

Research progress and prospect of low-carbon biological technology for nitrate removal in wastewater treatment

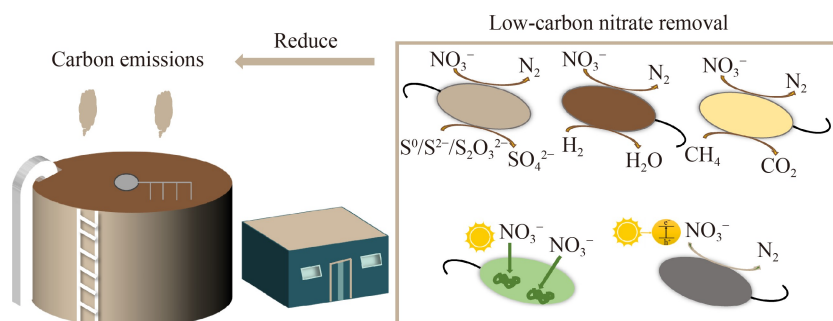
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HIGHLIGHTS

- n-DAMO achieves simultaneous nitrogen removal and methane emission reduction.
- Photosynthetic microorganisms achieve negative carbon emission by absorbing CO₂.
- PEDeN is an emerging low-carbon denitrification technology using photoelectrons.
- Solar-driven low-carbon nitrogen removal system is the future trend.

GRAPHIC ABSTRACT



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ABSTRACT

Wastewater treatment plants are the major energy consumers and significant sources of greenhouse gas emissions, among which biological nitrogen removal of wastewater is an important contributor to carbon emissions. However, traditional heterotrophic denitrification still has the problems of excessive residual sludge and the requirement of external carbon sources. Consequently, the development of innovative low-carbon nitrate removal technologies is necessary. This review outlines the key roles of sulfur autotrophic denitrification and hydrogen autotrophic denitrification in low-carbon wastewater treatment. The discovered nitrate/nitrite dependent anaerobic methane oxidation enables sustainable methane emission reduction and nitrogen removal by utilizing available methane *in situ*. Photosynthetic microorganisms exhibited a promising potential to achieve carbon-negative nitrate removal. Specifically, the algal-bacterial symbiosis system and photogranules offer effective and prospective low-carbon options for nitrogen removal. Then, the emerging nitrate removal technology of photoelectrochemical denitrification and the underlying photoelectron transfer mechanisms are discussed. Finally, we summarize and prospect these technologies, highlighting that solar-driven biological nitrogen removal technology is a promising area for future sustainable wastewater treatment. This review has important guiding significance for the design of low-carbon wastewater treatment systems.

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1 Introduction

The issue of global climate change has become one of the most prominent environmental concerns in the world.

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Several polar ice caps are approaching critical thresholds, necessitating urgent action to reduce carbon emissions (Lenton et al., 2019). Under the dual carbon goal, the Chinese government is also committed to striving for a carbon peak before 2030 and carbon neutrality before 2060 (Hao et al., 2022). According to a survey, wastewater treatment plants are one of the most primary energy consumers and sources of greenhouse gas

emissions (Lu et al., 2018). In 2019, Chinese wastewater treatment plants emitted 53.0 million metric tons of carbon dioxide equivalent (MtCO₂e) (Du et al., 2023). Wastewater treatment plants generate carbon emissions directly or indirectly during processes such as biological carbon production, wastewater treatment (nitrogen and phosphorus removal), sludge digestion, electricity generation, and system maintenance (Kyung et al., 2015; Lu et al., 2018).

As is well known, nitrogen pollution can lead to water eutrophication, and the nitrogen removal from wastewater is an important component of wastewater treatment. Biological nitrogen removal will produce greenhouse gases such as CO₂, nitrous oxide (N₂O) and methane (CH₄), exacerbating carbon emissions (Ross et al., 2020). CO₂ is produced during microbial degradation and organic matter metabolism (Gupta and Singh, 2012). When carbon sources are insufficient, N₂O is a byproduct of incomplete denitrification (Chen et al., 2019a). According to the IPCC report, in municipal sewage nitrogen removal, the N₂O produced during the aerobic process is 0.016 mg N₂O–N/mg TN removed and the energy consumption required to remove 1 kg NH₄⁺–N is 4.4 kWh (Shukla et al., 2019), which also implies that sewage nitrogen removal is an important source of energy consumption and carbon emissions. CH₄ is usually produced during anaerobic digestion (Wang et al., 2014). It is worth noting that the global warming potential of N₂O and CH₄ is much higher than that of CO₂ (Chen et al., 2019a; Roth et al., 2023).

Nitrate contamination has become one of the most important water quality issues globally (Hu et al., 2020). Improper treatment of nitrate in wastewater can easily lead to pollution of groundwater and drinking water, further posing risks to public environment and human health (Bijay-Singh and Craswell, 2021). Exposure to nitrate can result in both acute and chronic health issues, such as infant methemoglobinemia and the potential development of cancers in adults (Fewtrell, 2004). Biological denitrification converts nitrate into nitrogen gas (N₂) and is a crucial step in wastewater nitrogen removal, which involves the conversion of nitrate to nitrite by nitrate reductase (Nar), followed by the conversion of nitrite to nitric oxide (NO) by nitrite reductase (Nir). Subsequently, NO is further converted to N₂O by NO reductase (Nor), and finally, N₂O is transformed into N₂ by N₂O reductase (Nos) (Chen and Strous, 2013). In traditional nitrification-denitrification processes, external carbon sources such as glucose or acetate are added as electron donors to drive heterotrophic denitrification and the type of carbon source plays a decisive role in the final products (N₂, NH₄⁺ or N₂O) of denitrifiers (Carlson et al., 2020). N₂O is the product of incomplete heterotrophic denitrification whose production will be effected by carbon source, C/N ratio, initial nitrate concentration, temperature and pH value, among

them, organic carbon is a key factor (Wang et al., 2014; Lee et al., 2019). For example, when methanol is used as the only carbon source, N₂O will not be produced while the methanol is biotoxic and unsafe (Lee et al., 2019). Currently, commonly used carbon sources in practical applications include sodium acetate and glucose while they have respective drawbacks of being costly and having a slow denitrification rate and both inevitably cause N₂O emission (Lyu et al., 2017; Fu et al., 2022). Large amounts of external carbon source will easily lead to excessive sludge production, undoubtedly increasing the cost (Cui et al., 2019), which are serious issues in engineering application.

Therefore, it is necessary to replace traditional carbon sources with other clean or cheap electron donors to reduce carbon emissions and facilitate practical applications. At present, there have been reviews describing the role of anammox-related processes in low-carbon nitrogen removal from wastewater (Guo et al., 2022; Ma et al., 2023). A successfully operating anammox-related processes can achieve nitrogen removal rates of 5 kg N/(m³·d), which is more than 10 times the performance of traditional nitrification-denitrification processes (Lackner et al., 2014). Furthermore, it saves 100% of external organic carbon addition and reduces energy consumption by 60% (Lackner et al., 2014). Sulfur autotrophic denitrification combined with anammox can achieve low-carbon nitrogen removal while deeply treating nitrate (Li et al., 2022). Hydrogen autotrophic denitrification has less secondary pollution, the products are relatively clean and harmless, and N₂O emissions can be greatly controlled, which makes it an advantageous technology for deep nitrogen removal (Wang et al., 2022b). CH₄ is the cheapest and simplest organic carbon and it can be produced on the site of the wastewater treatment plants. It is worth noting that during nitrate/nitrite dependent anaerobic methane oxidation (n-DAMO), CH₄ can be taken as electron donors *in situ*, which can simultaneously achieve CH₄ emission reduction and nitrogen removal (Fan et al., 2023). In addition, light energy serves as the energy source for all life activities on the earth, photoautotrophic microorganisms and even some non-photoautotrophic microorganisms can convert light energy into chemical energy (Gomelsky and Hoff, 2011). This undoubtedly provides a new direction for promoting green and low-carbon development because it can reduce the use of chemicals. For example, microalgae can undergo photoautotrophic growth using water as an electron donor and assimilating nitrogen even in the absence of organic carbon (Winkler and Straka, 2019).

Based on this, this review highlights the roles and challenges of sulfur autotrophic denitrification, hydrogen autotrophic denitrification and n-DAMO in low-carbon wastewater treatment. This review places a particular emphasis on the role of light energy in wastewater nitrate

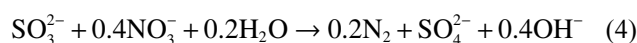
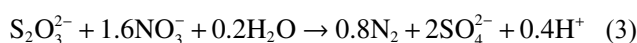
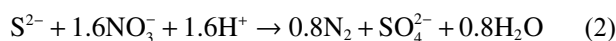
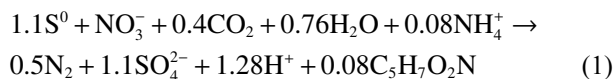
removal, including photosynthetic microorganisms, as well as the latest discoveries of photoelectrotrophic denitrification that use photoelectrons as donors, discussing their types and the involved mechanisms. The review provides a summary and outlook for these low-carbon nitrate removal technologies, further offering insights and guidance for the synergistic effect of pollution reduction and carbon emissions reduction in wastewater treatment.

2 Sulfur autotrophic denitrification

2.1 Sulfur autotrophic denitrification with various sulfur sources

Sulfur autotrophic denitrification (SAD) is an efficient and resource-utilizing biological wastewater treatment process, mediated by sulfur-oxidizing bacteria (SOB) which were first characterized and isolated in 1954 (Baalsrud and Baalsrud, 1954). In this process, sulfur is used to replace organic carbon as an electron donor for the reduction of nitrate to N_2 (Zhang et al., 2022b). Compared to other autotrophic denitrification processes, the reduced inorganic sulfur compounds are readily available and can be utilized at a significantly higher specific rate. Besides, compared with conventional denitrification processes, the sludge production rate decreases by about 55%, and the operational costs, and the emissions of CO_2 and N_2O could decrease by more than 80% (Pokorna and Zabranska, 2015; Hu et al., 2020; Rahimi et al., 2020; Pang and Wang, 2021). The cost of the reduced inorganic sulfur compounds ranges from 0.13 to 0.25 \$/kg, which is 8 times lower in organic carbon of heterotrophic denitrification (HD) (Wang et al., 2023a). Given these benefits, the SAD process has attracted a giant interest in wastewater treatment.

SAD has been demonstrated to be an efficient method for removing nitrogen (Wang et al., 2021c; Zhang et al., 2022a), nevertheless, there is a dearth of information about the SOB that mediated SAD and their metabolic properties. This knowledge gap represents a significant obstacle to the improvement and evolution of SAD. In the SAD process, four common sulfur compounds, elemental sulfur (S^0), sulfide (S^{2-}), thiosulfate ($S_2O_3^{2-}$), and sulfite (SO_3^{2-}), can be used as electron donors. Equations (1)–(4) described the SAD with various sulfur sources (Batchelor and Lawrence, 1978; Pokorna and Zabranska, 2015; Xue et al., 2022; Yuan et al., 2022).



S^0 is the most favorable electron donor for SAD due to its non-toxicity, low cost, and easy availability. Since the denitrification process consumes carbonate, the limestone (as an inorganic carbon source) is often mixed into S^0 in a certain proportion to achieve a better denitrification performance (Liang et al., 2022a; Liang et al., 2022b). $S_2O_3^{2-}$ is another higher preferred electron donor due to its high bioavailability (Wang et al., 2020; Deng et al., 2022; Wang et al., 2023a). Since $S_2O_3^{2-}$ provides electrons to SOB in the form of a solution, it can achieve a similar denitrification rate as the HD process. In SAD process, the nitrate is first reduced to nitrite and step reduced to N_2 like the HD process. During the process of denitrification, sulfur compounds (S^0 , S^{2-} , SO_3^{2-} , $S_2O_3^{2-}$) provide electrons through the electron transfer chain to reduce nitrogen oxides (Deng et al., 2019; Ma et al., 2020; Grubba et al., 2022). These compounds then participate in more complex reactions which ultimately convert into SO_4^{2-} (Wu et al., 2020; Zhang et al., 2022a). Compared to HD processes, the SAD process significantly reduces direct carbon emissions from wastewater treatment plants since CO_2 is not produced as a result of SAD (Wang et al., 2023a).

2.2 Wastewater treatment systems integrating the sulfur autotrophic denitrification

The SAD process provides several advantages, such as being economical, low carbon, and producing fewer greenhouse gas emissions, which make it an excellent choice for wastewater treatment plants looking to achieve energy-saving goals. In this system, organic carbons are not necessary, and the CO_2 is fixed to synthesize organic carbon for the growth of SOB (Pokorna and Zabranska, 2015; Yuan et al., 2022). Thus, SAD has the potential to become a major global contributor to negative carbon emissions. Additionally, the SAD process can create a better condition which enlarges the N_2O removal performance. The N_2O emissions produced through SAD process decreased by about 96% than the HD process (Yang et al., 2016). Most wastewater influent has a COD/N ratio lower than 6, which is difficult to provide enough electron donors to complete the denitrification process (Wang et al., 2023a). Adding a SAD unit after the conventional biological nitrogen removal process can achieve a more efficient nitrogen removal without adding additional carbon sources.

Extensive laboratory-scale studies have explored coupling the PN/A with the SAD process to enhance nitrogen removal performance (Deng et al., 2019; Zhang et al., 2020; Deng et al., 2021; Deng et al., 2022). These studies discovered that the anammox process can reduce the sulfate production rate during the SAD process by consuming the nitrite intermediate product (Fig. 1), which makes the SAD process more practical and efficient. By combining S^0 -driven SAD with the anaerobic ammonium oxidation process, the consumption of sulfur for every 1 g

of nitrogen treated can be reduced from 2.5 g to 0.43–0.55 g. The combination of FeS-driven SAD and anammox processes allows for the simultaneous removal of ammonium and nitrate (Wang et al., 2023c). Moreover, the SAD process can also be coupled with the sulfate-dependent anaerobic ammonium oxidation (sulfamnox) process, which convert the ammonia and sulfate to nitrate and sulfur. The intermediates produced in the process of SAD and sulfamnox can be utilized by each other to achieve stable denitrification (Liu et al., 2021b; Deng et al., 2022). Table 1 summarizes the nitrogen removal performance of different SAD-based coupling processes.

2.3 Challenges

The SAD process has great engineering application potential, however, there are still some issues that need to be resolved. Sulfate production is a problem in the SAD process, particularly in a $S_2O_3^{2-}$ or S^0 -based SAD process, which produces 4.07 g or 2.51 g SO_4^{2-} -S/g NO_3^- -N respectively (Zhang et al., 2022c). Higher concentration of SO_4^{2-} (> 400 mg SO_4^{2-} -S/L) can stress aquatic ecosystems (Wang et al., 2020). Furthermore, high concentration of sulfate can be reduced to H_2S by bacteria under anaerobic conditions, leading to serious odor, equipment corrosion (Brettar and Rheinheimer,

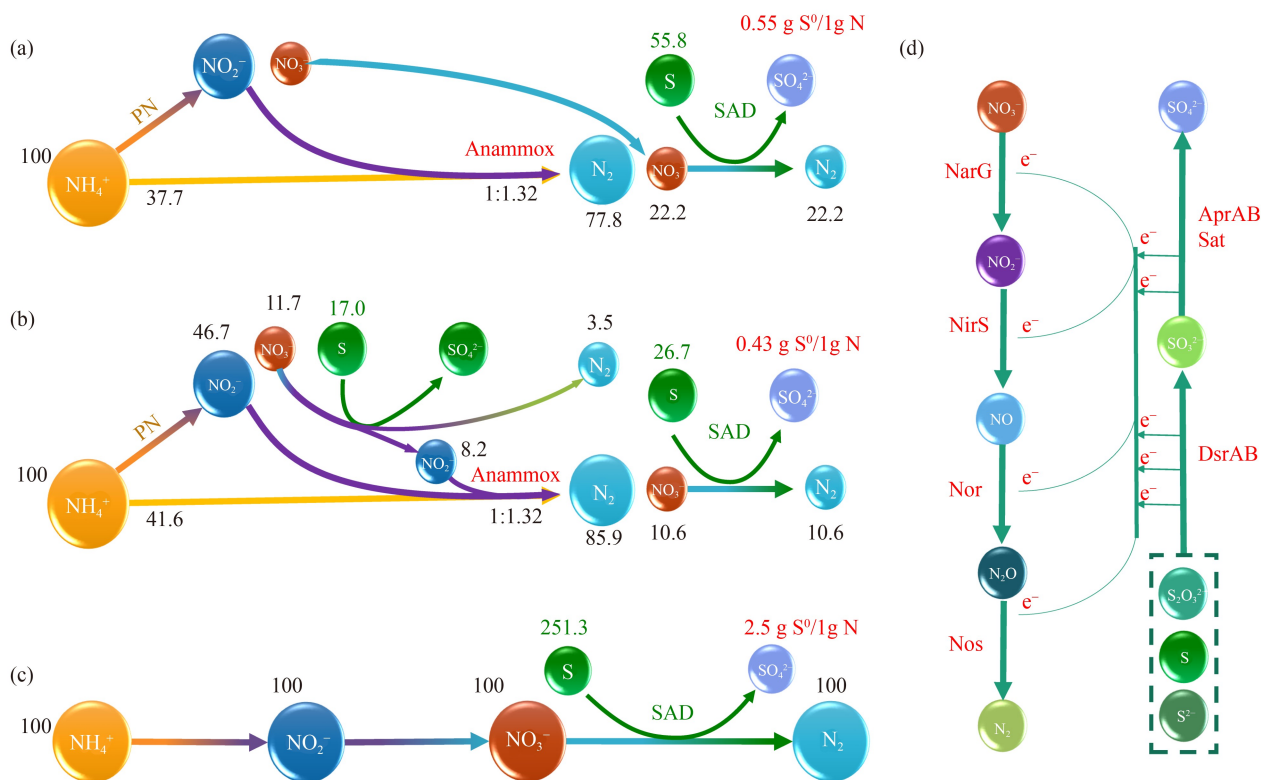


Fig. 1 (a) The SAD process is coupled after partial-nitrification/anammox; (b) the partial-nitrification/anammox are coupled with the SAD process in a single reactor; (c) partial-nitrification and SAD process; (d) inter-conversions between different forms of N and S mediated by SOB.

Table 1 The nitrogen removal performance of different SAD-based coupling processes

Process	Reduced sulfur compounds	Reactor	Seeding sludge	Performance	Ref.
Two-stage SAD-ANAMMOX	$S_2O_3^{2-}$	UASB	Activated sludge Anammox sludge	NRE: 82.5%	Deng et al. (2019)
Simultaneous SAD-ANAMMOX	–	EGSB	SAD sludge	NRE: > 95%	Zhou et al. (2021)
Simultaneous SAD-ANAMMOX	S^0	Upflow biofilm reactor	PN/A sludge	NRE: 98%	Zhang et al. (2020)
PN-SAD	S^0	Upflow-vibration biofilm reactor	Sludge obtained from a long-stable PN reactor	NRE: 97.1% NAR: 95%	Deng et al. (2020)
Sulfamnox-anammox-SAD	S^0	Upflow anaerobic bioreactor	Activated sludge	ARE: 70%	Zhan et al. (2023)

Note: EGSB: Expanded granular sludge bed; UASB: Upflow anaerobic sludge blanket; NRE: Total nitrogen removal efficiency; ARE: Ammonium removal efficiency; NAR: Nitrite accumulation rate.

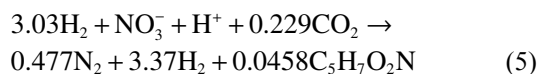
1991; Pan et al., 2013). Although S^{2-} is a better electron donor for the SAD process, the biogenic accumulation of sulfur could become another issue (Wang et al., 2023a).

There are currently two methods for reducing the production of SO_4^{2-} in SAD processes. The first is to couple SAD with sulfamox, which provides substrate for SOB to decrease the sulfate production rate while sulfamox is limited and remains at the proof-of-concept stage. The second method is partial SAD, which involves coupling with anammox to avoid excessive SO_4^{2-} production. This process reduces the production of SO_4^{2-} by decreasing the demand for reduced inorganic sulfur compounds. With appropriate S/N control, partial SAD can achieve NO_2^- production while the optimized operating conditions for the different systems that use reduced inorganic sulfur compounds have not yet been fully determined. Therefore, future studies should focus on the interaction between anammox and SOB to help the operation of partial SAD process.

3 Hydrogen autotrophic denitrification

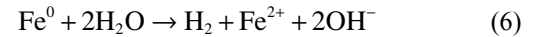
3.1 Production and source of hydrogen in hydrogen autotrophic denitrification

Denitrification is a natural process that occurs all the time in nature. In environments where there are no natural electron donors available, denitrifying microorganisms have the option to use hydrogen (H_2) as an energy source. To survive in such environments, these microorganisms use H_2 as an electron donor and nitrate as the final electron acceptor, which also called the hydrogen autotrophic denitrification (HAD). Equation (5) showed the chemical equation for the HAD process (Ma et al., 2020). H_2 is an excellent electron donor for denitrification and its high diffusivity through biofilms promotes the nitrate removal performance in attached growth systems (Ruiz-Romero et al., 2009; Di Capua et al., 2019).



H_2 is not common in the natural environment and is typically produced from hydrocarbon fuels or can also be generated biologically through dark or photo-fermentation processes (Ruiz-Romero et al., 2009; Di Capua et al., 2019). H_2 can also be produced by direct electrolysis in the bioreactor or by anaerobic corrosion of thermodynamically unstable Fe^0 , shown as Eq. (6), which have shown successful applications in enhancing nitrate removal rates (Zhu et al., 2019; Ma et al., 2020). As the delivery efficiency of H_2 in water is low, some researchers proposed that the most effective method is the *in situ* generation and direct uptake of H_2 by H_2 -consuming denitrifiers (Tian and Yu, 2020). An alternative approach that has gained interest is the supply

of H_2 through membrane aeration with micro-bubble or bubble-less technology, which is similar to *in situ* generation (Zhu et al., 2019; Ma et al., 2020; Tian and Yu, 2020).



Bacteria can use H_2 as a source of electrons to reduce different substances such as O_2 , NO_3^- , Fe^{2+} , SO_4^{2-} , and CO_2 (Burgdorf et al., 2006; Ruiz-Romero et al., 2009). These bacteria have enzymes called hydrogenases, which catalyze the reversible oxidation of H_2 into protons ($H_2 \leftrightarrow 2H^+ + 2e^-$). The electrons are then transferred to intermediates like NAD + or bc1 complex which introduce the electrons into the denitrification respiratory chain (Pang and Wang, 2021). There are three families of hydrogenases classified according to their metalcore: [NiFe], [FeFe], and [Fe] (Pang and Wang, 2021; Zhang et al., 2022a). Hydrogenotrophic bacteria that grow in environments without organic substrates have to assimilate mineral carbon. This process requires energy in the form of reduced co-enzymes (NADH, H^+ , FADH, H^+) and ATP. The enzymes involved in carbon assimilation are the carboxylases. For example, in the Calvin cycle, 1 CO_2 and 3 NADH, H^+ are consumed to produce glyceraldehyde-3-phosphate (Dijkhuizen and Harder, 1984).

Compared to SAD, HAD does not produce any harmful by-products such as sulfate, and it has a higher denitrification rate. The denitrification rate of HAD is about 6 times higher than SAD which is driven by S^0 , and the HAD process is clean with only water as a by-product and does not pose any harm to human health (Ma et al., 2020; Grubba et al., 2022). Besides, by optimizing hydrogen transfer in bacterial cells, denitrification kinetics comparable to HD can be achieved (Vasiliadou et al., 2006; Lee et al., 2010; Epsztein et al., 2016). These advantages make HAD a promising process for low-carbon wastewater treatment. At present, most research on HAD has focused on areas where there are high standards for controlling organic matter in the effluent, such as nitrogen removal from drinking water or groundwater (Ma et al., 2020; Tian and Yu, 2020).

3.2 Hydrogen autotrophic denitrification combined with multi-contaminant removal

Microbial electrolysis cells (MECs) have garnered significant attention for their ability to remove organic waste at the anode while generating H_2 at the cathode (Carmona-Martínez et al., 2015). Thus, the HAD process is often used in conjunction with MEC, making it useful for removing complex multi-pollutants. In addition, due to the low solubility of hydrogen, H_2 -based membrane biofilm reactor (H_2 -MBfR) can effectively help dissolve hydrogen and increase the denitrification rate (Jiang et al., 2020). It is an emerging nitrate removal technology that

uses inorganic carbon as the electron donor.

Currently, the process of integrating H₂-MBfR with MEC has become a low-carbon nitrogen removal technology that has attracted much attention. MEC can perform long-distance electronic compensation through H₂ to achieve nitrate reduction in MBfR (Han et al., 2023b). The process of gas diffusion membrane integrated into MEC (MMEC) can effectively combine the degradation of organic waste, generation of H₂ and reduction of nitrate, and the total nitrogen removal rate can exceed 90% (Liang et al., 2021). In addition, by combining MEC with multiple air cathode microbial fuel cells (MEC-MFC), the air cathode MFC can be used as the power source of MEC, thereby reducing external power supply and reducing system energy consumption (Wang et al., 2021a). Recently, there are also studies combining MEC with permeable bio-reactive barrier (PRB) to achieve efficient removal of organic waste and groundwater nitrate nitrogen (Han et al., 2023a). Table 2 summarizes the performance of various HAD-based coupling processes.

There is no clear way to determine the most effective method for treating wastewater that contains nitrates. Combining HAD with other processes in a single reactor is a promising approach to achieving integrated removal of multiple contaminants.

3.3 Challenges

HAD is a clean and efficient low-carbon sewage nitrogen removal technology while the low bioavailability of H₂ often limits the process due to the low aqueous solubility of 7.4×10^{-4} mol/L at 30 °C. Therefore, gas-permeable membranes such as porous membranes or hollow fiber membranes are often used to prevent hydrogen limitation and improve the denitrification rate (Di Capua et al., 2019; Dawood et al., 2020; Zhang et al., 2022a). Although these designs can increase the denitrification load to some extent, the nitrogen removal rate of HAD is still lower than HD (Matějů et al., 1992; Devlin et al., 2000; Ergas and Reuss, 2001).

It is important to note that there is a potential risk of using H₂ as an electron donor in wastewater treatment plants, as it can form flammable and explosive mixtures with air (Wu et al., 2018; Wu et al., 2020). H₂ is one of

the most expensive electron donors used in the autotrophic denitrification process. Although the cost of producing H₂ has become more cost-competitive in recent years, it is still around 3 USD/kg H₂ (Di Capua et al., 2019; Dawood et al., 2020). Furthermore, membrane operation and cleaning can also increase the operating costs. Therefore, finding lower-cost H₂ production methods and solving the safety issues of H₂ transportation are crucial to advancing HAD toward engineering applications.

4 Nitrate/nitrite-dependent anaerobic oxidation of methane (n-DAMO)

4.1 Combined n-DAMO-based processes

Nitrate/nitrite-dependent anaerobic oxidation of methane (n-DAMO) is a new biological nitrogen removal technology that uses nitrate or nitrite as the electron acceptor and CH₄ as the electron donor (Fan et al., 2023), among which *Candidatus Methyloirabilis oxyfera* belonging to n-DAMO bacteria and *Candidatus Methanoperedens nitroreducens* belonging to n-DAMO archaea work together (Li et al., 2023a). n-DAMO bacteria can use CH₄ as a carbon source to reduce nitrite to nitrogen, while n-DAMO archaea can use CH₄ to reduce nitrate to nitrite. n-DAMO can achieve stable nitrogen removal rates in both mainstream wastewater and sidestream wastewater treatment (Cai et al., 2015; Xie et al., 2018). Useful nitrogen removal rate was also observed in real wastewater treatment by n-DAMO (Lim et al., 2021).

The current combination processes based on n-DAMO can be divided into nitrification-n-DAMO process, nitrification-n-DAMO process and Partial nitrification (PN)-n-DAMO-Anammox (Fan et al., 2023). The nitrification-n-DAMO process uses nitrate as an electron donor and traditionally requires n-DAMO archaea and n-DAMO bacteria to complete. Recent studies have found that complete nitrate reduction and methane oxidation can be performed by a single *Methyloirabilis* bacterium, which expands our new understanding of the carbon and nitrogen cycles and provides new ideas for sewage treatment and methane emission reduction (Yao et al., 2024). Compared to nitrification-n-DAMO process,

Table 2 The nitrogen removal performance of various HAD-based coupling processes

Process	Apply of H ₂	Seeding sludge	Performance	Ref.
MEC-MBfR	Anodic oxidation and gas diffusion membrane	Activated sludge	NRE: > 90%	Liang et al. (2021)
MEC-MBfR	Applied negative pressure to MEC, changed to positive pressure to supply gas to MBfR	Anaerobic sludge	R NO ₃ ⁻ : 97.8%	Han et al. (2023b)
MFC-MEC	MFC as a power source; Anodic oxidation	Activated sludge	R NO ₃ ⁻ : 80.6%	Wang et al. (2021a)
MEC-PRB	Applied negative pressure to MEC, changed to positive pressure to supply gas to PRB	Anaerobic sludge	R NO ₃ ⁻ : 85.0% ± 0.8%	Han et al. (2023a)

Note: R NO₃⁻: Nitrate removal efficiency.

nitritation-n-DAMO process uses nitrite as an electron donor, which can reduce aeration and methane requirements (Wang et al., 2019). Besides, compared with the PN/A process, nitritation-n-DAMO does not have excessive nitrate accumulation and N₂O emission. The PN-n-DAMO-anammox process has attracted more and more attention due to its advantages of low aeration energy consumption, reduced greenhouse gas emissions and complete nitrogen removal. Table 3 summarizes the operating conditions and nitrogen removal performance of various PN-n-DAMO-anammox processes. Membrane biofilm reactors (MBfR) were widely used in water treatment due to their high methane transfer efficiency (Xie et al., 2018). PN/anammox/n-DAMO can also be integrated into a single MBfR (Liu et al., 2019). Useful nitrogen removal rates were observed by PN-n-DAMO-anammox processes in both low-concentration mainstream wastewater treatment and high-concentration sidestream wastewater treatment. Besides, it was worth noting that it cannot only use CH₄ biogas as an electron donor, but also use dissolved CH₄ in the effluent as an electron donor, which is very helpful for reducing CH₄ emissions and deep nitrogen removal (Liu et al., 2020a). It also maintains good stability at low temperatures. Integrating n-DAMO and anammox consortia in granular sludge also enables a very short hydraulic retention time (2.16 h) and dissolved CH₄ removal at 10 °C (Fan et al., 2021).

4.2 Metabolic characteristics and interactions of functional microorganisms in n-DAMO

n-DAMO is a sustainable technology with carbon and energy benefits. However, the metabolic characteristics and interactions of n-DAMO microorganisms are not fully understood. Clarifying this can provide insights for the optimization and advancement of n-DAMO-based wastewater nitrogen removal techniques. *Candidatus Mmethylomirabilis oxyfera* and *Candidatus Methanope-re-dens nitroreducens* are often used as model

microorganisms for n-DAMO bacteria and n-DAMO archaea. The n-DAMO bacterium uses nitrite as an electron acceptor and is an obligate anaerobic methane-oxidizing bacterium, but it can utilize methane through the aerobic methane oxidation pathway of particulate methane monooxygenase (pMMO) (He et al., 2018). n-DAMO archaea distantly related to ANME-2 archaea can utilize CH₄ by reversed methanogenesis pathway and take methyl-coenzyme M reductase (MCR) as a biomarker (Ding and Zeng, 2021). n-DAMO archaea usually have higher methane affinity relative to n-DAMO bacteria (He et al., 2018). The recently discovered n-DAMO bacterium *Candidatus Mmethylomirabilis sinica* can highly express nitrate reductase Nap. It has a high affinity for nitrate and can couple nitrate and methane to generate N₂ (Yao et al., 2024).

n-DAMO combined with anammox technology plays an important role in deep nitrogen removal of wastewater and CH₄ emission reduction. Anammox bacteria can consume nitrite and ammonium, and the nitrate generated can be utilized by n-DAMO archaea. The nitrite produced by n-DAMO archaea can be further utilized by anammox bacteria and n-DAMO bacteria (Li et al., 2023a). Taking advantage of this interaction, first using influent water containing nitrate and ammonium to stimulate the growth of n-DAMO archaea, and then adding nitrite can simultaneously promote the growth of anammox and n-DAMO archaea, achieving efficient nitrogen removal (Nie et al., 2020). This interaction is also evident in the stratification of the biofilm. Anammox bacteria on the surface of the biofilm utilizes substrates in the wastewater, while n-DAMO bacteria located in the middle layer primarily utilize nitrite produced by the innermost layer of n-DAMO archaea (Liu et al., 2023). Besides, interesting interactions also exist between n-DAMO and heterotrophic microorganisms. n-DAMO microorganisms can produce formic acid and acetic acid, which can be used by heterotrophic microorganisms as carbon sources to promote denitrification (Guo et al., 2023; Li et al., 2023a).

Table 3 The operational conditions and performance of different PN-n-DAMO-anammox processes

Reactor	Operational conditions	Biomass	Nitrogen removal rate (kg N/(m ³ ·d))	Ref.
MBfR	$T: 30 \pm 1 \text{ }^\circ\text{C}$ $C_{\text{NH}_4^+-\text{N}}: 470 \text{ mg/L}$ $C_{\text{NO}_2-\text{N}}: 560 \text{ mg/L}$	biofilm	1	Liu et al. (2020b)
one-stage MBfR	$T: 30 \pm 1 \text{ }^\circ\text{C}$ $C_{\text{NH}_4^+-\text{N}}: 1030 \text{ mg/L}$	biofilm	1.5	Liu et al. (2019)
UASB	$T: 30 \pm 0.5 \text{ }^\circ\text{C}$ $C_{\text{NH}_4^+-\text{N}}: 500 \text{ mg/L}$ $C_{\text{NO}_2-\text{N}}: 550 \text{ mg/L}$	granule	1	Liu et al. (2021a)
MBfR	$T: /$ $C_{\text{NH}_4^+-\text{N}}: 22 \text{ mg/L}$ $C_{\text{NO}_2-\text{N}}: 29.5 \text{ mg/L}$	biofilm	0.28	Xie et al. (2018)
MGSR	$T: 10\text{--}20 \text{ }^\circ\text{C}$ $C_{\text{NH}_4^+-\text{N}}: 23 \text{ mg/L}$ $C_{\text{NO}_2-\text{N}}: 47 \text{ mg/L}$	granule	0.94 (20 °C) 0.55 (10 °C)	Fan et al. (2021)

Note: T : temperature; $C_{\text{NH}_4^+-\text{N}}$: the concentration of NH_4^+-N ; $C_{\text{NO}_2-\text{N}}$: the concentration of $\text{C}_{\text{NO}_2-\text{N}}$; MGSR: membrane granular sludge reactor.

4.3 Challenges

It is important to note that the engineering application of n-DAMO presents certain challenges compared to conventional heterotrophic denitrification. This is because the electron donor in the n-DAMO process is CH_4 , which has low solubility. As a result, the denitrification rate of DAMO is lower than that of other heterotrophic denitrification processes (Wang et al., 2023b). Additionally, precise control of aeration pressure and aeration volume is necessary to prevent the escape of unused methane gas into the atmosphere. Moreover, methane is easy to burn and has certain risks in engineering applications.

The doubling time of n-DAMO microorganisms is usually more than two weeks, even longer than that of anammox bacteria, which is also an important challenge that limits its engineering applications (Wang et al., 2019). It is reported that nucleobase and betaine help promote the activity of n-DAMO microorganisms (Wang et al., 2019). In the future, it is necessary to further understand the metabolic characteristics and interactions of n-DAMO microorganisms, which will help enrich n-DAMO microorganisms and provide a theoretical basis for the integration of n-DAMO and AD technologies.

5 Photosynthetic microorganisms for low-carbon nitrate removal

SAD, HAD and n-DAMO may achieve nitrate removal with a low carbon emission. However, these technologies require the input of external additives (e.g., sulfur, H_2 and CH_4), and additional waste in the effluent of SAD that must be treated again, which makes the processes more expensive and complex. Photosynthetic microorganisms potentially provide a more sustainable carbon-neutral alternative to nitrate removal and recovery without external additives. Nitrogen in wastewater can be assimilated effectively by most photosynthetic microorganisms, such as photosynthetic bacteria and microalgae, which are able to utilize inorganic carbon as the carbon source and light as the energy source for growth, thus minimizing carbon emissions during the nitrogen removal process.

5.1 Simultaneous nitrate recovery and CO_2 reduction by microalgae

Microalgae as one of the most widespread photosynthetic microorganisms have a high photosynthetic capacity compared to higher plants and can assimilate nitrogen for the synthesis of amino acids and proteins (Han and Zhou, 2022). They are capable of assimilating all kinds of inorganic nitrogen, including nitrate, nitrite, ammonium, and even urea (Kumar and Bera, 2020). The flexible

nitrogen source requirement allows them to adapt to wastewater with various nitrogen forms. In addition, some *Cyanobacteria* are able to fix nitrogen to sustain treatment performance when the nitrogen source is depleted (Trebuch et al., 2023b). Ammonium as the most abundant form of inorganic nitrogen in municipal wastewater can be directly assimilated by microalgae without requiring conversion steps. Nitrate is taken up in microalgae by a common high-affinity transport system and enters the cells by diffusion. Nitrate is reduced to nitrite by nitrate reductase inside the cell, and nitrite is further reduced to ammonium by nitrite reductase (Fig. 2). Ammonium is then incorporated into carbon skeletons mainly through the operation of the glutamine synthetase-glutamate synthase cycle, achieving nitrogen recovery from wastewater (Han and Zhou, 2022; Kong et al., 2022).

Microalgae have a powerful photoautotrophic metabolism that allows them to utilize CO_2 effectively. According to the stoichiometry of biomass growth, 49 kg of NH_4^+ is required for mitigating per ton of CO_2 , which leads to an algal biomass production of 489 kg (Boelee et al., 2014). Thus, simultaneous CO_2 capture of industrial flue gas and wastewater treatment via microalgae recently have attracted increasing attention (Zhang et al., 2023b). CO_2 capture efficiency of 102.13 t/a was achieved through the cultivation of microalgae *Chlorella vulgaris* in a raceway pond (Valdovinos-García et al., 2020). In addition, the sulfur oxides (SO_x) and nitrogen oxide (NO_x) in the flue gas can be also removed by microalgae. For instance, *Chlorella* sp. could achieve fuel gas purification with the NO_x and SO_x removal efficiency of 62%–65% (Kumar et al., 2018). In addition to excellent CO_2 capture ability, the harvested algal biomass can be used for various applications such as biofertilizer, biofuel, or as a valuable resource for other industries (Han and Zhou, 2022). Consequently, the use of microalgae as a nature-inspired approach minimizes carbon emissions and achieves resource recovery during N removal. Nevertheless, a long hydraulic retention time (HRT) of a few days is usually required to achieve effective nitrogen removal. Besides, the poor settleability of unicellular microalgae necessitates an additional step for biomass separation. Reactor design, light condition, harvesting technology, and downstream process are essential factors that need to be considered in the microalgae-based system.

5.2 Photosynthetic bacteria-based nitrate removal

Photosynthetic bacteria (PSB) are the earliest bacteria on Earth, having existed for over 3 billion years (Blankenship, 2010). They are currently found in diverse environments, thriving in both aerobic and anaerobic conditions, such as soil, lakes, and oceans (Stomp et al., 2007). PSB exhibit remarkable tolerance to harsh environ-

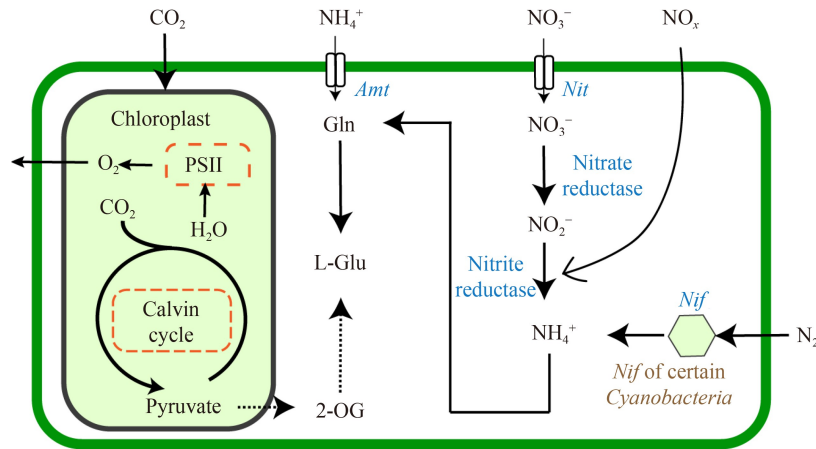


Fig. 2 Uptake and transport mechanism of inorganic nitrogen in algae and their fate.

mental conditions, including extreme temperatures, salinity, and acidic or alkaline conditions. Additionally, they possess the capability to utilize both organic and inorganic compounds as carbon sources (Blankenship, 2010). Different from microalgae, PSB perform anoxygenic photosynthesis that utilize light as solar energy without oxygen production. Moreover, PSB have bacteriochlorophyll that exhibits maximum absorption in the near-infrared (NIR) spectrum, around 804 nm (Stomp et al., 2007). Consequently, PSB do not demand high light intensities in comparison to microalgae, resulting in energy consumption reduction (Hülßen et al., 2018).

Nitrogen removal by PSB is mainly via assimilation, and some PSB are capable of denitrification. Similar to microalgae (Fig. 2), PSB can assimilate all forms of inorganic nitrogen, and reduce nitrate to ammonia for cell synthesis (Cao et al., 2020). In addition to nitrate assimilation, some heterotrophic PSB possess the genes and enzymes involved in denitrification. During the denitrification process, light energy is captured by photopigments and used to drive electron transport reactions, resulting in the production of reducing power (e.g., reduced ferredoxin or NADPH) that can be utilized in the nitrate reduction process (Liu et al., 2022). This feature allows for substantial energy savings compared to traditional heterotrophic denitrification. Purple non-sulfur bacteria (PNSB), represented by genera such as *Rhodospseudomonas*, *Rhodobacter*, and *Rhodospirillum*, are the most commonly utilized PSB in wastewater treatment. PNSB exhibit efficient absorption of diverse organic substances (with a biomass yield close to 1 g COD biomass/g COD removed), enabling the recovery of various value-added products through biomass harvesting (Liu et al., 2022). Therefore, the PSB-based system has a wide range of organic load, from municipal wastewater to aquaculture wastewater with a COD over 4000 mg/L (Hülßen et al., 2018). In general, PSB exhibit superior efficiency in terms of photoheterotrophic growth compared to microalgae and are particularly well-suited

for wastewater with a high carbon/nitrogen (C/N) ratio. However, PSB in wastewater are susceptible to contamination from other bacteria, especially in open systems. Consequently, closed systems such as photobioreactors are typically required, which raises the overall cost (Liu et al., 2022). In addition, developing an industrial-grade photobioreactor specifically for PSB-based wastewater treatment remains a challenging task. Urgent research is imperative in evaluating safety, ensuring stable operation, devising effective separation and purification methods, and designing scalable systems.

5.3 Algal-bacterial symbiotic interaction for nitrate removal

Algal-bacterial symbiosis (ABS) system, which stands for the integration of microalgae technology with conventional activated sludge methods, has demonstrated promising outcomes in improving treatment efficiency, reducing energy consumption, and minimizing carbon emissions (Peng et al., 2018). In the ABS system, algae capture dissolved CO_2 released through bacterial respiration to produce oxygen through photosynthesis. Moreover, bacteria can utilize the oxygen produced by algae to biodegrade pollutants (Li et al., 2023b). This process enables a reduction in both aeration energy costs and CO_2 generation. As a result of their cooperative interactions, microalgae and bacteria can establish mutually beneficial symbiotic relationships, resulting in enhanced performance compared to single microalgae or activated sludge systems (Zhou et al., 2022).

The ABS system achieves nitrogen removal through algal assimilation and bacterial nitrification-denitrification processes. In this system, nitrification primarily takes place during daylight hours with high photosynthetic activity, while denitrification predominantly occurs at night when photosynthesis and oxygen are not available (Peng et al., 2018). The symbiotic association between microalgae and bacteria favors the overall nitrogen

removal performance. Microalgae presence provides the necessary oxygen for the nitrification process, thereby reducing operational costs and mitigating the risks of pollutant volatilization during mechanical aeration. For instance, it was reported that the incorporation of microalgae into activated sludge communities resulted in a 2.7-fold increase in nitrification performance (Bankston et al., 2020). In terms of denitrification, the organic carbon released by microalgae can serve as a carbon source for denitrifying bacteria. Additionally, algae can utilize secondary metabolites, such as vitamins and amino acids, released by bacteria to improve nitrogen assimilation (Li et al., 2023b). Furthermore, signal transduction through interkingdom molecular signals, plays important roles in the nitrogen removal performance of the ABS system (Wu et al., 2022). Notably, nitrification-denitrification processes are sometimes compatible with microalgae due to the temporal oscillations in dissolved oxygen (DO) during the dual cycle, which requires careful parameter management to control the DO level, including light-dark cycle, nitrogen concentration, light intensity, and algae-bacteria ratio (Fallahi et al., 2021). Additionally, although the existence of bacteria can assist in flocculation of microalgae (Wang et al., 2022a), the biomass harvesting and separation of microalgae in suspended ABS remains a problem.

5.4 Photogranule as a new approach toward sustainable nitrate removal

Photogranule, also known as algae-bacterial granular sludge (ABGS), is an innovative granule-based ABS system used in wastewater treatment (Abouhend et al., 2018; Ji et al., 2020). Photogranules integrate microalgae, *Filamentous cyanobacteria*, Phototrophic bacteria, nitrifiers, and denitrifiers within a single biogranule (Trebuch et al., 2023a). In comparison to suspended ABS systems, photogranules exhibit a higher biomass density, resulting in a reduced HRT of 6–12 h

and rapid settling in just a few minutes (Ji and Liu, 2021). As a consequence of the high biomass density and availability of light, mature photogranules commonly establish physical-chemical gradients and anoxic zones, leading to biomass stratification (Trebuch et al., 2023a). Notably, filamentous cyanobacteria serve as crucial components within photogranules, contributing to the development and maintenance of granule structure (Abouhend et al., 2020). The *Filamentous cyanobacteria* can extensively colonize the entire photogranules and act as the backbone of the granule (Fig. 3(a)), which provides more binding sites for bacterial attachment (Kong et al., 2023a). Microalgae are primarily located in the outer layer of the photogranules, followed by photosynthetic bacteria. Aerobic bacteria, such as ammonium-oxidizing bacteria, reside close to the microalgae and *Cyanobacteria*, consuming the oxygen and creating an anoxic core for denitrifiers and other anaerobic bacteria. As a result, photogranules can simultaneously achieve nitrification and denitrification (SNAD) under light illumination. Due to the close spatial distance between algae and bacteria within the granule, the O_2 , CO_2 , and other metabolites could be effectively shared by each other. Therefore, mechanical aeration and external carbon sources are unnecessary for photogranules under proper light intensity, significantly lower CO_2 emissions than suspended ABS system.

The photogranules consist of a diverse bacterial community that works synergistically to remove pollutants (Fig. 3(b)). The coexisting bacteria not only supply essential vitamins for algal growth but also facilitate extracellular polymeric substances (EPS) biosynthesis by utilizing the partially derived nucleotide sugars from *Filamentous cyanobacteria*, which contribute to granule stability and nitrogen removal (Kong et al., 2023a). The complex carbon compound (i.e., polysaccharide, algal organic matter) released by algae can be decomposed to available carbon sources (i.e., formate, acetate) by certain degrading bacteria and utilized by denitrifying bacteria

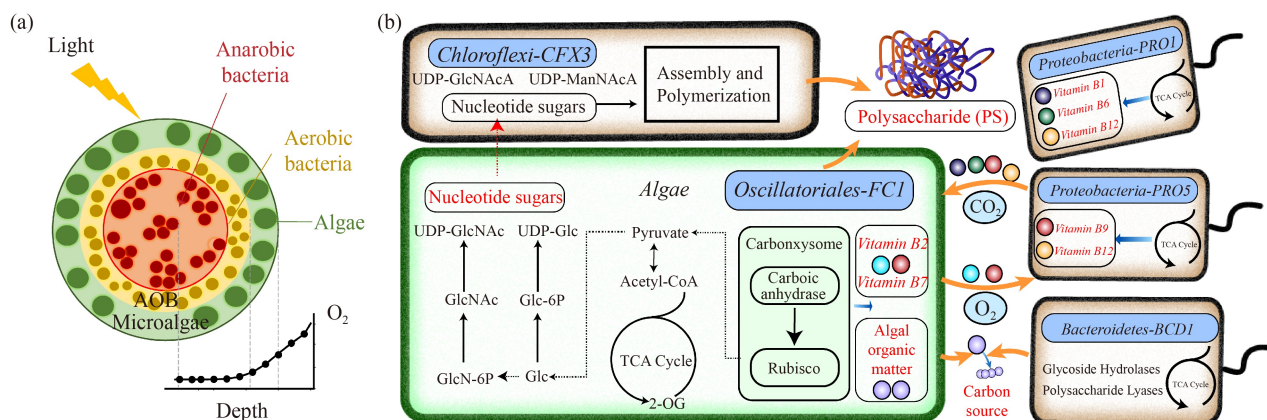


Fig. 3 (a) Confocal laser scanning (CLSM) microscopy image of a photogranule cross section; (b) Model of metabolite exchange in photogranules between *Filamentous cyanobacteria* and symbiotic bacteria.

(Kong et al., 2023a). Notably, photogranule can provide diverse niches for different bacteria, which allows the integration of various functional bacteria into the granules to improve the system performance. By successfully engineering methanotrophs within the photogranule communities, this engineered system achieved a high dissolved methane removal rate of 26 mg CH₄/(L·d), attributed to the potential syntrophic relationship between methanotrophs and *Filamentous cyanobacteria* (*Leptolyngbya* and *Phormidium*) within the the photogranules (Safitri et al., 2021). By incorporating polyphosphate accumulating organisms (PAOs) into photogranules, the overall phosphorus removal rate was improved by 6-fold (Trebuch et al., 2023c). By successfully assembling anammox bacteria into photogranules, a notable nitrogen removal rate of 294 mg N/(L·d) was achieved (Kong et al., 2023b). The interkingdom cross-feeding relationship between the microalgae (*Chlorella* sp.) and the anammox bacteria contributed to the overall nitrogen removal performance. Furthermore, the integration of anammox and photogranules was estimated to yield a negative carbon emission of 0.47 kg-CO₂/kg-N-removed, which offers a feasible approach for achieving carbon-neutral wastewater treatment (Kong et al., 2023b). The integration of various functional bacteria into photogranules enhances overall efficiency by providing diverse niches for different bacteria and facilitating synergistic pollutant removal. Table 4 compares the differences in nitrate removal technologies of several photosynthetic microorganisms, highlighting the role of photogranules in efficient nitrogen removal.

Life cycle assessment (LCA) has demonstrated the sustainability and environmental feasibility of laboratory-scale photogranules, and the energy neutral of the photogranule-based system may be within reach (Brockmann et al., 2021). Similarly, thermodynamic modeling support that photogranules achieve negative entropy, which entails the conversion of unavailable disordered energy (light) into available ordered energy of low entropy (Wang et al., 2023d). Overall, the potential

applications of photogranule-based wastewater treatment are promising while further researches are still necessary in several areas. These include the development of mature granulation technology to enhance the cultivation efficiency of photogranules, as well as finding ways to maintain the stability of the granules during long-term operation. Additionally, research on downstream treatment processes for harvested photogranules is needed to explore effective methods for further treatment of wastewater. These areas of research will contribute to the advancement and optimization of photogranule-based wastewater treatment systems.

5.5 Challenges

The application of photosynthetic microorganisms in nitrogen removal presents an innovative and eco-friendly approach. Successful implementation of these technologies in full-scale wastewater treatment plants may require infrastructure modifications. Challenges such as the availability of light, water turbidity, toxic pollutants, and invasive microorganisms need to be addressed for the reliable application of photosynthetic microorganisms. The combination of photosynthetic microorganisms with advanced pretreatment technology, membrane processes, or adsorption techniques, could enhance the overall performance. Despite these challenges, development efforts in engineer scale-up, downstream treatment processes, and resource recovery offer great promise for the future advancement and widespread implementation of this technology in practical wastewater treatment systems.

6 Photoelectrotrophic denitrification

6.1 The types of photoelectrotrophic denitrification

Light energy is a sustainable and renewable energy source that extends beyond its well-known role in

Table 4 The operational conditions and performance of different photosynthetic microorganisms-based processes

Process	Light conditions	Microorganisms	Nitrogen removal rate (mg N/(L·d))	Ref.
Microalgae-based system	White LED 100 mW/cm ²	<i>Chlorella</i> sp. <i>Phormidium</i> sp.	13.5	Kong et al. (2022)
PSB-based system	> 790 nm Filtered sunlight	<i>Rhodospseudomonas</i> sp. <i>Blastochloris</i> sp.	10.5	Hülßen et al. (2022)
PSB-based system	810–850 nm 12h dark/12h light 50 mW/cm ²	<i>Rhodospseudomonas</i> sp.	5.8	Sepúlveda-Muñoz et al. (2020)
ABS system	White LED 100 mW/cm ²	<i>Chlorella vulgaris</i> . Activates sludge	7.5	Huang et al. (2023)
Photogranule-based systems	White LED 200 mW/cm ²	<i>Chlorella</i> sp. Activates sludge	99.4	Ji et al. (2020)
Photogranule-based systems	White LED 100 mW/cm ²	<i>Ca. Brocadia</i> sp. <i>Chlorella</i> sp. Activates sludge	294	Kong et al. (2023b)

photosynthesis. Actually, many non-photosynthetic microorganisms also possess the ability to capture light energy (Gomelsky and Hoff, 2011). Non-photosynthetic aerobic denitrifier *Alcaligenes faecalis* and iron-oxidizing bacteria *Acidithiobacillus ferrooxidans* were first reported that able to convert solar energy into chemical energy through the photocatalysis of semiconductor minerals under light irradiation, and the growth rate is related to the intensity and wavelengths of light (Lu et al., 2012). Since then, more and more non-photosynthetic bacteria have been found to have the ability to utilize photoelectrons generated by semiconductors as a vital energy source for growth and metabolism (Sakimoto et al., 2016; Jin et al., 2021), which is also named photoelectrophily metabolism and has garnered significant attention in recent years due to its potential applications in sustainable energy production and environmental remediation. When the energy of incident light irradiation exceeds the bandgap between the valence and conduction bands of the photosensitizer, it generates photoelectrons and photoexcited holes, thereby leading to redox reactions (Pallikkara and Ramakrishnan, 2021). Except for semiconductors, dissolved organic matter (DOM) can also act as photosensitizers (Huang et al., 2022), inducing photoelectrons and offering an alternative avenue for some microorganisms to access light energy.

Importantly, bacteria can utilize photoelectrons generated by photosensitizers for denitrification, also known as photoelectrotrophic denitrification (PEDeN) (Cheng et al., 2017). *Thiobacillus denitrificans* is a non-photosynthetic bacterium that can directly use electrodes as electron acceptors for denitrification and is used as a model bacterium for research about PEDeN (Yu et al., 2015; Chen et al., 2019b). PEDeN uses photoelectrons as electron donors and does not have the problems of large amounts of remaining sludge, secondary pollution and difficult transportation in traditional denitrification (Fu et al., 2022). Besides, unlike general photocatalytic and photoelectrochemical systems, PEDeN does not require the supply of additional organic matter or electricity (Cheng et al., 2017), which also hints at its application potential in low-carbon wastewater treatment. According

to different photosensitizers, PEDeN systems can be divided into semiconductor-mediated systems and DOM-mediated systems. The various operational conditions and performance of different PEDeN systems are shown in Table 5.

TiO₂ as a low-cost, non-toxic, and stable semiconductor material, is widely used in the field of optoelectronics (Guo et al., 2019). The conduction band of TiO₂ exceeds the energy level required for nitrate reduction, thereby offering the potential for PEDeN through the utilization of photoelectrons by microorganisms (Wang et al., 2016). Microorganisms can almost entirely reduce nitrate to N₂ where TiO₂ serves as a photosensitizer, and light intensity would affect the removal rate (Cheng et al., 2017). When TiO₂@carbon paper used as a photosensitizer, the degradation of pyridine with high concentration and denitrification can also be achieved simultaneously (Shi et al., 2022). On the contrary, when using CdS as a photosensitizer, the product typically generated in PEDeN is N₂O rather than N₂, as sulfides, reactive oxygen species (ROS), and free nitrous acid (FNA) can all inhibit the activity of nitrous oxide reductase (Chen et al., 2019b; Chen et al., 2020a). Besides, Anthraquinone-2-sulfonate (AQDS) is an electron shuttle and is also photosensitive (Xu et al., 2022; Zeng et al., 2023). When AQDS is used as the photosensitizer, nitrate is almost completely converted into N₂O, and the quantum yield is higher than that of other semiconductor materials, acting as a capacitor (Chen et al., 2022). While N₂O can serve as a propellant for rocket launches (Kamps et al., 2019), it is also a potent greenhouse gas, drawing considerable attention toward the need for its emissions reduction (Ravishankara et al., 2009). Among the denitrification, N₂O reductase is very sensitive to environmental changes, especially oxygen, so the presence of ROS usually leads to the inhibition of its activity (Speir et al., 2023), causing the accumulation of N₂O. It has been reported that H₂O₂ and superoxide radicals ($\cdot\text{O}_2^-$) have a stronger inhibitory effect on N₂O reductase than O₂, moreover, higher concentrations of nitrate or nitrite may lead to increased production of ROS (Chen et al., 2020a).

Therefore, in order to achieve low-carbon denitrifica-

Table 5 The operational conditions and nitrogen removal performance of different photoelectrotrophic denitrification systems

Photosensitizer	Light conditions	Microorganisms	Nitrogen removal performance	Ref.
TiO ₂	380 ± 20 nm 30/45/60 mW/cm ²	Activated sludge	Under 60 mW/cm ² irradiation, the denitrification rate constant reaches 0.15 ± 0.02 h ⁻¹ , which is 36.36% higher under 30 mW/cm ² irradiation, and the nitrate is almost converted to N ₂ .	Cheng et al. (2017)
CdS	395 ± 5 nm 3.07 ± 0.14 mW/cm ²	<i>Thiobacillus denitrificans</i>	The denitrification rate constant is 0.081 ± 0.011 h ⁻¹ , the percentage of N ₂ O-N is > 96.4% ± 0.4%	Chen et al. (2019b)
CdS@Mn ₃ O ₄	395 ± 5 nm 3.07 ± 0.14 mW/cm ²	<i>Thiobacillus denitrificans</i>	The percentage of N ₂ -N exceeds 80%	Chen et al. (2020b)
DOM	395 ± 5 nm 1–10 mW/cm ²	<i>Thiobacillus denitrificans</i>	The denitrification rate constant is 0.06 ± 0.003 h ⁻¹ , percentage of N ₂ -N is 94.3% ± 5.5%	Huang et al. (2022)
AQS	400 ± 5 nm 0.62 mW/cm ²	<i>Thiobacillus denitrificans</i>	The percentage of N ₂ O-N is almost 100%	Chen et al. (2022)

tion, ROS should be cleared in time. Using Mn_3O_4 nanoenzyme as an *in situ* coating on the surface of bio-semiconductors can effectively suppress the production of N_2O by reducing ROS and serving as a physical protective layer, and the conversion of nitrate to N_2 can exceed 80% (Chen et al., 2020b). DOM in surface water appears to be an effective photosensitizer for N_2 production by PEDeN (Huang et al., 2022). DOM play a vital role in scavenging ROS (Wenk et al., 2013) thereby reducing inhibition of nitrous oxide reductase.

6.2 Photoelectron transfer mechanism involved in photoelectrotrophic denitrification

Since PEDeN is an emerging nitrate removal technology, it is necessary to clarify the mechanism to provide guidance for improving nitrogen removal performance. Microbial extracellular electron transfer pathways can be categorized into two modes: direct electron transfer mediated by cytochrome c on the outer membrane or conductive pili, and indirect electron transfer using redox substances as electron shuttles (Shi et al., 2016). Denitrifiers have been shown to have extracellular electron transport capabilities, the cytochrome c in the outer membrane and the redox substances in the extracellular polymeric substances help them absorb electrons from the electrode (Yu et al., 2015; Sathishkumar et al., 2020). However, there are still relatively few studies on the mechanism of microorganisms to take in photoelectrons.

First, a key of PEDeN is the generation of photoelectrons, which are generated when the intensity of the incident light exceeds the band gap of the photosensitizer, thus providing electron donors for the reduction of nitrate (Cheng et al., 2017). Therefore, the incident light is usually ultraviolet light with high energy rather than visible light, and this also requires photosensitizers such as semiconductor whose conduction band energy is higher than the energy level of nitrate reduction ($E(2\text{NO}_3^-/\text{N}_2) = +0.791 \text{ V vs. SHE at pH } 7$) (Benz et al., 1998). The conduction band energy levels of semiconductors such as TiO_2 and CdS are usually negative (Cheng et al., 2017; Cheng et al., 2018), so the photoelectrons are enough to reduce nitrate without inputting additional electrical energy. DOM is also capable of generating photoelectrons, which mainly due to its fulvic acid-like (FA) substances and some reducing functional groups (Huang et al., 2022). DOM undergoes intramolecular charge transfer to form a triplet-state DOM under light irradiation and then the photoelectrons are transferred to denitrifiers through membrane-bound cytochrome c, enabling nitrate reduction (Huang et al., 2022). AQDS is typically used as an analog for NOM. Similarly, AQDS undergoes electron transitions to become triplet-state AQDS under light irradiation, providing photoelectrons directly to denitrifiers. Additionally,

AQDS can act as a capacitor, releasing the majority of photoelectrons in the dark conditions (Chen et al., 2022).

Another important factor for achieving PEDeN is the ability of denitrifiers to transfer photoelectrons from extracellular to intracellular to reduce nitrate. The current mechanism for microbial photoelectron transfer is primarily mediated by membrane-bound proteins (cytochromes, ferredoxin, flavoproteins) and hydrogenases (H_2ase). In *Moorella thermoacetica*-CdS system, over an extended period (24 h), H_2ase dominate in the photoelectron uptake, leading to the production of hydrogen, which subsequently enters the Wood-Ljungdahl pathway, resulting in the production of acetic acid while in the short-term (less than 3 h), non- H_2ase enzymes play a significant role (Kornienko et al., 2016). Among membrane-bound proteins, cytochrome c appears to be the most important transfer mediator of photoelectrons. Research has found that the direction and intensity of the photoelectrons generated by CdS nanoparticles are controlled by the activation of cytochromes c (Katz et al., 2006). As for *Geobacter sulfurreducens*-CdS biohybrid, photoelectrons can be stored in OmcS (a cytochrome c in outer membrane) and further directly used by bacteria (Huang et al., 2019). In the *Shewanella oneidensis*-CdS biohybrid system, omcA and mtrC play a leading role in photoelectron transfer (Zhang et al., 2023a). Essentially, this is because the redox potential of these cytochrome c proteins is more favorable than the conduction band energy level of CdS, making it thermodynamically advantageous for electrons to transfer from CdS to them (Huang et al., 2021). It is worth noting that EPS also plays a crucial role in this system, and the redox substances within EPS will aid in the transfer of photoelectrons (Zhang et al., 2023a). Similarly, for denitrifier *Thiobacillus denitrificans*, a series of cytochrome c is thought to play an important role in denitrification (Beller et al., 2013). In *Thiobacillus denitrificans*-CdS system, cytc4 located in the periplasmic space is considered to be a key protein mediating extracellular photoelectron transfer (Fig. 4). Some of the photoelectrons are transferred to Nar through ubiquinone (Q/QH₂), and the other part is absorbed by cyt4 and further transferred to other enzymes involved in denitrification (Zhou et al., 2023).

6.3 Challenges

By using photoelectrons as the electron donor and requiring no external carbon source, PEDeN is a promising low-carbon wastewater treatment technology. While PEDeN is primarily conducted under light irradiation in the near-ultraviolet range (around 400 nm) at present, it should be further extended to broader response spectra, such as the visible light spectrum, and even under natural sunlight to achieve nitrate removal.

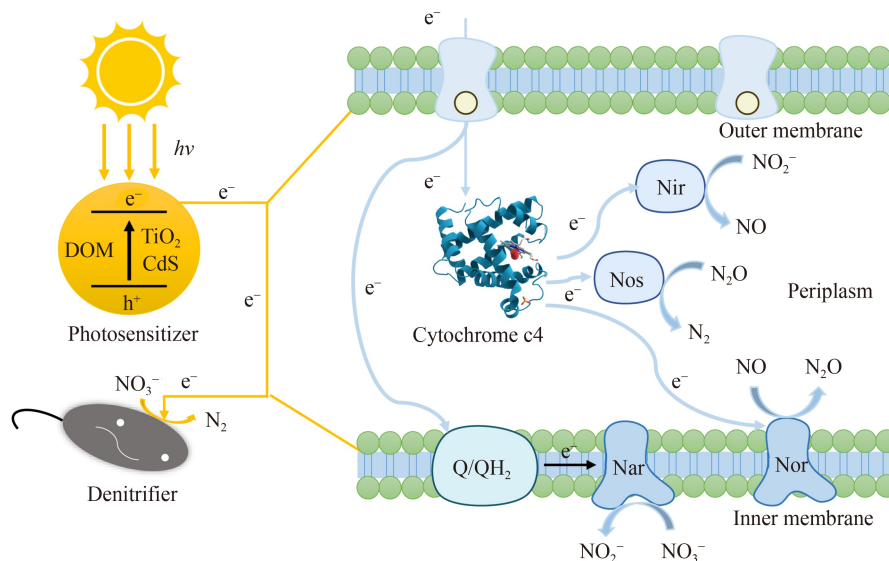


Fig. 4 The photoelectron transfer mechanism involved in photoelectrotrophic denitrification.

For example, semiconductor materials can be modified through doping and dye sensitization to respond to visible light (Dong et al., 2015). Some naturally occurring minerals such as hematite have been proven to be effective as photoelectric anodes (Li et al., 2021). Besides, *Thiobacillus denitrificans* is a model bacterium that is currently well studied. Actually, many non-photosynthetic bacteria have been shown to be able to use photoelectrons for metabolism. Therefore, we can continue to deeply explore the photoelectric metabolic capabilities of other nitrogen-cycling microorganisms, thereby broadening the application of photoelectric biological water treatment. The elucidation of the extracellular photoelectron transfer mechanism in microorganisms is crucial for enhancing the denitrification rate while current research in this area is limited. In the future, the use of multi-omics techniques and single-bacteria electrochemical imaging methods can further deepen our understanding of the complex relationship between microorganisms and photosensitizers. Additionally, this will provide recommendations for the design and optimization of future solar-driven nitrogen removal systems.

7 Future prospects

This study critically reviews the latest advancements and microbial mechanisms in autotrophic denitrification processes guided by SAD, HAD, n-DAMO, photosynthetic microorganisms-based system and PEDeN. SAD exhibits advantages of low cost for inorganic sulfur compounds, high bioavailability, and the potential for coupled treatment of various pollutants. HAD has the

advantages of less secondary pollution and clean products. n-DAMO is a sustainable wastewater treatment technology that can simultaneously achieve methane emission reduction and nitrogen removal. Photosynthetic microorganisms offer significant advantages in reducing carbon emissions and achieving resource recovery. PEDeN allows continuous utilization of solar energy, achieving low-carbon denitrification through photoelectrons. However, these technologies still face challenges in engineering implementation.

Cost remains a significant issue as SAD and PEDeN require external addition of inorganic sulfur compounds or semiconductor materials. In addition, SAD will produce by-products such as sulfate, which will undoubtedly cause expensive treatment costs. Besides, the safety and low solubility of H_2 in HAD also limit its large-scale application. Gas methane in the n-DAMO process is flammable and dissolved methane is difficult to recover. The availability of light and the turbidity of the water in a photosynthetic microbial system both have a huge impact on performance. Table 6 summarizes the advantages, disadvantages and challenges of various denitrification technologies.

Thus, we proposed several future research directions and objectives to address these limitations:

- 1) Expansion of the application of autotrophic denitrification through combined processes. For example, the combination of SAD and PN/A technology can enhance denitrification performance and reduce sulfate yield (Deng et al., 2019). The integration of HAD, MEC, and MBfR can effectively solve the problem of low H_2 solubility (Han et al., 2023b). Integrating various functional bacteria with photogranules can also help improve nitrogen removal performance, which can all help to aid the application of these processes (Kong et al.,

Table 6 The advantages, disadvantages, and challenges of various denitrification technologies

Process	Advantages	Disadvantages	Challenges
SAD	Low cost of sulfur compounds; high bioavailability	Excess SO_4^{2-} and H_2S emission	No identified optimal options to reduce the production of SO_4^{2-}
HAD	Clean and no by-products	Low bioavailability, high cost, poor safety of H_2	Finding the methods of H_2 production with lower cost and solving the safety issues of H_2 transportation
n-DAMO	Simultaneous Methane emission reduction and nitrogen removal	Low growth rate of n-damo microorganisms	Difficulty of recovering the dissolved methane, relatively low nitrogen removal rate
Photosynthetic microorganisms-based system	Negative carbon emissions; resource recycling	Prone to contamination, unstable, poor sedimentation	Solving issues such as light homogeneity, water turbidity, toxic contaminants, and invasive microorganisms
PEDeN	Sustainable and efficient	Unstable photosensitizer and requirement of additional semiconductor materials	Need to apply with a wider spectrum response range

2023b). The two-stage A/B process is currently a sustainable way to achieve energy-neutral for municipal wastewater (Kang et al., 2023). Autotrophic denitrification technology can be coupled with the carbon source recovery process. COD is recovered in the front stage, and autotrophic denitrification is performed after nitrification in the backstage to achieve deep nitrogen removal.

2) Exploration of nitrogen removal microorganisms that can utilize light energy and design of solar-driven nitrogen removal systems. It is worth noting that light energy is a sufficient clean energy source and the energy source for all life activities on earth. The Earth receives more light energy in one hour than humans consume in an entire year (Lewis and Nocera, 2006). Light energy can not only provide energy for photosynthetic bacteria and mediate biological sewage biological treatment processes (such as bacterial and algal symbiosis systems), but also provide energy for some chemoautotrophic bacteria (such as *Thiobacillus denitrificans*) to use photoelectrons for denitrification. Light energy can also drive the conversion of organic pollutants into chemicals, reduce chemical production costs and CO_2 emissions, and achieve efficient resource utilization of pollutants (Pi et al., 2023). In the future, we can use metagenomics techniques to explore nitrogen removal microorganisms that can utilize light energy and further design solar-driven nitrogen removal systems, which will be of great significance to carbon emission reduction. In this system, the homogeneity of light, shading effects between microorganisms, and algae growth are all factors worth considering (Wang et al., 2021b; Ahangar et al., 2023). In addition, photoelectric and photomagnetic coupling technologies will help further expand the development of low-carbon biological nitrogen removal technology (Zheng et al., 2024).

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Conflict of Interests The 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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References

- Abouhend A S, Mcnair A, Kuo-Dahab W C, Watt C, Butler C S, Milferstedt K, Hamelin J, Seo J, Gikonyo G J, El-Moselhy K M, et al. (2018). The oxygenic photogranule process for aeration-free wastewater treatment. *Environmental Science & Technology*, 52(6): 3503–3511
- Abouhend A S, Milferstedt K, Hamelin J, Ansari A A, Butler C, Carbajal-Gonzalez B I, Park C (2020). Growth progression of oxygenic photogranules and its impact on bioactivity for aeration-free wastewater treatment. *Environmental Science & Technology*, 54(1): 486–496
- Ahangar A K, Yaqoubnejad P, Divsalar K, Mousavi S, Taghavijeloudar M (2023). Design a novel internally illuminated mirror photobioreactor to improve microalgae production through homogeneous light distribution. *Bioresource Technology*, 387: 129577
- Baalsrud K, Baalsrud K S (1954). Studies on *Thiobacillus denitrificans*. *Archives of Microbiology*, 20: 34–62
- Bankston E, Wang Q, Higgins B T (2020). Algae support populations of heterotrophic, nitrifying, and phosphate-accumulating bacteria in the treatment of poultry litter anaerobic digestate. *Chemical Engineering Journal*, 398: 125550
- Batchelor B, Lawrence A W (1978). Autotrophic denitrification using elemental sulfur. *Journal of Water Pollution Control Federation*, 50(8): 1986–2001
- Beller H R, Zhou P, Legler T C, Chakicherla A, Kane S, Letain T E A, O'Day P (2013). Genome-enabled studies of anaerobic, nitrate-

- dependent iron oxidation in the *Chemolithoautotrophic* bacterium *Thiobacillus denitrificans*. *Frontiers in Microbiology*, 4: 249
- Benz M, Brune A, Schink B (1998). Anaerobic and aerobic oxidation of ferrous iron at neutral pH by chemoheterotrophic nitrate-reducing bacteria. *Archives of Microbiology*, 169(2): 159–165
- Bijay-Singh, Craswell E (2021). Fertilizers and nitrate pollution of surface and ground water: an increasingly pervasive global problem. *SN Applied Sciences*, 3(4): 518
- Blankenship R E (2010). Early evolution of photosynthesis. *Plant Physiology*, 154(2): 434–438
- Boelee N C, Temmink H, Janssen M, Buisman C J N, Wijffels R H (2014). Balancing the organic load and light supply in symbiotic microalgal–bacterial biofilm reactors treating synthetic municipal wastewater. *Ecological Engineering*, 64: 213–221
- Brettar I, Rheinheimer G (1991). Denitrification in the Central Baltic: evidence for H₂S-oxidation as motor of denitrification at the oxic-anoxic interface. *Marine Ecology Progress Series*, 77(2–3): 157–169
- Brockmann D, Gérard Y, Park C, Milferstedt K, Hélias A, Hamelin J (2021). Wastewater treatment using oxygenic photogranule-based process has lower environmental impact than conventional activated sludge process. *Bioresource Technology*, 319: 124204
- Burgdorf T, Lenz O, Bührke T, Van Der Linden E, Jones A K, Albracht S P J, Friedrich B (2006). [NiFe]-hydrogenases of *Ralstonia eutropha* H16: modular enzymes for oxygen-tolerant biological hydrogen oxidation. *Journal of Molecular Microbiology and Biotechnology*, 10(2–4): 181–196
- Cai C, Hu S, Guo J, Shi Y, Xie G J, Yuan Z (2015). Nitrate reduction by denitrifying anaerobic methane oxidizing microorganisms can reach a practically useful rate. *Water Research*, 87: 211–217
- Cao K, Zhi R, Zhang G (2020). Photosynthetic bacteria wastewater treatment with the production of value-added products: a review. *Bioresource Technology*, 299: 122648
- Carlson H K, Lui L M, Price M N, Kazakov A E, Carr A V, Kuehl J V, Owens T K, Nielsen T, Arkin A P, Deutschbauer A M (2020). Selective carbon sources influence the end products of microbial nitrate respiration. *ISME Journal*, 14(8): 2034–2045
- Carmona-Martínez A A, Trably E, Milferstedt K, Lacroix R, Etcheverry L, Bernet N (2015). Long-term continuous production of H₂ in a microbial electrolysis cell (MEC) treating saline wastewater. *Water Research*, 81: 149–156
- Chen D, Gu X, Zhu W, He S, Huang J, Zhou W (2019a). Electrons transfer determined greenhouse gas emissions in enhanced nitrogen-removal constructed wetlands with different carbon sources and carbon-to-nitrogen ratios. *Bioresource Technology*, 285: 121313
- Chen J, Strous M (2013). Denitrification and aerobic respiration, hybrid electron transport chains and co-evolution. *Biochimica et Biophysica Acta*, 1827(2): 136–144
- Chen M, Cai Q, Chen X, Huang S, Feng Q, Majima T, Zeng R J, Zhou S (2022). Anthraquinone-2-sulfonate as a microbial photosensitizer and capacitor drives solar-to-N₂O production with a quantum efficiency of almost unity. *Environmental Science & Technology*, 56(8): 5161–5169
- Chen M, Zhou X, Chen X, Cai Q, Zeng R J, Zhou S (2020a). Mechanisms of nitrous oxide emission during photoelectrotrophic denitrification by self-photosensitized *Thiobacillus denitrificans*. *Water Research*, 172: 115501
- Chen M, Zhou X F, Yu Y Q, Liu X, Zeng R J X, Zhou S G, He Z (2019b). Light-driven nitrous oxide production via autotrophic denitrification by self-photosensitized *Thiobacillus denitrificans*. *Environment International*, 127: 353–360
- Chen X, Feng Q, Cai Q, Huang S, Yu Y, Zeng R J, Chen M, Zhou S (2020b). Mn₃O₄ nanozyme coating accelerates nitrate reduction and decreases N₂O emission during photoelectrotrophic denitrification by *Thiobacillus denitrificans*-CdS. *Environmental Science & Technology*, 54(17): 10820–10830
- Cheng H Y, Tian X D, Li C H, Wang S S, Su S G, Wang H C, Zhang B, Sharif H M A, Wang A J (2017). Microbial photoelectrotrophic denitrification as a sustainable and efficient way for reducing nitrate to nitrogen. *Environmental Science & Technology*, 51(21): 12948–12955
- Cheng L, Xiang Q, Liao Y, Zhang H (2018). CdS-based photocatalysts. *Energy & Environmental Science*, 11(6): 1362–1391
- Cui Y X, Biswal B K, Van Loosdrecht M C M, Chen G H, Wu D (2019). Long term performance and dynamics of microbial biofilm communities performing sulfur-oxidizing autotrophic denitrification in a moving-bed biofilm reactor. *Water Research*, 166: 115038
- Dawood F, Anda M, Shafiqullah G M (2020). Hydrogen production for energy: an overview. *International Journal of Hydrogen Energy*, 45(7): 3847–3869
- Deng S, Peng Y, Zhang L, Wu L (2020). Advanced nitrogen removal from municipal wastewater via two-stage partial nitrification-simultaneous anammox and denitrification (PN-SAD) process. *Bioresource Technology*, 304: 122955
- Deng Y F, Ekama G A, Cui Y X, Tang C J, Van Loosdrecht M C M, Chen G H, Wu D (2019). Coupling of sulfur(thiosulfate)-driven denitrification and anammox process to treat nitrate and ammonium contained wastewater. *Water Research*, 163: 114854
- Deng Y F, Tang W T, Huang H, Qian J, Wu D, Chen G H (2021). Development of a kinetic model to evaluate thiosulfate-driven denitrification and anammox (TDDA) process. *Water Research*, 198: 117155
- Deng Y F, Zan F X, Huang H, Wu D, Tang W T, Chen G H (2022). Coupling sulfur-based denitrification with anammox for effective and stable nitrogen removal: A review. *Water Research*, 224: 119051
- Devlin J F, Eedy R, Butler B J (2000). The effects of electron donor and granular iron on nitrate transformation rates in sediments from a municipal water supply aquifer. *Journal of Contaminant Hydrology*, 46(1–2): 81–97
- Di Capua F, Pirozzi F, Lens P N L, Esposito G (2019). Electron donors for autotrophic denitrification. *Chemical Engineering Journal*, 362: 922–937
- Dijkhuizen L, Harder W (1984). Current views on the regulation of autotrophic carbon dioxide fixation via the Calvin cycle in bacteria. *Antonie van Leeuwenhoek*, 50: 473–487
- Ding J, Zeng R J (2021). Fundamentals and potential environmental significance of denitrifying anaerobic methane oxidizing archaea. *Science of the Total Environment*, 757: 143928
- Dong S, Feng J, Fan M, Pi Y, Hu L, Han X, Liu M, Sun J, Sun J (2015). Recent developments in heterogeneous photocatalytic water treatment using visible light-responsive photocatalysts: a review.

- RSC Advances, 5(19): 14610–14630
- Du W J, Lu J Y, Hu Y R, Xiao J, Yang C, Wu J, Huang B, Cui S, Wang Y, Li W W (2023). Spatiotemporal pattern of greenhouse gas emissions in China's wastewater sector and pathways towards carbon neutrality. *Nature Water*, 1(2): 166–175
- Epsztein R, Beliaevski M, Tarre S, Green M (2016). High-rate hydrogenotrophic denitrification in a pressurized reactor. *Chemical Engineering Journal*, 286: 578–584
- Ergas S J, Reuss A F (2001). Hydrogenotrophic denitrification of drinking water using a hollow fibre membrane bioreactor. *Journal of Water Supply: Research & Technology–Aqua*, 50(3): 161–171
- Fallahi A, Rezvani F, Asgharnejad H, Khorshidi Nazloo E, Hajinajaf N, Higgins B (2021). Interactions of microalgae-bacteria consortia for nutrient removal from wastewater: a review. *Chemosphere*, 272: 129878
- Fan S Q, Wen W R, Xie G J, Lu Y, Nie W B, Liu B F, Xing D F, Ma J, Ren N Q (2023). Revisiting the engineering roadmap of nitrate/nitrite-dependent anaerobic methane oxidation. *Environmental Science & Technology*, 57(50): 20975–20991
- Fan S Q, Xie G J, Lu Y, Zhao Z C, Liu B F, Xing D F, Ding J, Han H J, Ren N Q (2021). Mainstream nitrogen and dissolved methane removal through coupling n-DAMO with anammox in granular sludge at low temperature. *Environmental Science & Technology*, 55(24): 16586–16596
- Fewtrell L (2004). Drinking-water nitrate, methemoglobinemia, and global burden of disease: a discussion. *Environmental Health Perspectives*, 112(14): 1371–1374
- Fu X, Hou R, Yang P, Qian S, Feng Z, Chen Z, Wang F, Yuan R, Chen H, Zhou B (2022). Application of external carbon source in heterotrophic denitrification of domestic sewage: a review. *Science of the Total Environment*, 817: 153061
- Gomelsky M, Hoff W D (2011). Light helps bacteria make important lifestyle decisions. *Trends in Microbiology*, 19(9): 441–448
- Grubba D, Yin Z, Majtacz J, Al-Hazmi H E, Mąkinia J (2022). Incorporation of the sulfur cycle in sustainable nitrogen removal systems - a review. *Journal of Cleaner Production*, 372: 133495
- Guo Q, Zhou C, Ma Z, Yang X (2019). Fundamentals of TiO₂ photocatalysis: concepts, mechanisms, and challenges. *Advanced Materials*, 31(50): 1901997
- Guo X, Lai C Y, Hartmann E M, Zhao H P (2023). Heterotrophic denitrification: an overlooked factor that contributes to nitrogen removal in n-DAMO mixed culture. *Environmental Research*, 216: 114802
- Guo Y, Luo Z, Shen J, Li Y (2022). The main anammox-based processes, the involved microbes and the novel process concept from the application perspective. *Frontiers of Environmental Science & Engineering*, 16(7): 84
- Gupta D, Singh S K (2012). Greenhouse gas emissions from wastewater treatment plants: a case study of Noida. *Journal of Water Sustainability*, 2(2): 131–139
- Han F, Zhou W (2022). Nitrogen recovery from wastewater by microbial assimilation: a review. *Bioresource Technology*, 363: 127933
- Han Y, Feng Y, Wang N, Yang P, Ding G, An J, Liu J, Li N, He W (2023a). Liquid-gas phase transition enables microbial electrolysis cell to treat organic pollution and synchronously remediate nitrate-contaminated groundwater via hydrogenotrophic denitrification. *Journal of Cleaner Production*, 414: 137627
- Han Y, Yang P, Feng Y, Wang N, Yuan X, An J, Liu J, Li N, He W (2023b). Liquid-gas phase transition enables microbial electrolysis and H₂-based membrane biofilm hybrid system to degrade organic pollution and achieve effective hydrogenotrophic denitrification of groundwater. *Chemosphere*, 331: 138819
- Hao J, Gao F, Fang X, Nong X, Zhang Y, Hong F (2022). Multi-factor decomposition and multi-scenario prediction decoupling analysis of China's carbon emission under dual carbon goal. *Science of the Total Environment*, 841: 156788
- He Z, Feng Y, Zhang S, Wang X, Wu S, Pan X (2018). Oxygenic denitrification for nitrogen removal with less greenhouse gas emissions: microbiology and potential applications. *Science of the Total Environment*, 621: 453–464
- Hu Y, Wu G, Li R, Xiao L, Zhan X (2020). Iron sulphides mediated autotrophic denitrification: an emerging bioprocess for nitrate pollution mitigation and sustainable wastewater treatment. *Water Research*, 179: 115914
- Huang J, Cheng S, Zhang Y, Teng J, Zhang M, Lin H (2023). Optimizing aeration intensity to enhance self-flocculation in algal-bacterial symbiosis systems. *Chemosphere*, 341: 140064
- Huang M, Liu C, Cui P, Wu T, Feng X, Huang H, Zhou J, Wang Y (2021). Facet-dependent photoinduced transformation of cadmium sulfide (CdS) nanoparticles. *Environmental Science & Technology*, 55(19): 13132–13141
- Huang S, Chen M, Diao Y, Feng Q, Zeng R J, Zhou S (2022). Dissolved organic matter acting as a microbial photosensitizer drives photoelectrotrophic denitrification. *Environmental Science & Technology*, 56(7): 4632–4641
- Huang S, Tang J, Liu X, Dong G, Zhou S (2019). Fast light-driven biodecolorization by a *Geobacter sulfurreducens*-CdS biohybrid. *ACS Sustainable Chemistry & Engineering*, 7(18): 15427–15433
- Hülßen T, Hsieh K, Tait S, Barry E M, Puyol D, Batstone D J (2018). White and infrared light continuous photobioreactors for resource recovery from poultry processing wastewater: a comparison. *Water Research*, 144: 665–676
- Hülßen T, Stegman S, Batstone D J, Capson-Tojo G (2022). Naturally illuminated photobioreactors for resource recovery from piggery and chicken-processing wastewaters utilising purple phototrophic bacteria. *Water Research*, 214: 118194
- Ji B, Liu Y (2021). Assessment of microalgal-bacterial granular sludge process for environmentally sustainable municipal wastewater treatment. *ACS ES&T Water*, 1(12): 2459–2469
- Ji B, Zhang M, Gu J, Ma Y, Liu Y (2020). A self-sustaining synergetic microalgal-bacterial granular sludge process towards energy-efficient and environmentally sustainable municipal wastewater treatment. *Water Research*, 179: 115884
- Jiang M, Zheng J, Perez-Calleja P, Picioreanu C, Lin H, Zhang X, Zhang Y, Li H, Nerenberg R (2020). New insight into CO₂-mediated denitrification process in H₂-based membrane biofilm reactor: an experimental and modeling study. *Water Research*, 184: 116177
- Jin S, Jeon Y, Jeon M S, Shin J, Song Y, Kang S, Bae J, Cho S, Lee J K, Kim D R, et al. (2021). Acetogenic bacteria utilize light-driven electrons as an energy source for autotrophic growth. *Proceedings*

- of the National Academy of Sciences of the United States of America, 118(9): e2020552118
- Kamps L, Sakurai K, Saito Y, Nagata H (2019). Comprehensive data reduction for N₂O/HDPE hybrid rocket motor performance evaluation. *Aerospace*, 6(4): 45
- Kang D, Zhao X, Wang N, Suo Y, Yuan J, Peng Y (2023). Redirecting carbon to recover VFA to facilitate biological short-cut nitrogen removal in wastewater treatment: a critical review. *Water Research*, 238: 120015
- Katz E, Zayats M, Willner I, Lisdat F (2006). Controlling the direction of photocurrents by means of CdS nanoparticles and cytochrome c-mediated biocatalytic cascades. *Chemical Communications*, 13: 1395–1397
- Kong L, Feng Y, Du W, Zheng R, Sun J, Rong K, Sun W, Liu S (2023a). Cross-feeding between *Filamentous cyanobacteria* and symbiotic bacteria favors rapid photogranulation. *Environmental Science & Technology*, 57(44): 16953–16963
- Kong L, Feng Y, Sun J, Rong K, Zhou J, Zheng R, Ni S, Liu S (2022). Cross-feeding among microalgae facilitates nitrogen recovery at low C/N. *Environmental Research*, 211: 113052
- Kong L, Zheng R, Feng Y, Du W, Xie C, Gu Y, Liu S (2023b). Anammox bacteria adapt to long-term light irradiation in photogranules. *Water Research*, 241: 120144
- Kornienko N, Sakimoto K K, Herlihy D M, Nguyen S C, Alivisatos A P, Harris C B, Schwartzberg A, Yang P (2016). Spectroscopic elucidation of energy transfer in hybrid inorganic–biological organisms for solar-to-chemical production. *Proceedings of the National Academy of Sciences of the United States of America*, 113(42): 11750–11755
- Kumar A, Bera S (2020). Revisiting nitrogen utilization in algae: a review on the process of regulation and assimilation. *Bioresource Technology Reports*, 12: 100584
- Kumar P K, Vijaya Krishna S, Verma K, Pooja K, Bhagawan D, Himabindu V (2018). Phycoremediation of sewage wastewater and industrial flue gases for biomass generation from microalgae. *South African Journal of Chemical Engineering*, 25: 133–146
- Kyung D, Kim M, Chang J, Lee W (2015). Estimation of greenhouse gas emissions from a hybrid wastewater treatment plant. *Journal of Cleaner Production*, 95: 117–123
- Lackner S, Gilbert E M, Vlaeminck S E, Joss A, Horn H, Van Loosdrecht M C M (2014). Full-scale partial nitrification/anammox experiences: an application survey. *Water Research*, 55: 292–303
- Lee J W, Lee K H, Park K Y, Maeng S K (2010). Hydrogenotrophic denitrification in a packed bed reactor: effects of hydrogen-to-water flow rate ratio. *Bioresource Technology*, 101(11): 3940–3946
- Lee Y Y, Choi H, Cho K S (2019). Effects of carbon source, C/N ratio, nitrate, temperature, and pH on N₂O emission and functional denitrifying genes during heterotrophic denitrification. *Journal of Environmental Science and Health. Part A, Toxic/Hazardous Substances & Environmental Engineering*, 54(1): 16–29
- Lenton T M, Rockström J, Gaffney O, Rahmstorf S, Richardson K, Steffen W, Schellnhuber H J (2019). Climate tipping points—too risky to bet against. *Nature*, 575(7784): 592–595
- Lewis N S, Nocera D G (2006). Powering the planet: chemical challenges in solar energy utilization. *Proceedings of the National Academy of Sciences of the United States of America*, 103(43): 15729–15735
- Li J, Chen H, Triana C A, Patzke G R (2021). Hematite photoanodes for water oxidation: electronic transitions, carrier dynamics, and surface energetics. *Angewandte Chemie*, 133(34): 18528–18544
- Li J, Liu T, Mcilroy S J, Tyson G W, Guo J (2023a). Phylogenetic and metabolic diversity of microbial communities performing anaerobic ammonium and methane oxidations under different nitrogen loadings. *ISME Communications*, 3(1): 39
- Li S N, Zhang C, Li F, Ren N Q, Ho S H (2023b). Recent advances of algae-bacteria consortia in aquatic remediation. *Critical Reviews in Environmental Science and Technology*, 53(3): 315–339
- Li X, Shi M, Zhang M, Li W, Xu P L, Wang Y, Yuan Y, Huang Y (2022). Progresses and challenges in sulfur autotrophic denitrification-enhanced anammox for low carbon and efficient nitrogen removal. *Critical Reviews in Environmental Science and Technology*, 52(24): 4379–4394
- Liang B, Kang F, Wang Y, Zhang K, Wang Y, Yao S, Lyu Z, Zhu T (2022a). Denitrification performance of sulfur-based autotrophic denitrification and biomass-sulfur-based mixotrophic denitrification in solid-phase denitrifying reactors using novel composite filters. *Science of the Total Environment*, 823: 153826
- Liang B, Kang F, Yao S, Zhang K, Wang Y, Chang M, Lyu Z, Zhu T (2022b). Exploration and verification of the feasibility of the sulfur-based autotrophic denitrification integrated biomass-based heterotrophic denitrification systems for wastewater treatment: from feasibility to application. *Chemosphere*, 287: 131998
- Liang D, He W, Li C, Wang F, Crittenden J C, Feng Y (2021). Remediation of nitrate contamination by membrane hydrogenotrophic denitrifying biofilm integrated in microbial electrolysis cell. *Water Research*, 188: 116498
- Lim Z K, Liu T, Zheng M, Yuan Z, Guo J, Hu S (2021). Versatility of nitrite/nitrate-dependent anaerobic methane oxidation (n-DAMO): first demonstration with real wastewater. *Water Research*, 194: 116912
- Liu C, Liu T, Zheng X, Meng J, Chen H, Yuan Z, Hu S, Guo J (2021a). Rapid formation of granules coupling n-DAMO and anammox microorganisms to remove nitrogen. *Water Research*, 194: 116963
- Liu L Y, Xie G J, Xing D F, Liu B F, Ding J, Cao G L, Ren N Q (2021b). Sulfate dependent ammonium oxidation: a microbial process linked nitrogen with sulfur cycle and potential application. *Environmental Research*, 192: 110282
- Liu S, Li H, Daigger G T, Huang J, Song G (2022). Material biosynthesis, mechanism regulation and resource recycling of biomass and high-value substances from wastewater treatment by photosynthetic bacteria: a review. *Science of the Total Environment*, 820: 153200
- Liu T, Hu S, Yuan Z, Guo J (2019). High-level nitrogen removal by simultaneous partial nitrification, anammox and nitrite/nitrate-dependent anaerobic methane oxidation. *Water Research*, 166: 115057
- Liu T, Hu S, Yuan Z, Guo J (2023). Microbial stratification affects conversions of nitrogen and methane in biofilms coupling Anammox and n-DAMO processes. *Environmental Science & Technology*, 57(11): 4608–4618
- Liu T, Li J, Khai Lim Z, Chen H, Hu S, Yuan Z, Guo J (2020a). Simultaneous removal of dissolved methane and nitrogen from

- synthetic mainstream anaerobic effluent. *Environmental Science & Technology*, 54(12): 7629–7638
- Liu T, Lim Z K, Chen H, Wang Z, Hu S, Yuan Z, Guo J (2020b). Biogas-driven complete nitrogen removal from wastewater generated in side-stream partial nitrification. *Science of the Total Environment*, 745: 141153
- Lu A, Li Y, Jin S, Wang X, Wu X L, Zeng C, Li Y, Ding H, Hao R, Lv M, et al. (2012). Growth of non-phototrophic microorganisms using solar energy through mineral photocatalysis. *Nature Communications*, 3(1): 768
- Lu L, Guest J S, Peters C A, Zhu X, Rau G H, Ren Z J (2018). Wastewater treatment for carbon capture and utilization. *Nature Sustainability*, 1(12): 750–758
- Lyu W, Huang L, Xiao G, Chen Y (2017). Effects of carbon sources and COD/N ratio on N₂O emissions in subsurface flow constructed wetlands. *Bioresource Technology*, 245: 171–181
- Ma B, Liang Y, Zhang Y, Wei Y (2023). Achieving advanced nitrogen removal from low-carbon municipal wastewater using partial-nitrification/anammox and endogenous partial-denitrification/anammox. *Bioresource Technology*, 383: 129227
- Ma Y, Zheng X, Fang Y, Xu K, He S, Zhao M (2020). Autotrophic denitrification in constructed wetlands: achievements and challenges. *Bioresource Technology*, 318: 123778
- Matějů V, Čížinská S, Krejčí J, Janoch T (1992). Biological water denitrification: a review. *Enzyme and Microbial Technology*, 14(3): 170–183
- Nie W B, Xie G J, Ding J, Peng L, Lu Y, Tan X, Yue H, Liu B F, Xing D F, Meng J, et al. (2020). Operation strategies of n-DAMO and anammox process based on microbial interactions for high rate nitrogen removal from landfill leachate. *Environment International*, 139: 105596
- Pallikkara A, Ramakrishnan K (2021). Efficient charge collection of photoanodes and light absorption of photosensitizers: a review. *International Journal of Energy Research*, 45(2): 1425–1448
- Pan Y, Ye L, Yuan Z (2013). Effect of H₂S on N₂O reduction and accumulation during denitrification by methanol utilizing denitrifiers. *Environmental Science & Technology*, 47(15): 8408–8415
- Pang Y, Wang J (2021). Various electron donors for biological nitrate removal: a review. *Science of the Total Environment*, 794: 148699
- Peng L, Ngo H H, Guo W S, Liu Y, Wang D, Song S, Wei W, Nghiem L D, Ni B J (2018). A novel mechanistic model for nitrogen removal in algal-bacterial photo sequencing batch reactors. *Bioresource Technology*, 267: 502–509
- Pi S, Yang W, Feng W, Yang R, Chao W, Cheng W, Cui L, Li Z, Lin Y, Ren N, et al. (2023). Solar-driven waste-to-chemical conversion by wastewater-derived semiconductor biohybrids. *Nature Sustainability*, 6: 1673–1684
- Pokorna D, Zabranska J (2015). Sulfur-oxidizing bacteria in environmental technology. *Biotechnology Advances*, 33(6): 1246–1259
- Rahimi S, Modin O, Mijakovic I (2020). Technologies for biological removal and recovery of nitrogen from wastewater. *Biotechnology Advances*, 43: 107570
- Ravishankara A R, Daniel J S, Portmann R W (2009). Nitrous oxide (N₂O): the dominant ozone-depleting substance emitted in the 21st century. *Science*, 326(5949): 123–125
- Ross B N, Lancellotti B V, Brannon E Q, Loomis G W, Amador J A (2020). Greenhouse gas emissions from advanced nitrogen-removal onsite wastewater treatment systems. *Science of the Total Environment*, 737: 140399
- Roth F, Broman E, Sun X, Bonaglia S, Nascimento F, Prytherch J, Brüchert V, Lundevall Zara M, Brunberg M, Geibel M C, et al. (2023). Methane emissions offset atmospheric carbon dioxide uptake in coastal macroalgae, mixed vegetation and sediment ecosystems. *Nature Communications*, 14(1): 42
- Ruiz-Romero E, Alcántara-Hernández R, Cruz-Mondragon C, Marsch R, Luna-Guido M L, Dendooven L (2009). Denitrification in extreme alkaline saline soils of the former lake Texcoco. *Plant and Soil*, 319: 247–257
- Safitri A S, Hamelin J, Kommedal R, Milferstedt K (2021). Engineered methanotrophic syntrophy in photogranule communities removes dissolved methane. *Water Research X*, 12: 100106
- Sakimoto K K, Wong A B, Yang P (2016). Self-photosensitization of nonphotosynthetic bacteria for solar-to-chemical production. *Science*, 351(6268): 74–77
- Sathishkumar K, Li Y, Sanganyado E (2020). Electrochemical behavior of biochar and its effects on microbial nitrate reduction: role of extracellular polymeric substances in extracellular electron transfer. *Chemical Engineering Journal*, 395: 125077
- Sepúlveda-Muñoz C A, De Godos I, Puyol D, Muñoz R (2020). A systematic optimization of piggery wastewater treatment with purple phototrophic bacteria. *Chemosphere*, 253: 126621
- Shi H, Chen D, Jiang X, Li Y, Fan W, Hou C, Zhang L, Wang Y, Mu Y, Shen J (2022). Simultaneous high-concentration pyridine removal and denitrification in an electricity assisted biophotodegradation system. *Chemical Engineering Journal*, 430: 132598
- Shi L, Dong H, Reguera G, Beyenal H, Lu A, Liu J, Yu H Q, Fredrickson J K (2016). Extracellular electron transfer mechanisms between microorganisms and minerals. *Nature Reviews Microbiology*, 14(10): 651–662
- Shukla P R, Skea J, Calvo Buendia E, Masson-Delmotte V, Pörtner H O, Roberts D, Zhai P, Slade R, Connors S, Van Diemen R (2019). IPCC, 2019: Climate Change and Land: an IPCC Special Report on Climate Change, Desertification, Land Degradation, Sustainable Land Management, Food Security, and Greenhouse Gas Fluxes in Terrestrial Ecosystems. Cambridge: Cambridge University Press
- Speir S L, Tank J L, Taylor J M, Grose A L (2023). Temperature and carbon availability interact to enhance nitrous oxide production via denitrification in alluvial plain river sediments. *Biogeochemistry*, 165(2): 191–203
- Stomp M, Huisman J, Stal L J, Matthijs H C P (2007). Colorful niches of phototrophic microorganisms shaped by vibrations of the water molecule. *ISME Journal*, 1(4): 271–282
- Tian T, Yu H Q (2020). Denitrification with non-organic electron donor for treating low C/N ratio wastewaters. *Bioresource Technology*, 299: 122686
- Trebuch L M, Bourceau O M, Vaessen S M F, Neu T R, Janssen M, De Beer D, Vet L E M, Wijffels R H, Fernandes T V (2023a). High resolution functional analysis and community structure of photogranules. *ISME Journal*, 17(6): 870–879

- Trebuch L M, Schoofs K, Vaessen S M F, Neu T R, Janssen M, Wijffels R H, Vet L E M, Fernandes T V (2023b). N₂-fixation can sustain wastewater treatment performance of photogranules under nitrogen-limiting conditions. *Biotechnology and Bioengineering*, 120(5): 1303–1315
- Trebuch L M, Sohier J, Altenburg S, Oyserman B O, Pronk M, Janssen M, Vet L E, Wijffels R H, Fernandes T V (2023c). Enhancing phosphorus removal of photogranules by incorporating polyphosphate accumulating organisms. *Water Research*, 235: 119748
- Valdovinos-García E M, Barajas-Fernández J, Olán-Acosta M D L Á, Petriz-Prieto M A, Guzmán-López A, Bravo-Sánchez M G (2020). Techno-economic study of CO₂ capture of a thermoelectric plant using microalgae (*Chlorella vulgaris*) for production of feedstock for bioenergy. *Energies*, 13(2): 413
- Vasilidou I A, Pavlou S, Vayenas D V (2006). A kinetic study of hydrogenotrophic denitrification. *Process Biochemistry* (Barking, London, England), 41(6): 1401–1408
- Wang C, Wu Y, Hu W, Li Y (2021a). Autotrophic denitrification for nitrate removal from groundwater with an integrated microbial fuel cells (MFCs)-microbial electrolysis cell (MEC) system. *International Journal of Electrochemical Science*, 16(1): 150973
- Wang H, Wu P, Zheng D, Deng L, Wang W (2022a). N-Acyl-Homoserine Lactone (AHL)-mediated microalgal-bacterial communication driving *Chlorella*-activated sludge bacterial biofilm formation. *Environmental Science & Technology*, 56(17): 12645–12655
- Wang J, Hua M, Li Y, Ma F, Zheng P, Hu B (2019). Achieving high nitrogen removal efficiency by optimizing nitrite-dependent anaerobic methane oxidation process with growth factors. *Water Research*, 161: 35–42
- Wang J J, Huang B C, Li J, Jin R C (2020). Advances and challenges of sulfur-driven autotrophic denitrification (SDAD) for nitrogen removal. *Chinese Chemical Letters*, 31(10): 2567–2574
- Wang L, Qiu S, Guo J, Ge S (2021b). Light irradiation enables rapid start-up of nitrification through suppressing *nxrB* gene expression and stimulating ammonia-oxidizing bacteria. *Environmental Science & Technology*, 55(19): 13297–13305
- Wang Q, Jiang G, Ye L, Pijuan M, Yuan Z (2014). Heterotrophic denitrification plays an important role in N₂O production from nitrification reactors treating anaerobic sludge digestion liquor. *Water Research*, 62: 202–210
- Wang Q, Xu J, Ge Y, Zhang Y, Feng H, Cong Y (2016). Efficient nitrogen removal by simultaneous photoelectrocatalytic oxidation and electrochemically active biofilm denitrification. *Electrochimica Acta*, 198: 165–173
- Wang S, Wang Y, Li P, Wang L, Su Q, Zuo J (2022b). Development and characterizations of hydrogenotrophic denitrification granular process: nitrogen removal capacity and adaptability. *Bioresource Technology*, 363: 127973
- Wang S S, Cheng H Y, Zhang H, Su S G, Sun Y L, Wang H C, Han J L, Wang A J, Guadie A (2021c). Sulfur autotrophic denitrification filter and heterotrophic denitrification filter: comparison on denitrification performance, hydrodynamic characteristics and operating cost. *Environmental Research*, 197: 111029
- Wang T, Li X, Wang H, Xue G, Zhou M, Ran X, Wang Y (2023a). Sulfur autotrophic denitrification as an efficient nitrogen removals method for wastewater treatment towards lower organic requirement: a review. *Water Research*, 245: 120569
- Wang W, Zhang Y, Yin T M, Zhao L, Xu X J, Xing D F, Zhang R C, Lee D J, Ren N Q, Chen C (2023b). Prospect of denitrifying anaerobic methane oxidation (DAMO) application on wastewater treatment and biogas recycling utilization. *Science of the Total Environment*, 905: 167142
- Wang Y, Zheng X, Wu G, Guan Y (2023c). Removal of ammonium and nitrate through anammox and FeS-driven autotrophic denitrification. *Frontiers of Environmental Science & Engineering*, 17(6): 74
- Wang Z, Chen W, Wang J, Gao M, Zhang D, Zhang S, Hao Y, Song H (2023d). Exploring the mechanism and negentropy of photogranules for efficient carbon, nitrogen and phosphorus recovery from wastewater. *Chemical Engineering Journal*, 476: 146510
- Wenk J, Eustis S N, Mcneill K, Canonica S (2013). Quenching of excited triplet states by dissolved natural organic matter. *Environmental Science & Technology*, 47(22): 12802–12810
- Winkler M K H, Straka L (2019). New directions in biological nitrogen removal and recovery from wastewater. *Current Opinion in Biotechnology*, 57: 50–55
- Wu C, Qin Y, Yang L, Liu Z, Chen B, Chen L (2020). Effects of loading rates and N/S ratios in the sulfide-dependent autotrophic denitrification (SDAD) and anammox coupling system. *Bioresource Technology*, 316: 123969
- Wu J, Yin Y, Wang J (2018). Hydrogen-based membrane biofilm reactors for nitrate removal from water and wastewater. *International Journal of Hydrogen Energy*, 43(1): 1–15
- Wu X, Kong L, Pan J, Feng Y, Liu S (2022). Metagenomic approaches to explore the quorum sensing-mediated interactions between algae and bacteria in sequence membrane photo-bioreactors. *Frontiers in Bioengineering and Biotechnology*, 10: 851376
- Xie G J, Liu T, Cai C, Hu S, Yuan Z (2018). Achieving high-level nitrogen removal in mainstream by coupling anammox with denitrifying anaerobic methane oxidation in a membrane biofilm reactor. *Water Research*, 131: 196–204
- Xu J, Xie J, Wang Y, Xu L, Zong Y, Pang W, Xie L (2022). Effect of anthraquinone-2, 6-disulfonate (AQDS) on anaerobic digestion under ammonia stress: triggering mediated interspecies electron transfer (MIET). *Science of the Total Environment*, 828: 154158
- Xue M, Nie Y, Cao X, Zhou X (2022). Deciphering the influence of S/N ratio in a sulfite-driven autotrophic denitrification reactor. *Science of the Total Environment*, 836: 155612
- Yang W, Zhao Q, Lu H, Ding Z, Meng L, Chen G H (2016). Sulfide-driven autotrophic denitrification significantly reduces N₂O emissions. *Water Research*, 90: 176–184
- Yao X, Wang J, He M, Liu Z, Zhao Y, Li Y, Chi T, Zhu L, Zheng P, Jetten M S M, et al. (2024). Methane-dependent complete denitrification by a single *Methylomirabilis* bacterium. *Nature Microbiology*, 9: 464–476
- Yu L, Yuan Y, Chen S, Zhuang L, Zhou S (2015). Direct uptake of electrode electrons for autotrophic denitrification by *Thiobacillus denitrificans*. *Electrochemistry Communications*, 60: 126–130
- Yuan Z, Chen Y, Zhang M, Qin Y, Zhang M, Mao P, Yan Y (2022).

- Efficient nitrite accumulation and elemental sulfur recovery in partial sulfide autotrophic denitrification system: insights of seeding sludge, S/N ratio and flocculation strategy. *Chemosphere*, 288: 132388
- Zeng Y, Fu Q, Dionysiou D D, Zhang M, Li M, Ye B, Chen N, Gao J, Wang Y, Zhou D, et al. (2023). Triplet chromophoric dissolved organic matter regulating the phototransformation and toxicity of imidacloprid in paddy water. *Chemical Engineering Journal*, 474: 145636
- Zhan M, Zeng W, Liu H, Li J, Meng Q, Peng Y (2023). Simultaneous nitrogen and sulfur removal through synergy of sulfamox, anammox and sulfur-driven autotrophic denitrification in a modified bioreactor enhanced by activated carbon. *Environmental Research*, 232: 116341
- Zhang K, Kang T, Yao S, Liang B, Chang M, Wang Y, Ma Y, Hao L, Zhu T (2020). A novel coupling process with partial nitrification-anammox and short-cut sulfur autotrophic denitrification in a single reactor for the treatment of high ammonium-containing wastewater. *Water Research*, 180: 115813
- Zhang Q, Xu X, Zhang R, Shao B, Fan K, Zhao L, Ji X, Ren N, Lee D J, Chen C (2022a). The mixed/mixotrophic nitrogen removal for the effective and sustainable treatment of wastewater: from treatment process to microbial mechanism. *Water Research*, 226: 119269
- Zhang Q, Xu X, Zhou X, Chen C (2022b). Recent advances in autotrophic biological nitrogen removal for low carbon wastewater: a review. *Water*, 14(7): 1101
- Zhang S, Li C, Ke C, Liu S, Yao Q, Huang W, Dang Z, Guo C (2023a). Extracellular polymeric substances sustain photoreduction of Cr(VI) by *Shewanella oneidensis*-CdS biohybrid system. *Water Research*, 243: 120339
- Zhang X, An L, Tian J, Ji B, Lu J, Liu Y (2023b). Microalgal capture of carbon dioxide: a carbon sink or source? *Bioresource Technology*, 390: 129824
- Zhang X N, Zhu L, Li Z R, Sun Y L, Qian Z M, Li S Y, Cheng H Y, Wang A J (2022c). Thiosulfate as external electron donor accelerating denitrification at low temperature condition in S⁰-based autotrophic denitrification biofilter. *Environmental Research*, 210: 113009
- Zheng R, Feng Y, Kong L, Wu X, Zhou J, Zhang L, Liu S (2024). Blue-light irradiation induced partial nitrification. *Water Research*, 254: 121381
- Zhou S, Zhang Z, Sun Z, Song Z, Bai Y, Hu J (2021). Responses of simultaneous anammox and denitrification (SAD) process to nitrogen loading variation: start-up, performance, sludge morphology and microbial community dynamics. *Science of the Total Environment*, 795: 148911
- Zhou X, Huang S, Chen X, Jianxiong Zeng R, Zhou S, Chen M (2023). Mechanisms of extracellular photoelectron uptake by a *Thiobacillus denitrificans*-cadmium sulfide biosemiconductor system. *Chemical Engineering Journal*, 468: 143667
- Zhou Y, Zhou Y, Chen S, Guo N, Xiang P, Lin S, Bai Y, Hu X, Zhang Z (2022). Evaluating the role of algae in algal-bacterial granular sludge: nutrient removal, microbial community and granular characteristics. *Bioresource Technology*, 365: 128165
- Zhu T, Cai W, Wang B, Liu W, Feng K, Deng Y, Wang A (2019). Enhanced nitrate removal in an Fe⁰-driven autotrophic denitrification system using hydrogen-rich water. *Environmental Science. Water Research & Technology*, 5(8): 1380–1388