

Physico-chemical treatments for removal of recalcitrant contaminants from landfill leachate

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Abstract

In this paper, the technical applicability and treatment performance of physico-chemical techniques (individual and/or combined) for landfill leachate are reviewed. A particular focus is given to coagulation–flocculation, chemical precipitation, ammonium stripping, membrane filtration and adsorption. The advantages and limitations of various techniques are evaluated. Their operating conditions such as pH, dose required, characteristics of leachate in terms of chemical oxygen demand (COD) and $\text{NH}_3\text{-N}$ concentration and treatment efficiency are compared. It is evident from the survey of 118 papers (1983–2005) that none of the individual physico-chemical techniques is universally applicable or highly effective for the removal of recalcitrant compounds from stabilized leachate. Among the treatments reviewed in this article, adsorption, membrane filtration and chemical precipitation are the most frequently applied and studied worldwide. Both activated carbon adsorption and nanofiltration are effective for over 95% COD removal with COD concentrations ranging from 5690 to 17,000 mg/L. About 98% removal of $\text{NH}_3\text{-N}$ with an initial concentration ranging from 3260 to 5618 mg/L has been achieved using struvite precipitation. A combination of physico-chemical and biological treatments has demonstrated its effectiveness for the treatment of stabilized leachate. Almost complete removal of COD and $\text{NH}_3\text{-N}$ has been accomplished by a combination of reverse osmosis (RO) and an upflow anaerobic sludge blanket (UASB) with an initial COD concentration of 35,000 mg/L and $\text{NH}_3\text{-N}$ concentration of 1600 mg/L and/or RO and activated sludge with an initial COD concentration of 6440 mg/L and $\text{NH}_3\text{-N}$ concentration of 1153 mg/L. It is important to note that the selection of the most suitable treatment method for landfill leachate depends on the characteristics of landfill leachate, technical applicability and constraints, effluent discharge alternatives, cost-effectiveness, regulatory requirements and environmental impact.

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1. Introduction

Landfill is one of the most widely employed methods for the disposal of municipal solid waste (MSW). Up to 95% total MSW collected worldwide is disposed of in landfills [1]. After landfilling, solid waste undergoes physico-chemical and biological changes. Consequently, the degradation of the organic fraction of the wastes in combination with percolating rainwater leads to the generation of a highly contaminated liquid called “leachate”.

After the waste is disposed of in a landfill, there is a shift from a short acidic phase (aerobic) to much longer acidogenic and methanogenic phases (anaerobic decomposition). Leachate in the acidic phase contains high concentrations of volatile fatty acids (VFAs). With a molecular weight of less than 120 Da, VFA compounds constitute the majority of organic matter in leachate and are easily biodegradable [2]. The ratio of biological oxygen demand/chemical oxygen demand (BOD/COD) of young leachate in the acidic phase is in the range of 0.4–0.5 [3].

During the methanogenic phase, *methanogenic* bacteria such as *methanogenic archaea* degrade the VFAs and reduce the organic strength of leachate, leading to a pH higher than 7.0. After degradation, only humic-like compounds that have high molecular weight remain in the leachate [4]. Since BOD

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decreases much faster than COD, the ratio of BOD/COD in stabilized leachate is less than 0.1 [5]. Because of the potential of leachate to ultimately find its way into the groundwater, causing contamination with chemical species in dissolved or suspended forms, the generation of landfill leachate creates the potential for a long-term impact on the surrounding environment.

Of toxic compounds such as adsorptive organic halogen (AOX) compounds, humic acids and chloride compounds that remain in stabilized leachate, ammoniacal-nitrogen ($\text{NH}_3\text{-N}$) has been identified as one of the major toxicants to living organisms, as confirmed by toxicity analyses carried out using bioassay methods and test organisms such as *Salmo gairdneri* and *Onchorhynchus nerka* [6]. High concentration of untreated $\text{NH}_3\text{-N}$ can stimulate algal growth, deplete dissolved oxygen through eutrophication and have toxic effects on aquatic organisms [7].

Although the characteristics of landfill leachate depend on the type of MSW being dumped, the degree of solid waste stabilization, site hydrology, moisture content, seasonal weather variations, age of the landfill and stage of the decomposition in the landfill [8], the common features of stabilized leachate are high strengths of $\text{NH}_3\text{-N}$ (3000–5000 mg/L) and moderately high strength of COD (5000–20,000 mg/L), as well as a low ratio of BOD/COD (less than 0.1) [9]. A landfill site may still produce leachate with high concentration of $\text{NH}_3\text{-N}$ over 50 years after filling operations have ceased [10]. If not properly treated, leachate that seeps from a landfill can enter the underlying groundwater, thus posing potentially serious hazards to the surrounding environment and to public health.

As the treatability of landfill leachate depends on its composition and characteristics (Table 1) [11], the nature of the organic matter present as well as the age and structure of the landfill, different technologies including biological treatments, physico-chemical treatments, advanced oxidation process (AOP) as well as natural systems such as constructed wetlands [12–16] and leachate recirculation [17–20] have been developed in recent years, not only to minimize the

Table 1
Characterization of different types of landfill leachate [11]

Type of leachate	Young	Intermediate	Stabilized
Age of landfill (years)	<1	1–5	>5
pH	<6.5	6.5–7.5	>7.5
BOD/COD	0.5–1.0	0.1–0.5	<0.1
COD (g/L)	>15	3–15	<3
$\text{NH}_3\text{-N}$ (mg/L)	<400	NA	>400
TOC/COD	<0.3	0.3–0.5	>0.5
Kjehdal nitrogen (g/L)	0.1–2	NA	NA
Heavy metals (mg/L)	>2	<2	<2

generation of toxic contaminants from leachate, but also to comply with the increasingly stringent discharge standards in different countries (Table 2).

Due to its reliability, simplicity and high cost-effectiveness, biological treatment (suspended/attached growth) is commonly used for the removal of the bulk of leachate containing high concentrations of BOD [26]. When treating young (biodegradable) leachate, biological techniques can yield a reasonable treatment performance with respect to COD, $\text{NH}_3\text{-N}$ and heavy metals. However, when treating stabilized (less biodegradable) leachate, biological treatment may not be able to achieve the permitted maximum COD levels for direct or indirect discharge due to the recalcitrant characteristics of organic carbon in the leachate. As a result, the search for other effective and efficient technologies for the treatment of stabilized landfill leachate has intensified in recent years.

Physico-chemical treatments have been found to be suitable not only for the removal of refractory substances from stabilized leachate, but also as a refining step for biologically treated leachate. Prior to discharge, an additional effluent refining using physico-chemical treatments, such as chemical precipitation, activated carbon adsorption and ion exchange, can be carried out on-site.

Numerous research studies on the treatment of stabilized leachate using different types of physico-chemical treatments

Table 2
Maximum discharge standard from landfill leachate in some countries

Parameters	Maximum discharge standard (mg/L)				
	USA [21]	Germany [22]	France [23]	Hong Kong [24]	South Korea [25]
COD	NA	200	120	200	50
BOD ₅	NA	20	30	800	NA
$\text{NH}_3\text{-N}$	NA	NA	NA	5	50
Total nitrogen	NA	70	30	100	150
Phosphorus	NA	3	NA	25	NA
Cd(II)	0.01	0.1	NA	0.1	NA
Cr(III)	NA	0.5	NA	0.1	NA
Cr(VI)	0.05	0.1	NA	NA	NA
Ni(II)	0.013	1.0	NA	0.6	NA
Pb(II)	0.03	0.5	NA	NA	NA
Cu(II)	0.07	0.5	NA	1.0	NA
Zn(II)	0.3	2	NA	0.6	NA
Ag(I)	0.05	NA	NA	0.6	NA

NA: not available.

have been carried out worldwide in recent years. However, no attempt has so far been made to obtain a comprehensive overview of all the treatments mentioned above in terms of the optimum conditions for the removal of COD and $\text{NH}_3\text{-N}$ from landfill leachate.

In this article, the technical applicability and performance of all available physico-chemical treatments for landfill leachate are exhaustively reviewed. Their advantages and limitations in application are compared. To evaluate their treatment performances on the basis of COD, $\text{NH}_3\text{-N}$ and/or heavy metal removal, selected information on pH, dose required, strength of wastewater in terms of COD, $\text{NH}_3\text{-N}$ and heavy metal concentration, as well as treatment efficiency is presented.

2. Physico-chemical treatments for stabilized landfill leachate

2.1. Individual treatment

2.1.1. Coagulation–flocculation

Coagulation–flocculation has been employed for the removal of non-biodegradable organic compounds and heavy metals from landfill leachate [27–31], as shown in Table 3. The coagulation process destabilizes colloidal particles by the addition of a coagulant. To increase the particle size, coagulation is usually followed by flocculation of the unstable particles into bulky flocs so that they can settle more easily [32]. This technique facilitates the removal of suspended solids and colloid particles from a solution. The general approach for this technique includes pH adjustment and involves the addition of ferric/alum salts as the coagulant to overcome the repulsive forces between the particles [33].

The removal of heavy metals from stabilized leachate containing high concentrations of organic and inorganic matter was investigated using coagulation with FeCl_3 [27]. The metal removal performances were reported to be higher at pH 9.0 than at pH 4.0. The results demonstrated the effectiveness of precipitation at basic pH for the removal of heavy metals [27].

Another application of coagulation–flocculation for the removal of non-biodegradable organic compounds from hazardous landfill leachate was studied by Amokrane et al. [28]. Although the doses required were identical (0.035 mol/L of Fe or Al), with an initial COD concentration of 4100 mg/L, ferric chloride was found to give higher removal of organic compound (55%) than alum (42%). These results were in agreement with the previous study undertaken by Diamadopoulos [29] in the Thessaloniki landfill (Greece). At an initial concentration of 5690 mg/L and at pH 4.8, the maximum COD removal of 56% was achieved with 0.8 g/L of FeCl_3 , as compared to 39% with 0.4 g/L of $\text{Al}_2(\text{SO}_4)_3$. The results of both studies suggest that FeCl_3 is more effective than alum as a coagulant.

Table 3
Removal of recalcitrant compounds using coagulation–flocculation

Location of landfill	Species	Precipitant/ coagulant	Dose (g/L)	Initial concentration in leachate (mg/L)				COD/TOC	pH	Removal efficiency (%)		References
				COD	BOD	$\text{NH}_3\text{-N}$	Metal			COD	$\text{NH}_3\text{-N}$	
NA	Cd(II) Cu(II) Ni(II) Pb(II) Zn(II) Cr(VI)	FeCl_3	0.3	NA	NA	NA	0.5	NA	9.0	NA	97 74 82 98 95 87	[27]
Jeandela-incourt (France)		FeCl_3 $\text{Al}_2(\text{SO}_4)_3$	2.0 0.9	4100	200	1040	NA	2.9	4.5–5.0	55 42	NA NA	[28]
Thessaloniki (Greece)	NA	FeCl_3 $\text{Al}_2(\text{SO}_4)_3$	0.8 0.4	5690	2215	580	NA	NA	4.8	56 39	NA NA	[29]
Thessaloniki (Greece)	-	FeCl_3	1.5 5.5	5350 70900	1050 26800	940 3100	NA NA	NA NA	10.0 6.2	80 30	- -	[30]
Turkey	NA	$\text{Ca}(\text{OH})_2$	1.5 3.0 2.0	5350 70900 7000	1050 26800 NA	940 3100 700	NA NA NA	NA NA NA	10.0 6.2 12.0	38 40 86	- -	[31]

NA: not available.

In a similar study, the application of coagulation–flocculation for the treatment of stabilized leachate from the Thessaloniki landfill (Greece) was reported by Tatsi et al. [30]. Without pH adjustment, the addition of 1.5 g/L of FeCl₃ was able to increase the COD removal rate to 80%, while 1.5 g/L of Al³⁺ ions resulted in up to 38% reduction of COD (Table 3). These results were in agreement with another study carried out by Kargı and Pamukoglu in Turkey [31]. After 30 h of fed-batch operation, coagulation–flocculation treatment using 2 g/L lime achieved 86% COD removal with an initial COD concentration of 7000 mg/L [31].

Overall, it is found that coagulation–flocculation technique using FeCl₃ is effective for the removal of organic compounds and heavy metals. To improve the removal of COD from leachate, lime can be employed as a coagulant, as shown in Table 3. The other drawbacks of this technique include the high operational cost due to high chemical consumption, the sensitivity of the process to pH and the generation of sludge. It is important to note that the velocity gradient, settling time and pH play major roles in increasing the probability of the settling of colloidal particles.

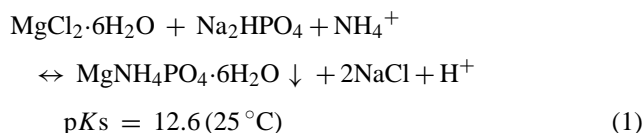
2.1.2. Chemical precipitation

Due to its capability, the simplicity of the process and inexpensive equipment employed, chemical precipitation has been employed for the removal of non-biodegradable organic compounds, NH₃–N and heavy metals from landfill leachate [34–38]. During chemical precipitation, dissolved ions in the solution are converted to the insoluble solid phase via chemical reactions. Typically, the metal precipitate from the solution is in the form of hydroxide. Struvite (magnesium ammonium phosphate (MAP)) or lime is usually employed as the precipitant, depending on the target of the removal [either NH₃–N or heavy metal].

The removal of ammoniacal–nitrogen from anaerobically pre-treated leachate in the Odayeri landfill (Turkey) was studied using struvite precipitation [34]. Using this technique, ammonia was converted into a nitrogen fertilizer such as urea. About 50% COD and 90% NH₃–N, with an initial COD concentration of 4024 mg/L and NH₃–N concentration of 2240 mg/L, were removed. The results indicate that struvite precipitation was more effective for the removal of NH₃–N than for organic compounds removal. These results were in agreement with another study conducted by Calli et al. [35]. In their study, struvite with a stoichiometric ratio of Mg:NH₄:PO₄ = 1:1:1 was employed to reduce the NH₃–N concentration in young leachate from the Komurcuoda landfill (Turkey). About 98% NH₃–N with an initial concentration of 3260 mg/L was precipitated at pH 7.5. In addition to NH₃–N, 20% COD was also removed under the same conditions.

In a similar study, the removal of ammonium from stabilized leachate in the WENT landfill (Hong Kong) was also carried out by applying struvite precipitation [36]. The precipitation of struvite forms an insoluble compound which can be easily separated from the liquid, as shown from the

following reaction:



With a 1:1:1 mole ratio of Mg²⁺, NH₄⁺ and PO₄³⁻, about 98% removal of NH₃–N with an initial concentration of 5618 mg/L was achieved at pH 9.0 after 15 min. The advantage of struvite precipitation is that the sludge produced after treatment may be utilized as a nitrogen fertilizer if the leachate does not contain any heavy metals. However, since COD was not significantly removed during the treatment, biological steps needed to be conducted after precipitation to reduce the organic loading of leachate [37].

The uptake of heavy metals such as Cu(II), Pb(II), Fe(II), Mn(II) and Ni(II) from young leachate in the Gaziantep landfill (Turkey) was explored by employing lime as a precipitant [38]. Table 4 shows that 8 g/L of lime was found to be reasonably effective for metal precipitation. In addition to lime addition, pH adjustment to 11.0 was suggested as a means to enhance metal precipitation. However, the drawbacks of chemical precipitation include the high dose of precipitant required, the sensitivity of the process employed to pH, the generation of sludge and the need for further disposal of the sludge.

2.1.3. Ammonium stripping

Due to its effectiveness, ammonium stripping is the most widely employed treatment for the removal of NH₃–N from landfill leachate [29,34,35,39,40]. Prior to anaerobic treatment, landfill leachate containing NH₃–N and the air phase are allowed to interact in a countercurrent flow in a stripping tower: NH₃–N is transferred from the waste stream into the air and is then absorbed from the air into a strong acid such as sulphuric acid or directly flux into the ambient air [41].

The treatment of stabilized leachate from the Thessaloniki landfill (Greece) using ammonium stripping for 24 h was investigated [29]. With an initial NH₃–N concentration of 2215 mg/L, about 95% NH₃–N was removed at pH 11.5. The NH₃–N removal was found to improve with an increasing flow rate, as a result of a greater interaction between the liquid and the air phases.

A laboratory-scale study of the treatment of young leachate from the Komurcuoda landfill (Istanbul) by using ammonium stripping for 12 h was carried out [35]. About 94% NH₃–N removal with an initial concentration of 3260 mg/L was achieved by adding 11 g/L of lime. However, under the same conditions, with air stripping, the COD removal was always lower than 15%. This suggests that ammonium stripping treatment alone was not effective for the removal of non-biodegradable compounds from young leachate. Subsequent treatment using biological methods such as nitrification would be required to improve the degradation of recalcitrant organic compounds from the leachate.

Table 4
Chemical precipitation for treatment of non-biodegradable organic compounds

Location of landfill	Species	Precipitant/ coagulant	Dose (g/L)	Initial concentration in leachate (mg/L)				pH	Removal efficiency (%)		References	
				COD	BOD	NH ₃ -N	Metal		COD	NH ₃ -N		Metal
Oyaderi (Turkey)	NA	Struvite	NA	4024	NA	2240	NA	2.48	50	90	NA	[34]
Komurcuoda (Turkey)	NA	Struvite	NA	47800	28500	3260	NA	NA	20	98	NA	[35]
WENT (Hong Kong)	NA	Struvite	NA	7511	1652	5618	NA	2.90	53	98	NA	[36]
Gaziantep (Turkey)	Cu(II)	Ca(OH) ₂	8.0	37026	15550	2430	0.11	NA	NA	NA	72	[38]
	Pb(II)						1.20				46	
	Fe(II)						6.60				78	
	Mn(II)						0.36				97	
	Ni(II)						2.00				51	

NA: not available.

Another study on the treatment of leachate from the Junk Bay landfill (Hong Kong) using ammonium stripping was carried out in laboratory scale [39]. About 10 g/L lime was used to adjust the pH of leachate to 11. After pH adjustment, approximate 90% NH₃-N with an initial concentration of 500 mg/L was removed after ammonium stripping for 12 h (Table 5). This can be due to the fact that at pH 11, most NH₃-N was in the form of NH₃ gas, thus resulting in a higher removal of NH₃-N. Under the same conditions, 47% COD removal was achieved. The results suggest that ammonium stripping was more effective for the removal of NH₃-N than for organic compounds removal.

The treatment of young leachate from the Mustankorkea landfill (Finland) was explored by separately employing ammonium stripping and nanofiltration (NF) [40]. At pH 11, ammonium stripping with 24 h of retention time was able to remove 89% NH₃-N and 21% COD with initial concentrations of 220 and 920 mg/L, respectively (Table 5). However, only 50% NH₃-N and 66% COD removal were achieved by nanofiltration alone at the same concentrations (Table 6). The results of ammonium stripping treatment were in agreement with those obtained in another study undertaken by Ozturk et al. [34] in the Oyaderi landfill (Turkey) using anaerobically pre-treated leachate, where 85% NH₃-N with an initial concentration of 1025 mg/L was removed by the stripping process alone.

As a whole, ammonium stripping gives a NH₃-N treatment performance in the range of 85–95% with concentrations ranging from 220 to 3260 mg/L. The reduction in COD, however, is relatively low of less than 47% with its concentration ranging from 500 to 47,800 mg/L. Prior to treatment, pH of leachate can be easily adjusted to basic conditions (pH 11–12) to improve the removal of NH₃-N by stripping process. Another advantage of this is that it is possible to meet the NH₃-N discharge standard using ammonium stripping [42]. In terms of operational cost, ammonium stripping was found to be more economically appealing than other treatments such as reverse osmosis or nanofiltration.

In spite of its advantages, the major drawbacks of ammonium stripping are the environmental impact due to the release of NH₃ gas into the atmosphere. Therefore, there is a need for further treatment of the gas with HCl or with H₂SO₄, thus increasing the operational cost of waste treatment due to chemicals. The other limitations of this technique are the CaCO₃ scaling of the stripping tower when lime is employed for pH adjustment, the need for pH adjustment of the treated effluent prior to discharge and the difficulty in removing ammonia with concentrations of less than 100 mg/L [43,44].

2.1.4. Membrane filtration

2.1.4.1. Nanofiltration. Due to its unique properties between ultrafiltration (UF) and reverse osmosis (RO) membranes, NF has found a place in the removal of recalcitrant organic compounds and heavy metals from landfill leachate [27,34,45]. This treatment process has the ability to remove particles with a molecular weight of higher than 300 Da as

Table 5
Treatment performance of ammonium stripping for removal of ammoniacal–nitrogen

Location of landfill	Chemical for pH adjustment	Dose (g/L)	Initial concentration in leachate (mg/L)			BOD/COD	COD/TOC	pH	Removal efficiency (%)		References
			COD	NH ₃ -N	BOD				COD	NH ₃ -N	
Thessalomiki (Greece)	Ca(OH) ₂	3.1	5690	2215	580	0.1	NA	11.5	NA	95	[29]
Oyaderi (Turkey)	Ca(OH) ₂	8	5730	1025	NA	NA	2.34	11.0	25	85	[34]
Komurcuoda (Turkey)	Ca(OH) ₂	11	47800	3260	28500	0.6	NA	11.0	<15	94	[35]
Junk Bay (Hong Kong)	Ca(OH) ₂	10	500	500	NA	NA	NA	11.0	47	90	[39]
Mustankorkea (Finland)	NA	NA	920	220	84	0.4	NA	11.0	21	89	[40]

NA: not available.

well as inorganic substances through electrostatic interactions between the ions and membranes. The significance of this membrane lies in its surface charges, which allow charged solutes smaller than the membrane pores to be rejected, along with bigger neutral solutes and salts.

Urase et al. [27] employed a NTR-7250 membrane to remove heavy metals. It was reported that more than 99% removal was achieved with initial metal concentrations of 0.69 mg/L of Cr³⁺ and 0.23 mg/L of Cu²⁺, respectively.

Treatment of anaerobically pre-treated leachate from the Odayeri landfill (Turkey) was undertaken using NF [34]. With an initial COD concentration of 3000 mg/L and NH₃-N concentration of 950 mg/L, about 89% COD and 72% NH₃-N removal could be achieved with total operating cost of US\$ 0.8 m⁻³.

In a similar study, the removal of heavy metals from stabilized leachate was carried out by employing NF [45]. Using a AFC-30 membrane, over 88% metal cations (Pb²⁺, Zn²⁺ and Cd²⁺) with initial concentrations of less than 0.70 mg/L were removed.

In general, NF has demonstrated a moderate treatment performance (>65% treatment efficiency) for the removal of organic compounds with COD concentration ranging from 920 to 3000 mg/L (Table 6). Due to the negatively charged groups on the membrane, NF is also effective for the removal of heavy metals, as shown in Table 6. The application of NF allows material dissolved in water to be separated into monovalent and divalent ions. Unlike RO, NF has a looser membrane structure, enabling higher fluxes and lower operating pressure for the treatment of leachate.

2.1.4.2. Reverse osmosis. With high fluxes and the ability to operate over wide temperature and pH range, RO is another alternative physico-chemical treatment for stabilized leachate. In RO application, any solvent that contains metal cations is passed through a membrane in such a way that the metal concentrations are reduced [46]. With 98–99% rejection rate for organic and inorganic contaminants, RO can be used for the removal of heavy metals, suspended/colloidal materials and dissolved solids from landfill leachate [47].

The treatment of young leachate from the Chung Nam landfill (South Korea) was carried out using an RO system [25]. About 96–97% removal of COD and NH₃-N was achieved with initial concentrations of 1500 and 1400 mg/L, respectively. The results suggest that RO greatly enhanced treatment efficiency by removing non-biodegradable organic compounds from landfill leachate.

The removal of dioxins, such as polychlorinated dibenzo-p-dioxins (PCDD), polychlorinated dibenzofurans (PCDF) and polychlorinated biphenyls (PCB), from the Yachiyo landfill (Japan) was studied using RO [48]. Complete removal of the three toxic substances with an initial concentration of 2.35 pg/L was achieved to meet the environmental legislation. With an initial COD concentration of 97.4 mg/L and NH₃-N concentration of 33.7 mg/L, reverse osmosis was found to achieve complete COD removal and 98% NH₃-N removal.

Table 6
Removal of organic and inorganic compounds using UF, NF or RO

Location of landfill	Kind of process	Type of membrane	Species	Pressure (bar)	Initial concentration (mg/L)				BOD/COD	pH	Rejection rates (%)			References
					COD	NH ₃ -N	Metal	BOD			COD	NH ₃ -N	Metal	
NA	NF	NTR-7250	Cr(III) Cu(II) Pb(II)	3	NA	NA	0.69 0.23 0.03	NA	NA	NA	NA	NA	100 99 93	[27]
Odayeri (Turkey)	NF	SW	NA	25	3000	950	NA	NA	NA	NA	89	72	NA	[34]
Mustankorkea (Finland)	NF	Desal 5-DL	NA	6–8	920	220	NA	84	0.40	7.6	66	50	NA	[40]
Spillepeng (Sweden)	NF	AFC-30	Pb(II) Zn(II) Cd(II)	20	2000	NA	0.61 0.50 0.03	NA	NA	NA	NA	NA	97 88 94	[45]
Chung Nam (South Korea)	RO	SW-4040	NA	NA	1500	1400	NA	450	0.30	NA	97	96	NA	[25]
Yachiyo (Japan)	RO	DT	Mn(II)	9–11	97.4	33.7	4.77	5	0.05	6.0	100	98	100	[48]
Pietramelina (Italy)	RO	SW30-2521	Cd(II) Zn(II) Cu(II)	52	3840	NA	0.50	1200	0.31	6.0	98	NA	100 97 99	[49]
Hedeskoga (Sweden)	RO	AFC99	Cr(III)	40	1254	541	0.02	125	0.10	7	95	82	NA	[50]
Spillepeng (Sweden)	RO	NA	NA	30	925	280	NA	NA	NA	6.5	98	98	NA	[53]
Wijster (Holland)		NA	NA	40	335	140	NA	NA	NA	6.5	98	98	NA	
Ihlenberg (Germany)	RO	NA	NA	36–60	1797	366	0.25	54	0.03	7.7	99	100	98	[54,55]
	NF			NA	17000	3350	NA	510	0.03	6.4	96	58	NA	
Lipówka (Poland)	RO	SS	NA	27.6	1780	743	NA	331	0.28	7–8	97	NA	NA	[56]
	UF	PVC	NA	3							52	NA	NA	

NA: not available.

A laboratory-scale study of the treatment of leachate from the Pietramelina landfill (Italy) was undertaken by applying RO [49]. At a pressure of 52 bar, the maximum rejection rate of COD with an initial concentration of 3840 mg/L was found to be 98%. At the same metal concentration of 0.5 mg/L, the rejection rate of Cd(II) ions was found to be slightly higher (100%) than that of Zn(II) ions (97%) and Cu(II) ions (99%).

In another study, the treatment of stabilized leachate from the Hedekosga landfill (Sweden) was conducted using RO on a pilot-scale [50]. The maximum removal of COD and NH₃-N with initial concentrations of 1254 and 541 mg/L was found to be 95 and 82%, respectively. The overall cost for a full-scale treatment was about US\$ 4.25 m⁻³. The treatment was inexpensive, when compared to that in the study carried out by Rudolph and Köppke that cost US\$ 7–10 m⁻³ [51]. For a complete treatment system where RO is the secondary step after a biological treatment, the overall cost varied between US\$ 35 and 40 m⁻³ [52].

Reverse osmosis was also employed for the treatment of leachate from the Spillepeng (Sweden) and the Wijster landfills (The Netherlands) [53]. The reduction of COD and NH₃-N was found to be 98%, with the initial concentrations as presented in Table 6. An adjustment of pH to below 6.5 was found to maintain the flux at a constant level of 16–31 L/(m² h) for 60 h.

A comparative study of the treatment of stabilized leachate from the Ihlenberg landfill (Germany) was evaluated using RO and NF [54,55]. At a permeate flux of 15 L/(m² h), RO gave a higher removal efficiency of COD (99%) and NH₃-N (100%), when compared to NF (COD: 96%; NH₃-N: 58%). Table 6 presents the initial concentrations of COD and NH₃-N used in this study.

The application of RO and UF for the treatment of stabilized leachate from the Lipówka landfill (Poland) was also explored [56]. With an initial concentration of 1780 mg/L, RO gave a higher COD removal efficiency (97%) than UF (52%). Due to the high loading and varying composition of leachate, a combination of biological treatment and RO is required for further purification of the leachate.

Overall, RO has been found to be highly effective for the removal of both COD and NH₃-N. More than 95% COD was removed with the initial concentrations of organic compounds ranging from 335 to 3840 mg/L, while more than 96% NH₃-N removal could be achieved with an initial concentration ranging from 33 to 1400 mg/L.

In spite of its advantages, the drawbacks of RO include a low retention of small molecules that pass through the membrane and membrane fouling, an undesirable deposition of suspended or dissolved substances on the external surface of membrane [57]. Other limitation of RO is high energy consumption. Peters [58] reported that the cost for energy consumption accounted for about 60–80% RO treatment. Therefore, the affordability of RO needs to be considered carefully during the selection of treatment in order to justify it as a solution.

As a whole, Table 6 shows that the characteristics of certain membrane affect its treatment performance for organic compounds and ammoniacal-nitrogen. It is widely known that the passage of water through the membranes also depends on porosity, material, hydrophilicity, thickness, roughness and charge of the membrane [59]. Table 7 summarizes the characteristics of some membranes and their treatment performance for landfill leachate.

It is apparent from Table 7 that membranes with polyamide or cellulose acetate as their skin material have higher removal of organic compounds and NH₃-N and can work at a wide range of temperature (5–35 °C), compared to those constituted of polyvinyl chloride. This can be due to the fact that polyamide composing the membranes has a higher porosity and hydrophilicity than other materials such as polysulphone or polyethylene-terephthalate [59]. It is important to note that the selection of appropriate membrane for leachate treatment depends on a number of factors such as the characteristics of wastewater, the nature and concentration of materials present in the leachate, pH and temperature [11].

2.1.5. Activated carbon adsorption

Among the treatment techniques reviewed above, adsorption is the most widely employed method for the removal of recalcitrant organic compounds from landfill leachate [60–73]. Basically, adsorption is a mass transfer process by which a substance is transferred from the liquid phase to the surface of a solid, and becomes bound by physical and/or chemical interactions. Due to its inherent physical properties, large surface area, microporous structure, high adsorption capacity and surface reactivity, adsorption using granular activated carbon (GAC) or powder activated carbon (PAC) has been receiving a considerable attention recently for the removal of organic and inorganic pollutants from contaminated wastewater.

In 1995, the removal of organic compounds from stabilized leachate in the Goslar landfill (Germany) was evaluated using GAC [61]. The results of the column studies showed that 91% COD removal with an initial concentration of 940 mg/L was achieved. The kinetic rate of adsorption was found to be affected not only by film diffusion, but also by the rate of adsorption and by the internal surface diffusion on the solid surface of an adsorbent.

The treatment of landfill leachate using a separate GAC adsorption, granular activated alumina (GAA) and/or ferric chloride (FC) was also carried out [62]. Among the three adsorbents investigated, GAC was found to be the most effective adsorbent for the removal of the heavy metals (Cd(II), Cu(II), Cr(III), Mn(II), Pb(II) and Zn(II)) (Table 8). About 80–96% heavy metals with an initial concentration of 184 mg/L were able to be removed at a pH range of 6–7.7 with 2 g/L of GAC. At equilibrium, the Freundlich isotherm was reported to be representative for GAC adsorption.

A comparative study of the removal of NH₃-N from stabilized leachate in the Burung Island landfill (Malaysia) was undertaken by using GAC and/or limestone [63]. About

Table 7
Characteristics of some membranes and their treatment performance for landfill leachate

Type of membrane	Type of application	Skin materials	Type of module	Pressure (bar)	Membrane area (m ²)	Temperature (°C)	Manufacturer	Removal (%)		References
								COD	NH ₃ -N	
SW	NF	NA	Spiral wound	25	2.0	25	NA	89	72	[34]
Desal 5-DL	NF	NA	Flat sheet	6–8	4.5×10^{-3}	NA	NA	66	50	[40]
DT	RO	Polyamide	NA	9–11	7.6	5–35	NA	100	98	[48]
SW30-2521	RO	NA	Spiral	52	NA	28	DOW (Italy)	98	NA	[49]
SW-4040	RO	Polyamide	Spiral wound	NA	6.7	NA	Filmtech (USA)	97	96	[25]
AFC99	RO	Polyamide	Tubular	40	9×10^{-1}	15	AFC99 (UK)	95	82	[50]
SS	RO	Cellulose acetate	Tubular	27.6	1.55×10^{-2}	25	NA	97	NA	[56]
PVC	UF	Polyvinyl chloride	Tubular	3	2.5×10^{-2}	25	NA	52	NA	[56]

NA: not available.

40% NH₃-N with an initial concentration of 1909 mg/L was removed with 42 g/L of GAC, while 56 g/L of lime was able to remove 19% NH₃-N under the same concentration. In spite of being less efficient, lime was found to be more cost-effective than GAC for NH₃-N removal.

In Greece, the adsorption of organic compounds from stabilized leachate in the Thessaloniki landfill was studied using powder activated carbon, varying the dose from 0.2 to 10.0 g/L [29]. About 95% COD with an initial concentration of 5690 mg/L was removed with 6 g/L of PAC. The Freundlich isotherm was found to be applicable for adsorption equilibrium, thus suggesting that multilayer adsorption occurred on the surface of PAC [64].

In addition to GAC or PAC, non-conventional materials that are locally available in large quantities, such as agricultural waste or industrial by-products, can be chemically modified and utilized as low-cost adsorbents [65,66]. The conversion of waste, representing an unused resource, into activated carbon which can be used as an adsorbent for water purification, would add considerable economic value, helping the industry reduce the cost of waste disposal and, most importantly, providing an inexpensive alternative to costly commercial activated carbon [67,68]. Low-cost adsorbents such as coconut shell charcoal [69,71] and zeolite [72] can be employed to reduce the COD loading of landfill leachate.

To improve the treatability of leachate from a local landfill in Turkey, Kargı and Pamukoglu employed zeolite and PAC in the treatment [72]. With an initial COD concentration of 7000 mg/L, the application of 5 g/L of PAC and/or zeolite, respectively, achieved 87 and 77% COD removal. With an initial NH₃-N concentration of 700 mg/L, however, 1 g/L of PAC and/or zeolite was able to remove 30 and 40% NH₃-N, respectively. The results indicate that PAC was slightly more effective than zeolite for COD removal at 5 g/L of dose, while zeolite had a higher removal efficiency for NH₃-N than PAC at 1 g/L of dose.

In general, the application of activated carbon adsorption (GAC or PAC) is effective for the removal of non-biodegradable compounds from leachate, but not for NH₃-N. More than 90% COD was removed with its concentration ranging from 940 to 7000 mg/L. However, the need for frequent regeneration of activated carbon column and the high cost of GAC may limit its application for the treatment of landfill leachate in developing countries.

2.1.6. Miscellaneous treatment technologies

2.1.6.1. Ion exchange. Ion exchange is a reversible interchange of ions between the solid and liquid phases where there is no permanent change in the structure of the solid. This treatment is capable of effectively removing the traces of metal impurities to meet the increasingly strict discharge standards in developed countries. Prior to ion exchange, the leachate should first be subjected to a biological treatment. Although the application of ion exchange is not commonly employed for the treatment of landfill leachate, it has received considerable interest in Germany for the removal of non-

Table 8
Treatment performance of adsorption technique on refractory compounds

Location of landfill	Species	Type of adsorbent	Dose of adsorbent (g/L)	Initial concentration in leachate (mg/L)			BOD/COD	pH	Removal efficiency (%)		Metal capacity (mg/g)	Commercial price (US\$/kg)	References
				COD	NH ₃ -N	Metal			COD	NH ₃ -N			
Goslar (Germany)	NA	GAC Filtra-sorb 400	9.1	940	NA	NA	0.003	7.5	91	NA	564	NA	[61]
NA	Cd(II)	GAC	2	NA	NA	184	NA	7.7	NA	NA	81.88	NA	[62]
	Cu(II)							6.0			80.04		
	Cr(III)							6.0			70.84		
	Mn(II)							6.0			71.76		
	Pb(II)							6.0			87.40		
	Zn(II)							7.0			77.28		
	Cd(II)	GAA	2	NA	NA	184	NA	8–9	NA	NA	31.28	NA	[62]
	Cu(II)							8–9			30.36		
	Cr(III)							8–9			15.64		
	Mn(II)							8–9			36.80		
	Pb(II)							8–9			37.72		
	Zn(II)							8–9			31.28		
	Cd(II)	FC	2	NA	NA	184	NA	3–7	NA	NA	10.12	NA	[62]
	Cu(II)							3–7			13.80		
Cr(III)	3–7							36.80					
Mn(II)	3–7							14.72					
Pb(II)	3–7							16.56					
Zn(II)	3–7							12.88					
Burung Island (Malaysia)	NA	GAC	42	3450	1909	NA	0.33	9.7	NA	40	NA	2.65	[63]
Japan	NA	GAC	6.4	221	332	NA	0.12	7.0	NA	NA	NA	NA	[64]
La Zoreda (Spain)	NA	GAC	20	5108	1876	0.4	0.20	7.0	93	NA	38.12	NA	[73]
NA	NH ₄ ⁺	PAC	5	7000	700	NA	NA	NA	87	NA	NA	NA	[72]
		Zeolite	5	7000	700	NA	NA	NA	77	NA	NA	NA	[72]
		PAC	1	7000	700	NA	NA	NA	NA	NA	30	NA	[72]
		Zeolite	1	7000	700	NA	NA	NA	NA	NA	40	NA	[72]
Thessaloniki (Greece)	NA	PAC	6	5690	2215	NA	0.10	NA	95	NA	NA	NA	[29]

NA: not available.

biodegradable compounds that contain humic substances [60].

In a study carried out by Rodríguez et al. [73], the removal of humic substances from stabilized leachate in the La Zoreda landfill (Spain) was evaluated using ion exchange resins such as Amberlite XAD-8, XAD-4 and Amberlite IR-120 and/or granular activated carbon adsorption. Among the adsorbents studied, GAC was found to achieve the highest removal of COD (93%), followed by Amberlite XAD-8 (53%), XAD-4 (46%) and IR-120 (31%) at the initial COD concentration of 5108 mg/L (Table 9). Unlike GAC adsorption, the synthetic resins gave low COD removal due to the effect of binding-site competition with heavy metals from the leachate.

The removal of ammonia from landfill leachate by ion exchange was compared to that by ozonation [74]. Ozonation is capable of converting nitrite to nitrate, but is less effective to convert ammonia to nitrate. While ion exchange can reduce the concentration of both nitrate and NH_4^+ ions to desired levels, it was reported that about 500 bed volume (BV) of ammonium with an initial concentration of 20 mg/L was removed at a pH range of 7–9 using ion exchange alone. Only 250 BV of ammonium was removed by ozonation treatment alone at the same pH range with an ozone consumption of 0.29 mg of NH_4^+ /mg of ozone [74]. The aeration (stripping) effect may be the reason for ammonia removal by ozonation.

In addition to organic compounds and ammonia, ion exchange using kaolinite was employed to study the sorption of Cd(II) and Ni(II) from leachate from a landfill in Italy [75]. It was reported that the removal of Ni(II) was higher (99%) than that of Cd(II) (90%) with an initial Ni(II) concentration of 0.94 mg/L and Cd(II) concentration of 0.002 mg/L. It was found that both metals were removed after contacting with kaolinite. Depending on the type of organic matter present and the ion exchange resin employed, ion exchange is effective for heavy metal removal from landfill leachate. After an aerobic pre-treatment, ion exchange normally achieves an excellent metal removal from effluents.

Unlike other physico-chemical treatments such as adsorption, coagulation–flocculation or membrane filtration, landfill researchers rarely employed ion exchange technique for the removal of heavy metals from leachate. This could be due to the fact that the concentration of heavy metals in the landfill leachate is low (less than 2 mg/L) [38], making heavy metals not the focus of pollutant removal in the treatment of leachate. In addition, the application of ion exchange for such purpose is not economically appealing due to high operational cost. Other limitation is that, prior to ion exchange, appropriate pre-treatment system such as the removal of suspended solids from leachate is required. As a result, data and information on the treatment results of ion exchange for heavy metal removal from leachate are rarely reported.

2.1.6.2. Electrochemical treatment. Electrochemical treatment such as membrane electrodialysis has also contributed to environmental protection in France [76] and Brazil [77]. In Rio Claro (Brazil), the electrodegradation of stabilized land-

Table 9
Uptake of organic contaminants by using ion exchange

Location of landfill	Species	Ion exchanger	Dose of resin (g/L)	Initial concentration in leachate (mg/L)				BOD/COD	pH	Adsorption capacity (mg/g)			Metal removal (%)	References
				COD	$\text{NH}_3\text{-N}$	BOD	Metal			COD	$\text{NH}_3\text{-N}$	Metal		
La Zoreda (Spain)	Ni(II)	Amberlite XAD4	NA	5108	1876	1025	0.690	0.20	7.0	20.33	NA	NA	NA	[73]
	Zn(II)	Amberlite XAD8	NA	5108	1876	1025	0.280	0.20	7.0	24.94	NA	NA	NA	
	Cu(II)	Amberlite IR-120	NA	5108	1876	1025	0.410	0.20	7.0	14.82	NA	NA	NA	
Italy	Ni(II)	Kaolinite	192	6378	NA	NA	0.940	NA	8.3	NA	NA	NA	99	[75]
	Cd(II)	Kaolinite	192	6378	NA	NA	0.002	NA	8.3	NA	NA	NA	90	

NA: not available.

Table 10
Electrochemical technologies for degradation of organic contaminants

Location of landfill	Rio Claro (Brazil)
Anode	TiO ₂
Cathode	Ti
Electrical current (A/m ²)	1160
Power consumption (kWh/m ³)	NA
Initial concentration in leachate (mg/L)	
COD	1855
NH ₃ -N	1060
BOD	463.75
BOD/COD	0.25
COD/TOC	1.5
pH	NA
Removal efficiencies (%)	
COD	73
NH ₃ -N	49
Metals	NA
Reference	[77]

NA: not available.

fill leachate was investigated by employing a flow electrochemical reactor [77]. Using a constant flow rate of 2000 L/h for 180 min and at a current density of 1160 A/m², the maximum removal of COD and NH₃-N with initial concentrations of 1855 and 1060 mg/L was found to be 73 and 49%, respectively (Table 10). The results suggest that electrodegradation was an alternative means to breakdown recalcitrant organic compounds in landfill leachate. Due to high energy consumption, however, this technology is more expensive than other treatment methods. As a result, this treatment technique has been investigated less extensively for the treatment of stabilized leachate.

2.2. Combined treatments

2.2.1. Combination of two or more physico-chemical treatments

The characteristics of coagulation–Fenton reaction for the treatment of stabilized leachate from the Metropolitan landfill (South Korea) were evaluated [78]. With an initial COD concentration of 417 mg/L, the removal of recalcitrant compounds (with molecular weights higher than 500 Da) improved from 48% by using coagulation alone to 73% by using combined treatments. The results suggest that the coagulation step enhanced Fenton oxidation for the removal of organic compounds from the leachate.

Another study of the treatment of stabilized leachate was conducted by comparing coagulation–flocculation to the Fenton oxidation (Fe(II)/H₂O₂) process in combination with GAC adsorption [79]. Fenton oxidation pre-treatment was found to improve the adsorption capacity of GAC for COD removal (443 mg/g of GAC) at pH 4.0, compared to coagulation–flocculation (193 mg/g of GAC) at pH 5.7. This might be due to the transformation of organic compounds into oxidation by-products that had smaller molecules than

the starting compounds. As a result, by-products were able to pass through the micropores of GAC.

Rivas et al. [80] combined a sequential coagulation–flocculation and Fenton oxidation process in their study. The aim of employing this combined process is to remove the colloidal particles present in the leachate. About 90% COD removal was achieved with an initial concentration of 7400 mg/L at pH 8.5 by using 0.8 g/L of FeCl₃.

A combination of coagulation and photo-oxidation (UV–vis) for the treatment of stabilized leachate from the Qingshan landfill (China) was explored [81]. This combined treatment with 0.5 g/L FeCl₃ as the coagulant was able to remove 64% COD with an initial concentration of 5800 mg/L. However, only 31% COD removal was achieved by UV–vis irradiation alone at the wavelength (λ) of 313 nm under the same COD concentration. This result suggests that the combined treatment is more effective than separate process.

In the Bordo Poniente landfill (Mexico), the transformation of recalcitrant organic compounds from stabilized leachate using a coagulation–ozonation coupling was investigated [82]. The use of iron(III) sulphate as the coagulant during pre-treatment was found to be effective. At pH 4–5, the two-step treatment with ozonation gave 78% COD removal with an initial concentration of 5000 mg/L.

The treatment efficiency of stabilized leachate from the Gramacho landfill (Brazil) by combining coagulation–flocculation and ozonation followed by ammonium stripping for 96 h was evaluated [83]. This combination was found to be effective for complete removal of NH₃-N with an initial concentration of 800 mg/L. When 3 mg/L of ozone was employed, ozonation was able to achieve 48% COD removal with an initial concentration of 3460 mg/L.

The treatment of stabilized leachate from the Saint-Nazaire landfill (France) using a combination of NF and coagulation was evaluated [84]. Using MPT-31 membrane with high negative charge, maximum COD and NH₃-N removal with initial concentrations of 2150 and 790 mg/L, respectively, was found to be 80 and 21%. By employing MPT-20 that has a low negative charge, a lower removal of COD (74%) and NH₃-N (12%) was achieved at the same conditions. The difference of treatment performance between the two membranes might be attributed to the electrostatic effects that affect the repulsion between the negatively charged membrane surface and humic-type compounds.

A pilot-scale study of the application of NF and PAC adsorption for the treatment of biologically pre-treated leachate from a landfill in Germany was also conducted [22]. Meier et al. [22] reported that the combined treatments were able to remove 97% COD with an initial concentration of 1450 mg/L. The result suggests that the combination of the two treatments improved the efficiency of COD removal, compared to the other study carried out by Marttinen et al. [40] that employed NF alone for the removal of about 66% COD with an initial concentration of 920 mg/L (Table 6).

The treatment of stabilized leachate from the Berg landfill (Germany) was explored using a combination of NF and

GAC adsorption/ozonation [85]. Due to its high rejection rate for organic compounds and high permeate flux, Desal 5K was selected as the NF membrane. An approximate 99% COD rejection with an initial concentration of 4000 mg/L was achieved. To avoid CaSO_4 scaling and for reasons of cost, HCl was used for pH adjustment.

Treatment of landfill leachate using a combination of ozonation and GAC adsorption column was carried out [86]. With an initial concentration of 205 mg/L, about 40% COD was removed when 6 mg of ozone/mg of dissolved organic carbon (DOC) was applied. Biodegradation inside the activated carbon beds was suggested to be the major mechanism for the removal of organic compounds from ozonated leachate.

Stabilized leachate from the Badazos landfill (Spain) was treated by integrating ozone and GAC adsorption [87]. Approximate 90% COD removal was accomplished with an initial concentration of 4970 mg/L. Using adsorption after the ozonation step allowed the formation of smaller molecules during ozonation, which are more suitable for adsorption than the big molecules present in the leachate. In addition, adsorption is capable of removing the remaining organic compounds and metal species in the leachate [88].

A combination of UF and biologically activated carbon was investigated for the treatment of young leachate in the Niagara landfill (USA) [89]. This technology integrated the adsorption of organic matter with cross-flow filtration in one unit. About 97% COD was removed with an initial COD concentration of 3050 mg/L (Table 11). Due to membrane fouling, it was observed that the addition of PAC was able to mitigate permeate flux deterioration.

A combination of RO and evaporation was adopted for the treatment of leachate from a landfill in Italy [90]. No significant difference was found in terms of $\text{NH}_3\text{-N}$ removal between two membranes (AD: 97%; SC: 98%). However, AD membranes gave a slightly higher COD rejection rate (88%) than SC membranes (80%). The maximum rejection of $\text{NH}_3\text{-N}$ for both membranes was at pH 6.4, suggesting that pH strongly affected the rejection performance.

2.2.2. Combination between physico-chemical and biological treatments

The treatment of hazardous leachate from the landfills in Brescia, Veneto, Fontana-fredda and Fossalta (Italy) using a combination of activated sludge (AS) and wet oxidation was also evaluated [91]. As presented in Table 12, only moderate COD removal (38–50%) was achieved with the initial COD concentration ranging from 3100 to 19,400 mg/L. Therefore, in order to meet the discharge standards, further biological treatment was required.

A new combined treatment consisting of AS and RO was developed to treat young leachate from the Mechernich landfill (Germany) [92]. Almost complete removal of both COD and $\text{NH}_3\text{-N}$ was achieved with initial concentrations of 6440 and 1153 mg/L, respectively. These results suggest that a combination of physico-chemical and biological treatment

was able to optimize the removal of recalcitrant compounds and ammonia from landfill leachate.

A combination of an upflow anaerobic sludge blanket (UASB) reactors and RO was studied for the treatment of stabilized leachate from the Bavel landfill (The Netherlands) [93]. The UASB reactor was employed for pre-treatment of leachate. Since recalcitrant compounds with initial COD and $\text{NH}_3\text{-N}$ concentrations of 35,000 and 1600 mg/L, respectively, were able to be completely removed from the leachate, the effluent was discharged to surface water without further treatment.

A two-stage treatment of young leachate from the Komurcuoda landfill (Turkey), consisting of UASB and struvite precipitation with the stoichiometric ratio ($\text{Mg}:\text{NH}_4:\text{PO}_4 = 1:1:1$), was undertaken by Altinbaş et al. [94]. About 85% $\text{NH}_3\text{-N}$ removal was achieved with an initial concentration of 2240 mg/L at pH 9.2. This result was in agreement with those of previous studies carried out by Yangin et al. [95] and Kabdaslı et al. [96]. At an initial $\text{NH}_3\text{-N}$ concentration of 2130 mg/L, 86% removal was achieved at pH 9.3. At the same conditions, a combination of struvite precipitation and UASB was able to remove 83% COD with an initial concentration of 8900 mg/L [95]. The overall treatment cost was US\$ 0.9 m^{-3} .

The application of GAC-nitrification for the treatment of stabilized leachate containing high concentrations of ammonia was investigated [97]. Approximately 93% $\text{NH}_3\text{-N}$ removal was accomplished with an initial concentration of 830 mg/L. However, at an initial concentration of 2450 mg/L, only 55% COD removal was achieved. These results demonstrate that a combination of physico-chemical and nitrification was not effective enough in removing non-biodegradable organic compounds from the leachate.

A comparative study of the treatment of stabilized leachate from the Penzberg landfill (Germany) by using a combination of an aerobic treatment and GAC adsorption was evaluated [98]. The biological reactor in combination with GAC adsorption was found to be able to ensure the discharge meet the standards set in local environmental legislation. The removal of COD and $\text{NH}_3\text{-N}$ with initial concentrations of 1980 and 130 mg/L, respectively, was found to be 65 and 97%.

A combination of aerobic pre-treatment, GAC adsorption and coagulation was studied for the treatment of stabilized leachate from the Minden-Heisterholz landfill (Germany) [99]. About 92% COD removal was achieved with an initial concentration of 1400 mg/L. The cost of leachate treatment, mainly due to chemical consumption, was found to be US\$ 2.3 m^{-3} .

As a whole, a combination of two treatments proves to be more efficient and effective than individual treatment. This could be due to the fact that a two-step treatment has the ability to synergize the advantages of individual treatments, while overcoming their respective limitations. A combined treatment is indeed capable of improving the effluent quality and minimizing the residue generated at a lower treatment cost than an individual treatment.

Table 11
Combined physico-chemical technologies for treatment of landfill leachate

Location of landfill	Type of hybrid treatment	Precipitant/adsorbent/ membrane	Dose (g/L)	Initial concentration in leachate (mg/L)			BOD/COD	COD/TOC	pH	Removal efficiency (%)		References
				COD	NH ₃ -N	BOD				COD	NH ₃ -N	
				Metropolitan (South Korea)	Coagulation + Fenton oxidation	FeCl ₃				0.8–1.0	417	
Badajoz (Spain)	Coagulation – flocculation + Fenton oxidation	Fe(II)/H ₂ O ₂	1.0									
		FeCl ₃	0.8	7400	NA	444	0.06	NA	8.5	90	NA	[80]
Wuhan (China)	Coagulation + photo- oxidation	Fe(III)/H ₂ O ₂										
Bordo Poniente (Mexico)	Coagulation + ozonation	FeCl ₃ , UV–vis	0.5	5800	NA	430	0.07	NA	7.6	64	NA	[81]
		Fe ₂ (SO ₄) ₃	2.4	5000	NA	50	0.02	NA	4–5	78	NA	[82]
Gramacho (Brazil)	Coagulation – flocculation + ozonation + ammonia stripping	O ₃	1.7 × 10 ⁻³									
		Al ₂ (SO ₄) ₃	0.7	3460	800	150	0.04	0.24	8.5	48	100	[83]
Badajoz (Spain)	Ozonation + adsorption	O ₃	3 × 10 ⁻³									
		O ₃ GAC	1.5 × 10 ⁻³ 5	4970	700	850	0.17	NA	8–9	90	NA	[87]
Germany	NF + adsorption	PAC	NA	1450	NA	NA	NA	NA	7.3	97	NA	[22]
Saint-Nazaire (France)	NF + coagulation	FeCl ₃ /MPT-31	1–1.5	2150	790	215	0.10	NA	7.5	80	21	[84]
Berg (Germany)	NF + adsorption + ozonation	Desal 5K GAC O ₃	– NA NA	4000	NA	NA	NA	NA	6.5	99	NA	[85]
Niagara (USA)	UF + adsorption	GAC	NA	3050	NA	1678	0.55	3.6	7.0	97	NA	[89]
Italy	RO + evaporation	AD SC	–	19900	30	4000	0.20	3.8	6.4	88 80	97 98	[90]

NA: not available.

Table 12
Combination of physico-chemical and biological treatments for degradation of recalcitrant contaminants

Location of landfill	Type of hybrid treatment	Precipitant/ adsorbent	Dose (g/L)	Initial concentration in leachate (mg/L)		BOD/COD	pH	Removal efficiency (%)		Treatment cost (US\$/m ³)	References
				COD	NH ₃ -N			COD	NH ₃ -N		
Brescia (Italy)	Wet oxidation + activated sludge	NA	NA	19400	NA	0.69	7.1	50	NA	NA	[91]
Veneto (Italy)	Wet oxidation + activated sludge			17350		0.75	7.3	26			
Fontanafredda	Wet oxidation + activated sludge			3100		0.35	7.3	27			
Fossalta (Italy)	Wet oxidation + activated sludge			4140		0.41	7.8	38			
Mechernich (German)	RO + activated sludge	-	-	6440	1153	0.70	NA	99	99	NA	[92]
Bavel (The Netherlands)	RO + UASB	-	-	35000	1600	-	7.4	99	99	NA	[93]
Komurcuoda (Turkey)	Struvite + UASB	Struvite	17.5	4024	2240	NA	9.2	NA	85	NA	[94]
Kemerburgaz (Turkey)	Struvite + UASB	Struvite	NA	8900	2130	NA	9.3	83	86	0.9	[95]
Istanbul (Turkey)	Struvite + ammonium stripping	Struvite	NA	4560	2170	NA	8.1	80	90	NA	[96]
NA	GAC + nitrification	GAC	2.7	2450	830	0.08	6.5–7.0	55	93	NA	[97]
Penzberg (Germany)	GAC + membrane bioreactors	GAC	NA	1980	130	0.06	NA	65	97	NA	[98]
Minden-Heisterholz (Germany)	GAC + coagulation + aerobic	GAC	0.8–1.2	1400	493	0.24	7.1	92	NA	2.3	[99]
		Ca(OH) ₂	0.6–1.2								

NA: not available.

3. Comparison of outstanding performances of physico-chemical for treatment of stabilized leachate

To evaluate the performances of all the physico-chemical treatments described above, a comparative study is presented in terms of pH, dose required (g/L) and initial concentrations range of COD, NH₃-N and heavy metals (mg/L) in leachate. Although it has a relative meaning due to different testing conditions (pH, temperature, strength of wastewater, seasonal climate and hydrology site), this comparison is useful to evaluate the overall treatment performance of each technique for helping the decision-making process.

Table 13 summarizes the best performance of individual treatments for the removal of COD and NH₃-N from stabilized leachate. It is found that ammonium stripping and struvite precipitation, respectively, achieved 94 and 98% NH₃-N removal with initial NH₃-N concentrations of 3260 and 5618 mg/L. An outstanding level of COD removal (95–98%) was also demonstrated by NF, RO and PAC adsorption with the initial COD concentration ranging from 3840 to 17,000 mg/L.

Among the combined treatments reviewed above, it is observed that the combination of RO and UASB (COD: 35,000 mg/L; NH₃-N: 1600 mg/L) and/or RO and activated sludge (COD: 6440 mg/L; NH₃-N: 1153 mg/L) demonstrated outstanding treatment performances with almost complete removal for COD and NH₃-N (Table 14). A combination of ozonation and GAC adsorption also achieved 90% COD removal with an initial concentration of 4970 mg/L.

As a whole, physico-chemical treatments have many advantages for the treatment of landfill leachate such as plant simplicity, ease of operation, insensitivity to temperature changes and the convenience of the process employed. Their benefits, however, are outweighed by the drawbacks such as high operational costs due to chemicals used, energy consumption and handling costs for sludge disposal. Physico-chemical treatments are therefore suitable for pre-treatment of stabilized leachate to complement the biological degradation process [100].

4. Comparison of treatment cost among physico-chemical technologies

Estimating a reliable treatment cost for landfill leachate is complicated due to many cost components such as collection system, pumping equipment and treatment facility. In addition, changes in the quality and quantity of leachate due to seasonal variations also contribute to the inconsistency of its treatment cost. Therefore, information on the treatment cost of landfill leachate is rarely reported.

Basically, treatment costs of landfill leachate vary, depending on its strength and quantity, the process employed, the local condition of a landfill site, the amount and composition of impurities, as well as the extent of purification [101,102]. The overall treatment cost includes construction costs as well

Table 13
Comparison of the outstanding removal performance of COD and/or NH₃-N from individual physico-chemical treatment

Type of treatment	Type of precipitant/coagulant/adsorbent/membrane/electrode	Dose (g/L)	Initial concentration in leachate (mg/L)			Pressure (bar)	BOD/COD	pH	Removal efficiency/rejection rate (%)		References
			COD	NH ₃ -N	Heavy metals				COD	NH ₃ -N	
Adsorption	PAC	6	5690	2215	NA	–	NA	NA	95	NA	[29]
Ammonia stripping	Ca(OH) ₂	11	47800	3260	NA	–	0.60	11.0	–	94	[35]
Precipitation	Struvite	NA	7511	5618	NA	–	0.22	8.5–9.0	–	98	[36,37]
Reverse osmosis	SW30-2521	–	3840	NA	0.5	52	0.31	6.0	98	NA	[49]
Nanofiltration	NA	NA	17000	3350	NA	NA	0.03	6.4	96	NA	[54,55]

NA: not available.

Table 14
Highlight of the prominent removal performance among combined treatments

Type of hybrid treatment	Precipitant/adsorbent/membrane	Dose (g/L)	Pressure (bar)	Initial concentration in leachate (mg/L)			BOD/COD	COD/TOC	pH	Removal efficiency (%)		References
				COD	NH ₃ -N	BOD				COD	NH ₃ -N	
Coagulation–flocculation + Fenton oxidation	FeCl ₃	0.5–0.8	–	7400	NA	444	0.06	NA	8.5	90	NA	[80]
	Fe(II)/H ₂ O ₂	NA										
GAC adsorption + NF + ozonation	GAC Desal 5K O ₃	– NA NA	8.5	4000	NA	NA	NA	NA	6.5	99	NA	[85]
GAC adsorption + ozonation	GAC O ₃	5	–	4970	700	850	0.17	NA	8–9	90	NA	[87]
		1.5 × 10 ⁻³	–									
GAC + UF	GAC	NA		3050	NA	1678	0.55	3.6	7.0	97	NA	[89]
RO + evaporation	AD	–	60	19900	30	4000	0.20	3.8	6.4	88	97	[90]
	SC	–								80	98	
RO + activated sludge	–	–	NA	6440	1153	4508	0.70	NA	NA	99	99	[92]
RO + UASB	–	–	NA	35000	1600	–	–	–	7.4	99	99	[93]

NA: not available.

as operational and maintenance costs (O&M). The construction costs normally depend on the effluent quality required and the capacity of the installation, while O&M costs cover manpower, energy, chemicals and maintenance. The manpower cost varies significantly from one country to another. To obtain an accurate assessment of the operational cost for leachate treatment, a pilot-scale study needs to be carried out [103].

The treatment cost for $\text{NH}_3\text{-N}$ removal using struvite precipitation has been reported to be one-fifth more expensive than that of ammonium stripping [36]. Depending on the type of precipitation and chemicals employed, the treatment cost of struvite precipitation varies between US\$ 2 and 4 m^{-3} [94].

Subject to the size and complexity of the RO plant, the overall treatment cost of landfill leachate in Germany is in the range of US\$ 2– 30 m^{-3} [58,104]. A combination of biological and physico-chemical treatments that could meet German requirements costs about US\$ 41 m^{-3} [105]. This cost was able to be reduced if the RO systems were supplied with a storage lagoon that could level out seasonal variations during the production of leachate. In addition, the combination of storage and pre-treatment of leachate can overcome membrane fouling, thus reducing the O&M cost for closed landfills [106].

Compared to RO, the treatment cost using evaporation and thermal oxidation is more expensive, ranging from US\$ 30 to 70 m^{-3} [107]. The evaporation of leachate using plastic film as a heat exchanger has been reported to be US\$ 4 m^{-3} [108].

Depending on the liner, pump and land costs, it is estimated that treatment cost for constructed wetlands in the USA was about US\$ $50,000 \text{ ha}^{-1} \text{ year}^{-1}$ [109]. Liners and land acquisition, pumps and piping are included as the basic costs of excavation and vegetation establishment. For large systems, a wetland requires O&M cost of US\$ $990 \text{ ha}^{-1} \text{ year}^{-1}$ [110].

Due to the expenditures on chemicals and plants construction and/or maintenance, the application of physico-chemical

treatment for stabilized leachate is relatively costly. Since no individual treatment can guarantee particular treatment efficiency, a combination of physico-chemical and biological treatments should be adopted to improve treatment performance and to reduce overall treatment costs.

5. Concluding remarks

Over the past two decades (1983–2005), considerable research has been carried out on the treatment of stabilized leachate using various types of individual and/or combined physico-chemical technologies. Although many different treatments can be applied, it is evident from a survey of 118 publications that not one of the individual physico-chemical techniques reviewed is universally applicable or highly effective for the purpose.

Table 15 summarizes the treatability of stabilized landfill by various physico-chemical techniques. Among the treatment techniques presented in Table 15, adsorption, membrane filtration and chemical precipitation have been the most frequently applied and studied worldwide for the removal of recalcitrant organic compounds from stabilized leachate. Both activated carbon adsorption and nanofiltration are effective for over 95% COD removal with the initial concentrations ranging from 5690 to 17,000 mg/L. About 98% removal of $\text{NH}_3\text{-N}$ with an initial concentration ranging from 3260 to 5618 mg/L has been achieved using struvite precipitation. A combination of physico-chemical and biological treatments has been demonstrated to be effective for the removal of COD and $\text{NH}_3\text{-N}$ from landfill leachate. Almost complete removal of both COD and $\text{NH}_3\text{-N}$ has been accomplished by a combination of RO and UASB with an initial COD concentration of 35,000 mg/L and $\text{NH}_3\text{-N}$ concentration of 1600 mg/L and/or RO and activated sludge with an initial COD concentration of 6440 mg/L and $\text{NH}_3\text{-N}$ concentration of 1153 mg/L.

Although individual physico-chemical treatments are suitable for the removal of heavy metals and for the

Table 15
Summary of the applications of physico-chemical treatments for stabilized landfill leachate

No.	Type of treatment	Target of removal	Remarks	References
1	Coagulation–flocculation	Heavy metals and suspended solids	High sludge production and subsequent disposal may be a problem	[111]
2	Chemical precipitation	Heavy metals and $\text{NH}_3\text{-N}$	Requires further disposal due to sludge generation	[112]
3	Ammonium stripping	Ammoniacal–nitrogen	Requires other equipments for air pollution control	[76]
4	Microfiltration	Suspended solids	Used after metal precipitation	[113]
5	Ultrafiltration	High molecular weight compounds	Costly and limited applicability due to membrane fouling	[114,115]
6	Nanofiltration	Sulphate salts and hardness ions, like Ca(II) and Mg(II)	Costly and requires lower pressure than reverse osmosis	[116]
7	Reverse osmosis	Organic and inorganic compounds	Costly and extensive pre-treatment is required prior to RO	[117]
8	Activated carbon adsorption	Organic compounds	Carbon fouling can be a problem and GAC adsorption is costly	[118]
9	Ion exchange	Dissolved compounds, cations/anions	Used as a polishing step after biological treatments and treatment cost is high	[60]

hydrolyzation of some organic compounds, a combination of two physico-chemical treatments or physico-chemical and biological treatments is required for optimum treatment of stabilized leachate. Overall, it is found that a combination of two physico-chemical treatments can maximize the removal of recalcitrant organic compounds from stabilized leachate, as reflected by a significant decrease of the COD values after treatment, while a combination of physico-chemical and biological treatments is required to achieve effective removal of $\text{NH}_3\text{-N}$ and COD with a substantial amount of biodegradable organic matter. In most cases, physico-chemical treatments are suitable for pre-treatment of stabilized leachate to complement the biological degradation process.

It is important to note that the selection of the most suitable treatment technology for stabilized landfill leachate depends on the characteristics of the wastewater, the legal requirements of the residual concentrations of $\text{NH}_3\text{-N}$, COD and heavy metals in discharge, the overall treatment performance compared to other technologies, age of a landfill, plant flexibility and reliability as well as environmental impact. Due to seasonal weather variations, it is also necessary to consider temporal fluctuations in the quantity and composition of leachate. Finally, economic parameters such as investment and operational costs (energy consumption, residual deposition and maintenance) also play major roles in this decision-making process. All the factors mentioned above should be considered to select the most effective and inexpensive treatment technology in order to protect the environment.

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