or HCl. Performances of this process can be evaluated in term of ammonia-nitrogen removal efficiency. Marttinen *et al*.[50] reported a 89% ammonia reduction at pH=11 and 20°C within 24h retention time. High rates of ammonia removal have been achieved by Cheung *et al*.[11] in spite of high initial ammonia concentration (0.5-0.7 g L^{-1} of N). Their results showed that 93% of 309-368 mg L^{-1} ammonia-nitrogen were removed in free stripping tanks with one day retention time. In recent works, 99.5% of ammonia reduction has been respectively attained by Silva *et al*.[63]. But a major concern about ammonia stripping is the release of NH_3 into the atmosphere so as to cause severe air pollution if ammonia can not be properly absorbed with either H_2SO_4 or HCl. Others drawbacks are the calcium carbonate scaling of the stripping tower, when lime is used for pH adjustment and the problem of foaming which imposes to use a large stripping tower[39].

Chemical oxidation: Chemical oxidation is required for the treatment of wastewater containing soluble organic non-biodegradable and/or toxic substance[48]. As Amokrane and co-workers[4] reviewed, commonly used oxidants such as chlorine, ozone, potassium permanganate and calcium hydrochloride for landfill leachate treatment resulted in COD removal of around 20-50%. The most processes based on direct reaction of oxidant (O_3-selective) with contaminates or via generated hydroxyl radicals (OH).

Advanced Oxidation Processes (AOP) have been proposed in recent years as an effective alternative for mineralization of recalcitrant organics in landfill leachate. The main purpose of AOP[23] is to enhance chemical oxidation efficiency by increasing generation of hydroxyl radicals. Most of them, except simple ozonation (O_3), use a combination of strong oxidants, e.g. O_3 and H_2O_2, irradiation, e.g. Ultra-Violet (UV), Ultra-Sound (US) or Electron Beam (EB) and catalysts, e.g. transition metal ions or photo catalyst. Table 2 lists typical AOP systems currently reported in the literature. All these processes have been recently reviewed by Wang *et al*.[73].

Table 2: List of typical AOP systems[45]

With irradiation	Without irradiation
Homogeneous System	
O_3/ultraviolet (UV)	O_3/ H_2O_2
H_2O_2/UV	O_3/OH^-
Electron beam	H_2O_2/Fe^{-2} (Fenton's)
Ultrasound (US)	
H_2O_2/ US	
UV/US	
H_2O_2/Fe^{-2}/UV(photo-fenton's)	
Heterogeneous systems	

TiO_2/ O_2/UV	electro-Fenton
TiO_2/ H_2O_2/UV	

Although many of researchers using ozonation have demonstrated the effectiveness in eliminating COD (reduction is about 50 to 70% in most cases)[7,64] most of them only used this process as tertiary treatment prior to discharge in the environment. Sometimes the treatment efficiency on stabilized leachates has been moderate[63]. After 1h of ozonation (1.3-1.5 gO_3/gCOD degraded), only 30% COD depletion was observed by Rivas *et al*.[60].COD lowering can be greatly enhanced combining oxidants (H_2O_2/O_3) or adding an irradiation system (H_2O_2/UV) (Table 2). Wable *et al*.[72], Bigot *et al*.[7] and Schulte *et al*.[62] reported organic matter removal efficiency as high as 90% for the O_3/H_2O_2 process. Concerning the H_2O_2/UV process, the BOD/COD ratio has been increased significantly from 0.1 to 0.45 by Qureshi *et al*.[57]. Also, Steensen[64] reported 85-90% of COD reduction with a biologically pre-treated leachate. Fenton and photo-Fenton processes allow COD decrease efficiency of, respectively, 45-75 and 70-78%. In term of biodegradability improvement, BOD_5/COD ratios close to 0.5 after oxidation have been reported in recent works using Fenton process[45]. Finally, a few papers reported photocatalytic treatment or electron-beam radiation treatment[5] of organic components from landfill leachates even at laboratory scale. These technologies have been applied to treat or degrade principally humic substances.

However, common drawbacks of AOP is the high demand of electrical energy for devices such as ozonizers, UV lamps, ultrasounds, which results in rather high treatment costs[45]. Also, for complete degradation (mineralization) of the pollutants to occur, high oxidant doses would be required, rendering the process economically expensive. Silva *et al*.[63] applied high ozone doses (until 3.0 g L^{-1}) to attain significant toxicity decrease. Furthermore, some intermediate oxidation products can actually raise the toxicity of the leachate. Among these processes and according to Lopez *et al*.[45], Fenton's process seems to be the best compromise because the process is technologically simple, there is no mass transfer limitation (homogeneous nature) and both iron and hydrogen peroxide are cheap and non-toxic. But Fenton's process required low pH and a modification of this parameter is necessary.

Ion exchange: Ion exchange is a reversible interchange of ions between the solid and liquid phases where there is no permanent change in the structure of the solid. This treatment is capable of effectively removing the traces of metal impurities to meet the increasingly strict discharge standards in developed countries. Prior to ion exchange, the leachate should first be subjected to a biological treatment. Although the application of ion exchange is not commonly employed for the treatment of landfill leachate, it has received considerable interest in Germany for the removal of non-biodegradable compounds that contain humic substances[15]. The removal of ammonia from landfill leachate by ion exchange was compared to that by ozonation[41].

The application of ion exchange is not economically appealing due to high operational cost. Other limitation is that, prior to ion exchange, appropriate pre-treatment system such as the removal of suspended solids from leachate is required.

Electrochemical treatment: Electrochemical treatment such as membrane electrodialysis has also contributed to environmental protection in France[2] and Brazil. In Rio Claro (Brazil), the electrodegradation of stabilized landfill leachate was investigated by employing a flow electro-chemical reactor[51]. Using a constant flow rate of 2000L h^{-1} for 180min and at a current density of 1160A m^{-2}, the maximum removal of COD and NH_3-N with initial concentrations of 1855 and 1060mg L^{-1} was found to be 73 and 49%, respectively. The results suggest that electrodegradation was an alternative means to breakdown recalcitrant organic compounds in landfill leachate. Due to high energy consumption, however, this technology is more expensive than other treatment methods. As a result, this treatment technique has been investigated less extensively for the treatment of stabilized leachate.

Membrane filtration:

Microfiltration (MF): Micro filtration is a low-pressure cross-flow membrane process for separating colloidal and suspended particles in the range of 0.05-10 microns (i.e., Fat). MF was used as a pre-treatment for another membrane process (UF, NF or RO) or in partnership with chemical treatments. But, it cannot be used alone. Only Piatkiewicz[56], in a polish study, reported the use of MF as prefiltration stage. No significant retention rate (COD reduction between 25-35%) was achieved.

Ultrafiltration (UF): Ultra filtration is a selective fractionation process utilizing pressures up to 10 bar. It concentrates suspended solids and solutes of molecular weight greater than 1,000. The permeate contains low-molecular-weight organic solutes and salts. UF is effective to eliminate the macromolecules and the particles, but it is strongly dependant on the type of material constituting the membrane. UF may be used as a tool to fractionate organic matter and so to evaluate the preponderant molecular mass of organic pollutants in a given leachate. Also, tests with membrane permeates may give information about recalcitrance and toxicity of the permeated fractions. Except Tabet *et al*.[68], UF was eliminated as a primary means for treating landfill leachate due to drastic existing regulations. These authors used membranes close to nanofiltration, leachate had a low organic matter content and local water standards were not so strict. However, Syzdek and Ahlert[67] suggested that UF might prove to be effective as a pre-treatment process for Reverse Osmosis (RO). UF can be used to remove the larger molecular weight components of leachate that tend to foul reverse osmosis membranes. The elimination of polluting substances is never complete (COD between 10 and 75%). More recently, UF has been applied to biological post-treatment of landfill leachate[67]. Finally, UF membranes have been successfully used in full scale membrane bioreactor plants by combination of membrane technology and bioreactors[68]. High treatment levels for landfill leachate have been achieved in such a process.

Nanofiltration (NF): Due to its unique properties between Ultrafiltration (UF) and Reverse Osmosis (RO) membranes, NF has found a place in the removal of recalcitrant organic compounds and heavy metals from landfill leachate[54]. This treatment process has the ability to remove particles with a molecular weight of higher than 300Da as well as inorganic substances through electrostatic interactions between the ions and membranes. The significance of this membrane lies in its surface charges, which allow charged solutes smaller than the membrane pores to be rejected, along with bigger neutral solutes and salts.

NF studied membranes are usually made of polymeric films with a molecular cut-off between 200 and 2000 Da. The high rejection rate for sulphate ions and for dissolved organic matter together with very low rejection for chloride and sodium reduces the volume of concentrate[42]. Few studies mention the use of NF to treat landfill leachates[42,50,70]. Nearly 60-70% COD and 50% ammonia were removed by NF, whatever membrane material and geometry (flat, tubular, or spiral wounded), with an average velocity of 3 m/s and a transmembrane pressure between 6 and 30 bars. Physical methods were used in combination with nanofiltration and it was found satisfactory for removal of refractory COD from the leachate used. COD removal was 70-80%[70]. Treatment of anaerobically pre-treated leachate from the Odayeri landfill (Turkey) was undertaken using NF[54].With an initial COD concentration of 3000mg/L and NH_3-N concentration of 950mg L^{-1}, about 89% COD and 72% NH_3-N removal could be achieved with total operating cost of US$ 0.8m^{-3}.

However, successful application of membrane technology requires efficient control of membrane fouling. A wide spectrum of constituents may contribute to membrane fouling in leachates nanofiltration: Dissolved organic and inorganic substances, colloidal and suspended particles[70]. In particular, natural organic matter fouling has recently gained interest[42].

Reverse Osmosis (RO): Reverse osmosis is a high-pressure, energy-efficient technique for dewatering process streams, concentrating low-molecular-weight substances in solution, or purifying wastewater. It has the ability to concentrate all dissolved and suspended solids. The permeate contains a very low concentration of dissolved solids. In the past, several studies, performed both at lab and industrial scale, have already demonstrated RO performances on the separation of pollutants from landfill leachate[42]. Values of the rejection coefficient referred to COD parameter and heavy metal concentrations higher than 98 and 99%, respectively. Tubular and spiral wounded modules were the first medium used in the early RO systems for the purification of landfill leachate starting in 1984. Depending on the salt content of the feed water and the operation time between the cleaning cycles, the operating pressure ranges between 30 and 60 bar at ambient temperature and the specific permeate flux reach 15 L^{-1} $h^{-1}m^{-2}$[42]. The average specific energy demand is low with less than 5 kWh m^{-3} of permeate for a recovery rate of 80%[55].

However, two issues have been identified and remain today, as major drawbacks for the implementation of pressure-driven membrane processes and particularly RO, to landfill leachate treatment: Membrane fouling (which requires extensive pre-treatment or chemical cleaning of the membranes, results in a short lifetime of the membranes and decreases process productivity) and the generation of large volume of concentrate (which is unusable and has to be discharged or further treated). In the early 1990's, steady improvement of membrane technology and striving for high water recoveries in landfill leachate treatment resulted in development of a high pressure RO system based on the DT-module and operating at transmembrane pressures of 120 and 200 bar. An adapted process permits to reduce certain salt fractions by controlled precipitation. This means an increase of the permeate recovery from about 80-90% with a concentration factor of 10 and a reduction of concentrate volume[42]

Table 3: Effectiveness of leachate treatments vs. leachate age

Type of treatment	Leachate age: Young	Medium	Old	Target of removal	Remarks
Channeling					
Combined treatment with domestic sewage	Good	Fair	Poor	Removal suspended solid	Excess biomass and nutrients
Recycling	Good	Fair	Poor	Improve leachate quality	Least expensive and low efficiency
Biological					
Aerobic processes	Good	Fair	Poor	Removal suspended solid	Hamper by refractory compound and Excess biomass
Anaerobic processes	Good	Fair	Poor	Removal suspended solid	Hamper by refractory compound, Longtime and biogas
Physico/chemical					
Coagulation/Flocculation	Poor	Fair	Fair	Heavy metals and suspended solids	High sludge production and subsequent disposal
Chemical precipitation	Poor	Fair	Poor	Heavy metals and NH3-N	Requires further disposal due to sludge generation
Adsorption	Poor	Fair	Good	Organic compounds	Carbon fouling can be a problem and GAC adsorption is costly
Oxidation	Poor	Fair	Fair	Organic compounds	Residual O_3
Stripping	Poor	Fair	Fair	NH3-N	Requires other equipments for air pollution control
Ion exchange	Good	Good	Good	Dissolved compounds, cations/anions	Used as a polishing step after biological treatments and treatment cost is high
Membrane filtration					
Micrfiltration	Poor	-	-	Suspended solids	Used after metal precipitation
Ultrafiltration	Poor	-	-	High molecular weight compounds	Costly and limited applicability due to membrane fouling
Nanofiltration	Good	Good	Good	Sulphate salts and, hardness ions	Costly and requires lower pressure than reverse osmosis
Reverse Osmosis	Good	Good	Good	Organic and inorganic compounds	Costly and extensive pre-treatment is required prior to RO

CONCLUSION

Optimal leachate treatment, in order to fully reduce the negative impact on the environment, is today's challenge. But, the complexity of the leachate composition makes it very difficult to formulate general recommendations. Variations in leachates, in particular their variation both over time and from site to site, means that the most appropriate treatment should be simple, universal and adaptable. The various methods presented in the previous sections offer each advantages and disadvantages with respect to certain facets of the problem. Suitable treatment strategy depends on major criteria:

- The leachate characteristics such as COD, BOD, NH_3-N and age of leachate. The knowledge of these specific parameters may help to select suitable treatment processes (Table 3)
- The final requirements given by local discharge water standards

During many years, conventional biological treatments and classical physico-chemical methods are being considered as the most appropriate technologies for manipulation and management of high strength effluents like landfill leachates. When, treating young leachate, biological techniques can yield a reasonable treatment performance with respect to COD, NH_3-N and heavy metals. When treating stabilized (less biodegradable) leachate, physico-chemical treatments have been found to be suitable as a refining step for biologically treated leachate, in order to remove organic refractory substances. The integrated chemical-physical-biological processes (whatever the order) ameliorates the drawbacks of individual processes contributing to a higher efficacity of the overall treatment.

In the recent years, with the continuous hardening of the discharge standards in most countries and the ageing of landfill sites with more and more stabilized leachates, conventional treatments (biological or physico-chemical) are not sufficient anymore to reach the level of purification needed to fully reduce the negative impact of landfill leachates on the environment. It implies that new treatment alternatives species must be proposed. Therefore, in the last 20 years, more effective treatments based on membrane technology has emerged as a viable treatment alternative to comply and pending water quality regulations in most countries.

Today, membrane processes and most particularly RO and NF offer the best solution and have been proved to be the more efficient, adaptable and indispensable means of both:

- Achieving full purification (rejection rates of 98-99% for RO)and
- Solving the growing problem of water pollution.

However, landfill leachate RO feasibility is highly conditioned by the control of concentrate treatment costs and the choice of the feed pre-treatment mode in order to reduce membrane fouling. Biological pre-treatment are often proved ineffective as RO pre-treatment[1,75]. On the contrary, lime precipitation appears like a promising option for the pretreatment of RO membranes and the removal of colloidal particles and organic macromolecules that arethe principal RO foulants of landfill leachates[75]. In the same way, microfiltration and ultrafiltration have proved to be suitable, provided that they are preceded byphysico/chemical process as lime precipitation[67].

Residue production, which constitute a capital environmental concern, still remain major hurdle, since it is usually unusable and has to be discharged, further treated or landfilled. The transport to an incineration plant equipped for the burning of liquid hazardous waste remains the preferred option (in spite of many controversies) but leads to high treatment costs.

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