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Recent advances in constructed wetlands methane reduction: Mechanisms and methods

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Constructed wetlands (CWs) are artificial systems that use natural processes to treat wastewater containing organic pollutants. This approach has been widely applied in both developing and developed countries worldwide, providing a cost-effective method for industrial wastewater treatment and the improvement of environmental water quality. However, due to the large organic carbon inputs, CWs is produced in varying amounts of CH_4 and have the potential to become an important contributor to global climate change. Subsequently, research on the mitigation of CH₄ emissions by CWs is key to achieving sustainable, low-carbon dependency wastewater treatment systems. This review evaluates the current research on CH₄ emissions from CWs through bibliometric analysis, summarizing the reported mechanisms of CH₄ generation, transfer and oxidation in CWs. Furthermore, the important environmental factors driving CH₄ generation in CW systems are summarized, including: temperature, water table position, oxidation reduction potential, and the effects of CW characteristics such as wetland type, plant species composition, substrate type, CW-coupled microbial fuel cell, oxygen supply, available carbon source, and salinity. This review provides guidance and novel perspectives for sustainable and effective CW management, as well as for future studies on CH_4 reduction in CWs.

KEYWORDS

constructed wetland, methane reduction, greenhouse gas, methanogen, methanotrophy main topic

Introduction

With rapid economic and industrial development, global climate change has become an increasingly critical concern, driven by the excessive emission of greenhouse gases (GHGs) such as carbon dioxide (CO₂), methane (CH₄) and nitrous oxide (Dreyfus et al., 2022). GHG emissions are continually increasing worldwide, resulting in a large amount of research which focused on methods to control GHG emissions, and the development of low-carbon systems (Nuamah et al., 2020).

Wastewater treatment is one of the most resource-intensive industrial practices. Constructed wetlands (CWs) are a well-established low-cost, energy-saving, multifunctional approach to sustainable wastewater treatment, that have been widely used for the treatment of various polluted water bodies (Yu et al., 2020, 2021). However, due to the large scale of wastewater discharged, the

widespread use of CWs would have an obvious environmental consequence in terms of GHG emissions. Studies have shown that the amount of GHGs emitted from CWs is 2-to 10-fold higher than from natural wetlands (Maltais-Landry et al., 2009). Current atmospheric CH₄ concentrations are more than 2.5-fold higher than pre-industrial levels (Zhang Q. et al., 2020), which is concerning as the global warming potential (GWP) of CH₄ in the carbon cycle is 34-fold stronger than that of CO₂. Therefore, despite CH₄ being present at much lower atmospheric concentrations than CO₂, its growth rate is considerably larger, making it one of the most important GHGs contributing to global warming (Guerrero-Cruz et al., 2021). Average CH₄ emissions are extremely variable by reviewing 158 studies data, ranging from 0.15 to 5,220 mg/ m²/h, which can disrupt earth radiation levels (Mander et al., 2014) It has been reported that a 1-fold increase in atmospheric CH₄ concentration would lead to tropospheric surface warming by 0.2-0.3 degrees, presenting a serious risk to human and environmental health (Dreyfus et al., 2022). CH₄ emissions originate from both anthropogenic and natural sources, with wetland ecosystems being the largest natural source, generating annual global CH₄ emissions of 177-284 Tg (Zhang et al., 2021b), 82% of which originate from CWs worldwide (Nuamah et al., 2020). CH₄ has become an important aspect of global carbon reduction, due to its potentially considerable role in future warming. According to the latest IPCC report (Dreyfus et al., 2022), in order to achieve the global temperature rise control target of 1.5°C, CH₄ emissions should be reduced by one-third by 2030 and nearly half by 2050. Achieving this goal is a necessary requirement for sustainable social and economic development (Zhang Q. et al., 2020). and effectively controlling CH₄ emissions from CWs is an essential aspect of reducing global CH₄ emissions.

The CH₄ emission by CWs is the terminal product of various processes in the production, transport, and oxidation of organic matter under anaerobic conditions (Shao et al., 2020). Methanogens and methanotrophs are important microorganisms that mediate functional communities in CWs, and are closely connected with the metabolism of CH₄ and carbon cycle processes (Bridgham et al., 2013). Disruption to the CH₄ source-sink balance of CWs is a direct driver of dramatic increases in atmospheric CH₄. In CW ecosystems, plants use assimilation to fix inorganic carbon from both the air and the water column (converting it into organic carbon), while also fixing organic carbon in the water column through substrate sequestration and uptake, resulting in wetlands serving as a carbon sink (Liu et al., 2022). When wetlands are subjected to long-term anaerobic conditions, plant debris, litter and the substrate gradually convert macromolecular organic matter into CH₄ and CO₂, which is released into the atmosphere through anaerobic microbial metabolic activities, resulting in wetlands also serving as a carbon source (Yan et al., 2012). CO₂ released from CWs can be captured from the atmosphere through photosynthesis, with the CO₂ fixed within plant biomass no longer contributing to the long-term carbon sink, and CO2 emissions from CWs are not considered as GHG because they are the natural fate of organic matter (Guo et al., 2020). The abundance, composition and activity of methanogens and methanotrophs are important determinants of CH4 emissions from CWs (Ji et al., 2021). In view of this, the growth of methanogens should be inhibited in CWs, ensuring a suitable environment is provided for the survival of methanotrophs, maintaining maximum conversion of the generated CH₄ to CO₂ and subsequently, reducing the contribution of CWs to the GWP. To stimulate the growth of methanotrophs, plant cover serving as a powerful carbon sink is used in horizontal subsurface flow CWs (HSSFCWs) which allow O₂ transported by the aerenchyma of plant roots (Mander et al., 2008). It is confirmed that plant diversity can also increase carbon sequestration in the substrate and substrate-based carbon sequestration not only completely offsets GWP based on CH4 and nitrous oxide emissions, but also simulates the conversion of CWs from a carbon source to a carbon sink (Du et al., 2018). Carbon uptake in vegetated wetlands decreases with increasing levels of salinity, mainly due to the inhibition of plant productivity (Sheng et al., 2015). Microbial transport and transformation are the main reason for the high carbon source consumption of CW-coupled microbial fuel cell (MFC) systems, with these processes regulated by microbial competition driven by environmental aspects, providing a novel approach to controlling CH₄ emissions from wetland systems (Liu et al., 2022). The contributions of carbon "source" and "sink" functions in CWs, as well as their relationship and interactions, are crucial to the material and energy cycles within CW ecosystems, global carbon dynamics and global climate change trends (Dreyfus et al., 2022).

If CWs are designed only to consider pollutant removal, the process may not fit with the current double-carbon philosophy, highlighting the importance of constructing effective wetland systems, that can achieve high pollutant removal performance while minimizing CH_4 emissions. There remains a lack of research on CH_4 emissions from CWs, making it difficult to accurately determine the mechanisms and processes of CH_4 emission from CWs. Furthermore, the important environmental control factors have not been comprehensively established, reducing the validity of CH_4 emission reduction measures, and limiting the effectiveness of CWs. Therefore, this review was conducted to explore the available research in this field.

Bibliometric analysis

Data collection sources

Data was collected using the Web of Science (WOS) Core Collection database, with this review only considering the Science Citation Index Expanded (SCI-Expanded). Based on the findings of previous research by Yu et al. (2022), the keywords used to screen the available research on CH4 in CWs included ("constructed wetland*" OR "treatment wetland*" OR "engineered wetland*" OR "artificial wetland*" OR "reed bed*" OR "man-made wetland*") and ("methane sequestration*" OR "methane capture*" OR "methane emission*" OR "CH₄ capture*" OR "CH₄ sequestration*" OR "CH₄ emission*" OR "methane reduc*" OR "CH4 reduc*"). The literature type was restricted to "article" and "review," with publications in English from 1991 to December 2021 included (1991 was the earliest publication date available in the database). After the initial data search, 108 articles investigating CH4 in CWs were selected. The analysis of these publications were performed using Microsoft Excel (2019), charts were generated using Origin (2019 learning version) and the S-curve was prepared using the logistic model in Loglet Lab 4. Analysis of keyword co-occurrence were performed using VOS viewer software (version 1.6.15). After exporting the 108 publications from the WOS platform in plain text form, the author keyword co-occurrence function of VOS viewer software was used to analyze keyword co-occurrence, with the thesaurus files then were merged (such as CH₄, methanes replaced with methane) and the minimum number of co-occurrences set to 3.

Analysis

Publication trend

The publication trend for studies on CH₄ in CWs is shown in Figure 1. In the long term, the number of publications has continually increased with fluctuations, reflecting the growing concern within the scientific community about the effect of CH4 emissions and the need to actively reduce CH₄ emissions in order to maintain global ecological security (Kasak et al., 2020). Prior to 2004, the publication number was relatively low and did not increase significantly from 2005 to 2013. However, a notable increase was observed after 2014, finally reaching 108 articles in 2021. Although the number of publications on this topic remains relatively low, work in this field is gaining importance constantly, as shown by the S-curve (R^2 =0.984) in Figure 1, which indicates that research is in the growth stage and is expected to reach the inflection point in 2026 and remain stable until 2040. Therefore, publications in this field are not expected to reach saturation over the next 15 years, highlighting the high potential for innovation and development. In this sense, a comprehensive review of the current state of knowledge is essential to help stimulate and guide future development and research on CH₄ in CWs.

Main topic

The results of keyword co-occurrence analysis are shown in Figure 2. The 6 most frequently co-occurring keywords were intercepted,

showing that research on the effects of CH_4 on GWP, plants, and microorganisms was closely linked to studies on CH_4 emissions. Studies by Chen et al. (2020c) and Maucieri et al. (2019) attracted attention as they found that different plant species were able to exert varying effects on CH_4 emissions. Furthermore, it has been shown that microbial diversity and abundance are a critical factor affecting CH_4 fluxes, resulting in the need for further research and validation (Zhang et al., 2021a). MFC technology is a promising approach to the control of CH_4 emissions from CWs. For example, Liu et al. (2022) reported that microbial competition in a CW-MFC system can convert unstable carbon sources to CO_2 rather than CH_4 , which can considerably reduce the contribution of CWs to global CH_4 emissions.

Production, transport, and oxidation of CH_4 in CWs

Production of CH₄ in CWs

Plants

Plants play a vital role in CH_4 emissions, however due to contradictory results having been reported, the production of CH_4 by plants was previously only considered in terms of a channel for soilatmospheric gas exchange. However, some terrestrial plants were first demonstrated to release CH_4 under aerobic conditions by Keppler et al. (2006), with more recent studies proving evidence that lignin, pectin, and cellulose can all serve as precursors for CH_4 formation





(Bruhn et al., 2012). The production of CH_4 by plants has been reported as a defense strategy against environmental stress factors, such as cutting damage, increased temperatures, UV radiation, and the disturbance of cytochrome c oxidase activity (Zhang et al., 2012). Once plants trigger stress responses, ROS, such as O_2 ⁻⁻ and H_2O_2 , can be overproduced within plants, further exacerbating the degradation of cellular material, leading to the production of CH_4 (Bruhn et al., 2012; Wang et al., 2023). In addition, some of the identified CH_4 producing terrestrial plants are used in CW applications, such as *Phragmites australis* and *Thalia dealbata*. However, CH_4 emissions from wetland hydrophyte plants under aerobic conditions have not been investigated to date, highlighting

a key gap in knowledge that requires future research.

Microorganisms

Anaerobic microorganisms play the major role in CH_4 production. Under anaerobic conditions, anaerobic hydrolytic microbes, fermentative microbes, and hydrogen-producing acetogens can decompose organic matter from wastewater, substrate materials, and plant biomass (Liu et al., 2022), forming simple inorganic (e.g., CO_2 and H_2) and organic compounds (e.g., acetate; López et al., 2019), that are subsequently converted to CH_4 by methanogens (specialized anaerobic archaea; Malyan et al., 2016). Anaerobic environments occur widely in the substrate layer of CWs. For example, HSSFCWs are designed to purify wastewater through anaerobic pathways (Engida et al., 2020), while anaerobic microzones have been identified in surface flow CWs (SFCWs) due to water flow layering over substrate, and vertical subsurface flow CWs (VSSFCWs) due to longterm operation causing microbial oxygen (O₂) consumption rates to exceed the reoxygenation rate (Maucieri et al., 2017). Methanogens are divided into seven orders, belonging to Euryarchaeota (Table 1). Most methanogens are hydrogenotrophic, with only Methanosarcinales being acetoclastic (Table 1; Bridgham et al., 2013; Zhang et al., 2021c). Methanosarcina are fast-growing organisms that can utilize high acetate concentrations, in contrast to Methanosaeta (Lu Y. et al., 2015). Newly discovered methanogens have been classified as belonging to Euryarchaeota (such as Methanomassiliicoccales, Methanofastidiosa, and Methanonatronarchaeia; Dridi et al., 2012; Nobu et al., 2016; Sorokin et al., 2017), as well as non-Euryarchaeota (such as Verstraetearchaeota, Bathyarchaeota, and Geoarchaeota (Evans et al., 2015; Vanwonterghem et al., 2016; Guo et al., 2020). Based on the results of this review, three methanogenic metabolic pathways exist in CWs: H₂/CO₂ reduction, methyl cracking, and acetate fermentation. The hydrogenotrophic pathway is more energetically favorable to methanogenesis than the acetoclastic pathway (Zhang et al., 2018a), although the acetoclastic pathway has been reported to dominate in freshwater wetland ecosystems, accounting for more than 67% of CH₄ emissions (Zhang et al., 2018b). Novel methanogens have also been found to utilize a fourth methanogenic pathway, involving the reduction of methyl compounds by H₂, as originally observed in *Methanobacteriales* (Dridi et al., 2012) and Methanomicrobiales (Sprenger et al., 2007), then later found in Methanomassiliicoccales (Dridi et al., 2012), Methanofastidiosa (Nobu et al., 2016), Bathyarchaeota (Evans et al., 2015), and Verstraetearchaeota (Vanwonterghem et al., 2016). The specific equations are shown in Eqs 1-7.

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \tag{1}$$

TABLE 1 Taxonomy of major methanogens in *Euryarchaeota* phylum.

Class	Order	Family	Genus	Major CH₄ production pathway
Methanobacteria	Methanobacteriales	Methanobacteriaceae	Methanobacterium	Hyrogenotrophic,Methylotrophic
			Methanobrevibacter	Hyrogenotrophic, Methylotrophic
			Methanosphaera	Hyrogenotrophic, Methylotrophic
			Methanothermobacter	Hyrogenotrophic, Methylotrophic
		Methanothermaceae	Methanothermus	Hyrogenotrophic, Methylotrophic
Methanococci	Methanococcales	Methanococcaceae	Methanococcus	Hyrogenotrophic, Methylotrophic
			Methanothermococcus	Hyrogenotrophic, Methylotrophic
		Methanocaldococcaceae	Methanocaldococcus	Hyrogenotrophic
			Methanotorris	Hyrogenotrophic
Methanomicrobia	Methanomicrobiales	Methanomicrobiaceae	Methanoculleus	Hyrogenotrophic
			Methanomicrobium	Hyrogenotrophic
			Methanofollis	Hyrogenotrophic
			Methanogenium	Hyrogenotrophic
			Methanolacinia	Hyrogenotrophic
			Methanoplanus	Hyrogenotrophic
		Methanospirillaceae	Methanospirllum	Hyrogenotrophic
		Methanocorpusculaceae	Methanocorpusculum	Hyrogenotrophic
		Methanoregulaceae	Methanolinea	Hyrogenotrophic
			Methanoregula	Hyrogenotrophic
			Methanosphaerula	Hyrogenotrophic
		Methanocalculaceae	Methanocalculus	Hyrogenotrophic
	Methanosarcinales	Methanosarcinaceae	Methanosarcina	Hyrogenotrophic, Aceticlastic, Methylotrophic
			Methanococcoides	Aceticlastic, Methylotrophic
			Methanohalobium	Aceticlastic, Methylotrophic
			Methanohalophilus	Aceticlastic, Methylotrophic
			Methanolobus	Methylotrophic
			Methanomethylovorans	Methylotrophic
			Methanimicrococcus	Methylotrophic
			Methanosalsum	Methylotrophic
		Methanosaetaceae	Methanosaeta	Aceticlastic
		Methermicoccaceae	Methermicoocus	Methylotrophic
		Methanotrichaceae	Methanothrix	Aceticlastic
	Methanocellales	Methanocellaceae	Methanocella	Hyrogenotrophic
Methanopyri	Methanopyrales	Methanopyraceae	Methanopyrus	Hyrogenotrophic
Thermoplasmata	Methanomassiliicoccales	Methanomassiliicoccaceae	Methanomassiliicoccus	Methylotrophic

Malyan et al. (2016).

$$4CH_3OH \rightarrow 3CH_4 + CO_2 + 2H_2O \tag{2}$$

$$2(CH_3)_2S + 2H_2O \rightarrow 3CH_4 + CO_2 + 2H_2S$$

$$2(CH_3)_2 NH + 2H_2O \rightarrow 3CH_4 + CO_2 + 2NH_3$$
(4)

$$4CH_3COCOOH + 2H_2O \rightarrow 5CH_4 + 7CO_2 \tag{6}$$

 $\mathrm{CH}_3\mathrm{COOH} \rightarrow \mathrm{CH}_4 + \mathrm{CO}_2$

$$CH_{3}OH + H_{2} \rightarrow CH_{4} + H_{2}O$$
⁽⁷⁾

(3)

(5)

Transport of CH₄ in CWs

The transport of CH₄ occurs mainly via three processes: (1) direct CH₄ transport through molecular diffusion from the water and substrate column; (2) direct CH₄ transport through the ebullition flux process from substrate column; and (3) plant-mediated CH₄ transport from the substrate column via plant aerenchym (Figure 3; Thangarajan et al., 2013; Kasak et al., 2020). Ebullition plays a major role in direct CH₄ transport process, producing three-fold more CH4 fluxes than molecular diffusion by Bonetti et al. (2021), while plant-mediated CH₄ transport is the dominant mode of CH₄ release among three processes (Liu et al., 2017), accounting for about 70% of the total- CH_4 emissions (Li et al., 2010). Moreover, the plant-mediated transport can also transport O2 to plant organ (Silvey et al., 2019). Therefore, investigations into the plant-mediated transport process are required to further our understanding of the role of plants in CW CH4 contributions. Plant-mediated CH4 transport mechanisms can be classified as molecular diffusion or convective transport processes. Molecular diffusion rates depend primarily on the CH4 gradient between plant roots and above-ground parts, along with the interior of the plant organ and the atmosphere, with the capacity for diffusion being susceptible to ambient temperatures (Joabsson et al., 1999). In contrast, gas movement by convective transport relies on the pressure difference between the inner-and outer-plant environment (Orr, 1992). Different plant species exhibit a diverse range of CH₄ transport processes, with convective transport processes typically more effective than molecular diffusion processes (Xu H. et al., 2021). In a recent study by Feng et al. (2022), a novel plant-girdling method was developed, removing the epidermis and subepidermal sclerenchyma to disrupt the O2 transport pathway, suppressing O_2 release and increasing CH_4 emissions, verifying that wetland plants aerenchyma play a vital role in the transport of CH_4 .

Oxidation of CH₄ in CWs

During CH₄ production and transport processes, CH₄ oxidation is a significant factor affecting CH₄ fluxes. CH₄ oxidation can occur *via* aerobic or anaerobic pathways. Aerobic oxidation of CH₄ (Table 2) mainly occurs at micro-interfaces where CH4 and O2 coexist, such as the substrate-air and water-air interfaces, the plant rhizosphere, and within plant tissues (Figure 3; Bonetti et al., 2021). Aerobic CH₄ oxidation is a chemical process with rapid reaction rates, depending on the concentration of O2. Under anaerobic conditions, microbes use electron acceptors other than O₂ to oxidize CH₄, including sulfate (sulfate-reduction-dependent anaerobic methane oxidation, SAMO), NO_2^- / NO_3^- (nitrite-dependent anaerobic methane oxidation), metal oxides (e.g., Fe³⁺ and Mn⁴⁺ anaerobic methane oxidation, metal-AOM), and direct interspecies electron transfer (Wegener et al., 2015; Guerrero-Cruz et al., 2021). Among these, SAMO is driven by anaerobic methanotrophs and sulfate-reducing bacteria, while nitrite-dependent AMO is performed by Candidatus Methylomirabilis oxyfera and Candidatus Methanoperedens nitroreducens (Cui et al., 2015). Additionally, metal-AMO is thermodynamically easier than SAMO (occurring at a 2-to 10-folds faster rate than SAMO) because metal-AMO produces more energy, accelerating the AMO process (Zhang et al., 2022). Mn-AMO has been estimated to reduce total-CH₄ emissions by 66%



Aerobic methanotrophs types	Class	Order	Family	Genus	Formaldehyde assimilation pathway	Remark
Type I	γ-Proteobacteria	Methylococcales	Methylococcaceae	Methylomonas	RuMP pathway	Psychrophiles
				Methylobacter		Psychrophiles
				Methylosarcina		Thermophiles
				Methylomicrobium		/
				Methyllohalobius		Haloalkaliphiles
				Methylosphaera		/
				Methylosoma		Halophiles
				Methylothermus		Thermophiles
				Methylovulum		Psychrophiles
				Crenothrix		/
				Clonothrix		/
Type X	γ-Proteobacteria	Methylococcales	Methylococcaceae	Methylococcus	RuMP pathway; low levels of enzymes of the serine pathway	Thermophiles
				Methylocaldum		Thermophiles
				Methylogaea		1
Type II	a-Proteobacteria	Rhizobiales	Methylocystaceae	Methylocystis	Serine pathway	Acidophiles
				Methylosinus		Acidophiles
			Beijerinckiaceae	Methylocella		Acidophiles
				Methylocapsa		Acidophiles
				Methyloferula		Acidophiles
Others	Verrucomicrobia	Methylacidiphilales	Methylacidiphilaceae	Methylacidiphilum	A variant of the serine pathway	1

Malyan et al. (2016); Zhu et al. (2016).

Acidophiles means growth at pH of 3.8–5.5; psychrophiles means growth at 5°C-10°C but not above 20°C; thermophiles means growth>45°C; halophiles means growth at 15% NaCl; haloalkaliphiles means growth at 12% NaCl and at pH of 9–11; /means no data.

(Liu et al., 2020) and Fe-AMO has the potential to decrease CH_4 emissions by 2-fold if it was to oxidize 10% of CH_4 worldwide (Egger et al., 2015). Among the different types of CWs, the use of VSSFCWs may be preferable for the reduction of CH_4 emissions as they have a suitable aerobic/anaerobic interface due to intermittent flooding, which is beneficial for both aerobic oxidation and AOM processes (Zhang et al., 2022).

Methyl coenzyme M reductase (mcrA) and particulate methane monooxygenase (pmoA), are key enzymes in CH₄ production and oxidation, resulting in their use as phylogenetic biomarkers for methanogens and methanotrophs (Chen et al., 2020b). Methanotrophs are the only known CH₄ biosinks, oxidizing another component of methanogenesis as carbon sources and producing CO₂, consuming at least 10% of atmospheric CH₄ in the process (O'Connor et al., 2010). CH_4 emissions have been found to positively correlate with the abundance of mcrA (Zhang et al., 2021d), and negatively correlate with the abundance of pmoA (Xu H. et al., 2021). However, no significant relationship was observed between pomA and CH₄ emissions, suggesting that other factors also mediate their function and activity (Zhang et al., 2021d). The mcrA/pmoA ratio can be used to explore the quantitative relationship between CH₄ production, oxidation and emissions, with a higher pmoA/ mcrA ratio indicating higher CH4 oxidation potential and lower CH₄ emissions (Zhao et al., 2022).

Environmental factors influencing CH₄ reduction in CWs

Temperature

Temperature influences the production and oxidation of CH₄. The optimum temperature for methanogenesis is typically 35°C-40°C (Ralf, 2007), with low temperatures impairing the activity of methanogens and fermentative bacteria (Barbera et al., 2015) by reducing the rate of organic matter degradation and hence, substrate availability for CH₄ production (Zhu et al., 2007; Maucieri et al., 2019). CH₄ production has been found to positively correlate with temperature to some extent under sufficient substrate availability conditions (Bhattacharyya et al., 2013; Zhao et al., 2022). For example, methanogenesis rate at 12°C is significantly lower than that at 30°C (Maucieri et al., 2019). Temperature not only influence microbial activity, but also affect the succession of dominant methanogenic archaea. According to Lu Y. et al. (2015), methanogens were dominated by Methanosarcinaceae (utilizing acetate and H₂/CO₂ substrates), while as temperature lower, Methanosaetaceae dominate the methanogens (using acetate for CH₄ production). Methanotrophs is temperature non-sensitive species and their optimum temperature is 25°C. CH_4 can be oxidized either at low to-2°C or at high to 30°C (Zhang et al., 2021a).

As a result of microbial activity influenced by temperature, CWs in warm season can release higher CH_4 (3.4%–42%) than the cool

or cold season (López et al., 2019; Maucieri et al., 2019; Shao et al., 2020). Johansson et al. (2004) found that CH₄ fluxes in SFCWs were strongly driven by season, fluctuating from-375 mg/m²/d to 1739 mg/m²/d for the spring to autumn period. Wang et al. (2019) concluded that average CH₄ emission in summer is 1.7 times higher than in winter, which was also proved by Zhao et al. (2014), D'Acunha and Johnson (2019), and Liu et al. (2019a). Therefore, in order to reduce the release of CH₄, it is recommended to control the temperature at an appropriate time of higher temperatures in summer.

Water table position

Water table position determines the degree of anaerobiosis inside CWs. High water tables can exhibit slower rates of atmospheric O_2 diffusion, creating a larger anoxic zone which is beneficial to CH₄ production (McInerney and Helton, 2016). Henneberg et al. (2015) compared CH₄ emissions in CW mesocosms with 0 cm, -10 cm, and -20 cm water tables, showing that CH₄ emissions were much higher in the 0 cm water table treatment system than the -20 cm system both with and without vegetation. This phenomenon may partly be due to higher levels of CH₄ dissolution occurring at increased water depths (Zhou et al., 2020). When the water table is below the substrate surface, the CH₄ produced is oxidized as it travels through the water layer due to diffusion and ebullition, resulting in a reduction in CH₄ emissions (Bonetti et al., 2021). When the water table is high, CH_4 emissions are increased as most CH4 enters plant root systems in the deeper anaerobic layer, before being transported to the atmosphere through the aerenchyma (Henneberg et al., 2015). A significant positive correlation has been reported between CH4 emission rates and water table positions (Liu et al., 2009), with studies also documenting that net CH4 fluxes are reduced in systems with a lower water table (Bridgham et al., 2013).

Redox potential

Redox potential (Eh) is an indicator of the internal O₂ level in CWs, which determines the activity of microbes and various enzymes, and is closely linked to CH4 production and oxidation processes (Liu et al., 2009). Different microbial groups require varying Eh conditions, with aerobic microbes generally requiring an Eh between +300 and + 400 mV. Parthenogenic anaerobic microbes typically have a cut-off Eh of +100 mV, with aerobic respiration occurring at Eh levels above this point and anaerobic respiration occurring at lower Eh levels. Specialized anaerobic bacteria typically require an Eh of-200 to-250 mV. The conversion of organic matter to CH4 occurs via the general anaerobic digestion pathway, while methanogens at the end of the respiratory chain require a strong reducing environment and very low Eh conditions (optimally-350 mV), with methanogenic processes initiated at Eh conditions < -200 mV (Chen et al., 2020b). The CH₄ production potential of a system increases with decreasing Eh (Liu et al., 2019b; Shao et al., 2020), and principal component analysis studies have also demonstrated that CH4 fluxes are positively correlated with Eh conditions (Liu et al., 2019a).

The mechanisms and methods of CH₄ reduction in design of CWs

CH₄ generation, transport, and oxidation are the three main processes that contribute to net CH₄ emissions from CWs (Bridgham et al., 2013), which are influenced by CWs design. Schemes implemented for the reduction of CH₄ emissions have mainly depended on the reduction of CH₄ generation and the promotion of CH₄ oxidation (Mander et al., 2014). which are affect by CW types, plant species, substrate types, the effect of CW-MFC systems, O₂ supply, available carbon source, and salinity. Therefore, a comprehensive analysis is required to provide a basis for the effective regulation and operation of CWs, while also actively mitigating GWP contributions (Figure 4; Liu et al., 2017; Maucieri et al., 2017).

Selection of suitable CW type

CWs are classified as SFCWs, HSSFCWs and VSSFCWs due to their varying structures and characteristics, resulting in significant differences in CH₄ emission profiles (Zhang et al., 2021a). SFCW systems consist of wastewater flowing over a substrate layer (Maucieri et al., 2017), while VSSFCW systems are gradually infiltrated by wastewater being applied to the surface layer by intermittent feeding, with the wetland bed maintaining an aerobic state with strong reoxygenation capabilities (Liu et al., 2019a). In contrast, wastewater is applied to HSSFCWs through the substrate layer by horizontal percolation in a mixed environment with aerobic, anoxic, and anaerobic degradation pathways, where the bed is submerged for a prolonged period of time causing anoxia (Zhou et al., 2020).

 CH_4 emissions from SSFCWs have been found to be significantly lower than from SFCWs (Table 3). In a survey by VanderZaag et al. (2010), SFCWs were found to emit 2-to 3-fold more CH_4 as a percentage of carbon removal than SSFCWs. The conditions associated with the highest CH_4 fluxes from SFCWs and the lowest fluxes from VSSFCWs were studied by Liu et al. (2009). The reason for the observed variation



CW types	Substrate types	Vegetation types	CH₄ fluxes (mg/m²/h)	Wastewater types	Reference
SF	Sand	Phragmites australis	0.09	Domestic Wastewater	Zhu et al. (2007)
SSF		Phragmites australis	0.16		
SF		Unvegetated	0.37		
SSF		Unvegetated	0.14		
SF	Pea-stone and gravel	Typha latifolia	9.3	Dairy farm wastewater	VanderZaag et al. (2010)
SSF			4.9		
SF	Sand	Phragmites australis	61.67	Municipal Wastewater	Gui et al., 2007
HSSF			8.18		
VSSF			3.23		
SF	Sand	nd Phragmites australis	26	Domestic Wastewater	Liu et al. (2009)
HSSF			5.4		
VSSF			1.7		
SF	Cobble	Cyperus alternifolius	0.64	River Wastewater	Zhang et al. (2021a)
HSSF	Gravel	Cyperus alternifolius	0.15		
VSSF	Gravel	Cyperus alternifolius	0.42		
HSSF	Gravel	Cyperus papyrus	22.70±1.9	Municipal Wastewater	Bateganya et al. (2015)
HSSF	Gravel	Unvegetated	38.30±3.3	-	
VSSF	Sand and gravel aggregates	Cyperus papyrus	3.30 ± 0.4		
VSSF	Sand and gravel aggregates	Unvegetated	13.60 ± 1.4		
HSSF	Gravel	Phragmites australis	31.8	Piggery farm wastewater	Zhou et al. (2020)
VSSF	Gravel	Phragmites australis	6.6		

TABLE 3 CH₄ emissions in different types of CWs.

was that SFCWs exhibit very low reoxygenation rates, that are insufficient for the complete oxidation of organic matter and readily create anaerobic conditions that promote CH₄ release (Søvik et al., 2006), with this effect usually observed in wetland systems that do not have routine harvesting of above-ground plant biomass, providing a continual supply of organic carbon (Hernandez et al., 2018). However, SSFCWs increase the contact time in the aerobic headspace and rhizosphere, which hinders the movement of gases (Zhu et al., 2007), resulting in more CH₄ oxidation occurring in SSFCWs than in SFCWs and lower CH₄ fluxes. Liu et al. (2009) also discovered that Eh conditions < -100 mV were primarily found in SFCWs, while no Eh was identified in VSSFCWs, supporting the association between Eh conditions and the high CH₄ fluxes observed in SFCWs.

CH₄ fluxes from HSSFCWs were found to be higher than those from VSSFCWs (Table 3). For example, Bateganya et al. (2015) found that HSSFCWs emit more CH₄ than VSSFCWs, irrespective of plant growth. Zhou et al. (2020) designed a hybrid CW system that generated about 4.8-fold lower CH₄ fluxes in the VSSF bed section than the HSSF bed section. The reason for this difference may be attributed to VSSFCWs having efficient O₂ transport, while HSSFCWs contain anoxic-anaerobic conditions (Mander et al., 2008), exhibiting negative Eh levels (-100 mV to-500 mV) and low dissolved oxygen (DO) concentrations (< 2 mg/L; López et al., 2015). In contrast, the VSSF system was found to be higher than the HSSF system in a study by Zhang et al. (2021a), due to the potential for CH₄ emissions to be affected by competition between various methanogens. A life cycle assessment also concluded that the environmental impact of CH₄ emissions from VSSFCWs was half or less than those from HSSFCWs (Fuchs et al., 2011).

CWs are increasingly being designed with composite structures that are more valid and robust for the treatment of a wide range of wastewater types (Liu et al., 2019a), However, these composite CW systems often have a negative effect on CH_4 fluxes (Zhou et al., 2020). For example, a monitoring study found that CH₄ emissions from VSSF-HSSF-SF CW was higher than from both VSSFCW and SFCW (Liu et al., 2009). Liu et al. (2019a,b) and Liu et al. (2018) designed integrated VSSFCWs that consist of alternating multifunctional layers of aerobic-anoxic-anaerobic-anoxic-aerobic conditions, leading to the accumulation of methanogens and increased CH₄ emissions (Zhang et al., 2021a). Waste resource conservation and reducing environmental influence have become priorities in sustainable engineering design. Therefore, the small occupation area of VSSFCWs, along with their high treatment efficiency and relatively low level of CH4 fluxes, make VSSFCW systems more extensively used, and a more CH₄ flux can be reduced via an enhanced O₂ transfer approach (Liu et al., 2019b).

Plant species selection

The relationships between CH_4 emissions and plant occurrence and diversity in CWs remain unknown (Han et al., 2019), as plants are able to both produce CH_4 independently, and mediate or influence CH_4 emission pathways. For example, organic matter and root exudates (such as sugars and amino acids) synthesized by plants *via* photosynthesis, can provide electron donors (Chen et al., 2020c; Liu et al., 2022), while root exudates also release degradable carbon which increases the number of methanogens and methanotrophs (Zhang et al., 2018b). Furthermore, plant root-secreted O₂ can provide electron acceptors (Kasak et al., 2020), with the intensity of O₂ secretion varying depending on the plant species, biomass, temperature, O₂ concentration, and photointensity conditions (Feng et al., 2022). In typical VSSFCWs, O₂ released by plant roots can provide approximately 0.43–1.12% of the biochemical oxygen demand (Zhang et al., 2014). The rhizosphere is a crucial zone for the production and oxidation of CH₄, with plant species composition, diversity, and planting density affecting the release of CH₄ (Chen et al., 2020c).

It has been observed that vegetation-covered CWs produce less CH₄ than those without vegetative cover (Zhu et al., 2007; Bateganya et al., 2015). However, contradictory results have been reported by Silvey et al. (2019), with this variation potentially due to plant species variation. Chen et al. (2020c) reported that planting Cyperus alternifolius resulted in CWs having lower CH4 fluxes than unvegetated CWs, while planting Phragmites australis and Canna indica had the opposite effect, highlighting the varying effect of different species on CH₄ fluxes. Firstly, some plants have been found to suppress CH₄ fluxes (Table 4). A monitoring study by Bateganya et al. (2015) found that planting Cyperus papyrus was more effective for the suppression of CH₄ fluxes regardless of the CW types, due to the extensive belowground rhizome network facilitating O2 transfer (Xu et al., 2019; Zhang et al., 2021a). Cyperus alternifolius is a ciliate with large root surface areas and root lengths of approximately 20 cm, providing them with the capacity to reach the bottom of CWs and release high amounts of root O_{22} leading to a more extensive aerobic environment (Chen et al., 2020c). The presence of Oenanthe javanicade has been shown to reduce the carbon concentration in wastewater, minimizing the production of CH₄ (Zhao et al., 2016; Han et al., 2017). In contrast, some plant species enhance CH₄ emissions (Table 4; Du et al., 2018; Maucieri et al., 2019). Juncus effusus has a significant capacity to transport CH₄ (Henneberg et al., 2015), and Rumex japonicus possesses a high root biomass capable of secreting low molecular weight substances (Zhao et al., 2016), accelerating microbial activity and increasing CH₄ emissions (Du et al., 2018). Overall, the contribution of plants to CH₄ emissions remains unclear. For example, Phragmites australis has an extensive rhizome system that typically penetrates substrate depths of 0.6-1.0 m, and has been reported to effectively reduce CH₄ fluxes (Xu et al., 2019), while other studies have reported that Phragmites australis possesses highly developed aerenchyma that allow more efficient gas transfer and increase CH₄ emissions (Sheng et al., 2015; Chen et al., 2020c). Moreover, some studies have proposed that certain plant species has no overall impact on CW CH₄ emissions (Han et al., 2019; Maucieri et al., 2019). Instead, plant characteristics such as root porosity and belowground biomass can regulate microbial community competition, O₂ transfer efficiency to the root system, and carbon source inputs, leading to varying levels of CH4 productivity in different species (Maucieri et al., 2017; Zhang et al., 2018b). The average CH₄ fluxes of submerged plants are generally lower than those of emergent plants (Niu et al., 2015; Zhang et al., 2018a). O₂ and carbon inputs from emergent plants can significantly affect the methanogenic community structure and methanogenic pathways, while submerged plants are primarily subject to regulation by DO and nitrogen levels (Zhang et al., 2018a). Furthermore, in contrast to the comparatively less-rigid forb species, some structurally rigid graminoids exhibit larger aerenchyma, which increases their ability to transport O₂ between the roots. Mesocosms containing Asclepias incaranta were found to have average CH4 fluxes that were 8-fold higher than those of mesocosms containing Alisma triviale (Silvey et al., 2019). Therefore, the selection of suitable plant species composition is essential to minimize CH_4 emissions from CWs.

Plant species diversity has been widely studied in recent years, and its contribution to CH4 emissions has increasingly being investigated. Several studies have observed a positive correlation between CH₄ emissions and plant species diversity (Du et al., 2018), as high species diversity increases carbon source available and promotes CH₄ emissions (Zhang et al., 2012). Some studies have also found that plant species diversity have no significant influence on CH₄ emissions (Zhao et al., 2016; Han et al., 2019), as in high ammonium environments, denitrification has a higher capacity to produce thermodynamic processes than methanogenesis (Chen et al., 2020c). In addition, denitrifying bacteria have a competitive organic substrate advantage, limiting CH₄ formation (Zhang et al., 2018a), in which high ammonium loading and plant biomass can perform CH4 offsetting functions, leaving CH₄ emissions unchanged overall (Han et al., 2019). Moreover, plant density has been reported to have no influence on CH₄ emissions (Hernandez et al., 2018). In conclusion, species characteristics remain a critical driver although more research is required in this field.

The contribution of plant harvesting to CH4 emissions is also significant (Feng et al., 2022). Non-harvested wetland plants can promote methanotrophic CH₄ consumption by transporting O₂ to the roots and substrate through aerenchyma (Zhu et al., 2007). However, the dying plant biomass provides an abundant source of bioenergy and promotes methanogenesis, with the potential to generate 10-40% of annual atmospheric CH₄ emissions worldwide (Juutinen et al., 2003; Keppler et al., 2006), highlighting the need for plant harvesting to be carefully managed. The time and manner of harvesting of aboveground macrophytes also affects CH₄ emissions from CWs (Kasak et al., 2020). Ensuring that harvesting occurs at the end of the growing season (i.e., before nutrient transfer to belowground plant structures) can significantly reduce CH4 emissions. However, biomass harvesting during peak periods of plant growth and soil microbial activity has been shown to significantly enhance CH4 emissions (Barbera et al., 2015), due to the rapid release of CH₄ accumulated in the vascular system of plant stalks (Kasak et al., 2020). Therefore, effective planning to optimize harvesting time and method can effectively reduce CH4 emissions from managed wetlands, and thus enhance their multiple ecological benefits.

Substrate amendment

The substrate forms the backbone of CWs, providing support for the growth of plants and microbes (Ji et al., 2021). Recently, novel substrate amendment schemes have been applied in CWs, with the addition of substances such as biochar, iron oxides, manganese oxides, zeolite, walnut shell, activated alumina, and ferric-carbon, which are gradually displacing traditional substrates (such as sand, gravel, and ceramsite) and improving the treatment efficiency of CWs. Some reviews have focused on the impact of substrate on CH_4 emission reduction. However, none of these have emphasized the role of enhanced substrates compared to conventional substrates (Yu et al., 2022; Zhao et al., 2022). Therefore, in order to achieve sustainable and low-environmental impact CW operations, we summarized multifarious functional substrates used in CWs for enhancing the CH_4 emissions reduction.

TABLE 4 CH_4 emissions in CWs planted different vegetation.

Vegetation types	CWs types	Substrate types	CH₄ fluxes (µg/m²/h)	Wastewater types	References
Emergent vegetation					
Phragmites australis	SFCW	1	2,640	Agricultural wastewater	de Klein and van der Werf (2014)
Phragmites australis			1820		
Unvegetated			7,430	-	
Unvegetated			19,810		
Cyperus papyrus	HSSFCW	Gravel Sand and gravel	$22,700 \pm 1900$	Municipal wastewater	Bateganya et al. (2015)
Unvegetated			38,300±3,300		
Cyperus papyrus			$3,300 \pm 400$		
Unvegetated			$13,600 \pm 1,400$		
Juncus effusus	SFCW	1	$20,380 \pm 1930$	Sewage treatment water	Ström et al. (2006)
Phragmites australis			13,880±3,190		
Typha latifolia			9,380±1990		
Unvegetated			$200 \pm 2,580$		
Arundo domax	HSSFCW	Gravel	25,170	Municipal wastewater	Maucieri et al. (2014)
Phragmites australis			21,160		
Unvegetated			18,100		
Phragmites australis	HSSFCW	Gravel	$20,220 \pm 6,700$	Municipal wastewater	López et al. (2015)
Schoenoplectus Californicus			18,120±1,130		
Rumex japonicus	VSSFCW	Sand	310	Synthetic wastewater	Niu et al. (2015)
Oenanthe hookeri	_		200	-	
Phalaris arundinacea			290		
Iuncus effusus			190		
Unvegetated	_		140		
Rumex japonicus	1	Fine sand	285 ± 102.5	Synthetic wastewater	Zhao et al. (2016)
Oenanthe javanica			21.67±58.33		
Phalaris arundinacea			140 ± 117.08		
Juncus effuses	_		154.58 ± 114.58		
Rumex japonicus	/	Coarse sand	245.83 ± 8.75		
Oenanthe javanica			256.25±8.33		
Phalaris arundinacea			250±7.5		
Iuncus effuses			240 ± 6.25		
Rumex japonicus	VSSFCW	Sand	285.8	Synthetic wastewater	Han et al. (2017)
Oenanthe javanica			232.08		
Phalaris arundinacea			242.92		
Juncus effuses			210.83		
Typha orientalis	VSSFCW	Sand and gravel	10,100	River wastewater	Zhang et al. (2018b)
Cyperus alternifolius	-		15,100	-	
Arundo domax			12,500		
Iris pseudacorus			19,400		
Thalia dealbata			7,100		
Phragmites australis	HSSFCW	SSFCW Gravel	30,000	Municipal wastewater	Maucieri et al. (2019)
Arundo donax	-		34,000		
Chrysopogon zizanioides			45,000		
Miscanthus×giganteus			59,000		
Unvegetated			25,000	1	

(Continued)

TABLE 4 (Continued)

Vegetation types	CWs types	Substrate types	CH₄ fluxes (µg/m²/h)	Wastewater types	References
Lolium perenne	VSSFCW	Sand	0.011	Synthetic wastewater	Han et al. (2019)
Cichorium intybus			0.025		
Medicago sativa			0.033		
Rumex japonicus			0.014		
Canna indica	SSFCW	Gravel	13.66 ± 24	Synthetic wastewater	Chen et al. (2020c)
Cyperus alternifolius			-34.60 ± 8.12		
Phragmites australis			21.88 ± 2.51		
Unvegetated	_		-5.32 ± 7.14		
Submerged vegetation		•			·
Potamogeton crispus	/	/	5,700	Municipal wastewater	Zhang et al. (2018a)
Myriophyllum spicatum			1,600		
Hydrilla verticillata			4,000		

/ means no data.

Carbon-rich substrate types

Biochar is an organic carbon-enriched product that is considered a promising alternative substrate (Zhuang et al., 2022). Ji et al. (2021) showed that biochar-based CWs contained a higher pmoA/mcrA ratio than none-biochar CWs, with the addition of biochar having an inhibitory effect on CH₄ fluxes, possibly due to biochar promoting the secretion of O₂ from plant roots, increasing CH₄ oxidation (Ji et al., 2020). However, Chen et al. (2020a) and Guo et al. (2020) proposed contrasting conclusions, finding that CH4 fluxes were consistently higher in biochar-added CWs than CWs without added biochar. This may be due to biochar enhancing direct interspecies electron transfer between methanogens and Geobacteraceae, while also providing organic matter to methanogens, resulting in the stimulation of CH₄ emissions (Liu et al., 2012). Overall, the influences of biochar on CH₄ emissions remains unclear (Table 5), with the reported differences primarily caused by variations in the raw biochar materials, operating conditions, and the properties of the microbial community within the system (Chen et al., 2020a), highlighting the need for further research to determine the role of biochar in regulating CH₄ emissions (Zhuang et al., 2022). Walnut shell is also loaded with high concentrations of organic matter. Xu G. et al. (2021) showed that CH4 emissions from walnut shell substrate were 14.8-fold higher than from the control substrate, due to the release of large amounts of degradable organic carbon from walnut shell, resulting in high CH₄ fluxes.

Electron-exchange substrate types

Highly crystalline iron oxides and manganese oxides are electron-exchange substrates that are abundant and readily available, making them highly suitable for use as CW substrate materials (Zhang et al., 2021b; Yu et al., 2022). Cheng et al. (2021a) reported that CWs using iron oxide substrates emitted less CH₄, finding that iron oxide increased CH₄ emissions by promoting electron transfer between *Geobacter* and methanogens, while also directly inhibiting the activity of methanogens and some enzymes involved in CO₂ reduction, and promoting the AOM process under the influence of dissimilated metal-reducing bacteria, ultimately resulting in a reduction in CH_4 emissions overall (Cheng et al., 2021b). Mn ore substrates have been found to reduce CH_4 emissions from CWs (Liu et al., 2020). In addition, Cheng et al. (2021b) observed that both Mn ore and iron ore substrates inhibited CH_4 emissions, with Mn ore reported to be more effective due to the fact that Mn ore promotes AOM processes mainly by competing for organic substrates and providing electron acceptors, almost completely inhibiting CH_4 production (Xu G. et al., 2021). However, the use of iron oxide as a substrate has more complex implications, such as different forms and valences of iron affecting CH_4 production (Cheng et al., 2021a).

Adsorption substrate types

Fe-C is widely used as a substrate in wastewater treatment, utilizing chemistry-coupled biological processes for the removal of pollutants (Dong et al., 2020). Zeolite, a common silicate mineral, is considered to be a high-performance gas adsorption material, due to its relatively well-developed pore network and skeletal configuration (Wang Y. et al., 2020), with well characterized CH₄ adsorption, storage, and oxidation capabilities (Wang H. et al., 2020; Zhou et al., 2020). Zhao et al. (2022) monitored CH₄ emissions from laboratory-scale CWs containing different substrates, showing that systems utilizing Fe-C and zeolite as substrates all exhibited lower average CH₄ fluxes than the control groups (Table 5), as Fe-C can compete with methanogens for substrate in the presence of iron-reducing bacteria, inhibiting the production of $\rm CH_4.$ In addition, $\rm Fe^{3+}$ has a high Eh as an electron acceptor (Bond and Lovley, 2002), and the larger surface area of activated carbon facilitates biofilm generation, promoting CH₄ oxidation. Zeolite incorporation into the substrate has been found to significantly reduce CH₄ emissions from CWs. Zhou et al. (2020) discovered that CWs containing zeolite substrate exhibited reduced CH4 fluxes by about 2-fold compared to those of gravel substrate CWs, due to the porous structure of zeolite improving local atmospheric DO concentrations and reducing methanogen activity. The results of these studies are further supported by the observation that Fe-C and zeolite

TABLE 5 CH₄ emissions in CWs with different substrates.

Substrate types	CW types	Vegetation	CH₄ fluxes (mg/m²/h)	Main operation conditions	References
Biochar and ceramsite ^a	SSFCW	Lythrum salicaria	0.17 ± 0.11	Non-aeration	Ji et al. (2020)
Ceramsite			0.25±0.19	Non-aeration	
Biochar and ceramsite			-0.058 ± 0.077	Aeration	
Biochar and ceramsite			0.12 ± 0.14	Tidal flow	
Biochar and gravel	/	Typha latifolia	0.2192-0.477.0	Non-aeration	Guo et al. (2020)
Gravel			0.1274-0.2708	Non-aeration	
Biochar and coarse gravel	SSFCW	Canna indica	0.023	Non-aeration	Chen et al. (2020a)
Coarse gravel			0.003	Non-aeration	
Biochar and coarse gravel	SFCW		0.03	Non-aeration	
Coarse gravel			-0.029	Non-aeration	
Ceramsite	SSFCW	Lythrum salicaria	0.24 ± 0.01	Non-aeration	Ji et al. (2021)
Biochar and ceramsite			0.08 ± 0.08	Non-aeration	
Biochar and ceramsite			0.03±0.03	Aeration	
Gravel	VSSFCW	Cyperus alternifolius	229.17±10	Non-aeration	Liu et al. (2020)
Mn ore			125.42 ± 15.83		
Gravel and sand	VSSFCW	SFCW Iris pseudacorus	17.08	Non-aeration	Xu G. et al. (2021)
Mn ore, gravel and sand			2.00		
Walnut shell, gravel and sand	-		252.30		
Activated alumina, gravel and sand			6.43		
Quartz sand	VSSFCW /	0.059-0.061	Non-aeration	Cheng et al. (2021a)	
Iron ore and quartz sand			0.048-0.051		
Gravel and quartz sand	VSSFCW	Yellow calamus	0.06	Non-aeration	Cheng et al. (2021b)
Iron ore, gravel, and quartz sand			0.05	_	
Mn ore, gravel, and quartz sand			0		
Gravel	VSSFCW	Phragmite australis	31.8	Non-aeration	Zhou et al. (2020)
Zeolite			16.6		
Gravel	VSSFCW	SFCW Acorus calamus	0.41±0.07	Aeration	Zhao et al. (2022)
Zeolite and gravel	-		0.20±0.03		
Fe-C and gravel			0.31 ± 0.04		
Fe-C, zeolite, and gravel			0.21±0.03	-	

^ameans the mix of two or three types of substrates.

/means no data.

substrates contained a reduced abundance of the functional gene mcrA and a significantly increased abundance of pmoA (Zhao et al., 2022). Activated alumina is also a powerful adsorption substrate, as shown in a study by Xu G. et al. (2021) in which activated alumina significantly reduced CH₄ emissions from CWs due to its strong adsorption capacity, leading to reduction in organic matter content and inhibiting methanogenic microbial activity (Alvarez and Cervantes, 2012). In addition, sand, ceramsite and gravel have also been utilized as adsorption fillers (Table 5; Wang Y. et al., 2020), but they have been gradually replaced due to poor CH₄ adsorption capacities (Ji et al., 2020; Cheng et al., 2021a; Zhao et al., 2022). With a survey of CWs finding that mcrA and pmoA were not detectable in all gravel substrates (Chen et al., 2020c), which was attributed to the fact that methanogens and methanotrophs do not easily attach to gravel so it is gradually replaced.

CW-coupled MFC systems

MFCs are a low-environmental impact energy utilization technology (Wang et al., 2019; Zhang et al., 2021d). CWs (especially VSSFCWs) have unique water quality conditions with substantial redox gradients across their vertical profile, with DO and Eh levels increasing from the subsurface to the surface zones (Maucieri et al., 2017). As CWs and MFC systems function under similar conditions, it is feasible to control CH₄ emissions using CW-coupled MFC systems (Zhang et al., 2021b,c). Methanogens and electrogenic bacteria require similar living conditions, such as an anaerobic environment and low Eh, resulting in competition for the substrate at CW-MFC anodes, which may inhibit the power generation performance of the CW-MFC (Liu et al., 2017; Zhang K. et al., 2020). A comprehensive understanding of the competitive mechanisms between methanogens and electrogens would help maximize the potential advantages of CW-MFC systems (Zhang et al., 2021e), along with determining the influences of circuit condition, plant type, external resistance, substrate type, and hydraulic retention time (HRT).

Open/closed circuit in CW-MFC

Open or closed circuit systems are one of the essential factors affecting CH₄ fluxes from CW-MFCs. Liu et al. (2022) found that the CH₄ emissions from non-planted CW-MFCs, were about 0.21 ± 0.01 mg/ m²/h higher in open-circuit systems than in closed-circuit systems, while in the planted open-circuit system the CH4 emissions were $0.46 \pm 0.02 \text{ mg/m}^2/\text{h}$ higher than in the closed-circuit planted system. A study by Xu et al. (2021b) yielded CH₄ fluxes of 6.37-7.28 mg/m²/h and 7.43-8.36 mg/m²/h for closed and open circuit CW-MFC systems, respectively. Zhang et al. (2021e) noted that running a MFC in CWs can suppress a third of all CH₄ emissions. These studies show that CW-MFCs can effectively reduce CH4 emissions from CWs, with similar findings also reported by Zhang K. et al. (2020) and Zhang et al. (2021c), with the main contributory factor being the bioanode. In anaerobic environments, methanogens are dominant in CWs due to the absence of current transmission, resulting in an increase in the production of CH₄ (Zhang et al., 2021d). In contrast, because organic matter is more readily available for electrochemically active bacteria (EAB) in closedcircuit CW-MFC systems, electrical stimulation of EAB growth results in a current that inhibits methanogens (Liu et al., 2022). In addition, electrons from the anode may compete with methanogens as the anode layer of the CW-MFC has a higher Eh, allowing electron-producing bacteria to capture electrons more easily than methanogens, leading to a reduction in CH_4 emissions (Zhang et al., 2021e).

Plant rhizosphere location

 CH_4 emissions are significantly affected by plants in CW-MFC systems. Recently, an increasing number of studies have demonstrated that plant root location has an influential effect on CH_4 emissions in CW-MFC systems (Liu et al., 2017). When the plant rhizosphere is in the cathode layer, electron acceptors provided by root-secreted O_2 favor cathode reactions and power generation (Zhang et al., 2021e). Since CW-MFC anodes are typically in an anoxic state rather than an anaerobic state, the small amount of O_2 secreted by plant roots has little effect on the O_2 levels in the anode environment (Zhang et al., 2021d). Therefore, plant rhizospheres in the anodic zone can provide photosynthetic organic matter as an alternative energy source, increasing CH_4 emissions. Zhang et al. (2021e) reported that CW-MFC systems with rhizospheres in the cathode zone emit less total-CH₄ (29.21 mg/m²/d) than those with rhizospheres at the anode (33.01 mg/m²/d). Zhang et al. (2021d) observed that growing *Typha* orientalis and *Cyperus alternifolius* in the cathode zone of a reactor, resulted in lower CH₄ emissions than when grown at the anode. Zhang et al. (2021c) reported a similar conclusion, with methanogens becoming more active as the organic matter content of the rhizosphere increases, resulting in an increase in CH₄ emissions. Additionally, the O₂ provided by plant roots has been found to have less of an impact on lowering CH₄ emissions than the organic matter in the root system (Lu L. et al., 2015).

The presence of plants and the selected plant species also influence CH_4 emissions in CW-MFC systems. Liu et al. (2022) reported that CH_4 emissions increased by $0.48 \pm 0.02 \text{ mg/m}^2/\text{h}$ in closed-circuit CW systems with plants, compared to the non-planted group. A similar conclusion was reached by Xu et al. (2021b), with a 21.79% reduction in average CH_4 emissions from non-planted reactors, as compared to those with plants. Zhang et al. (2021d) reported lower CH_4 emissions in systems with anode grown *Typha orientalis* (3.9 mg/m²/h), than with anode grown *Cyperus alternifolius* (4.5 mg/m²/h). A study comparing the effects of three plant species, *Typha orientalis*, *Thalia dealbata*, and *Cyperus alternifolius*, found that CH_4 emissions from the CW-MFC were highest in the *Typha orientalis* system, and lowest in the *Cyperus alternifolius* system (Zhang K. et al., 2020). These results emphasize the importance of further research to determine the effect of different plant species on CH_4 emissions from CW-MFC systems.

The important role that plants play in CH_4 emissions from CW-MFC systems is becoming increasingly apparent, with both the plant species and root location influencing CH_4 emissions, although root location appears to have a more pronounced effect (Zhang et al., 2021d). Therefore, focusing on plant selection to ensure that plants with inhibiting methanogenesis or poor CH_4 transport are utilized, with their roots located in the cathodic zone, has been shown to be more advantageous for the sustainable development of wastewater treatment systems, in terms of reactor power generation and CH_4 reduction.

External resistance

The long-term application of external resistance in CW-MFC systems affects the microbial community structure and biochemical metabolism in the anode biofilm (Zhang et al., 2021c). Liu et al. (2022) observed that an external resistance of 1,000 Ω leads to an increase in CH_4 emissions by $0.67 \pm 0.01 \text{ mg/m}^2/\text{h}$ compared to a 100Ω resistance. Wang et al. (2019) found that CH₄ emissions tended to increase with the addition of external resistances $>500 \Omega$. This effect occurs due to variation in microbial metabolic activities, electron transfer rates and substrate utilization kinetics, under varying external resistance conditions (Liu et al., 2022). Higher external resistance loads slow down the flow of electrons to and from EAB, which encourages methanogens to consume more substrate and increases CH₄ emissions (Liu et al., 2017). However, it has also been found that insufficiently low external resistances lead to rapid electron flow rates, which are not conducive to the sustainable utilization of CW-MFC systems, and eventually leads to a decline in power generation (Nikhil et al., 2018). Therefore, a suitable relatively low external resistance should be applied, ensuring that CW-MFC systems generate maximum levels of power, while releasing minimum CH₄ (Liu et al., 2022).

Additional influencing factors

Substrate. CH₄ emissions in CW-MFC systems are also driven by substrate type. A Mn CW-MFC system was shown to generate lower CH₄ emissions (53.44–66.64 mg/m²/h) than a clinopyrite CW-MFC system (62.69–88.02 mg/m²/h; Zhang et al., 2021b). Furthermore, Xu et al. (2021b) observed a 25.42% reduction in average CH₄ emissions from Mn-based reactors compared to graphite granule-based reactors. This may be due to the occurrence of Mn-driven Mn-AOM lowering CH₄ emissions, while dissimilatory metal reduction processes encourage competition between EABs and methanogens, as well as increasing the growth of EABs on Mn ore anodes, further inhibiting the growth of methanogens (Li et al., 2019; Zhang et al., 2021b, 2022). Therefore, different substrate materials have varying electron acceptor functions and significantly affect CH₄ emissions, with Mn exhibiting high prospects as a substrate for CH₄ emissions reduction.

HRT. Due to the relatively high organic load in CW-MFCs, there is a positive correlation between CH_4 emissions and HRT. Lower HRTs result in a higher load and increased CH_4 emissions (Zhang et al., 2021e). It was observed that when the HRT in plant systems increased, CH_4 fluxes tended to reduce, with higher organic matter contents stimulating the activity of inactive microorganisms, consuming DO, and promoting the growth of methanogens (Wang et al., 2019). HRT is an extremely vital operating parameter for conventional wetlands (Zhang K. et al., 2020). A longer HRT can prolong the contact time between microorganisms and wastewater (Zhang et al., 2021d), ensuring the effective removal of organic matter as well as the suppression of CH_4 emissions. Finally, it has been found that CW-MFC systems have a stronger CH_4 reduction effect when operated in continuous flow mode than in batch mode (Wang et al., 2019).

Other methods

Oxygen supply strategy

Shortage of DO in CWs due to prolonged saturation and rapid microbial metabolism is a major limitation to the removal of organic matter from conventional CWs (Wang et al., 2022). Intermittent aeration and tidal flow are considered to be the most effective oxygenation strategies to directly influence CH4 emissions, allowing the manipulation of DO conditions in CWs (Table 5; Ji et al., 2020), providing additional O₂ for the inhibition of CH4 biochemical processes and the acceleration of CH4 oxidation, reducing CH₄ fluxes (Ji et al., 2021). D'Acunha and Johnson (2019) found that CH₄ fluxes could be reduced by 60.7% using intermittent microaeration under optimal aeration conditions. Furthermore, Zhao et al. (2022) observed increased CH4 fluxes in aerated sections of CWs compared to non-aerated sections, which was attributed to CH4 production primarily occurring in non-aerated sections, while CH4 emissions were enhanced by agitation and blowing in aerated sections, with the change in emissions consistent with the aeration rate and dissolved CH₄ concentration. Ji et al. (2020) showed that only a slight decrease in CH₄ fluxes occurred under tidal flow oxygenation conditions, although Ji et al. (2021) concluded that the total CH4 fluxes were slightly higher due to the loss of large amounts of CH4 from the empty substrate after drainage. However, the use of intermittent aeration increases operational costs and energy input requirements, making it more suitable for use in CWs in relatively concentrated communities or limited land areas. In contrast, the tidal flow process involves no significant additional costs or maintenance expenses.

The available carbon sources in CWs can be supplied internally or externally. Internal carbon sources mainly include plant roots or microbial secretions, plant deadfall, organic matter decomposition, and substrates (Liu et al., 2019a), while external carbon sources include biodegradable carbon, natural plant material, and soluble carbon from natural organic matter (Liu et al., 2019b). To balance the nutritional ratio in CWs, additional carbon sources must be provided in some CWs, directly affecting CH₄ emissions (Chen et al., 2020b). C/N ratios represent the relative amount of carbon and nitrogen available in the wastewater, with different C/N ratios promoting or inhibiting microbial activity (Guo et al., 2020). Yan et al. (2012) found that CH₄ fluxes in CWs with C/N ratios of 2.5, 5, and 10 were $1.36 \pm 0.09 \text{ mg/m}^2/\text{h}$, $2.02 \pm 0.07 \text{ mg/m}^2/\text{h}$, and 2.34 ± 0.15 mg/m²/h, respectively, showing that CH₄ fluxes were lowest with low C/N ratio conditions. Corbella and Puigagut (2015) also reported a positive correlation between CH₄ emissions and influent C/N conditions, which may be attributed to increases in C/N causing rapid O₂ consumption, resulting in a lower Eh and higher CH₄ production (Guo et al., 2020). However, Chen et al. (2020b) analyzed CWs with influent C/N ratios of 0, 5, 10, 15, and 20, showing that CH₄ emissions did not differ significantly, reaching a minimum value of $-42.49 \text{ mg/m}^2/\text{h}$ at a C/N ratio of 10. Zhao et al. (2014) combined SSFCWs with an earthworm eco-filter and found that CH4 emissions increased in accordance with the influent C/N ratio regardless of the sequence of treatment, as the earthworm activity greatly increased the permeability of the SSFCW. Therefore, lower C/N ratio conditions are more favorable for CH₄ emissions reduction. Liu et al. (2019a) observed that the addition of urea as an external carbon source at concentrations of 0, 12.1, 30, 45, 61, and 80 mmol/L, resulted in a trend of increasing and then decreasing CH_4 emissions, achieving the lowest emission level with a urea concentration of 80 mmol/L. Similar results were obtained by Liu et al. (2019b) using ethanol as an external carbon source, with results showing that Eh values after the addition of ethanol (except for the highest ethanol concentration of 32 mmol/L) were higher than the control groups, resulting in more CH4 emissions. Excessive carbon source concentration increases the water purification load. Therefore, when external carbon sources are provided, the dosage should be adjusted to ensure optimal water purification performance with CH₄ emission control.

Salinity control

With increasing levels of water scarcity, desalination, or the direct utilization of seawater in coastal areas is becoming increasingly common, generating large quantities of saline wastewater. CWs are increasingly being used for the treatment of saline wastewater in response to different regional treatment strategies (Shao et al., 2020). The salinity of the influent affects not only the growth of wetland plants but also the structure, abundance and activity of microbial communities (Sheng et al., 2015; Xiao et al., 2016). According to Shao et al. (2020), CH₄ emissions were higher at salinities of 0 and 0.5%, continually decreasing with increasing salinity, with a substantial decrease occurring at 1.0%. Sheng et al. (2015) also observed that CH₄ emissions decreased significantly with increasing salinity due to inhibition of methanogen growth and activity (Xiao et al., 2016). In addition, salinity has been repeatedly shown to inhibit CH₄ emissions in tidal wetlands, due to competition between sulfate and methanogens for substrates in tidal waters (Poffenbarger et al., 2011; Marton et al., 2012). In summary, the effect of salinity on CH₄ fluxes in CWs is pronounced, with emissions promoted by low salinity and inhibited by high salinity conditions.

Conclusions and perspectives

Global climate change is a complex phenomenon. CWs are a highly productive green technology for surface source pollution control, which are designed to treat wastewater using the natural processes of plants, substrates and microorganisms, utilizing carbon cycling processes that are an essential part of the global system, generating both environmental and economic benefits. Currently, CWs have the potential to be restored using known and innovative land management practices, providing significant opportunities for carbon sequestration and CH4 offsetting. CH4 emissions from SFCWs have been found to be significantly higher than from SSFCWs, while CH₄ emissions from VSSFCWs were significantly lower than from HSSFCWs. Several species-specific communities have been shown to facilitate the operation of energy-efficient and low-emission CWs. In addition, the selection of an appropriate substrate is also critical for CH4 mitigation. In CW-MFCs, the combination of biological and bio-electrochemical methods can effectively control CH₄ emissions from CWs, although it is essential to maintain a stable balance between the systems CH₄ production rate and power generation capacity. In addition, the O₂ supply strategy, carbon source concentrate, and salinity control are key operational aspects that should be optimized to reduce the CH₄ production potential of the system.

As research has progressed in this field, tremendous advances have been made in the control of CH_4 emissions from CWs. However, there are still some aspects that require further investigation:

- When construction budgets and operating conditions allow, the use of integrated CWs is more effective for wastewater treatment, and optimization of the design of integrated systems for CH₄ reduction will increase the development potential of CWs.
- The effect of different plant species on CH₄ emissions in CW systems requires further research, with the rational application of plant litter as a substrate, such as biochar, may be beneficial to the overall sustainability and low-environmental impact of the CWs.
- Different anode materials can function as distinct types of electron acceptors, influencing the oxidation of CH₄ in CW-MFC systems. Therefore, the mechanism of effect anode materials on CH₄ production and emissions requires further investigation.
- Despite CWs being an essential ecosystem, most of the previously reported results were obtained from controlled mesoscale

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experiments, resulting in the need for future research to include field assessments conducted over long time spans.

Author contributions

GY was responsible for data curation, formal analysis, and investigation. JC wrote the manuscript draft. GW, HC, and JH contributed to conceptualization, formal analysis, and visualization. YL, WW, FS, YM, QW, MW, TL, ZS, JS, and ZY provided feedback on the manuscript. All authors contributed to the article and approved the submitted version.

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Conflict of interest

WW and FS were employed by Hunan Pilot Yanghu Reclaimed Water Co., Ltd. YM was employed by Hunan Rongantai Ecological Technology Co., Ltd. QW and MW were employed by CCCC-TDC Environmental Engineering Co., Ltd. TL and ZS were employed by China Railway Wuju Group the First Engineering Co., Ltd.

The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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