

**TRENDS AND GAPS ON  
NEW EMERGING POLLUTANTS (NEPS)  
IN LANDFILL LEACHATE  
OVER THE PAST 10 YEARS (2010-2020)**

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**SCHOOL OF CIVIL ENGINEERING  
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NEW EMERGING POLLUTANTS (NEPS)  
IN LANDFILL LEACHATE  
OVER THE PAST 10 YEARS (2010-2020)

by

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## **ABSTRACT**

New Emerging pollutants (NEPs) are substances that have recently occurred widely in the environment and recognised as a possible environmental or public health risk. In the new era, even with the new evolving technologies, NEPs still exists, which merit appropriate control. However, there is no sufficient information to assess the risk of NEPs. Landfilling is the most conventional method in disposing of municipal solid waste for the past many years. One of its major drawbacks is the production of highly contaminated leachate. Hence various types of alarming pollutants have evolved through leachate over the years. This study was set to conduct a quantitative approach in reviewing and analysing various past and present studies related to NEPs in landfill leachate. This research will involved a combination of Critical literature review (CLR) and Systematic literature review (SLR) based research, concluding results on NEPs from various studies. It was found from the review that NEPs in leachate consist of different compositions, a wide range of concentrations, and could be treated by various means. The leachate treatment for NEPs is mostly adapted from existing wastewater treatment technology. There is a growing trend in the discharge of NEPs in landfill leachate over the past 10 years. However, there are still gaps in the treatment technology as most of the current systems are not meant for advanced parameters such as NEPs. These results had provided future perspectives on the management of NEPs in landfill leachate as supported by various inputs from scholars in the field.

## **ABSTRAK**

Bahan pencemaran baru muncul (NEPs) adalah bahan yang baharu dijumpai kebelakangan ini secara meluas di persekitaran dan diakui boleh memberi kemungkinan risiko kepada persekitaran atau kesihatan awam. Namun, di era baru, walaupun dengan teknologi yang semakin maju, NEPs masih muncul dan mewajarkan kawalan tertentu. Walaupun demikian, tiada informasi yang mencukupi untuk menilai risikonya. Tapak pelupusan kambus tanah merupakan kaedah yang paling konvensional dalam pengurusan sisa pepejal perbandaran sejak bertahun-tahun yang lalu. Salah satu kelemahan utama tapak pelupusan kambus tanah adalah penghasilan larut lesap yang tercemar. Oleh itu, pelbagai jenis NEPs yang telah dan sedang berada dalam larut lesap hingga kini. Kajian ini dilakukan secara kuantitatif dalam mengkaji dan menganalisis pelbagai kajian terdahulu dan terkini berkaitan NEPs. Kajian ini melibatkan penyelidikan berasas kajian literatur kritikal (CLR) dan kajian literatur sistematik (SLR) untuk merumuskan hasil kajian dari berbagai penyelidikan berkaitan NEPs. Hasil kajian menunjukkan bahawa terdapat pelbagai komposisi NEPs dalam larut lesap dengan kepekatan dan kaedah olahan yang berbeza. Kebanyakan kaedah olahan NEPs dalam larut lesap berasal dari kaedah olahan air sisa. Terdapat trend yang semakin meningkat dalam pelepasan NEPs dari larut lesap kambus tanah sejak 10 tahun yang lalu. Masih terdapat kekurangan dalam teknologi olahan NEPs semasa disebabkan olahan sediaada tidak disasarkan untuk parameter lanjutan NEPs. Hasil kajian ini telah memberikan perspektif masa depan mengenai pengolahan NEPs dalam pengurusan larut lesap kambus tanah seperti disokong oleh berbagai sarjana dalam bidang ini.

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# CHAPTER 1

## INTRODUCTION

### 1.1 Overview

Landfilling has been an ongoing process for the past many years and is the most conventional method in disposing of municipal waste. It has been used not only in Malaysia but in other developing countries such as European Union countries and the USA (Banch *et al.*, 2019). The proportion and composition of waste in landfills in recent decades has changed, mainly as higher demand and improved quality of consumer products. Products such as paints, oils, batteries, electrical products, and pharmaceuticals potentially deposited at landfills contain hazardous additives. The same goes for common chemicals that are used to strengthen certain properties of the consumer products, such as flame-retardants, plasticisers, surface-active substances, and fragrances. When these unique beneficial additives end up in landfill and decomposed, subsequent chemical compounds, some of which bring negative effects, are released into landfill leachate (Eggen *et al.*, 2010). Consequently, landfill leachate became more complicated in its composition of contaminants which include organic and inorganic pollutants. Due to its ability to infiltrate into the ground and surface water reservoirs, it can develop severe and chronic threats to the environment and health. Therefore, one of the major drawbacks is the production of highly toxic and contaminated liquid at the landfill which is known as leachate (Reshadi *et al.*, 2019).

In this new era with growing industrial and municipal activities, even with the new evolving technologies and developments, new pollutants emerge as well. New emerging pollutants (NEPs) are substances that have recently been shown to take place widely in the human daily surrounding and living environment. After decades of

disposal and decomposition of waste at the landfill that result in the release of chemical compounds, NEPs can be recognised as a possible environmental, wildlife or public health risk. However, there is inadequate information to examine the impact (Ramakrishnan *et al.*, 2015).

Literature has distinguished NEPs into few classes based on their daily use, such as pharmaceuticals and personal care products, surfactants and their degradation products, plasticisers, pesticides, fire retardants, pesticides, and nanomaterials (Ramakrishnan *et al.*, 2015). In China, the two most frequently researched NEPs classes are Phthalate esters (PAEs, with 15 studies) and pharmaceuticals and personal care products (PPCPs, with 13 studies). It is found out that the concentrations of nine different NEPs groups ranged widely from 0.03 µg/L (organochlorine pesticides) to 4500 µg/L (alkylphenol polyethoxylates/bisphenol analog) (Qi *et al.*, 2018).

This research evaluates the trend in NEPs in landfill leachate transformation from the year 2010 to 2020. There has been a significant amount of research into the incidence and behaviour of NEPs in various settings, especially water and wastewater treatment (Ramakrishnan *et al.*, 2015). Researchers have shown a growing interest in landfill leachate studies in terms of valuable and innovative landfill leachate treatment method (Reshadi *et al.*, 2019).

Also, research has proven the impact of NEPs in landfill leachate and have led to stricter requirements of pollution control as well as the implementation of new discharge standards. However, the NEPs treatment in landfill leachate has not been widely reported. The development and implementation of an integrated leachate treatment process that incorporates various physical, biological, and chemical technologies may be a viable choice for reducing leachate contamination levels (Gao *et*

*al.*, 2015). These changes have led to newly developed treatment technologies in leachate management. For instance, application of such as advanced oxidation and reverse osmosis (targeted for micropollutants), nano-filtration (targeted for pharmaceutical substances) and ozonation (targeted for tetracycline antibiotics) will remove the targeted substances in landfill leachate in a more effective manner (Ramakrishnan *et al.*, 2015; Luo *et al.*, 2018). Hence, the current state of knowledge on these advanced technologies for NEPs removal in landfill leachate will be reviewed.

## **1.2 Problem Statement**

A problem arises when unfamiliar types of waste produced by the rapid growth of diverse industries are being disposed of in the landfill and decomposes into distinctive molecules after a long period of time. NEPs produced in landfill leachate are contributing as a possible threat to the environment (contamination of surface water and groundwater) and public health (Ramakrishnan *et al.*, 2015). Nonetheless, people began to question: what are the NEPs that are present in landfill leachate? (Eggen *et al.*, 2010). Where do these pollutants come from? (Ramakrishnan *et al.*, 2015). Most of the research works reported in the literature is mainly restricted to specific pollutants groups, thus seeming like a huge portion of the research is missing and yet to be organised. The research gaps on NEPs in landfill leachate over the past 10 years (2010-2020) remains unexplored. The knowledge is important to evaluate its future perspective, which could lead us to better management of NEPs in the years to come.

## **1.3 Research Questions**

1. What are the differences in the type of NEPs and their characteristics in landfill leachate in the year 2010 to 2020? How is it treated?

2. How are the evolving trends of research on NEPs in landfill leachate from the year 2010 to 2020?
3. What are the research limitations on NEPs in landfill leachate over the past 10 years?
4. What is the future perspective in the management of NEPs in landfill leachate?

#### **1.4 Research Objectives**

The above research questions will be addressed by the following research objectives:

1. To conduct a review on the presence of NEPs in leachate, their common compositions, concentrations and their associated treatment methods.
2. To evaluate the trend and gaps on NEPs in landfill leachate over the past 10 years.
3. To present a future perspective on the management of NEPs in landfill leachate.

#### **1.5 Significance of Study**

The findings of this study will redound to the benefit of society on exploring the trends and gaps on NEPs in landfill leachate of the past 10 years based on studies around the world. To the best of our understanding, limited research has been undertaken to examine this issue. The review would lead to recent data, the presence of types of NEPs in leachate in terms of their common compositions, concentrations, and their associated treatment methods. This research will also provide a unique, quantitative analysis towards the research trends of NEPs in landfill leachate, which identified the scholars, institutions, regions, keywords, etc., which are important information for better management of NEPs in future. The data would be useful to be used for practitioners to understand the challenges, impacts and the control of NEPs in landfill leachate.

## CHAPTER 2

### LITERATURE REVIEW

#### 2.1 Landfilling and leachate

Improper management of municipal solid waste management (MSW) may lead to environmental problems, especially due to poor landfill management. Landfilling is one of the preferred methods of waste disposal due to its economic and simple process (Eggen *et al.*, 2010; Banch *et al.*, 2019). Landfills serve as the ultimate destination for a diverse range of waste from residential, industrial, and commercial sources (Andrews *et al.*, 2012; Masoner *et al.*, 2016).

Leachate contains large amounts of organic and non-organic pollutants; generated when precipitation percolates through waste, and also due to biochemical processes in wastes cells and inherent water content of wastes themselves (Qi *et al.*, 2018). Several studies have been carried out to understand the chemical processes and to identify toxic and hazardous substances which potentially pollutes the abundant environment and even penetrates to groundwater (Eggen *et al.*, 2010; Banch *et al.*, 2019). The collection system of solid waste, landfill age, type and composition of discarded waste, the solubility of waste component, site and hydrological factors, compaction of solid waste and design and operations of the landfill are some of the factors that change the characteristics of leachate (Banch *et al.*, 2019). In addition, the volume of landfill leachate generated at the respective landfill is determined by its surface area and topography, as well as regional climatic conditions such as rainfall and cover type (Kapelewska *et al.*, 2018).

According to the flowchart adapted from Ramakrishnan's (2015), hazardous NEPs are originated from industrial production or services with illegal disposal. They

can come from the disposal of products and packaging from consumers. All the disposal items ended up in MSW landfill and will undergo physical/chemical transformation or biological transformation after some time. The presence of NEPs can be found in appearance in both gas and leachate.

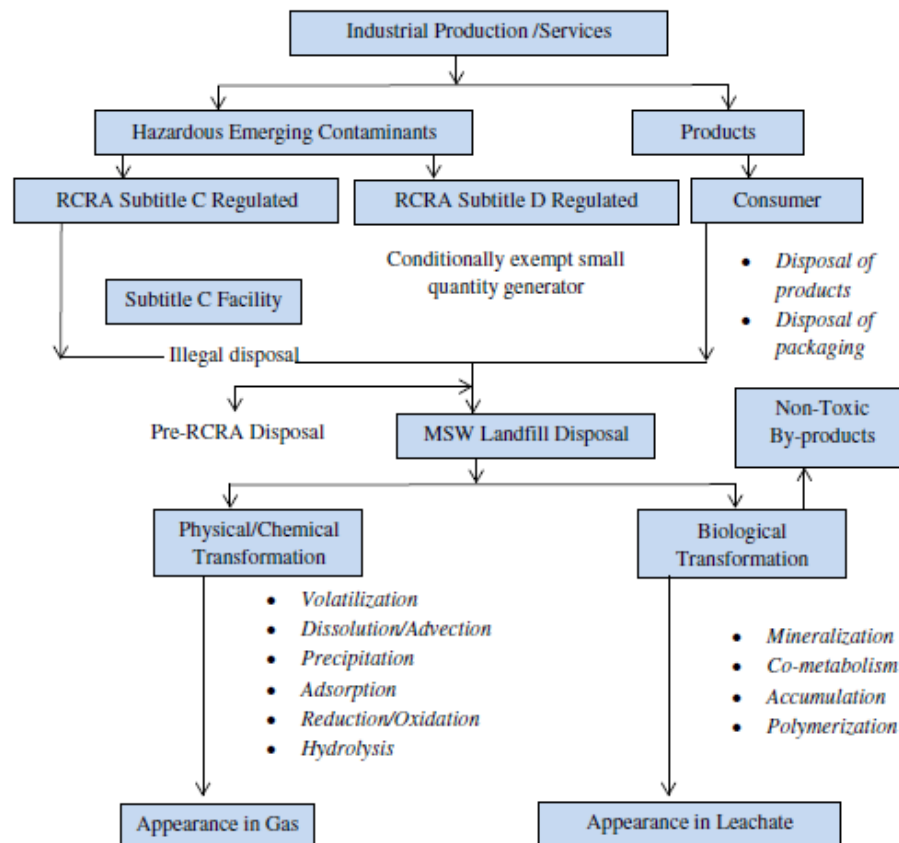


Figure 2.1: Origin, pathway, fate, and transformation of emerging contaminants along with municipal solid waste (MSW) and hazardous pollutants in the US. Adapted from (Ramakrishnan *et al.*, 2015).

## 2.2 New Emerging Pollutants (NEPs)

### 2.2.1 Definition

The understanding around the term New emerging pollutants (NEPs) has been rapidly evolved around landfill leachate. In the nearest decade, the first review article

utilising the term NEPs in municipal landfill leachate is traced back to 2010 when Trine Eggen defined the term as additives with unique beneficial improvement properties in daily use that bring negative environmental impacts after decomposition (Eggen *et al.*, 2010).

Ramakrishnan (2015) has first introduced another term that upholds a similar meaning which is emerging contaminants (ECs). The author provides another easy understanding definition towards NEPs which are categories of contaminants that have recently been shown to occur widely in the environment and have attracted the serious concern of society. These substances are identified as being a possible environmental or public health threat, but yet to have sufficient data to determine their effect (Ramakrishnan *et al.*, 2015).

The term Contaminants of emerging concern (CEC) is also used to refer NEPs. The US Geological Survey suggested that CECs are “any synthetic or naturally occurring chemical that is not commonly monitored in the environment but has the potential to enter the environment and cause known or suspected adverse ecological and/or human health effects” (Qi *et al.*, 2018). Jason R. Masoner stated in his paper in 2016 whereby the effect of CECs occurring in the environment has been recognised as a global phenomenon, and there is still much to be understood regarding these chemicals. Despite that, exposure to CECs has been linked to negative effects on ecosystem health, according to an increase in number of evidence (Masoner *et al.*, 2016).



### 2.2.2 Type

Over the last few decades, many of the NEPs detected in landfills are xenobiotic organic compounds, including per- and polyfluoroalkyl substances (PFAS), organophosphate esters (OPE), bisphenols, and various pharmaceuticals (Propp *et al.*, 2021). By tracing back to the year 2012, Williams J. Andrews found out that leachate that has been deposited at landfills potentially contain many organic compounds found in consumer products such as pharmaceuticals, plasticisers, disinfectants, cleaning agents, fire retardants, flavorings, and preservatives, known as NEPs (Andrews *et al.*, 2012).

In 2014, the dominant categories of NEPs included pharmaceuticals and personal care products, surfactants and their degradation products, plasticisers, pesticides, fire retardants, pesticides, and nanomaterials are discussed in particular in the paper “Emerging contaminants in landfill leachate and their sustainable management” (Ramakrishnan *et al.*, 2015). This categorisation is applied similarly in 2016, in Yu. N. Vodyanitskii’s research where he integrated the NEPs into class such as organic chemicals of industrial, agricultural and domestic use, plasticisers, surfactants, personal care products, and pharmaceuticals (Vodyanitskii and Yakovlev, 2016). In the same year, a total of 129 out of 202 NEPs were found out during Jason. R. Masoner’s study, including prescription pharmaceuticals, industrial chemicals, non-prescription pharmaceuticals, household chemicals, steroid hormones, and plant/animal sterols (Masoner *et al.*, 2016).

Over the past two decades, PAEs and PPCPs were the 2 most frequently studied NEPs classes in landfill leachate China, followed by APEOs/BPA, sPAHs, PFCs and FRs while OCPs, cVMS and sterol are the infrequent ones (Qi *et al.*, 2018). Apart from

that, the New emerging pollutants studied in Victoria R. Propp's research included artificial sweeteners (ASs), PFAS, organophosphate esters (OPE), pharmaceuticals, bisphenols, sulfamic acid, perchlorate, and substituted phenols (Propp *et al.*, 2021).

On top of that, Microplastics (MPs) are new contaminants that have been found in large quantities in both aquatic and terrestrial ecosystems. Landfills are containers for a collective load of plastic waste from industry and households, but there is no research on MPs occurrences in landfill systems (Su *et al.*, 2019). Furthermore, the existence of nanoparticles (NPs) as a kind of NEPs is undeniable. This is because the incorporation of these NPs into the solid waste stream is unavoidable as NPs are gradually introduced into consumer goods with coating, surface modified or to achieve specific properties (Bolyard *et al.*, 2013).

### **2.2.3 Composition**

For environmental and public health concerns, it is important to evaluate the chemical composition and genotoxic potential of chemicals discharged from industrial and domestic sources such as sanitary landfills (Gajski *et al.*, 2012). The chemical composition of NEPs compounds can affect the basic parameters of landfill leachate, such as pH, COD, BOD, TOC, Kjeldahl Nitrogen, heavy metals and organic compounds.

Leachate nowadays may contain chemicals such as humic and fulvic acids, phthalic esters, pesticides, and several other (emerging) organic micropollutants (perfluorinated compounds-PFCs, pharmaceuticals and personal care products, polyaromatic hydrocarbons-PAHs), inorganic compounds (chloride, sulfate, bicarbonate and carbonate, sulfide species, alkali and alkaline earth metals, iron and

manganese), high concentration of nitrogen compounds and heavy metals (Silva *et al.*, 2013). The basic composition of leachate is grouped into 4 categories of pollutants (Ghosh, *et al.*, 2017).

Table 2.1: List of categories of pollutants and their compounds

Category	Expressed as	Compounds
Dissolved organic matter	Chemical Oxygen Demand (COD) Total Organic C (TOC)	CH <sub>4</sub> Volatile fatty acids More refractory compounds (fulvic-like & humic-like compounds)
Inorganic macro components	-	Ca <sup>2+</sup> , Mg <sup>2+</sup> , Na <sup>+</sup> , K <sup>+</sup> , NH <sub>4</sub> <sup>+</sup> , Cl <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , HCO <sub>3</sub> <sup>-</sup>
Heavy metals	-	Ca, Na, Mg, K, Fe, Mn, As, Cd, Co, Cr, Cu, Hg, Ni, Pb, Zn
Xenobiotic organic compounds	-	Aromatic hydrocarbons Phenols Chlorinated aliphatics

Table 2.2: List of New emerging pollutants (NEPs) classes and their compounds

Sources	Classes	Compounds
(Qi <i>et al.</i> , 2018)	PAEs	di-n-butyl phthalate di (2-ethylhexyl) phthalate dimethyl phthalate diethyl phthalate
	PPCPs	Sulfadiazine Sulfamethoxazole Sulfamethazine Erythromycin Trimethoprim
	APEOs/BPA	Bisphenol A
(Kapelewska <i>et al.</i> , 2018)	Parabens	Mehtylparaben Ethylparaben Propylparaben Buthylparaben
	Sunscreen agents and insect repellent	Benzophenone Benzophenone 2 Benzophenone 3 3-(4-methylbenzylidene) camphor N,N-diethyltoluamide (DEET)
	Phenols	4-n-Octylphenol 4-n-Nonylphenol Bisphenol A Triclosan

	Pharmaceuticals and hormones	Diclofenac Carbamazepine Estrone β-Estradiol Diethylstilbestrol
(Masoner <i>et al.</i> , 2016)	Household chemical	Acetophenone Benzophenone Bisphenol A (BPA) Camphor D-Limonene Ethyl citrate Galaxolide Menthol N, N-Diethyltoluamide (DEET) Skatol Tri(2-chloroethyl) phosphate Tri(dichlorisopropyl) phosphate Tributylphosphate Triclosan
	Industrial chemicals	1,4-Dichlorobenzene 1-Methylnaphthalene 2-Methylnaphthalene 4-Cumylphenol 4-Nonylphenol 4-Nonylphenol monoethoxylate 4-Nonylphenol diethoxylate 4-tert-Octylphenol 4-tert-Octylphenol monoethoxylate 4-tert-Octylphenol diethoxylate Anthracene Bromoform Diethyl phthalate Diethylhexyl phthalate Isophorone Isopropylbenzene Methyl-1H-benzotriazole Naphthalene Para-cresol Pentachlorophenol Phenantrene Phenol Triphenyl phosphate
	Non-prescription pharmaceuticals and degrades	1,7-Dimethylxanthine Acetaminophen Caffeine Chloroxylenol Chlorpheniramine Cimetidine Cotinine Dextromethorphan

		Diphenhydramine Famotidine Fexofenadine Ibuprofen Lidocaine Loratadine Nicotine Piperonyl butoxide Pseudoephedrine Ranitidine
	Pesticides and degradates	Atrazine Carbaryl
	Plant and animal sterols	3-beta-Coprostanol beta-Sitosterol Cholesterol Stigmastanol
	Prescription pharmaceuticals and degradates	10-Hydroxy-amitriptyline Acyclovir Albuterol Amphetamine Antipyrine Atenolol Bupropion Carbamazepine Carisoprodol Codeine Dehydronifedipine Diltiazem Duloxetine Erythromycin Fenofibrate Fluconazole Fluvoxamine Glipizide Glyburide Lamivudine Loperamide Lorazepam Meprobamate Metaxalone Metformin Methadone Methocarbamol Metoprolol Morphine Nadolol N-Desmethyldiltiazem Orlistat Oseltamivir Oxazepam

		Oxycodone Paroxetine Pentobarbital Pentoxifylline Phenazopyridine Phendimetrazine Phenytoin Prednisolone Prednisone Primidone Propoxyphene Quinine Raloxifene Sulfadimethoxine Sulfamethizole Sulfamethoxazole Tamoxifen Temazepam Theophylline Thiabendazole Tramadol Triamterene Trimethoprim Venlafaxine Verapamil Warfarin
	Steroid hormones	17-beta-Estradiol cis-Androsterone Dihydrotestosterone Epi-testosterone Estriol Estrone
(Kapelewska <i>et al.</i> , 2018)	Parabens	Methylparaben (MP) Ethylparaben (EP) Propylparaben (PP) Butylparaben (BP)
	Sunscreen agents and insect repellent	Benzophenone (BPh) Benzophenone 2 Benzophenone 3 3-(4-methylbenzylidene) camphor (4-MBC) N,N-diethyltoluamide (DEET)
	Phenols	4-n-Octylphenol 4-n-Nonylphenol Bisphenol A Triclosan
	Pharmaceuticals and hormones	Diclofenac Carbamazepine Estrone β-Estradiol

		Diethylstilbestrol
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### **2.3 New Emerging Pollutants (NEPs) removal in leachate management**

Even if landfilling is recognised as an economical and easy operation, one of the most difficult problems to address is leachate disposal, as it can be emitted from the start of landfill operations until decades after closure (Banch *et al.*, 2019). Unfortunately, leachate is rarely contained in older landfills and those without liners, and it is more likely to slowly percolate into the groundwater.

There are two main strategies for minimising the amount of NEPs in landfill leachate and avoiding downstream impacts from the viewpoint of environmental sustainability. This includes disposal of landfill leachate to minimise a load of NEPs and reduction in the mass of disposed of items containing NEPs (Ramakrishnan *et al.*, 2015). A future perspective on leachate management includes treatment of leachate, legislation, disposal and handling of leachate and source control.

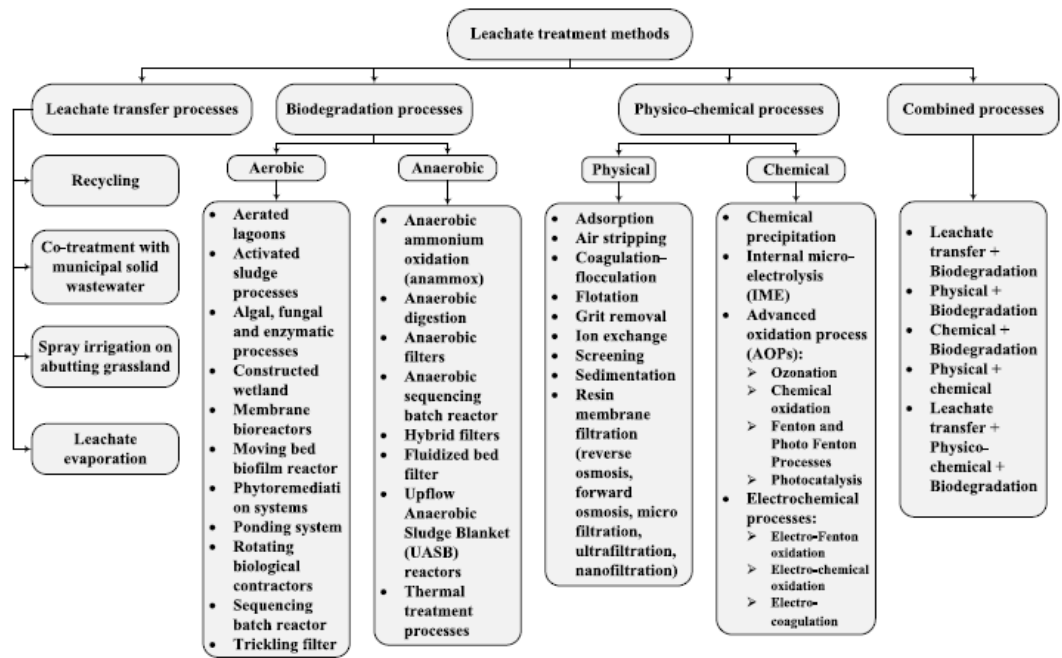


Figure 2.2: Leachate recovery, reuse and degradation methods. Adapted from (Jagaba *et al.*, 2021)

For newly developed landfills and those with liners, the leachate is collected and treated on-site with traditional treatment (biological and physical/chemical treatment), membrane treatment, or transported off-site to a sewage treatment facility (Qi *et al.*, 2018). Coagulation, flocculation, reverse osmosis, ion exchange, filtration, precipitation, ozonation, and adsorption are indeed some of the methods and processes used to treat landfill leachate (Reshadi *et al.*, 2020). However, the efficiency of leachate treatment has been questioned, especially in the case of products which that have a specialised waste treatment process (Marcoux *et al.*, 2013).

When compared to traditional methods, advanced treatment processes such as membrane treatment and advanced oxidation will achieve higher removal efficiencies (up to 100%) for NEPs compounds (Qi *et al.*, 2018). According to research done in 2017, it is found out that aged refuse bioreactors had a lot of potential for removing antibiotics and ARGs from leachate, however highly dependant on reactor configuration, operating conditions and types of antibiotic and ARGs (Su *et al.*, 2017).



Apart from that, membrane bioreactor (MBR) with post-treatment reverse osmosis (RO) and nanofiltration (NF) is commonly effective for the removal of PFAAs from the aqueous phase (Yan *et al.*, 2015). This statement is also proven in 2017, whereby the implication of full-scale membrane bioreactor (MBR) in landfill leachate treatment can largely reduce the concentration of specific individual PPCPs (Sui *et al.*, 2017).

### 2.3.1 Membrane bioreactor (MBR)

Membrane bioreactor (MBR) has been shown to be a suitable and effective choice for leachate treatment. The membrane bioreactor has 3 activated sludge tanks and an external ultrafiltration unit. Apart from that, pre-denitrification and two-stage nitrification are used in the membrane bioreactor configuration (Wintgens *et al.*, 2014).

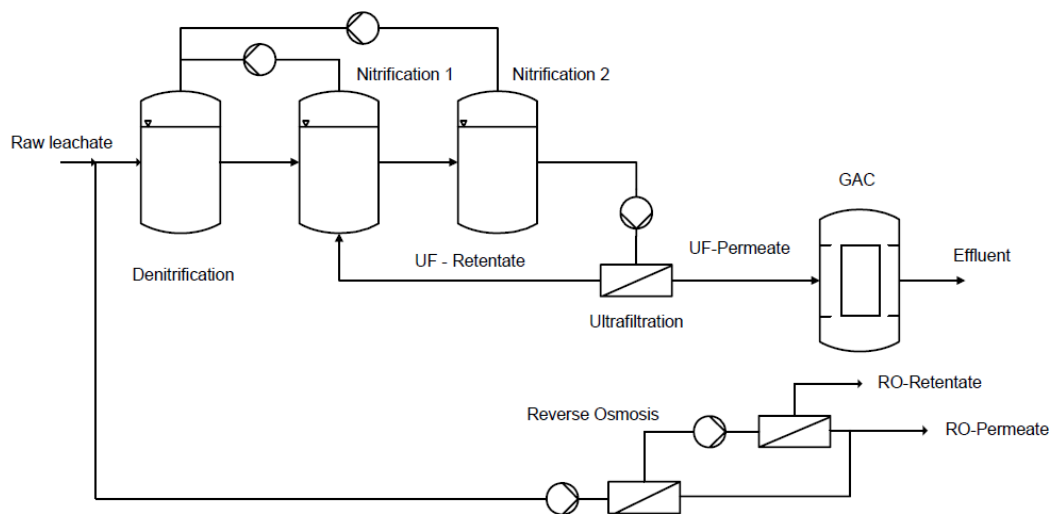


Figure 2.3: Membrane bioreactor configuration at a landfill leachate treatment plant. Adapted from (Wintgens *et al.*, 2014)

A combined anaerobic-membrane bioreactor system effectively removed organochlorine pesticides, PAHs, and 4-nonylphenol (Xu *et al.*, 2008). A submerged hollow fibre membrane bioreactor was used to treat landfill leachate with pH = 7.4–7.8;

conductivity = 8,000–10,000 mg/L; COD = 1,400–2,800 mg/L; BOD<sub>5</sub> = 650–1,300 mg/L; diacetone alpha-keto-gulonic acid (DAG) = 80–430 lg/L; diacetone sorbose (DAS) = 1,420–2,570 lg/L; and propyphenazone= 85–130 lg/L. Its COD removal efficiency was reported as 23% whereas DAG, DAS, and propyphenazone were approximately 69, 29, and 17 % respectively (Ramakrishnan *et al.*, 2015).

### **2.3.2 Coagulation and flocculation**

Because of its simplicity and efficacy, the coagulation-flocculation process is one of the most essential and commonly used leachate treatment procedures. This treatment only achieves moderate COD and TOC removal, and it has drawbacks: sludge is formed, and when typical chemical coagulants are used, an increase in the concentration of aluminium or iron in the liquid phase may be noticed (Silva, Dezotti and Sant'Anna, 2004). Coagulation and flocculation process with different coagulant can have effects on the removal of COD, turbidity and suspended solids which contributed to the colour of landfill leachate (Aziz *et al.*, 2007).

However, research showed that dissolved organic macromolecules (DOM) and micropollutants, such as phthalic acid esters (PAEs) in landfill leachate, can be treated using a complexation-flocculation process. Among the tested coagulants (ferric chloride, aluminium sulfate and poly aluminium chloride (PAC)), PAC was the most efficient For removing DOM and PAEs (Zhang and Wang, 2009).

### **2.3.3 Constructed wetland (CW)**

Landfill leachate is often processed at wastewater treatment plants, which necessitates transportation and treatment that is relatively costly. However, in many

circumstances, landfill leachate may be treated more efficiently and cost-effectively locally in the landfill area. The constructed wetland treatment system is known as engineered systems that use natural processes to treat pollutants. It often consists of an equalization tank, aeration lagoons, sedimentation tank, reed beds and polishing ponds. Biodegradation (aerobic, anoxic, anaerobic), phytoremediation (plant uptake, biodegradation enhancement), and sorption (adsorption, absorption) are all the included components in CWs (Garcia-Rodríguez *et al.*, 2014).

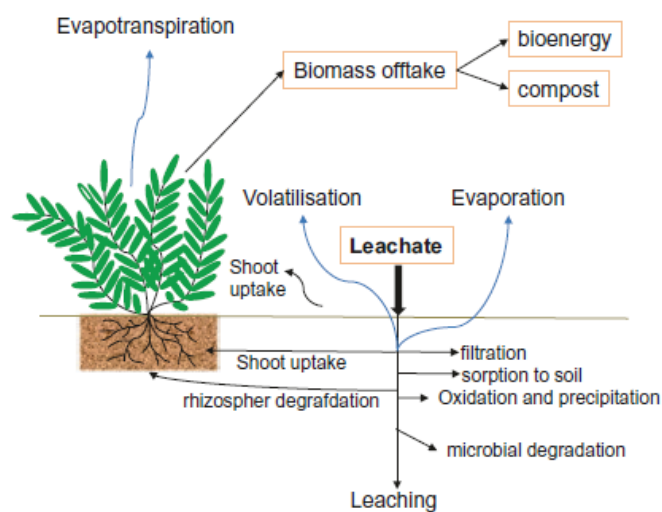


Figure 2.4: Treatment process and mechanisms during landfill leachate phytoremediation. Adapted from (Jones, Williamson and Owen, 2006)

Many NEPs, such as pesticides, pharmaceutical and personal care products (PPCPs), and endocrine-disrupting chemicals, are thought to be cost-effectively removed using constructed wetlands (CWs). The overall removal of PPCPs and EDCs in the hybrid CW system were greater than 77.2%. Several compounds in particular such as BPA, CA and SMZ, had excellent removal efficiencies (>99.99%) (Yi *et al.*, 2017). This is further supported by Yin's research, where the researcher found out that 61% of total PFAS and 50-96% of individual PFAS are removed by the CW treatment system (Yin *et al.*, 2017).

### 2.3.4 Nitrification

The nitrification/denitrification process involves the elimination of ammonium with microbial. During the traditional nitrification-denitrification process, ammonia is converted to nitrate under aerobic conditions, which is then reduced to  $N_2$  under anoxic conditions (Thakur and Medhi, 2019). The nitrification process is suitable for the treatment of age landfill leachate with rich nitrogen concentrations and poor  $BOD_5/COD$  ratios ( $<0.2$ ) to remove ammonium and degrade some organic material (Gao *et al.*, 2015).

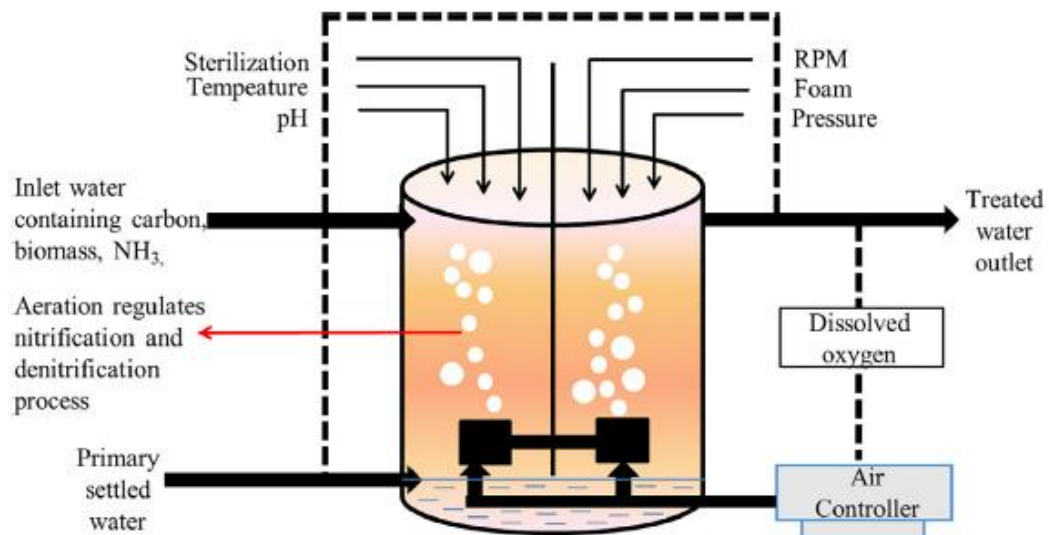


Figure 2.5: Schematic diagram for a lab-scale bioreactor for monitoring operational conditions during nitrogen removal. Adapted from (Thakur and Medhi, 2019).

When leachate undergoes a series of a process combining nitrification/anaerobic/ozonation has achieved a better removal efficiency of methamphetamine (79%), ketamine (85%) and MDMA (60%). (Lu *et al.*, 2016) According Qi's study, nitrification combining by the anaerobic process can achieve a removal efficiency of PPCPs up to 85% (Qi *et al.*, 2018).

### 2.3.5 Advanced Oxidation Process (AOP)

Advanced Oxidation Processes (AOPs) are a subset of oxidation procedures that have a common chemical feature: the generation of OH radicals (Gao *et al.*, 2015). The high oxidative capability and efficiency of AOPs make it a common tertiary treatment technique for removing the most recalcitrant organic and inorganic pollutants. The AOP process combining with biological processes have proven a successful conversion of recalcitrant contaminants such as pharmaceuticals, pesticides and new emerging organic micropollutants into smaller and more biodegradable components (Gao *et al.*, 2015).

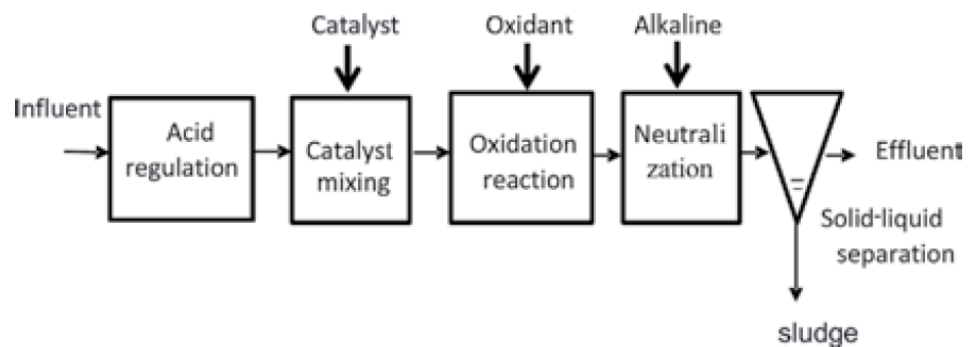


Figure 2.6: Schematic diagram for Fenton process. Adapted from (Bustillo-lecompte, 2020)

As demonstrated by a variety of lab and industrial-scale operations, ozone oxidation has a wide range of uses in the treatment of landfill leachate. For ozonation procedures with 1.24 mg O<sub>3</sub>/mg COD, conversion efficiencies of DAG, DAS, and propyphenazone in landfill leachate were 11, 20, and 99%, respectively; note that COD removal was around 56% (Ramakrishnan *et al.*, 2015). The fenton-oxidation process is also one of the popular processes in removing pollutants in landfill leachate. In Qi's study, the Fenton process is targeted to remove BPA, and PAEs compounds up to 20%-100% removal efficiency (Qi *et al.*, 2018). Besides that, an oxidation ditch with granular

activated carbon can achieve a better benzophenone removal of up to 80% (Lu *et al.*, 2016).

### 2.3.6 Adsorption-Activated carbon

Adsorption of traditional adsorbents, including activated carbon, has been commonly used with significant results (Reshadi *et al.*, 2020). Adsorption is a surface phenomenon that occurs whereby organic and inorganic substances are removed. Physical and chemical bonds formed, and species are attracted to the surface of a highly porous structure. In practice, activated carbon adsorption is frequently utilised as a stage in a leachate treatment train that includes chemical, physical, and biological components (Gao *et al.*, 2015).

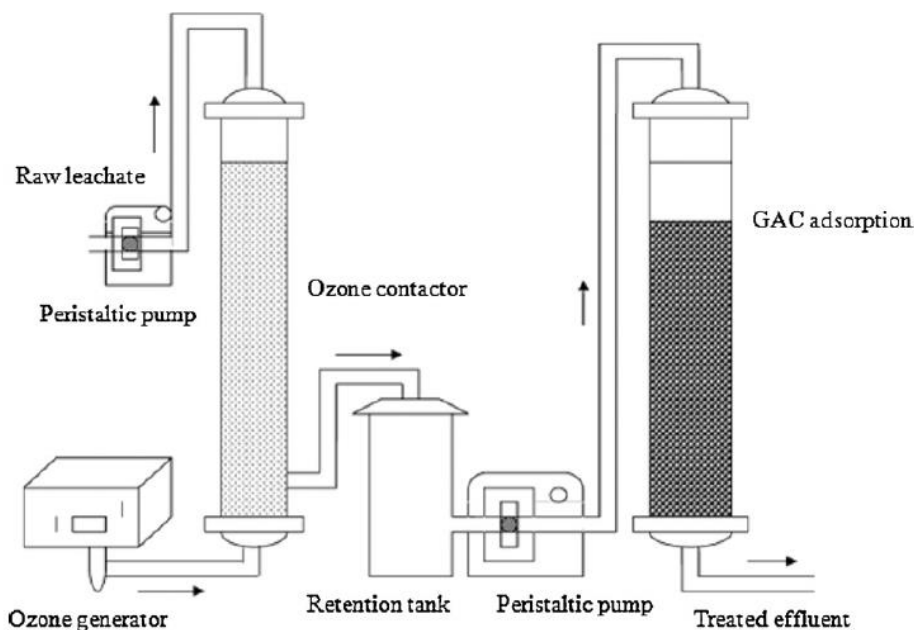


Figure 2.7: Schematic diagram of the integrated ozone – GAC adsorption system. Adapted from (Foo and Hameed, 2009)

In the removal of PFAAs from leachate, an adsorption approach using activated carbon was reported to be relatively efficient (removal efficiency ranged between 70 and 99%) (Hamid *et al.*, 2018). There is also a study stating that the maximal adsorption

capacities of PFOS and PFOA on alumina, according to the sorption isotherms, are  $0.252 \mu\text{g}/\text{m}^2$  and  $0.157 \mu\text{g}/\text{m}^2$ , respectively (Wang and Shih, 2011). The high level in the occurrence of adsorption indicates that the possibility of such a method can be a potential treatment for NEPs.

### 2.3.7 Aged refuse bioreactors

The aged refuse reactor is a dynamic biological system with a wide variety of microbiota. As a result, a thorough study of the microbial population, antibiotics, and ARGs is critical for probing the dynamics of the leachate resistive (Su *et al.*, 2017).

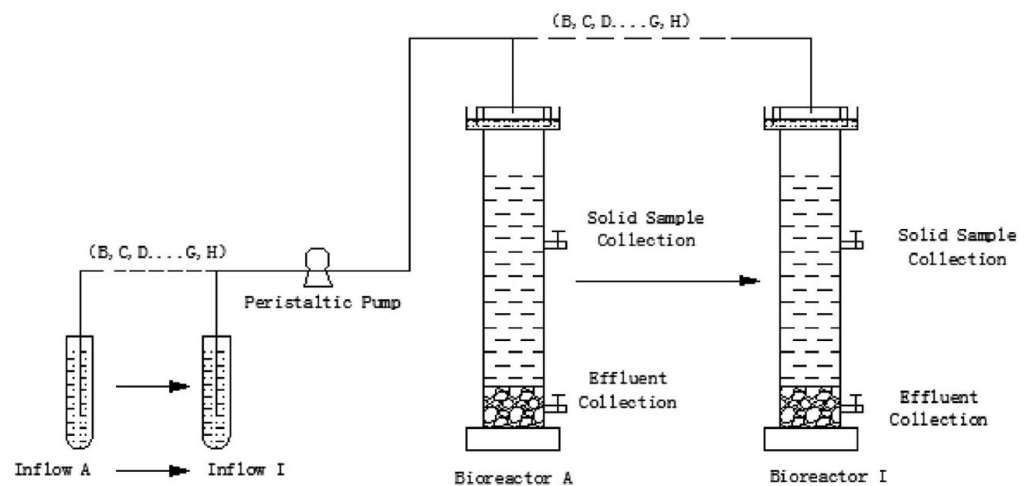


Figure 2.8: Schematic diagram of Aged Refuse Bioreactor. Adapted from (Wang *et al.*, 2017)

Antibiotics and antibiotic resistance genes (ARGs) were removed from leachate using an old refuse bioreactor, and the removal efficiency was excellent. The total removal efficiency for the detected antibiotics was around 76.75%, with sulfanilamide and macrolide removal efficiencies exceeding 80% (Su *et al.*, 2017). According to Qi's study, three-stage horizontal and tower aged refuse bioreactors can efficiently achieve 48.4 to 76.8% removal of PPCPs (Qi *et al.*, 2018).

### 2.3.8 Membrane filtration

Various techniques are used in membrane filtration as landfill leachate treatment, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO).

Table 2.3: Type of membrane filtration, its targeted group and its characteristics.  
Adapted from (Gao *et al.*, 2015)

Type	Targeted group	Characteristics
Microfiltration (MF)	microorganisms, small particles, large molecules, emulsion droplets and large colloidal	Used as pretreatment process with other membrane processes. Combined with chemical treatment processes.
Ultrafiltration (UF)	larger molecular weight components of leachate (organic biodegradable macromolecules and non-biodegradable ones)	Separation dependant on particle size and molecular weight cut-offs (MWCO) Pretreatment method for reverses osmosis.
Nanofiltration (NF)	Recalcitrant organic compounds and heavy metals	Looser membrane structure, enabling higher fluxes and lower operating pressure. Remove of particles by electrostatics interactions
Reverse osmosis (RO)	heavy metals, suspended/colloidal materials and dissolved solids	Can be a main step or a single post-treatment step High fluxes and the ability to operate over a wide temperature and pH range

Apart from that, that nanofiltration could be used to treat landfill leachate with pH = 7.39–7.84, conductivity = 7,560–9,690 IS/m, COD = 574–803 mg/L, and total pharmaceutical mass concentration = 27–55 mg/L. It was able to achieve more than 90% rejection of COD and three pharmaceuticals intermediate. Membrane systems have a high efficacy in treating NEPs in landfill leachate, regardless of whether or not additional treatment of the reject stream is necessary (Ramakrishnan *et al.*, 2015). >95% of PFAAs were removed from leachate and biologically treated leachate (e.g.,



membrane bioreactor, followed by RO or NF) using high-pressure membrane filtration techniques such as reverse osmosis (RO) and nanofiltration (NF) (Hamid *et al.*, 2018).

## **2.4 Overview**

This literature audit has given insights into the researchers' understanding of the terminology of NEPs in landfill leachate and their associated treatment method. This is significant because various drawbacks and challenges on NEPs' determination have been identified and clarified. Numerous topics and study areas that focuses on the presence of NEPs in landfill leachate and the application of various technologies for the removal of NEPs group found in landfill leachate has been gaining attractions from researchers around the world.

Over the years, there has been so much research and discussion conducted on the NEPs in wastewater, including their respective treatment technique in the wastewater treatment plant. Many treatment methods that currently applicable in the treatment of landfill leachate are adaptations of wastewater treatment methods. It is important to conduct more extensive studies to stimulate and model the overall treatment processes that target all kinds of NEPs found specifically in landfill leachate. Every individual physical/chemical, or biological systems should be designed and modified with targeted NEPs removal in landfill leachate. In terms of leachate management, there is a lack of concrete pre-emptive control measures employed to manage NEPs in landfill leachate. Through extensive literature review, it is noted that there is a lack of preventive measures such as legal directives, recycling programmes and controlling environmental pollution sources. Nevertheless, researchers should consider not just the technical aspects of their findings but also the viability of putting them into practice from both an environmental and economic standpoint.