

Review

Novel and Conventional Technologies for Landfill Leachates Treatment: A Review

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Abstract: Municipal solid waste final disposal represents an environmental burden worldwide since landfilling, or open dumping, is still the preferred solution for the end of life of solid discarded materials. This study aims to review the technological innovations applied for landfill leachate treatment, taking into consideration the experiences obtained during the past years and the solutions which have been implemented. The review showed that both biological and physiochemical treatments are not able to achieve the requested water quality level, according to the limits established by regulations, whether applied in a single treatment or multiple treatments. In order to respect sustainable release limits to guarantee environmental protection, the construction of depuration systems and combining biological and physiochemical treatment methods is considered of the utmost importance. The review looks at possible joint applications of different treatment techniques reviewed by other studies and considers the state of the art of current research. Combined technical solutions suggested within the 2016 peer-reviewed papers are presented and discussed as a sustainable way to effectively treat landfill leachate, giving particular attention to feasible solutions for developing countries.

Keywords: landfill leachate; biological treatments; innovative technologies; combine treatment solutions; developing countries; review

1. Introduction

Municipal solid waste (MSW) final disposal represents an environmental burden worldwide since landfilling, or open dumping, is still the preferred solution. A lack of suitable technological choices, especially in developing countries [1,2], means environmental pollution and risk for the health of the population due to the exposure to toxic compounds caused by the release of untreated leachate [3] and the dispersion of micro-pollutants into the atmosphere [4]. Dump sites in developing and emerging countries suffer from the lack of leachate treatment and caption facilities, causing the contamination of water bodies and soil and threatening human health [5,6].

Generally, the project developer's objective for leachate management should be the implementation of highly cost-effective systems with low area requirements, and simple implementation and maintenance [7]. To date, there are clear directions for the reduction of MSW inflow to dump sites, following circular economy and 3Rs (reduce, reuse and recycle) principles which consist of opportunities to improve waste management through introducing waste introduction back into productive use [8].

However, final disposal remains the common solution. Also, developed countries did not improve the “zero-waste” perspective effectively [9]; for instance, in Italy 47% of MSW is disposed to landfills, while in Greece the number is over 80%, according to recent surveys [10]. This is due to several reasons, which include economic, environmental, political, technological and social aspects.

Landfill rainwater infiltrations, added to other chemical and physical phenomena, support the waste in undergoing a series of decomposition phases which allows the generation of leachate [11]. Despite that, the composition of leachate varies significantly among landfills depending on the waste composition, waste age, and landfilling technology [12]. Pollutants in MSW landfill leachate can be divided into four main groups: dissolved organic matter, quantified as chemical oxygen demand (COD); inorganic macro-components; heavy metals; and xenobiotic organics. However, other compounds may be found in leachate from landfills such as borate, sulfide, arsenate, barium, lithium, mercury and cobalt [11].

For these reasons, landfill leachate treatment became a relevant technical challenge, since many technologies are available to treat different leachate typologies. Notwithstanding, holistic approaches are essential to find suitable treatment techniques in order to safeguard environmental and human livelihood, as suggested in other reviews [13]. Moreover, due to the variability in leachate composition, there is a lack of unique and flexible ways to treat it [14]. That is why it is important to introduce integrated, modular, multi-stage, systems as well as options for coupling technologies with various physical-chemical or other biological processes which should treat each leachate typology as a function of its pollutant concentrations [7].

The present review aims to summarize and analyze the recent developments in landfill leachate treatment, particularly for MSW final disposal sites, focusing on the quantitative and qualitative aspects of pollutant removal, describing the most common biological, physical and chemical treatments applied. Moreover, discussions about the combination of physical-chemical and biological processes are carried out in order to suggest which arrangements are useful in treating landfill leachate, giving examples from recent years. For that purpose, Section 3.3 summarizes a review of some leachate treatment technologies applied together in order to achieve a significant reduction of biological and chemical pollutants while Section 4 discusses the results found. Leachate transfer systems such as recycling and combined treatment with domestic sewage are not discussed singularly, though they are mentioned within the associated methodologies. We suggest this review for policy-makers and plan developers who need to choose the most feasible solution for handling disposal site releases.

2. Leachate Characteristics and Main Issues

Landfill leachate production is a consequence of the MSW decomposition associated with storm water filtration. Biochemical reactions, established within the waste, allow the generation of leachate and byproducts in a sequence of four main stages [14]:

- Step 1: aerobic phase;
- Step 2: anaerobic and acidogenic phase;
- Step 3: methanogenic phase (unstable);
- Step 4: stable methanogenic phase.

Risks associated with the dispersion of leachates are mainly due to high pollutant concentrations and the non-homogenous nature of the substances. This aspect plays a key role in implementing treatment processes, since there is no single treatment available for all landfill leachate typologies [14]. The non-homogenous nature of the pollutants is due to the numerous biochemical reactions diffuse within the waste piles that contribute to leachate generation. According to other studies [14,15], generally the leachate composition ranges are 100–50,000 mg/L chemical oxygen demand (COD), 3–25,000 mg/L biological oxygen demand (BOD), 5–11 pH, 13–5000 suspended solids (SS), and 10–13,000 mg/L nitrates ($\text{NH}_4\text{-N}$), which are functions of the numerous factors that influence the compound quality; the most important factor is the waste age. Indeed, time influences the COD, BOD_5

and the ratio of BOD₅:COD, since they decrease with the time while the pH value increases [15]. Typically, according to other reviews [13,16], young leachate is characterized by a high organic fraction (BOD₅/COD < 0.6) and low pH (<6.5) while old leachate (six years old or older) presents low biodegradability (BOD₅/COD < 0.1), high ammonium amounts (>400 mg/L) and pH > 7.5.

Another important aspect is the evaluation of leachate amounts that will be produced by the landfill. Indeed, disposal costs and individual technical component dimensions, such as leachate collection sewers and impermeable layers, can be estimated within a project even if leachate flows are known. Leachate amounts, very often, are obtained by simple empirical expression functions of hydrological balances. Equation (1) gives an example [17]. Considering a landfill sector already filled and covered such as a drainage basin, in which both waste layers and the final capping have already been deposited, the amount of leachate is related to the mass balance relative to the water inflows and outflows in the sector:

$$L = P - R + R^* - ET + J + IS + IG + (\Delta US - \Delta UW) + b \quad (1)$$

where:

L = volume of leachate;

P = rainfall;

R = surface runoff;

R* = surface runoff from external areas;

ET = evapotranspiration;

J = irrigation and/or recirculation of leachate;

IS = infiltration water from surface water bodies;

IG = infiltration water from groundwater;

ΔUS = variations of water content in the capping material;

ΔUW = variation of water content in the amount of disposed waste;

b = production or consumption of water associated with the different aerobic and anaerobic biochemical degradation reactions of organic substances.

Water balances highlighted a few aspects [15]:

- leachate is formed mainly in wet and rainy areas;
- leachate is extremely variable and follows the precipitation trends;
- leachate amounts are a function of the efficiency of the coverage of capping and continue to be generated for long periods.

During operative periods, the application of the hydrologic balance method is very difficult due to the inability to take into account all the variables that contribute to the balance. Managerial variables are the ones that affect mostly leachate production, such as waste disposition methods or the land slope, while the post-closing phase allows physical and geometrical parameters to be constant and clearly definable. As a result, leachate composition and flow may vary considerably case by case [14]; hence, specific surveys are required for each specific disposal site in order to implement the most suitable leachate treatment plant.

3. Review of the Main Landfill Leachate Treatment Technologies

According to other reviews [7,18], landfill leachate treatments are generally classified into three main groups:

- (a) biological processes (aerobic or anaerobic);
- (b) chemical and physical processes;
- (c) a combination of physical-chemical and biological processes.

These processes are going to be introduced and discussed in the sections below, with particular attention on combined treatment processes, as an answer for the technical issues and pollutant removal rates required.

3.1. Biological Treatment

Biological treatments are useful to treat leachate with high amounts of organic substances. The effectiveness of the treatment decreases with the increase in the landfill age, since biodegradable organic matter reduces over time and landfill leachate is getting stabilized. Biological treatment can be divided into aerobic and anaerobic:

- Aerobic treatment allows reducing organic pollutants and is able to accomplish nitrification processes. It exhibits rapid removal kinetics, low sensitivity for the presence of toxic substances and considerable efficiency in ammonia stripping. As disadvantages, there is a remarkable production of excess sludge and great energy costs due to the high amount of oxygen required.
- Anaerobic and anoxic processes are based on the activity of microorganisms able to break down organic matter within the environment with no dissolved oxygen. Notwithstanding the several benefits of the anaerobic treatment, the application processes are limited, mainly due to the low growth rate of anaerobic microorganisms, ineffective $\text{NH}_4\text{-N}$ removal and poor retention of biomass [19]. These processes do not require aeration systems, and thus treatment costs are contained, also allowing energy recovery by biogas collection and exploitation. They are characterized by low reaction kinetics and low biomass growth as compared to aerobic systems.

Aerobic systems may act as a combined aerobic–anoxic–anaerobic system whether low oxygen is provided or oxygen-deprived zones are created by diffusion or mixing conditions. As a result, this allows combining removal processes: anaerobic and aerobic treatments are applied together for wastewater management mostly to remove organic substances and nitrogen-based compounds by nitrification and denitrification processes. Nitrification involves two steps—the oxidation of ammonia to nitrite by ammonium-oxidizing bacteria (AOB), followed by the oxidation of nitrite by nitrite-oxidizing bacteria (NOB), whereas denitrification involves denitrifying bacteria for the conversion of nitrate to nitrogen gas [20].

The use of biological treatment processes, aerobic, anaerobic or anaerobic/aerobic, for municipal landfill leachate usually results in low COD and BOD removal values, mainly because of the high ammonia concentration and the presence of refractory organic compounds. To improve the efficiency of the biological treatment, pre-treatment to reduce the concentration of these two components has to be considered [21]. The principal biological treatments are presented below and pollutant removal yields are summarized in Table 1.

Table 1. Leachate pollutant removal rates by biological treatment.

Technology	References	Pollutant Removal Rates (%)										
		COD	BOD	NH ₄ -N	TN	PO ₄ ³⁻	Cl ⁻	Heavy Metals	Fe	SS	SO ₄ ²⁻	Turbidity
Aerobic Methods												
Lagoons	[22]	40	64	77		42	27		30		44	
Constructed Wetlands	[23]	50	59	51		53	35		84		49	
Rotating Biological Contactors	[24]	38	80	98								
Sequencing Batch Reactor	[25,26]	76, 85	84, /	65, 55		23, /	26, /			62, /		
Trickling Filters	[26]	49	77	59.5		56				73		72
MBBR	[27]	60–81		92–95								
FBBR	[28]	85			80	70						
MBR	[29,30]	71, 79	93, 99	63, 60		87, /						
SHARON	[31]			90–98								
Anaerobic and Anoxic Methods												
UASB	[32,33]	42, 55–75	/, 72–95	48, /						45, 45		
SAMBR	[34]	90	88							100		
AF	[35,36]	90, 90										
Anammox	[37,38]	62, 14–16			94, 80–94							

Note: /: Not Available.

3.1.1. Aerobic Treatments

Aerated Lagoons

One of the simplest forms of onsite treatment of landfill leachate, usually applied in developing countries, is aerated lagoons (or stabilization ponds) where treatment occurs via biological oxidation. However, if the aerated lagoon method is adopted as a full-scale leachate treatment, hydraulic retention times (HRT) need to be fully evaluated since they can be significant. In the most practical full-scale applications, HRT is determined by the inflow of leachate flow rates into the reactors which are usually simply ponds dug in the soil [39]. COD removal can vary between 1% and 95% since higher COD removal efficiencies may be associated with younger leachates, which hold higher concentrations of COD. Nitrification accounts for 63% of ammonium removal but a significant part (37%) of the ammonium could not be accounted for by nitrification [39]. Other studies, also reported in previous reviews [15], introduced leachate treatment plants where a two-stage anaerobic/facultative lagoon system, with an anaerobic section characterized by a 32-day retention time and a 22.8 °C temperature and a facultative section characterized by 240-day retention time and a 14.3 °C temperature, achieved removal rates equal to COD 40%, BOD 64%, $\text{NH}_4\text{-N}$ 77%, $\text{NO}_3\text{-N}$ 63%, P 42%, Cl^- 27%, SO_4^{2-} 44%, Fe 30% [22]. Despite that, this treatment is suggested in countries with extensive free areas, with no energy available and low economic funds.

Constructed Wetlands (CW)

CWs for wastewater treatment involve the use of engineered systems that are designed and constructed in order to use natural processes. Indeed, these systems are designed to mimic natural wetland systems, utilizing wetland plants, soil, and associated microorganisms to remove contaminants from wastewater effluents [40]. Overall, studies into land irrigation treatment of landfill leachate conclude that this management practice may offer an environmentally acceptable, economic and effective form of treatment [41]. The extensive root system of the weed provides a large surface area for attached microorganisms, increasing the potential for the decomposition of organic matter: Nitrogen and phosphorus are removed through plant uptake (with harvesting) while ammonia is removed through volatilization and nitrification/denitrification [40].

Despite its potential treatment, CW use on a full scale in developed countries has not been extensively pursued due to poor performance in winter, as CW gives its optimum growing temperature range between 20 °C and 30 °C, and for the economic feasibility of the systems due to large land acquisition [40].

CWs were developed as a pilot integrated system for old sanitary landfill sites, achieving high removal efficiencies proving that leachate can be treated effectively [42]. For instance, Bulc [23] introduced a CW consisting of three interconnected beds, two with a vertical flow and one with a horizontal flow stage, covering 311 m² with an intermittent hydraulic load of 0.5 cm/d, filled with sand media and planted with reeds. Removal efficiencies were: COD 50%, BOD₅ 59%, ammonia nitrogen 51%, total phosphorus 53%, sulfides 49%, chlorides 35%, and Fe 84%, with negative effects for nitrate and sulfates [23]. Despite that, in the case of vertical subsurface flow CWs, the COD removal efficiency can be improved by introducing the intermittent recirculation of the treated wastewater from the bottom to the top of the bed and intermittent artificial aeration supplied at the bottom of the bed: 90% COD removal has been achieved at a pilot scale [43]. Moreover, vertical flow CWs were used to remove phenol, bisphenol A (BPA), and 4-tert-butylphenol (4-t-BP) from synthetic young and old leachate with removal percentages of: phenol (88%–100%) > 4-t-BP (18%–100%) ≥ BPA (9%–99%) [44], and chromium from contaminated irrigation water with a total chromium removal range from 55% to 61% [45].

In tropical regions CWs should achieve not only pollutant removal but also landfill leachate volume-saving, reducing environmental pollution and preventing the uncontrollable dispersion of polluted leachate [46]. So, the potential for the application of wetland technology in developing countries is enormous as most of the emerging cities worldwide have warm tropical and subtropical

climates that are conducive for higher biological activity and productivity. Furthermore, tropical and subtropical regions are known to sustain a rich diversity of biota that may be used in wetlands, guaranteeing a better performance of wetland systems. Although land may be a limiting factor in dense urban areas, CWs are potentially well suited to smaller communities where municipal land surrounding schools, hospitals, hotels and rural areas is not in short supply [40].

Aerated Reactors

Aerated reactors are aerobic treatments, based on the use of continued aeration with a pre-established large bacterial population (activated sludge), mainly used for municipal wastewater treatment. Activated sludge is the mixture of microorganisms, which are intensified by their confinement within reactors, that allows the degradation of pollutant compounds. In order to achieve biological nitrogen removal it is necessary to introduce a denitrification step since the process may provide pre-/post-denitrification or complete denitrification, depending on the biodegradable internal carbon source. However, the activated sludge process is not adequate for the leachate treatment due to the significant disadvantages of this treatment [15]:

- high sludge production, which involves considerable costs for disposal;
- significant energy demand;
- the presence of inhibitor microorganisms due to the high concentrations of $\text{NH}_4\text{-N}$.

Therefore, alternative techniques should be developed to enable more efficient COD and nitrogen removal in order to save economic outgoings and energy consumption.

Rotating Biological Contactors (RBCs)

An RBC is an attached growth bioreactor that offers an alternative technology to the conventional activated sludge process. An RBC unit typically consists of a series of closely spaced large, flat or corrugated discs that are mounted on a common horizontal shaft and are partially or completely submerged in wastewater [47]. The shaft continually rotates by a mechanical motor or a compressed air drive and a biofilm is established onto the entire surface area of the media, which metabolizes the organic materials contained in the wastewater. In aerobic processes, the rotation of the media promotes oxygen transfer and maintains the biomass in aerobic conditions, also providing turbulence to the surface, allowing the removal of excess solids from the media. The performance of RBCs depends upon several design parameters such as rotational speed, organic and hydraulic loading rates, hydraulic retention time (HRT), RBC media, temperature, wastewater and biofilm characteristics, dissolved oxygen levels, effluent and solids recirculation, step-feeding and medium submergence [47].

Leachate treatment from an old solid waste landfill with RBCs was carried out by a few studies, removing a maximum of 80% BOD₅, 38% COD, and 98% of the ammonia in the leachate. So, while the RBC treatment was successful in removing nitrogen species and a large fraction of the soluble organic matter from the leachate, a significant amount of non-degradable COD remained after treatment [24]. Hence, RBC is not suited to treat landfill leachate alone and should be joined with other methods such as biological anaerobic systems [48] or physical-chemical technologies. Moreover, RBCs have some operating problems such as difficulty in the maintenance of an appropriate biofilm thickness under adverse conditions [47].

Sequencing Batch Reactor (SBR)

SBRs are systems enabled to treat wastewater or landfill leachate within the same tank. The operation systems used in aerobic conditions comprise four steps: (1) feeding; (2) aeration; (3) settling; and (4) discharge [49].

Due to the consumption of carbon sources in the aeration stage, wastewater does not have a sufficient carbon source in the following anoxic denitrification stage and it leads to low total nitrogen (TN) removal efficiency [50]. Moreover, the organics biodegradation rate in the SBR decreases as the

influent ammonium increase, suggesting ammonium pre-treatment in these cases [50]. Therefore, SBR cycles should be modified to effectively use the carbon source contained in the landfill leachate with low C/N ratios (<4) and to achieve advanced nitrogen removal without the addition of external carbon sources. A modified SBR can be made by anaerobic–aerobic–anoxic sequence processes and can be developed in order to use landfill leachate organics fractions. The feature of the modified SBR process is the addition of the anaerobic stage after the feeding, so that microorganisms can store the organic matter and supply a carbon source for endogenous denitrification after the aeration stage [51]. However, denitrification cannot be implemented; hence, ammonia reduction does not achieve high rates. Obtained removals are equal to 85% for COD and 55% for ammonia [25]. According to Aluko et al. [26], leachate treatment by SBR allows achieving removal rates such as: COD 76%, PO_4^{4-} -P 23%, ammonia 64.83%, suspended solids 62.28%, BOD₅ 84.06%, chloride 26.31%, along with the removal of metals such as cadmium, iron and zinc ($>60\%$).

Trickling Filters (TFs)

TFs consist of a fixed bed of granular material where wastewater flows and generates a biofilm. Usually aerobic conditions are maintained by forced air flowing through the bed or by natural convection. There are numerous types of filling materials, all of which contribute to the removal of pollutants in different ways which can be reused materials: plastic, wood or rubber from tires [52,53]. TFs cannot be applicable for wastewater treatment with a high organic matter concentration, such as leachates from young landfills, due to the unavoidable obstructions caused by the biomass and large amounts of big materials fractions. Therefore, TFs can be used only for the biological treatment of old leachate.

As suggested by other studies, by means of TF COD removal can achieve the 49%, PO_4 -P removal the 56%, and ammonia removal the 59.50%, along with significant reductions in suspended solids (73.17%), turbidity (71.96%) and BOD₅ (76.69%). However, the concentration of NO_3 -N rises due to the nitrification process [26]. However, in specific conditions, NH_4 -N removal reaches 90% and can be used as a good option for ammonia removal [54].

Moving Bed Bioreactor (MBBR)

MBBR is a biofilter that combines a high specific surface without the possibility of clogging. In this reactor, the biofilm is growing on carriers circulating inside the tank shaped to maximize growth by protecting the biofilm from abrasion [55]. The media carriers may have different shapes and sizes depending on the application. They can occupy different volumes in the reactor, from 30% to 60%, depending on the amount of required biomass.

Since the reactor generally consists of the oxidation, nitrification and denitrification stages, it is designed to be particularly effective for the removal of nitrogen, phosphorus and COD. The efficiency of leachate nitrogen removal could reach values around 90%, but only 20% for COD [56]. Improving operating conditions, the MBBR process allows removing 85%–90% of the ammonia and 60%–81% of the COD contained in landfill leachate [27]. Moreover, introducing aerobic conditions after anaerobic stages, COD removal reaches 92%–95% while ammonia reaches over 97%, making the MBBR process attractive and promising to apply in the treatment of highly concentrated wastewater [57].

MBBR treatment is an advantageous process for the reduction of more than 70% of the volumes needed for the tanks, thanks to the more efficient kinetics; continuous running, as MBBR are not subject to clogging; removal of organic and nitrogen compounds through a single process; and low sensitivity to toxic substances. However, it requires ventilation with medium to large bubbles which means large amounts of energy are required.

Fluidized Bed Bioreactors (FBBR)

FBBR are initially fixed beds of solid particles brought to a fluidized state by an upward stream of gas or liquid as soon as the flow rate of the fluid exceeds a certain limiting value. With further

increasing the flow rate the bed begins to expand uniformly, reaching the fluidized condition [58]. The fluidized bed can be made by different materials, such as lava rock [59] or sulfur particles [60], but the most common is sand.

Fluidized-sand beds are an efficient, relatively compact, and cost-competitive technology for removing dissolved wastes from wastewater. Individual fluidized-sand beds can treat both small and large flows, with single beds treating as much as 190 L/s of water flow [61]. A fluidized-sand bed is approximately a linear flow reactor where biofilm grows around individual sand grains, decreasing the density of the particles, causing the particles to migrate toward the top of the biofilter, and increasing the total expansion of the biofilter bed. The actively growing microbial biomass usually requires management, which is typically accomplished by siphoning the thickest and oldest biofilm-coated particles that migrate to the top portion of the bed. Smaller sand sizes (effective size of 0.13–0.25 mm) allow the attachment of more biofilm, but require increased management to prevent the sand bed from overflowing at the biofilter outlet [62].

FBBRs with anoxic and aerobic beds are implemented to treat municipal wastewater, employing lava rock as a carrier media for the simultaneous removal of carbon, nitrogen and phosphorus. This treatment was successfully demonstrated to remove 95% TSS, 93% COD, 97% $\text{NH}_4\text{-N}$, $\text{NO}_3\text{-N}$, 78% total nitrogen and 79% $\text{PO}_4\text{-P}$, with outflow concentrations as low as 4–6, 18–20, 0.35–0.7, 5.8–6.5, 7.5–8.3 and 0.7 mg/L, respectively [59]. Liquid-solid circulating FBBRs were used to optimize carbon and nutrient removal capabilities from leachate using 600 μm lava rock with a total porosity of 61%. Maintaining liquid recirculation with the amount of solids in the columns and air injected, the technology achieved COD, nitrogen, and phosphorus removal efficiencies of 85%, 80%, and 70%, respectively, at nutrients loading rates of 2.15 kg COD/($\text{m}^3\cdot\text{d}$) [28].

Generally, FBBRs can be relatively easy to manage because they do not filter solids from the passing flow and the actively growing microbial biomass in the expanded bed can be readily harvested by siphoning the lightest biofilm-coated particles from the top of the bed. On the down-side, fluidized-sand beds are relatively complex to design [61]. Sometimes FBBRs are not aerated, as trickling filters are. Therefore, they should always be designed with a cascade column placed immediately downstream to strip dissolved carbon dioxide and bring the dissolved oxygen up to near 90% saturation. Moreover, whether water flow through a FBBR ceases for more than approximately 6–24 h, depending upon conditions, the static but biologically active bed can turn anaerobic, resulting in a significant loss in nitrification capacity [61]. So, the FBBR can be a feasible way to treat landfill leachate; however, technological foresight must be taken into account, considering specific landfill streams and economic conditions. Indeed, FBBRs allow saving space while increasing technological issues.

Membrane Biological Reactor (MBR)

The MBR system combines a conventional biological activated sludge process with a membrane separation process. The membrane bioreactors are divided, depending on the positioning of the membrane with respect to the organic sector, in submerged membranes (S-MBRs) and outer membranes (R-MBRs) [63]. In the first case the membranes are immersed within wastewater, in the oxidation tank. Through a pump, a slight depression is created inside the filter module, allowing the treated effluent to pass through the membranes, thus obtaining an efficient solids separation without the need to proceed with further sedimentation treatments. In the second case, membranes are positioned external to the aeration tank: the effluent from the oxidation tank is pumped into the membrane filtration module, while the sludge is recirculated into the bioreactor.

Removal efficiency rates, both for old and young leachate, change as a function of the membrane type, which can be a flat-sheet MBR and a hollow-fiber MBR: PO_4^{3-} , BOD and COD removal rates are similar and achieve 87%–81%, 92%–93%, and 71%–68%, respectively. Despite that, the flat-sheet membrane achieved significantly higher total nitrogen and ammonium removal rates when compared to the rates for the hollow-fiber membrane: 61.2% vs. 49.4% for TN and 63.4% vs. 47.8% for ammonium [29]. Temperature also plays a vital role in the removal efficiency; therefore,

a mesophilic temperature is usually applied (30–45 °C) for the treatment. The COD removal can increase to 79%, while the percentage of the ammonia removal decreases from 75% to 60% along with the gradual increase of the BOD removal at 97%–99% [30]. These results indicate that treatment of leachate with MBR, applying a temperature slightly above 45 °C, is suitable for the removal of COD and BOD, especially in the case in which the leachate to be treated has a high concentration of organic matter. However, this system is not efficient for the removal of nitrogen, and as a result, the appropriate pretreatment of wastewater is often considered as a necessary procedure and the coagulation/flocculation process was proposed recently as a relevant viable option for MBRs [64].

The MBR effectively produces a clarified and substantially disinfected effluent. In addition, it concentrates the biomass and reduces the necessary tank size, increasing the bio-treatment process efficiency. MBRs thus tend to generate treated waters of higher purity with respect to dissolved constituents such as organic matter and ammonia, both of which are significantly removed by bio-treatment [65]. However, membrane fouling is considered the main drawback, due to the interaction between the membrane and fouling agents, leading to the membrane's efficiency deterioration [66,67]. Moreover, membrane fouling was found to be most severe at pH 5.5, followed by at pH 8.5, while the fouling at pH 6.5 and 7.5 was quite similar and less severe than that at pH 5.5 and 8.5 [68], identifying MBR technology as an expensive and dangerous technical choice for leachate treatment.

Despite that, MBR produces less sludge than a suspended biomass system, is a more compact structure with a high biomass concentration, produces a high-quality effluent, is a versatile technology that can treat both old and young landfill leachate and is effective in organic micropollutant removal, particularly bisphenol A and nonylphenol [69].

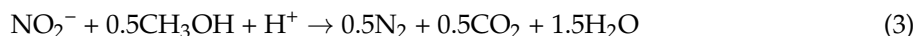
Membrane-Aerated Biofilm Reactor (MABR)

An emerging technology that has the potential to increase energy efficiency in biological wastewater treatment is the membrane-aerated biofilm reactor (MABR), also known as the membrane biofilm reactor (MBfR). The biofilm grows on a gas-permeable membrane in order to play the role as both a support and an oxygen-supplying source for the biofilm [70]. The main advantage of the MABR, in contrast to other membrane technologies, is that the membrane does not act as a filter for the wastewater, but rather it is used to deliver the oxygen required for wastewater treatment without any bubble formation, thus overcoming the oxygen supply constraint [71,72].

MABR for landfill leachate treatment can achieve 80%–99% of nitrification with oxygen transfer rates as high as 35 g O₂/m²-day [73]. When the influent COD/N rate is 5 or higher, the COD, ammonium and total nitrogen removal efficiencies reach 83.7%, 93.1% and 84.6%, respectively [74]. The results suggested that MABR technology offers a good option for effective leachate treatment; however, it is used mostly for urban wastewater with a high BOD concentration and low chemical pollutant amounts.

Single Reactor High Activity Ammonium Removal Over Nitrite (SHARON)

The SHARON process is a partial nitrification system to oxidize half the influent ammonia nitrogen to nitrite. As this partial nitrification process is limited to nitrite rather than nitrate in the conventional process, 25% of the aeration energy is saved, 30% of the sludge is reduced, and overall 20% less CO₂ is emitted [20]. This method is usually operated in a continuous stirred tank reactor (CSTR) or swim-bed reactors (SBRs) without biomass retention, where aerated and non-aerated periods are alternated to reach nitrification and denitrification processes, respectively. In aerobic conditions, the pH undergoes variations as a consequence of nitrification and dioxide carbon stripping processes. During nitrification, the pH decreases (Equation (2)) while in anoxic conditions the pH increases due to the denitrification process (Equation (3)) [75].



Of the various processes, the SHARON process appears to be a practicable way to treat landfill leachate to substantially reduce the concentration of ammonium, reaching 90%–98% of ammonium removal. This can be achieved as long as operations are carried out at an elevated temperature and pH [31].

The process requires relatively little initial investment because a simple well-mixed tank reactor of modest dimensions without sludge retention is sufficient. The process does not produce chemical sludge and has a relatively low production of biological one. Compared to the traditional nitrification and denitrification via nitrate, the SHARON process demands 25% less aeration energy and 40% less added carbon [31].

3.1.2. Anaerobic and Anoxic Treatment

Up-Flow Anaerobic Sludge Blanket (UASB)

The UASB system is an anaerobic treatment characterized by a metabolically active granular sludge with significant settling characteristics ensuring that excellent biomass retention occurs independent of the hydraulic retention time (HRT), achieving soluble COD removal rates between 85% and 90% [76]. Retention of active sludge within the UASB reactor enables good treatment performance at high organic loading rates. Natural turbulence caused by the influent flow and the biogas production provides good wastewater-biomass contact in UASBs, hence higher organic loads can be applied in UASB systems. Therefore, less reactor volume and space are required while, at the same time, high-grade energy is produced as a biogas that can be exploited. Several configurations can be imagined for a wastewater treatment plant including a UASB reactor; in any case, they must include screens for coarse material and drying beds for the sludge. However, the effluent from UASB reactors usually needs further treatment in order to remove remnant organic matter, nutrients and pathogens. This post-treatment can be accomplished in conventional aerobic systems such as stabilization ponds, activated sludge plants, and others [77].

The UASB reactor normally operates at temperatures between 20 °C and 35 °C, reaching around 70% removal of COD, and reaching 80% at temperatures around 35 °C. For instance, landfill leachate inflow to UASBs with a HRT of 1.5 days, an organic loading rate of 6.73 kg COD/m³-day and with an operation temperature of 35 °C can be treated and achieve removal efficiencies of total COD, soluble COD, total solids, volatile solids, total phosphorus, ammonia nitrogen of 42.2%, 58.1%, 45.3%, 68.2%, 44.3%, 47.8%, respectively [32]. At lower temperatures, such as 13–14 °C, 50%–55% COD and 72% BOD₇ removals were obtained with organic load rates of 1.4–2 kg COD/m³-day, while up to 75% COD and up to 95% BOD₇ removals were achievable at temperatures of 20 °C with 4 kg COD/m³-day organic loads rate [33]. The main disadvantage of this treatment is the high sensitivity to toxic substances and the limited NH₄-N removal [78]. Therefore, the application of UASB for landfill leachate treatment is not common, at least applied with other technologies.

Submerged Anaerobic MBR (SAMBR)

The SAMBR is an anaerobic bioreactor coupled with membrane filtration. It has the ability to produce effluents similar in quality to those generated during aerobic treatment, while recovering energy and producing substantially less residuals. The majority of studies at the bench scale and pilot scale indicated an adequate treatment performance at HRTs comparable to those used in aerobic treatment and at low temperatures [79].

For municipal wastewater, using SAMBR technology, the removal of the total suspended solids, soluble COD and BOD were, respectively, 100%, 90% and 88% [34]. The behavior of landfill leachate treated by SAMBR is similar with a HRT equal to two days and a leachate percentage within the reactor of about 20%. However, a gradual decrease in organic removals was observed as the leachate percentage increased [80] and HRT decreased [81].

The advantage of the process is biogas production, exploitable for energy recovery (0.160 L/g COD removed) [82] concurrently with organics removal. Nevertheless, high energy amounts are required and technical issues are common due to membrane fouling.

Anaerobic Filter (AF)

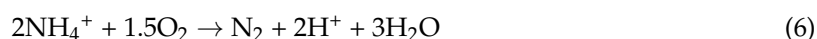
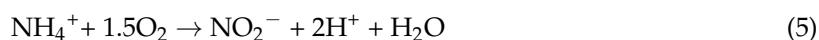
The AF is a fixed-bed biological reactor mostly used for food industries' wastewater treatment. It uses a submerged bed of packed media such as pieces of polyvinyl chloride (PVC) plastic, etched glass and baked clay. Commercial media for use in AF are available either in the form of loose fill or modular blocks fabricated from corrugated plastic sheets. The reactor packed with media with an open-pored surface texture exhibited 78% COD removal efficiency, likely due to its higher retention of attached biofilm [83]. The biomass is grouped into three main parts: biofilm at the bottom, which is the largest amount; biofilm at the top, which has the highest specific methanogenic activity; and nonattached biomass. The bacteria remain in the filter, providing a long solid retention time, even though the HRT is much shorter. Soluble organic compounds in the influent wastewater pass in close proximity to the biomass and diffuse into the surfaces of the attached or granulated solids. Here they are converted to intermediates and to end products, specifically methane and carbon dioxide [84].

As introduced by other reviews [35], the ability of the anaerobic filters to reduce COD in leachate from a partially stabilized (COD 3750 mg/L; BOD/COD = 0.3) landfill and from a relatively new landfill (COD 14,000 mg/L; BOD/COD = 0.7) has been demonstrated by Henry et al. [85], where substantial COD removals (90%) at organic loadings between 1 and 2 kg COD m³/d were achieved at room temperature (21–25 °C) with HRTs of 24–96 h without any supplemental phosphorus injection. However, the accumulation of the dissolved sulphide concentration in an AF, when treating a highly alkaline, high-strength and sulphate-rich landfill leachate, inhibits both the activity of the sulphate reduction and methanogenic processes. Wang et al. [36] suggested adding FeCl₃ in order to improve the process performance and methane production. Indeed, during the initial period when FeCl₃ was not added, COD was removed but with a declining efficiency. The efficiency of COD removal increased at the point of FeCl₃ addition, which reduced soluble sulphides and allowed the establishment of an active methanogenic population to work in parallel with the sulphate-reducing population. At a loading of 0.76 kg COD m³/d, on average 90% of the influent leachate COD was removed. This declined progressively as the process loading increased step-wise, reaching a level of 73% at a loading of 4.58 kg COD m³/d; after this point there was a sharp decline in the COD removal efficiency when the loading was increased to 7.63 kg COD m³/d at a nominal retention time of three days. Therefore, the gradual decline in the COD removal efficiency could be a result of the decreasing retention time in the reactor [36].

AF has several advantages over aerobic and anaerobic processes. It is more suitable for handling high-pollutional-load wastewaters as it presents high substrate removal efficiencies at short hydraulic retention times and high organic loading rates. It tolerates high hydraulic and organic overloadings and it operates at lower HRTs, thus requiring smaller volumes. It is less expensive to construct, to operate and to maintain since the volume of the sludge is low. It has adequate performance at ambient temperature, and is less sensitive to moderate changes in pH or temperature, rendering AF extremely useful for the treatment of seasonal effluents, such as landfill leachate [84]. Nevertheless, it should be joined with other treatment solutions as its removal yields must be improved for aggressive wastewater treatment.

Anaerobic Ammonium Oxidation (Anammox)

Anammox is a novel technology developed in recent years that has a potential technical advantage and economic efficiency for nitrogen removal from landfill leachate when compared with conventional biological nitrogen removal processes [31]. The Anammox process occurs in nature at both low and high temperatures and salinities. Anammox bacteria converts ammonium and nitrite to nitrogen gas using CO_2 as their carbon source for growth while the nitrite required for their growth may be provided by aerobic ammonium-oxidizing bacteria [86]. Anammox (Equation (4)), nitrification (Equation (5)) and the final results of their combination (Equation (6)) are reported below:



Therefore, Anammox systems are influenced by factors such as influent nitrite and organic concentrations and need to be optimized. Indeed, Anammox is a slow-growing autotrophic bacteria (fed by CO_2) and if organic matter is present, heterotrophic bacteria (fed by organics) can grow faster, competing with Anammox, which is overcome within the process. Indeed, the influent organics concentration should be kept below 800 mg/L, which facilitates Anammox [87].

Wang et al. [37] introduced a combined continuous-flow process of nitrification and Anammox applied to treat mature leachate. An anoxic/aerobic reactor and an up-flow anaerobic sludge blanket (UASB) were combined. The anoxic/aerobic treatment reduces COD and $\text{NH}_4\text{-N}$, while nitrogen removal is achieved in the subsequent UASB via Anammox. Under concentrations of influent ammonia and COD which were respectively 1330 mg/L and 2250 mg/L, the removal efficiencies of TN and COD reached 94% and 62% respectively [37]. Another study included partial nitrification using a sequencing batch reactor (PN-SBR) followed by a Anammox hybrid reactor (HAR), which consists of a suspended biomass layer in the bottom part and a bio-carrier bed in the upper part, run at the influent total ammonia concentrations (TAN) of 500 mg N/L and 1000 mg N/L. The obtained TN removals were $93\% \pm 1\%$ and $81\% \pm 1.2\%$ at nitrogen loading rates of $4.3 \text{ kg TN/m}^3 \text{ d}$ and $8.3 \text{ kg TN/m}^3 \text{ d}$, respectively, while the COD removal ranged from 14% to 46% [38]. Hence, Anammox is a sustainable and suitable solution for leachate treatment even if coupled with other treatment systems such as reactors designed for nitrification/ denitrification, as suggested by other reviews [7]. Particularly, Anammox is highly recommended for old leachate with non-biodegradable COD and lots of nitrogen.

3.2. Physical-Chemical Treatment

Physical-chemical treatments are necessary when the leachate quality and landfill age do not allow achieving release standards by only applying a biological treatment. Indeed, biological treatments have poor efficiency against certain substances such as metals or halogenated bio-refractory organic compounds (AOX). Therefore, physical-chemical treatments are suggested for the removal of refractory substances from stabilized leachate, and also as a refining step for biologically treated leachate. Prior to discharge, an additional effluent-refining step using physical-chemical treatments, such as chemical precipitation, activated carbon adsorption and ion exchange, can be carried out on-site [88]. Chemical-physical treatments are manifold and each of them contributes differently to leachate treatment. For instance, in ion-exchange, adsorption and membrane filtration are the most frequently studied for the treatment of heavy metal wastewater [89]. The most important solutions are reported in this section and the pollutant removal efficiencies are proposed in Table 2.

Table 2. Leachate pollutant removal rates by physical-chemical treatment.

Technology	References	Pollutant Removal Rates (%)										
		COD	BOD	NH ₄ -N	TN	PO ₄ ³⁻	Cl ⁻	Heavy Metals	Fe	SS	SO ₄ ²⁻	Turbidity
Flocculation/Coagulation	[90]	79								90		93
Membrane Process												
MF	[91]									99.6		98.3
NF	[92,93]	/, 96	/, 42	/, 57				70, /			/, 92	
RO	[93,94]	99, /		99.9, /				98, 97				
Air Stripping	[95–97]			89, 85, 99.5								
Adsorption	[88,98]	/, 90		/, 40		84, /		/, 80–96	77, /			
Chemical Precipitation	[89,96]	50, /		85, /				/, 92–100				
Ion Exchange	[89,99]			/, 94				55–100, /				
AOP	[100–102]	50, 70, 81		/, /, 83								
Fenton	[103–105]	60, 45, 85										
Photo-catalysis	[106,107]	56, 60										
Electrochemical Processes												
EC	[108]	40–70		10–25								
EO	[109]	64–70		15–61								

Note: /: Not Available.

3.2.1. Flocculation-Coagulation

Flocculation is a treatment typically applied to enhance the ability of a treatment process to remove non-settleable colloidal solids (such as surfactants, heavy metals, fatty acids and humic acids). In particular, this process allows achieving the efficient removal of turbidity, SS and color, achieving 79% COD, 93% turbidity and 90% SS removal [110].

Flocculation can be introduced, combining coagulant compounds, such as pre-treatment before MBR in order to prevent fouling issues [111]. Indeed, it is often used upstream of biological treatment or reverse osmosis, or downstream with leachate final cleaning functions. The main coagulants are aluminum sulfate, ferrous sulfate, ferric chloride and chorine ferric sulfate [112]. Landfill leachate characterized by low pH values and high concentrations of organic material (22,520 mg/L COD) can be treated effectively by FeCl_3 ; flocculants that are able to achieve better removal efficiencies are [90]:

- FeCl_3 (3000 mg/L): it removes 67.3% of COD and 87% of turbidity;
- FeCl_3 (3000 mg/L) added with polyelectrolyte in variable quantities: it removes 64% of COD and 100% of turbidity.

In order to determine the best coagulant and/or flocculants, another important factor must be considered, such as sludge production. According to Assou et al. [90], the most suitable coagulant is FeCl_3 at the concentration of 3000 mg/L with a sludge production of 700 ml/L. Recently, relatively new types of coagulation reagents were developed in order to increase the efficiency of the coagulation-flocculation process, such as pre-polymerized coagulants and an alternative mixture of aluminum, ferric, and silicate salts incorporated together in a new product [113], which exhibited better coagulation efficiency [114]. According to other studies, the addition of ferric or aluminum coagulants to fresh leachates resulted in a 25%–38% reduction of COD values while higher removal capacities were obtained for the partially stabilized leachates and the corresponding reduction exceeded 75% when the pH value was adjusted to 10. Nevertheless, almost complete removal of color was obtained in all cases [115]. Therefore, old leachate is more suited for coagulation pretreatment processes.

As regards the leachate organic load, this treatment does not seem to be in a condition to give, by itself, results capable of achieving significant removal rates. However, a coagulant such as ferric chloride allows removing color in similar trend as COD, turbidity and SS, suggesting that can be a viable way to manage leachate problems [116].

3.2.2. Separation Treatments with Membrane Filtration

Depending on the size of the pores, the membrane physical separation process is classified by micro-filtration (MF), ultra-filtration (UF), nano-filtration (NF); furthermore, another technology is reverse osmosis (RO) which is an inverse pressure-driven technology applied within water desalination or water potabilization plants [117]. UF is able to remove compounds in a field between 10^{-3} and 10^{-2} μm , while for the lower dimensions reverse osmosis is applied. The main materials used in the construction of the membranes are cellulose acetate and aromatic polyamides; the arrangement may be in floors, tubular modules, spirals and hollow fibers. The membranes are subject to phenomena of clogging and fouling by a series of substances (organic, inorganic), which undermine the efficiency and represent the main issue for applying these technologies for leachate treatments; it is therefore necessary to include the washing steps.

The membrane separation treatments, in particular NF and RO, are considered one of the most efficient treatments for landfill leachate as regards COD, SS and organics removals. This is because they are flexible for all types of leachate: old, middle and young [118]. RO is the most suitable solution for leachate treatment, concerning pollutant removal [7,15]. Nevertheless, this method also provides some disadvantages:

- High transmembrane pressure: 50–60 bar required to win the osmosis pressure; it means that high energy amounts are necessary;

- The fouling phenomenon which entails frequent surface cleaning processes.

Ameen et al. [91] suggested passing a leachate sample through a conventional coagulation process before being filtered through a hollow-fiber MF membrane. As a result, the hollow-fiber MF membrane decreased the turbidity, color, total suspended solids, total dissolved solids and volatile suspended solids in the leachate by 98.30%, 90.30%, 99.63%, 14.71% and 20%, respectively. Therefore, this study provided the indication that MF is capable of removing a high percentage of solids from leachate and might be considered as a polishing stage after onsite biological treatment for sanitary landfill leachate [91]. Alternatively, NF is a membrane process between RO and UF. During NF, substances are rejected by two principles: the rejection of neutral species by size (molecules larger than 200–300 g/mole are rejected), and the rejection of inorganic ions due to electrostatic interactions between the ions and the membrane. Metals such as cadmium, chromium, nickel, lead and zinc have a retention higher than 70% [92] while BOD₅ has a retention of 42%, COD of 96%, ammonia of 57% and sulphate of 92% [93]. Finally, RO is a very effective instrument for the purification of landfill leachate whether all design criteria and specific requirements for landfill leachate have been taken into consideration. Indeed, retention rates can achieve 99%, 99.9% and 98% for COD, ammonium and heavy metals, respectively [93]. The pressure has an advantageous effect on the COD rejection, which increases from 96% to 98% when the operating pressure is increased from 20 to 53 atm, while the removal of metals such as zinc, copper and cadmium is always higher than 97% [94].

3.2.3. Air Stripping

Air stripping is a physical-chemical method often applied with other technical solutions for ammonia removal [119]. It consists of organic constituent separation through exposure to air or steam flows. The volatile organic compounds (VOCs) are freed from the aqueous phase to the gas phase until the state of equilibrium is reached. The process is applied for the treatment of old leachates, reaching high efficiencies with increasing retention times and temperatures. As reported by other reviews [15], an ammonium removal of around 89% at pH 11 and at a temperature of 20 °C for 24 h of treatment can be achieved [95], while 85% [96] and 99.5% [97] of ammonium removal can be reached with 17 h and 120 h of treatment, respectively. Consequently, this process requires generally high HRT and pH values.

Besides the leachate quality, in particular pH and the ammonia nitrogen concentration, which affects the removal of ammonia by stripping, the container configuration is another important factor in ammonia stripping because the majority of ammonia is desorbed at the surface. As such, 65%–74% of ammonia nitrogen can be removed in free stripping tanks with one day of retention time [120]. Then, the contaminated gas, which causes a foaming phenomenon, is treated with H₂SO₄ and HCl, which absorbs the ammonia extracted from the leachate. These substances must be able to absorb most of the ammonia to prevent the emission of NH₃ in the atmosphere, causing serious air pollution problems.

3.2.4. Adsorption by Activated Carbon (AC)

Adsorption is a surface phenomenon by which a multi-component fluid (gas or liquid) mixture is attracted to the surface of a solid adsorbent and forms attachments via physical or chemical bonds [121]. Since our early history, AC was the first widely used adsorbent. However, adsorbents dealing with new porous materials are recommended for ecologically friendly processes, important methods of sustainable development [122] and development of appropriate technologies suited for each specific area. Indeed, it has been extensively proved that any cheap material with a high carbon content and low levels of inorganic compounds can be used as a raw material for the production of activated carbons: various carbonaceous materials such as coal, lignite, nutshells, wood, tobacco stems and peat are used in the production of commercial activated carbon [123]. Also, the production of AC derived from food waste is important from both environmental and economical viewpoints. The processing and transformation of this waste into AC would lay a foundation to eliminate the problems of disposal

and management of food waste materials, while providing a value-added end product for water and wastewater treatment that could potentially expand the carbon market [124].

Absorption treatment by means of AC is preferred among other chemical treatments in order to treat old leachate. Nevertheless, leachate contains aromatic compounds and condensed structures that might have less accessible sites of a granular activated carbon (GAC) surface which means a lower degree of diffusive adsorption [125]. However, the main disadvantage is the frequent regeneration of AC columns and the consequent high consumption of powdery carbon, required to avoid clogging issues and continue adsorption processes.

Adsorption is often used along with the biological processes for the effective treatment of leachate. Indeed, non-biodegradable organic matter, part of the inert COD and color are reduced by GAC adsorption in order to achieve acceptable concentration levels for subsequent biological treatment as it can reduce more than 85% of the non-biodegradable material [126] or 80% of the total organic carbon [127]. Moreover, coffee grounds activated carbon can be an optimal solution for the removal of total iron (77%) and PO₄-P (84%) with optimum iron removal at pH < 5 and pH > 11 for PO₄-P removal [98]. Other reviews reported that GAC technologies are suitable solutions to treat landfill leachate, reaching high removal rates for COD (90%), ammonium (40%) and heavy metals (80%–96%), but the need for frequent regeneration of the activated carbon column and the high cost of GAC may limit its application for landfill leachate treatment in developing countries [88].

3.2.5. Chemical Precipitation

Chemical precipitation is a solution suited for the removal of heavy metals and inorganic compounds. The processes include chemical injections that react with heavy metal ions which dissolve within the leachate to form insoluble precipitates. The formed precipitates can be separated by sedimentation or filtration; therefore, the treated water is then decanted and appropriately discharged or reused. The conventional chemical precipitation processes include hydroxide precipitation and sulfide precipitation with a heavy metal removal efficiency around 92%–100% [89].

Chemical precipitation is also useful for NH₄-N precipitation using magnesium ammonium phosphate (MAP), called struvite, a sedimentation that has been studied and applied to different types of wastewater [128]. During MAP precipitation, COD was not significantly reduced, and thus a biological treatment process may need to be included to remove COD. Li et al. [128] used chemical precipitation in order to treat leachate with an initial NH₄-N concentration of 5618 mg/L. They reduced the ammonium concentration to 112 mg/L (98% removal) within 15 min, while the pH was controlled between 8.5 and 9.0, producing a minimum MAP solubility [128]. Struvite precipitation was also applied at the stoichiometric ratio (Mg:NH₄⁺:PO₄^{3−} = 1:1:1) to anaerobically pre-treat raw landfill leachate effluent having an influent ammonium concentration of 2240 mg/L: maximum ammonia removal was observed as 85% at a pH of 9.2, as well as TKN removal, while, on the other hand, organic nitrogen removal could not be achieved. Besides, the total COD removal reached around 50% [96].

The removal of heavy metals from aqueous solutions has been traditionally carried out by chemical precipitation for its simplicity and inexpensive capital cost. However, chemical precipitation is usually adapted to treat high-concentration wastewater containing heavy metal ions and it is ineffective when the metal ion concentration is low. Furthermore, chemical precipitation is not economical and can produce a large amount of sludge to be treated with great difficulty [89].

3.2.6. Ion Exchange

Ion exchange is a reversible interchange of ions between the solid and liquid phases capable of effectively removing ions and dissolved organic matter from water and wastewater [88]. Ion-exchange resins are polymeric means, joined by covalent bonds, with active functional groups. These functional groups are related to mobile ions that can be exchanged with an ion of the same charge dissolved within the liquid compound [129], making ion exchange a well-established, environmentally friendly technology with metal recovery opportunities [130].

Besides synthetic resins, natural zeolites, naturally occurring silicate minerals, have been widely used to remove heavy metals from aqueous solutions due to their low cost and high abundance. Clinoptilolite is one of the most frequently studied natural zeolites that has received extensive attention due to its selectivity for heavy metals with a removal efficiency of 55% for Pb, 93.6% for Ni and 100% for Zn [89]. The optimum results attained by Bashir et al. [99] indicate that 6.0 min of contact time was required to achieve 94.2% of $\text{NH}_4\text{-N}$ removal when the cationic resin dosage and shaking speed were 24.6 cm^3 and 147 rpm (revolutions per minute), respectively. According to this study, ion exchange resins can be used for the efficient removal of ammoniacal nitrogen from semi-aerobic stabilized landfill leachate [99]. Other studies obtained the highest removal percentages with the chelating resin; Zn removal percentages of 93% were achieved with this resin, although lower results were achieved for Cd (50%). This type of resin, based on the iminodiacetate group, is of the weak cationic type and presents a high selectivity for heavy metals such as Cd and Zn versus sodium, calcium and magnesium ions, when working at pH values of between 4 and 8 [131]. Janin et al. [130], combining M4195 and IR120 resins in four successive columns, succeeded to recover copper and chromium at 94% and 81%, respectively [130], presenting ion exchange as a suitable solution for heavy metal removal from landfill leachate.

According to other reviews, in order to guarantee more efficient pollutant removal amounts, leachate should be subjected to biological treatments and appropriate pre-treatment systems before ion exchange, such as suspended solids removal. In any case, ion exchange is not economically appealing due to its high operational cost. Moreover, this method is suited only for metal and ammonia removal, making ion exchange a poor technological choice for leachate treatment in developing countries [7,35,88].

3.2.7. Chemical Oxidation and Advanced Oxidation Processes (AOP)

Chemical oxidation aims at the mineralization of the contaminants to carbon dioxide, water and inorganics or, at least, at their transformation into harmless products [132]. The main oxidants used for leachate treatment are chlorine, ozone, potassium permanganate and calcium hydrochloride [112]. In recent years, there has been a growing interest towards advanced oxidation processes (AOP) using the combination of strong oxidizing agents such as O_3 and H_2O_2 , together with ultra-violet radiation (UV) or ultrasound (US). Indeed, the three most applied AOP techniques are $\text{O}_3/\text{H}_2\text{O}_2$, $\text{H}_2\text{O}_2/\text{UV}$ and O_3/UV [132]. Many processes are based on the direct reaction of the oxidant with the contaminants while AOP is characterized by the generation of OH radicals as reactive species able to oxidize halogenated organics and improve the biodegradability of recalcitrant organic pollutants [7,15,18].

Numerous studies have shown that the removal of COD obtained by ozonation is around 50%–70% [100,101] and many of these studies have applied the process as a tertiary treatment before discharging the treated effluent into sewage or surface water. Using $\text{O}_3/\text{H}_2\text{O}_2$ as an oxidizing agent has been reported with organic matter removal efficiencies around 90% [133,134], while with a combination of persulfate and hydrogen peroxide ($\text{S}_2\text{O}_8^{2-}/\text{H}_2\text{O}_2$), under optimum operational conditions, 81% COD and 83% $\text{NH}_4\text{-N}$ removals were achieved [102]. Moreover, De Morais and Zamora [135] demonstrated that UV/ H_2O_2 and photo-Fenton treatment allow the total organic carbon content to be efficiently removed in lower than 60 min reaction times with 97% and 89% TOC removal and 56% and 58% COD removal, respectively, a fact that confirms the great degradation capacity of both photochemical processes.

A suitable application of AOPs to leachate treatments must take into account that they make use of expensive reactants such as H_2O_2 and/or O_3 and therefore it is obvious that their application should not replace, whenever possible, the more economic treatments such as the biological degradation [132]. Indeed, a significant decrease in the overall leachate treatment cost could be achieved by combining AOPs with a biological process [136]. Moreover, only wastes with relatively small COD contents ($\leq 5.0 \text{ g/L}$) can be suitably treated these techniques since higher COD contents would require the consumption of too-large amounts of expensive reactants [132]. As a result, the drawbacks of AOP are

the high demand of electrical energy and high costs as well as the control system that is required to allow the treatment to act properly.

Fenton Process

Among various AOPs, Fenton's reagent ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$) is one of the most effective methods of organic pollutant oxidation [137]. Fenton's mixture has been used (either alone or in combination with other treatments) as a chemical process for the treatment of a wide range of wastewaters. It is a system based on the generation of very reactive oxidizing free radicals, especially hydroxyl radicals, which have a stronger oxidation potential than ozone. The efficiency of this process depends on several variables such as temperature, pH, hydrogen peroxide, ferrous ion concentration and treatment time. Chemical oxidation using Fenton's reagent, under optimum conditions, has proven to be a viable alternative to the oxidative destruction of organic pollutants in mixed waste chemicals, with a COD removal of up to 78%–89% [138]. However, metals in the Fenton's residue present great potential for environmental contamination, and require an administration system and control of their final disposal [138].

The Fenton reaction initiated by Fe^{2+} and H_2O_2 and the Fenton-like reaction initiated by Fe^{3+} and H_2O_2 are used for the treatment of leachates since they can significantly remove recalcitrant and toxic compounds and increase leachate biodegradability, as the BOD_5/COD ratio of the leachate can increase from 0.2 up to 0.5 [103]. It has been demonstrated that Fenton's reagent is able to destroy toxic compounds in wastewaters such as phenols and herbicides. Production of OH radicals by Fenton's reagent occurs by means of the addition of H_2O_2 to Fe^{2+} salts (7).



This is a very simple way of producing OH radicals using neither special reactants nor a special apparatus [132].

Some studies stated that, by Fenton processes, COD removal efficiencies range from 45% to 85%, with a decolorization efficiency as high as 92%, while little ammonia is reduced [103–105]. Optimal pH values reported for conventional, photo- and electro-Fenton processes for landfill leachate treatment range between 2.0 and 4.5; COD removal increased with the increasing temperature, although a further temperature increase may significantly cause inefficient H_2O_2 decomposition which offsets the increase of COD removal [105]. More recently, Silva et al. [139], successfully designed a photo-Fenton plant for the treatment of biologically oxidized landfill leachate considering the combined use of solar light, through CPCs (compound parabolic collectors) technology, and artificial radiation (UV lamps); the photo-Fenton reaction, at optimum conditions, showed a removal efficiency between 57% and 80%. Lopez et al. [103] identified that COD could be removed by the Fenton's pre-treatment at about 60% of the initial value (10,540 mg/L) while Gotvajn et al. [140] suggested that Fenton's oxidation is not a viable treatment option for replacement of the existing biological treatment since the final level of the COD removal (80%) was attained in 5 min but the biodegradability increased, expressed as the BOD_5/COD ratio (0.27–0.78), and some other parameters (BOD_5 , concentration of ammonium and nitrogen) were affected by the higher temperature of the process (20–49 °C).

Consequently, Fenton's reaction can be effectively exploited to treat landfill leachate; however, it is particularly appropriate for stabilized or mature leachate. It represents a process applicable to the treatment of highly toxic leachate with faster kinetics [105] but with the typical high costs of AOP processes, which comes from the reagent's consumption (especially of H_2O_2) [141], that required combined technology to obtain good pollutant removal.

Photocatalysis

Photocatalytic processes make use of a semiconductor metal oxide as a catalyst and of oxygen as an oxidizing agent [6]. Many catalysts have been tested so far, although TiO_2 seems to have the most

interesting attributes such as high stability, good performance and low cost [20,21]. The initiating event in the photocatalytic process is the absorption of the radiation with the formation of electron-hole pairs. The considerable reducing power of formed electrons allows them to reduce some metals and dissolved oxygen with the formation of the superoxide radical ion O_2^- whereas remaining holes are capable of oxidizing adsorbed H_2O or HO^- to reactive HO radicals. These reactions are of great importance in oxidative degradation processes due to the high concentration of H_2O and HO^- adsorbed on the particle surface [132].

A pilot-scale semi-batch-mode photocatalytic system used for the degradation experiments of leachate characterized to have low biodegradability ($BOD/COD = 0.011$) was studied by Cho et al. [106], reaching only 52% COD, 79% TOC removal after 5 h of illumination time, while after 10 h of photocatalytic oxidation, 56% COD and 88% TOC were removed. Another study tested a mature landfill leachate by UV- TiO_2 photocatalysis with a removal of COD, DOC, and color of above 60%, 70% and 97%, respectively; under optimal conditions, the ratio of BOD/COD was elevated from 0.09 to 0.39, representing a substantial improvement in biodegradability. Moreover, within the same study, esters were produced during the photocatalytic process and ketones, hydrocarbons, aromatic hydrocarbons, hydroxyl-benzenes, and acids were degraded during the photocatalytic oxidation processes: 37 out of 72 kinds of organic pollutants in the leachate remained after 72 h treatment [107].

Photoassisted titanium dioxide (TiO_2) oxidation shows a great potential to treat landfill leachate. Despite the relatively good efficiency of TiO_2 photocatalysis to deal with landfill leachate, several technical challenges need to be addressed, such as [142]:

- post-separation methods of titanium after water treatment;
- depth of light penetration into the aqueous titanium suspension;
- low quantum efficiencies of the degradation process on the irradiated catalyst;
- the catalyst turnover number and catalyst poisoning also need to be further investigated.

Use of UV irradiation is also an obstacle to scale-up this technology. Certain modifications of the TiO_2 semiconductor may resolve this issue to an extent by reducing the band gap of TiO_2 . However, most of these changes are achieved by doping or by making composite catalysts. Covering all these aspects, it is believed that further research is needed toward sunlight-irradiated TiO_2 photocatalysis based on the charge transfer mechanism in order to make this technology more cost-effective and commercialized [142].

3.2.8. Electrochemical Processes

Electrochemical processes are treatments mainly applied for old leachates and consist of an electrochemical cell composed of an anode and a cathode which are functional for pollutant reduction. The main processes studied and reported in the literature are electro-coagulation (EC) and electro-oxidation (EO).

Electro-Coagulation

EC is a process that uses electrodes useful for supplying ions within the solution, allowing the suspended contaminants, emulsified or dissolved, to form agglomerates. Coagulating ions are produced in situ and three different stages can be identified:

- i) formation of the coagulating electrode through the sacrificial electrolytic oxidation;
- ii) pollutants and suspended particles being destabilized and emulsions breaking;
- iii) aggregation of destabilized phases and the formation of flakes.

There are many factors able to influence the electro-coagulation process, such as the reactor design, electrode material and current leachate density and conductivity (the removal of polluting particles increases with the density). Despite higher removal efficiencies achieved with aluminum,

iron is considered the best electrode because it is less toxic, requires less energy, and is less sensitive to inhibitor phenomena [37]. Also, the distance between the electrodes could affect the efficiency of the removal of pollutants present in the leachate. However, some studies are discordant and it is clear that distance is also a function of the operating conditions [143]. The disadvantage is that electro-coagulation corresponds to high energy consumption and, therefore, it is necessary to find a compromise between the removal efficiency and consumption of power required [144].

Electrical conductivity plays an important role in EC because, when it is low, the current efficiency decreases, and greater applied potential is required to prevent electrode passivation, leading to additional power consumption. NaCl is the most common electrolyte used to increase the conductivity. Landfill leachate generally has high conductivity values and the addition of an electrolyte is not required. COD removal remains, on average, in the 40%–70% range, while the respective nitrogen removal is 10%–25% [108].

EC has proven to be a fast and efficient technology for leachate treatment. It requires simple equipment, easy operative conditions and no need for the addition of chemicals. Unfortunately, it presents some disadvantages such as the need to regularly replace the sacrificial electrode, due to oxidation reactions, and the significant generation of sludge during the process. Sludge contains large amounts of iron and other recalcitrant pollutants, and thus a specific treatment before its disposal is required.

Electro-Oxidation

EO consists of applying a voltage or a current, constant with time, to the compound which needs to be treated [109]. The electrolytic cell is the reactor where the whole process takes place and EO can be developed in two distinct ways: indirect or direct.

- Direct oxidation allows the polluting particles to exchange electrons directly with the anode surface. This method does not appear to be effective in the degradation of organic material; despite that, it promotes the formation of very powerful oxidizing agents that are used for indirect oxidation;
- Indirect EO takes place when high chlorine compounds are concentrated within the leachate. The active chlorine is oxidized by the anode producing hypochlorite, which has a strong oxidation effect with respect to the organic compounds [145]. This reaction is particularly suitable for saline leachates, where many pollutants are removed, such as ammonium, and with the presence of metal ions (Ag^+ , Fe^{3+} , Co^{3+} , Ni^{2+}).

Electrochemical oxidation easily removed 64%–70% of COD, and 15%–61% of ammonium with an energy consumption that varies between 0.377 and 0.740 kWh/L, depending on the treated sample [89].

The Boron-Doped Diamond (BDD) anode is an EO method that is economical due to fast oxidation, high current efficiency and a low energy consumption. These electrodes are therefore considered suitable for landfill leachate treatment regarding their properties: an inert surface with low adsorption properties and outstanding corrosion stability, even in acidic conditions [146]. High-quality BDD electrodes with excellent electrochemical property deposition on niobium (Nb) substrates by the hot filament chemical vapor deposition (HFCVD) method are an effective technique for landfill leachate treatment, as 87.5% COD and 74.06% $\text{NH}_4\text{-N}$ removal were achieved after 6 h treatment, with a specific energy consumption of $223.2 \text{ kWh}\cdot\text{m}^{-3}$. BDD is confirmed as the most studied, followed by Ti/PbO_2 , $\text{Ti/RuO}_2\text{-IrO}_2$ and graphite [147]. However, the treatment effectiveness, as for EC, depends on the electrode material, voltage, current density and pH [145,148].

EO provides a simple and feasible method for the treatment of leachate from landfills, offering high efficiencies without the problem of sludge production, contrary to EC. Using appropriate experimental conditions, this method could remove a large portion of COD and ammonia, while significantly reducing color, without the accumulation of refractory organic substances. The most significant

disadvantage is the high treatment operation costs, due to the high energy consumption. To overcome this disadvantage, there are two options:

- i) implement this technology combining other techniques, either as a pre-treatment or as a finishing step;
- ii) introduce renewable energy within the system.

However, EO technology could be applied to leachates in order to reduce the concentration of the refractory organic matter and ammonium. Using this technology alone it appears that it is not possible to achieve the limits for discharging into sewage, except where local limits for carbonaceous substances and nitrogen are decidedly more permissive. Although high energy consumption and a potential chlorinated organic formation may limit its application, EO is a promising and powerful technology for the treatment of landfill leachate, especially for low BOD₅/COD or high toxic landfill leachate [109].

3.3. Combination of Physical-Chemical and Biological Processes

Treatment combination is an efficient and suitable to treat leachate as practicable in different areas and for dissimilar leachate pollutant concentrations. As reported by other reviews, conventional treatments are not sufficient to obtain high performances in contaminant removal because no flexible and universal way to treat landfill leachate exists; instead, it is specific, case by case [13,14]. Taking into account the leachate age, season, climatic conditions, regulation criteria and pollutant concentration, leachate treatment plants are forced to integrate chemical-physical and biological stages [18] which ameliorates drawbacks and contributes to a higher treatment efficiency [35].

Membrane technology, such as reverse osmosis, was noted as a viable treatment technology to comply with strict release concentrations, as reported by older reviews [15,35]; however, fouling issues and high energy requirements are effective barriers that need to be overcome, specifically for developing countries [149]. Moreover, methods such as membrane and carbon adsorption only transfer the pollutant from one stream to another, changing the environmental issue to another sector [7].

Many studies introduced combined technologies in order to enhance treatment efficiencies. Henderson et al. [150] introduced an anaerobic filter with an RBC removing 80% and 90% of COD, and reaching over 95% removal for ammonia. Słomczyńska and Słomczyński [151] and Neczaj et al. [25] showed that by applying an ultrasonic pre-treatment before SBR, the efficiency in terms of COD and NH₄-N removal can be substantially increased, reaching an efficiency rate of almost at 70% for a leachate concentration in influent of 5%, 10% and 15%. Kargi and Pamukoglu [152] proposed a coagulation treatment before an aerated biological reactor together using powder activated carbon (PAC) and zeolite powder as adsorbent materials, with approximately 77% and 87% of COD removed with PAC and zeolite, while Heavey [153] pretreated the leachate with peat adsorbents, accomplishing very high removal efficiencies of 100% for ammonia and BOD, 69% for COD. Chen et al. [57] investigated the performance of a MBBR with an anaerobic-aerobic arrangement to treat landfill leachate for the simultaneous removal of COD and ammonium, achieving a COD removal efficiency of 91%. Liang and Liu [154] proposed a partial nitrification reactor (PNR), an anaerobic ammonium oxidation (Anammox) reactor and two underground soil infiltration systems with average removal efficiencies of 97% NH₄-N, 87% TN and 89% COD obtained. Hasar et al. [155] introduced a treatment configuration consisting of ammonia stripping, coagulation/flocculation, A/An (Aerobic/Anaerobic) MBR and reverse osmosis, where leachate could be used even for all the reuse applications at the optimal conditions because the final COD value decreased to less than 4 mg/L. Di Iaconi et al. [156,157] suggested a pre-treatment step for nitrogen recovery, as struvite followed by ozone, enhancing the biological degradation carried out in a SBBGR system (sequencing batch biofilter granular reactor), which was able to meet the current discharge limits even in the case of highly stabilized municipal landfill leachates. Tugtas et al. [158] proposed the bio-electrochemical treatment of anaerobically pre-treated landfill leachate in batch and continuous-flow two-chambered microbial fuel cells (MFCs)

Xie et al. [159] introduced the integration of membrane separation with an anaerobic bioreactor to form an anaerobic membrane bioreactor (AnMBR), resulting a stable COD removal efficiency of $62.2\% \pm 1.8\%$ when the reactor is fed with raw leachate with $13,000 \pm 750$ mg/L COD. Liu et al. [160] performed an innovative combined treatment process of air stripping, Fenton, SBR and coagulation, finding high COD and $\text{NH}_4\text{-N}$ removal rates of 92.8% and 98%, respectively. Amaral et al. [161] suggested a landfill treatment configuration consisting of an association of air stripping, MBR, and a NF (nano-filtration) membrane which has shown excellent performance, especially regarding the removal of COD (80%–96%), ammonia (85%–95%), color (98%–99.9%), and phosphorus (78%–99.8%). Gao et al. [162] introduced an integrated approach with the continuous recirculation of ozonated autotrophic nitrogen removal (ANR) effluent, showing the possibility to reach a COD removal as high as 40% in the ANR with a slight decrease in nitrogen removal of around 70%–80% when compared with no leachate ozonated recirculation. Zhang et al. [163], in order to explore the feasibility of energy-free denitrifying, surveyed a self-powered device in which an ammonia/nitrate coupled redox fuel cell (CRFC) reactor served in removing nitrogen and harvesting electric energy, at the same time as 26.2% $\text{NH}_4\text{-N}$ and 91.4% $\text{NO}_2\text{-N}$ removal.

In 2016 many other combined treatments were introduced, achieving high pollutant removal rates and enhancing treatment efficiencies, allowing project developers to implement technological choices suited for specific landfill release management issues. So, in the next sections we specifically review some modern combined technologies suggested in 2016, summarized in Table 3, evaluating the pros and cons of each, suggesting joint methods as a reliable way for leachate treatment.

3.3.1. Combined Treatments Introduced in 2016

SAMBR–MBR (Synthetic Leachate, London)

Trzcinski and Stuckey [164] introduced an anaerobic-aerobic combined treatment in order to handle a synthetic leachate from the organic fraction of municipal solid waste with the COD at 5.72–26.78 g/L, ammonia-nitrogen at 7–140 mg N/L and phosphorus at 3.9–24 mg P/L. They used a submerged anaerobic membrane bioreactor (SAMBR), followed by an aerobic membrane bioreactor (AMBR). The COD removal in the SAMBR was in the range of 89.7%–98% (average: 94.5%), while the total COD removal increased into the range of 92.2%–98.4% (average: 96.1%) due to the polishing action of the AMBR.

The results demonstrated the excellent resilience of the SAMBR to organic shocks at loading rates as high as 19,800 mg COD/L day, and HRTs as low as 0.4 days. Between 13.6% and 50.8% (average: 26%) of the SAMBR-permeated COD was degraded in the AMBR. However, calcium (160 mg/L) in the leachate was found to precipitate on the SAMBR membrane, causing severe fouling [94].

SBBGR–EO (Italy)

Moro et al. [165] suggested an aerobic treatment combined with physical-chemical technology. In particular, a sequencing batch biofilter granular reactor (SBBGR) was used to remove biodegradable carbon and nitrogen species from raw leachates ($\text{BOD}/\text{COD} = 0.2$) and the biological effluent coming from the SBBGR was treated using two electrodes by electrochemical oxidation.

The SBBGR was able to remove 46.8% of the COD, leaving high residual concentrations in the effluent (2050 mg/L). As for the nitrogen content, SBBGR treatment led to the extensive removal of ammonia and total nitrogen species (i.e., 99.9% and 83.5%, respectively) with residual concentrations of 2.1 and 339.6 mg/L, respectively, and 55.2%–10.1% of SS and color. However, the performance of the electro-oxidative process applied to biological effluent has revealed residual COD concentrations equal to 1200, 500, 192 and 50 mg/L (98% removal) after 240 min for experiments carried out with 33, 83, 133 and 200 mA/cm².

Table 3. Leachate pollutant removal rates by combined treatments.

Technologies	Reference	Combined Treatments	BOD/COD	Pollutant Removal Rates (%)						
				COD	NH ₄ -N	Total N	BOD	PO ₄ ³⁻	SS	Heavy Metals
SAMBR, BR	[164]	A, An	>0.5	96.1						
SBR, EO	[165]	A, P/C	0.2	98	99	83.5			55.2	
SBR, Fenton-like, SBR	[166]	A, P/C, A	0.17–0.57	95	95		95			
Activated Sludge, Coagulation, PhotoFenton	[167]	A, P/C, P/C	0.07–0.13	96	62–99				88	
PhotoFenton, MBR	[168]	P/C, A	0.18	96	88		90.2	100	95.5	
Trickling filters, EC	[169]	A, P/C	0.09	80	94		94	98		
Fenton, Aerated Biomass	[170]	P/C, A	0.16–0.27	83	95				46	
Aerobic SBR, Adsorption	[171]	A, P/C	<0.1	43	96					24–100
CW, Adsorption	[172]	A, P/C	0.2	86.7	99.2					87–89
MBR, UF, EO	[173]	A, P/C	0.14–0.3	94	77		97	53		
MBR, PAC to activated sludge, NF	[174]	A, P/C, P	0.3	94	97			99		99

Notes: A = Aerobic; An = Anaerobic; P/C = Physico/Chemical.

According to the results, the effluent obtained using the combined biological and electro-oxidation treatment had a COD value such that could be discharged into the sewerage system after having used a current density corresponding to 83 mA/cm² or 133 mA/cm², and it could even be discharged into receiving water bodies if a current density equal to 200 mA/cm² was used for 240 min. Nevertheless, the electrochemical oxidation carried out on landfill leachate is only effective on nitrogen ammonia through indirect electro-oxidation, and it is not effective on other nitrogen species. In any case, where ammonia is present, 82% is removed by the end of the test at 200 mA/cm². The toxicity does not appear to be affected; in fact, the EO of raw leachate as well as biological effluent leads to a decrease in the toxicity, regardless of the applied current density.

In conclusion, this study proved that electrochemical treatment with new patented electrode potentialities, alone or in combination with a new type of biofilter, strengthens the process [165].

SBR–Fenton-Like–SBR Post-Oxidation (Estonia)

Klein et al. [166] introduced an aerobic–physic-chemical–aerobic combined treatment for young landfill leachate (BOD/COD = 0.57–0.17). The proposed technological scheme consisted of an activated sludge pre-treatment combined with a Fenton-like process enhanced by continuous sludge reuse and followed by an activated sludge post-oxidation. Biological leachate pre- and post-treatments were carried out in sequencing batch reactors (SBRs) operated at three 8 h cycles per day with three feeding pulses per cycle. A settling period (30 min) was applied and aeration was performed intermittently (1 h anoxic/1 h aeration). The concentration of dissolved oxygen (DO) was kept between 0.7 and 2.5 mg/L during the aeration phase.

During the first operational period, the BOD₇ removal efficiency was 99%, leaving a mean effluent BOD₇ concentration of 60 mg/L. The COD removal efficiency was 86%, leaving a mean COD concentration in the effluent of 1490 mg/L. A BOD₇/COD ratio of 0.04 in the effluent indicated low biodegradability, which meant that biological wastewater treatment methods were not further applicable. Biological pre-treatment removed 99% of the NH₄-N, 83% of phenols and 86% of lignin and tannins.

The Fenton-like process allows increasing the BOD₇/COD (from 0.04 to 0.55), where the COD removal efficiency was 75%. The mean COD removal efficiency was 60% which leaves 600 mg/L of COD in the effluent. During the operational period, the Fenton-like treatment increased BOD₇ by 65% on average and the BOD₇/COD ratio increased from 0.04 to 0.30. Furthermore, it was found that 100% of the NO₂-N was oxidized to NO₃-N.

The effluent of the chemical treatment contained substantial concentrations of organics; therefore, the wastewater needed an additional treatment stage before discharge. Therefore, the biologically and chemically treated landfill leachate was subjected to a biological post-treatment reactor for final purification. The results showed that activated sludge treatment was effective at removing easily degradable organic material since the BOD₇ and COD removal efficiencies were 94% and 49%, respectively. When the mean value of the COD in the effluent of the chemical treatment reactor was 600 mg/L, then the mean COD value in the effluent of the reactor was 290 mg/L. The mean BOD₇/COD ratio in the effluent was 0.03, which indicated that the easily degradable fraction was oxidized in the reactor. Additionally, 98% of the NH₄-N, 6% of phenols and 23% of lignins and tannins were removed in the biological post-treatment process.

So, this combined treatment process resulted in a more than 95% removal of each measured parameter (COD, BOD, NH₄-N). Overall, the combined technological scheme with continuous ferric sludge reuse in the Fenton-like stage proved to be a promising alternative for landfill leachate treatment [166]. However, COD concentrations are too high to be introduced into water bodies, and a post combination with municipal wastewater is recommended for this case.

Aerobic Lagoon–Activated Sludge Biological Pre-Oxidation–Coagulation–Photo-Fenton (Portugal)

Silva et al. [167] surveyed a methodology for the treatment of landfill leachates, after aerobic lagooning, involving an aerobic activated sludge biological pre-oxidation (ASBO), a coagulation/sedimentation step (240 mg Fe^{3+} /L, at pH 4.2) and a photo-oxidation through a photo-Fenton (PF) reaction (60 mg Fe^{2+} , at pH 2.8) combining solar and artificial light.

The ASBO process applied to a leachate after aerobic lagooning, with high organic and nitrogen content (1.1–1.5 g C/L; 0.8–3.0 g N/L) and low biodegradability ($\text{BOD}_5/\text{COD} = 0.07\text{--}0.13$), is capable of oxidizing 62%–99% of the ammonium-nitrogen. The coagulation/sedimentation stage led to the precipitation of humic acids, promoting a marked change in the leachate color, from dark-brown to yellowish-brown (related to fulvic acids), accompanied by a reduction of 60%, 58% and 88% of the DOC, COD and TSS, respectively. The photo-Fenton (PF) system promoted the degradation of the recalcitrant organic molecules into more easily biodegradable ones. The PF oxidation step ($\text{Fe}^{2+}/\text{H}_2\text{O}_2/\text{UV-Vis}$; 60 mg Fe^{2+} /L, pH 2.8) using artificial ($\sim 1.3 \text{ kW/m}^3$) and solar radiation was used to degrade the most recalcitrant organic compounds. These three first steps of the multistage treatment strategy are able to remove 80% of the organic matter present in the sanitary landfill leachate, obtaining, at the end, a biodegradable effluent, able to be oxidized in a final biological process, in order to fulfill the discharge limits imposed by legislation to sewage systems [167]. Indeed, in order to fully comply with the regulation, a subsequent biological denitrification stage should also be employed to release the treated leachate to water bodies.

In conclusion, aerobic biological oxidation allows achieving almost complete removal of ammonia, and a physical-chemical process, followed by a 14 h settling phase, promotes the precipitation of the humic acids and the sedimentation of the suspended solids generated, maximizing the light penetration in the downstream photoreactor. Further, the PF oxidation steps, using artificial and solar radiation, are used in order to degrade the most recalcitrant organic compounds, through the generation of powerful reactive chemical species, such as hydroxyl radicals, turning them into simpler and easily biodegradable organic compounds. This combined process helps us to understand how a joint method allows the removal of different pollutants from landfill leachate.

Photo-Electro-Fenton Process–Membrane Bio Reactor (India)

Nivya and Pieus [168] projected a physico-chemical and aerobic treatment, combining a photo-electro-Fenton process (PEF) followed by MBR treatment. The BOD/COD ratio of the original sample was 0.18 and it was treated by a reaction mixture of H_2O_2 (57.6%) and a current density of 140.5 A/m^2 at pH 2.9. After 45 min, the treated samples were allowed to settle for 2 h and then introduced into the aerobic reactor. The MBR consisted of a hollow-fiber membrane module with a pore size of $0.1 \mu\text{m}$.

The percentage pollutant removal efficiencies of the synthetic landfill leachate pollutants TSS, BOD, COD, ammonia-nitrogen, phosphate, sulphate, sulphide and chloride are 89.3%, 71.9%, 83.6%, 65%, 100%, 58%, 92.3% and 65% in the PEF process, respectively. The biodegradability in terms of the BOD/COD ratio of synthetic wastewater is increased from 0.19 to 0.34; therefore, the effluent could be biologically treated using MBR. After MBR treatment, the percentage removal efficiencies of pollutants were increased to 95.5%, 90.2%, 96.2%, 88.3%, 100%, 82.7%, 93.3% and 88.2%, respectively [168].

From this study it is proved that utilizing MBR as a post-treatment after PEF increased the pollutant removal efficiency, and the combined technology is a viable way to treat old landfill leachate.

Trickling Filters—Electro-Coagulation (Magnesium-Based Anode) (Canada)

Oumar et al. [169] introduced a combination of biofiltration (BF) (or trickling filters) and electro-coagulation (EC) processes for the treatment of sanitary landfill leachate ($\text{BOD}/\text{COD} = 0.09$). The biofilter system used a PVC column filled with indigenous microflora bacteria fixed on an organic support mainly comprised of peat and wood shavings. The calcite was added in the filter media

in order to buffer the pH and to provide a mineral carbon source for autotrophic microorganisms. The electrochemical tests were then conducted in a batch recirculation mode with a liquid flow entering in the top of the cell.

The BF process showed a satisfactory result in terms of ammonia, BOD₅, turbidity and phosphorus removal with removal percentages of 94%, 94%, 95% and >98%, respectively. However, the COD removal was low (13% on average). Electro-coagulation of bio-treated landfill leaching using a magnesium-based anode had enabled the reduction of the residual COD; with a current density of 10 mA/cm² and 20 min of treatment time, the COD removals were 52%, 41% and 27% in the presence of 1.0, 2.0 and 3 g/L of NaHCO₃, respectively. Therefore, around 53% of the residual COD and 85% of the true color were removed by applying a current density of 10 mA/cm² and a treatment time of 30 min. A significant increase in the pH was recorded after the EC treatment. The pH passed from 8.4 before treatment to 10.6 after 30 min of treatment [169].

The combination of these two technologies allow achieving 94%, 94%, 95% and >98% removal of ammonia, BOD₅, turbidity and phosphorus, respectively, with the activity of the biofiltration, while 80% of COD can be removed by the EC. So, this study proved the feasibility of combined BF and electro-coagulation processes for the effective treatment of mature sanitary landfill leachate, although it cannot achieve suitable concentration for release into water bodies.

Fenton Process–Passive Aerated Immobilized Biomass (PAB) (Egypt)

Ismail and Tawfik [170] implemented a passive aerated immobilized biomass (PAB) reactor after a Fenton process in order to treat a medium-old landfill leachate (BOD/COD = 0.16–0.27). Landfill leachate Fenton oxidation was performed for 15 min at pH 3.5, with a molar ratio H₂O₂/Fe⁺² of 5 and a H₂O₂ dosage of 25 ml/L. Then, the pre-treated effluent was fed to the passive aerated immobilized biomass reactor. The PAB consisted of three segments separated by open spaces (10 cm height) for the diffusion of natural oxygen from ambient air, avoiding energy consumption, and created fully aerobic conditions where the dissolved oxygen concentration exceeded 5.0 mg/L in the bulk liquid. Landfill leachate flowed from the top of the reactor and trickled downwards due to gravity, contacting the immobilized biomass on the sponge surface, which has pore size of 0.63 mm and is packed into a net-like cylindrical polyvinyl chloride ring of 3 cm diameter and 3 cm length.

The whole process allowed the removal of 83% of the total COD, 46% of TSS, 95% of NH₄⁺; therefore, the effluent quality was compliant with local release standards (COD = 1100 mg/L and TSS = 600 mg/L) for discharge into the sewerage network [170]. However, this solution might be not applicable for high leachate streams or for full-scale plants, as smell and ineffective aerations can be an important issue.

Aerobic SBR–Zeolite Adsorption (Malaysia)

Lim et al. [171] studied an aerobic sequencing batch reactor (ASBR) for the treatment of locally obtained, very old landfill leachate with initial ammonia-nitrogen and COD concentrations of 1800 and 3200 mg/L, respectively. ASBR was aerated using a fine air bubble diffuser and an air pump located at the bottom of the reactor with a superficial air upflow velocity of 1.0–1.2 cm/s to provide aeration to the system throughout the entire treatment process. After seven days of biological degradation by the reactor system, the adsorption experiment (physical removal) was carried out using 10% of the zeolite to adsorb the effluent discharged from the system.

ASBR removed 65% of the ammonia-nitrogen and 30% of the COD during seven days of treatment time. Thereafter, zeolite polished the NH₄-N and COD content. The results obtained are promising as the adsorption of leachate by zeolite further enhanced the removal of the ammoniacal nitrogen and COD up to 96% and 43%, respectively. Furthermore, this combined biological-physical treatment system was able to significantly remove heavy metals from the wastewater at 100% aluminum, 44% vanadium, 63% chromium, 75% magnesium, 24% cuprum and 85% plumbum [171].

Co-Treatment Constructed Wetland–Adsorption by ZELIAC/Zeolite (Iran)

Mojiri et al. [172] conducted a study to co-treat landfill leachate and municipal wastewater by using a CW system which contains two substrate layers of adsorbents, namely ZELIAC and zeolite. Zeolite, activated carbon, limestone, rice husk ash, and Portland cement have been ground, passed through a 300 mm mesh sieve, and mixed to prepare ZELIAC. Zeolite and activated carbon were present in ZELIAC; therefore, ZELIAC could function both as an adsorbent and ion-exchanger.

The contact time (h) and leachate-to-wastewater mixing ratio (%; v/v) were considered as independent variables. An air pump was used to supply air to the wetland. The landfill leachate displayed a high-intensity color (1817 Pt. Co) and contained high concentrations of COD (2301 mg/L), $\text{NH}_4\text{-N}$ (627 mg/L), Ni (4.6 mg/L), and Cd (2.5 mg/L). The BOD_5 was 461 mg/L, and a low biodegradability ratio ($\text{BOD}_5/\text{COD} = 0.20$) was observed (age > 15 years).

The removal efficiency of Ni varied from 68.3% (reaction time = 12 h and leachate-to-wastewater mixing ratio = 80%) to 86.9% (reaction time = 42 h and leachate-to-wastewater mixing ratio = 20%). Optimum Ni removal (86.0%) was observed at a contact time of 49.0 h and a leachate-to-wastewater mixing ratio of 20.0%. Moreover, the removal efficiency of Cd ranged from 68.4% to 88.9% with optimum Cd elimination (87.1%) achieved at a contact time of 51.3 h and a leachate-to-wastewater mixing ratio of 20.0%. Removal efficiencies decreased as the leachate ratio in the leachate and wastewater mixture increased. At the optimum contact time (50.2 h) and leachate-to-wastewater mixing ratio (20.0%), the removal efficiencies of the color, COD and ammonia contents were 90.3%, 86.7%, 99.2%, respectively [172].

Hence, CWs with adsorption systems are a suitable way to remove COD and heavy metals from landfill leachate, making this combined treatment a suitable method for developing countries with extended free areas and with poor economic funds.

MBR–UF–EO (Québec, Canada)

Zolfaghari et al. [173] suggested the combination of a membrane bioreactor (MBR) equipped by ultra-filtration (UF) and an electro-oxidation process (EO) with a boron-doped diamond electrode (BDD) in order to effectively treat highly contaminated, medium-old landfill leachate ($\text{BOD}/\text{COD} = 0.14\text{--}0.3$). The membrane bioreactor used in this study is a submerged hollow-fiber UF with a nominal pore size of $0.04\text{ }\mu\text{m}$ and a total filtration surface area of 0.047 m^2 . Instead, the EO reactor was equipped with an anode and a cathode with an inter-electrode gap of 2 cm. The rectangular anode electrode was made of niobium coated with boron-doped diamond (Nb/BDD), while the cathode was made of titanium with the same physical characteristics of the anode.

For MBR, an organic load rate of 1200 mg/L COD and a sludge retention time of 80 days were considered as the optimum operating conditions in which the COD, TOC, $\text{NH}_4\text{-N}$ and phosphorous removal efficiencies reached the averages of 63%, 35%, 98% and 52%, respectively, while the best performance of the EO process was at the current intensity of 3 A with a treatment of time of 120 min. By using the combination of MBR and EO as the tertiary treatment, at the optimum operating condition for both processes, the COD, TOC and ammonium concentrations were decreased to 89, 57 and 65 mg/L, respectively, equal to COD 94%, BOD 97%, NH_4^+ 77%, PO_4^{3-} 53% removal.

The main problem associated with EO processes was the considerable toxicity increase mainly associated with the presence of ammonia, and the residual concentrations of radicals and organic-chlorinated compounds. The utilization of a radical scavenger after electro-oxidation or keeping electro-oxidized landfill leachate in a storage tank before introduction into the aeration basin could be a solution to this challenge [173].

MBR–PAC to Activated Sludge–NF (Iran)

Peyrav et al. [174] introduced a bench-scale integrated process based on submerged aerobic powdered activated carbon and a membrane Bioreactor (PAC-MBR) that has been used and established for landfill

leachate treatment with an approximate BOD/COD of 0.3, reflecting a high non-biodegradable portion within the leachate. The biological treatment under aerobic conditions was performed in batch mode and the bioreactor was aerated vigorously with an air compressor at a HRT of 24 h. The continuous agitation ensures a homogeneous mixture of the leachate and prevents biomass settling.

Although the hybrid treatment method of landfill leachate yielded rather high degrees of purification, and the introduction of PAC again improved its quality, the effluent discharge standards still have not been reached. So, the permeate of PAC-MBR systems was subjected to a further purification step with the NF. The various concentrations of the effluent COD from PAC-MBR systems were between 550 and 850 mg/L, whereas the COD values decreased to below 50 mg/L in the permeate stream and there were substantial COD removals by NF. So, the NF process complemented the PAC-MBR treatment more effectively, with above 94% COD removal. TKN and $\text{NH}_4\text{-N}$ were removed by PAC-MBR from 88% to 91%, and by the NF process from 96% to 97%, respectively. Moreover, during the NF process, the removals of chromium (III) and nickel were increased to 100% and 99%, respectively.

Hence, the results indicated that PAC-MBR treatment and NF membrane filtration can be integrated to enhance the removal rate of the trace organic contaminants in a wide range. Indeed, the COD removal rate reached up to 75% by the PAC-MBR unit and improved up to 94% by the NF process, and a considerable amount of TKN, which mainly consists of $\text{NH}_4\text{-N}$, was efficiently removed. The phosphorus content of the feed leachate reached to below the discharge limit value by a 99% reduction during the hybrid treatment system while the heavy metal removal efficiency amounted to $99\% \pm 2\%$ [174].

NF, joined with pre-treatment technologies, can be considered a viable way to treat landfill leachate, as it is able to reduce the pollutant concentration amounts in accordance with the needs of water bodies. However, these methodologies generated activated carbon waste and the energy necessary for the treatment is high, due to the introduction of NF and MBR. However, they can be suitable for cities in developed countries, where economic funds are available and release regulations are strict.

4. Discussion

As reviewed in this paper and in other previous studies [7,15,18], landfill leachate represents a complex issue that can be tackled by numerous technologies. Environmental, economic, social and seasonal changes are only a few characteristics that affect leachate treatment choices, making the development of a new project a real challenge for engineers and local policy-makers.

Past studies had already highlighted the difficulty of treating landfill leachate by stand-alone conventional chemical/physical or biological treatments due to the high percentage of high-molecular-weight organic materials and biological inhibition caused by the presence of heavy metals, suggesting a combination of technologies in order to achieve high pollutant removal rates [175]. Indeed, biological treatments are useful to decrease organic pollutants, but they are not able to reduce heavy metal concentrations or inorganic chemical compounds usually found within landfill leachate; on the contrary, physical-chemical treatments are not suited to treat young leachates as they are not able to effectively reduce organics fractions. As a result, stand-alone technologies are not useful for leachate pollutant reductions and a combination is suggested in order to achieve regulation limits for the release to water bodies.

Combined treatments are a considerable way to reduce harmful molecules within landfill leachate, and in the last 30 years they have been used and proposed in different forms. Single, combined choices are not suited for all cases and every circumstance must be studied in order to evaluate the most suitable tool that should be developed. For instance, developing countries are typical areas where this issue has more importance; indeed, a lack of public regulation, know-how, economic funds and local awareness of environmental pollution are barriers that stop the construction of suitable solutions and that incentivize open dumping [176]. In these regions the word 'feasible' is the most effective, since cheaper solutions need to be considered. For that purpose, membrane filtration, chemical oxidation and

ion exchange cannot be chosen as effective methods, whereas aerated lagoons, constructed wetlands, adapted adsorption technologies and SBR could be the most suitable approaches.

Figure 1 sums up the COD and $\text{NH}_4\text{-N}$ removal yields of each technology reviewed in this paper, giving a comparison with an average removal rate of the combined technologies suggested in 2016 (dashed lines). However, no leachate age or composition, pros or cons, state of the study areas or particular conditions were taken into account in this comparison, giving only a general view of the maximum pollutant removal achieved in the surveys stated in each article. As a result, it is obvious that high amounts of COD and ammonia can be removed at the same time only by reverse osmosis (as reported in previous reviews [15]) and by combined methods. Nevertheless, RO has many disadvantages such as high cost and a large amount of energy required, as explained in Section 3.2.2. Combined treatments, instead of a single technology, allow removing pollutants and recalcitrant contaminants with high yields and in an adaptable manner, responding to the variable issues given by landfill leachate.

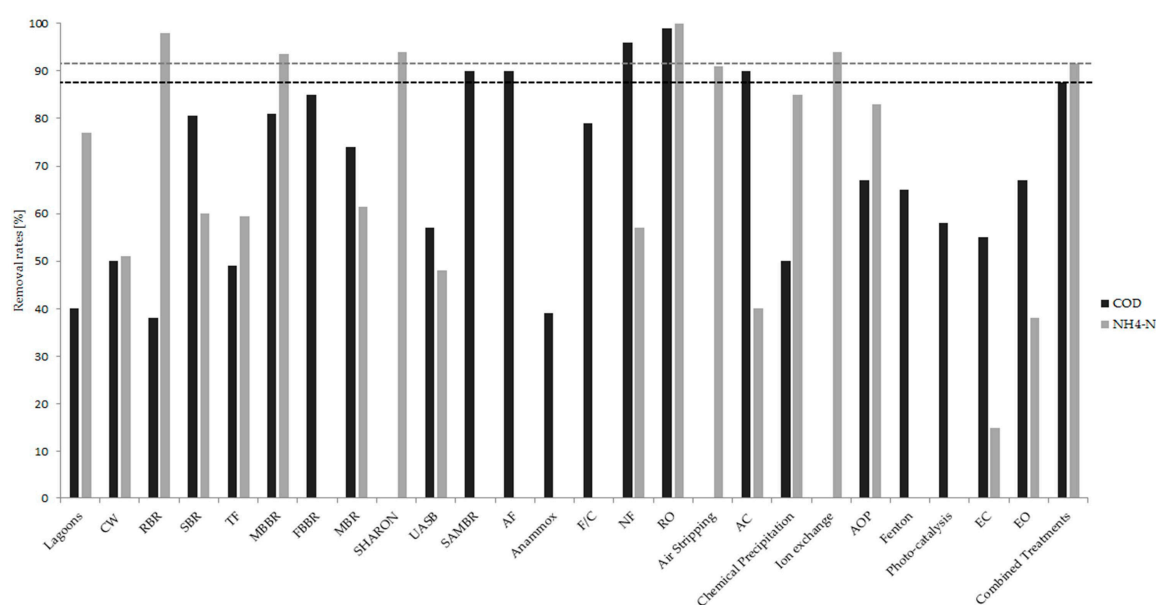


Figure 1. Leachate treatment COD (black) and $\text{NH}_4\text{-N}$ (gray) removal rates. Comparison between single and combined treatments.

As reported in Section 3, the combination of aerated biological treatments and physical-chemical ones is the most used. Indeed, as is visible in Table 3, the results obtained allow considering these practices as reliable for COD and $\text{NH}_4\text{-N}$ removal, with special emphasis on AOP technologies with biological ones, also suggested in previous years and considered reliable solutions [177–179]. However, all of them do not achieve concentrations low enough to release the wastewater to water bodies and further treatments must be applied such as membrane filtration. The introduction into sewage systems is the application suggested, as the treatments allow achieving COD concentrations around 300–1000 mg/L COD and heavy metal removal, so the compounds can be finally manage in another treatment plant (activated sludge systems). However, this possibility is not always applied as landfill leachate is far from cities. An example is given in developing countries, where leachate, once partially handled, is discharged to water bodies directly, affecting the health of the environment and population. This is due to the fact that sewage systems are not fully constructed and do not cover all areas around or treatment plants are still not developed. Another issue is the application of membrane filtration or MBR technologies suggested by the majority of authors in 2016. While these are useful and effective practices to treat landfill leachate, these technologies are cost-effective and large amounts of energy are required. Therefore, the application of such technologies is not feasible within low- to middle-income

countries and the application cannot be considered sustainable even if applied with renewable energy. Moreover, the risks of fouling and clogging are particular issues that required know-how and expertise, as the solutions are not fully replicable.

The majority of untreated landfill leachate is visible in developing countries, and feasible solutions are required, as reported in a few studies [180]. The last studies proposed effective solutions for leachate treatment in developed countries, helping plan developers to choose the best solutions for each particular case study. However, efforts must be implemented for introducing sustainable and inexpensive solutions in order to guarantee environmental protection worldwide.

5. Concluding Remarks

Landfills, or open dumps, are still the preferred solution for MSW end of life worldwide, particularly in developing countries. Therefore, landfill leachate represents a common environmental burden that also affects the health of the population. The presented review sums up the most important technical solutions for leachate treatment that have a key role in releasing landfill leachate to sewage systems or water bodies, highlighting strengths and potential threats. Biological and physical-chemical combined solutions are presented as a reliable way which allows achieving high pollutant removal rates for young, medium and old landfill leachate. As a result, policy-makers and project developers have suitable tools to introduce effective treatment plants for specific landfill conditions. Developing countries are disadvantaged as, often, emerging cities do not have economic funds and sufficient know-how to introduce integrated technologies within the systems. In any case, the general objective of policy-makers should be the implementation of plans focused on waste energy recovery and recycling as they are good options to reduce landfill inflows. At the same time, improving leachate treatment, especially in landfills constructed for big urban areas, is required in order to protect water bodies and the surrounding soils. In any case, other studies need to be developed to detect an effective treatment method for low- to middle-income countries in order to provide solutions for environmental protection and sustainable cities worldwide.

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