

Treatment of landfill leachate with different techniques: an overview

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ABSTRACT

Landfill leachate is characterised by high chemical and biological oxygen demand and generally consists of undesirable substances such as organic and inorganic contaminants. Landfill leachate may differ depending on the content and age of landfill contents, the degradation procedure, climate and hydrological conditions. We aimed to explain the characteristics of landfill leachate and define the practicality of using different techniques for treating landfill leachate. Different treatments comprising biological methods (e.g. bioreactors, bioremediation and phytoremediation) and physicochemical approaches (e.g. advanced oxidation processes, adsorption, coagulation/flocculation and membrane filtration) were investigated in this study. Membrane bioreactors and integrated biological techniques, including integrated anaerobic ammonium oxidation and nitrification/denitrification processes, have demonstrated high performance in ammonia and nitrogen elimination, with a removal effectiveness of more than 90%. Moreover, improved elimination efficiency for suspended solids and turbidity has been achieved by coagulation/flocculation techniques. In addition, improved elimination of metals can be attained by combining different treatment techniques, with a removal effectiveness of 40–100%. Furthermore, combined treatment techniques for treating landfill leachate, owing to its high chemical oxygen demand and concentrations of ammonia and low biodegradability, have been reported with good performance. However, further study is necessary to enhance treatment methods to achieve maximum removal efficiency.

Key words | biological treatment, chemical treatment, landfill leachate, organic pollutants

HIGHLIGHTS

- Membrane bioreactors and integrated biological techniques could remove up to 100% of ammonia.
- Enhanced elimination of metals can be gained by combining different treatment methods.
- Better elimination efficiency for suspended solids has been achieved by coagulation/flocculation.

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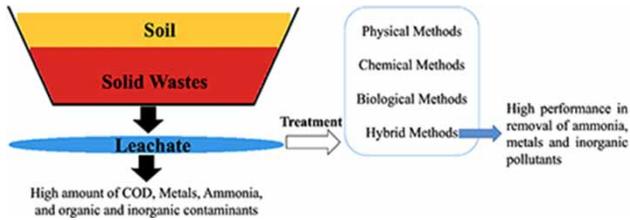
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GRAPHICAL ABSTRACT



INTRODUCTION

Urban solid waste landfills are commonly used for household, industrial nonhazardous and commercial solid wastes as well as nonhazardous sludge (Mojiri *et al.* 2016a). Sanitary landfilling continues to be employed in waste management plans despite its potentially hazardous effect on the environment (Mojiri *et al.* 2017). Compared with other methods, such as incineration, sanitary landfilling generally entails lower operation costs (Gotvajin & Pavko 2015). Waste may undergo a series of biological and physicochemical transformations after being landfilled, thereby producing extremely polluted wastewater called leachate. Such wastewater may pollute nearby ground and surface water as well as soil (Zamri *et al.* 2017).

Landfill leachate is characterised by high chemical and biological oxygen demand (COD, BOD) and often consists of high concentrations of organic contaminants, heavy metals, toxic materials, ammonia and inorganic materials as well as refractory compounds, such as humic substances (Chávez *et al.* 2019) as well as contaminants of emerging concern (Eggen *et al.* 2010). The characteristics of landfill leachate may differ depending on the degradation procedure, climate, hydrology conditions and age of a landfill. Ecological pollution and health issues are commonly connected to the insufficient treatment of landfill leachate (Mojiri *et al.* 2016a).

Minimising risks to the environment and human health is a serious concern in open dumping and sanitary landfills (Xaypanya *et al.* 2018). Appropriate key techniques for landfill leachate treatment consist of biological methods and chemical and physical processes. However, a comprehensive assessment of landfill leachate, including its

characteristics, influences and treatment techniques, is lacking. Thus, this article serves to provide such a critical review.

LANDFILL LEACHATE AND ITS CHARACTERISTICS

Leachate forms when water penetrates waste in a landfill and transfers certain forms of contaminants (Mojiri *et al.* 2017). Municipal landfill leachate contains pollutants that can be categorised into four key groups, namely, organic contaminants and substrates, inorganic compounds, heavy metals, total dissolved solids (TDS) and colour (Mojiri *et al.* 2016a). Based on its age, landfill leachate may be divided into three key groups (Table 1), namely, young, intermediate and old (Aziz 2012; Tejera *et al.* 2019). Aziz (2012) and Vaccari *et al.* (2019) stated that in ‘young’ landfills (i.e. the acid phase),

Table 1 | Leachate characteristics and treatability based on the landfill age

| Age (years) | Young 0–5 | Intermediate 5–10 | Old >10 |
|---------------------------|---------------|----------------------|------------|
| pH | <6.5 | 6.5–7.5 | >7.5 |
| COD (mg/L) | >10,000 | 5,000–10,000 | <5,000 |
| BOD ₅ /COD | 0.5–1.0 | 0.1–0.5 | >0.1 |
| NH ₃ -N (mg/L) | <400 | – | >400 |
| H.M | Medium to low | Low | Low |
| VFA/HFA | VFA (80%) | VFA (5–30%) + HFA | HFA (80%) |
| Biodegradability | High | Medium | Low |

H.M, heavy metals; VFH, volatile fatty acids; HFA, humic and flavic acids. (Sources: Aziz 2012; Yadav & Dikshit 2017; Tejera *et al.* 2019).

leachate is characterised by low pH levels, high concentrations of volatile acids and simply degraded organic matter. In mature landfills (i.e. the methanogenic phase), leachate methane production and pH are high, and the organic materials present are mainly humic and fulvic fractions. However, there is a slightly difference in some other studies (Wang *et al.* 2018a, 2018b) due to the waste characteristics based on the countries. Table 2 shows the characteristics of landfill leachate around the world. Based on Table 2, most concentrated landfill leachates were located in China with COD (mg/L, 28,000) and in Riyadh (Saudi Arabia) with Fe (167.6 mg/L) for concentrated landfill leachate.

Colour and TDS

Colour is a common pollutant in landfill leachate. The decomposition of certain organic compounds, such as humic acid (HA), may cause water to turn yellow to dark brown (Naveen *et al.* 2016). Gotvajn & Pavko (2015) emphasised that substances and particles produce colour and turbidity. TDS display the integrative influence of certain cations and anions, such as calcium, chlorides, magnesium, sodium, potassium and bicarbonates, on water/wastewater. Furthermore, TDS can be produced from small amounts of dissolved organic matter (Sakizadeh 2019) and may inhibit or diminish the biological degradation of dissolved organic carbon (Hanson *et al.* 2019). Hussein *et al.* (2019) expressed that high electrical conductivity and TDS may specify dissolved organic and inorganic substances in samples.

Organic and inorganic pollutants, and heavy metals

The organic composition of leachate varies depending on waste characteristics, the age of a landfill and climatic conditions (Mojiri *et al.* 2016a). Urban solid waste and landfill leachate contain a wide variety of organic compounds (Scandelai *et al.* 2019). In landfill leachate, dissolved organic matter makes up 80% of total organic compounds and is generally composed of refractory humic substances and volatile fatty acids (Jiang *et al.* 2019). Such refractory organics may not be efficiently degraded by conventional biological treatments. Dissolved organics may be signified by BOD₅ and COD (Samadder *et al.* 2017). Moreover, persistent organic pollutants may be found in landfill

leachate. Scandelai *et al.* (2019) indicated that various organic compounds with medium and low polarity, such as amines, alcohols, carboxylic acids, aldehydes, benzothiazolone, ketones, phenols, chlorinated benzenes, phosphates, nitrogen compounds, pesticides and aromatic and polyaromatic hydrocarbons, have been frequently noticed in leachate. Contaminants of emerging concern – pharmaceuticals, personal care products, surfactants, plasticisers, fire retardants, pesticides and nanomaterials – are also found in many municipal landfills, requiring attention on their management (Ramakrishnan *et al.* 2015; Qi *et al.* 2018).

Inorganic macro components, such as sulphates, chloride, iron, ammonia, aluminium and zinc, comprise anions and cations (Agbozu *et al.* 2015). Talalaj (2015) argued that landfill leachate generally consists of large amounts of compounds, 80–95% of which are inorganic and approximately 52% are organic. Inorganic ions contain chloride (Cl⁻), nitrites and nitrates, cyanide (CN⁻), sulphides (S⁻) and sulphates (SO₄²⁻). Moreover, inorganic cations contain ammonia and ferrous (Talalaj 2015).

One of the most toxic contaminants in landfill leachate is heavy metals. In most developing countries, the segregation of nonhazardous wastes from hazardous wastes before disposal into a landfill is uncommon (Edokpayi *et al.* 2018); therefore, several heavy metals in high concentrations have been reported in the landfill leachates (Chuangcham *et al.* 2008). Removal of heavy metals is a difficult task; consequently, we pay more attention to the removal of metals from landfill leachate in this study. Dan *et al.* (2017a) reported that the most common heavy metals in landfill leachate are chromium (Cr), manganese (Mn), cadmium (Cd), lead (Pb), iron (Fe), nickel (Ni) and zinc (Zn). Metal concentrations in young (acetogenic) leachate are generally higher than those in old leachate (Dan *et al.* 2017a).

LANDFILL LEACHATE TREATMENT METHODS

The different landfill leachate treatment methods are shown in Figure 1 and Table 3.

Biological treatment methods

The biological degradation of contaminants results from the metabolic activities of microorganisms (Gotvajn &

Table 2 | Characteristics of landfill leachate around the world

| Remarks | COD (mg/L) | BOD ₅ | BOD ₅ /COD | Ammonia (mg/L) | Heavy metals (mg/L) | | | | | Location | References |
|--|-------------|------------------|-----------------------|----------------|---------------------|-------|-----------|----------|-------|--|----------------------------------|
| | | | | | Fe | Mn | Zn | Cd | Ni | | |
| Concentrated leachate | 28,000 | 950 | 0.04 | 3.50 | 30.00 | 4.03 | 17.80 | NR | 3.70 | MSW incineration plants, China | Ren <i>et al.</i> (2018) |
| Semi-aerobic | 935 | 83 | 0.09 | 483 | 7.9 | NR | 0.6 | NR | NR | Pulau Burung, Malaysia | Kamaruddin <i>et al.</i> (2015) |
| – | 6,140 | 558 | 0.09 | 1,856 | NR | NR | NR | 0.01 | NR | Heimifeng, Changsha, China | Hu <i>et al.</i> (2016) |
| Covered landfill | 24,040 | 15,021 | 0.59 | 2,281 | 10.37 | NR | 0.96 | NR | 0.95 | Istanbul Kömürçüoda Landfill, Turkey | Akgul <i>et al.</i> (2013) |
| – | 2,350 | NR | NR | 310 | NR | NR | 0.05 | 0.02 | 0.54 | Sivas, Turkey | Atmaca (2009) |
| Sanitation landfill | 2,305 | 105 | 0.04 | 1,240 | NR | NR | NR | NR | NR | Beijing, China | Wang <i>et al.</i> (2016) |
| Semi-aerobic | 1,343 | 96 | 0.07 | NR | 3.41 | 0.17 | 2.3 | NR | 0.17 | Matuail landfill, Bangladesh | Jahan <i>et al.</i> (2016) |
| – | 10,400 | 1,500 | 0.14 | NR | 11.16 | NR | 3.00 | 0.03 | 1.33 | Mavallipura landfill, India | Naveen <i>et al.</i> (2014) |
| – | 17,003 | NR | NR | NR | 167.61 | 10.83 | 0.18 | NR | 0.50 | Riyadh City, Saudi Arabia | Al-Wabel <i>et al.</i> (2011) |
| Semi-sanitary | 3,380 | 760 | 0.22 | 1,150 | NR | NR | 1.35–1.60 | 0.13–0.3 | NR | Nonthaburi Landfill, Thailand | Xaypanya <i>et al.</i> (2018) |
| Concentrated landfill leachate | 1,281 | NR | – | 14.2 | NR | 0.692 | – | – | 0.233 | Jiangsu Province, China | Cui <i>et al.</i> (2018) |
| – | 7,700 | 1,300 | 0.16 | 1,780 | 10.03 | NR | 1.06 | NR | NR | Xiangtan, China | Hu <i>et al.</i> (2011) |
| – | 3,308–3,540 | 823–1,274 | 0.24–0.35 | 1,006–1,197 | NR | NR | NR | NR | NR | Nam Binh Duong, Vietnam | Luu (2020) |
| – | 781 | 1,16 | 0.14 | 212 | 21 | NR | NR | NR | NR | Jones County Municipal Landfill, Iowa, USA | Nivala <i>et al.</i> (2007) |
| Sanitation landfill | 4,737 | NR | NR | 1,897 | NR | NR | NR | NR | NR | Virginia, USA | Iskandar <i>et al.</i> (2017) |
| NR | 765 | 70 | 0.09 | 342 | 2.6 | NR | 0.07 | NR | NR | Saint-Rosaire's City, Québec, Canada | Oumar <i>et al.</i> (2016) |
| Old and active landfill | 1,380 | NR | NR | 665.2 | NR | NR | NR | 0.004 | NR | Jakuševac landfill, Zagreb, Croatia | Dolar <i>et al.</i> (2016) |
| Operated for 2 years (very young). Non-hazardous wastes, no fermentable wastes | 260 | 47 | 0.18 | 187 | NR | NR | NR | NR | NR | France | Ricordel & Djelal (2014) |
| – | 3,847 | 388 | 0.11 | 3,158.98 | 21.50 | NR | NR | 1.70 | NR | Ouled Fayet landfill site, Algeria | Boumechhour <i>et al.</i> (2012) |
| Sanitation landfill | 4,425–4,860 | 433–588 | 0.09–0.12 | NR | NR | NR | NR | NR | NR | Sao Carlos, Brasil | Ferraz <i>et al.</i> (2014) |
| – | 1,013 | NR | NR | 398.02 | 6.84 | 0.42 | NR | 6.26 | NR | Guaratinguetá, Brasil | Peixoto <i>et al.</i> (2018) |

NR, not reported.

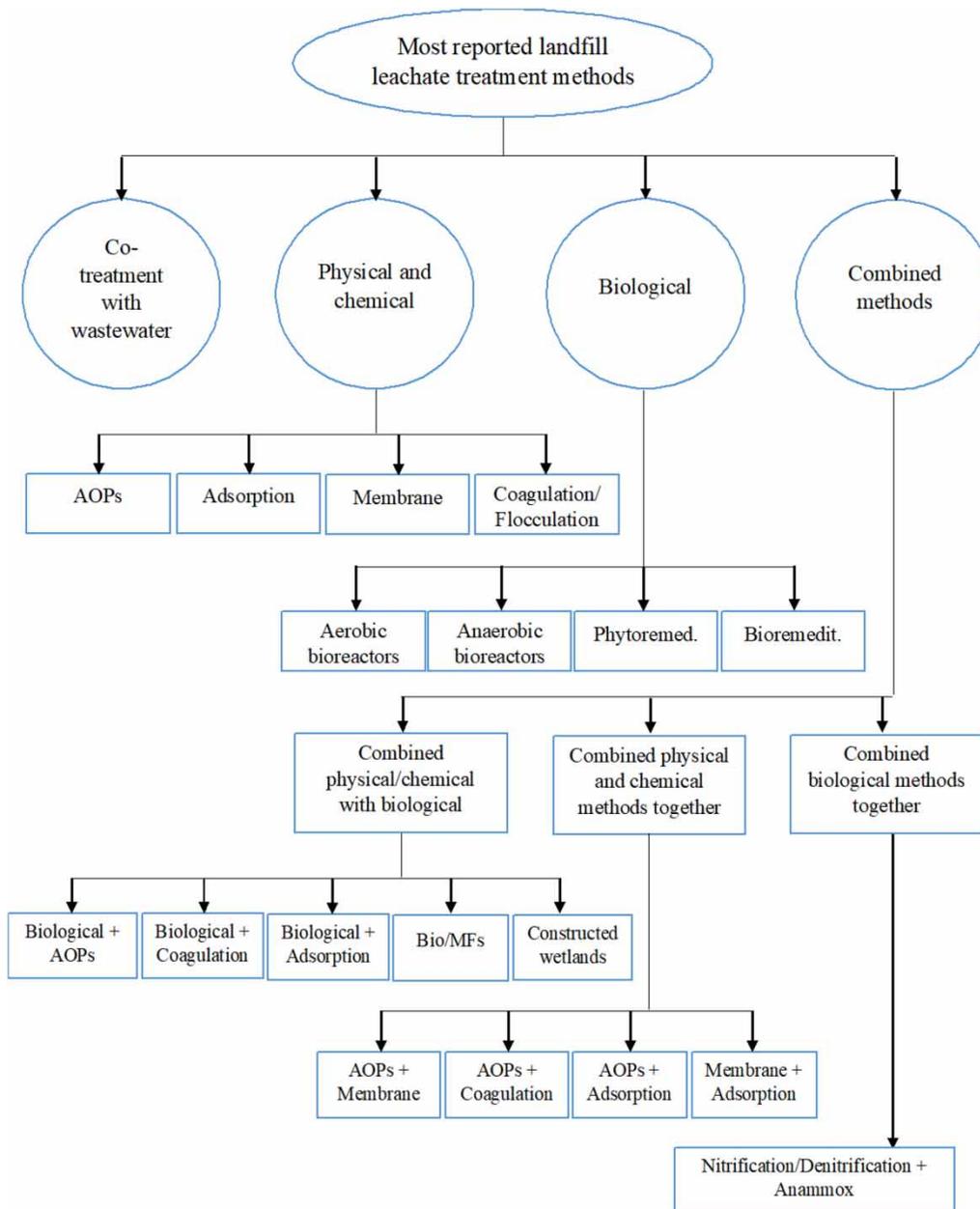


Figure 1 | Common landfill leachate treatment methods.

Pavko 2015). Owing to their cost effectiveness, biological techniques are commonly used to eliminate nutrients (e.g. ammonia) and organic compounds; however, such techniques may not be able to efficiently remove heavy metals and nonbiodegradable organics (Miao et al. 2019). Biological methods are classified into two main groups: (i) aerobic biological procedures

and (ii) anaerobic biological procedures (Dabaghian et al. 2019).

Bioreactors

Bioreactors have been applied for treating wastewaters during several years because these methods are simple and

Table 3 | Reported landfill leachate treatment methods

| Compounds | Removal (mg/L) or Removal efficiency (%) | Treatment method | Remarks | Category | References |
|--------------------------|--|---|--|--|--------------------------------|
| Ammonia | 94.5% | Adsorption/Photo-Fenton-Ozone | Pre-treatment was done via activated carbon (Sawdust) activated by H ₃ PO ₄ . After the adsorption process, the leachate was moved to a solar photo-Fenton/O ₃ process. | Advanced oxidation process/Adsorption | Poblete & Pérez (2020) |
| COD | 95.1% | | | | |
| Colour | 95.0% | | | | |
| HA (ABS ₂₅₄) | 97.9% | Electrocoagulation/Fiber filtration | Anodic electrodes were arranged in parallel. After electrocoagulation with aluminium or iron electrodes, the treated landfill leachate was applied to two stages of fiber filters. | Advanced oxidation process/Coagulation/Adsorption | Li <i>et al.</i> (2017) |
| COD | 94% | | | | |
| As | 87% | | | | |
| Fe | 96% | | | | |
| P | 86% | | | | |
| COD | 3,381.9 mg/L | Electro-catalytic ozonation | The current density was 42.1 mA/cm ² , and ozone concentrations varied 100–400 mg/h. This method increased biodegradability index from 0.27 to 0.45. | Advanced oxidation process | Ghahrchi & Rezaee (2020) |
| BOD | 1,521 mg/L | | | | |
| Ammonia | 90% | Supercritical water oxidation (ScWO)/Zeolite | ScWO was operated under a pressure of 23 MPa at 600 and 700 °C, without the addition of oxidants. Zeolite was used by following ScWO. | Advanced oxidation process/Adsorption (ion-exchange) | Scandelai <i>et al.</i> (2020) |
| Nitrite | 100% | | | | |
| Nitrate | 98% | | | | |
| Colour | 98% | | | | |
| Turbidity | 98% | | | | |
| COD | 74% | | | | |
| COD | 83.3% | Kefir grains/Ag-doped TiO ₂ photocatalytic | Biological pre-treatment was done in 250 mL beakers containing 50 mL of leachate inoculated with Kefir grains. Then, leachate was moved for treatment by using Ag-doped TiO ₂ photocatalytic. | Advanced oxidation process/biological method | Elleuch <i>et al.</i> (2020) |
| Ammonia | 70.0% | | | | |
| Cd | 100% | | | | |
| Ni | 94.0% | | | | |
| Zn | 62.5% | | | | |
| Mn | 53.1% | | | | |
| Cu | 47.5% | | | | |
| COD | 68% | | | | |
| Colour | 97% | | | | |
| HA (UV-254) | 83% | | | | |

(continued)

Table 3 | continued

| Compounds | Removal (mg/L) or Removal efficiency (%) | Treatment method | Remarks | Category | References |
|--------------------------------|--|--|--|---|----------------------------------|
| COD | 97.8% | Fenton process | The Fenton reaction was done by adding powdered ferrous sulphate and an appropriate $H_2O_2:Fe^{2+}$ ratio. | Advanced oxidation process | Roudi <i>et al.</i> (2018) |
| COD HA | 90.2% 93.7% | Coagulation-flocculation/ Microelectrolysis-Fenton processes | Landfill leachate was treated by chemical flocculation with polyaluminium chloride (PAC) as flocculant, and subsequently purified by microelectrolysis-Fenton process. Concentration of H_2O_2 (mg/L) varied 2.66–4. | Advance oxidation process/Coagulation-flocculation | Luo <i>et al.</i> (2019) |
| COD Colour Ni | 88.2% 96.1% 73.4% | Electro-ozonation/adsorbent augmented SBR | At first stage, the raw concentrated leachate was treated by electro-ozonation reactor. The electro-ozone reactor was reinforced by a cross-column ozone chamber to develop ozone gas diffusion. Furthermore, the ozone reactor was supported with anode and cathode plates (Ti/RuO ₂ –IrO ₂ , 18 cm × 8 cm). After that leachate was moved to the second reactor (SBR + Composite adsorbent). | Advanced oxidation process/biological/adsorption | Mojiri <i>et al.</i> (2017) |
| Colour Turbidity Ammonia | >90% >90% >90% | EO/Coagulation | Al ₂ (SO ₄) ₃ with dosage of 50 g/L was added as coagulant. And two stainless steel plates were applied as electrodes. Sodium sulphate 0.1 mol/L was added to the leachate in order to improve the conductivity of the solution. | Advanced oxidation process/coagulation | de Oliveira <i>et al.</i> (2019) |
| COD Ammonia | 36% 99% | UV _{solar} /O ₃ /H ₂ O ₂ /S ₂ O ₈ ²⁻ / Zeolite | Ozone, hydrogen peroxide and UV _{solar} were considered in the same reactor with leachate to produce a high amount of hydroxyl radicals, which have a short life. The S ₂ O ₈ ²⁻ was added directly. Then, treated leachate was treated by zeolite. | Advanced oxidation process /adsorption | Poblete <i>et al.</i> (2019) |
| COD | 91% | UV-based sulphate radical oxidation process/ Coagulation-flocculation | For coagulation-flocculation (pre-treatment), ferric chloride (FeCl ₃) was used, with COD: FeCl ₃ ratio = 1:1.3, as the coagulant. Then, leachate was treated by UV-based sulphate radical oxidation process (UV-SRAOP). For UV/SRAOP, the sulphate radical was produced using UV-activated persulphate (UV/PS) and peroxymonosulphate (UV/PMS). | Advanced oxidation process/Coagulation-flocculation | Ishak <i>et al.</i> (2018) |
| Colour COD Ammonia | 100% 88% 79% | Ozone/catalyst (ZrCl ₄) | Zirconium tetrachloride was added, dosage 1.2 g (COD/ZrCl ₄), as a catalyst to ozone reactor. | Advanced oxidation process | Abu Amr <i>et al.</i> (2017) |

| | | | | | |
|----------|------------|--|--|---------------------------------------|--|
| COD | 16.5% | Vermiculite/Ozonation | Rotating packed bed reactor was used to provide greater gas diffusion to the medium. Optimum operation conditions were as follows: rotation of 915 rpm, pH of 5.8 and ozone flow of 3.9 L/min. Biodegradability was increased (BOD ₅ /COD), from 0.13 to 0.49 by this treatment method. | Advanced oxidation process | Braga et al. (2020) |
| Colour | 40.5% | | | | |
| COD | 72% | MAC/Ozonation | MnCe-ACs were produced by impregnating Mn and Ce oxides onto granular activated carbon surfaces. MnCe-AC was added to a cylinder and ozone was added from bottom of the reactor. | Advanced oxidation process/Adsorption | Wang et al. (2015a, 2015b) |
| HA | 91% | | | | |
| COD | 100% | Activated carbon (Oat hulls) | Oat hulls adsorbents were activated with phosphoric acid and pyrolysed (N ₂ atmosphere) at 350 and 500 °C. | Adsorption methods | Ferraz & Yuan (2020) |
| Colour | 100% | | | | |
| COD | 51.0% | Activated carbon (Coffee wastes) | The washed coffee was oven-dried at 105 °C for 24 h prior to activation. And then it was activated via H ₃ PO ₄ . | Adsorption methods | Chávez et al. (2019) |
| Ammonia | 32.8% | | | | |
| Chlorine | 66.0% | | | | |
| Bromine | 81.0% | | | | |
| Copper | 97.1% | | | | |
| COD | 93.6% | Zero-valent iron nanofibers/ reduced ultra-large graphene oxide (ZVINFs/rULGO) | At the optimum condition, pH, dosage of ZVINFs/rULGO and reaction time were 3, 1.6 g/L and 45 min. | Adsorption methods | Soubh et al. (2018) |
| Ammonia | 84.8% | | | | |
| COD | 77.3% | Silica nanoparticle | At the optimum condition, pH and dosage of adsorbent were 6 and 90 min. | Adsorption methods | Pavithra & Shanthakumar (2017) |
| Colour | 82.5% | | | | |
| COD | 49% | Zeolite Feldspar Mineral Composite Adsorbent | Samples were shaken for 5 h with 200 rpm at pH 7. | Adsorption methods | Daud et al. (2016) |
| Ammonia | 45% | | | | |
| COD | 65.5–92.1% | Amino acid modified bentonite | Batch experiments were done under contact time 20–100 min, pH 2–11 and bentonite dosage of 10–40 g/L. | Adsorption methods | Hajjizadeh et al. (2020) |
| Pb | 99.2 | MS@GG | MS@GG was produced by modification of melamine sponge (MS) with polydopamine (PDA) and then coat with glutathione/graphene oxide. | Adsorption methods | Feng et al. (2019) |

(continued)

Table 3 | continued

| Compounds | Removal (mg/L) or Removal efficiency (%) | Treatment method | Remarks | Category | References |
|---|--|---|--|-------------------------------|-------------------------------|
| COD Ammonia TSS Fe Zn Cu Cr Cd Pb As | 53.5% 91.3% 60.2% 89.7% 94.6% 94.1% 89.9% 17.2% 93.7% 86.4% | Tannin-Based Natural Coagulant | Tannin dosage and pH were 0.73 g and 6, respectively. | Coagulation/flocculation | Banch <i>et al.</i> (2019) |
| COD Colour SS | 61.9% 98.8% 99.5% | Polyaluminium chloride and <i>Dimocarpus longan</i> Seeds as Flocculants | A coagulation–flocculation process using a combination of Polyaluminium chloride (PACl) as a coagulant and <i>Dimocarpus longan</i> seed powder (LSP) as coagulant aid was done. | Coagulation/flocculation | Aziz <i>et al.</i> (2018) |
| COD Ammonia Turbidity | 66.9% 43.3% 96.2% | Red earth as coagulant | The optimal pH and the optimal coagulant dosage were 5.0 and of 9,000 mg/L, respectively. | Coagulation/flocculation | Zainol <i>et al.</i> (2018) |
| COD | 45% | Ferric chloride as coagulant and a cationic flocculant AN 934-SH polyelectrolytes as flocculant | The pH was fixed at 6.3. Optimum condition was 7.2 g/L FeCl ₃ and 0.2 mL/L Flocculant. | Coagulation/flocculation | Taoufik <i>et al.</i> (2018) |
| COD Ammonia | 94.6% Up to 88.9% | Using membrane processes of NF and RO | A working pressure and flow rate were set at 15 bar and 750 mL/min. The surface area of the membranes was 10.7 cm. | Membrane | Košutić <i>et al.</i> (2015) |
| COD BOD Ammonia | 17.5–48.5% 45.4–81.6% 50–98.8% | Using <i>Aspergillus flavus</i> | The <i>A. flavus</i> strain were isolated form leachate contaminated soil. | Bioremediation with the fungi | Zegzouti <i>et al.</i> (2020) |
| COD Ammonia Mn Cu Se | 40% 50% 40% 60% 52% | Using <i>Brevibacillus panacihumi</i> strain ZB1 | The pure colonies of <i>B. panacihumi</i> strain ZB1 were grown in sterile nutrient broth in the incubator shaker for 24 h. About 10% (v/v) of the <i>B. panacihumi</i> strain ZB1 was used to treat the raw leachate sample in the 200 mL conical flask. The leachate sample was treated anaerobically for 21 days and followed by 21-days aerobic treatment. | Bioremediation | Er <i>et al.</i> (2019) |

| | | | | | |
|--------------------------|-------|--|---|--------------------------------|--|
| Ammonia | 90% | Using <i>Chlorella</i> sp. | After growing the <i>Chlorella</i> sp., it was inoculated for experimental studies. | Bioremediation with microalgae | Ouaer et al. (2017) |
| COD | 60% | | | | |
| Ammonia | 83% | Using <i>Chlamydomonas</i> sp. SW15aRL | The <i>Chlamydomonas</i> sp. strain SW15aRL, previously isolated from a sample of raw leachate in 2014 from a landfill site, was maintained in raw leachate or diluted raw leachate samples with a phosphate concentration adjusted to a molar N:P ratio ~ 16:1 prior to the experiments. | Bioremediation with microalgae | Paskuliakova et al. (2018a) |
| Leachate Pollution Index | 74.7% | Using garbage enzyme | The garbage enzyme (fermented mixture of jaggery, organic waste and water in the ratio 1:3:10) was applied. | Bioremediation/Enzyme | Rani et al. (2020) |
| COD | 67% | Using <i>Colocasia esculenta</i> , <i>Gynerium sagittatum</i> and <i>Heliconia psittacorum</i> . | Plants were transplanted in a constructed wetland with a gravity flow ($Q = 0.5 \text{ m}^3/\text{d}$). | Phytoremediation/wetland | Madera-Parra (2016) |
| Cd | 80% | | | | |
| Pb | 40% | | | | |
| Hg | 50% | | | | |
| COD | 75% | Using <i>Imperata cylindrica</i> | Contact time was ranged from 0 to 30 days. | Phytoremediation | Moktar & Tajuddin (2019) |
| Pb | 56.3% | | | | |
| Cd | 16.2% | | | | |
| Zn | 6.5% | | | | |
| COD | 81.0% | Using <i>Typha latifolia</i> | Flow rate of 5 L/day and a HRT of 22 days were used. | Phytoremediation/wetland | Yalçuk & Ugurlu (2020) |
| Ammonia | 60.0% | Using <i>Canna indica</i> | | | |
| COD | 84.0% | | | | |
| Ammonia | 56.0% | | | | |
| COD | 86.7% | Using <i>Typha domingensis</i> | Plants in a reactor with two kinds of substrates including zeolite and ZELIAC. 20% of landfill leachate was mixed with 80% of domestic wastewater at optimum condition. | Wetland/co-treatment | Mojiri et al. (2016b) |
| Ammonia | 99.2% | | | | |
| Colour | 90.3% | | | | |
| Ni | 86.0% | | | | |
| Cd | 87.1% | | | | |
| COD | 93% | Membrane bioreactor + Activated sludge | Membrane sequenced batch bioreactors were inoculated indigenous leachate bacteria or activated sludge. | Bioreactor/Membrane | Azzouz et al. (2018) |
| Fe | 71% | Membrane bioreactor + Indigenous leachate bacteria | | | |
| Zn | 78% | | | | |
| COD | 95% | | | | |
| Fe | 71% | | | | |
| Zn | 74% | | | | |

(continued)

Table 3 | continued

| Compounds | Removal (mg/L) or Removal efficiency (%) | Treatment method | Remarks | Category | References |
|--------------------------------------|--|---|--|--------------------------|---------------------------------|
| COD TOC Ammonia Phosphorous | 63% 35% 98% 52% | Membrane bioreactor | Organic load rate of 1.2 gCOD/L/day and sludge retention time of 80 days were selected. | Bioreactor/Membrane | Zolfaghari <i>et al.</i> (2016) |
| Ammonia TN | >98% >90% | Membrane bioreactor | DM filtration was conducted in a submerged configuration inside the aerobic bioreactor. | Bioreactor/Membrane | Saleem <i>et al.</i> (2018a) |
| COD Ammonia | 80% 78% | Air stripping, and aerobic and anaerobic biological processes | For aerobic reactor, the activated sludge system was applied. And for anaerobic reactor, the upflow anaerobic fixed bed reactor was used. | Bioreactor/Air Stripping | Smaoui <i>et al.</i> (2020) |
| Colour COD Ammonia TSS | 85.8% 84.8% 94.2% 91.8% | SBR and coagulation | Sequential treatment via SBR followed by coagulation was applied. Aluminium Sulphate was used as coagulant. | Bioreactor/Coagulation | Yong <i>et al.</i> (2018) |
| COD | >70% | Anaerobic Sequencing Batch Biofilm Reactor | Biomass from the bottom of a landfill leachate stabilisation pond was immobilized in polyurethane foam cubes as inoculum. | Bioreactor | Contrera <i>et al.</i> (2018) |
| COD Ammonia | 30% 65% | Aerobic sequencing batch reactor (ASBR) | Air upflow velocity was set at 1.0–1.2 cm/s. | Bioreactor | Lim <i>et al.</i> (2016) |
| TN | 95.0% | Partial-denitrification and Anammox | Firstly, leachate diluted with municipal sewage. And two USB reactors were used. | Integrated bioreactor | Wu <i>et al.</i> (2018) |
| TN | 98.7% | Partial nitrification, simultaneous anammox and denitrification | During the aerobic phase, the DO was maintained below 0.5 mg/L. | Integrated bioreactor | Zhang <i>et al.</i> (2019) |
| Ammonia TN | 98% 90% | DM bioreactor | DM filtration was conducted in a submerged configuration inside the aerobic bioreactor provided with a hydrostatic water head of 8 cm. And the initial inoculum was collected from the aerobic bioreactor in a municipal wastewater treatment plant. | Bioreactor/Membrane | Saleem <i>et al.</i> (2018b) |
| COD Ammonia | 99% 99% | Activated sludge process/RO | Biological pre-treatments followed by RO. | Bioreactor/Membrane | Talalaj <i>et al.</i> (2019) |

reliable, and highly cost-effective (Gotvajn & Pavko 2015). But, the main drawbacks of bioreactor treatments involve temperature issues and leachate toxicity for microbial communities (Lippi et al. 2018).

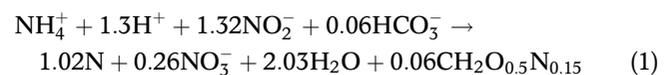
Aerobic bioreactors. Aerobic treatments are the most commonly applied biological procedures. Aerobic reactors involve sustained aeration with large pre-established bacterial populations (i.e. activated sludge) (Torreta et al. 2017). The activated sludge process requires high concentrations of microorganisms, mainly bacteria, fungi and protozoa, to eliminate organic matter from wastewater (Rajasulochana & Preethy 2016). According to Wang et al. (2018a, 2018b), the activated sludge process may efficiently eliminate biodegradable organic material by completely transforming it into carbon dioxide and water. The sequencing batch reactor (SBR) is the most common method for treating landfill leachate. The SBR consists of several time-oriented periodic stages, and its batch operation may enhance process efficacy (Yong et al. 2018).

One of the main drawbacks of this technique involves the need for high concentrations of dissolved oxygen in biofilm reactors for denitrification (Payandeh et al. 2017).

Anaerobic bioreactors. Anaerobic methods generally demonstrate better landfill leachate treatment performance than aerobic treatment techniques owing to the high COD and high BOD/COD ratio of landfill leachates (Azreen & Zahrim 2018). Anaerobic approaches are effective biotechnological treatments for concentrated organic wastewater. Such methods are energy efficient and environmentally friendly owing to their low production of sludge and biogas (Gamoñ et al. 2019). Anaerobic treatment involves the biological decomposition of organic or inorganic matter without oxygen molecules. Key drawbacks of this technique include long retention time, its sensitivity to temperature changes and low elimination efficiency (Azreen & Zahrim 2018). The anaerobic activated sludge process may require upflow anaerobic sludge blanket (UASB) and expanded granular sludge blanket (EGSB) reactors for the purification of landfill leachate. In a UASB reactor, wastewater flows through a sludge bed with high microbial activity (Gotvajn & Pavko 2015). Meanwhile, an EGSB is a

third-generation anaerobic bioreactor that is characterised by high volumetric loading (Wang et al. 2018a, 2018b).

Anaerobic ammonium oxidation (anammox). Anammox bacteria transform ammonium (an electron donor) and nitrite (an electron acceptor) into nitrogen gas, using CO₂ as the carbon source for growth (Torreta et al. 2017). The most commonly applied mechanism of the anammox process is presented by the following equation (Gamoñ et al. 2019):



Anammox bacteria are considered monophyletic and comprise six candidate genera, namely, *Candidatus jettenia*, *Candidatus anammoxoglobus*, *Candidatus brocadia*, *Candidatus scalindua*, *Candidatus anammoximicrobium* and *Candidatus kuenenia* (Mojiri et al. 2020). Remarkably, other types of contaminants, such as high COD and heavy metals, can affect anammox activities. Therefore, the anammox reactors are often combined with other treatment methods (Kumar et al. 2016).

Nitrification and denitrification process. The denitrification and nitrification processes involve the microbial elimination of ammonium. Ammonia is transformed into nitrate under an aerobic condition, which in turn is reduced to N₂ by an anoxic condition during a conventional nitrification-denitrification process (Thakur & Medhi 2019). In the process, firstly, ammonia is oxidised by ammonia-oxidising bacteria into nitrite (NO₂⁻). Secondly, NO₂⁻ is converted into nitrate by nitrite-oxidising bacteria. Finally, the denitrification of nitrate into N₂ is performed by heterotrophic bacteria during the anoxic step (Miao et al. 2019). Generally, this step is integrated into other treatment techniques owing to the effects of other pollutants on the process.

Phytoremediation

Phytoremediation methods employ the capability of plant-soil systems to degrade and inactivate potential toxic elements in leachate (Song et al. 2018). The benefits of phytoremediation include (1) low-cost installation and energy

consumption and (2) the elimination of the pollutants from landfill leachate (Madera-Parra 2016).

Daud et al. (2018) used *Lemna minor* to treat landfill leachate. More than 70% of metals, 39% of COD and 47% of BOD are removed during a 15-day contact time. Daud et al. (2018) and Song et al. (2018) said that several aquatic plants, such as *Colocasia esculenta*, *Pistia stratiotes*, *Eichhornia crassipes*, *Phragmites australis*, *Azolla filiculoides*, *Typha domingensis*, *Hydrilla verticillata*, *Azolla caroliniana*, *Salvinia Cucullata*, *Heliconia psittacorum*, *Azolla pinnata*, *L. minor*, *Lemna gibba*, *Lemna aequinoctialis*, *Gynerum sagittatum* and *Spirodela polyrhiza* can be used to treat leachate. Plants with a remarkable metal-accumulating ability are categorised as hyperaccumulator (Tangahu et al. 2011). Hyperaccumulation is a vital factor for the success of phytoremediation (Alaboudi et al. 2018). Hyperaccumulator plants can be recognised by the translocation factor (TF) and the bioconcentration factor. TF (Equation (2)) is an indication of the plant's capability to translocate metals from its root to its shoot (Ndimele et al. 2014). BCF (Equation (3)) shows the accumulation of metals in plant tissues. Plants with BCF values of more than 2 or TF values more than 1 are considered as hyperaccumulator (Mellem et al. 2009). Table 4 illustrates the concentration of metals in roots and shoots of plants during removing metals by phytoremediation or constructed wetlands.

$$TF = \frac{\text{Concentration of metals in aerial parts}}{\text{Concentration of metal in roots}} \quad (2)$$

$$BCF = \frac{\text{Concentration of metal in plant tissues}}{\text{Concentration of metal in substrate (water)}} \quad (3)$$

Bioremediation

Moris et al. (2018) stated that bioremediation involves biologically removing contaminants from the environment. Its benefits include cost-effective and environmentally-friendly techniques. The use of microalgae, algae and other fungi and bacteria for the bioremediation of landfill leachate has been reported in the literature (Moris et al. 2018; Spina et al. 2018). Paskuliakova et al. (2018a) claimed that algae can eliminate inorganic and simple organic compounds, whereas a few complex substances may undergo a certain

degree of biotransformation. According to Paskuliakova et al. (2018b), microalgae that have been employed to treat landfill leachate include the *Scenedesmus*, *Chlamydomonas* and *Chlorella* genera as well as cyanobacteria and other phylogenetic. Moreover, major bacteria that have been utilised for landfill leachate treatment include *Firmicutes*, *Actinobacteria*, *Proteobacteria*, *Brevibacillus panacihumi* strain ZB1 and *Pseudomonas putida* (Moris et al. 2018; Michalska et al. 2020).

Co-treatment of landfill leachate and urban wastewater with biological methods

To enhance the biodegradability of landfill leachate and BOD/COD ratios, researchers have mixed domestic wastewater with landfill leachate before treatment (Mojiri et al. 2016a). Ranjan et al. (2016) used an SBR for the co-treatment of urban wastewater and landfill leachate. With a hydraulic retention time (HRT) of 6 days and a landfill leachate concentration of 20% v/v, 93, 83, 70 and 83% of ammonia, nitrite, COD and turbidity, respectively, were removed.

Mojiri et al. (2017) emphasised that owing to high COD and BOD/COD ratios, comparing landfill leachate treatments with methods used for domestic wastewater is difficult. Thus, a combined system should be applied to treat leachate. Li et al. (2020) employed denitrification/partial nitrification–anammox to eliminate nitrogen from intermediate landfill leachate. At optimum conditions, total nitrogen (TN) removal rate and TN elimination efficacy were 0.45 m³/d and 96.7%, respectively. The denitrification–nitrification–anammox process demonstrates two vital points, that is, the improvement of degradable COD in wastewater to realise nitrate removal and the improvement of autotrophic bacteria growth. Pirsheh et al. (2017) utilised a combined aerobic–anaerobic/biogranel activated carbon SBR for landfill leachate treatment. This biodegradable landfill leachate treatment demonstrates high performance.

Physical and chemical treatment methods

Adsorption and ion-exchange

Erabee et al. (2018) expressed that adsorption has been broadly applied for the treatment of landfill leachate.

Table 4 | TF and BCF during remediation of metals by plants

| Metal | Plant | Concentration in influent ($\mu\text{g/L}$) | Accumulation in root ($\mu\text{g/g}$) | Accumulation in shoot/leaves ($\mu\text{g/g}$) | TF | BCF | Remarks | References |
|-------|------------------------------|---|--|--|------|------|--|-----------------------------|
| Zn | <i>Water hyacinth</i> | 1,420 | 1,100 | 600 | 0.58 | 1.3 | Mixing ration of landfill leachate and tap water (75%) | Abbas et al. (2019) |
| Pb | | 770 | 600 | 360 | 0.68 | 0.7 | | |
| Cu | | 620 | 400 | 400 | 0.63 | 0.5 | | |
| Fe | | 1,120 | 800 | 650 | 0.53 | 1 | | |
| Ni | | 1,410 | 750 | 500 | 0.57 | 1.25 | | |
| Zn | <i>Water lettuce</i> | 1,420 | 1,300 | 660 | 0.6 | 1.2 | Mixing ration of landfill leachate and tap water (75%) | Abbas et al. (2019) |
| Pb | | 770 | 650 | 350 | 0.5 | 0.6 | | |
| Cu | | 620 | 520 | 250 | 0.58 | 0.5 | | |
| Fe | | 1,120 | 1,000 | 500 | 0.5 | 1 | | |
| Ni | | 1,410 | 1,200 | 470 | 0.5 | 1.1 | | |
| Zn | <i>Lemna minor L.</i> | 1,470 | NR | NR | NR | 0.78 | BCF reported after 3 days | Daud et al. (2018) |
| Pb | | 830 | | | | 0.46 | | |
| Cu | | 690 | | | | 0.63 | | |
| Fe | | 1,170 | | | | 0.76 | | |
| Ni | | 1,210 | | | | 0.58 | | |
| Zn | <i>S. globulosus</i> | 106–887 | 49.98 | 82.81 | NR | NR | After 15 days | Ujang et al. (2005) |
| Ni | | 17–96 | 20.37 | 12.5 | | | | |
| Cu | | 8–31 | 11.11 | 12.78 | | | | |
| Cr | | 30–123 | 26.11 | 24.65 | | | | |
| Pb | | Jun-51 | 7.43 | 8.91 | | | | |
| Zn | <i>E. sexangulare</i> | 106–887 | 124.93 | 206.32 | NR | NR | After 15 days | Ujang et al. (2005) |
| Ni | | 17–96 | 6.58 | 21.28 | | | | |
| Cu | | 8–31 | 5.99 | 12.06 | | | | |
| Cr | | 30–123 | 28.52 | 38.68 | | | | |
| Pb | | Jun-51 | 6.1 | 24.87 | | | | |
| Pb | <i>A. selengensis</i> | 4,080 | 404.79 (10^3) | 65.37 (10^3) | NR | NR | – | Wang et al. (2018a, 2018b) |
| Cd | | 790 | 24.71 (10^3) | 2.90 (10^3) | | | | |
| Cr | | 6,120 | 765.59 (10^3) | 127.99 (10^3) | | | | |
| V | | 14,180 | 645.21 (10^3) | 156.57 (10^3) | | | | |
| Mn | <i>Vetiveria zizanioides</i> | 490 | 121.55 (10^3) | 48.12 (10^3) | NR | NR | pH was set at 7. | Roongtanakiat et al. (2007) |
| Fe | | 16,150 | 1,430.07 (10^3) | 62.31 (10^3) | | | | |
| Cu | | 60 | 4.30 (10^3) | 2.45 (10^3) | | | | |
| Zn | | 4,090 | 82.31 (10^3) | 14.27 (10^3) | | | | |
| Pb | | 50 | 4.50 (10^3) | 0.69 (10^3) | | | | |
| Al | <i>Typha domingensis</i> | 6,560 | 303,910 | NR | 0.14 | 46.3 | Industrial wastewater was treated by phytoremediation. | Hegzay et al. (2011) |
| Fe | | 10,460 | 154,680 | NR | 0.18 | 40.4 | | |
| Zn | | 3,870 | 117,640 | NR | 0.11 | 30.3 | | |
| Pb | | 990 | 14,870 | NR | 0.35 | 15.2 | | |

(continued)

Table 4 | continued

| Metal | Plant | Concentration in influent ($\mu\text{g/L}$) | Accumulation in root ($\mu\text{g/g}$) | Accumulation in shoot/leaves ($\mu\text{g/g}$) | TF | BCF | Remarks | References |
|-------|-----------------------|---|--|--|------|------|---|----------------------|
| Cu | <i>Echhornia</i> | 101.3 | NR | NR | 5.08 | 0.61 | Contaminated water was treated by phytoremediation. | Pandey et al. (2019) |
| Zn | <i>crassipus</i> | 259.4 | NR | NR | 3.64 | 0.91 | | |
| Ni | | 7 | NR | NR | 7.63 | 1.83 | | |
| Pb | | 28.5 | NR | NR | 1.73 | 0.88 | | |
| Fe | | 1,026.8 | NR | NR | 1.04 | 0.92 | | |
| Cr | <i>Acorus calamus</i> | 11,390 | 64,480 | 7,980 | NR | NR | - | Sun et al. (2013) |
| Fe | <i>Linn.</i> | 20,350 | 22,310 | 4,860 | | | | |
| Cu | | 45 | 1,590 | 650 | | | | |
| Zn | | 7,720 | 9,970 | 3,930 | | | | |
| Cr | <i>Juncus</i> | 11,390 | 30,450 | 15,470 | NR | NR | - | Sun et al. (2013) |
| Fe | <i>effusus L.</i> | 20,350 | 77,290 | 14,090 | | | | |
| Cu | | 45 | 650 | 730 | | | | |
| Zn | | 7,720 | 13,290 | 540 | | | | |

NR, Not Reported.

Advantages of this method include its ease of operation, the simplicity of its design, its insensitivity to toxic substances and its ability to remove a variety of contaminants (Chávez et al. 2019). Different adsorbents and their performance are shown in Table 5.

In adsorption, the pollutants can adhere to the surface of the adsorbent over several mechanisms (Figure 2). The surface of the adsorbent has specific characteristics that allow the attachment of the adsorbate. Adsorption occurs under certain conditions, a reversible phenomenon which is named desorption, is applicable. In desorption, the adsorbates can be released from the surface of the adsorbent and got back to the liquid (Bello & Raman 2019).

Modified activated carbon (MAC), which is produced by immersing granular activated carbon (2.0 g) in a KMnO_4 solution (30 mg/L) for 6 h, was created to treat landfill leachate. Approximately 99% of ammonia and 86% of zinc can be removed by MAC in a contact timespan of 120 min. The Langmuir adsorption capacity (mg/g) of this adsorbent for the removal of ammonia and zinc is 0.16 (Erabee et al. 2018). Zamri et al. (2017) used an ion-exchange resin to treat landfill leachate, with a maximum adsorption capacity (mg/g) based on a pseudo second-order kinetic model of 13.4, 13.5, 14.2, 33,333.3, 10,000.0 and 50,000.0

for Cr^{6+} , Al^{3+} , Cu^{2+} , COD, ammonia and colour, respectively.

Advanced oxidation processes

Advanced oxidation processes (AOPs) that apply a combination of oxidants and catalysts to produce hydroxyl radicals ($\cdot\text{OH}$) in solutions, such as ultraviolet (UV), Fenton, ozonation and electrochemical oxidation (EO) methods, have garnered interest for the degradation of hazardous organic compounds or biorefractory in wastewater (Särkkää et al. 2015). However, the main drawback of AOPs is high capital and operating costs.

In an EO process, contaminants are eliminated either by (a) direct EO in which organics are oxidised by moving electrons to an anode directly or (b) indirect EO in which certain electroactive species that act as mediators are produced to conduct the degradation procedure (Mandal et al. 2017). The EO of organics in metal oxide anodes was described by Ukundimana et al. (2018) as follows (Equations (4)–(6)).

Water is electrolysed via anodic catalysis to generate adsorbed hydroxyl radicals.

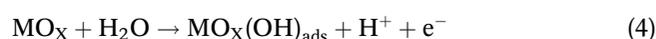


Table 5 | Adsorbents reported for landfill leachate treatment

| Pollutants in landfill leachate | Adsorbent | Adsorption isotherm | Adsorption capacity (mg/g) | Remarks | References | |
|---------------------------------|---|---------------------|----------------------------|--|---|----------|
| TSS | Activated carbon (AC) | Langmuir | 1.77 | AC was derived from coconut shell. AC was modified by heating at 600 °C. | Erabee <i>et al.</i> (2018) | |
| Ammonia | | | 3.18 | | | |
| Zn | | | 0.02 | | | |
| Mn | | | 0.06 | | | |
| Cu | | | 0.07 | | | |
| S ²⁻ | | | 0.02 | | | |
| COD | AC | Langmuir | 272.75 | AC was derived from walnut shell. | Mahdavi <i>et al.</i> (2018) | |
| Colour | AC | Langmuir | 555.55 | AC was derived from sugarcane bagasse. | Azmi <i>et al.</i> (2015) | |
| COD | | | 126.58 | | | |
| Ammonia | | | 14.61 | | | |
| Colour | | | Freundlich | | | 0.67 |
| COD | | | 0.20 (10 ⁻²) | | | |
| Ammonia | | | 3.0 (10 ⁻⁷) | | | |
| Pb | AC | Pseudo-second order | 0.03 | AC was derived from sugarcane bagasse. | Salas-Enrriquez <i>et al.</i> (2019) | |
| Cu | | | 0.01 | | | |
| Ni | | | 0.01 | | | |
| Zn | | | 0.01 | | | |
| Colour | | | Biochar | | | Langmuir |
| COD | Biochar | | 35.71 | | | |
| Ammonia | | | 500.00 | | | |
| COD | Biochar | Pseudo-second order | 490 | Biochar was derived from coconut shell at high temperature, and it is activated via microwave heating. | Lam <i>et al.</i> (2020) | |
| COD | Biochar | Freundlich | 5.80 | Biochar was derived from Miscanthus at 450. | Kwarciak-Kozłowska <i>et al.</i> (2019) | |
| FA | Magnetic graphene oxide | Langmuir | 82.16 | - | Zhang <i>et al.</i> (2016) | |
| HA | | | 106.50 | | | |
| Pb | | | 45.50 | | | |
| Bisphenol A | Bentonite modified by hexadecyl trimethyl ammonium bromide (HTAB) | Pseudo-second order | 10.44 | The HTAB-bentonite was synthesized by cation exchange with HTAB solution (20 mmol/L) over stirring. | Li <i>et al.</i> (2015) | |
| Ni | Red mud | Langmuir | 11.06 | Batch experiments were done with neutral pH, adsorbent dosage of 10 g/L and shaking speed of 75 rmp. | Ayala & Fernández (2019) | |
| Zn | | | 12.04 | | | |
| Cd | | | 12.57 | | | |
| Ni | | Freundlich | 2.08 | | | |
| Zn | | 4.40 | | | | |
| Cd | | 3.79 | | | | |

(continued)

Table 5 | continued

| Pollutants in landfill leachate | Adsorbent | Adsorption isotherm | Adsorption capacity (mg/g) | Remarks | References |
|---------------------------------|---|---------------------|----------------------------|--|-------------------------------------|
| Ammonia | Zeolites (Clinoptilolite) | Langmuir | 17.45 | – | Pauzan <i>et al.</i> (2020) |
| Bisphenol A | High silica Y-type zeolite powder | Pseudo-second order | 141.0 | Batch experiments were done in temperature room for 4 h at pH = 7. | Chen <i>et al.</i> (2015) |
| Colour | Zeolites | Langmuir | 0.01 | Activated zeolites were produced by heating to 250 °C. | Aziz <i>et al.</i> (2020) |
| COD | | | $3.0 (10^{-4})$ | | |
| Ammonia | | | $8.9 (10^{-5})$ | | |
| Colour | Zeolites | Langmuir | 42.55 | – | Bashir <i>et al.</i> (2017) |
| COD | | | 0.22 | | |
| Ammonia | | | 0.31 | | |
| Pb | MS@GG | Pseudo-second order | 253.80 | MS modified with PDA and then coated with glutathione/graphene oxide (GG) | Feng <i>et al.</i> (2019) |
| HA | Aminated Magnetic Nanoadsorbent | Langmuir | 181.82 | Amino-functionalized $\text{Fe}_3\text{O}_4@/\text{SiO}_2$ nanoparticles were produced by surface functionalization of $\text{Fe}_3\text{O}_4@/\text{SiO}_2$ nanoparticles using (3-aminopropyl) trimethoxysilane (APTMS) as the silylation agent. Batch experiments were done at neutral pH and shaken speed 150 rpm. | Wang <i>et al.</i> (2015a, 2015b) > |
| Pb | $\text{Fe}_3\text{O}_4@/\text{Mesoporous Silica-Graphene Oxide Composites}$ | Langmuir | 333.33 | – | Wang <i>et al.</i> (2013) |
| Cd | | | 166.67 | | |

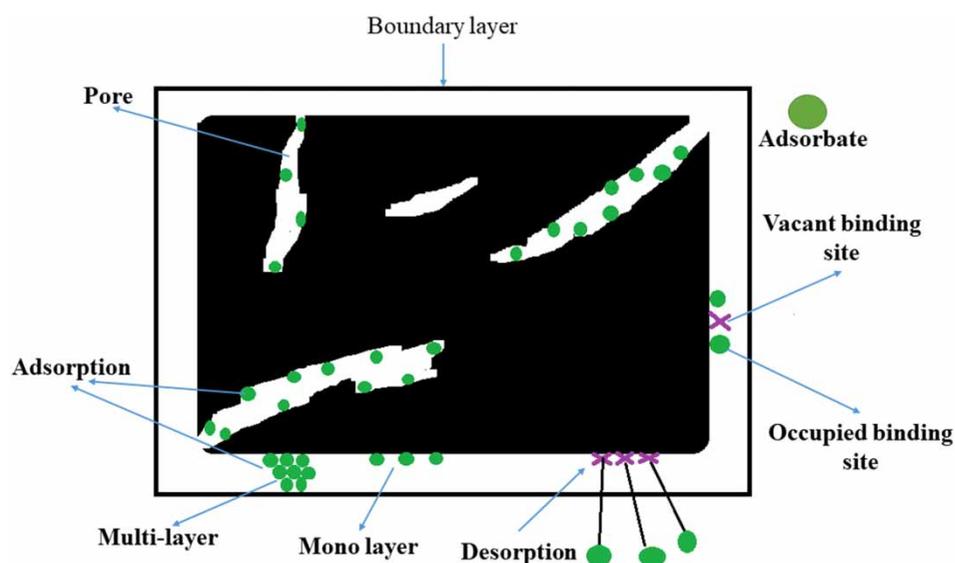
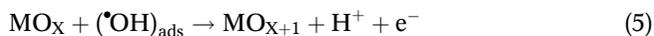


Figure 2 | Basic model of adsorption (Source: Bello & Raman 2019).

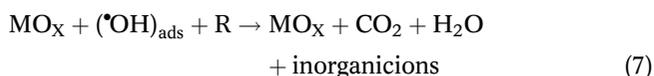
Adsorbed hydroxyl radicals at metal oxide (MO_x) electrodes (except for BDD and Pt) may form chemisorbed active oxygen.



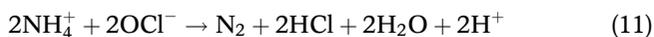
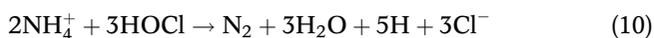
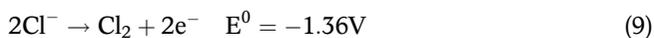
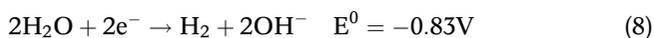
Meanwhile, the hydroxyl radicals will react to one another to form molecular oxygen to complete the electrolysis of the water molecules.



Organic pollutants (R) in landfill leachate can be oxidised via the mechanisms illustrated in Equation (7) by reacting to the physisorbed hydroxyl radicals $\text{MO}_x(\bullet\text{OH})$ formed by Equation (6).

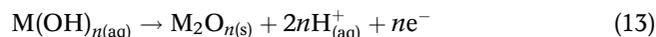
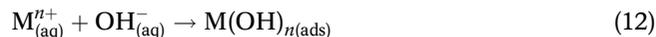


When electricity is applied to wastewater, oxygen gas derived from the breakup of water molecules and chlorine gas is produced in a chloride ion solution (Equations (8) and (9)). Hypochlorous acid (HOCl) and hypochlorite ion (OCl^-) are vital ions responsible for the indirect oxidation of ammonium to nitrogen gas (Equations (10) and (11)) (Ghimire et al. 2020). EO has been deemed effective for ammonium elimination (Mandal et al. 2017).



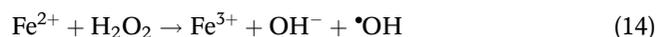
In an EO procedure, the formation of metal oxide on an anode relies on the pH of the electrolyte and metal ion. Yasri & Gunasekaran (2017) indicated that a metallic hydroxide film might form on an anode in an alkaline

media for transition metals (Equations (12) and (13)).



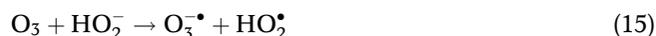
EO, BDD, Ti/Pt, Ti/PbO₂, Ti/SnO₂, Ti/Pt/SnO₂-Sb₂O₄, Ti/RuO₂-IrO₂ and graphite have been commonly applied as electrodes for the treatment of landfill leachate (Ukundimana et al. 2018). Among the benefits of EO, the breakdown of high molecular organic compounds, the absence of sludge and the complete mineralisation of organics are its most significant advantages (Mandal et al. 2017).

The Fenton process has been commonly employed for the oxidation of different organics from wastewater, as it exhibits a high oxidation potential of 2.72 V (Nakhate et al. 2018). Fe(II) ions are oxidised into Fe(III) in the presence of excess H₂O₂ (Equation (14)). This reaction mechanism displays the activation of H₂O₂ in the presence of Fe(II) ions to form hydroxyl radicals that can oxidise organic compounds (Gautam et al. 2019). This classic Fenton reaction may be assisted by electric currents (i.e. the electro-Fenton process) or UV irradiation (i.e. the photo-Fenton process), thereby considerably enhancing its efficacy (Seibert et al. 2019). Singa et al. (2018) argued that compared with other AOPs, the Fenton process includes benefits such as an easy implementation operation, high efficiency and the lack of an energy requirement for H₂O₂ activation.



Ozone is a powerful oxidant, with a redox potential of 2.07 V in an alkaline solution. Consequently, O₃ can oxidise organic and inorganic substances. Gautam et al. (2019) claimed that the key drawbacks of landfill leachate treatment through ozonation include the following. (1) Leachate is a complex wastewater with high organic compounds; hence, high amounts of ozone are required. (2) Ozone mass transfer from a gas to a liquid is low. The ozonation of pollutants may be performed by two techniques, namely, direct and indirect ozonation (Wang & Chen 2020).

A direct O₃ molecule reaction with contaminants involves oxidation–reduction reactions (e.g. reactions between O₃ and HO₂[−]/or O₂^{•−}; Equations (15) and (16); Wang & Chen 2020).



An indirect reaction by •OH is revealed in the following equation (Nilsson 2018):



UV treatment has been generally used to degrade aquatic organic compounds and kill microbes. During the absorption of UV light, electrons are transferred to oxygen molecules that convert O₂ and contaminant molecules into radicals (Equations (18) and (19)).



UV treatment may result in the homolytic cleavage of the chemical bonds of contaminants, thereby causing the formation of two radicals (Mishra *et al.* 2017).

Approximately 99.9% of diethyl phthalate (DEP; organic pollutant) is removed from landfill leachate through the ozone/hydrogen peroxide process (O₃/H₂O₂) at an initial concentration of 20 mg/L DEP and 120 min of ozonation (Mohan *et al.* 2019).

Membrane technology

The use of different membrane technology to treat wastewater has gained considerable attention (Dabaghian *et al.* 2019). Membrane separation involves the selective filtration of influent through different-sized pores (Warsinger *et al.* 2016). Microfiltration (MF), dynamic membranes (DMs), nanofiltration (NF), ultrafiltration (UF) and reverse osmosis (RO) are the main membrane processes employed in landfill leachate treatment (Dabaghian *et al.* 2019). The advantages of using membranes include low overall energy requirements, simplicity and high efficiency (Siyal *et al.* 2019).

DMs may provide a new approach by exploiting fouling as a means for solid–liquid separation. A DM is specified as a self-forming and regenerative fouling surface formed by the removal of colloids, suspended solids and microbial cell particles through a coarse underlying support material (Saleem *et al.* 2018b; 2019). For this purpose, cheap materials, such as filter cloths, have been applied as underlying support to develop DMs (Saleem *et al.* 2019).

MF and UF are categorised as low-pressure (<2 bar) processes. Separation by MF is primarily performed by sieving. However, this process is generally limited to the elimination of organic colloids, suspended solids or particles and bacteria owing to fairly large pore sizes (approximately 0.1–1.0 μm). UF membranes likewise operate mainly via sieving but contain a broader separation range compared with MF and rely on pore sizes between 0.01 and 0.1 μm to remove pathogens, particles and colloids (Warsinger *et al.* 2016).

Meanwhile, NF can eliminate ions that contribute substantially to osmotic pressure; thus, it allows operation pressures that are lower than those used in RO. Pre-treatment is required for heavily contaminated wastewater for NF to be effective (Nqombolo *et al.* 2018).

Among the new procedures for landfill leachate treatment, RO is one of the most promising and effective techniques (Yao 2017). The RO process separates contaminants into two streams, namely, permeate (filtrate) and highly polluted concentrates, which are often recirculated into the waste body (Talaaj 2019). Pertile *et al.* (2018) removed 43% of COD and 63% of BOD from landfill leachate through MF, with a transmembrane pressure of 0.5–1.4 bar.

Coagulation and flocculation

Fundamentally, coagulation facilitates the destabilisation of fine particles (colloids) from wastewater to form a floc that can be settled simply (Achak *et al.* 2019). Coagulation/flocculation efficacy relies on selected coagulants/flocculants. Coagulants are generally trivalent-metal inorganic salts, such as aluminium sulphate, polyaluminium chloride and ferric chloride (Wei *et al.* 2018). Lippi *et al.* (2018) stated that the main advantage of this treatment is its high effectiveness in removing organic matter, suspended solids and

humic acids. However, drawbacks include the cost of chemicals and the management of generated sludge.

Nascimento *et al.* (2016) utilised natural chitosan as a coagulant for landfill leachate treatment. The removal rate for colour and turbidity was 80 and 91.4%, respectively, with a chitosan dosage of 960 mg/L and a pH of 8.5. Nithya & Abirami (2018) removed 85.2% of turbidity from landfill leachate via pine bark as a natural coagulant, with a pH of 7 and a coagulant dosage of 4 g/mL.

Hybrid physical/chemical methods

To improve removal efficiency and decrease energy consumption, several physical/chemical treatment methods have been combined to treat landfill leachate. Xiang *et al.* (2019) posited that hybrid processes, especially AOPs, combined with other treatments may be promising approaches for saving energy. Four integrated systems for combined physical/chemical methods have been identified.

AOPs combined with membranes. The integration of membrane filtration with AOPs may efficiently mitigate membrane-fouling problems, thereby enhancing overall separation performance (Pan *et al.* 2019). Santos *et al.* (2019) removed 94–96% of COD and 96–99% of colour from landfill leachate by combining the Fenton, NF and MF processes. Santos *et al.* (2019) indicated that the concentration of dissolved solids may be high after an AOP–Fenton process owing to the presence of organic matter that has not been completely oxidised and the addition of salts and acid/basic agents. Thus, the use of membranes can resolve this issue.

AOPs combined with coagulation. According to Chen *et al.* (2019), this integrated method can reduce the concentration of organic pollutants and increase the biodegradability of wastewater by altering the molecular structure of residual organics. Gautam *et al.* (2019) identified energy intensiveness, electrode passivation and the formation of chlorinated organics as the main drawbacks of electrocoagulation methods. Integrated photoelectrooxidation and activated carbon can remove 70.3% of COD, 58.3% of ammonia and 58.4% of TN (Klauck *et al.* 2017). Chen *et al.* (2019) eliminated 88.3% of COD, 98.8% of colour and

94.3% of UV254 from landfill leachate by using a combined coagulation–ozonation process.

AOPs combined with adsorption. The integration of AOPs with adsorption has been suggested to improve pollutant removal efficiency, specifically, metals from landfill leachate. Bello & Raman (2019) stated that complex organic contaminant can be degraded by AOPs but complete mineralisation is not mostly practical and some intermediate contaminants are frequently generated. Therefore, combining AOPs and adsorption could remove these intermediates. Integrated H₂O₂–granular activated carbon can reduce 97.3% of COD and increase biodegradable ratio by 116% (Eljaiek-Urzola *et al.* 2018). Eljaiek-Urzola *et al.* (2018) stated that integrating H₂O₂ with activate carbon can improve the decomposition of peroxide in free radicals and enhance performance. Jafari *et al.* (2017) removed 99.8% of tetracycline, as emerging pollutants, from aqueous solution by Heterogeneous Fenton: activated carbon–Fe₃O₄.

Membrane filtration combined with coagulation or adsorption. According to Alimoradi *et al.* (2018), coagulants or adsorbents have been applied sequentially to membranes to eliminate suspended and colloidal substances from wastewater, thereby reducing organic load and hindering membrane fouling. Gkotsis *et al.* (2017) emphasised that the use of coagulants in MBR systems could contribute significantly to reducing transmembrane pressure. Apart from that, Alimoradi *et al.* (2018) stated that coagulation pre-treatment delays the reversible and irreversible fouling by improving sludge filterability and by eliminating soluble microbial products, respectively. Alimoradi *et al.* (2018) removed more than 90% of Al by integrated coagulation-membrane bioreactor. 99.2% of COD, 100% of suspended solids and 97.3% of total organic carbon were removed by combined coagulation and membrane (Boluarte *et al.* 2016). 100% of 4-chlorophenol, 78–100% of oxidation intermediates from wastewater by integrated catalytic oxidation and adsorption (Arsene *et al.* 2013).

Hybrid physical/chemical and biological methods

Biological ways are frequently employed to treat landfill leachate. However, a biological procedure alone is not efficient enough to eliminate the bulk of refractory contaminants in

landfill leachate (Wu *et al.* 2010). Therefore, researchers (Mojiri *et al.* 2016b) have suggested integrated biological methods and physical/chemical techniques to improve biodegradability ratios and increase biological performance in treating landfill leachate. Five commonly applied combined treatment methods have been identified.

Integrated adsorption and biological treatment methods

Adsorption can be employed to diminish contaminants and leachate toxicity to provide favourable growth conditions for microbial growth (Er *et al.* 2018). Munz *et al.* (2007) listed the advantages of combination of adsorption, such as activated carbon, and biological methods as: protecting microorganisms from load pick of inhibiting organic and inorganic compounds, improving refractory organics, improving sludge settleability and dewaterability capacity. Besides, the application of the adsorption technique together with the biological method leads to a reduction of the quantity of adsorbent employed for the wastewater treatment process (Yi *et al.* 2018). Sawdust added to an SBR can remove 99% of COD and 95% of ammonia (Mohajeri *et al.* 2018). More than 60% of ampicillin was eliminated by integrating adsorption and biodegradation (Shen *et al.* 2010). Ammonia was removed at more than 70% from landfill leachate by integrated adsorption and biological treatment (Yi *et al.* 2018).

Integrated membrane and biological treatment methods

Generally, the membrane bioreactor is a vital innovation in treating wastewater treatments since it overcomes the disadvantages of the conventional activated sludge process, such as producing excess sludge, requiring secondary clarifiers, and limitations with elimination of recalcitrant (Iorhemen *et al.* 2016). Among anaerobic biological methods, the anaerobic membrane bioreactor (AnMBR) system, which decouples HRT from solid retention time (SRT), is feasible for treating heavy wastewater, such as leachate (Abuabdou *et al.* 2020). Regarding the drawbacks of membrane bioreactors, Abuabdou *et al.* (2020) argued that starting an AnMBR in temperatures below 20 °C may result in the reduction of biomass growth, thereby causing a long SRT for stabilisation. Xu *et al.* (2019) removed more than 90% of sulphonamides and tetracyclines by using a membrane

bioreactor. More than 90% of COD was removed from landfill leachate by AnMBR (Zayen *et al.* 2010).

Integrated AOP and biological treatment methods

He *et al.* (2020) expressed that integrating AOP techniques, as a pre-treatment, leads to readily biodegradable intermediates for biological posttreatment. Therefore, it has a positive impact for treating wastewaters, such as landfill leachate. Researchers (He *et al.* 2020; Xia *et al.* 2020) reported that zone oxidation, photocatalyst and EO are promising pre-treatment methods to enhance biodegradability of refractory contaminants. A combined semiaerobic aged refuse biofilter and ozonation process can eliminate 92.1% of colour and 61.4% of UV₂₅₄ from landfill leachate (Chen *et al.* 2019). More than 70% of aromatic pollutants, such as p-aminophenol, by hybrid reactor including ozone pre-treatment and bioreactor (Xia *et al.* 2020). COD concentration was decreased to less than 50 mg/L by combined photocatalytic pre-oxidation reactor with SBR (He *et al.* 2020). Integrated ozonation and membrane bioreactor removed up to 99% of pharmaceuticals, such as Etodolac (Kaya *et al.* 2017). 100% of sulfadiazine, 97% of total organic carbon, 94% of BOD₅ and 97% of COD were eliminated by ozonation and membrane bioreactor (Lastre-Acosta *et al.* 2020).

Integrated coagulation and biological treatment methods

Coagulation/flocculation can be applied as pre-treatment and posttreatment with biological treatment methods (Niazi 2018; Güvenç & Güven 2019). Employed coagulation/flocculation as a pre-treatment leads to improvement of the biodegradability and reduces COD, colour and metals in landfill leachate. These advantages can enhance the treatment of landfill leachate with biological methods. The use of the coagulation/flocculation as a posttreatment can remove refractory pollutants, such as metals, COD and organics. Niazi (2018) expressed that biological treatment results the degrading dissolved and colloidal organics which transform to active biomass. The active biomass in reject water produced from the biological method can get more dissolved organics and colloidal solids from the wastewater which is eliminated by coagulation. An integrated coagulation and anaerobic bioreactor process can

remove 72% of COD and 70% of total organic carbon (Yadav *et al.* 2016).

Constructed wetlands

Mojiri *et al.* (2016b) suggested that the constructed wetland (CW) system was engineered to increase water quality. A wetland system comprises permeable substrata, such as gravel, which is typically planted with emergent wetland plants, such as *Schoenoplectus*, *Typha*, *Phragmites* and *Cyperus*. Dan *et al.* (2017b) expressed that degradable organic carbon and ammonia can be efficiently removed from landfill leachate by CW systems. Nitrogen pollutants can be removed by adsorption through substrate, absorption through plant roots, volatilisation in ammonia forms, biological degradation and biochemical transformation into N₂ (Gottshall *et al.* 2007; Badejo *et al.* 2018). Zhuang *et al.* (2019) expressed that more than 50% of nitrogen can be eliminated by microbial activities, such as the nitrification/denitrification process, while around 25% of nitrogen may be absorbed by plant roots. Up to 89% of ammonia removal using a CW was reported by Mannarino *et al.* (2006).

The majority of phenolic compounds are removed by microbial activities and adsorption through substrate (Rossmann *et al.* 2012). Dan *et al.* (2017a) removed 88–100% of phenols, 18–100% of 4-tert-butylphenol and 9–99% of bisphenol A by using a vertical flow-constructed wetland. Apart from organic contaminants, heavy metals can be removed by CW systems.

According to Dan *et al.* (2017b), various mechanisms, such as the adsorption of soil or substrates as well as particulates and soluble organics, the precipitation of insoluble salts and the uptake of aquatic plants and microorganisms, may affect metal removal via CW systems. Ujang *et al.* (2005) removed up to 92.2% of Zn, 96.8% of Ni, 99.5% of Cu, 87.5% of Cr and 98.1% of Pb by using a CW which contained *E. sexangulare* and media.

CONCLUSIONS

Landfill leachate often possesses significant pollution potential with high concentrations of organic and inorganic contaminants. Primary landfill leachate treatment

techniques consist of physical, chemical and biological methods. Owing to high concentrations of contaminants in landfill leachate and its low biodegradability, integrated treatment methods and co-treatment with wastewater are strongly recommended. Membrane filtration and integrated biological methods (nitrification/denitrification/anammox) have demonstrated high performance in removing nitrogen and ammonia from landfill leachate. Moreover, coagulation/flocculation methods have exhibited high efficiency in removing suspended solids and turbidity, with a removal rate of more than 90%. Bioremediation has demonstrated varied removal efficiency for COD, ranging from 17.5 to 60% depending on bacteria or algae species, thereby failing to show high performance in reducing COD. Finally, physical/chemical treatments have exhibited high performance in removing heavy metals.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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