Mitigation of hazards and risks of emerging pollutants through innovative treatment techniques of post methanated distillery effluent - A review

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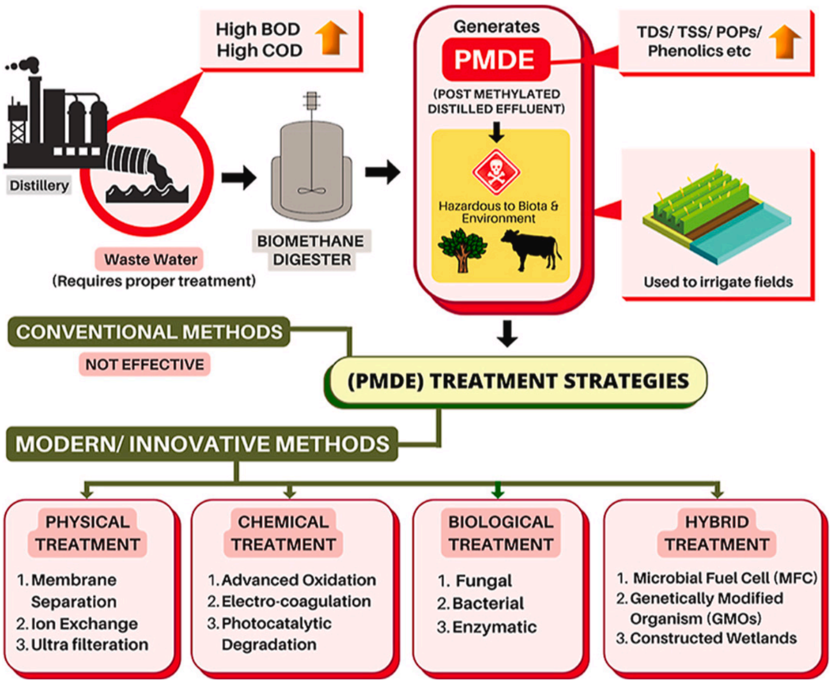
Highlight

- PMDE toxicity to ecosystems and human health examined.
- PMDE treatment using advanced membrane separation technologies presented.
- Advanced oxidation, electrocoagulation, photo & sono catalytic degradation reviewed.
- Biological treatment e.g., enzyme treatment of PMDE discussed.
- Hybrid treatment e.g., MFC, GMO and constructed wetland are evaluated.

Graphical Abstract



Abstract

Distillery wastewater has high biological and chemical oxygen demand and requires additional treatment before it can be safely discharged into receiving water. It is usually processed through a biomethanation digester and the end product is the post-methanated distillery effluent (PMDE). Research have shown that PMDE released by molasses-based distilleries is a hazardous effluent that can cause harm to the biota and the environment; it contains elevated amount of total dissolved solids (TDS), total suspended solids (TSS) and excess levels of persistent organic compounds (POPs), heavy metals, phenolic compounds, and salts. The practice of wastewater reuse for irrigation in many water scarce countries necessitates the proper treatment of PMDE before it is discharged into receiving water. Convention methods have been in practice for decades, but innovative technologies are needed to enhance the efficiency of PMDE treatment. Advance physical treatment such as membrane separation technology using graphene, ion-exchange and ultrafiltration membranes; chemical treatment such as advanced oxidation methods, electrocoagulation and photocatalytic technologies; biological treatment such as microbial and enzymatic treatment; and hybrid treatment such as microbial-fuel cell (MFC), genetically modified organisms (GMO) and constructed wetland technologies, are promising new methods to improve the quality of PMDE. This review provides insight into current accomplishments evaluates their suitability and discusses future developments in the detoxification of PMDE. The consolidated knowledge will help to develop a better management for the safe disposal and the reuse of PMDE wastewater.

Keywords

Distillery industry, Advanced oxidation, Membrane filters, Catalysis, Innovative treatment techniques

1. Introduction

According to The Central Pollution Control Board of India (CPCB) (http://www.cpcb.nic.in), there are about 400 distilleries in India with a combined production capacity of around ~4 billion L of alcohol. Molasses-based distilleries generate ~15–20 L of wastewater per L of ethanol produced, with high biological oxygen demand (BOD) (8–10,000 mgL^{-1}), chemical oxygen demand (COD) (40–45000 mg L^{-1}), and a dark brownish colour effluent (Tripathi et al., 2021a, b). The dark brown colour is primarily attributed to melanoidin, an intricate biopolymer produced by the non-enzymatic maillard reactions during the fermentation production process of sugarcane molasses in distilleries (de Salles Pupo et al., 2020; Santal et al., 2016). The wastewater is usually processed through a biomethanation digester to lower the high BOD and COD, and the end product is the post-methanated distillery effluent (PMDE) (Naik et al., 2010). PMDE is a complex effluent generated after the treatment of spent wash in an anaerobic digestion process. It contains persistent organic compounds, heavy metals, and phenolics compounds; it has high levels of total dissolved solids (TDS), total suspended solids (TSS), BOD, COD, and sulfate (Chaturvedi et al., 2021; Tripathi et al., 2021a,b; Pereira et al., 2021; Mikucka and Zielinska, 2020´ ; Neogi et al., 2021). The resulting high redox potential is harmful to the biota and causes serious environmental pollution. Wastewater is a valuable resource for Indian farmers, especially in arid regions, for crop irrigation. Many livestock also drink from river or body of water that receives distillery discharge. Extra treatment is therefore essential to ensure the safe disposal and reuse of PMDE. The is a need to evaluate different advanced technologies and understand their pros and cons in treating PMDE (Pereira et al., 2021; Iqbal et al., 2019; Bhat et al., 2019; Mazzeo et al., 2018; Leme and

Marin- Morales, 2009). This review aims to provide state-of-the-art information on the advanced treatments of molasses-based wastewater, knowledge of these innovative technologies will lead to more efficient and the environment.

2. Post methanated distillery effluent (PMDE): properties and composition

The raw material of distillery is sugar cane molasses produced from citrus, wood sugar, candy and sugarcanes; it is a dark, viscous by- product of the cane sugar refined process. It contains reduced polymer sugars, which can react further during enzyme hydrolysis to form fermentable sugar. In normal caramel molasses, the moisture content is generally between 17 and 25%, sugar (sugar, glucose and fructose) between 40 and 50% and polysaccharides (dextrin, pentosans, polyuronic acids) between 2% and 5%. Molasses contains sucrose (480–520 g L^{-1}), glucose (80–120 g L^{-1}), fructose (80–120 g L^{-1}), polyphenols (16–25 g L^{-1}), antioxidants (5–9 g L^{-1}), calcium (2003 mg L^{-1}), magnesium (1118 mg L^{-1}), sodium (114 mg L^{-1}), and potassium (8002 mg L^{-1}). PMDE origination and mechanism is presented in Fig. 1.

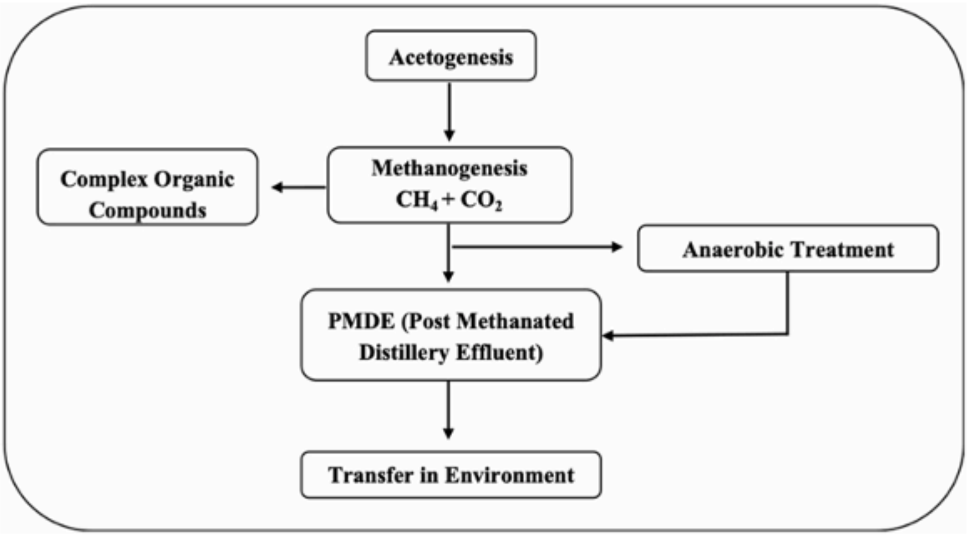
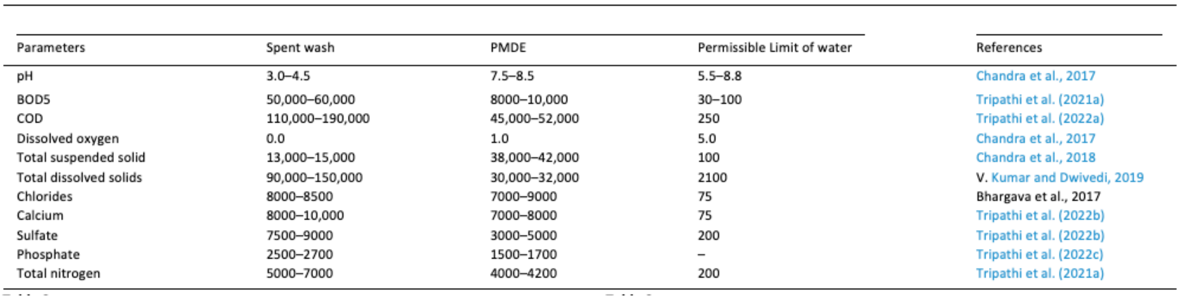


Fig. 1. PMDE origination & mechanism involved before its discharge in environment.

In the distillery process, sucrose from the molasses is transformed biologically into cell energy with ethanol and carbon dioxide as by products. Sugarcane or sugar molasses imparts an ethanol-producing substratum (Ummalyma et al., 2018). Sugar molasses is the primary source of sucrose in concentrated sugar ($C_{12}H_{22}O_{11}$). Only 50% of the molasses contains reducing sugars, the remaining contents are nitrogen, phosphorus, solid sediment and ash remnants of fermentable and unfermented sugars. In molasses, fermentable sugars are used directly, while fermentable sugars such as starch, cellulose, or pectin may not be used until hydrolysed. Ethanol is formed by the application of enzymes and reactions (Ali et al., 2020; Ashok kumar et al., 2019; Chaurasia et al., 2020; Han et al., 2020; Lakshmikandan et al., 2020; Mao et al., 2020; Zabed et al., 2020). It is used in the form of absolute or remedied spirit that is baptised from the entire process. Molasses is manufactured in huge amount and disposed of in landfills during fermentation and distillation. Further, rectified spirit and anhydrous ethanol etc. are produced post distillation (Chandra et al., 2018). Physical or chemical methods are costly to treat distillery wastewater and the resulting secondary pollutants are highly toxic to aquatic life. The spent wash and left-over biomasses distillery wastewater are usually treated using biological methods such as anaerobic digestion involving archaeal, a phylogenetically distinct group of microbes that produces methane (Santal et al., 2016; Krzywonos et al., 2017; Chandra et al., 2018; Wilk et al., 2019). These bacteria are extremely oxygen sensitive and require anaerobic conditions. The formation of methane by methanogens is known as methanogenesis or biomethanation. In the anaerobic digestor, the spent wash and biomass undergo several stages: hydrolysis, acidogenesis, acetogenesis and methanogenesis (Fig. 1). When the process of methanogenesis is complete, the PMDE is discharged. The PMDE contains heavy metals, persistent organic pollutants (POPs), phenolics compounds and sulfides which contributes to the toxicity of the effluent and inhibit PMDE biodegradation during their tertiary treatment. PMDE features intense brown color, high BOD, COD, salts and plant-rich nutrients (Table 1). Despite areduction in BOD load from approximately 50,000 mgL^{-1} to 8000–5000 mg L^{-1}, further treatment is still required. The suspension solids in the effluent also impart water turbidity, reduce penetration of sunlight and impairment of the biological activities of aquatic life. An economically viable and environmentally safe disposal method is therefore necessary for dealing with the large volumes of PMDE (Krzywonos et al., 2017). Properly treated effluent may be reused to irrigate agricultural soils, surfaces and soil water in controlled condition

Table 1. Properties of spent wash and an aerobically treated distillery effluent (PMDE).



Parameters	Spent wash	PMDE	Permissible Limit of water	References
pH	3.0–4.5	7.5–8.5	5.5–8.8	Chandra et al., 2017
BOD5	50,000–60,000	8000–10,000	30–100	Tripathi et al. (2021a)
COD	110,000–190,000	45,000–52,000	250	Tripathi et al. (2022a)
Dissolved oxygen	0.0	1.0	5.0	Chandra et al., 2017
Total suspended solid	13,000–15,000	38,000–42,000	100	Chandra et al., 2018
Total dissolved solids	90,000–150,000	30,000–32,000	2100	V. Kumar and Dwivedi, 2019
Chlorides	8000–8500	7000–9000	75	Bhargava et al., 2017
Calcium	8000–10,000	7000–8000	75	Tripathi et al. (2022b)
Sulfate	7500–9000	3000–5000	200	Tripathi et al. (2022b)
Phosphate	2500–2700	1500–1700	–	Tripathi et al. (2022c)
Total nitrogen	5000–7000	4000–4200	200	Tripathi et al. (2021a)

3. Toxicity profile and environmental pollution of PMDE wastewater

The distillery industry's alcohol production generates a considerable volume of wastewater each day (Bharagava and Chandra, 2010; AIDA, 2004). Physicochemical indices such as BOD, COD, TSS, and microbiological pollution are all exacerbated by this effluent. The impact on human health is listed in Tables 2 and 3. For example, aniline, chlorpyrifos, and other organic pollutants including phthalic acid derivatives, hazardous metals such as arsenic, lead, chromium, antimony, are all included in PMDE. Melanoidins' pigments are coloured using heavy metals [DPI] (Rosa et al., 2020; Hubadillah et al., 2020). Axenic conditions for aquatic fauna and flora both can be caused by the transport of these pollutants along with wastewaters, which persist in the environment (water/soil) for a long period of time, posing severe health hazards to living organisms, and reducing soil fertility and photosynthetic activity of aquatic plants (Xue et al., 2019; Ashok kumar et al., 2019; Kang et al., 2020) (Fig. 2). Tables 2 and 3 presents the health impact of inorganic and organic pollutants present in PMDE respectively. A cost-effective and environmentally benign method for treating PMDE before it is disposed of in the environment is thus urgently required.

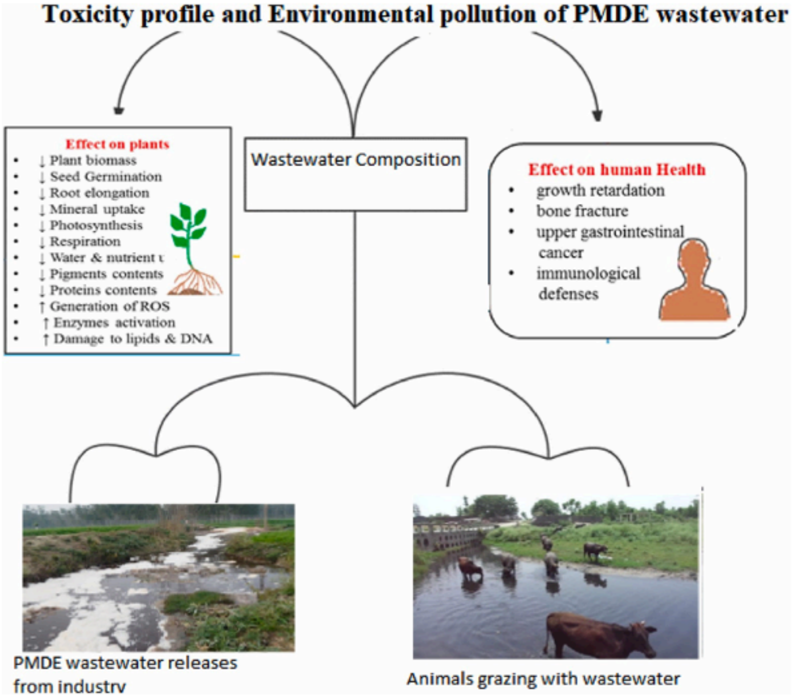


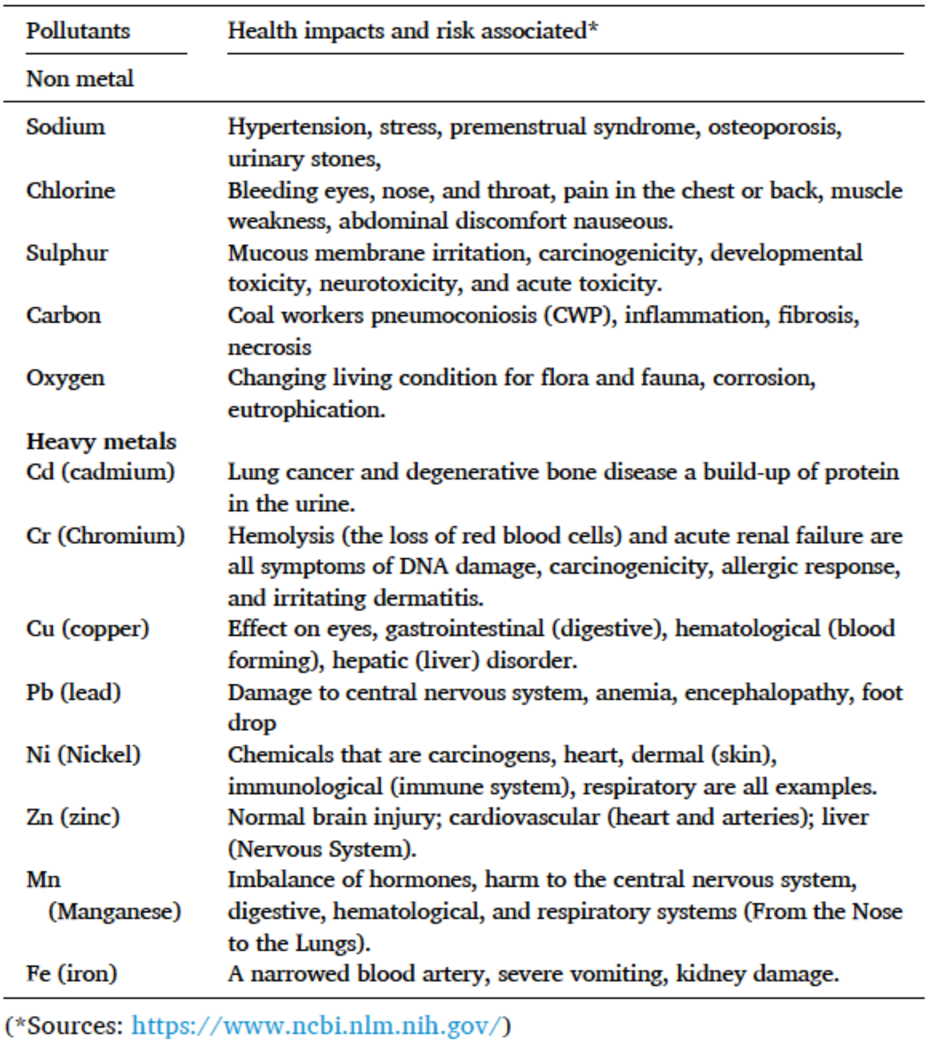
Fig. 2. Showing the PMDE industry effluent and their effects on animals and flora & fauna.

4. By-product of PMDE: melanoidin structure and mechanism

The by-product of PMDE (i.e., sugarcane molasses-based distillery wastewater) is the primary source of melanoidin. In addition, PMDE releases other mutagenic and carcinogenic complexes into the environment (Chowdhary et al., 2018a, b; Tripathi et al., 2021a). Melanoidins is a nitrogenous heterocyclic dark pigment polymer, it is produced by high-temperature chemical reactions between amino and carbonyl chemicals (Wang et al., 2011). The carbonyl groups of reducing sugars react with the amino groups of amino acids, peptides, and proteins to form these molecules. Melanoidin's fundamental skeleton is made up of carbohydrates and amino acids, according to prior research (Cãmmerer et al., 2002; Tripathi et al., 2022a, b, c). Melanoidin is the result of a three-step process. Sugar-amine condensation is the first step in the process. The Strecker degradation process is used to degrade the amino acid in the second step, which includes dehydrating and fragmenting the sugar. Through aldehyde amine condensation, brown-coloured heterocyclic nitrogenous chemicals are created towards the end of the second step of the Maillard reaction. Melanoidins are the high-molecular-weight molecules generated at this point (Liu et al., 2020). The majority of research on melanoidin as an antioxidant has focused on low-concentration foods including baked goods, cooked meat, roasted coffee, honey, and sweet wine (Shaheen et al., 2021).

During heating, roasting, grilling, and freezing, sugar and free amino groups are reduced, resulting in the creation of these compounds (Lund and Ray, 2017). However, various research has shown that excessive levels of melanoidin in the environment are a pollutant owing to its anionic characteristics and complexation with other environmental contaminants and heavy metals emitted from industrial operations (Chandra et al., 2017; Tripathi et al., 2021a, b, c, d, e).

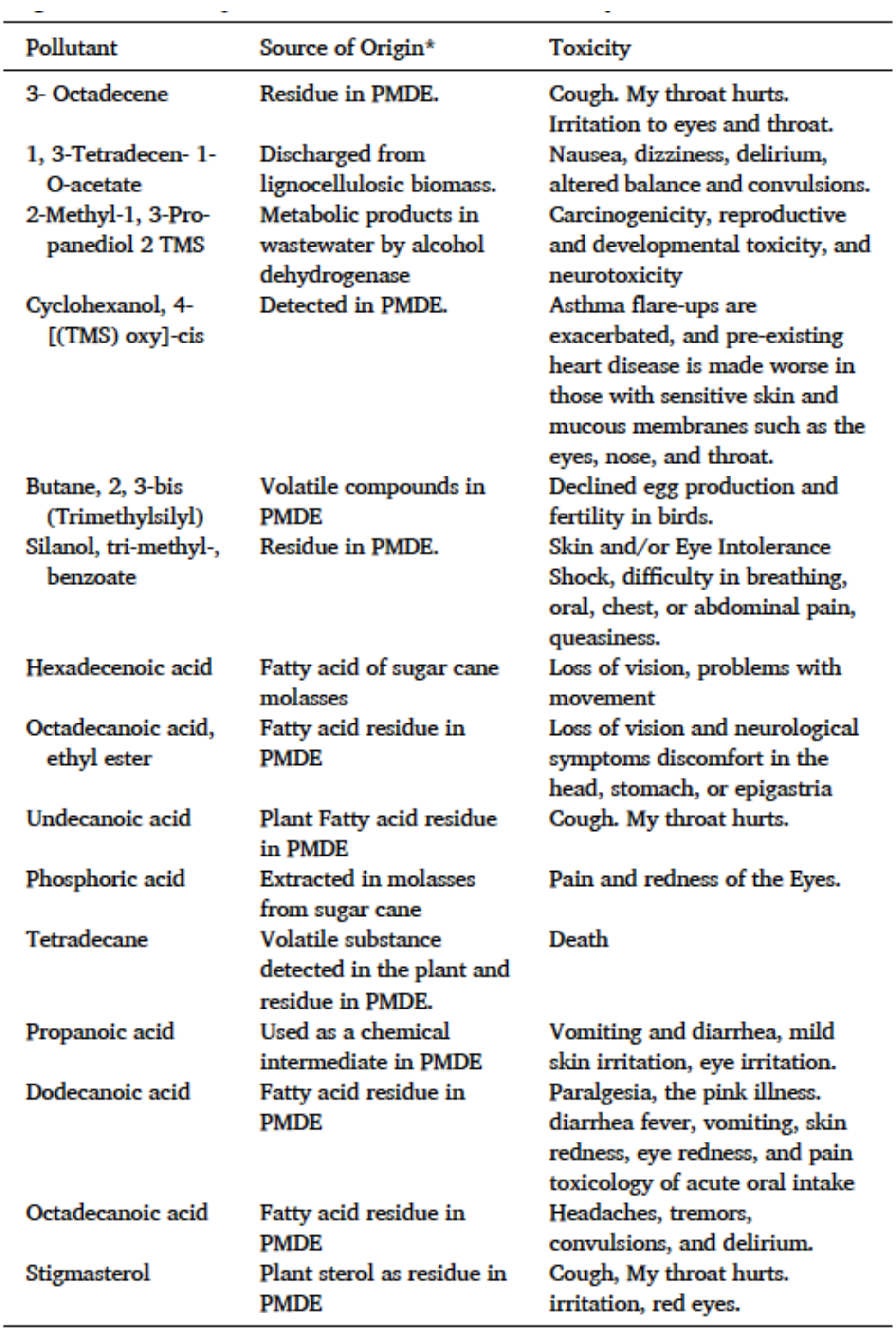
Table 2. Various inorganic metallic and non-metallic pollutants present in PMDE and their health impacts and risk associated.



Pollutants	Health impacts and risk associated*
Non metal	
Sodium	Hypertension, stress, premenstrual syndrome, osteoporosis, urinary stones,
Chlorine	Bleeding eyes, nose, and throat, pain in the chest or back, muscle weakness, abdominal discomfort nauseous.
Sulphur	Mucous membrane irritation, carcinogenicity, developmental toxicity, neurotoxicity, and acute toxicity.
Carbon	Coal workers pneumoconiosis (CWP), inflammation, fibrosis, necrosis
Oxygen	Changing living condition for flora and fauna, corrosion, eutrophication.
Heavy metals	
Cd (cadmium)	Lung cancer and degenerative bone disease a build-up of protein in the urine.
Cr (Chromium)	Hemolysis (the loss of red blood cells) and acute renal failure are all symptoms of DNA damage, carcinogenicity, allergic response, and irritating dermatitis.
Cu (copper)	Effect on eyes, gastrointestinal (digestive), hematological (blood forming), hepatic (liver) disorder.
Pb (lead)	Damage to central nervous system, anemia, encephalopathy, foot drop
Ni (Nickel)	Chemicals that are carcinogens, heart, dermal (skin), immunological (immune system), respiratory are all examples.
Zn (zinc)	Normal brain injury; cardiovascular (heart and arteries); liver (Nervous System).
Mn (Manganese)	Imbalance of hormones, harm to the central nervous system, digestive, hematological, and respiratory systems (From the Nose to the Lungs).
Fe (iron)	A narrowed blood artery, severe vomiting, kidney damage.

(*Sources: https://www.ncbi.nlm.nih.gov/)

Table 3. Organic pollutants present in PMDE and their toxicity



Pollutant	Source of Origin*	Toxicity
3- Octadecene	Residue in PMDE.	Cough. My throat hurts. Irritation to eyes and throat.
1, 3-Tetradecen- 1-O-acetate	Discharged from lignocellulosic biomass.	Nausea, dizziness, delirium, altered balance and convulsions.
2-Methyl-1, 3-Propanediol 2 TMS	Metabolic products in wastewater by alcohol dehydrogenase	Carcinogenicity, reproductive and developmental toxicity, and neurotoxicity
Cyclohexanol, 4-[(TMS) oxy]-cis	Detected in PMDE.	Asthma flare-ups are exacerbated, and pre-existing heart disease is made worse in those with sensitive skin and mucous membranes such as the eyes, nose, and throat.
Butane, 2, 3-bis (Trimethylsilyl)	Volatile compounds in PMDE	Declined egg production and fertility in birds.
Silanol, tri-methyl-, benzoate	Residue in PMDE.	Skin and/or Eye Intolerance Shock, difficulty in breathing, oral, chest, or abdominal pain, queasiness.
Hexadecenoic acid	Fatty acid of sugar cane molasses	Loss of vision, problems with movement
Octadecanoic acid, ethyl ester	Fatty acid residue in PMDE	Loss of vision and neurological symptoms discomfort in the head, stomach, or epigastria
Undecanoic acid	Plant Fatty acid residue in PMDE	Cough. My throat hurts.
Phosphoric acid	Extracted in molasses from sugar cane	Pain and redness of the Eyes.
Tetradecane	Volatile substance detected in the plant and residue in PMDE.	Death
Propanoic acid	Used as a chemical intermediate in PMDE	Vomiting and diarrhea, mild skin irritation, eye irritation.
Dodecanoic acid	Fatty acid residue in PMDE	Paralgesia, the pink illness. diarrhea fever, vomiting, skin redness, eye redness, and pain toxicology of acute oral intake
Octadecanoic acid	Fatty acid residue in PMDE	Headaches, tremors, convulsions, and delirium.
Stigmasterol	Plant sterol as residue in PMDE	Cough, My throat hurts. irritation, red eyes.

(*Sources: https://www.ncbi.nlm.nih.gov/).

5. Innovative treatment technology for PMDE wastewater

To comply with technology-based treatment standards, innovative processes to treat industrial wastewater often involve toxicity reduction technologies. A number of technologies are available to treat PMDE, they can be broadly categories into physical, chemical and biological treatment. Physical require space and they are relatively expansive (e.g., lagoon and floating method), requiring infrastructure investment. Chemical treatment result in pollutant removal via ion exchange or chemical precipitation or using advance oxidation process such as the Fenton process, although effective, this chemical treatment may result in acidifying the environment, high cost of reagents and production of high volume of ferric sludge. The biological methods are more environmentally friendly, trickling filter and activated sludge process have been successfully used to treat PMDE (Tripathi et al., 2022a;b, c). Advances and innovation of treatment are being promoted but many of these technologies are still in their infancy. Below are some of the innovative technologies being developed to treat PMDE and the pros and cons of the different treatment technologies are presented in Table 4 (see Table 4) (see Table 6) (see Table 5).

Table 4. Different treatment techniques, their preferences/advantages and limitations for treating PMDE wastewater

Innovative Treatment techniques	Merits	Demerits	References
Physical Treatment			
Membrane separation using graphene	Growth on various substrates; low Temperature; simple fabrication procedure	Agglomerate after solvent evaporation; having functional groups after reduction; difficult to mass production	Hubadillah et al. (2020)
Ion-exchange membranes	Lower energy costs, ow dynamic Binding capacity for large beads	Traditional bead-based chromatography	Tian et al., 2019
ultrafiltration membrane	Superior quality water, Lower energy requirements, Feasibility, Ability to produce water	Retention of small species, Flux may decline due to membrane fouling.	
Chemical Treatment			
Advanced oxidation method	Powerful Antioxidant Properties, Optimizes the pH Levels from Our Bodies, boosts our Immunity, Helps with Weight Loss	The BOD/COD ratios increased. Maintaining pH value	Rosa et al., 2020
Electrocoagulation	Lighter particles removed, Lighter particles removed, Faster particle removal, more rapid start-up, thicker sludge's	More complex/more maintenance, Higher -energy consumption	Jiang and Zhang, 2018
Photocatalytic degradation	Lighter particles removed,	Much Sensitive for human body, more complex/more maintenance	Ceretta et al. (2020)
Biological Treatment			
Fungal Treatment	Acceptable naturalness, Effectiveness and potentially high specificity, Protection of biodiversity in managed ecosystems	Can be very costly, they can have a short shelf life	Zhou et al. (2019)
Bacterial treatment	Bacteria are used to clean up oil spills through the bioremediation process, microorganisms are also used to produce pure compounds, and bacteria have been genetically altered to produce proteins such as insulin, growth hormones, and antibodies.	Cause diseases and infection, cause death, bacterial disease caused by sexual contact.	Kadam et al., 2018
Enzymatic Treatment			
Lignin peroxidises	Uses in Bio-refinery, textile, Energy, bioremediation, cosmetology, and dermatology industries. do not need pre-conditioning to a particular pollutant tolerate a wide range of environmental conditions	Hyper pigmentation, skin- lightning, complex hetero-polymer.	Haq and Raj, 2018
Manganese peroxidase			Rybczyńska-Tkaczyk et al., 2020
Laccase			Zhou et al. (2019)
Hybrid Treatment			
Microbial-fuel cells	Eco-friendly, cost-effective, no toxic metabolite generation, and effective wastewater treatment and bioelectricity generation	Low power generation, electron transfer, limited surface area of electrode, high cost of platinum is a major limitation to MFC application and economic viability	Miran et al. (2018); Sun et al. (2020)
Genetically modified organisms (GMOs)	Potential to accelerate decolorization of PMDE	Not suitable due to banned filed applications, possible harm to environment and biodiversity, horizontal gene transfer	Maksoud et al. (2020); Chandanshive et al. (2020)
Constructed wetland	Eco-friendly, efficient dye removal, self-regeneration, no energy requirement, recharging ground water, providing excellent habitat for aquatic and wildlife, no biosolids and sludge generation	Require large land area, costly to design and construction, facilitate mosquito breeding, high monitoring requirement, expensive to install, require skill and management, required long period for vegetation and establishment for optimal treatment efficiency	Kadam et al. (2018); Tara et al. (2019)

5.1 Innovative physical treatment technology

5.1.1 Membrane separation using graphene

Due to the high concentration of contaminants in PMDE wastewater, water emulsion separation is still a pressing problem. Because of the simplicity of its operation and the high effectiveness of its separation, membrane technology is much sought after in the water separation industry. Membrane fouling may be aided by the hydrophobicity features of the membrane, which can reduce the lifespan of the membrane. Carbon nanotubes, nanoporous graphene, and graphene oxide have all been researched in recent decades as potential materials to utilize in the production of high-capacity membranes for PMDE wastewater desalination and treatment. Graphene and graphene oxide-based composite membranes may be split into two groups: (i) freestanding membranes built entirely of graphene or graphene oxide, and (ii) polymeric/ceramic membranes modified with graphene-based nanomaterials. If a nanomaterial is added to a membrane casting solution and then the membrane is formed from a polymer and nanomaterial combination, polymeric membranes may be modified that way. In comparison to thin film composite polyamide membranes, nanomaterial membranes may achieve much greater water/permeate fluxes.

5.1.2 Ion-exchange membranes

Water desalination and electrolysis procedures use ion exchange membranes (IEMs) as an essential component for treatment. As a result, they have been explored and developed for use in innovative energy conversion and storage systems, as well as efficient desalination and wastewater treatment procedures. Fixed charges in the polymer matrix characterize an important subclass of dense polymeric membranes known as IEMs. Counter-ions (ion pairs with opposing charges) may flow through these membranes while similarly charged ions are blocked (co-ions). First discovered by Donnan, IEM permselectivity for counter- ions is referred to as the Donnan effect or exclusion (towards co-ions). Many industrial procedures, such as electrodialysis (ED), diffusion dialysis (DD), and electrolysis, are dependent on IEMs because of their ion permselectivity. There are several novel applications for IEMs, including as membrane capacitive deionization (MCDI), RED, MFCs, and ion exchange membrane bioreactors, in addition to the more established ones like flow batteries and ED/DD (IEMBs). For PMDE desalination, IEMs with low fixed charge density are also being explored as chlorine-resistant membranes.

5.1.3 Ultra-filtration membrane

Due to advancements in membrane performance and decreased prices, ultra-filtration (UF) technology has been more widely employed in PMDE wastewater treatment systems. However, ultra-filtration technology's principal drawback is membrane fouling. Natural organic matter (NOM) is generally considered safe for human consumption. There are a variety of harmful and carcinogenic disinfection byproducts (CDBPs) that may be produced when NOM in effluent water reacts with disinfectants. Many CDBPs have been found in chlorinated water, including trihalomethanes, haloacetic acids, haloacetonitriles, and other haloketones, to name just a few of the more frequent ones (Zhang and Minear, 2006; Jiang and Zhang, 2018). In UF effluent, DBPs are formed because of the organic matter's properties. To date, the effects of pretreatment on DBP production after ultrafiltration have not been studied.

5.2 Innovative chemical treatment technology

5.2.1 Advanced oxidation method

For the elimination of refactory contaminants from industrial wastewaters, advanced oxidation processes (AOPs) are deemed quick, and competitive. In the 1980s, AOPs were originally tested for drinking water treatment before being extensively used for industrial wastewater treatment (Naushad et al., 2019a; Ceretta et al., 2020). Ozone (O_3), H_2O_2, and a wide variety of catalysts, including Fe_2O_3, ZnO, CdS, TiO_2, GaP, and ZnS have all been employed in AOPs, together with high-energy radiation like UV light (Bilinska et al., 2019; Pazdzior et al., 2019; Hien et al., 2020; Tripathi et al., 2021f). Highly oxidative and non-reactive species, such as hydroxyl (OH•) and sulfate radicals (SO_4^-), are formed in AOPs, which function as mediators in electron transfer, hydrogen abstraction, and radical addition processes (Naushad et al., 2019a; Asgari et al., 2020). As a result, AOPs may effectively degrade or convert a wide range of refactory effluent contaminants into CO_2 and water (Naushad et al., 2019a; Rosa et al., 2020). However, this technology is prohibitively costly, needs a great deal of chemical energy, contains a number of time-consuming steps, and produces a substantial quantity of secondary pollutant sludge (Naushad et al., 2019b). A number of other processes, including as ozonation, photo-Fenton, ion exchange and electro-oxidation, are now in general use for the breakdown and mineralization of PMDE.

5.2.2 Electrocoagulation

Recalcitrant PMDE wastewater may be effectively treated with the use of an electrochemical method known as electrocoagulation (EC). To eliminate PMDE's harmful contaminants, the EC method uses electrical energy to dissolve iron (Fe) and aluminum (Al) (Biliska et al., 2019; Mironyuk et al., 2019). In the EC treatment process, several researchers have utilized a variety of electrodes, including iron, stainless steel, aluminum, mild steel, and graphite (Bilinska et al., 2019; Bener et al., 2019). Toxic refactory pollutants in synthetic and actual PMDE wastewater could be degraded and mineralized effectively using a variety of electrode combinations, such as Al/Al, Fe/Al, and Fe/Fe. Aluminum, for example, greatly decreases BOD, COD, and TSS in PMDE treatment (Bilinska et al., 2019; Maksoud et al., 2020). Bener et al. (2019) recently employed the electrocoagulation procedure to treat PMDE wastewater in preparation for reusing it as irrigation water in agriculture. This technique was found to eradicate 94.9 percent pigmentation, 83.5 percent viscosity, 64.7 percent TSS, 42.2 percent TOC, and 18.6 percent COD from PMDE wastewater.

5.2.3 Photo and sono catalytic degradation

For the treatment of PMDE wastewaters, it is a promising new method. The photoactivation of semiconductors in the UV-TiO_2 process is triggered by irradiation with electron-hole pairs that form as a consequence of band gap excitation (Arcanjo et al., 2018; Naushad et al., 2019a; Fazal et al., 2020). These activities may take place at or near the surface of the photoexcited particles (Naushad et al., 2019a; Pa´zdzior et al., 2019), according to the authors. The hydroxyl radical may be formed as a result of the light's potential hole reacting with electron donors (Naushad et al., 2019a; Rosa et al., 2020). Hydroxyl radicals may oxidize organic molecules, resulting in their mineralization into corban dioxide and water. These semiconductor catalysts include TiO_2, CdS, ZnO and copper nanoparticles (CuNPs) as well as SA/TiO_2, Fe_2O_3, TiO_2/UV, Bi_2O_3-Zio, $S_2O_8^{-2}$/Fe^{2+} and ZnS, which create highly reactive species such as H_2O_2, O_3, and O^{-2} in the process (Ashok kumar et al., 2019; Fazal et al., 2020; Ceretta et al., 2020). As far as mineralization and decontamination of PMDE effluent is concerned, these species are among the most successful (Arcanjo et al., 2018; Ceretta et al., 2020). TiO_2 is one of the most often utilized semiconductors in the treatment of PMDE effluent (Naushad et al., 2019b). A biochar-TiO_2 composite for the treatment of PMDE wastewater, for example, was reported by Fazal et al. (2020) to have a 99.20% photodegradation efficiency of simulate wastewater. The co-precipitation procedures to produce $CuO/Cu(OH)_2$ nanostructures that were used in the photocatalytic degradation of reactive green 19 A dye and the removal of PMDE. PMDE wastewater treatments have also made use of sonolysis, a low-tech chemical procedure. RCPs, organics, and organic compounds from PMDE may be reduced and detoxified using the sono-catalytic method. There are a variety of semiconductors that have been effective in catalyzing the reduction of organic loads, suspended solids, and content, as well as other potentially harmful substances (Pazdzior et al., 2019; Ashok kumar et al., 2019). However, due to low elimination efficiency, high costs, and environmental concerns, it is not an ideal solution (Areerob et al., 2018; Naushad et al., 2019a). Nanocomposites such as Fe_3O_4-graphene/ZnO, CdSe, gold quantum dots (GQDs), and TiO_2-BC have also been used to remove a variety of dyes, including Methylene Blue (MB), Methyl Orange (MO), Rhodamine B (RB), Reactive Blue 69 (RB), and other inorganic refactory chemicals from the PMDE under ideal environmental conditions (Areerob et al., 2018; Ashok kumar et al., 2019; Asgari et al., 2020; Ahmad et al., 2020, 2021). The US/UVZnO/PS composite was capable of removing and mineralizing acid blue 113, COD, and TOC from legitimate PMDE wastewater under acceptable situations.

5.3 Innovative biological treatment technology

5.3.1 *Fungal treatment process*

In bioremediation process, fungi are used in the degradation and detoxification process is known as myco-remediation. The primary role of fungi in the ecosystem is decomposition carried out by fungal mycelium. In microorganisms, fungi are unique in that they secrete a variety of extracellular enzymes. Lignocellulose decomposition is classified as the most important mineralization event in the earth’s nutrient cycle (Bennett and Faison, 1997). It determines the right fungal species to target a particular pollutant. The importance of fungi in the environment is that organic and inorganic substrates are decomposed and processed e.g., *P. chrysosporium, F. flavus, A. fumigates* etc. Cooke and Noel (1979), for example, recommended for the use of fungus in wastewater treatment since fungi demonstrated greater levels of organic matter decomposition.

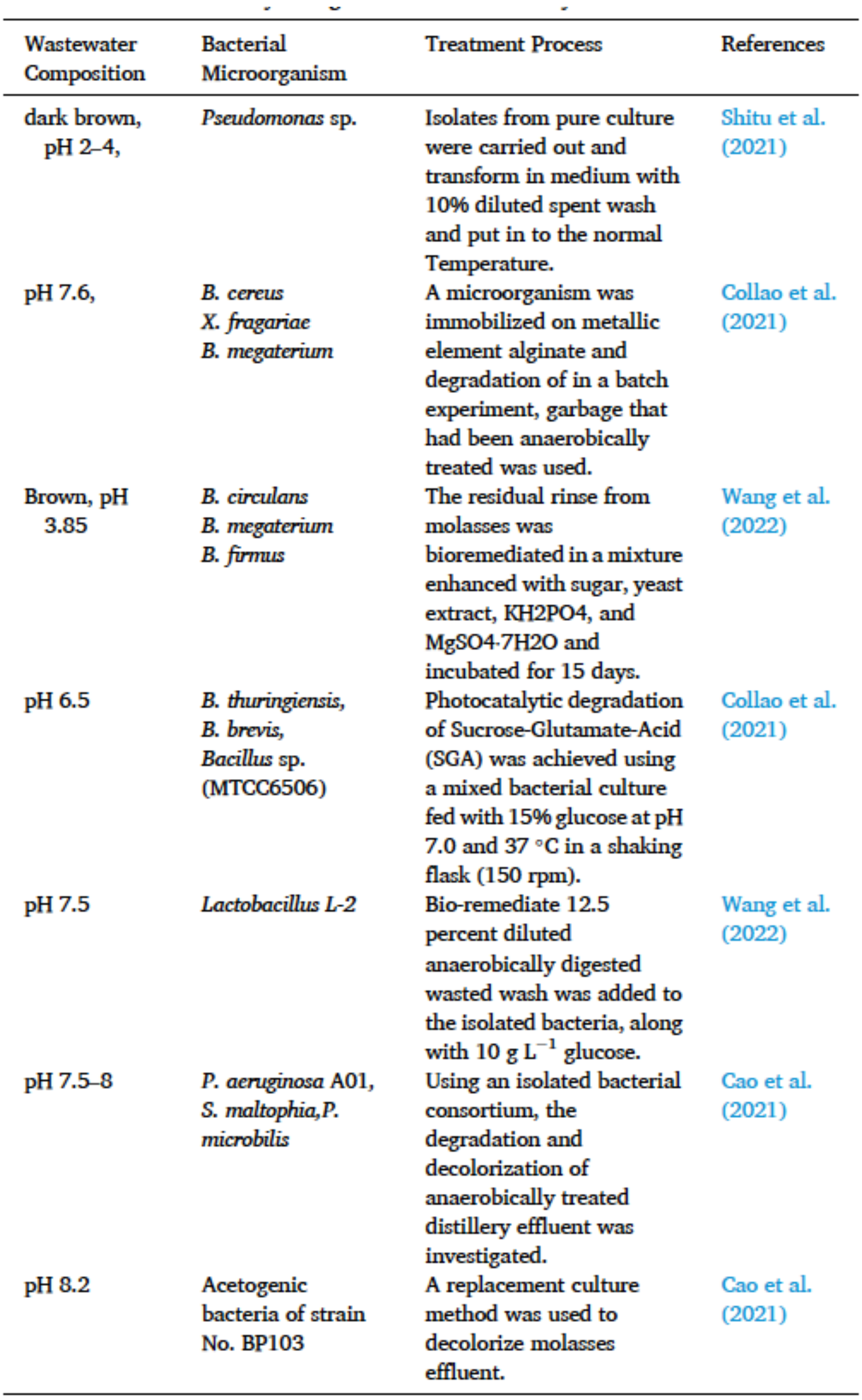
Table 5. Wastewater treatment using myco-remediation

Wastewater Composition	Fungal Microorganism	Treatment Process	References
Greenish dark brown, pH 7.2	*C. cladosporioides*	In a batch experiment, fungi were utilized to decompose 100 mL of 10% diluted PMDE supplied with carbon and nitrogen sources.	Noman et al. (2019)
pH 5.2	*Penicillium* sp. *P. decumbens*	In Methanation Process of Molasses, PMDE diluted to 50%.	Copete-Pertuz et al. (2021)
Brown, pH 3.85<	*A. fumigatus*	PMDE was degraded in a mixture containing glucose, yeast extract, KH2PO4, and MgSO4·7H2O, as well as fungi, and incubated for a few days.	Kumar et al. (2021)
Dark-brown, pH 4.1	*Taramites* sp. I-62	PMDE was added to the culture medium at a concentration of 20% (v/v) and cultured for 7 days at 28 °C.	Sharma et al. (2022)
NR	*P. chrysosporium*	The development rate of a rotating biological contactor (RBC) is boosted when polyurethane foam (PUF) and scouring web (SW) are introduced.	Singh, 2017
Dark brown, pH 4.3	*F. flavus*	On a polyurethane foam cube, segregated fungal development was impacted by decolorized 10% diluted PMDE.	Kumar et al. (2021)
pH 3.9	*T. pubescent* MB 89	To cure 10% PMDE, the segregated fungal development was impacted in flask cultures and a bubble lift bioreactor.	Noman et al. (2019)

5.3.2 *Bacterial treatment process*

Bacteria play an important role in bioremediation, a potent, cost- effective, and environmentally acceptable alternative to physicochemical techniques (Singh et al., 2022). Various bacterial species, including as *Pseudomonas* sp.*, B. cereus, B. thuringiensis, and X. fragariae,* have been identified as having the ability to mineralize and decolorize hazardous chemical pollutants present in PMDE. For bioremediation of PMDE waste products, free or immobilized cells are being researched extensively. When compared to free-cell culture, immobilization of bacteria on inert support materials such as alginate, polyacrylamide, agar, styrene, and polymer is far more favourable with benefits of compact carrier pellets body, high retention of biomass, extensibility of culture, and a faster removal technique. The chemical composition of the wastewater, as well as nutrient, pH, heat, oxygen, and inoculum size, all affect bacteria permeability in PMDE wastewater treatment.

Table 6. Wastewater treatment using bacterial community



Wastewater Composition	Bacterial Microorganism	Treatment Process	References
dark brown, pH 2–4,	*Pseudomonas* sp.	Isolates from pure culture were carried out and transform in medium with 10% diluted spent wash and put in to the normal Temperature.	Shitu et al. (2021)
pH 7.6,	*B. cereus* *X. fragariae* *B. megaterium*	A microorganism was immobilized on metallic element alginate and degradation of in a batch experiment, garbage that had been anaerobically treated was used.	Collao et al. (2021)
Brown, pH 3.85	*B. circulans* *B. megaterium* *B. firmus*	The residual rinse from molasses was bioremediated in a mixture enhanced with sugar, yeast extract, KH2PO4, and MgSO4·7H2O and incubated for 15 days.	Wang et al. (2022)
pH 6.5	*B. thuringiensis*, *B. brevis*, *Bacillus* sp. (MTCC6506)	Photocatalytic degradation of Sucrose-Glutamate-Acid (SGA) was achieved using a mixed bacterial culture fed with 15% glucose at pH 7.0 and 37 °C in a shaking flask (150 rpm).	Collao et al. (2021)
pH 7.5	*Lactobacillus L-2*	Bio-remediate 12.5 percent diluted anaerobically digested wasted wash was added to the isolated bacteria, along with 10 $g\ L^{-1}$ glucose.	Wang et al. (2022)
pH 7.5–8	*P. aeruginosa* A01, *S. maltophia,P. microbilis*	Using an isolated bacterial consortium, the degradation and decolorization of anaerobically treated distillery effluent was investigated.	Cao et al. (2021)
pH 8.2	Acetogenic bacteria of strain No. BP103	A replacement culture method was used to decolorize molasses effluent.	Cao et al. (2021)

5.3.3 *Enzymatic treatment process*

In the breakdown and decontamination of industrial wastewater pollutants as well as the rehabilitation of polluted areas, several enzymes are claimed to be involved (Kumar et al., 2020; Xiang et al., 2020). Table 4 lists the many kinds of intracellular and extracellular proteins that may be produced by various microorganisms during the breakdown and mineralization of harmful chemical contaminants. In the distillery sector, contaminants are broken down and cleaned up by laccase, peroxidase, and polyphenol oxidase enzymes (Zhou et al., 2020). Comparatively to other enzymes, laccase is the most efficient in degrading and decolorizing stubborn organic pollutants for wastewater treatment (Cao et al., 2019; Rybczynska-Tkaczyk et al., 2020). Because these colorless aromatics are so persistent in anaerobic settings, aerobic conditions are required for the subsequent mineralization of these aromatic intermediates. The peroxidase enzymes produced by *Halomonas* sp. GT and *Bjerkanderaadusta* CCBAS 930 degrade and decolorize PMDE in the presence of these organisms (Tian et al., 2019; Rybczynska-Tkaczyk et al., 2020). The lignin peroxidase enzymes produced by *Bacillus circulans* BWL1061 have also been shown to decolorize methyl blue dye (Liu et al., 2017). Laccase (EC1.10.3.2) has shown great promise in the breakdown and decolorization of a variety of organic pollutants, including PMDE and the more common crystal violet and methyl violet. Unlike other electron acceptors, it does not need a co-factor (Dauda and Erkurt, 2020; Zhou et al., 2019). Laccase enzymes use ABTS (2,2-azino-bis-3-ethylbenzothiazoline-6-sulfonic acid) as a redox mediator because of its low molecular weight. PMDE/wastewater degradation and decolorization are much improved when this redox mediator is present (Zhuang et al., 2020). Laccase generated by *Phomopsis* sp. may be employed as a mediator in the degradation of distillery effluent, according to Navada and Kulal (2020). More than only lignin peroxidase (LiP), tyrosinase, peroxide, and polyphenol oxidase are also found to have a key role in the breakdown and decolorization of organic pollution emissions from distillery businesses (Chandanshive et al., 2018; Cao et al., 2019; Zhuang et al., 2020). Additionally, these enzymes have been shown to break down polychlorinated biphenyls, phenols, and polyaromatic hydrocarbons (PAHs) (Liu et al., 2017; Haq and Raj, 2018). Due to its cost-effectiveness as well as environmental compatibility, enzyme breakdown and mineralization of PMDE has often been seen as an affordable feasible solution.

5.4 Innovative hybrid treatment technology

5.4.1 Microbial fuel cells

The electrons and protons created by the oxidation of various organic and inorganic compounds may be directly transferred to electrodes owing to the MFC system's utilization of microorganisms as catalysts (Miran et al., 2018; Sun et al., 2020; Chauhan et al., 2022). For the degradation and mineralization of PMDE wastewaters and the production of electricity with decreased CO_2 emissions. This is a new, advanced, and long-term strategy to consider (Oon et al., 2020). In order to effectively treat PMDE, many electrodes have been investigated in MFCs, with oxygen (O_2) and platinum (Pt) generally found to be the most suitable electron acceptor in MFCs systems. However, these electrodes have many limitations such as high cost, poor kinetics of oxygen reduction reaction (ORR) (Sun et al., 2020; Maksoud et al., 2020). As a result, MFCs have been outfitted with electrodes made of nickel, manganese, and copper, which are more efficient in reducing oxygen (ORRs) (Oon et al., 2020). An excellent ORR catalyst may be made from cobalt, for instance. A mixed community of sulfate reducing Proteobacteria, *Desulfovibrio*, and Deltaproteobacteria found by Miran et al. (2018), can decolorize Acid red 114 dye to a high degree (89.4%, 48.2%, and 52.7%), while also generating up to 258 ± 10 mWm^{-2}.

5.4.2 Genetically modified organisms (GMOs)

RCPs that are resistant to biodegradation because to their complex chemical structure and high stability were found in PMDE (Fazal et al., 2020; Tripathi et al., 2021a,b,c). Conventional secondary treatment of PMDE is not always successful with these methods (Maksoud et al., 2020; Chandanshive et al., 2020). The use of genetically modified organisms in the treatment of PMDE may be a viable option in the long run. Many scientists have genetically engineered microbes and employed them in the treatment of PMDE. When the laccase (*LacTT*) gene from *Thermus thermophilus* SG0.5JP17-16 was introduced into *Pichia pastoris*, it increased the plant's ability to break down and decolorize a variety of organic contaminants including reactive black and phenol red. A thermo-alkali-stable laccase gene purified from *Klebsiella pneumoniae* was cloned into *E. coli* and used for the rapid degradation and decolorization of various RCPs such as bromophenol blue, mordant black 9, reactive brilliant blue X-BR, cotton blue, azophloxine, reactive brilliant blue K-GR, congo red, reactive brilliant blue KN-R, malachite green and reactive dark blue M-2GE (Liu et al., 2017). Laccase isolated from

Pleurotus sp. MAK-II exhibited great resistance to several pigments as diazo dye, congo red, anthraquinone, remazol blue coloured and decolorization in the existence of violuric acid as catalyst (Manavalan et al., 2015).

5.4.3 Constructed wetland

Constructed wetland (CW) systems use plants' natural ability to remove PMDE wastewater contaminants as a remediation and treatment method. Degradation/mineralization/removal of PMDE, phenolic detergent, emulsifier, volatiles, toxins, and heavy metals from industrial wastewaters was achieved by using natural compounds such as wetland vegetation as a medium and associated bacterial ecology for destruction (Kadam et al., 2018; Tara et al., 2019; Oon et al., 2020; Shukla et al., 2021). Both aerobic and anaerobic conditions may be accommodated by CWs, with horizontal subsurface flow (HSFC) and vertical subsurface flow (VSFC) the most common. The scenario is inverted under anaerobic settings, when color is successfully eliminated but organic molecules are not decomposed effectively, where organic load is sufficiently removed but color is not greatly decreased (Hussein and Scholz, 2018; Tara et al., 2019; Oon et al., 2020). CW has been shown in several investigations to successfully decolorize and mineralize organic and inorganic compounds derived from PMDE. The removal of color (74%), COD (81%), BOD (72%), TDS (32%), and heavy metals such as Cr (97%), Fe (89%), Ni (88%) and Cd (72%), using a vertical-flow pilot-scale wetland supplemented with bacterial endophytes, was shown to be successful (Hussein and Scholz, 2018). A pilot-scale wetland with *Phragmites australis* reported (89%) COD; (91%) BOD; and (96%) of TDS removal from distillery bleaching effluent in a horizontal and vertical flow system (Hussein et al., 2019). With the addition of the microbial fuel cell (MFC), created wetlands were able to remove organic contaminants, as well as produce bioelectricity (Oon et al., 2020). Even more importantly, the incorporation of various bed-materials with the boosts therapy efficacy of CWs systems (Kadam et al., 2018; Tara et al., 2019; Oon et al., 2020).

Because CWs are economical, environmentally acceptable, widely accepted, and create bioenergy and bioelectricity, they offer a viable option for the breakdown and detoxification of PMDEs.

6. Current challenges and key issues

PMDE may be treated using a variety of classic and cutting-edge approaches. PMDE treatment using physicochemical approaches seems to be successful. However, these technologies have a high operating cost and produce unwanted secondary pollutants. Microbiological remediation is less expensive, environmental-friendly, and acceptable to public, although it is less efficient compared to physicochemical methods. In order to reduce the environmental and public health dangers, further research is needed from the laboratory size to pilot scale. The creation of transgenic microbial strains with specific genes for the removal of organic contaminants from wastewater is also an important tool to improve biological treatment efficiency, but its application requires careful control to avoid unintentional release. PMDE decomposition and detoxification may also be achieved by the use of a combination of physical, chemical, and biological processes. Contaminated sites may be efficiently remedied using built wetlands, which are environmentally favourable, developing technologies but it requires larger areas and upfront construction costs. Microbial fuel cell technology has the added advantage of generating bioelectricity from the treatment process, however, this technology is in its infancy and the power generated is still limited.

7. Conclusions and future perspectives

Alcohol production is a worldwide industry; the treatment of PMDE wastewater and its safe reuse present a global challenge. PMDE contains multiplex organic, inorganic and EDCs compounds, and they play an important role in environmental pollution and health hazards. Many studies found cytotoxicity, genotoxicity and carcinogenetic compounds in PMDE wastewater that persist in the environment for many years and cause harmful effects on the biota. The ineffective treatment of PMDE is of great concern. Conventional technologies are not completely effective and innovative methods or hybrid technologies are required to treat PMDE much efficiently. The innovative treatments hold many promises, especially in the biotreatment of PMDE as they are low-cost, sustainable and environment-friendly, but all these technologies have their drawbacks, either in terms of high running cost, requirement of new expensive infrastructure or currently at a low technology transfer level. More investment is needed in making these technologies more assessable and available to the distillery industry.

Author contributions

ST-Conceptualization and writing DP-Data analysis, review and revise the manuscript; AN-Review and incorporate the corrections in the manuscript; PC & RC-Supervision, investigation, generation of data and writing original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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