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Future and challenges of co-biofilm treatment on ammonia and Bisphenol A removal from wastewater

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ABSTRACT

Ammonia-nitrogen (NH_3–N) is one of the most frequent pollutants in wastewater which utilizes dissolved oxygen in water and produces eutrophication in water bodies, while Bisphenol A (BPA) is an emerging pollutant that disrupt endocrine hormones and system operation even at extremely low doses. This review discusses the levels of ammonia and BPA in domestic wastewater and their effects. The treatment of these contaminants through a biological process is emphasized. The removal mechanisms are explained, and a new 'co-biofilm' treatment is introduced to remove ammonia and BPA simultaneously. Co-biofilm treatment is a hybrid technology of moving bed biofilm reactor (MBBR) and water hyacinth for wastewater treatment. This hybrid technology provides co-biofilm treatment between biocarriers in MBBR and the roots of water hyacinth. In this technology, the extensive of aeration used in the MBBR and the slowness of water hyacinth's capability in removing contaminants could be improved. Further studies are suggested to optimize the performance of this environmentally friendly system.

1. Introduction

Water pollution has become a severe environmental problem in developing countries. Clean and safe drinking water is essential for life. However, many waterborne diseases come from polluted drinking water resources. For example, only 20 % of Pakistan's population has a reliable and treated water supply, while the others are forced to use untreated water [1]. Ammonia and Bisphenol A (BPA) are the primary pollutants in the water. Both have adverse effects on humans and the environment. Ammonia could be formed in aquatic ecosystems in two ways: unionized ammonia (NH_3) and ionized ammonia (NH_4^+) based on pH and temperature [2,3]. Uncharged NH_3 is the most toxic because it can dissolve in lipids. Natural ammonia does exist in the waterways, but human activities increase ammonia or ammonium ion concentrations.

Furthermore, as reported by previous researchers, nitrogen removal processes are not available in most wastewater plants [4]. Thus, the wastewater effluent from industry or sewage treatment plants discharged into a river or water body does not meet the standard guidelines of the authority. The effluent from these sources will cause ammonia to accumulate in the bodies of water and elevate ammonia concentrations. Usually, the ammonia concentration will naturally dilute in waterways before it reaches a drinking water treatment plant. Unfortunately, the ammonia discharged into rivers from various point and non-point sources makes this difficult [5]. Besides, the high ammonium concentration in water bodies will affect the continuous drinking water treatment plants [6]. According to a previous report, ammonia concentrations measured at 2 mg/L in surface water bodies would pose a challenge for the chlorine disinfection process in conventional drinking water treatment plants [7].

According to Zielinska et al. [8] in 1981, Aleksandr P. Dianin was the

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earliest researcher who synthesized BPA during a search for synthetic estrogen. The mixture of acetone and phenol formed BPA [9]. In the 1940s and 1950s, BPA was found to be beneficial in the plastics industry. BPA utilization was widely used, especially in epoxy resins and polycarbonate plastics [10,11]. Due to increasing BPA usage, it is ubiquitous in the environment, including domestic wastewater [10,12]. BPA is categorized as an endocrine disruptor compound (EDC), which is toxic to living things [13]. BPA acts as an estrogen mimic [14–16]. Rochester [17] stated that there are 91 studies relating to BPA and human health. More than 90 % of the people tested had a detectable amount of BPA in their urine [14]. BPA esters in polycarbonate and epoxy resins go through hydrolysis, resulting in BPA's occurrence in food, beverages, and the environment [18].

Many foods have direct contact with BPA via their packaging, including the inner coating of cans and the caps of bottles and jars. Humans have been exposed to BPA through food intake. Konieczna et al. [19] stated that BPA could enter human bodies through inhalation, orally, and through the skin. That also explains its existence in some people's urine. The continuous discharge of BPA in water bodies makes it pseudo-persistent even though its concentration is low [20]. Also, though BPA is always deficient in wastewater, it is not easy to entirely remove it by conventional biological treatment [21].

Because of the effects of ammonia and BPA on humans and the environment, finding an efficient treatment for their elimination, even at a low dosage, is essential. In general, the removal of ammonia and BPA could be carried out through chemical, biological, or physical methods, or a combination of those technologies. Examples of ammonia removal technologies include adsorption [22], air stripping [23], biological treatment, chlorination [24], biofiltration [25], chemical precipitation [26], ion exchange [27], and supercritical water oxidation [29]. The removal technologies for BPA include adsorption [13], biological treatment [30], advanced oxidation [31], photocatalytic [32], ultrafiltration [33] and phytoremediation [34].Some reports mentioned that biological treatment for ammonia removal is cheaper than chemical treatment [35]. Nonetheless, all these technologies have both advantages and disadvantages.

Co-biofilm technology is a combination of biofilm bioreactors such as moving bed biofilm reactor (MBBR) or sequence batch biofilm reactor (SBBR) with aquatic plants such as water hyacinth. Biofilm reactors have been widely applied in wastewater treatment and are reported to have high removal efficiency [36]. The distinctive biofilm structure and effective mass transfer performance provide biofilm bioreactors excellent benefits in improving the simultaneous removal of organic matter and nitrogen in wastewater [37]. Aquatic plants utilized in phytoremediation, such as *Eichhornia crassipes*, *Pistia stratiotes*, and others, are naturally adapted for growth in contaminated water bodies. Phytoremediation is a plant-mediated pollutant removal process employed for water system decontamination [38]. Co-biofilm refers to the biofilm development on the biocarrier and roots of the aquatic plant, which are the keys to the removal of pollutants. This system is also suggested to enhance the slowness of phytoremediation and help reduce aeration used in aerobic bioreactors through the oxygen transfer by the aquatic plants.

Due to a lack of reports on the co-biofilm discussion so far, this review would like to do a comprehensive study on the simultaneous removal of ammonia and BPA from wastewater, particularly biological removal treatment via co-biofilm technologies.

2. Ammonia and Bisphenol A (BPA) in domestic wastewater

2.1. Sources of ammonia and BPA

Countries like Pakistan and China found that sewerage is the primary source of contamination due to its freely released into drinking water supplies. Recently, elevated concentrations of ammonia and BPA were highlighted as the primary pollutants in drinking water sources [39]. These pollutants that contribute to water pollution come from point and non-point sources. The primary point sources are industrial effluents, domestic sewage, and aquaculture [40]. Meanwhile, agriculture and landfills are the primary non-point sources when heavy rainfall occurs, causing farming runoff and leachate from landfills. Unfortunately, pollutants from non-point sources are difficult to control [41]. Thus, it would be good if the point sources could be monitored and controlled.

Inorganic pollutants and nitrogen compounds come from sludge waste and domestic and industrial effluents, and they exist naturally in the waterways [42]. The escalation of the economic and human lifestyle has increased wastewater production. Wastewater rich in nutrients and organic and inorganic matter is discharged in the effluents [43]. Excess ammonia excreted from the human body through urine contributes to the ammonia concentrations in domestic sewage. Waste from industrial activities may result from pharmaceuticals, chemical fertilizers, coal gasification, and petroleum refining [2]. Elevated nitrate pollution has primarily been contributed by the agricultural and livestock farming sectors, which use ammonia in fertilizers [44]. A high ammonia concentration was observed in wastewater effluent from the food and agricultural industries [45]. Most previous researchers stated that ammonia pollution had been contributed by agricultural diffusion, industry, and landfill leachate [46–48].

Other researchers have reported that the source of BPA contamination is anthropogenic [49,50]. Generally, BPA is in the atmosphere from dust, food and beverages, water, wastewater, and humans (saliva, sweat, and urine) [50]. Clearly, the widespread evidence of BPA in the surrounding area shows no way to identify the natural sources of this contamination. BPA exposure to humans and the environment could occur because of BPA production, processing, treatment, and hydrolysis of polycarbonate and epoxy resins [51]. The role of BPA as an intermediate in epoxy resins and polycarbonate (PC) plastics results in its presence in numerous plastic products [16]. For instance, BPA is widely used in food and beverage packaging, dentistry, electrical equipment, and other plastic products [52–54].

Humans cannot avoid exposure to BPA. According to some reports, unreacted BPA could leach out due to contact with drinks and foods because its ester linkage undergoes hydrolysis at high temperatures or when in dilute acid or alkali solutions [49] [55]. Nevertheless, Michałowicz [50] reviewed in his paper that BPA leaching is negligible below room temperature. Hence, the various ways of BPA migration result in the presence of BPA pollutants in many municipal and industrial wastewaters in Korea and Finland [21,56–58]. Meanwhile, in the United States, BPA is found in various river water samples taken from industrial, residential, and commercial areas [59].

2.2. Health and environmental effects

The ammonium ion itself has no significant unfavorable impact on human health if the intake is lower than the amount that can be detoxified [60]. But according to Suárez et al. [61], long-term ammonia intake seems to increase ammonia levels in the blood, which can cause hyperammonemia. Hyperammonemia is when ammonia has been overproduced, impairing detoxification [62]. Excessive nitrogen compounds could lead to the blue-baby syndrome in infants [6]. Besides, inhalation of ammonia could lead to bronchiectasis problems [63].

Excessive ammonia could harm the environment. Ammonia could harm living organisms in the soil and aquatic life because of its toxicity [64]. Increasing nutrients in water bodies such as nitrate, phosphate, and silicate lead to eutrophication (which leads to nutrient enrichment and oxygen depletion) [64–67]. For instance, ammonia itself could turn into nitrogen, which results in algal growth. Therefore, the outcome of the phenomenon leads to oxygen depletion and, as a consequence, the death of the aquatic organism [29,43,46]. As concluded by previous reviews, the interactions of BPA with androgen, aryl hydrocarbons, estrogen, and peroxisome proliferator-activated receptors could disrupt sex hormone function (resulting in male and female infertility).

Furthermore, interaction with endocrine system function, leptin, insulin, and the immune system could also lead to hormone-sensitive cancers such as prostate and breast cancers, and metabolism problems such as polycystic ovary syndrome (PCOS) [16,19,50,55,68]. Also, Valentino et al. [52] reported that BPA is an obesogenic compound, an artificial chemical that could increase metabolic syndrome, obesity, and diabetes, even at low chronic consumption. The concentration of 0.05 μg/mL is the US Environmental Protection Agency's (US EPA) threshold dose; 0.02 μg/mL is the concentration of unconjugated BP-A shown in human serum; and 0.01 μg/mL denotes the European Union's acceptable daily intake [69,70]. Besides the adverse impact of BPA on humans, the same problem also applies to aquatic life. In particular, BPA could also have unfavorable effects on wildlife reproduction, such as prompting a genetic disorder and disrupting the embryonic growth of fish, amphibians, annelids, crustaceans, mollusks, and even insects [68].

2.3. Level of ammonia and BPA

A small dose of unpurified ammonia has high toxicity [69]. Ammonia concentration in river waters is of much concern because its high level can disrupt the operation of drinking water treatment plants [6]. Also, according to the World Health Organization (WHO) [70], drinking water that has an ammonia concentration above 0.2 mg/L would have an unpleasant taste and odor. According to the WHO, the ammonia level guideline in water bodies is 50 mg/L [74]. In Malaysia, the permitted level of ammonia in treated and untreated drinking water is under 1.5 mg/L [6]. According to the Department of Environment (DOE) Malaysia, regulated ammonia concentrations for domestic effluent are 20 mg/L for standard A and 10 mg/L for standard B. The average ammonia concentration in municipal wastewater is between 10 and 200 mg/L [2]. Several studies that reported NH_4^+N levels in real municipal wastewater are summarized in Table 1. The ammonia concentration is quite high for aquatic life. Moreover, other point and non-point sources of ammonia discharged into water bodies would elevate the ammonia concentration. Hence, the ammonia concentration will be unfavorable and could cause problems for the drinking water treatment plant, particularly the disinfection process.

According to the European Food Safety Authority (ESFA) [79], the tolerable daily intake (TDI) for BPA is 4000 ng/kg per day. Wang et al. [80] reviewed previous literature and found that the average BPA in municipal effluent wastewater is 188 ng/L. The BPA concentrations in several municipal wastewater plants are presented in Table 2. Previous literature showed that the range of BPA concentrations in municipal wastewater treatment plant (MWTP) effluents was between 0 and 3100 ng/L. The incomplete removal of BPA at the final treatment of municipal wastewater contributes to the high levels of BPA in water bodies. However, previous literature mentions that the continuous leaching of resins and plastics in water bodies will also elevate BPA concentrations [16,20]. As reported by previous researchers, the degradation of BPA by AOB in nitrifying systems was successful in the presence of nitrite through abiotic nitration [84]. The degradation of BPA occurred after the ammonia was degraded to nitrite. So far, knowledge of the reaction between ammonia and BPA has not been reported. Detailed information on ammonia and BPA degradation is addressed in another section of this review.

Table 1
NH_4^+N concentrations from several sources of effluent reported by previous studies.

No.	Types of wastewaters	Origin	Effluent NH4+N concentration (mg/L)	References
1	Sludge from municipal wastewater	–	45	[75]
2	Municipal wastewater	–	42	[73]
3	Municipal wastewater	South Africa	135	[77]
4	Municipal wastewater	–	28.19	[78]
5	Inoculated sludge	Beijing, China	41.91–67.05	[76]
6	Effluents of secondary wastewater (ESWW)	Qatar	58–65.78	[80]
7	Domestic wastewater	Yangzhou, China	(47.8–72.6)	[78]

Table 2
BPA concentrations in several municipal wastewater treatment effluents.

No.	Types of wastewaters	Origin	Concentrations of BPA effluent (ng/L)	References
1	The wastewater treatment plant	Taipei, Taiwan, China	0.39	[85]
2	The municipal treatment plant	Stonecutters Island, Hong Kong, China	131–1550	[86]
3	Three Sewage treatment plants	Shenzhen, China	34–3099.6	[87]
4	The municipal treatment plant (60 % domestic, 40 % hospital, institutions, industries)	Steinhäule, Germany	162	[88]
5	Wastewater treatment plant (100 % domestic)	Xiamen, China	16.8–544	[89]
6	The wastewater treatment plant	Xiamen, China	0–162	[90]
7	Municipal Wastewater Treatment Plant (urban sewage)	Rome, Italy	Around 20–70	[88]
8	The municipal wastewater plant	Northern, Greece	20–48	[91]

3. Biological removal of ammonia and BPA

Removing ammonia and BPA from domestic wastewater is crucial as it affects water quality. In many countries, conventional biological wastewater treatment has been used for a long time. However, the effectiveness of traditional wastewater treatment has been limited due to the emergence of new challenges [92]. Biological treatment is one promising treatment that is more effective for contaminant removal. The physical-chemical process of the nitrogen removal process is costly because of the processing, energy consumption, and chemicals used.

3.1. Ammonia removal technologies

Instead of physicochemical ammonia removal technology, biological ammonia removal technology is one promising method because it is cost-effective and considered to be a green approach for domestic wastewater treatment. Currently, natural oxidation of ammonia through a biofilm process is an excellent idea for an alternative, especially for low pollutant concentrations [93–95]. Moreover, the augmentation of microorganisms in a particular process condition enhances the degradation efficiency of tenacious organic pollutants [96,97].

Biological wastewater treatment applies bioreactor technology combined with a subsequent process of solid-liquid separation. The separation of biomass/water related to the natural mechanism strongly affects the removal efficiency [96]. Due to their distinctive potential, several types of standard bioreactors have been implemented in municipal and industrial wastewater treatment [92,99,100]. A summary of the bioreactors used in wastewater treatment is presented in Table 3.

A membrane bioreactor (MBR) is one of the favored technologies for wastewater treatment. MBR technology is a membrane separation process combined with a bioreactor. The same applies to the common wastewater treatment process. The bioreactor in this system implements microorganisms from any activated sludge in the aerated tank. A porous

Table 3
Summary of biological ammonia removal technologies.

No.	Type of treatment	Type of treated water	Treatment condition	Summary of efficiencies	References
1	Microalgal-bacterial granular sludge process	Synthetic wastewater	•A lab-scale of 60 mL glass reactor filled with matured microalgal-bacterial granules.	•At 6 h of non-aerated conditions, the removal efficiencies achieved for influent organics, ammonia and phosphorus is 92.69 %, 96.84 % and 87.16 %.	[116]
2	Up-flow aerated submerged fixed-film (ASFF) bioreactor	Real municipal wastewater	•A lab-scale of 14.8 L bioreactor with polypropylene packing media.	•At hydraulic retention time (HRT) 2.5 h and 8 h, the ammonia removal efficiency obtained is 78.9 % and 94.0 %.	[117]
3	Algal-bacteria biofilms affixed with rotating contactors	Municipal anaerobic digester filtrate	• Involving three pilot-scale reactors. • Each reactor in the pilot-scale system comprises multiple Algaewheel™ rotating algal contactors (RACs)	• At hydraulic retention times (HRTs) of 0.5–2 days, the total ammonia nitrogen (TAN) removal achieved is in the range of 45 % to 60 %. • From the overall treatment, >95 % TAN was oxidized to nitrite, meanwhile 27 to 36 % subsequently evolved as $N_{2,}$ and 3–11 % only deteriorated to nitrate.	[118]
4	Bio augmented multistage bio filter	Real municipal wastewater	• Implementation of biological wastewater treatment using bioaugmentation at lab scale • Three inert plastic tanks with a 25 L volume each is used. • The first tank was used for the sedimentation step, followed by the second tank, which was filled with 20 cm of gravel to act as a gravel biofilter, and the third tank was filled with 20 cm of sand to act as a sand biofilter.	• 99.99 % of tested microbial species removal is achieved. • After the sand filtration, the maximum removal efficiencies of H_2S, COD, BOD_5, total solids (TS), total dissolved solids, total suspended solids, ammonia, nitrate, phosphorus, and oil and grease achieved is 85, 93.4, 83.5, 37, 49.2, 93.4, 100, 55.7, 76.6 and 76.6 %, respectively.	[119]
5	Carbon-based moving bed biofilm reactor	Coal pyrolysis wastewater	• By implementing the solvent and alkali extraction in pre-treatment at pilot scale • Two different carriers, lignite activated coke (LAC) based MBBR and activated carbon (AC) based MBBR, are compared.	• LAC-based MBBR showed more efficient removal than AC-based MBBR. • The maximum phenol removal is 94.61 % and 88.68 %, and NH_4^+-N is 91.03 % and 75.67 % for LAC and AC-based MBBR, respectively, after NH_4^+-N exceeds 320 mg/L.	[120]
6	Aerobic up-flow submerged attached growth reactor (SAGR)	Gold mining wastewater	• A total of six SAGRs, each packed with locally sourced pea gravel (estimated specific surface area of 297 m^{-2} m^{-3}) • The two sets of three SAGRs were operated at HRTs of 0.45 days, 1.20 days, and 2.40 days.	• Over 98 % ammonia removal was achieved. • Free ammonia could harm performance at high pH	[121]
7	Step-feed three-stage integrated anoxic/oxic biological aerated filter (STIAOBAF)	Synthetic domestic wastewater	• Enhanced nitrogen removal through the combination of anoxic denitrification with aerobic simultaneous nitrification and denitrification (SND • The influent flow distribution ratio (IFDR) of the three reactors was optimized using response surface methodology	• The maximum TN removal efficiency achieved is 81.4 % at optimized influent flow distribution ratio (IFDR) of three reactors which is 32 %:49 %:19 %.	[122]
8	Tidal flow constructed wetland (TFCW)	Anaerobically digested dispersed swine wastewater (ADSWW)	• Pilot-scale • The theory of the dynamic process of rapid-adsorption and bio regeneration in biozeolite for N removal was proposed.	• The removal efficiencies achieved 73.79 %, 72.99 % and 70.71 % for COD, NH_4N and TN, respectively, at even 16 °C. • Nitrogen removal could be summarized as follows: nitrification-denitrification (80.32 %) > substrate adsorption (18.91 %) > plant uptake (0.77 %).	[106]
9	Biological folded non-aerated filter (BFNAF)	Synthetic inorganic wastewater	• 40 L lab reactor • By response surface method, the COD/N ratio and the HRT were 5.39 and 10.83 h.	•The maximum values of 88.62 ± 0.81 %, 76.12 ± 0.57 %, and 50.48 ± 1.02 % of NH_4^+, COD and TN removal efficiencies were achieved.	[123]
10	Anaerobic baffled reactor	Raw municipal wastewater	• 1000 L pilot reactor • HRT = 12 h	•In the first cell, the TSS and total COD removals were 75 ± 15 % and 43 ± 14 %, but only 20 % of the total methanation was observed for the complete ABR.	[105]
11	Integrated anaerobic fluidized-bed membrane bioreactor (IAFMBR)	Domestic wastewater	•Granular activated carbon (GAC) used as a carrier.	•The COD removal of 75.8 %, 73.6 % and 54.1 % was achieved at 8, 6 and 4 h HRT, respectively, resulting in a methane yield of 140, 180 and 190 L CH_4(STP)/kg COD removed and a conversion of 45.2 %, 53.1 % and 43.8 % of COD into methane in biogas.	[104]
12	Biological aeration filter (BAF)	Raw domestic sewage	• Two coupled BAFs were built up. • To improve the nitrogen performances backwash system was conducted in BAF2	• By heterotrophic denitrification pathway in BAF1, nitrogen removal concentration achieved was 21.4 mg/L. • Meanwhile, in BAF2, through an anammox pathway, the nitrogen removal elevated from 8.6 mg/L to 22.8 mg/L concentration. • In the aeration process applied, the maximum total nitrogen removal achieved up to 44.2 mg/L in both BAFs.	[124]
13	Enriched aerobic/anoxic biological filter (EABF)	Actual domestic sewage	• The implementation of two cylindrical biological reactors, which are aerobic and anoxic.	•At HRT of 12 h, the removal efficiencies of NH_4^+-N, TN and COD were 97.6 %, 86.9 % and	[122]

(continued on next page)

Table 3 (*continued*)

No.	Type of treatment	Type of treated water	Treatment condition	Summary of efficiencies	References
			• The volume of iron-based microbial coupling carrier (IBMC) is 4.5 L	85.3 %, respectively, in the EABF-filled IBMC with DO of 3.5 mg/L and reflux ratio of 5.5:1.	
14	Moving bed biofilm reactor (MBBR)	Actual domestic sewage	•This study applied six lab-scale MBBRs with a working volume of 9 L (16 cm in diameter, 45 cm in height) with a hydraulic retention time of 12 h and a DO concentration of 3–5 mg/L.	• Simultaneous biofilm growth enhancement and ammonia transformation was obtained via exogenous N-acyl homoserine lactones. • >20 % of ammonium, >90 % of TOC and nitrate were removed	[123]

membrane with pore diameters in the range of 0.05 to 0.1 μm performs the separation of microorganisms and treated wastewater [101]. Developing a membrane filtration system helps lengthen the solid retention time (SRT) in the bioreactor, thus increasing the biomass concentration and the small size of floc formed by intensive aeration of a membrane [102]. As depicted in Fig. 1, the activated sludge flocs and bacteria are rejected due to the small pores of the membranes, and clean water permeates through the membrane.

As stated by previous researchers, a membrane bioreactor is the hybridization of a membrane process (microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO)) with a biological suspended growth reactor [6,92,103]. In MBR, the process uses a semipermeable membrane to split up contaminants in fluids based on their electric charge or relative size, and it does not change their properties. The residue on the membrane is called the retentate or concentrate, while the filtrate fluid is called the permeate. According to Soni-Bains et al. [90], the benefits of MBR are smaller plant footprints, due to the second clarifier, and the tertiary filtration process is discarded. Also, it can be planned to lengthen the sludge period so that the sludge production can be lowered, increasing effluent quality and loading rate capability. However, biofilms attached to the surface of the membrane could be a limitation of MBR due to the reduction in permeate flux or the elevation of the transmembrane pressure (TMP). This is also called membrane fouling [6].

In 2014, Gao et al. [102] integrated an anaerobic fluidized bed with a membrane bioreactor to treat domestic wastewater. This system achieved 75.8 % removal of COD, and up to 53.1 % of the removed COD was converted into biogas. Hahn and Figueroa [103] also mentioned the ability of an anaerobic baffled reactor to treat domestic wastewater with up to 75 % COD removal capacity. Constructed wetlands showed good performance in treating swine wastewater with ammoniacal nitrogen removal up to 72.9 % and total nitrogen removal up to 70.7 %. The total nitrogen removal was contributed by denitrification, medium adsorption, and plant uptake [106]. Constructed wetlands promise good nitrogen removal efficiency from wastewater [107]. Enhancement of nitrogen removal efficiency using wetlands may be carried out by selecting the appropriate plant species [108,109], augmentation of nitrogen oxidizing bacteria [110,111], optimization of treatment

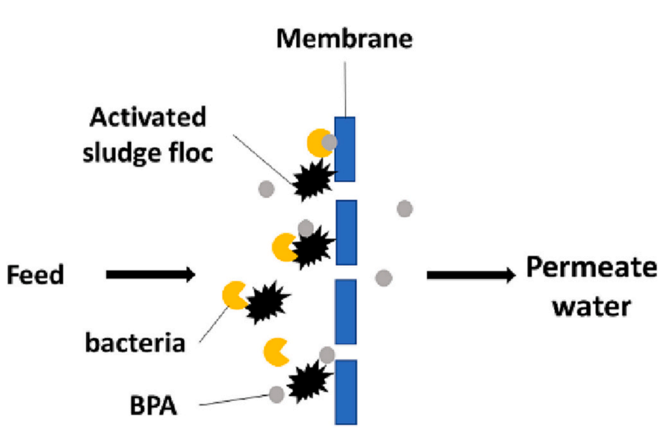

Fig. 1. The schematic diagram of the MBR concept.

conditions [112,113], and integration/hybridization of the treatment processes [114,115].

3.2. BPA removal technologies

The biodegradation of BPA by microorganisms has attracted much attention because it is environmentally friendly and has low operational costs. In this section, the authors focus on the removal of BPA by bacteria. In this treatment, emerging contaminants like BPA are degraded to simpler compounds by microorganisms and could also be biomineralized to carbon dioxide and water [127]. Several treatment technologies applied to BPA removal are constructed wetlands [128], activated sludge processes [129], membrane bioreactors [130], and aerated biological filters [131].

The successful removal efficiency of BPA could be achieved through biological treatment compared to the primary chemically assisted treatment [132]. Biodegradation was prompted by increased nitrification and HRT in the bioreactors. It is noteworthy that the simultaneous removal of ammonium ions and BPA could be achieved because nitrifying bacteria can degrade BPA. A summary of biological treatments for BPA removal is shown in Table 4.

A constructed wetland (CW) is an engineered system of biological-based wastewater remediation imitating a natural wetland, but it is more controllable [133]. The constructed wetland design system includes soil, wetland vegetation, and an accumulated microbial community for wastewater treatment [134]. The CW system comprises biodegradation (biological), sorption (physicochemical), and oxidation (chemical) removal mechanisms that are connected between plants, the soil, and the substrate [128,135]. Initially, the mechanism of pollutant removal takes place on the soil, gravel, and plant roots, where adsorption occurs [136]. Next, the adsorbed contaminants are degraded by microbes in the plant rhizosphere [137]. The plant is protected from pollutant toxicity due to the biodegradation processes of microbes located in the rhizosphere [138]. The design of constructed wetlands may be categorized as horizontal flow (HFCW), surface/subsurface flow (SFCW), and vertical flow (VFCW) [133].

In a vertical flow constructed wetland (VFCW) of lab-scale size, phenol removal from synthetic young and old leachate was >4-tert-butylphenol and BPA [138]. The findings stated that efficient removal of BPA and 4-tert-butylphenol from old leachate is better in the CWs planted with *Phragmites australis* compared to the unplanted CWs. Moreover, the existence of plants would help in the growth of BPA- and 4-t-BP-degrading bacteria and provide a carbon source for other probable microbes in CWs. A CW system is simple, eco-friendly [139], generates little secondary waste, is easy to maintain, and is suitable for implementation in remote areas [140].

In a conventional activated sludge bioreactor, the biomass generated from the growth of microorganisms with the dissolved oxygen existing in aerated tanks during wastewater treatment is called activated sludge [141]. The 'activated' term originated from the large number of bacteria and other microorganisms present in the biomass [142]. Conventional activated sludge worked efficiently compared to other methods (primary settling, chemical precipitation, aeration volatilization) because most of the removal of EDCs was contributed by biodegradation. Previous research found that the microbial community has the greatest influence on BPA elimination in activated sludge [143]. Moreover, non-

Table 4
Summary of biological treatment for BPA removal.

No.	Type of treatments	Type of water	Type of microorganism	Summaries/findings of studies	References
1	Constructed Wetlands (Activated Carbon Medium)	Synthetic Wastewater contaminated with EDCs	–	•98–100 % of BPA removal obtained.	[147]
2	Nitrifying Activated Sludge	Mineral Salt Medium + BPA in acetone	–	•Declines in concentrations of both BPA and NP occurred concurrently with oxidation of ammonium (NH_4^+) into nitrate (NO_3) by nitrifying sludge	[148]
3	Aerobic Granules Sludge in Sequencing Reactors	BPA-rich Wastewater	–	•More than 97 % BPA removal achieved	[149]
4	Bioreactor (peroxidase-mediated bioprocess)	Synthetic BPA solution	• *Pseudomonas* spp. and *Bacillus* spp. • Peroxidase generating bacteria	•Infusing H2O2 to accelerated	[150]
5	Continuous flow reactor (Nitrifying system with immobilized biomass)	Wastewater	•Heterotrophic bacteria	• BPA removal increased from 87.1 ± 5.5 % to 92.9 ± 2.9 % with increased influent BPA concentrations. • AOB community does not affect the BPA concentrations in the influent.	[151]
6	Bioremediation	BPA-treated cultures	•Picocystis sp. (Extremophilic microalgae)	• The experiment was conducted in 500 mL Erlenmeyer Flasks • At 25 and 75 mg/L BPA, the BPA removal efficiencies achieved were 72 % and 40 %.	[127]

BPA degraders, such as ammonia-oxidizing bacteria, promote co-metabolism or the activity of BPA degraders [141]. According to Adav et al. [142], the efficiency of activated sludge could be enhanced by replacing it with aerobic granules. Traditionally, the elimination of BPA in the activated sludge system was good, but the sludge's toxicity was high due to the high concentration of BPA [129].

Next, the biological aerated filter (BAF) system utilizes bioreactors where the biomass is attached to the media in the suspending medium [132]. Wastewater treatment plants (WWTPs) revealed that high removal of BPA was achieved (95 %) compared to activated sludge (68 %). In contrast, the results of BPA removal from municipal WWTPs with a BAF were lower than activated sludge. About 50 % removal of BPA was found after activated sludge treatment.

A comparison between MBR and a conventional activated sludge reactor (CSAR) in treating BPA resulted in MBR being slightly better at removing BPA than CSAR [146]. In addition, the HRT factor was observed to have no significant effect on the removal of BPA by the MBR system. Since low BPA removal was determined by sludge adsorption, the researchers believed that BPA was significantly removed through biodegradation due to the presence of the BPA biodegradation product 4-hydroxy-acetophenone. Ouarda et al. [127] revealed that at low BPA concentrations, the process of BPA removal was done through adsorption onto the activated sludge due to the hydrophobic characteristic of BPA, and only a slight reduction was contributed by biodegradation. In contrast, the high removal efficiency of BPA (99 %) was achieved through biodegradation when the initial BPA concentration was 15 mg/L. The results also revealed that when the BPA concentration was 20 mg/L or greater, the ammonia removal activity by heterotrophic bacteria stopped [130].

4. Ammonia and Bisphenol A (BPA) removal mechanism

4.1. Biodegradation of ammonium ion

Treatments proposed in this review paper focus on the biodegradation of ammonium through nitrification and denitrification. Nitrification and denitrification processes are well-known mechanisms that must be maintained during ammonium ion removal, especially from wastewater. Nitrification and denitrification can occur chemically and biologically [153]. Nitrification is described as converting ammonium ions into nitrate, while denitrification is a stage of transforming oxidized nitrogen into gaseous products. Both nitrification and denitrification rates must be maintained to obtain high removal performance [151]. Simultaneous aerobic-anaerobic treatment was developed to completely degrade ammonium ions in a single reactor [155]. This concept can be achieved by using a multi-stage reactor or bacteria with attached-suspended growth in one system [156]. The nitrification process is mainly carried out by well-known autotrophic bacteria, *Nitrosomonas* and *Nitrobacter*, which degrade ammonium ions into nitrate by obtaining a carbon source from the air and using oxygen as the electron acceptor. The process then continues with denitrification carried out by heterotrophic bacteria, which use nitrate as the electron acceptor while obtaining carbon from other organic materials [155].

4.2. Biodegradation of Bisphenol A (BPA)

BPA is a synthetic organic compound used in plastic materials such as polycarbonate and epoxy resins. The degradation of BPA is mainly conducted using a chemical method. Still, concern is emerging about utilizing such a large amount of organic solvent, which is characterized as a toxic compound to living organisms and the environment. Biodegradation of BPA is an alternative technology to treat BPA waste. Biodegradation of BPA occurs by bacteria and fungi breaking down the chemical structure of BPA into a more straightforward structure and less harmful compounds. Several bacterial degradation mechanisms can even achieve complete degradation of BPA to CO_2 and H_2O.

Several bacteria species are known to have the capability of degrading BPA. Most of the biodegradation has occurred under aerobic conditions. Vijayalakshmi et al. [154] demonstrated the aerobic degradation of BPA using *Pseudomonas aeruginosa* PAb1. The breakdown into phenol and hydroquinone can be achieved under optimized conditions using this species, while a more complex pathway was followed to transform BPA into acetone (Fig. 2). Different species of bacteria may possess other mechanisms for BPA degradation. As described by Kolvenbach et al. [155], *Sphingomonas* sp. strain TTNP3 could also degrade BPA into phenol via complex enzymatic reactions initiated by monooxygenase. Daâssi et al. [156] mentioned a different mechanism of BPA degradation by a fungal species of *Coriolopsis gallica*. The degradation mechanism was activated by BPA oxidative cleavage, which produces several organic acid compounds (Fig. 3). Vijayalakshmi [154] mentioned that *Pseudomonas aeruginosa* PAb1 was able to perform carbon-carbon cleavage of BPA resulting in the formation of 4-hydroxy isopropenyl benzene and isopropenyl benzene as intermediate products, while phenol, hydroquinone, p-hydroxy benzoic acid, and acetophenone were the final degradation products. Hydroquinone and phenol were also mentioned by Kolvenbach et al. [158] as the final products produced by the *Sphingomonas* sp. strain TTNP3 via hydroxylation, carbon-carbon cleavage, and ipso substitution mechanisms. Molkenthin [157] mentioned that by-products of BPA degradation exhibited less acute toxicity to *Vibrio fischeri*. Similarly, Olmez-Hanci [158] also

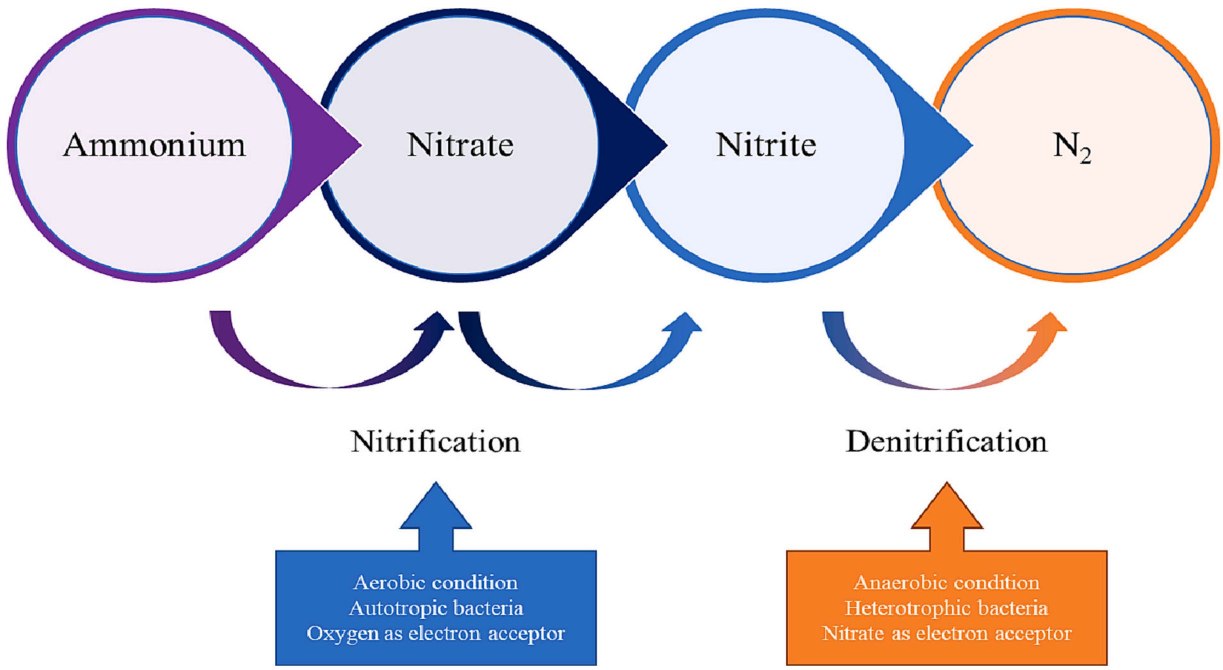

Fig. 2. Pathway of ammonium biodegradation.

mentioned that the degraded BPA presented less toxicity (reduced up to 80 %) to *V. fischeri*. Table 5 summarizes the biodegradation mechanisms of BPA by several bacterial and fungal species.

4.3. Factors affecting the biodegradation of ammonium ion

Simultaneous nitrification and denitrification are proposed as a compact technology to biodegrade ammonium ions in a single reactor. Some factors that affect the efficiency of these processes include the carbon source, dissolved oxygen concentration, pH, initial concentration, and biofilm formation. Also, efficiency is related to diffusion parameters inside the system.

Carbon is the main energy source for denitrifying bacteria. The availability of readily biodegradable carbon increases the simultaneous nitrification and denitrification processes for ammonium ion removal. The ratio of COD/N needs to be kept in a specific range to achieve the highest reduction of ammonium ions. Pochana and Keller [163] mentioned that a ratio of 3.5 to 4.5 is the optimum ratio for simultaneous nitrification and denitrification. He [167] updated this information with the statement that increasing the COD/N ratio results in higher ammonium removal. A COD/N ratio of 40 gave a total reduction of N of up to 92 % [167]. The addition of a readily biodegradable carbon source like acetate was also stated to increase the overall removal of ammonium ions [168].

Oxygen becomes the limiting factor for simultaneous nitrification and denitrification. Nitrification requires aerobic conditions, while denitrification requires anaerobic conditions [169]. At specific dissolved oxygen (DO) concentrations, the rate of nitrification and denitrification might occur equally, thus providing a desirable condition for simultaneous processes. Pochana and Keller [163] mentioned a critical DO of 0.5 mg/L to achieve that concurrent process. DO values higher than 0.5 will result in more nitrification process. Similarly, Gogina and Gulshin [167] also mentioned that DO values ranging from 0.4 to 0.5 mg/L were optimum for simultaneous processes of nitrification and denitrification. He [164] mentioned that a DO concentration of 1.5 mg/L results in the effective removal of N up to 97 %. Liu [168] confirmed that OD ranged from 1 to 1.2 mg/L, while Tan [169] mentioned that the range of 0.5 to 1 mg/L was enough to provide a nitrification process, and denitrifying bacteria could work simultaneously. Thus, this range was preferable to achieve high N removal.

Both nitrifying and denitrifying bacteria work under normal pH conditions, ranging from 6 to 9. The pH range of 7.6–8.5 was favorable for operation conditions during simultaneous nitrification and denitrification for N removal [167,169,173]. The initial concentration of pollutants also affects the biodegradation performance [174]. Increasing the initial concentration decreased the overall removal efficiency [175,176]. This condition was related to the capability of bacteria to tolerate the influent concentration. It was also suggested that nitrifying bacteria can accept shock loading, while denitrifying bacteria cannot [177].

The thickness of the biofilm formed during treatment played an important role in the simultaneous nitrification and denitrification processes. This condition is highly correlated with the mechanisms of oxygen and food diffusion through the biofilm layer [166]. A thick biofilm layer creates a situation where aerobic bacteria are on the biofilm surface and anaerobic bacteria are in the inner biofilm. Thus, simultaneous nitrification and denitrification can be achieved [169]. Increasing the thickness of the biofilm was suggested to perform simultaneous nitrification and denitrification more effectively [166,169], with 80 μm proposed as a median of effective biofilm thickness [166].

4.4. Factors affecting the biodegradation of Bisphenol A (BPA)

Maximum removal of BPA through biodegradation can be achieved by maintaining optimum conditions for the degradation processes. Several factors that affect the biodegradation efficiency of BPA must be considered. Those factors include the initial concentration of BPA, operational pH, dissolved oxygen availability, and microbial species. These factors affect the microbial activity during treatment, which is then correlated with the overall removal of BPA.

Biodegradation using microbial species needs to consider the selected microbe's tolerability and resistivity to the pollutant concentration. Several bacteria may possess the ability to tolerate high concentrations of pollutants, while other species don't. The initial concentration of BPA that will be treated using the biodegradation method needs to be considered, along with the microbial capability to tolerate that concentration. Vijayalakshmi [154] mentioned the

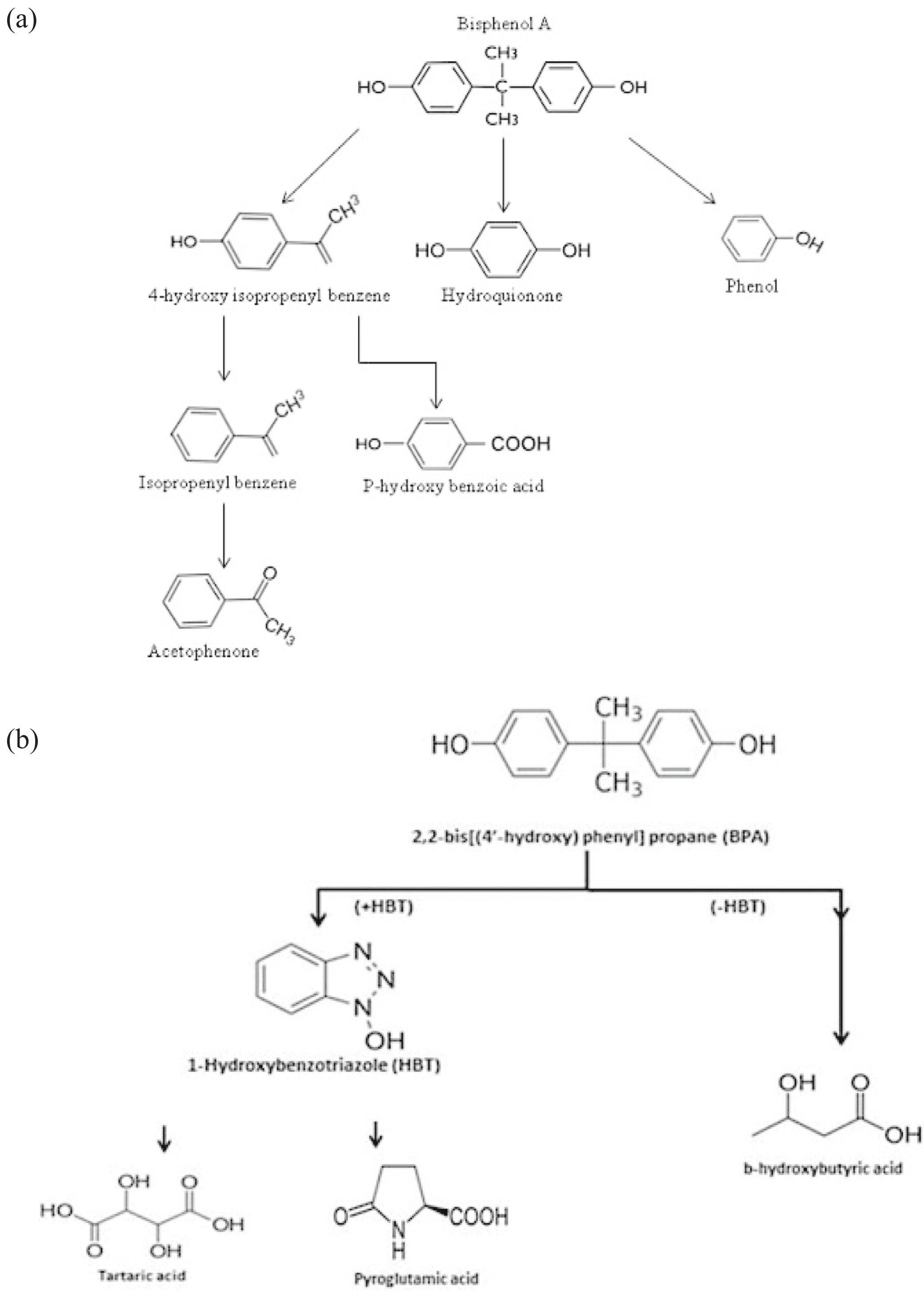

Fig. 3. BPA degradation pathway by (a) *Pseudomonas aeruginosa* PAb1 [157] (b) *Coriolopsis gallica* [159].

decreasing growth of *P. aeruginosa* PAb1 up to 45 % in the exponential phase with increasing the initial BPA concentration from 5 μM to 35 μM. Optimization of the removal efficiency using the Box Behnken Design also showed that increasing the BPA concentration from 10 to 40 μM decreased the overall removal from 100 % to 83.46 %. Treatment of BPA in a batch bioreactor showed a longer time to achieve complete removal as the initial concentration increased, indicating a negative effect of a high initial concentration on the microbes inside [150].

The pH affects the electrostatic interaction of BPA and the adsorbent during the removal using adsorption technology [178] while also affecting the growth of microbial species inside the reactor during the biodegradation. The optimum pH condition is highly related to microbial enzymatic activity. Aravind [176] reported that the base condition of pH was more favorable for biodegradation, with pH 9 showing the optimum electro-oxidation biodegradation condition. In contrast with Aravind [176], many researchers reported that the optimum state of BPA removal using the biodegradation method was under acid conditions. The selection of acid conditions relates to the fact that the enzymatic reactions of most microbes are in the range of pH 4–7. Mokhtar [177] stated that the optimum pH condition for BPA by immobilized

Table 5
Biodegradation mechanism of BPA.

No.	Species	Condition	Mechanism	Intermediate product(s)	Final product(s)	Source
1	*Pseudomonas aeruginosa* PAb1	A batch reactor in basal medium	Carbon-carbon cleavage	4-Hydroxy isopropenyl benzene, isopropenyl benzene	Phenol, hydroquinone, P-hydroxy benzoic acid, acetophenone	[157]
2	*Sphingomonas* sp. strain TTNP3	A batch reactor in a glass vessel system	Hydroxylation, carbon-carbon cleavage, *ipso* substitution	Phenomium ion, quinol, carboncationic compound	Hydroquinone, phenol	[158]
3	*Coriolopsis gallica, Bjerkandera adusta, Trametes versicolor*	Batch reactor using fungal laccases	Laccase catalytic oxidation	Tartaric acid, β-hydroxybutyric acid	Glycerol, β-hydroxybutyric acid	[159]
4	*Pseudomonas* spp., *Bacillus* spp.	Batch bioreactor with nutrient addition	Peroxidase-mediated degradation	–	CO_2, H_2O	[150]
5	Consortium bacteria dominated by *Citrobacter freundii*	A batch reactor in MSM medium	Cytochrome P450, P450 monooxygenase, ammonia monooxygenase, extracellular laccase	Valeric acid, undec-2-enyl ester, Benzophenone, Benzeneacetic acid	Phenol	[162]
6	Consortium bacteria	Batch reactor in MSM medium	Photolytic with light and dark reaction	2,2-Bis(4-hydroxyphenyl)-1-propanol, 1,2-bis (4-hydroxyphenyl)-2-propanol, 4,4-dihydroxy-alpha-methylstilbene, 2,2-bis(4-hydroxyphenyl) propanoic acid, 2,3-bis(4-hydroxyphenyl)-1,2-propanediol, p-hydroxyphenacyl alcohol	*p*-Hydroxybenzaldehyde, *p*-hydroxyacetophenone, *p*-hydroxybenzoic acid, *p*-hydroquinone, hydroxy-BPA	[163]
7	*Bacillus* sp. GZB	A batch reactor in microcosms medium	–	–	Phenolic compounds	[164]
8	*Bacillus* sp. GZB	A batch reactor in mineral medium	Carbon cleavage, hydroxylation, dehydration, demethylation	*p*-Benzenediol, 4-(2-Hydroxypropan-2-yl) phenol, 4-(Prop-1-en-2-yl) phenol, 1-(4-hydroxyphenyl) ethenone, 4-Hydroxybenzaldehyde,	Benzoic acid, 2-Hydroxypropanoic acid, 2-Methylbutanoic acid	[165]

laccase was pH 5. This was indicated by the lowest residual BPA and the highest enzymatic activity. Following Mokhtar [177], Taghizadeh et al. [178] also stated that the optimum pH for BPA removal needs to be maintained at 4.5 due to the high stability and activity of the enzyme. Laccase is the enzyme that plays an essential role in BPA removal [159,180–182], while some research mentions the highest activity of this enzyme in the range of pH 4.5–6 [183–186].

The availability of DO plays an important role in supporting microbial species growth inside the reactor. Most of the BPA biodegradation research was conducted in aerobic conditions. Only a limited number discussed anaerobic biodegradation. Different species of microbes require other conditions of available oxygen to achieve maximum BPA removal. Under aerobic conditions, consortium bacteria isolated from a contaminated lake in Taihu, China, can remove 70 % of BPA from an initial concentration of 10 mg/L [187]. *P. aeruginosa* PAb1 has a promising capability of removing 100 % of the BPA from the 35 μM solution under aerobic conditions [157]. *Bacillus* sp. GZB showed an excellent capability to remove BPA under both aerobic and anaerobic conditions due to its facultative aerobic characteristic. *Bacillus* sp. GZB showed an average removal of BPA, reaching 35.9 % in the various treatment conditions [164,165]. Co-metabolic degradation with a bioelectrochemical process showed that the species *Azoarcus* sp. dominated the anaerobic microbial consortium, with overall BPA removal above 80 % [188]. An anaerobic sludge treatment reactor removed up to 84.1 % BPA from contaminated sludge after 280 days of treatment [189]. The reason behind limited research on the anaerobic biodegradation of BPA is related to the final degradation product. Aerobic degradation can perform complete degradation of BPA producing CO_2 and H_2O. In contrast, anaerobic degradation produces methane as the final product, which still needs further treatment. Due to finding BPA in sludge, anoxic and anaerobic biodegradation of BPA will be very interesting to conduct. The diffusion of oxygen into sludge is minimal, creating an anoxic and even anaerobic condition at a certain depth; thus, more knowledge of this matter will be required.

The microbial species also play an important role in determining the BPA removal processes. Due to the different enzymatic reactions, different species of microbes will perform different pathways in degrading BPA. Certain species, like *Pseudomonas* sp. [157] and *Bacillus* sp. [164] were shown to have the capability to perform solely the degradation of BPA. The utilization of a single microbial species needs to consider its overall performance since microbes do not have a solitary function in their natural ecosystem. Several species need a consortium living environment to perform interlinked mechanisms in executing biodegradation [162–164,189]. The appropriate selection of microbial species, whether single or consortium cultures, as related to the operational condition may result in the highest contaminant removal.

5. Potential of co-biofilm treatment system for simultaneous ammonia and BPA removal

In this review paper, the author would like to introduce the combination of MBBR and water hyacinth (WH), known as a co-biofilm treatment system, to treat ammonia and BPA simultaneously (Fig. 4). The schematic diagram of the modified MBBR is shown in Fig. 5. MBBR

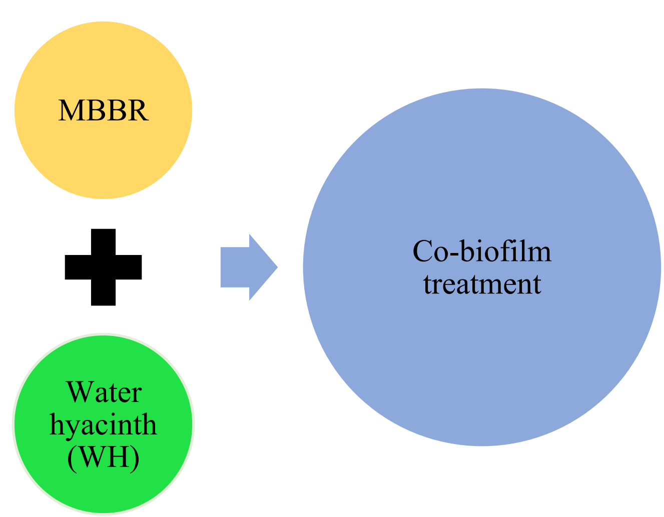

Fig. 4. Combination of MBBR and WH for co-biofilm treatment.

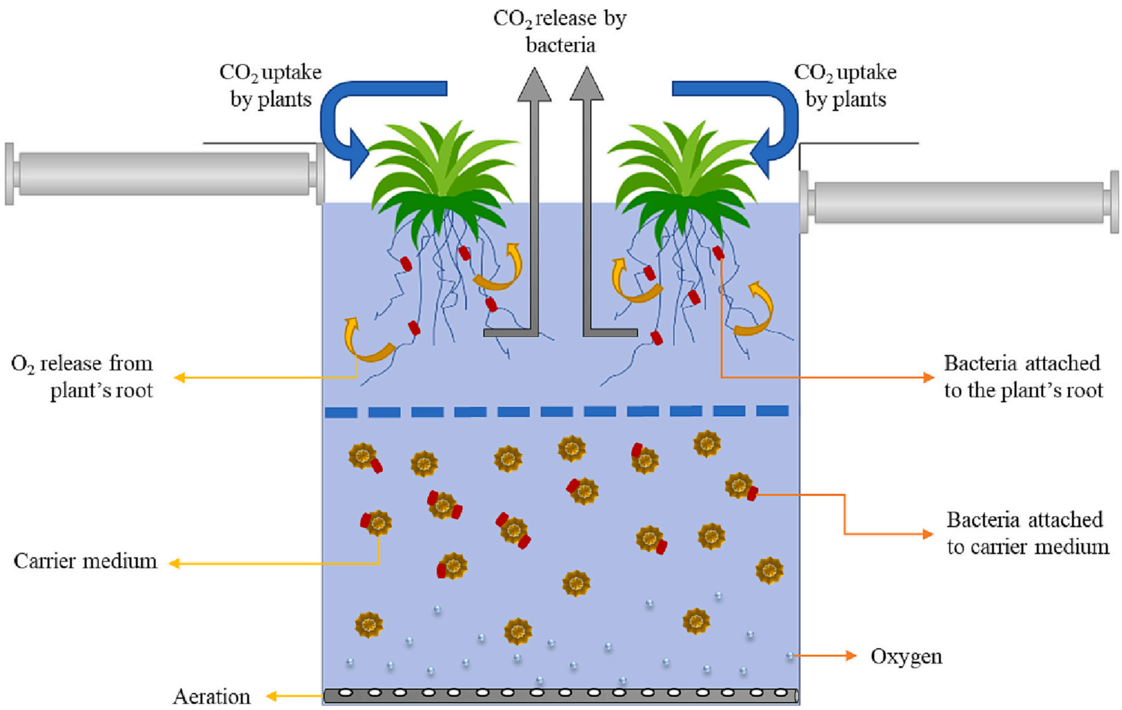

Fig. 5. Schematic diagram of modified MBBR.

Table 6
Summary of MBBR application in wastewater treatment.

Type of wastewater	MBBR configuration and volume	Aeration		Media			Hydraulic retention time (HRT)	Removal performances	Reference
		Flow rate (L/min)	Dissolved oxygen (DO) (mg/L)	Type and shape	Specific surface area (m^2/m^3)	Filling ratio			
Conventional activated sludge (CAS) WWTP effluent	Three identical glass reactors (3/each)	300 L/h	–	Anoxkaldnes™ K5	–	50 %	–	Ammonia = 95 % Atenolol and diclofenac ≥50 %	[204]
Secondary wastewater (ESWW) of municipal wastewater	Three identical jacketed MBBRs R1, R2, and R3 (2.0 L/each)	–	7.4–8.8	**R1** AnoxKaldness-K3 **R2** AnoxKaldness-K5 **R3** AnoxKaldness-M	 500 800 1200	 30 % 30 % 30 %	5.25	**R1** NH_4^+-N = 87.3 %, **R2** NH_4^+-N = 71.8 % **R3** NH_4^+-N = 47.2 %	[80]
Synthetic livestock and poultry breeding wastewater (LBPW)	Two MBBRs (3.0 L/each)	–	4–5	**K-MBBR** K1 cylindrical ring with some short flanks and a cross-like structure **P-MBBR** PVA gel, a very porous material shaped like 4-mm spherical beads, is a typical biomass carrier	500 1000	40 % 40 %		**K-MBBR** COD = 97.88 % NH_4^+-N = 96.03 % TN = 83.96 % **P-MBBR** COD = 96.12 % NH_4^+-N = 82.98 % TN = 77.04 %	[197]
Synthetic recirculating aquaculture systems (RAS) wastewater	Two lab-scale MBBRs (5.5 L/each)	–	5.5–8.5	**R1** AnoxKaldnes K5 **R2** Novel sponge biocarriers SB	 800 NA	 20 % 20 %	6	**R1** Ammonia = 86.67 ± 2.4 % **R2** Ammonia = 91.65 ± 1.3 %	[198]
Synthetic wastewater	Two parallel aerobic MBBR (working volume = 3 L)	–	–	Biological filler (copolymers)	980	40 %	10	Sulfamethoxazole (SMX) = 80.49 % Nitrate-N = 94.70 % Ammonia-N = 96.09 %	[199]
Simulated saline RAS wastewater	The four identical cylindrical reactors (Working volume = 10 L)		>5.5	High-density polyethylene (HDPE) biocarriers (type K5, AnoxKaldnes™)	800	30 %	24	**R4** Nitrite = 99.6 % Ammonia =95.3 % **R1** Nitrite = 94.1 % Ammonia =89.4 %	[200]

is one of the technologies that could efficiently polish pollutants from WWTP effluents. However, the continuous aeration needed for the system makes it not cost-effective. Compared to conventional activated sludge (CAS), one of the disadvantages of MBBR is the increased energy cost due to the need for aeration to promote carrier movement [190]. Besides, continuous aeration has a better nitrification capacity than intermittent aeration. In biofilms, high humic substance/polysaccharide ratios resulted in structural deterioration. Detached biomass had a larger proportion of humic compounds than biofilm. High humic substance/polysaccharide ratios in EPS are caused by intermittent aeration [188]. Biofilm forms on a media surface through a series of steps that include initial attachment, microcolony formation, and biofilm growth and development [192].

Moreover, the purpose of the biofilm depends on the bacteria community developing it [193]. The presence of water hyacinth is believed to help maintain the nitration process during the non-aerated mode of the MBBR. Previous research stated that oxygen was supplied by atmospheric diffusion, algal photosynthesis, and root release from WH [194]. The first two methods directly transfer oxygen into the water column, whereas the attached bacterial biofilm obtains oxygen released by plant roots. The release of oxygen from plant roots appears to be a significant source of oxygen in the lagoons. Besides, the implementation of water hyacinth in wastewater treatment was observed to be excellent in removing nitrogen and phosphorus, even during heavy rainfall [195]. To the best of the author's knowledge, there are no reported studies that have combined these two systems.

5.1. Application of a moving bed biofilm reactor in wastewater treatment

Because of its high nutrient removal and recovery capabilities, MBBR has shown tremendous promise in the circular economy. Furthermore, a number of researchers have used the MBBR method to remediate emerging pollutants [196]. Table 6 presents the application of MBBR in ammonia removal. Previously, Ashkanani [77] studied the influence of bio-carrier types on the operation of MBBR. Three distinct Anox Kaldness bio-carriers (K3, K5 and M) were implemented to treat real wastewater discharge. The findings revealed that the greater surface area of the bio-carrier tends to clog during high and low loading rates for MBBR under the nitrifying process. Moreover, ammonia removal was found to be low for a greater specific surface area of bio-carrier, resulting from the constraint of mass oxygen transfer.

Additionally, the effect of carrier type and C/N ratio has a significant impact on the nitrogen removal (NR) performance of MBBRs inoculated with heterotrophic nitrification-aerobic denitrification (HN-AD) microbes [197]. Two types of bio carriers, polyvinyl alcohol gel (PVA) and Kaldness (K1), have shown different nitrogen removal (NR) efficiencies at different C/N ratios. At a C/N ratio of 10.96 %, nitrogen removal efficiency was achieved with K1 as the carrier. However, by using a PVA carrier, the performance of NR was observed to be more stable than with K1 at various C/N ratios because the porosity of PVA gel was more favorable for the growth and augmentation of HN-AD bacteria, particularly *Acinetobacter* and *Paracoccus*, which had the highest species richness (16.37 %) at low C/N ratios. The utilization of an innovative sponge bio-carrier (SB) in MBBR was performed and compared with K5 plastic carriers to treat recirculating aquaculture system wastewater [198]. The ammonia removal efficiency achieved by SB in the MBBR was 91.7 %, which was high compared to K5 (86.7 %) at an optimum HRT of 6 h. *Nitrosomonas* and *Nitrospira* were the most common genera in the nitrifying community, coexisting with the heterotrophic genera *Hyphomicrobium*, *Mesorhizobium*, *Zhizhongheella*, and *Klebsiella* spp. Furthermore, the microbial community analysis revealed that SB has good biocompatibility, which encourages the growth of Nitrospira and Nitrosomonas in co-existence with denitrifying bacteria, resulting in the higher nitrification achievement of the bioreactor.

The removal of nitrogen and sulfamethoxazole (SMX) degradation occurred simultaneously in the MBBR bioaugmented with the *Achromobacter* strain JL9 [199]. The research revealed that the bioaugmented MBBR functioned effectively compared to the non-bioaugmented reactor, where the results showed high removal of ammonia-N, nitrate-N, and SMX. Besides that, the C/N ratio affected the relative abundance of the dominant genus, *Achromobacter*, in the MBBR system. The findings stated that the C/N ratio influences the number of sulphonamide resistant strains. In addition, the bacterial community formations in the MBBR system were adjusted by bioaugmentation and the C/N ratio. The bioaugmentation strain does not affect the stability of SMX degradation efficiency. Bioaugmentation contributes more to SMX degradation than the C/N ratio.

The removal of harmful nitrite and ammonia from recirculating aquaculture wastewater in MBBR using an expeditious method has been studied recently [200]. In the study, four MBBRs were filled with various fresh and mature biofilm carrier mixtures. The findings indicated that the reactor filled with mature biofilm showed higher removal of nitrite and ammonia compared to the one filled with fresh biofilm. In addition, the mature biofilm reactor showed a rapid rate of ammonia removal as compared to another reactor. The most plentiful heterotrophic bacteria in all reactors studied were *Planktosalinus*, *Marinobacter* and *Nitrospina*. In conclusion, using mature biofilms in MBBR has increased bacterial diversity while improving nitrite and ammonia removal efficiency. MBBR with mature biofilms performed exceptionally well in removing high nitrite and ammonia levels in 56 days or less.

To date, research conducted on the removal of BPA using MBBR has not been found. Considering its great performance in treating ammonia, simultaneous removal of ammonia and BPA may also be obtained using a MBBR. Hybridization of this technology to achieve higher removal performance can be conducted by integrating it with other treatment processes [201,202], while modification of parts inside the MBBR may also become an alternative option [203].

5.2. Application of water hyacinth (WH) in wastewater treatment

Eichhornia crassipes also known as water hyacinth (WH) is an invasive macrophyte plant. This plant had been extensively used for purification during wastewater treatment. Compared to other aquatic plants, the effectiveness of pollutant removal is higher [205]. Nonetheless, studies on its efficacy in the elimination of BPA are limited. However, BPA was believed to have been largely removed with the aid of WH. This is based on the results of 52.1 % BPA removal in a constructed wetland planted with WH [140]. Moreover, previous studies found that the presence of WH greatly expedited the removal of pesticides in water, with removal efficiency ranging from 66 to 79 % after 30 days of treatment [206]. Meanwhile, the performance on the elimination of ammonia by water hyacinth in wastewater treatment was excellent.

A constructed wetland planted with WH was operated to treat effluent from the oxidation pond of domestic wastewater treatment [207]. The presence of WH improved the removal of nitrogen by 29.4 %. The high ammonia removal efficiency of 81 % was attributed to the increased total nitrogen removal efficiency. Bacteria can eliminate ammonia during growth of periphyton and phytoplankton, uptake by WH, nitrification, and the release of ammonia gas into the atmosphere. The buffering effect of WH, which reduces pH in the marsh to near neutral values, makes the latter method implausible. A low nitrification rate is expected because of the low DO levels of roughly 2.0 mg/L in the influent and 0.5 mg/L in the effluent. Nitrification is relatively slow at DO concentrations <2 mg/L. Still, the reaction may proceed more quickly in the rhizosphere, where DO concentrations are higher due to oxygen transport and release through the air spaces (aerenchyma tissue) of macrophytes like *Eichhornia crassipes* stems and root zones. The rate of nitrification decreases dramatically below pH 6.0 and is almost nonexistent above pH 8.0.

Fazal [205] applied CW using WH to treat mixed industrial wastewater that contained high concentrations of heavy metals (Cd, Ni, Hg, and Pb), nutrients (phosphates and ammonia), and COD. The constructed wetland with a water hyacinth plant was able to remove pollutants. The hydrophytes (WH) demonstrated their ability to thrive in high nutrient concentrations while also removing considerable amounts of nutrients. Compared to anaerobic sludge (51.7 %), the wetland containing water hyacinth was more effective at eliminating ammonia by 71.6 %.

Water hyacinth and water lettuce (*Pistia stratiotes*) were used in studies to remove nutrients from domestic wastewater [209]. *P. stratiotes* outperformed *Eichhornia crassipes* in decreasing nitrate-nitrogen and ortho-phosphates, whereas WH does better than *P. stratiotes* in terms of reducing ammoniacal-nitrogen (AN) and nitrite-nitrogen. On average, WH lowered the NH_4-N concentration by 72 %, *P. stratiotes* reduced it by 83 %, and the control concentration was reduced by 95 %. Algae dramatically decreased the content of NH_4-N.

The best conditions for removing AN from wastewater (semiconductor effluent) utilizing a WH-based phytoremediation technique were explored [210]. The experimental design was a face-centered central composite design (CCD) in which four operational variables were studied (pH, retention time, macrophyte density, and salinity) as well as five responses (AN removal efficiency, fresh biomass growth, COD, BOD, and TSS). The maximum AN removal efficiency of 77.48 % (beginning AN concentration = 40 mg/L) was achieved through numerical optimization at the following optimum conditions: pH 8.51, retention duration of 8.47 days, macrophyte density of 21.39 g/L, and salinity of 0 g NaCl/L. The values predicted by the models coincided adequately with the experimental values, implying that the response surface approach was trustworthy and practicable for developing experimental designs employing phytoremediation process optimization.

Qin et al. [195] used self-designed fabrications in situ on a pilot scale for 30 days to examine the efficacy of phytoremediation of urban wastewaters by WH under an extreme rainfall event. The findings imply that WH has significant N and P removal capabilities, even in the presence of low DO concentrations (1 mg/L) and high ammonium ion concentrations (NH_4^+-N > 7 mg/L). Even during extreme precipitation events, WH can be used for water treatment to reduce the levels of NH_4^+-N, dissolved organic nitrogen (DON), and phosphate (PO_4^{3-}). Furthermore, during the high rainfall event, DO rose due to wet deposition, runoff, and surface flows, resulting in alterations in nitrification and denitrification processes, which drastically altered nitrogen forms in urban wastewater. Significantly, NH_4^+-N and DON were almost fully removed (>99 %) from wet deposition, runoff, and surface flows during high precipitation. Still, this removal was hampered by increased DO from wet sediment, runoff, and surface flows.

The effectiveness of sequencing-batch mode constructed wetlands (CW) in treating synthetic wastewater approximating low-strength sewage (CWs) was studied [211]. Six CWs with three substrates (gravel, light expanded clay, and clay bricks) were planted with WH, one for each substrate, to test the practicality of utilizing a floating macrophyte in CWs and determine the most optimal substrate. WH improved COD removal in gravel systems, boosting removal efficiency from 37 % in the unplanted system (CWG—U) to 60 % in the planted system (CWG—P). The vegetated CW with clay bricks (CWB-P) performed best for TKN and TAN removal, with 68 and 35 % maximum removal efficiencies, respectively. Plant direct absorption ranged from 4 to 74 % of total nitrogen removed and from 26 to 71 % of total phosphorus removed in CWG—P, CWC-P, and CWB-P. As a result, bricks were the best material for CW treatment performance, showing the possibility of their usage as a low-cost substrate. The application of WH influenced the removal of COD in the gravel system (CWG—P) and TAN in the clay brick system (CWB-P). On the other hand, direct plant absorption ranged from 4 to 74 % of total nitrogen removed and 26 to 71 % of total phosphorus removed in CWG—P, CWC-P, and CWB-P. In conclusion, plant uptake was most likely the predominant method for nutrient removal in the assessed CW with inert substrates (gravel and LECA). In contrast, the adsorption of nitrogen and phosphorus onto the substrate was the most important in the presence of clay bricks.

Aquatic plants are extremely important in wastewater purification. According to Ismail et al. [209], WH can be used to extract nutrients from wastewater. The WH phytoremediation treatment reduced pH, COD, and ammonia nitrogen. Compared to the literature, the majority of the reduction occurred on day 12 and was connected with the WH optimal growth rate, which was day 15. The steady mode of operation was discovered to delay the optimum growth of WH; however, it did not affect wastewater's nutrient removal rate or efficiency [205].

Furthermore, based on the findings, it is possible to deduce a correlation between nutrient intake (removal rate) and biomass growth rate. COD was reduced from 135 to 2 mg/L, which is considered a 95 % reduction. Compared to the standard, a similar reduction was observed for ammoniacal nitrogen (85 %) from 6.1 to 0.3 mg/L.

5.3. Future perspectives and challenges of co-biofilm treatment

In recent years, the practicality of MBBRs has been explored as an alternative approach to polish micropollutants from contaminated effluent. They are more cost-effective than ozone and activated carbon and more reliable than CAS in their biodegradation of pharmaceuticals and other micropollutants [213]. With its small footprint and high-performance capability in carbon and nitrogen removal, an MBBR is an excellent choice for small decentralized facilities or upgrades to existing facilities [214]. However, MBBR face slow start-up and poor treatment effectiveness, particularly at low temperatures [126].

Co-biofilms technology, by the addition of WH, provides a longer microbial retention time, reducing the microbial washout and promoting the growth of slow-growing microbes such as nitrifying bacteria and NA degraders. This benefit can be achieved by selecting the appropriate root's length of WH and controlling the DO inside the system. Furthermore, *co*-biofilm is frequently composed of more dense extracellular polymeric substances (EPS) than microbial floc; EPS protects against environmental stress [215] which may enhance the system's performance, especially during start-up.

Meanwhile, WH is one of the microphytes used in the phytoremediation process due to its great potential. The cover of WH over the water surface hinders sunlight and oxygen from penetrating the surface, slowing the photosynthesis of submerged aquatic plants to produce oxygen to support marine life. Thus, controlling WH growth is critical to preventing damage to the underwater system [217]. It is best to avoid using a WH plant to treat wastewater with a high phosphorus level. Phosphorus concentrations of 20 mg/L aided WH growth, whereas >40 mg/L phosphorus caused WH toxicity. Another limiting factor for water hyacinth development is high salinity in the water medium [217].

Considering that research using MBBR for BPA removal is currently lacking, the modification of MBBRs with WH may open a new possibility for good simultaneous removal of ammonia and BPA from wastewater [218]. However, the disadvantages of adopting MBBR in wastewater treatment are inadequate aeration and low energy efficiency in carrier mobilization [222]. In order to overcome this issue, the roots of WH substitute for the medium inside MBBR, thus facilitating the attachment and growth of bacteria as co-biofilm medium and supplying oxygen for the system via root transfer [219], while simultaneously performing uptake of pollutants [220,221]. Analysis of the operational conditions is suggested to be conducted in this initial stage. Research related to the number of WH to be used, pollutant loads, operational pH, and DO are crucial parameters affecting the performance of the co-biofilm reactors. This theoretical approach seems very promising for obtaining great simultaneous removal of organic compounds, nutrients, and micropollutants. Additionally, parametric optimization can be performed to obtain the optimum condition for treatment [112,113] following the application of MBBR-WH.

6. Conclusions

This review explains the importance of eliminating ammonia and BPA from wastewater. Biological treatment was highlighted in this paper as it is environmentally friendly and cost-effective. MBBR and WH applications for removing ammonia were observed to work effectively. Besides, the combination of MBBR and WH was recommended to enhance the polishing of ammonia and BPA from wastewater. This combination would enhance the aeration for MBBR and overcome the slowness of WH phytoremediation. Further research is required to determine the new system's effectiveness, which is co-biofilm treatment for reducing pollutants in wastewater, especially nitrogen compounds and micropollutants.

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.

Data availability

No data was used for the research described in the article.

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