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Review

Biological Treatment, Advanced Oxidation and Membrane Separation for Landfill Leachate Treatment: A Review

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Abstract: Landfill leachate, especially when produced in old municipal solid waste landfills, is a challenging type of wastewater which contains a variety of toxic substances. The existence of pollutants in the landfill leachate is primarily due to inadequate solid waste separation at the source. Pretreatment on site is a must for the landfill leachate to be safely released into the environment. One-step treatment is insufficient since landfill leachate has a complex composition that spatially and temporally varies. Often, the landfill leachate and municipal wastewater are treated together. Biological treatment is a routine technique which is applied to landfill leachate less than five years old. The concentration of easily biodegradable organic matter in the young landfill leachate declines with time and, as a result, the application of physical and chemical treatment processes is required. The goal of the current work is to investigate the usefulness and capability of the most efficient and widely available technique/s for landfill leachate treatment, to identify the main challenges and strengths of each technology and seek the optimum solution.

Keywords: landfill leachate; AOPs; photochemical treatment; activated sludge; membrane separation

1. Introduction

Despite progress in municipal solid waste (MSW) management and several alternative treatment methods, landfilling remains the most widely used disposal method, according to Schiopu and Gavrilescu [1]. Landfills do have some advantages in terms of investment and operational costs over other methods such incineration, especially if biogas is utilized for the production of energy. However, the management of leachate is a major environmental issue [2]. Leachate is a dark liquid generated by the anaerobic and aerobic decomposition of organic fractions in the waste; it contains a variety of organic pollutants, nutrients, inorganic salts and heavy metals [1]. The composition of leachate depends largely on its age as shown in Table 1. Leachates are divided into 3 groups depending on the age of the landfill: young, medium age and old. The age of young leachate is less than 1 year, while that of old leachate age is more than 5 years. Medium-aged leachate is between 5 and 10 years old [3].

Table 1. Composition of leachates [4,5].

Leachate Age	Age (Year)	pH	NH_4–N (mg/L)	TN (mg/L)	COD (mg/L)	BOD_5/ COD	TOC/ COD
Young	<1	7.2 ± 1.0	2162 ± 1385	1665 ± 1612	24,805 ± 22,982	0.46 ± 0.21	<0.3
Medium	1–5	7.7 ± 0.7	1070 ± 285	1421 ± 416	5239 ± 2618	0.23 ± 0.09	0.3–0.5
Medium	>5	8.2 ± 0.3	1616 ± 1557	1939 ± 1715	2652 ± 1786	0.121 ± 0.07	>0.5

The composition and the wide variation by age and landfill site renders leachates a particularly challenging wastewater requiring a combination of treatment technologies tailored for a specific landfill [6]. According to Wiszniowski et al., leachate treatment methods can be classified into two groups: (1) physico-chemical and (2) biological treatment techniques [7]. Physico-chemical treatment processes are mostly applied to increase treatment efficiency and as additional treatment processes for the elimination of bio-refractory substances that can limit the effect of biological oxidation treatment methods. Physical leachate treatment processes include adsorption, air-stripping, membrane filtration, and sedimentation [8], while coagulation-flocculation, chemical-electrochemical oxidation and precipitation methods are mainly used as chemical treatment techniques [9].

Biological processes are the most commonly used treatment method for the reduction of organic load in landfill leachates [10]. Although it is highly effective and simple in operation, it suffers from the presence of refractory non-biodegradable organics, heavy metals and elevated ammonium content of the landfill leachate. Biological treatment can successfully eliminate ammonium content present in leachate and according to Lo, aerobic biological treatment achieved more than 99% N–NH_4 removal efficiency within 20–40 days [11]. The application of anaerobic and aerobic reactors as a sequential system has proven to be an efficient leachate treatment method. For example, Kettunen et al. demonstrated 85% and 80% removal of COD and ammonium, respectively [12]. Advanced Oxidation Processes (AOPs) are known as useful post-treatment methods for the removal of organic contaminants in landfill leachates [13]. AOP treatment is based on photocatalytic activation in the presence of a catalyst, where organic pollutants are oxidized and, after a sequence of chain reactions, are degraded into less hazardous products [13]. The modified Fenton process, i.e., in the presence of UV and based on Fe(II) + H_2O_2 reaction, strongly promoted organic pollutant degradation rate and showed COD removal of up to 70% in landfill leachate [14]. Conventional membrane separation processes—reverse osmosis (RO), nano-, ultra- and micro-filtration (NF, UF and MF)—are widely used in developed countries for landfill leachate treatment. Generally, the membranes are applied after a physical, chemical or physico-chemical pretreatment step [3]. Another widely used sequence is membrane bioreactor (MBR), which is followed by the NF process [15,16]. Recent developments in the forward osmosis (FO) process have also initiated applications of FO membranes in landfill leachate treatment [17]. The main idea of the combination of different treatment techniques is the improvement of landfill leachate treatment efficiency. For example, the combined application of aerobic and anaerobic biological treatment followed by the reverse osmosis method was investigated in the work of Park et al., and organic pollutant treatment efficiency with improved biodegradability of BOD content reached 98% [18]. Marttinen et al. investigated the impact of physico-chemical methods on leachate treatment by nanofiltration at the first stage, followed by ammonia stripping and ozonation; the results indicated that the COD removal efficiency improved from 66% at first stage up to 90% after the combined treatment [19]. Another work by Steensen studied biological treatment of landfill leachate sequenced using the chemical oxidation method integrated with ozone/fixed bed catalyst and UV/H_2O_2 techniques as a post-treatment phase for the elimination of non-biodegradable organic content [20]. Inglezakis et al. studied the treatment of landfill leachate by applying a combined physical (stripping and adsorption), biological and photochemical process treatment, reaching 85%–100% total carbon removal [6].

In the present paper the application of the most promising and widely used technologies in landfill leachate treatment, namely biological treatment, advanced oxidation, and membrane separation, are presented and evaluated focusing on efficiency, advantages, drawbacks and recommendations.

2. Biological Treatment Processes

The choice of the optimal landfill leachate treatment method is challenging and depends on several variables such as the leachate quantity, quality, age, adjustability of plant and operating conditions [21]. There are two major challenges in the treatment of landfill leachates by biological treatment; the high concentration of toxic, non-biodegradable compounds resulting in low BOD/COD ratio, and high ammonium content, which can inhibit the activity of the activated sludge. Pre-treatment is frequently applied to facilitate the biological treatment [6]. Biological processes are considered a cost effective way to remove total nitrogen [22]. In particular, ammonium is removed in aerobic bioreactors by conversion to nitrates and nitrogen is removed using endogenous denitrification [23]:

$$NO_3^- + cBOD \rightarrow NO_2^- + CO_2 + H_2O$$

$$NO_2^- + cBOD \rightarrow N_2 + CO_2 + H_2O$$

or an anaerobic NH_4^+ oxidation (ANAMMOX) process [23]:

$$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + 2H^+ + H_2O$$

$$NO_2^- + 0.5O_2 \rightarrow NO_3^-$$

ANAMMOX is considered as a low-cost alternative to conventional denitrification systems and NH_4^+ is converted to N_2 with NO_2^- as an electron acceptor [7].

The bioreactors can employ aerobic, anoxic and anaerobic steps. Briefly, the aerobic step requires oxygen and produces large amounts of waste sludge, which increases the operational costs, is highly effective in the removal of biodegradable organics and oxidizes ammonium to nitrates; the anoxic step does not require oxygen, removes biodegradable organics and reduces nitrates to nitrogen. It is anaerobic, can produce energy and remove phosphorous, but is less efficient in the removal of biodegradable organics. Alvarez-Vazquez et al. published a review on landfill leachate, including data of full-scale units [4]. Data from 166 plants showed that 4% use activated carbon and ion exchange, another 4% chemical oxidation, 4% air stripping, 5% filtration, 11% flocculation/coagulation, and 70% biological treatment. Of the 120 plants using biological treatment, 21% use aerobic lagoons, 18% up-flow anaerobic sludge blanket reactor (UASB), 17% conventional activated sludge process (CAS), 15% sequential batch reactors (SBRs), 8% membrane bioreactors (MBRs) and 21% several other bioreactor types.

A review on 188 papers reports that CAS, SBRs and UASB reactors can remove over 90% of COD with a concentration ranging from 3500–2000 ppm and 80% of ammonium removal with a concentration ranging from 100–1000 ppm [24]. Recently Wang et al. published a review on the use of activated sludge reactors for the treatment of landfill leachates [22]. The data show that aerobic SBRs can achieve 70–85% COD removal and MBRs 70–100% for influent COD between 1348–5445 ppm. Moreover, SBRs are the preferred process for landfill leachate ammonium removal, reaching levels of 95–99%, while MRBs can achieve removals of 80–95%. The ammonium removal is enhanced by adding an additional carbon source to the reactors. The UASB process demonstrates high retention efficiency and has been frequently applied in the treatment of landfill leachates operating with a hydraulic retention time (HRT) of 1.25–2 days and very high influent COD up to almost 50,000 ppm, with a removal rate of 76–80%. Anaerobic MBRs have shown good removal rates of 62–95% but with considerably lower influent COD up to 13,000 ppm.

A literature review shows that the most used type of activated sludge bioreactor for landfill leachate treatment is the SBR due their simple structure and large capacity (Figure 1) [22].

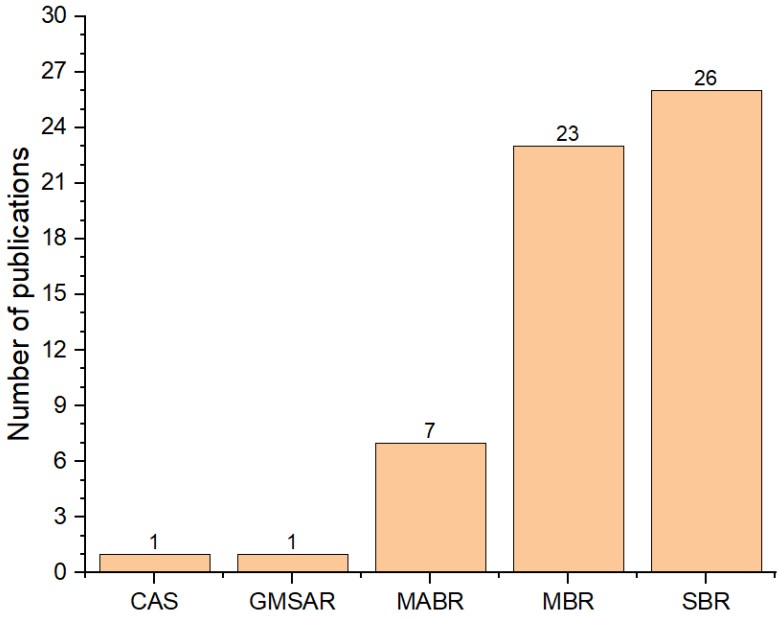

Figure 1. The number of publications focusing on a particular type of reactor.

The same chamber is used for wastewater and landfill leachate treatment in SBRs. The treatment is comprised of four steps; feeding, aerobic/anoxic/anaerobic reaction, settling and discharge [25]. One of the advantages of SBRs in the treatment of landfill leachate is the ability to carry out nitrification and denitrification in a single tank and its adaptability when influent composition fluctuates [26,27]. However, the anoxic denitrification step demonstrates low TN retention, which is a result of carbon consumption in the aeration zone [25]. Furthermore, the number of scientific works focusing on full-scale SBRs (treating landfill leachate) optimization is not high [28]. MBRs comprise a CAS process with the clarifier replaced by a membrane, which mostly removes suspended solids and retains higher activated sludge concentrations in the reactor. MBRs are attractive alternative because of their high sludge concentration leading to high COD removal [22]. In addition, they generally produce better quality effluents than CAS systems, thus requiring reduced post-treatment, while they also have a much reduced footprint compared CAS systems [4]. The major drawback of MBRs is membrane fouling, which creates operational problems. The HRTs of full scale units vary between 2 and 45 days and COD removal is between 60–100% [4]. Full-scale anaerobic and aerobic MBRs have similar performance when operating at similar HRT values (5–20 days) with average COD retentions around 85%. However, anaerobic MRBs do not remove ammonia, and thus post-treatment is necessary by stripping or, more usually, nitrification [4].

In MABRs the biofilm grows on a membrane which is permeable for a gas, and acts as O_2 source and support for the biofilm [25]. The main advantage of the MABR over other bioreactors is the much more efficient oxygen supply, and thus, reduced operational costs. Landfill leachate treatment by MABR may reach up to 99% of nitrification with a 35 g O_2/m^2-day transfer rate [25]. Syron studied the performance of a pilot-scale MABR for the treatment of landfill leachate [29]. The results showed that with an average HRT of about 5 days, the reactor achieved 80–99% nitrification and 30–90% COD removal. The authors conclude that optimization of MABR may facilitate the development of efficient and low-energy leachate treatment techniques. In UASB, the leachate flows upwards (the entrance is located at the bottom of the reactor) through an anaerobic sludge blanket, which consists of bacteria that have formed into granules and are therefore retained in the reactor. UASB

leachate treatment improves biomass retention and promotes the elimination of soluble COD by up to 90%; nevertheless widespread application of this method is limited due to the low ammonium removal and reactivity to toxic materials [25]. Anaerobic processes produce biogas, comprised of methane and carbon dioxide, which may be used to generate electricity in large plants or for heating the reactor.

As mentioned above, a combination of biological, chemical and physical methods significantly improves lachate treatment efficiency. Biological treatment followed by reverse osmosis was proven to be powerful, achieving the elimination of more than 95% of COD and up to 77.3% $N–NH_4^+$ from landfill leachate [30,31]. Application of reverse osmosis after biological treatment demonstrated up to 99.5% COD and 99.8% ammonium retention [21]. Inglezakis et al. studied the impact of the photochemical post-treatment method (after physical and biological processes); the results demonstrated improved TOC removal (from 37% to 59%); the method was proven to almost entirely remove TN and eliminate up to 85% of TC, however, did not show significant influence on ammonia elimination [6]. Innovative landfill leachate treatment with integration of the first stage (biological oxidation, coagulation, photo-Fenton method), and second stage (biological oxidation) was investigated by Silva et al. and resulted in 62–99% ammonium nitrogen and 88% TSS removal [32]. The leachate was collected from an aerated lagoon at a leachate treatment plant with a COD concentration of around 4000 and $N–NH_4^+$ between 200–2170 ppm [32].

Finally, it should be noted that Robinson et al. rightfully argue that there is a large gap between academic research and the reality of leachate treatment plant design and operation, and thus data from full scale plants are of particular importance [33]. Table 2 presents recent studies on MABR.

Table 2. Review of biological reactors used for landfill leachate treatment.

Reactor Type	Country	Experimental Conditions	COD Influent mg/L	$N–NH_4$ Influent, mg/L	BOD/COD Ratio	% COD Removal	% $N–NH_4$ Removal	% TN Removal [1]	Year	Ref
CAS	Brazil	pH = 6; HRT = 16 h; Q = 44 m^3/h	12–42 kg/h	1.2–4.2 (as TKN)	-	80–96	80–96 (as TKN)	61–97	2017	[34]
GMSAR	Malaysia	Anaerobic; T = 37 °C; pH = 7	270–239	717	-	71	-	-	2020	[35]
MABR	Ireland	Aerobic; T = 21–27 °C; pH = 7.5–8; HRT = 4–7.5 d; polydimethyl siloxane (PDMS) membrane	1000–3000	500–750	<0.2	30–90	80–99	-	2015	[29]
MABR	China	Aerobic; T = 25 ± 0.5 °C; pH = 7.5 ± 0.05; HRT = 19 h; PDMS membrane	362.10 ± 17.16	26.82 ± 0.72	-	94.34	-	78.21	2019	[36]
MABR	China	T = 23 ± 2 °C; pH = 8.7–9.5; HRT = 14–32 h; polyvinylidene fluoride (PVDF) membrane	367–2158	21.4–74.3	-	88.4–97.1	38.3 ± 3.7	90<	2019	[37]
MABR	China	Anaerobic-anoxic-aerobic; T = 25 ± 1 °C; pH = 8.7–9.5; HRT = 24 h; PDMS membrane	30.03–181.81 ± 2.50	-	-	82.40	-	88.52	2020	[38]
MABR	China	Aerobic; T = 25.0 ± 2.0 °C; pH = 7.5–8.5; PVDF membrane	5298.2	543	-	82.0	80.3	76.7	2021	[39]
MABR	Malaysia	Anaerobic; pH = 8–8.6; HRT = 1–4 d; PVDF membrane	5678	-	-	70–76	-	-	2021	[40]
MABR	Italy	Aerobic; T = 19 ± 4.3 °C; pH = 7.2 ± 0.09; ZeeLung membrane	100–990	32.1–37.1 ± 8.7	-	89	93	-	2022	[41]
MBR	China	Aerobic-anoxic/oxic; HRT = 168 h; flat sheet membrane	4000–20,000	1450–2100	-	81	99	75	2017	[42]
MBR	Japan	Aerobic; pH = 7.5–8.0; HRT = 12 h; PVDF membrane; membrane permeate flux (J) = 0.4286 m^3/m^2d^{-1}	9273	119	0.71	96	90	85 (as TKN)	2017	[43]
MBR	Algeria	Aerobic; T = 25 °C; pH = 7.5; HRT = 12 h; UF Carbosep M5 membrane	10,500	311.51	0.52	93–95	-	-	2018	[44]

Table 2. *Cont.*

Reactor Type	Country	Experimental Conditions	COD Influent mg/L	N–NH_4 Influent, mg/L	BOD/COD Ratio	% COD Removal	% N–NH_4 Removal	% TN Removal [1]	Year	Ref
MBR	Italy	T = 29–37 °C; pH = 7.5; gel GPT-BW 30 membrane; J = 7.1–32.5 LMH	1368 ± 422	35 ± 46	-	95	57.4–77.3	-	2018	[30]
MBR	Brazil	Aerobic; T = 25–30 °C; pH = 3.5; HRT = 48 h; poly(etherimide) (PEI) membrane; J = 20–32 LMH	1552–6899	844–1815 (NH_3–N)	0.03–0.1	68	53 ± 19 (NH_3–N)	-	2019	[45]
MBR	Canada	Aerated submerged MBR; T = 20 ± 1 °C; HRT = 48.8–51.3 h; Zee-Weed, ZW-1 membrane; J = 7.4 LMH	1038–1096	219–1040 ± 33	-	-	23.8–99.7	-	2019	[46]
MBR	China	Anaerobic-anoxic-oxic MBR (AAO-MBR); T = 23 ± 2 °C; HRT = 37.1–95.3 h; PVDF membrane	385–442	17–58	-	89	-	78	2019	[47]
MBR	China	Anaerobic fluidized bed MBR AMFBR); T = 21.5 −31.5 °C; HRT = 10 d; hollow fiber (HF) membrane	385	42	-	89.5–90.9 ± 2.8	91.9–92.4 ± 2.4	-	2019	[48]
MBR	Thailand	Two-stage activated sludge (AS) system; HRT = 12–24 h	<200	<40	-	97–98	91–94 (NH_3–N)	88	2019	[49]
MBR	Brazil	MBR-NF FMF-NF; T = 25 °C; pH = 7–8; HRT = 12–24 h; PEI membrane	3238–3374	431–448	-	22–27	27	-	2020	[50]
MBR	China	pH = 3–10	5000 ± 1000	1500 ± 500	<0.1	25.21–47.37	-	-	2020	[51]
MBR	China	MBR-Ozonation	477.9	684 (NH_3)	0.07	51.13	-	-	2020	[52]
MBR	Thailand	Anaerobic-aerobic; pH = 6.9–7.5; HRT = 24 h; HF membrane	3200–5800 ± 610	25 (NH_3–N)	-	95–98	80–84 (NH_3–N)	83–88	2020	[53]
MBR	Thailand	Anaerobic-aerobic; pH = 6.9–9.2; HRT = 1–2.5 d; PVDF membrane	2873 – 28,889	51–130 (NH_3–N)	0.5–0.76	70–96	97–100 (NH_3–N)	58–97 (as TKN)	2020	[54]
MBR	Thailand	Anaerobic-aerobic; pH = 4.35–6.46; HRT = 4 d; polyethylene (PE)	19,296 – 26,012	410.4–648.5 (NH_3–N)	-	99.3	99.2	94.3 (as TKN)	2020	[55]

Table 2. *Cont.*

Reactor Type	Country	Experimental Conditions	COD Influent mg/L	N–NH_4 Influent, mg/L	BOD/COD Ratio	% COD Removal	% N–NH_4 Removal	% TN Removal [1]	Year	Ref
MBR	Turkey	Anaerobic MBR; T = 35 ± 1 °C; HRT = 12–48 d; polypropylene (PP) membrane; Net flux = 0.5–5.5 LMH	7014 ± 250	1000 ± 200	0.1–0.3	62–98	90	87	2020	[56]
MBR	Turkey	Electro-MBR; T = 25 ± 4 °C; pH = 8–9.1; HRT = 5 d; HF membrane; Net flux = 1 LMH	65,000	1.815	0.55	3–15	78–86 (NH_3–N)	-	2020	[57]
MBR	Turkey	Sequencing batch MBR; T = 25 ± 4 °C; pH = 7.5–8.5; HRT = 15 d; PVDF membrane; J = 72 LMH	18,656 ± 12,098	3090 ± 84 (NH_3)	-	98	99 (NH_3)	-	2020	[58]
MBR	Turkey	MBR-UF-Electrooxidation; pH = 3.5–10	15,475	2342	-	≥97	≥99	-	2020	[59]
MBR	Brazil	Electro-MBR; pH = 7; polyethersulfone (PES)/PES-graphene oxide (GO) membrane; J = 7.2 LMH	863 ± 183	-	-	66–68	-	-	2021	[60]
MBR	China	Anaerobic baffed MBR (ABMBR); HRT = 6 d; polymethyl methacrylate (PMMA) membrane	12,700	1583.16 ± 101.03	-	80.38–91.2	21.56–99.4 (NH_3–N)	-	2021	[61]
MBR	India	Electro-Fenton MBR (Batch); pH = 7; HRT = 4 d; PP membrane	23,200 ± 2054	2196 ± 106 (NH_3–N)	0.15–0.25	90.62	84.66 (NH_3–N)	-	2021	[62]
MBR	Thailand	Anaerobic MBR; T = 29.5 ± 1.3 °C; pH = 6.9 ± 0.8; HRT = 3 d; chlorinated polyethylene (PE) membrane; J = 0.18 m^3/m^2day^{-1}	342–5253	140–685 (NH_3–N)	-	47.4–79.4 ± 13.7	63.1–99.9 (NH_3–N)	74.6–99.8 (as TKN)	2021	[63]
SBR	Canada	Activated sludge (AS) SBR/granular sludge (GS) SBR; T = 20 ± 2 °C; pH ≥ 6.5	448–654	225 ± 21	-	70	73 ± 8	55	2017	[64]

Table 2. *Cont.*

Reactor Type	Country	Experimental Conditions	COD Influent mg/L	N–NH_4 Influent, mg/L	BOD/COD Ratio	% COD Removal	% N–NH_4 Removal	% TN Removal [1]	Year	Ref
SBR	Canada	ASBR/GSBR; T = 20 ± 2 °C; pH ≥ 6.5	810 ± 83	128 ± 5	1.5	67–87	99	56 ± 12	2017	[65]
SBR	China	SBR with aeration phase; T = 25 ± 1 °C; pH = 7.5–8.5	3820	1010	0.32	>85	100	>95	2017	[66]
SBR	Estonia	Bio-Chemical SBR; pH = 7.5 ± 0.3; HRT = 23–28 d	11,800 ± 4000	1170 ± 330	0.57 ± 0.17	100	86	-	2017	[67]
SBR	Poland	T = 37 °C; HRT = 2 d	4125	775	0.15	5–10	46–78	-	2017	[68]
SBR	Vietnam	Partial nitritation (PN)-SBR; T = 28–32 °C; HRT = 3.85 d	2770 ± 85	3096 ± 542	2 ± 1	11	-	-	2017	[69]
SBR	Canada	Aerobic SBR	1050–1400	890–1100	<0.1	25	99.7	-	2018	[70]
SBR	China	PN-SBR/Integrated fermentation and denitritation (IFD-SBR); T = 21–26 °C; HRT = 44.7–65 h/20.5–100 h	2109 ± 200	1736 ± 40	-	19.7	25	95	2018	[71]
SBR	Greece	Twin SBR; HRT = 11.67 d	1295–1819	562–1627	0.25	1.5 ± 7.8	98–99	71 ± 12	2018	[28]
SBR	Iran	Biological ASBR; T = 32–34 °C; pH = 7–8.33; HRT = 4.1 d	10,500	461	0.68	81.2	-	-	2018	[72]
SBR	Kazakhstan	Aerobic, anaerobic, combined aerobic/anaerobic SBR; pH = 7–12; HRT = 1.5 d	2758–2998 (as TOC)	1918–2398 (100% removal by stripping)	-	24.8–29.6 (as TOC)	-	-	2018	[6]
SBR	Poland	Aerobic SBR; T = 35 °C; HRT = 3d	7758	980	0.12	90	92	-	2018	[73]
SBR	Portugal	Aerobic SBR; T = 20–30 °C; HRT = 4.7–7 d	400	8	0.05	54	-	86	2019	[74]
SBR	Slovenia	T = 14–23 °C; pH = 6.5–7.5	671	211	0.3	-	95	-	2018	[16]
SBR	China	pH = 7.2 ± 0.1; HRT = 18 h	4500–12,490	1734–2350	-	50	48.4–23.4	25	2019	[75]
SBR	China	Biological SBR	2080	2875	0–0.8	66.4–81.5	-	-	2019	[76]
SBR	India	Biological SBR; pH = 7.5; HRT = 15 h	600–2100	-	-	70.5–73.2	-	-	2019	[77]

Table 2. *Cont.*

Reactor Type	Country	Experimental Conditions	COD Influent mg/L	$N–NH_4$ Influent, mg/L	BOD/COD Ratio	% COD Removal	% $N–NH_4$ Removal	% TN Removal [1]	Year	Ref
SBR	Italy	PN-SBR	6617 ± 3444	1196 ± 616	0.20	>70	94	50	2019	[78]
SBR	Poland	T = 23 ± 2 °C	2510 ± 22	399 ± 21	-	3.5	-	-	2019	[79]
SBR	Poland	Aerobic SBR; T = 18–20 °C	3600–4500	750–990	0.11	93	98	-	2019	[80]
SBR	Poland	Anaerobic ammonium oxidation-SBR; T = 33 ± 1 °C; pH = 7.8 ± 0.2; HRT = 1d	1670–2070	320–470	0.02–0.80	42 ± 2	91–99.9	78.9–89.9	2019	[81]
SBR	Portugal	Multistage SBR; T = 24–33.5 °C; pH = 4.2	3596–8302	858–2480	0.04–0.47	90 ± 1	-	71 ± 4	2019	[82]
SBR	United Kingdom	Aerobic SBR; T = 23–25 °C; pH = 7.2–7.8; HRT = 16 d	3520	142.5	-	58.6	99.8	-	2019	[83]
SBR	China	PN-SBR and Integrated fermentation and denitrification (IFD)-SBR; T = 25–28.2 °C;	2039 ± 217	1760 ± 126	-	100	83	98.3	2020	[84]
SBR	Malaysia	T = 20–25 °C; pH = 7 ± 1; HRT = 1.7 d	1673–2343.3	1789–2127.5	0.1	49.9–84.5	53.2–93.1	-	2020	[85]
SBR	Poland	Aerobic-SBR + RO; polyamide (PA) membrane; J = $11.3 \cdot 10^{-6}$ L/$m^2 s^{-1}$	720–2180	400–910	0.06–0.82	18.8–20.0	98.9–99.0	46.7–86.4	2021	[86]

[1] TKN = Total Kjeldahl nitrogen.

3. Advanced Oxidation Processes

Advanced Oxidation Processes (AOPs) are highly efficient in the removal of resistant organic compounds in water [87–89]. Since AOPs are widely used in wastewater treatment, they are applicable in the treatment of landfill leachates as well [90]. AOPs proceed through the production of highly oxidative agents such as hydroxyl radicals .OH, which are able to oxidize a sufficient number of salts, mineral acids, and organic compounds, converting them ultimately into H_2O and CO_2. The mechanism of AOPs involves activation of hydroxyl radicals that can consequently degrade high concentrations of contaminants to less than 5 μg/L [91]. Nevertheless, application of AOPs for leachate treatment is limited due to potential formation of toxic by-products [91].

In AOPs, the most common generated oxidative agent is hydroxyl radical possessing a highly oxidative nature, which can be obtained by several methods. The hydroxyl radicals are mainly generated in two ways:

- Photochemical: UV/H_2O_2, UV/O_3, $UV/O_3/H_2O_2$, photo-Fenton/Fenton-like, photocatalytic oxidation [91];
- Non-photochemical: ozonation at elevated pH (>8.5), O_3/H_2O_2, O_3/catalyst, Fenton (H_2O_2/Fe^{2+}) [91].

The reaction rate of photochemical processes can be enhanced by integration of ozone, hydrogen peroxide, semiconductors, and metal salts [92]. According to the literature, water/wastewater that undergoes photochemical AOP treatment will benefit from [93]:

- Increased reaction rates compared to similar methods without any light source;
- Mild temperature, pressure and pH conditions;
- Possibility to select among a variety of oxidants;
- Additional benefits regarding the organoleptic characteristics of purified water.

The efficiency of pollutant removal by photochemical processes is influenced by different parameters, including the reactor configuration and physicochemical properties of the medium, which can significantly affect generation rate of free radicals [93,94].

Compared to conventional leachate treatment techniques, photochemical AOPs have high capital and treatment cost due to elevated turbidity of landfill leachate. Moreover, the cost of the treatment might be further increased due to the need for the repeated replacement of UV lamps, the intensity of which is significantly reduced by fouling (generated by ultraviolet-absorbing films). However, the costs of energy sources and reagents (O_3 and H_2O_2) are the main barriers for photochemical application of AOPs at an industrial scale [95].

Nowadays, the most commonly used chemical in AOPs is Fenton reagent, which is usually a mixture of Fe(II) salt (catalyst) and H_2O_2 (oxidant) that are used to break down Persistent Organic Pollutants (POPs) [96,97]. In Fenton processes, $OH^{\cdot}$ formation is the main part of the classic or free radical Fenton mechanism. The conventional Fenton reaction without involvement of organics includes the following [98]:

$$Fe^{2+} + H_2O_2 \rightarrow Fe^{3+} + OH^{\bullet} + OH^{-}$$

$$Fe^{3+} + H_2O_2 \rightarrow Fe^{2+} + HO_2^{\bullet} + H^{+}$$

$$OH^{\bullet} + H_2O_2 \rightarrow HO_2^{\bullet} + H_2O$$

$$OH^{\bullet} + Fe^{2+} \rightarrow Fe^{3+} + OH^{-}$$

$$Fe^{3+} + HO_2^{\bullet} \rightarrow Fe^{2+} + O_2H^{+}$$

$$Fe^{2+} + HO_2^{\bullet} + H^{+} \rightarrow Fe^{3+} + H_2O_2$$

$$2HO_2^{\bullet} \rightarrow H_2O_2 + O_2$$

When ferric ions (Fe^{3+}) are used to promote the disintegration of H_2O_2 into hydroxyl radicals, the reaction is called Fenton-like [99]. The reaction rate of Fenton is higher than

Fenton-like reactions [100]; this could be explained by the reduced reactivity of iron (III) towards hydrogen peroxide [101].

The Fenton reagent efficiency depends on concentration of H_2O_2, catalyst, hydrogen/hydroxide ions, and temperature; these are the main parameters defining the regeneration ability of iron and the rate of oxidation of organics [102]. According to Kuo, pH 3 is the most optimal for Fenton reactions; it is a result of an established stable system between H_2O_2 and Fe^{2+} in acidic environments [103]. At pH > 4, iron (II) ions become less stable and may be transformed into iron (III) ions, consequently forming ferric hydroxo complexes. Furthermore, in a basic environment, hydrogen peroxide becomes unstable and easily breaks down into H_2O and oxygen, subsequently losing its oxidizing potential [103,104]. Hence, pH tuning is of high importance for wastewater treatment by Fenton processes.

Another work focused on the investigation of the influence of hydrogen peroxide concentration and temperature on the dewaterability of sludge [105]. Poor dewaterability results in large amounts of sludge that have to be dewatered, which accounts for 35–50% of the total wastewater treatment cost [106]. Therefore, increasing the dewaterability of sludge can facilitate the dewatering process, and thus, considerably decrease the total operating cost. Neyens et al. studied the influence of several parameters on the dewaterability of sludge by means of specific resistance to filtration measurements [107]. The authors concluded that below 40 °C, sufficient sludge dewaterability was achieved by using H_2O_2 at high concentrations and prolonged reaction times, while at higher temperatures (particularly in the range of 80–90 °C), substantially smaller amounts of H_2O_2 were needed to achieve sufficient levels of sludge dewaterability, especially if the treatment pH ranged from 2.5–3.

Degradation of organic pollutants by Fenton oxidation processes can be decelerated in the presence of different ions such as sulfate, chloride, fluoride, bromide, and phosphate [108]. The deceleration is explained by scavenging of hydroxyl radicals, deposition of iron and less reactive dissolved iron (III) complex. Ammonia content in landfill leachate cannot be fully oxidized by OH. regardless of their high oxidation potential [98]. In addition, operation process issues such as foaming may lead to reduced removal performance. Foaming mainly occurs due to CO_2 released by carbonate compounds and organic foaming agents in an acidic environment [98].

Water and wastewater treatment with application of the Fenton processes allows several benefits, including: a lack of requirement for power input; utilization of safe, easy-to-apply, and relatively cheap chemical agents; a non-complex and adjustable system that can be used in plants that are in operation already [108].

Application of the Fenton process before the biological treatment of landfill leachate indicated that the AOP alone is capable of decolorizing a mature leachate by 90–95% [109]. Compared to mature and medium leachates, young leachates contain larger quantities of biodegradable organic substances [110]. With leachate aging, the concentration of non-biodegradable compounds increases [111]. The presence of humic acid (non-biodegradable compound) leads to elevated turbidity of the leachate [112]; molar mass of humic substances increases as leachate matures due to a higher degree of humification [113]. During the Fenton process, large organic substances break down into smaller fragments, resulting in improved leachate turbidity.

The Fenton process involves the following drawbacks [97]:

- A high concentration of chemicals is required to reduce the pH level of raw wastewater to 3 and to convert the treated solutions into inactive form before the discharge;
- Production of iron sludge (as a result of the conversion of Fe3+ to ferric-hydroxo complexes) which must be treated before disposal;
- Treatment cost is not cheap and risks exist due to logistics related to hydrogen peroxide;
- Absolute mineralization is not achievable because of production of iron(III)-carboxylic acid complexes that hydroxyl radicals are not able to disintegrate fully.

Several scenarios can be applied to reduce the disadvantages mentioned above. For instance, the concentration of hydrogen peroxide can be decreased by operating conditions

optimization. Production of iron sludge can be prevented/minimized by application of ion-exchange resins, iron-exchanged Nafion membranes and solid iron-containing catalysts [108]. It seems that it makes sense to include the sludge discharge in the cost of the treatment process [114]. Several works have reported that both chemical oxygen demand and flocs can be efficiently treated by chemical coagulation [114,115].

When UV light is used, Fenton processes are called photo-Fenton processes. The photo-Fenton processes need smaller amounts of catalyst, and they lead to a reduction in the amounts of precipitates and sludge. However, the implementation of the photo-Fenton processes requires additional power; therefore, minimization of energy should be considered [116]. In acidic environments, the primary form of the Fe^{3+} ion is $[Fe(OH)]^{2+}$. High amounts of Fe^{3+} reduce the efficiency of Fenton treatments [108]. This, however, can be avoided in the photo-Fenton process, since the reductive photolysis of $[Fe(OH)]^{2+}$ regenerates iron (II) ions that facilitates the reaction and generates more hydroxyl radicals [117].

The photo-Fenton processes have two main features:

(1) Due to the photolysis, the Fe^{3+} and Fe^{2+} are reduced to form more hydroxylic radicals [118]:

$$(Fe - OH)^{2+} + h\nu \rightarrow Fe^{2+} + OH^{\cdot}$$

(2) The ferric carboxylates are exposed to the photo-decarboxylation [119]:

$$Fe(III)(RHCO_2) \rightarrow Fe^{2+} + CO_2 + RH^{\cdot}$$

$$RH^{\cdot} + O_2 \rightarrow RHO_2^{\cdot} \rightarrow \text{products}$$

From the above equations, it is seen that the smaller amount of iron catalyst required leads to a decrease in the final sludge volume; in addition, the treatment of carboxylates can be handled more efficiently [118]. Specifically, in comparison with simple Fenton processes, the implementation of UV radiation in the photo-Fenton process required 32 times less Fe^{2+} to achieve the same level of removal of contaminants, while the volume of the sludge was reduced from 25% to 1% [119]. Photo-Fenton processes are considered a powerful method for the treatment of biologically pre-treated landfill leachates, as it has been found that 86% COD removal was achieved and more than 95% of the color was removed [120]. The application of photo-Fenton processes is also practical for degrading organic compounds, such as nitrobenzene and anisole [121], ethylene glycol [122], 4-chlorophenol [123] and herbicides [124]. These organic compounds are classified as harmful pollutants [125]. The photo-Fenton process can occur in three main UV ranges with use of artificial or sun light, specifically UVC (λ < 285 nm), UVB (λ = 285–315 nm), and UVA (λ = 315–400 nm); UVA, UVB and UVC irradiations demonstrate different disintegration rates of organic pollutants [102]. The main obstacles to the utilization of photo-Fenton processes on an industrial scale are associated with high operating costs because of the operation and maintenance of the UV lamp. In particular, the high electricity consumption is a major drawback of using UV radiation on a large scale. The health risk in the long term should be taken also into consideration, even if all safety measures are applied.

Hydrogen peroxide (H_2O_2) as oxidant or titanium dioxide (TiO_2) as a photocatalyst under UV illumination are commonly utilized in photochemical processes. In many cases, they are used in combination to achieve better results. Rocha et al. showed that a combination of H_2O_2 and sunlight diminished the number of aromatic compounds four times more than the usage of TiO_2 alone [126]. The decomposition of H_2O_2 produced $OH^{\cdot}$ radicals, which were responsible for the degradation of organic contaminants in the leachate:

$$H_2O_2 + h\nu = 2OH^{\cdot}$$

A municipal sanitary leachate extracted from Curitiba, Brazil, was treated with 3000 mg L^{-1} H_2O_2 under artificial UV light. It was observed that 97.2% and 55.5% TOC and COD removal was achieved, respectively, after 60 min. The COD removal can be improved by increasing the initial concentration of H_2O_2, as increased H_2O_2 concentrations favor

the generation of more hydroxyl radicals. Regarding the effect of solution pH, the most favorable initial value of pH for the UV/H_2O_2 process is 3.0 in terms of total organic carbon removal. The temperature significantly affects UV/H_2O_2 treatment. The decomposition of hydrogen peroxide has been reported to occur faster at higher temperatures in comparison with lower temperatures. Shu argues that if hydrogen peroxide is used alone, namely without the existence of UV irradiation, acceptable COD and color removal can be achieved (more than 50%); however, the necessary time for this operation is 600 min, which is a quite long period [127]. Specifically, the treatment with hydrogen peroxide alone resulted in 67.9% decolorization and 79.2% COD removal after 600 min. UV irradiation significantly reduced the time required to obtain these results. It has to be noted, however, that the leachate has a high absorption capacity of UV light, which interferes with the production of hydroxyl radicals. In such cases, higher concentrations of H_2O_2 are necessary; Chys et al. reported that the optimum ratio of hydrogen peroxide to COD was 6.1 $\frac{g\ H_2O_2}{g\ COD_0}$, which was considered undesirable in financial terms [128]. Specifically, in the same study the costs of Fenton, UV/H_2O_2 treatment and ozonation based on the prices of chemicals used in each process were estimated, taking into account also the work of Canizares et al. [129]. The results of cost estimation revealed that the H_2O_2/UV process was the most expensive one among the methods examined, with a cost equal to 13.0 $\frac{€}{kg\ COD\ removed}$, while the costs of ozonation and Fenton treatment were 3.1 $\frac{€}{kg\ COD\ removed}$ and 2.5 $\frac{€}{kg\ COD\ removed}$, respectively.

Among other things, leachates contain compounds that are refractory to biological oxidation or may be toxic to microorganisms [130]. The list of such compounds contains phenols, pesticides, non-biodegradable chlorinated solvents etc. [131]. AOPs, particularly Fenton and photo-Fenton-based processes, can be efficiently used to remove such compounds and improve the biodegradability of leachates, especially in the case of mature leachates that are characterized by low BOD_5/COD ratios [132]. The application of these AOPs has led to the improvement of biodegradability [133] and biotoxicity reduction in treated leachates. In the following table (Table 3), 44 selected studies (Figure 2) on the treatment of landfill leachates using AOPs are presented.

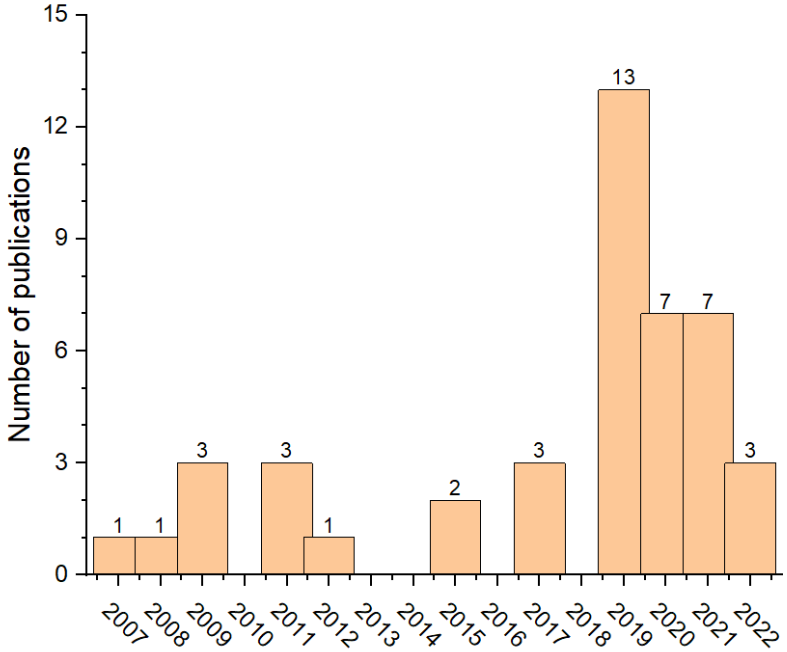

Figure 2. Number of publications focusing on AOPs according to the year published.

Table 3. Landfill leachate treatment by AOPs.

AOPs	Country	Experimental Conditions	Pretreatment	Initial COD, mg/L	Initial pH of the Raw Leachate	BOD_5/COD	Leachate Age	Influent NH_4 +-N (mg/L)	COD Removal (%)	Color Removal (%)	Year	Ref
Electro-Fenton	Turkey	pH_0 = 3; EF reactor; $[H_2O_2]_0$ = 2000 mg/L; I = 3 A; reaction time = 20 min	-	2350	8.36		-	310	72	90	2009	[134]
Electrochemical Oxidation	China	pH = 5–8; current density = 50 to 100 mA/cm^2; proportion of N_2 = 52.2% to 82.06%; electrolysis time = 4 h	-	37.08–43.37	7.60–7.95	0.13–0.18	-	2126–2716	48–70	-	2019	[135]
Electrochemical oxidation graphite/PbO_2 electrode	India	T = 31 °C; $[Cl^-]$ = 1244–4361 mg/L; $[HCO_3^-]$ = 610–7320 mg/L; $[NO_3^-]$ = 12–1483 mg/L; $[SO_4^{2-}]$ = 252–6082 mg/L; electrolysis time = 2 h	-	750 ± 9.4	7.9 ± 0.1	-	-	242 ± 4.3 (NH_3)	35.1–67.2 ± 1.5	-	2020	[136]
Electro-Fenton	India	pH = 2.85; H_2O_2 = 55.75%; current density = 129.5 A/m^2; reaction time = 40 min	MBR	23,200 ± 2054	8.1 ± 0.5	0.15–0.25	-	2196 ± 106 (NH_3–N)	90.62	-	2021	[62]
Electro-persulphate oxidation	Malaysia	pH = 4; [S 2 O 8 2-] = 0.88 g/L; current density = 44.6 mA/cm 2; contact time = 68.3 min	-	330—781	7.18—8.11	-	-	156–187 (NH 3–N)	45.7	97.3	2020	[137]
Electrocatalytic ozonation/Fe-EDTA	Iran	pH = 1.01–2.71; [ozone] = 400 mg/h; current conc. = 100 mA; reaction time = 3 h	-	11,387 ± 146.12	9.3 ± 0.23	-	Mature	-	39–79.7	-	2021	[138]
Electro-coagulation/ persulfate	Vietnam	pH = 2–4; current density = 35 mA/cm^2; aeration time/mixing time rate = 1.3; cycle time = 9 h	CF-SBBR	5119.34	7.0–8.5	-	-	1410.05	9.0–46.8	3.0–95.8	2021	[139]
Electrooxidation and PMS/UV/$CuFe_2O_4$	Iran	pH = 6.3; T = 25–28 °C; PbO_2 anode; current density = 25–50 mA/cm^2; time = 150 min; pH = 5.7; PMS = 10–15 mM; CFNPs = 0.15 g/L; time = 120 min	Electro-coagulation	8580 ± 200	6.4 ± 0.05	0.3	-	1176 ± 20	60–95.6	99.9	2020	[140]
Fe_2O_3 nanoparticles/ electroflotation	Malaysia	pH = 2–10; [Fe_2O_3] adsorption = 5–35 g/L; contact time = 10–120 min; electroflotation pH = 2–10; current density = 10–50 A/m^2; time = 15–60 min	-	2214 ± 12	8.4 ± 1	-	Mature	454 ± 4 (NH_3)	96	<100	2021	[141]
Fenton	USA	pH_0 = 3; $[H_2O_2]_0$ = 0.24 M; H_2O_2/Fe^{2+} (molar ratio) = 3	-	1100–1300	8.18	<0.05	Mature	300	61	-	2007	[118]
Fenton	Canada	pH_0 = 3.5; $[H_2O_2]_0$ = 650 mg/L; H_2O_2/Fe^{2+} (molar ratio) = 1:19; reaction time = 60 min	-	5700 ± 300	7.8 ± 0.3	-	Mixed	530 ± 10	66	-	2009	[142]
Fenton	Spain	pH_0 = 2.5; $[H_2O_2]_0$ = 0.075 M; $[Fe^{2+}]$ = 0.05 M	-	6118.75	8.34	-	Young	1965.00	>80	-	2009	[119]

Table 3. *Cont.*

AOPs	Country	Experimental Conditions	Pretreatment	Initial COD, mg/L	Initial pH of the Raw Leachate	BOD_5/COD	Leachate Age	Influent NH_4^+-N (mg/L)	COD Removal (%)	Color Removal (%)	Year	Ref
Fenton	Portugal	pH_0 = 3; Batch, $[Fe^{2+}]$ = 4 mmol/L; H_2O_2/Fe^{2+} (molar ratio) = 3; reaction time = 40 min	-	743 ± 14	3.5 ± 0.1	-	Mature	714 ± 23	46	-	2011	[143]
Fenton	Slovenia	Fe^{2+}/H_2O_2 (molar ratio) = 1:13.3	-	2455 ± 50	8.4	0.06	-	597 ± 40	80	-	2011	[144]
Fenton	Portugal	pH_0 = 2.9–3.1; batch; $[Fe^{3+}]$ = 20 mg/L; $[H_2O_2]_0$ = 17.6 mM; reaction time = 96 h	Coagulation/ flocculation process	5700	7.8	0.07	Mature		89		2015	[145]
Fenton	India	pH = 3.1; $[Fe^{2+}]$ = 0.04 mol/L; $[H_2O_2]$ = 0.075 mol/L; reaction time = 36 min	-	8900 ± 120	8.80 ± 0.10	0.20	-	1150 ± 20 (TN)	61	-	2021	[146]
Fenton/filtration	Mexico	pH_0 = 4; $[COD]/[H_2O_2]$ = 9; $[Fe^{2+}]/[H_2O_2]$ = 0.6; reaction time = 60 min	-	4268–7610	7.9–8.5	-	-	1210	90.8	95.7	2019	[147]
Fenton with electrolysis	China	$[H_2O_2]$ = 0.187 mol/L; UV current density = 20.6 mA/cm^2; inter-electrode gap = 1.8 cm	-	2500	8.5	-	-	2917	70	-	2019	[148]
Heterogeneous ZVI Photo-Fenton	Spain	pH_0 = 7.0; [alumZVI microspheres] = 5 g/L (iron source); $[H_2O_2]/[COD]$ = 2.125; UV irradiation time = 150 min	Coagulation	4961 ± 496	8.2 ± 0.1	0.03 ± 0.01	Mature	-	62	>90	2019	[149]
Microelectrolysis-Fenton	China	pH = 3.20; $[H_2O_2]$ = 3.57 g/L; [Fe-C] = 104.52 g/L	Chemical flocculation with (PAC)	6880 ± 180	8.50 ± 0.5	<0.08	-	2402 ± 48	90.27	-	2019	[150]
Microwave-persulfate	India	pH = 5.5; $[FeCl_3]$ = 1 g/L; MW-PS system at [PS] = 10 g/L	Coagulation/ flocculation	960–1550	7.35–8.2	-	-	1136–2293 (NH_3)	73–89	86	2019	[151]
Oxidation	Turkey	pH = 11; $[S_2O_8]^{2-}/[Zn^{+2}]$ = 2 g/12 g; reaction time = 120 min	-	2300	8.6	0.07	-	1870	88	98	2020	[152]
O_3/H_2O_2	Portugal	pH_0 = 7; $[H_2O_2]_0$ = 400 mg/L; ozone = 5.6 gO_3/h; reaction time = 60 min	-	743 ± 14	3.5 ± 0.1	-	Mature	714 ± 23	72	-	2011	[143]
Ozonation	China	pH = 6; $[H_2O_2]$ = 9 mL 30% $[H_2O_2]$/L leachate; $[O_3]_0$ = 25 g Nm^{-3} ± 5%; T = 25 °C	Biological	-	3–9	-	Mature	-	41.5–78.9	-	2022	[153]
Ozonation (O_3)/sonication (US)/Fe^{2+}/H_2O_2	Malaysia	$[H_2O_2]_0$ = 60 mM; COD conc. = 1500 ppm; $[Fe^{2+}]$ = 30 mM, pH = 7; O_3 flow rate = 20 LPM; O_3 production = 3.5 g/h; US = 100 W and 20 kHz	-	4850–5150	8.6–8.8	-	-	-	95	100	2017	[154]

Table 3. *Cont.*

AOPs	Country	Experimental Conditions	Pretreatment	Initial COD, mg/L	Initial pH of the Raw Leachate	BOD_5/COD	Leachate Age	Influent NH_4 +-N (mg/L)	COD Removal (%)	Color Removal (%)	Year	Ref
Ozonation–adsorption/adsorption–ozonation	Malaysia	pH = 8.2; O_3 = 27 g/Nm^3; reaction time = 60 min; adsorption zeolite = 160 g/L; contact time = 120 min	-	2100–2580	8.2–8.4	0.019–0.02	Mature	1000–1160	75	82	2021	[155]
Persulfate oxidation	China	pH = 6; [pyrite] = 9.28 mM; dimensionless oxidant = 0.25	-	760–790	5.8–6.0	-	-	11–25	45	-	2022	[156]
Persulfate oxidation with magnetic $CuFe_2O_4$/RGO nanocatalyst	Iran	pH = 2–11; reaction time = 5–60 min	-	2600	8	0.38	Mature	2560 (NH_3N)	65.1	58	2019	[157]
Photo-Fenton	Spain	pH_0 = 3–3.5, batch; [Fe^{2+}] = 2000 mg/L; $[H_2O_2]_0$ = 10,000 mg/L; reaction time = 60 min	-	3300–4400	7.5–8.3	-	Mature	1800–2350	86	>95	2008	[120]
Photo-Fenton	Brazil	pH = 7.8, [Fe^{3+}] = 90 mg/L; UV = 5.8 kJ/L; [H_2O_2] = 220 Mm	-	3324	8	0.28	-	-	-	52.5	2019	[158]
Photo-Fenton	Portugal	pH = 2.7–3; [$Fe2^{+}$] 60 mg/L; [H_2O_2] = 35.1–121 mM; reaction time = 2–4 h/overnight	SBR/ coagulation/ sedimentation	3596–8302	4.2	0.04–0.47	Mature	858–2480	90 ± 1	-	2019	[74]
Photo-Fenton-like homogeneous	Spain	pH_0 = 5; [ferric chloride] = 2 g/L; [H_2O_2]/[COD] = 2.125; UV irradiation time = 30 min	Coagulation	4961 ± 496	8.2 ± 0.1	0.03 ± 0.01	Mature	-	70	97	2019	[149]
Photocatalytic oxidation (•OH radical based/sulfate radical based)	China	pH_0 = 3; [total oxidant] = 0.048 mol/L in UV-H2O2, UV-PMS/H2O2 and UV-PMS processes; n(H2O2)/n(PMS) = 6:4 in the UV-PMS/H_2O_2	-	5680	7.86	-	Mature	1456.76	28.59–37.39	-	2020	[159]
Photolytic ozonation/ ozonation/Peroxone	India	pH = 9; ozone = 5 g/h per liter of leachate; λ = 254 nm; contact time = 60 min	-	5000–17,000	7.8–8.2	-	-	770–3400	45–72	-	2020	[160]
Potassium persulfate oxidation	China	pH = 5; [PS] = 0.05 M; [Fe_2O_3/CO_3O_4/EG] = 0.1 g	-	14,000	8.98	-	-	3120	67.1	-	2019	[161]
Solar photo-Fenton	Portugal	pH_0 = 2.9–3.1; batch, [Fe^{3+}] = 20 mg/L; $[H_2O_2]_0$ = 23.5 mM; 116 mM of H_2O_2 consumption; reaction time = 11.5 h	Coagulation/ flocculation process	5700	7.8	0.07	Mature	-	75	-	2015	[145]
Sulfate radical based oxidation	China	T = 60 °C; [$Na_2S_2O_8$] = 80–160 meq/L	-	1096	8.2	-	-	560 (NH_3)	76–81	-	2019	[162]

Table 3. *Cont.*

AOPs	Country	Experimental Conditions	Pretreatment	Initial COD, mg/L	Initial pH of the Raw Leachate	BOD_5/COD	Leachate Age	Influent NH_4 +-N (mg/L)	COD Removal (%)	Color Removal (%)	Year	Ref
$UV/S_2O_8^{2-}$	China	pH = 5; [persulfate] = 1.5 g/L	ARB-based biological technique	1700–2300	7.9–8.2	-	Mature	1040–1237	81	-	2017	[163]
UV/TiO_2	China	pH = 5; $[TiO_2]$ = 1 g/L	ARB-based biological technique	1700–2300	7.9–8.2	-	Mature	1040–1237	82	-	2017	[163]
$UV_{solar}/O_3/H_2O_2/S_2O_8^{-2}$	Chile	Solar-batch reactor; irradiation time = 250 min; $[S_2O_8^{-}2]$ = 0.2 g/L; $[H_2O_2]_0$ = 0.67 g/L; solar irradiation = 50.4 kJ/L; Q_{uv} = 3 gO_3/L	-	9172	8.7	-	-	3030	28.9	76.8	2019	[164]
UV/Persulfate	Malaysia	pH = 11.5; UV/PS, ammonia stripping flow rate = 50 mL/min; [PS] = 12 mM	-	1039 ± 150	6	0.07	-	549 ± 15 (NH_3N)	91	5–68	2021	[165]
$UV/PMS/Fe^{2+}$	Iran	pH = 7.25; Fe^{2+} = 0.46 g; PMS/Fe^{2+} = 2.54 g/g	-	10,780	7.4	0.52	-	1300 (NH_3N)	67.16	-	2022	[166]
ZVI Fenton-like	Portugal	pH_0 = 2, ZVI as an iron source; [iron shavings] = 25 g/L; $[H_2O_2]_0$ = 13.40 g/L; reaction time = 120 min	Biological processing	2047 ± 15	8.0	0.61	-	-	48	-	2012	[167]
ZVAl based H_2O_2 and persulfate oxidation	India	pH = 1.5; $[ZVAl]_0$ = 10 g/L; acid washing time = 20 min,	-	4896	8.3	0.04	Mature	-	83	63.7	2020	[168]

4. Membrane Separation Processes

Among commonly used water and wastewater treatment processes, membrane separation is probably the most efficient technology available. Membrane treatment of landfill leachate is mainly implemented in developed countries; the MBR technique is widely used in North America and Europe, while RO is mainly applied in North America. Table 4 summarizes studies focused on landfill leachate treatment by pressure-driven membrane processes and it can be seen that developing countries are also expressing interest in treating landfill leachate. For example, there are 9 published papers from Asia, 15 from Europe, Australia and the USA, and 10 from Brazil and Iran. RO and NF are the main membrane processes used for treatment of landfill leachates. Along with that UF, MF and FO have been also employed (Figure 3).

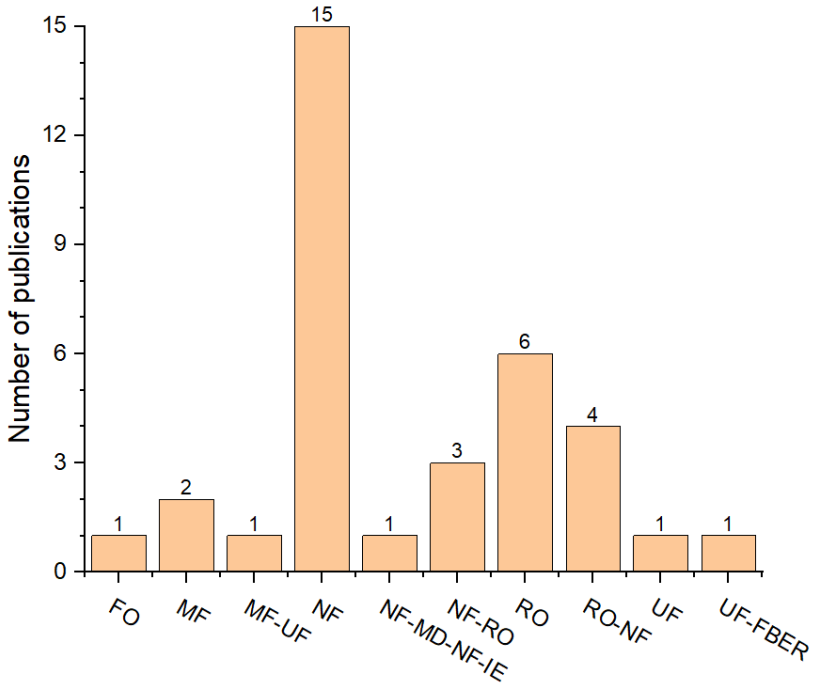

Figure 3. Number of publications dedicated to the particular membrane process/es.

The majority of studies applied a pretreatment step before membrane separation, such as screen and granular filtration, precipitation, coagulation, sedimentation, ion exchange, acidification, adsorption, membrane separation and MBR.

Literature review shows that only the combination of pretreatment—physical, chemical or biological—and membrane separation allows an efficient treatment of landfill leachates. Electrocoagulation was found to be more efficient than chemical coagulation due to higher coagulation concentration and formation of polymeric aluminum. On the other hand, chemical coagulation demonstrated better results than adsorption, while ion exchange showed a greater potential than coagulation for leachate treatment. Existence of both aerobic/anaerobic ammonia-oxidizing organisms and denitrifying bacteria in MBR placed before NF treatment allowed the simultaneous removal of nitrogen and organic carbon.

The membranes retain pollutants due to size exclusion, charge repulsion, hydrophobic/hydrophilic interactions removal mechanisms and their combinations. For instance, RO process can achieve 95–99.5% COD removal while MBR followed by RO/or NF/or FO sequence also showed quite high COD removal, i.e., 88–99.53%. Kwon et al., 2008; Chaudhari and Murthy 2010; Dolar et al., 2016; Istirokhatuna et al., 2018; Pertile et al., 2018; Kulikowska et al., 2019 reported less than 80% COD removal [169–174]. Combination of NF with the AOPs demonstrated quite good COD retention results, i.e., 87–96% [175,176].

It has been found that retention of ions in membrane processes depends on transmembrane pressure, electrolyte concentration, valency, diffusion and hydration energy.

Values of water flux drastically varied from study to study, i.e., minimum and maximum water flux consisted 1.6 and 2556 LMH, respectively. Dong et al., 2014 observed that in short-term experiments orientation of FO membrane was playing an important role [17]. For example, water flux in active layer facing feed solution configuration was higher than in in active layer facing draw solution orientation. Silva et al., 2018 demonstrated 64–71% water flux decline that was explained by concentration polarization phenomena. The authors claimed that there are two main resistances in the NF process, i.e., membrane resistance and concentration polarization. Ameen et al. observed that there are two types of fouling in MF membranes [177]. The first is due to deposition of colloidal contaminants on the membrane's surface and the second is a result of high negative pressure or substances in leachate that irreversibly affect the membrane's structure. Rukapan et al. reported that RO membranes can be efficiently cleaned by base with subsequent acid treatment [178].

Table 4. Landfill leachate treatment by membrane separation techniques.

Process	Pretreatment	Material [4] Geometry [5]	Mode	Pore Size	Velocity (or Flow Rate)	P (Bar)	Country [6]	Initial COD (mg/L)	Initial pH	BOD/COD	Initial $N–NH_4^+$ mg/L	Flux (LMH)	COD Removal (%)	Year	Ref
FO	MBR	CTA	Cross-flow	-	25 cm/s	-	China	696 ± 20 (after MBR)	-	-	-	18–8	98.6	2014	[17]
MF	Coagulation	HB	Cross-flow	100 nm	-	0.133–0.467	Malaysia	30,000–50,000; <2000	-	-	-	18–1.6	-	2011	[177]
MF	-	PEI	Cross-flow	100 kDa	8.9 m/s	0.5–1.5	Brazil	1182	-	-	-	20–40	43	2018	[174]
MF UF	-	Ceramic membranes	Cross-flow	300 kDa, 5 kDa	17 L/min	0.3 MPa	Poland	788 ± 61	8.12 ± 0.03	0.07 ± 0.005	0.07 ± 0.005 (NH_3)	37–20 (UF)	73.1	2020	[179]
NF RO	Sieve screen filtration	RT (NF) FS (RO)	Cross-flow	-	-	1.96–5.88	Korea	1100–1380	7.7–8.5	-	-	3–23	52 (NF) 95 (RO)	2008	[173]
NF	-	PA	Cross-flow	300 Da	-	5–20	India	56,521/ 109,205	6.7/6.8	0.24–0.32	-	20–270	47–85	2010	[169]
NF	Electro coagulation or coagulation	PA	Dead-end	0.84 nm/ 1.28 nm	-	5	Australia	-	7.3	-	-	-	-	2010	[180]
NF	MBR/UF	PA/HF	Cross-flow	200 Da	-	7	China	4670–6700	-	-	820–960	10–15	92–96 (total)	2010	[15]
NF	Ammonia stripping MBR	-	Cross-flow	-	-	14–15	Turkey	24,000	5.5–8.5	-	2313	-	99 (total)	2012	[181]
NF	Air stripping MBR	SW	Cross-flow	-	160 L/h	10	Brazil	4044 ± 544	8.12 ± 0.18	0.03 ± 0.02	1716 ± 386	6–7	88 ± 5 (NF) 91 ± 5 (total)	2015	[182]
NF	Precipitation MF	PSA (NF) PEI/HF (MF)	Cross-flow	500 nm (MF)	144 L/h (NF)	10 (NF)	Brazil	2848 ± 523	7.9 ± 0.8	-	-	-	94 ± 8	2015	[183]
NF	MBR/AC	-	Dead-end	-	-	5	Iran	51,000	8.2	0.3	1951	-	98	2016	[184]
NF-MD NF-IE [1]	Electro coagulation	PTFE/FS (MD) PA/FS (NF)	Dead-end	220 nm (MD)	-	5 (NF)	Australia	-	8.41	-	-	-	-	2016	[185]
NF RO	Coagulation	PA (RO)	Cross-flow	-	2 m/s	20 (RO); 15 (NF)	Poland	5079	8.2	-	-	14.4–61.2	87.5 (NF) 97.6 (RO)	2017	[186]
NF	Air stripping MBR	PA	Cross-flow	200 Da	2.4 L/min	8–12	Brazil	6900	8.5	-	2200	6–14.5	99 ± 1	2018	[187]
NF	Anoxic-aerobic MBR	PA/SW (NF)	Cross-flow	200–400 Da	-	41 (max)	Poland	1745–3947	7.12–7.29	-	631–1371	-	99.53	2018	[188]

Table 4. *Cont.*

Process	Pretreatment	Material [4] Geometry [5]	Mode	Pore Size	Velocity (or Flow Rate)	P (Bar)	Country [6]	Initial COD (mg/L)	Initial pH	BOD/COD	Initial $N–NH_4^+$ mg/L	Flux (LMH)	COD Removal (%)	Year	Ref
NF-RO RO-NF	-	PA/FS	Dead-end	200 Da (NF)	-	20–30 (NF); 50–60 (RO)	Germany (S)	1900	8.01	-	-	5–120	-	2018	[189]
NF	MF	-	Cross-flow	200 Da	-	4–6	Indonesia (S)	1781	8.4	-	-	590–295 87–31	13–39 94–97	2018	[171]
NF	AOP-Fenton, MF	Dow Filmtec NF90	Cross-flow	-	96 L/h	10	Brazil	2848 ± 523 7.9 ± 0,8	-	-	1319 ± 350 (NH_3)	7.8	94–96	2019	[176]
NF	Coagulation–flocculation	TriSep Cellulose Acetate (CA)	Cross-flow	500–700	60 L/h	8	Brazil	4137 ± 30	7.84 ± 0.08	0.07	4137 ± 30	16–11	94	2019	[190]
NF	-	Membrane SR100 Membrane NP030	Cross-flow	200–400	120 L/h	6–9	Brazil	2258 ± 230	-	0.21	14.8 ± 1	4.8–11	88–90	2020	[191]
NF	Air stripping/MBR	FilmTec NF90 2540 NF membrane	Cross-flow	200–400	144 L/h	7.5	Brazil	3238–3374	8.5–8.6	-	431–448	-	22–27	2020	[50]
NF	MBR, Fenton	FilmTec NF90 membrane	Cross-flow	-	2.4 L/min	10	Brazil	2910 ± 44	8.23	8.82 ± 0.17	0.17 ± 0.15 (NH_3)	8.9 ± 1.6	87.24	2021	[175]
RO	Sand filter cartridge filter	PA/DT	Cross-flow	-	-	21–50	China	9080	6.8–7.41	-	-	15–17	~99.5	2008	[192]
RO	Ammonia stripping coagulation flocculation MBR	PP	Cross-flow	-	-	11	Turkey	8500–19,200	6.45–7.50	0.4–0.7	1100–2150	5–22	99.1–99.5 (total)	2009	[193]
RO	Coagulation sedimentation sand filtration MF	-	Cross–flow	-	0.1–0.12 m/s	15–25	Thailand	1280–5790	8.58–8.96	-	-	-	~99	2012	[178]
RO NF	Coagulation magnetic ion exchange	PAR/FS	Cross-flow	-	0.2 m/s	14	USA	2225–2915	7.5–7.71	0.02 – 0.14	-	2556/1656	-	2012	[27]
RO NF	Coagulation filtration	-	Cross-flow	-	750 mL/min	15	Croatia	1720	8.05	< 0.27	-	< 80	>95	2015	[194]

Table 4. *Cont.*

Process	Pretreatment	Material [4] Geometry [5]	Mode	Pore Size	Velocity (or Flow Rate)	P (Bar)	Country [6]	Initial COD (mg/L)	Initial pH	BOD/COD	Initial $N–NH_4^+$ mg/L	Flux (LMH)	COD Removal (%)	Year	Ref
RO	Sand filter cartridge filter acid addition	PA/DT	Cross-flow	-	-	65	Poland	1266	7.45	0.64	-	-	97.03	2015	[31]
RO NF	Coagulation Adsorption UF	-	Cross-flow	100 Da (RO) 150–300 Da (NF)	750 mL/min	15	Croatia	1370/ 747	7.94/7.68	0.19	-	<135, <75–100	>37	2016	[170]
RO [2]	UF	PA	Cross-flow	-	-	10–65	Italy	1368 ± 422 (after UF)	7.2 (after UF)	-	35 ± 46 (after UF)	7–33	95–99.5	2018	[30]
RO	SBR	Polyamide film	Cross-flow	-	8 dm^3/ min	3.8 MPa	Poland	3720	7.7	-	910	-	99.9 ± 0.1	2022	[195]
UF	Adsorption	TiO_2	Cross-flow	5 kDa	7–11 L/min	8	Poland	1560 ± 102	8.36	0.11	-	8/9/96	~15–75	2019	[172]
UF/Fixed Bed Electro chemical Reactor	UF	Flat-sheet carbon membrane coated with TiO2	Dead-end	0.47 µm	-	-	China	389	7.6	-	69.6	-	82	2019	[196]

[1.] Ion exchange. [2] Multistage. [3] Activated carbon. [4] PAR: polyaromatic, PA: polyamide, PP: polypropylene, PSA: polysulfonamide, CTA: cellulose triacetate, PEI: polyetherimide, PTFE: polytetrafluoroethylene. [5] RD: rotary disk, DT: disk tube, HF: hollow fiber, SW: spiral wound, FS: flat sheet. [6] S: synthetic landfill leachate.

5. Summary

Referring to the data presented above, it can be seen that all discussed methods are promising technologies for landfill leachate treatment. The efficiency of the treatment depends on the pretreatment method applied and operational conditions. For instance, COD removal by biological treatment may vary from 1.5 to 99.5%, by AOPs—from 9 to 96% and for membrane separation—from 13 to 99.9%. In spite of the fact that some research groups reported relatively high COD removal, in majority of cases, the retention was lower than 90%. Comparison of the COD of landfill leachate after treatment with the discharge limits—50–300 mg/L for COD in Europe, 100 mg/L for COD in China and 140 mg/L for BOD in USA (BOD to COD ratio is provided in Table 1)—leads us in the direction of multi-stage landfill leachate treatment development [197–199]. The sequential treatment, however, requires a very careful design. For example, the biological reactor is a traditional, inexpensive and decently efficient method for treatment of high-organic-content liquids. However, it is slow and sensitive, and could be interrupted by toxic compounds. In the case of the combination of bio-treatment with membrane filtration, the performance could be improved, while such drawbacks as increased capital and operational cost will be brought onto the picture. Moreover, in case a membrane process is operated in cross-flow rather than dead-end mode (85% of the papers mentioned in Table 4 were focusing on the cross-flow regime), non-negligible amounts of landfill leachate concentrate will be produced. The landfill leachate retentate—a waste management issue [197]—possesses increased concentrations of organic pollutants, nutrients, inorganic salts and heavy metals. Retentate recirculation to the landfills is considered the most uncomplicated and affordable method for the concentrate treatment. This treatment approach may facilitate biological degradation of the landfill leachate (due to increased moisture). On the other hand, however, high contaminant concentrations may hinder the activity of microorganisms. Earlier studies suggested mixed recirculation of young and concentrated leachates to avoid the problem of hindrance [200]. AOPs can decrease the sludge volume by improving the sludge dewaterability and be complimentary used to remove non-biodegradable and toxic substances. AOP, like biological and membrane separation, is a non-ideal treatment process too; its disadvantages include the significant cost of treatment, sensitivity in regard to the landfill leachate turbidity and UV lamp replacements.

6. Conclusions

The literature review showed that three methods investigated in this study could be both poorly and highly efficient in landfill leachate treatment. The treatment efficiency is a multifunctional parameter affected by operational conditions, characteristics of landfill leachate and treating medium. It also should be kept in mind that to achieve adequate effluent quality, several physico-chemical and biological processes have to be combined. Successful treatment could be achieved through the study of pilot/full-scale plants. Along with that, this article provides an opportunity to run an analysis of variance (by applying data presented in the manuscript) to find optimal solutions. The authors hope that the presented work inspires colleagues from industry and academia to explore novel efficient approaches for landfill leachate treatment.

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