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Incorporation of the sulfur cycle in sustainable nitrogen removal systems - a review

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Abstract

In wastewater treatment systems, sulfur (S) removal processes are generally based on heterotrophic sulfate (SO_4^{2-}) reduction by sulfate reducing bacteria and S-dependent autotrophic denitrification by sulfur oxidizing bacteria. A combination of either two cycles (N and S) or three cycles (N, S and C) appears to be a viable approach to sustainable wastewater treatment, resulting in energy savings and reduction of sludge production. This review shows how the S cycle can be coupled with the other cycles in single systems for efficient N and S removal. Operating conditions, advantages, limitations and challenges of such systems are described. S removal processes are generally based on heterotrophic sulfate (SO_4^{2-}) reduction by sulfate reducing bacteria and S-dependent autotrophic denitrification by sulfur oxidizing bacteria. In terms of pH and temperature, the optimum conditions are determined by the narrowest ranges for heterotrophic SO_4^{2-} reduction (pH of 7-7.6, T = 28-30°C). The combined processes allow for almost complete N removal, while the efficiency of SO_4^{2-} removal can reach up to 75%. Among all the processes linking the N, S and C cycles, SANI (sulfate reduction, autotrophic denitrification and nitrification integrated) has been best recognized. Recently, the growing attention has been paid to the novel sulfammox process, which involves SO_4^{2-} dependent, anaerobic ammonia oxidizing bacteria. Numerous systems have been developed to combine SO_4^{2-} reduction, S-dependent autotrophic denitrification and partial nitritation/anammox processes. The coexistence of several bacterial groups and their competition for the substrates is thus a key issue to be considered. Specific inhibitors for each bacterial group also need to be recognized before full-scale implementations. Moreover, modeling the transformations of S

compounds has been incorporated with respect to all the processes responsible for those transformations.

Keywords: sulfur-dependent autotrophic denitrification; heterotrophic sulfate reduction; sulfammox; sulfur cycle; microbial community; mechanistic model

1. Introduction

High concentrations of ammonium (NH_4-N) lead to eutrophication of surface waters and pose a threat to the aquatic life and human health (Qin et al., 2021). NH_4-N can effectively be converted to nitrogen gas by combined nitrification-denitrification, but this method has a few important disadvantages, including a high demand of energy and carbon, and high sludge production. On the other hand, sulfate (SO_4^{2-}) is a type of the secondary pollutant because reduction of sulfide (S^{2-}) under anaerobic conditions is harmful for the aquatic environment (Hao et al. 2014). S compounds have not been widely used as substrates in wastewater treatment processes. Simultaneous removal of these two compounds (N and S) from wastewater, with or without involving the carbon (C) cycle, can be a viable approach to the sustainable wastewater management. In particular, this approach may be an effective alternative in the case of many types of industrial wastewater, which are characterized by high concentrations of pollutants, such as NH_4-N, SO_4^{2-} (>1000 mg/L of both N and S) and chemical oxygen demand (COD) (> 60,000 mg COD/L) (Rikmann et al., 2016, Jarvis and Younger 2000, Chapman, 1992).

A viable sustainable approach to biological wastewater treatment comprises a combination of nitrogen (N), sulfur (S) and carbon (C) removal. Lower operating costs result from the use of some products in one process as the substrates in other processes and the use of shared reactors. Moreover, no carbon is needed for S-dependent autotrophic denitrification, less sludge is generated, and the environmentally neutral compounds, such as nitrogen gas (N_2) and elementar sulfur (S^0), are the final products of biochemical reactions (Lin et al., 2018).

Conventional nitrification/denitrification for N removal is now being replaced by more sustainable N-shortcut processes, such as “nitrite shunt” or deammonification. In the case of S compounds, biological removal is based on heterotrophic SO_4^{2-} reduction by sulfate reducing

bacteria (SRB) and S-dependent autotrophic denitrification by sulfur oxidizing bacteria (SOB). Recently, the growing attention has been paid to the novel sulfate reducing ammonia oxidizing (sulfammox) process, which involves anaerobic ammonium oxidizing bacteria (AAOB). These bacteria use SO_4^{2-}, instead of nitrite nitrogen (NO_2^--N), as an electron acceptor to oxidize NH_4^+-N under anaerobic conditions.

The growing importance of using the combined N, S and C cycles in biological wastewater treatment processes has been confirmed by the increasing number of review papers on various aspects of S transformations. According to Web of Science database, 15, 12 and 3 review papers have been published specifically on S-dependent autotrophic denitrification, heterotrophic reduction of SO_4^{2-} and sulfammox (–see Figure S1 in the Supporting Information (SI)). Several papers focused on particular issues, including a detailed description of mechanisms of the individual processes, responsible microorganisms, reactors used, optimal operational conditions or inhibiting factors in S-dependent autotrophic denitrification (Wu et al., 2021, Cui et al., 2019, Lin et al., 2018), heterotrophic sulfate reduction (Sinharoy et al., 2020b) and sulfammox (Liu et al., 2021, Grubba et al., 2021).

However, only a combination of either two cycles – (N and S) or three cycles (N, S and C) would be the rational approach to wastewater treatment in order to save energy and the amount of sludge generated, especially for NH_4^+-N and SO_4^{2-} rich industrial wastewater. Due to the variety of N, S and C removal processes, the research interests have been shifting to the use of single- and multi-stage systems based on the combination of several processes, such as heterotrophic sulfate reduction, S-dependent autotrophic denitrification, nitrification, denitrification, anaerobic ammonia oxidation (anammox) and sulfammox (Wu et al., 2020, Yuan et al., 2020, Sun et al., 2018, Liu et al., 2017, Qian et al., 2015a, b, c, Jiang et al., 2013, Wang et al., 2009b).

Only two review papers (Hao et al., 2014, Show et al., 2013) described simultaneously S-dependent autotrophic denitrification and heterotrophic sulfate reduction. Hao et al. (2014) described a relationship between the N, S, C and P cycles in biological wastewater treatment systems. These authors focused on the acceptors and electrons used in the transformations of S compounds, key microorganisms, developed technologies, factors influencing the process performance, and achieved SO_4^{2-} reduction efficiencies. In the review of Show et al. (2013), existing models of the transformations of S compounds were additionally described (see – Table S1 in SI).

The present review provides updated results of research on S transformations, which have been revised and extended with new understanding and discoveries. A novel aspect is the inclusion of sulfammox in these transformations as no paper has synthesized autotrophic S-dependent denitrification, heterotrophic sulfate reduction and the sulfammox process in one review. In addition, the present study describes how sulfammox can increase the efficiency of N and S removal. Various process configurations and technologies, which are based on the three (N-S-C) cycles, are described and compared in terms of their efficiency. Moreover, modeling the transformations of N, S and C compounds has been incorporated with respect to all processes responsible for those transformations. Such a review provides a deeper insight into the conversions of S in biochemical processes, including sulfammox.

2. Single S-dependent biochemical processes integrating N, S and C conversions

There are three known processes combining sulfur and nitrogen conversions: S-dependent autotrophic denitrification, heterotrophic sulfate reduction and autotrophic sulfammox. The detailed description of those processes, including the metabolic mechanisms, biochemical reactions, influencing environmental factors can be found in the SI (S1-S3).

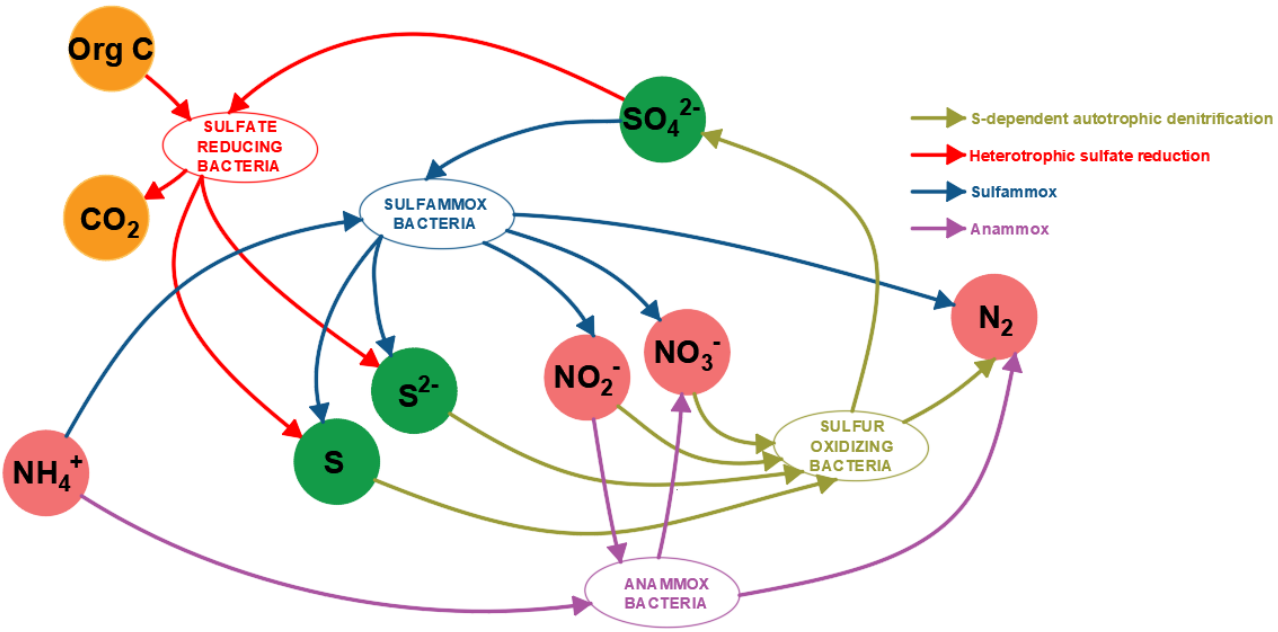
S-dependent autotrophic denitrification consists of oxidation of S compounds, including S^{2-}, S^0, thiosulfate ($S_2O_3^{2-}$) and sulfite (SO_3^{2-}), coupled with reduction of NO_3^--N and/or NO_2^--N. *T. denitrificans*, *Thiomicrospira denitrificans*, *Thiobacillus versutus*, *Thiosphaera pantotropha* and *P. denitrificans* are the known microorganisms responsible for that process. *P. denitrificans* is the chemotrophic α-proteobacteria which can grow on organic monocarbon compounds (methanol, methylamine) while using reduced forms of S and hydrogen as electron donors in denitrification (Baker et al., 1998). *T. denitrificans* belongs to β-proteobacteria that can use $S_2O_3^{2-}$ and thiocyanates under aerobic conditions, and additionally S^{2-} and S^0 under anaerobic conditions. *Sulfurimonas denitrificans* belongs to the ε-proteobacteria and is capable of oxidizing SO_3^{2-}, $S_2O_3^{2-}$ and S^0, while both NO_3^--N and oxygen are used as electron acceptors. *T. thioparus* is one of the representatives of autotrophic denitrifiers that reduce NO_3^--N to NO_2^--N by oxidation of S^{2-} (Tang et al., 2009). Although autotrophic denitrifying bacteria are chemolithotrophic, there are many denitrifying bacteria capable of adapting to autotrophic, heterotrophic and even mixotrophic growth (*P. versatus*, *P. denitrificans*, *Beggiatoa sp*.) (Pokorna and Zabranska, 2015).

Heterotrophic sulfate reduction is SO_4^{2-} reduction which takes place in two independent different paths. The first is the use of organic electron donors, which are also the carbon source for the SRB. The second is the use of inorganic electron donors, which must be supplemented with a carbon source, such as CO_2 (Sinharoy et al., 2020a). The SRB can be divided into 7 phylogenetic lines, including five for bacteria and two for archaea. Most of the SRB found in sulfate reduction reactors belong to 23 genera within *Deltaproteobacteria* (*Desulfovibrio, Desulfobacteraceae, Desulfobulbaceae, Syntrophobacteraceae, Desulfomicrobium, Desulfohalobium*). Another SRB belong to the gram-positive genera *Clostridia* (*Desulfotomaculum, Desulfosporosinus and Desulfosporomusa*). Three lineages, *Nitrospirae* (*Thermodesulfovibrio*), *Thermodesulfobacteria* (*Thermodesulfobacterium*) and

Thermodesulfobiaceae (*Thermodesulfobium*), contain only thermophilic SO_4^{2-} reducing agents. Archaeal SRB are *Euryarchaeota* and *Crenarchaeota* (Muyzer and Stams, 2008).

In a novel sulfammox process, NH_4^+-N is oxidized to N_2, whereas SO_4^{2-} plays the role of an electron acceptor which is reduced to S^0 under anaerobic conditions. *Brocadia Anammoxoglobus Sulfate* (Liu et al., 2008) is a functional microorganism responsible for simultaneous removal of NH_4^+-N and SO_4^{2-} and ended the conversion of NH_4^+-N and SO_4^{2-} by producing NO_2^--N as an intermediate. The second isolated species, *Bacillus Benzoevorans*, is responsible for carrying out the entire sulfammox reaction (Cai et al., 2010). *Verrucomicrobia* has also been reported to be involved in the sulfammox process (Rikmann et al., 2016). Some *Proteobacteria*, which may potentially perform sulfammox, include the following species: *Sulfurimonas, Desulfuromonadales, Desulfovibrio, Desulfuromonas, Desulfobulbus, norank Rhodobacteraceae* and *Thiobacillus* (Rios-Del Toro et al., 2018, Wang et al. 2017).

The key issues and challenges of S-dependent autotrophic denitrification, heterotrophic sulfate reduction and sulfammox are presented in Table 1. Figure 1 below shows the interactions between S-dependent autotrophic denitrification, heterotrophic sulfate reduction and sulfammox process.



115

116 Figure 1. Interactions between S-dependent autotrophic denitrification, heterotrophic sulfate
117 reduction, anammox and sulfammox process

118

119 **3. Operational conditions and performances of single S-dependent processes**

120 Each of the discussed processes (S-dependent autotrophic denitrification, heterotrophic sulfate
121 reduction and sulfammox) can be carried out independently, as evidenced by numerous
122 studies (Tables 2 and 3). However, the challenge is to combine these processes, in either
123 single- or multi-stage systems, in order to make biological wastewater treatment systems
124 more efficient.

125 ***3.1. S-dependent autotrophic denitrification***

126 In S-dependent autotrophic denitrification, the most frequently used electron donors are S^0
127 and S^{2-} (Table 2). The experiments were mainly carried out in packed bed reactors, but several
128 other types of reactors were also used. The reported rates of denitrification varied in a wide
129 range - from 0.03 to 8.13 kg $N/m^3/d$, depending mainly on the temperature and influent NO_3^--

N concentrations. The effects of pH in the investigated range (6.0-9.0) and S concentrations were less significant. For a detailed description of previous research related to S-dependent autotrophic denitrification, see the SI (S1). This process allowed for the efficient (>90%) removal of N and S^{2-} (Yang et al., 2016, Jing et al., 2010) with the NO_3^--N concentration in the range of 20-1230 mg N/L (Zhu et al., 2019, Zou et al., 2016, Kim et al., 2004).

During S-dependent autotrophic denitrification, SO_4^{2-} can be produced from different electron donors. Frequently, the S balance in the process is not 1/1 for the removed electron donor to SO_4^{2-} produced (Zou et al., 2016). In Table 2, the initial donor concentrations and the amount of SO_4^{2-} produced are similar. The observed imbalances result from the production of other S intermediates. The most common electron acceptor is NO_3^--N, but several studies comparing NO_3^--N and NO_2^--N have been reported (Sun and Nemati, 2012, Moraes et al., 2012, Jing et al 2010).

Different aspects of S-dependent autotrophic denitrification have been addressed in several reviews (Wu et al. 2021, Cui et al. 2019, Lin et al. 2018, Sabba et al. 2016). Wu et al. (2021) summarized all types of biofilm denitrification in terms of the reactor configuration, microbial transformations, factors influencing the process, and especially focused on N_2O emissions. The coexistence of S-dependent denitrification with anammox was also reported and S-driven denitrifiers were identified, including *Thiobacillus denitrificans* and *Thiobacillus thioparus*.

Cui et al. (2019) described S-dependent autotrophic denitrification in terms of the functional enzymes, electron donors, types of reactors, and operational factors. They also emphasized a significant advantage regarding S-dependent autotrophic denitrification compared to heterotrophic denitrification with respect to N_2O emissions. It was shown that

autotrophic denitrification mediated by S compounds (S^0, S^{2-}) emitted significantly less N_2O than heterotrophic denitrification with methanol, ethanol or acetate.

Sabba et al. (2016) focused mainly on SO_3^{2-} and its occurrence in the environment, chemistry, microbiology, and the role in denitrification. It was emphasized that SO_3^{2-} is an intermediate in the S oxidation pathway and should be chosen as the most economical electron donor. Lin et al. (2018) focused primarily on S oxidation, including biological gas desulfurization, phototrophic S^{2-} oxidation, S-dependent autotrophic denitrification, biological sulfur oxidation associated phosphorous removal, dye treatment. They also indicated viable applications of the products, such as Li batteries, production of S concrete by mixing S^0 with aggregates, biologically produced S fertilizer, oxidation of S^{2-} in microbiological fuel cells, and reclamation of metals from sewage sludge.

3.2. Heterotrophic sulfate reduction

Table 3 presents the diversity of research carried out so far on heterotrophic SO_4^{2-} reduction in terms of the electron donor, type of reactor and operating conditions. Most studies have been carried out in the gas lift reactor and fluidized-bed reactor. Both organic and inorganic donors were used, including carbon monoxide, methane, methanol, ethanol, hydrogen, crab shell, compost and many others. The use of different donors resulted in a different SO_4^{2-} reduction efficiency. A detailed description of the research can be found in SI (S2). The use of different electron donors and SO_4^{2-} content resulted in a wide range of SO_4^{2-} removal efficiencies (51-98%) and rates (0-3400 mg SO_4^{2-}/L/d). Nielsen et al. (2019) used methanol and ethylene glycol which resulted in reduction of SO_4^{2-} by 71.2% and 36.9%, respectively. The decrease of SO_4^{2-} concentration was limited to 13.8 and 5.3%, respectively, with the use of peat and straw. Low temperatures (below 10°C) significantly affected the SO_4^{2-} removal rates. For example, Virpiranta et al. (2019) carried out studies at various temperatures (22°C,

177 16°C, 6°C) and found gradually decreasing SO_4^{2-} removal rates, i.e. 169, 98 and 13-42 mg
178 SO_4^{2-}/L/d, respectively.

179 Sulfate reduction is less popular compared to S-dependent autotrophic denitrification,
180 but that process has also been addressed in several reviews (Kumar et al. 2021, Costa et al.,
181 2020, Sinharoy et al., 2020b, Serrano et al., 2019, Van den Brand et al., 2015). Kumar et al.
182 (2021) and Costa et al. (2020) focused on the use of SO_4^{2-} reduction for treatment of metal-
183 rich wastewater and recovery of these metals, showing a high degree of SO_4^{2-} reduction (>
184 90%) along with the efficient (> 99%) recovery of metals (Fe, Zn, Cd, Cu).

185 Similarly, Sinharoy et al. (2020b) described treatment of acid mine drainage (AMD)
186 with biological reduction of SO_4^{2-}. Heavy metals present in AMD can be removed by S^{2-}
187 precipitation. The review discussed various gaseous substrates, such as H_2, CO, CH_4, as
188 electron donors that could be used in this process. It was emphasized that only the
189 microorganisms capable of using gaseous substrates are appropriate for the AMD treatment
190 systems.

191 Serrano et al. (2019) focused on the optimum conditions for SRB. They presented the
192 recommended conditions for biomass, electron donor and acceptor and an experimental setup
193 of three SRB tests: (1) to assess the activity of SRB culture, (2) to determine the reduction
194 potential of an electron donor, and (3) to determine the possibility of using various sources of
195 SO_4^{2-} as an electron acceptor. They collected methodologies and results from many
196 publications and recommended setup and monitoring conditions to increase the comparability
197 and reproducibility of the SRB tests. Sodium sulfate and lactate were used as an electron
198 acceptor and electron donor, respectively.

199 Van den Brand et al. (2015) analyzed important parameters, such as pH, organic
200 substrates, COD/SO_4^{2-} ratio, substrate composition, SO_4^{2-}, salt, temperature and DO. They

found that the presence of SRB reduced pathogens, heavy metals and sludge produced. Sulfate reduction, autotrophic denitrification and nitrification integrated (SANI) was identified as a process combining the advantages of SRB and S-dependent autotrophic denitrification. However, they indicated that in order to ensure the benefits of using SRB, a sufficient SO_4^{2-} concentration in the influent wastewater would be required to maintain the COD/ SO_4^{2-} ratio below 0.67.

3.3. Sulfammox

Sulfammox is a new process that has been addressed in the literature, especially review papers, only very recently. Sulfammox has mainly been carried out in an upflow anaerobic sludge bed reactor and circulating flow reactor (Table 3). The obtained SO_4^{2-} removal efficiencies are normally much lower compared to heterotrophic sulfate reduction. However, sulfammox is an important process linking the N and S cycles, therefore the effect of sulfammox on the overall reduction of SO_4^{2-} and NH_4^+-N should not be neglected. In the studied systems, the typical influent concentrations of SO_4^{2-} ranged from 80 to 360 mg/L (Qin et al., 2021, Zhang et al., 2019b) and the highest obtained SO_4^{2-} removal efficiency was 45% (Zhang et al., 2019a). A detailed description of the research can be found in the SI (S3).

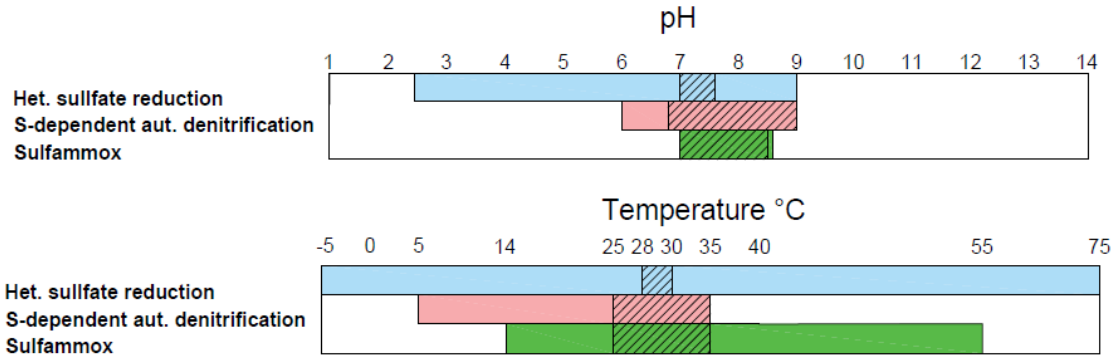
Liu et al. (2021) summarized the current understanding of sulfammox, including the mechanisms, responsible microorganisms and factors influencing the process. It was emphasized that the understanding of sulfammox has improved significantly in recent years, but more attention should be paid to recognizing the microbial community and its metabolic pathways. In addition, a variety of sulfammox end products were described that could be substrates for various N and S (anammox, S-dependent autotrophic denitrification) processes and coexist together in wastewater treatment systems. However, a challenge for the process is to ensure optimal environmental factors, such as temperature, pH, DO, for its practical

applications. It was also emphasized tha t residual organic carbon could have a significant positive effect on sulfammox, but this requires further research. A significant limitation of sulfammox is that the process was mostly investigated under laboratory scale. Practical applications should focus on implementations at low temperatures in full-size reactors.

In order to increase the efficiency of S removal in the sulfammox process, it is important to maintain the optimal pH of 8.5 and temperature of 30°C (Cai et al., 2010). The N/S ratio is also an important factor affecting that efficiency. When increasing the influent NH_4^+-N concentration from 166-666 mg N/L to 1000-2000 mg N/L, then the SO_4^{2-} removal efficiency increased from 64% to 71%. However, after increasing the influent NH_4^+-N concentration further to >3000 mg/L, the SO_4^{2-} reduction efficiency decreased to 28% (Wang et al., 2017). Also, reducing the concentration of SO_4^{2-} from 223 to 154 mg/L had a positive effect on the removal of SO_4^{2-} in the sulfammox process (Zhang et al., 2020). The N/S ratio also influenced the SO_4^{2-} removal efficiency, as the SO_4^{2-} removal efficiency at N/S = 2:1 and 4:1 was 38.8% and 30.5%, respectively (Zhang et al., 2019a).

3.4. Optimal conditions for S-dependent autotrophic denitrification, heterotrophic sulfate reduction and the sulfammox process

Figure 2 shows a summary of the reported pH and temperature ranges and their optimal values for the three S-dependent processes. The overall optimum conditions are explicitly determined by the narrowest ranges for heterotrophic sulfate reduction, which are 7-7.6 and 28-30°C for pH and temperature, respectively. The processes of S-dependent autotrophic denitrification, heterotrophic sulfate reduction and sulfammox can occur simultaneously with deammonification or its component processes, i.e. partial nitritation and anammox.



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248

249 Figure 2. Ranges of pH and temperatures and their optimal values ("[▨]" – optimum

250 conditions) reported in literature for the S-dependent processes

251

252 For comparison, for partial nitritation, the optimal ranges were 25-35°C for

253 temperature (Zhu et al. 2008, Kanders et al., 2014) and 7-8.6 for pH, with the optimal value of

254 8 (Jaroszyński et al., 2011). On the contrary, too low temperatures (10 - 15°C) cause the

255 excessive activity of NOB (Kouba et al. 2017), which can grow faster than AOB under such

256 conditions (Hellinga et al. 1998). The optimal pH range for NOB is 6 - 7.5, with the

257 maximum at 7 (Yin et al., 2016). For the anammox process, the optimal temperature and pH

258 is respectively 35-40°C (Dosta et al., 2008) and 6.7-8.3 (Jetten et al., 2001). The

259 recommended ranges for efficient deammonification are as follows: T = 20 - 35°C (Kanders

260 et al. 2014) and pH of 7.5-8 (Oshiki et al., 2011).

261 When coupling sulfammox with S-dependent autotrophic denitrification and heterotrophic

262 SO_4^{2-} reduction to increase the efficiency of S removal, it is important to keep the optimal

263 temperature of 28-30°C and pH of 7-7.6. The N/S ratio should be adjusted based on the

264 stoichiometry of all the processes involved, so that products of one process can be the

265 substrates for another process. Deviations from the optimal ratio can cause either production

of unwanted residues or bacterial competition for the substrates. SRB can compete with sulfammox bacteria for SO_4^{2-}. Moreover, heterotrophic SO_4^{2-} reduction and sulfammox contribute to formation of S^{2-} and/or S^0, which is the substrate for S-dependent autotrophic denitrification. Too intensive production of S^{2-} may lead to the persistence of this toxic compound in the effluent. The presence of carbon in heterotrophic SO_4^{2-} reduction may also contribute to the development of heterotrophic bacteria responsible for heterotrophic denitrification. Then NO_3^--N and/or NO_2^--N may become limited due to their use in both autotrophic and heterotrophic denitrification. In such a case, it is recommended to use full or partial nitrification to produce NO_3^--N and/or NO_2^--N. The competition and interactions of microorganisms participating in the aforementioned processes are shown in Figure 1.

4. Wastewater treatment systems integrating the N-S-C cycles

4.1. Systems integrating the sulfur cycle with nitrification-denitrification - Sulfate reduction, Autotrophic denitrification and Nitrification Integrated (SANI) and its modifications

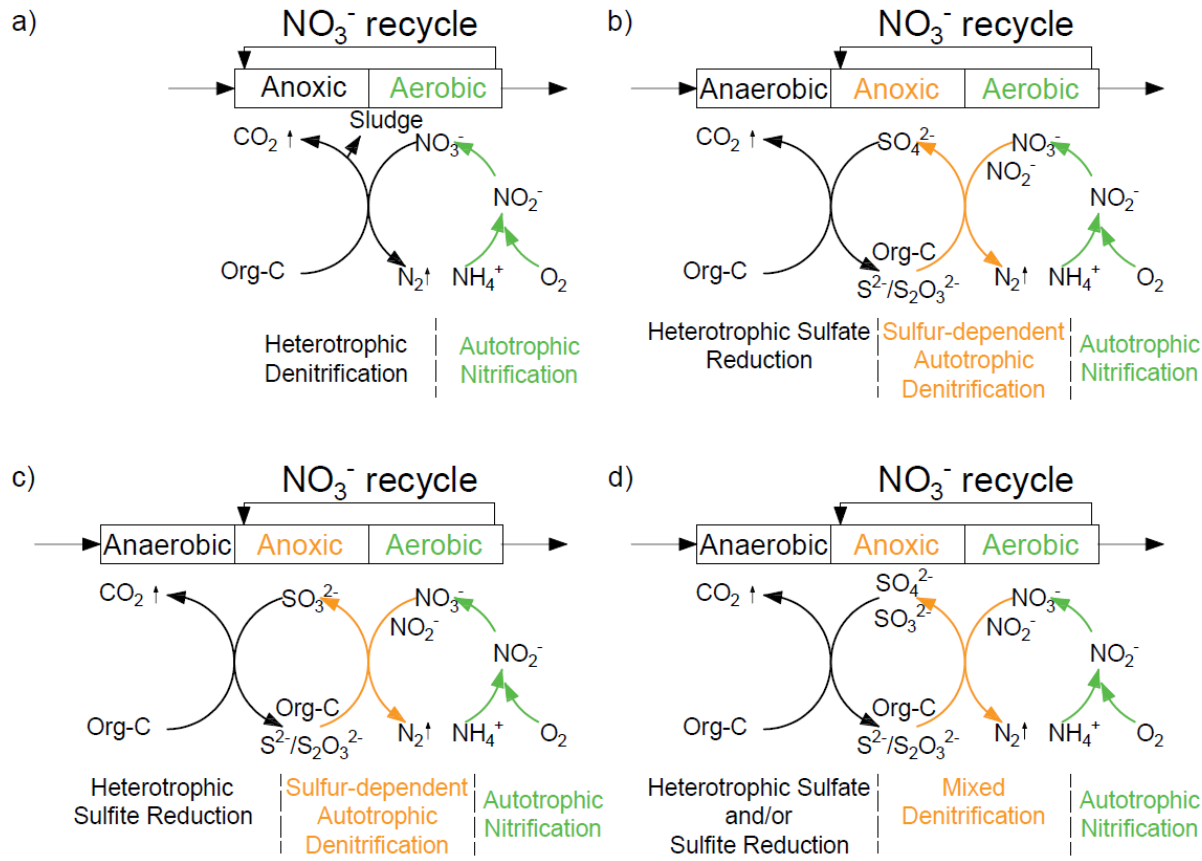
Biological SO_4^{2-} reduction along with biological oxidation of S in the form of SO_3^{2-}, S^0 or $S_2O_3^{2-}$ are two main pathways responsible for S conversions in wastewater treatment systems (Cardoso et al., 2006). An integrated process for SO_4^{2-} reduction, autotrophic denitrification and nitrification (SANI) was aimed to primarily remove organic compounds and N (Wang et al., 2009b). This process was originally developed for saline wastewater in Hong Kong and demonstrated there in full-scale (Wu et al., 2016, Wang et al., 2009b).

With that innovative approach, the conventional wastewater treatment, incorporating C and N cycles, can be extended with the S cycle, as shown in Figure 3. In the first anaerobic zone, COD is removed by SRB, which results in SO_4^{2-} reduction to S^{2-}. In the second anoxic zone, autotrophic reduction of NO_3^--N occurs with dissolved S^{2-} formed in the first zone. In the third aerobic zone, NH_4^+-N is oxidized to NO_3^--N, which is then recirculated to the second anoxic zone (Wang et al., 2009b). The SANI process and its modifications combine the

advantages of energy saving, reduced sludge production and smaller footprint. Wang et al. (2009b) noted that the total cost reduction for SANI would be >50% for a WWTP with an influent flow rate of 10,000 m^3/d.

The SANI process can be used for treatment of SO_4^{2-}-poor wastewater provided that low-cost and S-rich sources are available. For example, wet flue gas desulfurisation (FGD) systems used in boilers, coal-fired furnaces and power plants, can be reduced to alkaline flue gas sorption for production of liquid waste containing SO_4^{2-} and SO_3^{2-} (Srivastava and Jozewicz, 2001). Such a waste stream can be co-treated in the main wastewater stream in wet FGD-SANI after removing suspended solids and heavy metals (Qian et al., 2013).

The Mixed Denitrification (MD) - SANI process has also been proposed (Qian 2015a,b,c). MD-SANI generates $S_2O_3^{2-}$, S^{2-}, and some volatile fatty acids (VFA), which are subsequently converted in both heterotrophic denitrification (VFA) and autotrophic denitrification (S^{2-} and $S_2O_3^{2-}$) (Qian et al., 2015a). It should be noted that the latter process is induced faster by $S_2O_3^{2-}$ than S^{2-} (Cardoso et al., 2006). Figure 3b-d shows the SANI, FGD-SANI and MD-SANI processes depending on the available substrates.



306

307 Figure 3. Biological wastewater treatment systems using a) conventional heterotrophic

308 denitrification with autotrophic nitrification b) SANI c) FGD-SANI d) MD-SANI

309

310 ***4.2. Systems integrating the S cycle with anammox-based nitrogen removal processes***

311 In recent years, the growing attention has been paid to N removal using the anammox process.

312 The anammox process completely eliminates the need for organic C source, reduces the

313 amount of sludge produced by 80% and related energy costs for aeration by 60% compared to

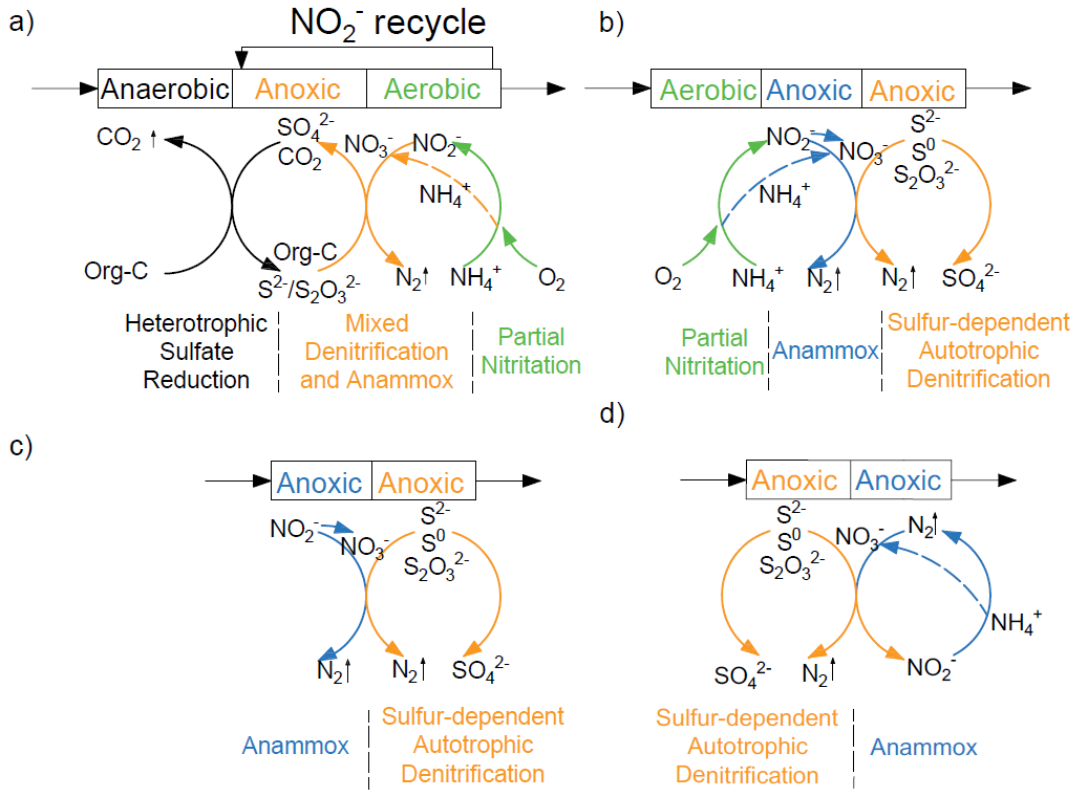
314 conventional nitrification/denitrification. The anammox process also has economic

315 advantages in the context of co-treatment of wastewater containing S compounds, especially

316 S^{2-} (Kosugi et al., 2019).

317 The anammox-based systems for combined N and S removal comprise (1) Sulfate
318 Reduction, Denitrification/Anammox and Partial Nitrification (SRDAPN), (2) Partial
319 Nitrification/Anammox and S-dependent autotrophic Denitrification (PNASD), (3) Anammox
320 and S-dependent autotrophic Denitrification (ASD), and (4) S-dependent autotrophic Partial
321 Denitrification/Anammox (SPDA).

322 The SRDAPN process is similar to the SANI process, but enhanced with anammox
323 (Figure 4a). As a consequence, instead of full nitrification, only PN is needed to produce NO_2^-
324 -N (Kosugi et al., 2019).



325

326 Figure 4. Wastewater treatment systems using the anammox process a) SRDAPN b) PNASD
327 c) ASD d) SPDA

328

329 The PNASD process uses PN/A to remove NH_4^+-N under aerobic (PN) – anoxic
330 (anammox) conditions. With S-dependent autotrophic denitrification, the produced NO_3^--N
331 can further be reduced to N_2, as shown in Figure 4b. The PNASD process has been
332 implemented as both two-stage (Dasgupta et al., 2017) and one-stage system (Yuan et al.,
333 2020).
334 The PNASD system can also be limited to an ASD system that ignores the share of
335 PN, as shown in Figure 4c. Then the NO_2^--N acceptor for anammox is not obtained from the
336 conversion of NH_4^+-N, but supplied from external sources. Accordingly, the costs of energy
337 used to produce NO_2^--N by AOB in PN are neglected, but the costs of process substrates
338 increase. The residual NO_3^--N from anammox can be removed along with S compounds (S^{2-},
339 S^0, $S_2O_3^{2-}$) by S-dependent autotrophic denitrification. The ASD process has been
340 implemented in both one-stage (Guo et al., 2016) and two-stage (Sun et al., 2018) systems.
341 If NO_2^--N can be obtained by partial autotrophic denitrification of NO_3^--N with
342 oxidation of S compounds (S^{2-}, S^0, $S_2O_3^{2-}$), then it can be used as a substrate in the anammox
343 process. Liu et al. (2017) and Wu et al. (2019) used a UASB reactor to perform S-dependent
344 denitrification with S^{2-} (Liu et al. 2017) and $S_2O_3^{2-}$ (Wu et al., 2019) for NH_4^+-N removal
345 from wastewater, as shown in Figure 4d.

346 ***4.3. Systems including the sulfammox process***

347 Both sulfammox and anammox incorporate "anaerobic" oxidation of NH_4^+-N. The
348 coexistence of both processes was found in marine sediments (Rios-Del Toro et al., 2018) and
349 anaerobic sludge (Rikmann et al., 2016). In conventional sulfammox, SO_4^{2-} is an electron
350 acceptor, which is reduced to S^0 or S^{2-}, while NH_4^+-N is oxidized to N_2, NO_2^--N and/or NO_3^--
351 N. Sulfammox may occur on its own, as shown in Figure 5a. Alternatively, the formed NO_2^--

N may be used as an electron acceptor for anammox in the combined Sulfammox/Anammox (SA) system (Figure 5b).

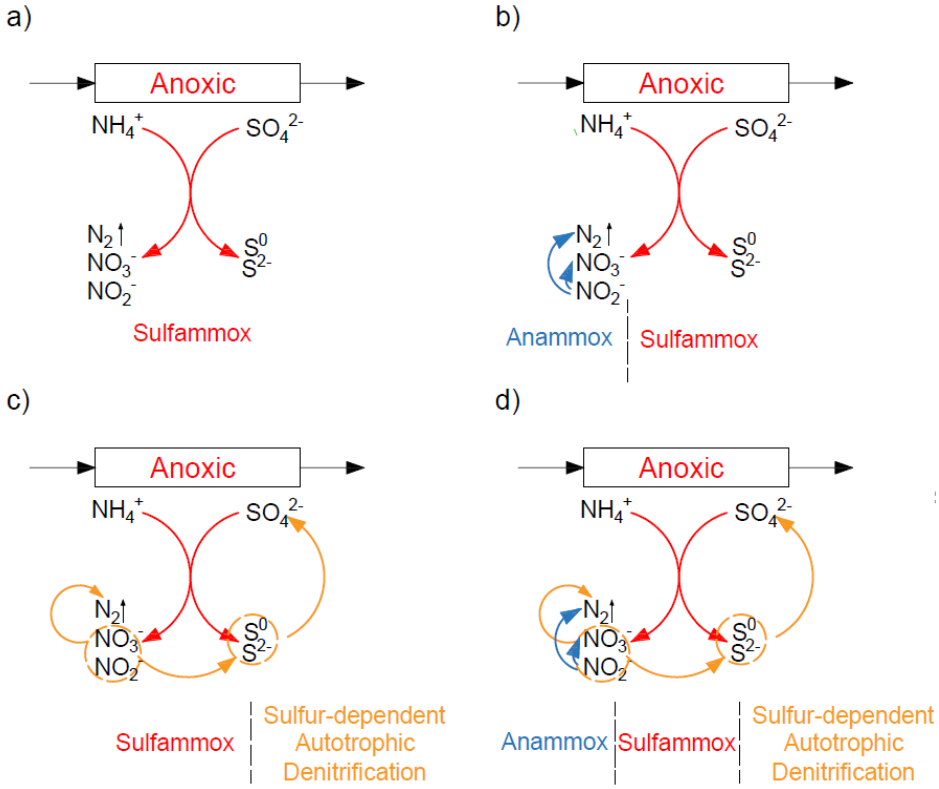


Figure 5. Wastewater treatment systems incorporating the sulfammox process a) Sulfammox b) SA c) SSD d) SASD

As NO_2^--N and NO_3^--N are generated in sulfammox, the process can be combined with autotrophic S-dependent denitrification in an Sulfammox - S-dependent autotrophic Denitrification (SSD) system, as shown in Figure 5c (Liu et al., 2021, Grubba et al., 2021). The formed S^0 and S^{2-} in sulfammox can be oxidized again to SO_4^{2-}, while NO_X-N are reduced to N_2. The SSD system can be expanded with anammox in SASD (Sulfammox – Anammox - S-dependent autotrophic denitrification), as shown in Figure 5d. In this case,

364 NO_2^--N can be reduced by both AAOB and autotrophic denitrifiers (Liu et al., 2021, Grubba
365 et al., 2021).

366 **5. Operational conditions and performances of the systems integrating the N-S-C**
367 **cycles**

368 The biochemical processes associated with the C, N and S conversions and the
369 microorganisms responsible for those conversions can be found in the SI (Figure S2).

370 ***5.1. SANI, FGD-SANI, MD-SANI***

371 The S cycle, which is part of the SANI process, ensures a more efficient use of electrons (Wu
372 et al., 2020) and eliminates the production of toxic S^{2-} (Qian et al., 2015c). In addition, it
373 reduces sludge production by 90% compared to the conventional biological N removal
374 processes. This is possible due to very low yield coefficients of the microorganisms
375 responsible for SO_4^{2-} reduction, autotrophic denitrification and nitrification, i.e., 0.02 kg
376 VSS/kg COD, 0.01 kg VSS/kg NO_3^--N and 0.07 kg VSS/kg NH_4^+-N, respectively (Lu et al.,
377 2011, Wang et al., 2009b). In addition, there are other significant reductions, including energy
378 consumption by 35% (Lu et al., 2011), greenhouse gas emission (GHG) by 36% (Lu et al.,
379 2011), and the space required for the process of wastewater treatment and sludge handling by
380 30%–40% (Liu et al., 2016).

381 As shown in Table 4, SANI shows a relatively high level of performance compared to
382 the conventional systems. The efficiencies of SO_4^{2-}, total nitrogen (TN) and COD removal
383 vary in the ranges of 72-98%, 55-74% and 82-97%, respectively (Hao et al., 2015, Lu et al.,
384 2009). The SANI modifications (FGD-SANI and MD-SANI), which use wastewater streams
385 from wet flue gas desulphurization, reveal even a greater performance potential (Qian et al.,
386 2015a, b, Jiang et al., 2013). The biological reduction of SO_3^{2-} in FGD-SANI and MD-SANI
387 provides more energy for bacterial growth, which is associated with a higher sludge efficiency

compared to the biological reduction of SO_4^{2-} (Jiang et al., 2013). Moreover, SO_3^{2-} is an intermediate in SO_4^{2-} reduction, which may result in faster reduction by SRB.

Jiang et al. (2013) found that the removal rates of specific organics in the SO_3^{2-} and SO_4^{2-} reducing reactors were similar. At the extremely low temperatures (<10°C), incomplete reduction of SO_3^{2-} in an anaerobic reactor (Figure 3c) resulted in accumulation of $S_2O_3^{2-}$ and reduction in the removal rate of organics. However, the anoxic and aerobic reactors (Figure 3c) still provided a high removal efficiency of organics (>94%), while NH_4^+-N and NO_3^--N were almost completely removed.

The MD reaction can lead to a much higher reduction of NO_3^--N and NO_2^--N compared to the S^{2-} based SANI process (Qian et al., 2015a). Qian et al. (2015b) reported that the denitrification rate increased sevenfold in MD-SANI compared to SANI. Furthermore, in comparison with SANI, FGD-SANI shows higher TN and COD removal efficiencies (98% and 94%). The complete removal of SO_3^{2-} and TN was achieved in MD-SANI, while the COD removal efficiency in that process was 81% (Table 4).

5.2. SRDAPN and PNASD - challenges resulting from the combination of aerobic and anaerobic conditions

The presence of S^{2-} in the influent wastewater imposes a significant risk of inhibition of the AAOB responsible for anammox. Threshold levels of S^{2-} inhibiting AAOB were found in the range of <1 to 64 mg S/l (Jin et al., 2013, Carvajal-Arroyo et al., 2013, Dapena-Mora et al., 2007). The study by Wiśniewski et al. (2019) determined the half maximal inhibitory concentration (IC_{50}) under two different S^{2-} conditions. The IC_{50} was 4.25 mg H_2S-S/L at a constant S^{2-} concentration of 11 mg TS-S/L and pH in the range 7-7.9 vs. 4.67 mg H_2S-S/L at a varying concentration of S^{2-} ranging from 1 to 15 mg TS-S/L and a constant pH of 7. The decrease in AAOB activity was due to the pH-dependent non-ionized form of H_2S. In

addition, heterotrophic bacteria may coexistence with AAOB but also outcompete AAOB at high influent C/N ratios (Chamchoi et al., 2008).

The PNASD process has been implemented in both one- and two-stage systems. The two stage-systems are easier to maintain and allow to avoid the negative impact of S^{2-} on AAOB and the competition between AOB and SOB for DO (Sahinkaya and Kilic, 2014a).

Zhang et al. (2020) used S^0 for denitrification and observed only a small effect, when DO was kept at the level of 0.4-0.8 mg/L. When the DO concentration increased to 1.2 mg O_2/L, the concentrations of NO_3^--N and SO_4^{2-} also increased. This indicates excessive oxidation of S^{2-} or its reduced compounds in aerobic systems. Under non-limited DO conditions, autotrophic SOB can readily utilize oxygen, which leads to accumulation of SO_4^{2-}. On the other hand, too low DO concentrations in the PNASD process can reduce the NO_2^--N production rate in PN.

5.3. ASD, SPDA and sulfammox systems - coexistence of AAOB and denitrifiers

Under anaerobic conditions, the combination of anammox process and S-dependent autotrophic denitrification can work with high removal efficiencies of TN (88-96%) and S (90-100%) (Table 4). AAOB and T. *denitrificans* can assist in the combined N and S removal without inhibition by S^{2-} (Guo et al., 2016). In that study, most of S^{2-} was oxidized to S^0 at the influent ratios of NH_4^+-N/S^{2-} and NO_2^--N/S^{2-} at 1.74 and 2.2-2.27, respectively. Two S forms can accumulate depending on the S/N ratio in the reactor, i.e., SO_4^{2-} (at S/N ratio <1) or S^0 (at S/N ratio >1) (Cardoso et al., 2006).

When NO_2^--N is fed to the anammox process, S-dependent autotrophic denitrification may occur. When both NO_2^--N (anammox substrate) and NO_3^--N (anammox product) are simultaneously present in the influent, the latter form is the preferred electron acceptor for

denitrification (Guo et al., 2016). However, a small portion of NO_2^--N can also be used by T. *denitrificans* and increase the overall efficiency of N and S removal.

Instead of complete denitrification, partial reduction to NO_2^--N can be achieved. This approach is advantageous for the Partial Denitrification/Anammox (PD/A) systems by continuously producing NO_2^--N for anammox (Wu et al., 2019). In addition, the consumption of electron donors can be reduced in comparison with the conventional biological nitrogen removal processes. The reported TN removal efficiencies exceeded 90% in SPDA (Table 4).

The novel sulfammox process has been applied in SO_4^{2-} and NH_4^+-N-rich wastewater treatment systems. One of the intermediates in the sulfammox reaction is NO_2^--N, which can be used by either AAOB or S-dependent autotrophic denitrification along with the residual NO_3^--N from anammox. Wu et al. (2020) combined sulfammox and anammox and obtained high removal efficiencies of NH_4^+-N (98.5%) and SO_4^{2-} (53%). Furthermore, the sulfammox and anammox processes can also be combined with S-dependent autotrophic denitrification (Rios-Del Toro et al., 2018).

6. Modeling N, S and C conversions in wastewater treatment systems

Modeling has been proven to be an effective tool to understand complex, interrelated N, S and C transformations (Show et al., 2013). In principle, two modeling approaches are possible, including empirical models, such as artificial neural networks (ANNs), and mechanistic models based on the Activated Sludge Model (ASM) family.

6.1. Artificial neural networks (ANNs)

The ANN model does not require a detailed process description, and it can be established by simple input and output parameters. Therefore, the ANN has been known for a long time as a tool in setting control mechanisms and performance models of biological wastewater treatment processes (Choi and Park, 2001). Wang et al (2009a) developed an ANN model to

monitor a denitrifying S^{2-} removal (DSR) process. The proposed model revealed that the comparative influences of four input factors on DSR performance were as follows: hydraulic retention time (HRT) > S^{2-} concentration > C/S ratio > N/S ratio. Even though the ANN model is capable of predicting an intricate function between input and output parameters, it cannot help in understanding mechanisms of the complex biochemical processes.

6.2. Mechanistic models

The International Water Association Activated Sludge Models (ASMs) No. 1, 2, 2d and 3 (Henze et al., 2000) describe conversions of organic C and N compounds (ASM1 and ASM3), and additionally P compounds (ASM2 and ASM2d). However, to simplify the model structure, all the ASMs only considered NO_3^--N reduction as a one-step heterotrophic process using readily biodegradable organic compounds as electron donors. Moreover, one-step NH_4^+-N oxidation to NO_3^--N was the only autotrophic N transformation.

S-dependent autotrophic denitrification and the synergistic and competitive relationships among microorganisms were subsequently integrated with the ASMs. On one hand, developing realistic models is essential for practical applications in simultaneous N, C and S removal systems. On the other hand, due to the complex interactions between autotrophic and heterotrophic denitrifiers, developing an exhaustive model and appropriate control strategy becomes challenging. The existing models (Table 5) have been used in bench-scale reactors to predict the process involving intricate metabolic pathways with synthetic substrates. However, further work is still necessary to confirm the models in practical applications with real wastewater.

A detailed description of the mechanistic models can be found in SI (S4).

7. Implications of combining the N, S and C cycles in wastewater treatment systems

7.1. Processes application opportunities

S^0 and S^{2-} are considered good alternatives to organic matter in the denitrification process due to the absence of organic residues in the treated wastewater. It is thus strongly recommended to use S-dependent autotrophic denitrification instead of heterotrophic denitrification, especially for wastewater with a low organic content. Attention should also be paid to the water-insoluble S^0, which can physically be removed from wastewater and reused for production of sulfuric acid, pesticides, fertilizers, in construction (Lin et al., 2018). It is economic, effective and readily available source of electrons. On the other hand, $S_2O_3^{2-}$ is readily bioavailable and may mediate a higher rate of denitrification compared to S^0 and H_2S. S^{2-} is often used in municipal and industrial areas requiring desulphurization. Depending on the local conditions, S-dependent autotrophic denitrification can occur with a wide spectrum of S compounds. Moreover, it can get them from the initial SO_4^{2-} reduction stage in the integrated systems combining N-S-C cycles.

Biological SRB-based methods are a sustainable way of treating AMD compared to physico-chemical methods (Sinharoy et al., 2020b). SRB are capable of using toxic metals in their metabolism, thus reducing environmental and human health problems. SRB can grow in a wide range of environmental conditions, which provides many opportunities for the development of technologies based on their metabolism, with SO_4^{2-} reduction being recognized as a key step in all S- dependent processes (Hao et al., 2014).

Among the various gaseous substrates for SO_4^{2-} reduction, H_2 is most energetic for SRB. The resources that can be recovered from this process are metal sulfides and S^0, which has also been identified by Kumar and Pakshirajan (2020) as a potential substrate for S-dependent autotrophic denitrification.

The combination of the N, S and C cycles could lead to the development of economically feasible and sustainable wastewater treatment systems that produce less sludge and reduce carbon footprint compared to the existing systems. The SANI process has already been used in several full-scale wastewater treatment installations in Hong Kong due to the practice of flushing toilets with seawater (Jiang et al., 2013). The process can also be applied to freshwater wastewater, even in cold inland areas that do not contain enough SO_4^{2-} or SO_3^{2-} rich wet flue gas desulphurization (Qian et al., 2015a, b, Jiang et al., 2013). It can also be adapted to treat industrial wastewater by adding SO_4^{2-}, seawater or some SO_4^{2-}-rich wastewater. Lu et al. (2009, 2012) suggested that the SANI process could be a good solution in densely populated cities to treat saline wastewater as an economic source in terms of water scarcity and wastewater treatment in water-poor coastal areas.

Other technologies that include anammox and SANI processes have discovered the advantages of AAOB coexisting with SRB, SOB, and AOB. In addition, compared to the SANI process, the combination of SO_4^{2-} reduction, denitrification/anammox and partial nitrification will further reduce aeration energy consumption due to the lack of full nitrification required for NO_3-N production. The presence of anammox in the SRDAPN process resulted in an increased NO_2^--N removal efficiency by over 30% (Kosugi et al., 2019).

For wastewater with a low organic content, PNASD can be considered a viable option. The two-step PNASD system was more efficient for N and S removal, and easier to maintain than the one-step system (where bacteria competed for DO) (Dasgupta et al., 2017). Moreover, it has also been proven that the process can be applied in a single reactor under mainstream conditions (Yuan et al., 2020).

Instead of combining the heterotrophic SO_4^{2-} reduction with anammox, sulfammox can replace or accompany both processes by using a SO_4^{2-} dependent AAOB. Recent studies

have proposed the use of sulfammox based on the combined reduction of NH_4^+-N and SO_4^{2-}. If SO_4^{2-} was reduced to S^{2-} or S^0 with organic compounds, this process would be replaced with sulfammox, while eliminating the addition of external carbon. Another suggested solution is to combine the sulfammox process with heterotrophic SO_4^{2-} reduction in order to increase the reduction rate of SO_4^{2-}. Moreover, if sulfammox is used upstream of an S-dependent autotrophic denitrification reactor, it contributes to oxidation of NH_4^+-N to N_2 (which increases the overall efficiency of NH_4^+-N removal) or NO_2^--N and NO_3^--N (which can be used in S- dependent autotrophic denitrification). By combining sulfammox and anammox, the efficiency of NH_4^+-N removal and SO_4^{2-} reduction to S^0 can be simultaneously increased (Liu et al., 2021, Grubba et al., 2021).

7.2. Advantages and disadvantages of two cycles or three cycles in wastewater treatment

The advantages and disadvantages of the systems based on the N-S-C cycles and their coupling are summarized below.

Advantages:

1. Approximately 35% reduction in energy consumption and up to 90% reduction in sludge production compared to full nitrification-denitrification.
2. Reduction or even no external carbon dosing for S-dependent autotrophic denitrification.
3. For the combined processes, almost complete N and S^{2-} removal and up to 75% efficiency of SO_4^{2-} removal.
4. Products of one process used as the substrates for another process.
5. When replacing heterotrophic denitrification with S-dependent autotrophic denitrification, carbon consumption is reduced by 100%. If heterotrophic SO_4^{2-} reduction is replaced by sulfammox, carbon consumption is also reduced by 100%.
6. Removal of a few harmful compounds (NH_4^+, NO_2^-,SO_4^{2-}, S^{2-}) in one system.
7. Approximately 30–40% reduction of volumes required for wastewater and sludge treatment processes.
8. Reduction of GHG emissions by 36% compared to conventional nitrification-denitrification.

Disadvantages:

1. Limited use in cold regions due to the high optimal temperature range (28-30°C).

2. Complex interactions and competition for substrates between the functional microorganisms.

3. Greater complexity of the systems potentially resulting in higher investment costs.

4. The operating conditions must be compatible with all the N-S-C processes.

5. Some substrates/products involved in one process may be inhibitors for other processes, e.g. S^{2-}.

7.3. Processes application limitations

One of the most important limitations of technologies combining N, S and C cycle processes is the narrow optimal range of temperature (28-30°C) and pH (7-7.6). Thus, cold weather in inland areas also restricts the use of coupled systems.

An important factor that should be considered when implementing technologies containing the S-dependent autotrophic denitrification process is the inhibition of this process caused by S^{2-} (Cardoso et al., 2006) as well as NO_2^--N, NO_3^--N and free nitric acid (FNA) (Cui et al., 2019). Even though S^0 is an inexpensive and non-toxic electron donor, but it provides a low denitrification rate due to its low solubility. The use of smaller S granules with a larger surface area improves the reaction efficiency, however it can cause low porosity and clogging and fouling of the reactors due to small S grain size or cracking (Wu et al., 2021). Moreover, as S^0 and H_2S reveal a much lower rate of NO_3^--N reduction, mainly the use of $S_2O_3^{2-}$ is recommended in the process. However, its natural content of wastewater is rather limited due to its instability (Cui et al., 2019).

In the case of heterotrophic SO_4^{2-} reduction, the presence of DO, NO_3^--N and NO_2^--N inhibits reduction of SO_4^{2-} and enhances oxidation of S^{2-} to S^0 or SO_4^{2-} (Mohanakrishnan et al., 2009). Moreover, the activity of SRB is inhibited by heavy metals, including Pb and Cd

(Sinharoy and Pakshirajan, 2019b). The toxicity of heavy metals depends mainly on the type of metal, responsible microorganisms, presence of other pollutants, and process conditions (Mal et al., 2016). Therefore, the systems based on heterotrophic SO_4^{2-} reduction cannot be used for wastewater rich in heavy metals. Moreover, a significant limitation is the limited number of microorganisms that are able to carry out SO_4^{2-} reduction with the use of gaseous substrates. Moreover, the low gas-liquid mass transfer also makes it difficult to scale-up the process.

A significant limitation in the implementation of integrated systems connecting N-S-C cycles is also the insufficient knowledge about the mechanism of sulfammox and responsible microorganisms. Until now, there has been no genomic evidence to support the ability of AAOB to use SO_4^{2-} as an electron acceptor. The growth rate of potential functional bacteria is also low, which limits their unambiguous identification (Liu et al., 2021). Moreover, the organic matter present in the wastewater stimulates the survival of heterotrophic bacteria, including denitrifiers. This leads to a competition between these bacteria and the sulfammox bacteria, thus destroying the sulfammox process.

7.4. Processes application challenges

Using specific N, S and C removal processes independently of each other is much easier to maintain than the processes combining these cycles. To link those processes in the combined technologies as presented in this review, it is important to recognize the effects of S^{2-} on N removal processes, such as autotrophic/heterotrophic denitrification and anammox, as well as the competition between AOB and SOB for DO. S^{2-} and organic matter, which are fed to an anaerobic compartment, can inhibit AAOB in anammox-coupled systems (Kosugi et al., 2019). Chen et al. (2018) showed that DO can react with S^{2-} while reducing the NO_3^--N removal rate. In addition, S^{2-} was reduced to S^0 and then converted to SO_4^{2-g} due to the

presence of DO. These findings highlight the challenges faced by single-stage integrated systems.

In order to avoid the inhibition of SO_4^{2-} reduction by heavy metals, it is recommended to use an upstream reactor in order to remove metals from AMD using S^{2-}. In order to use SO_4^{2-} reduction coupling systems, it is also necessary to consider selection of the appropriate type of reactor, use of resistant microorganisms, and presence of other pollutants. Designing novel reactor configurations with high gas-liquid mass transfer can also help in applying the process in full scale. Moreover, instead of obtaining pure gases, a cost-effective solution would be production of gaseous substrates by thermochemical or biochemical methods from various compounds (e.g. waste) (Sinharoy et al., 2020b).

In the case of sulfammox, more research is needed to identify potential applications and integration with other systems. The key enzymes involved in the metabolism of NH_4^+-N and SO_4^{2-} should also be investigated. For this purpose, it is important to develop appropriate reactor configurations and create operational conditions that can enrich functional bacteria and allow for simultaneous removal of NH_4^+-N and SO_4^{2-}. Under non-limited NO_3^--N conditions, the SO_4^{2-} concentration may increase due to S-dependent autotrophic denitrification. The role of organic matter also requires further investigation with regard to the existence of the sulfammox process.

The combination of anammox, S-dependent autotrophic denitrification and sulfammox processes is challenging due to the different requirements of the microorganisms responsible for each process. The S-dependent autotrophic denitrification process may result in the production of SO_4^{2-} from S^{2-} or S^0, which negatively affects sulfammox, where SO_4^{2-} must be reduced to S^0 (Liu et al., 2021). More focused research on the coexistence of sulfammox with other bacteria and the development of a mechanistic model are needed to better understand and predict N and S dynamics. Moreover, the S/N ratio also plays an important role in

634 determining the S-dependent autotrophic denitrification end products, requiring a closer look
635 at the N and S dynamics. On the other hand, in order to avoid fouling and clogging of the
636 reactors due to the presence of S^0, it is important to search for the appropriate sulfur grain
637 size.

638 Wang et al. (2009b) identified three main challenges for the SANI process. First of all,
639 it is the low efficiency of both SO_4^{2-} reduction during heterotrophic and S-dependent
640 autotrophic denitrification reduction. Secondly, high concentrations of SO_4^{2-} are required,
641 which may increase residual S^{2-} in the treated wastewater. Thirdly, transfer of NO_3^--N from
642 the nitrification reactor to the S-dependent autotrophic denitrification reactor can also be
643 difficult.

644 **8. Conclusions**

645 In terms of sustainability, the combination of N-S-C cycles processes has a few important
646 benefits, including energy savings and lower sludge production. The combined processes
647 allow for almost complete N and S^{2-} removal, while the efficiency of SO_4^{2-} removal can reach
648 up to 75%.

649 Among all the processes linking the N-S-C cycles, SANI has been best recognized,
650 but is rather not applicable in the case of wastewater with low organic content. Instead, it is
651 worth of considering the sulfammox process that can reduce SO_4^{2-} and increase NH_4^+-N
652 removal rate under anoxic conditions without the addition of external carbon.

653 Practical applications of the reviewed systems still face many challenges, especially in
654 the single-stage configurations. In particular, the coexistence of several bacterial groups
655 (AOB, AAOB, sulfammox bacteria, SOB, SRB) and their competition for the substrates is a
656 key issue to be considered. Moreover, practical applications of the coupled S and N/C cycles
657 require realistic models. However, due to the complex interactions between autotrophic and

heterotrophic denitrifiers, development of a mechanistic model and appropriate control strategy becomes challenging.

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Table 1. Key issues and challenges of S-dependent autotrophic denitrification, heterotrophic sulfate reduction and sulfammox

Topic	Process		
	S-dependent autotrophic denitrification	**Heterotrophic sulfate reduction**	**Sulfammox**
Key issues	consists of oxidation of S compounds, including S^{2-}, S^0, thiosulfate ($S_2O_3^{2-}$) and sulfite (SO_3^{2-}), coupled with reduction of NO_3^--N and/or NO_2^--N	SO_4^{2-} reduction, which involves the use of organic electron donors or inorganic electron donors, which must be supplemented with a carbon source	NH_4^+-N is oxidized to N_2, whereas SO_4^{2-} plays the role of an electron acceptor and is reduced to S^0 under anaerobic conditions
Challenges and opportunities	a) a good alternative to heterotrophic denitrification due to the lack of carbon dosing; b) reduction of toxic S^{2-}; c) the possibility of treating wastewater poor in organic content; d) residual SO_4^{2-} in wastewater; e) a long incubation time is needed before a fully adapted culture is obtained; f) precise control strategy (from S^{2-} to S^0) and novel S^0 recovery technology at the source; g) acclimation and adjustment of microorganisms: the concentration of S^{2-} should be controlled; maintaining the denitrification efficiency of autotrophic denitrification systems at low temperatures; alkalinity and pH control is necessary to prevent the formation of NO_2^--N; influence of the N/S ratio on the reactions and bioproducts, the optimal N/S ratio = 0.5-0.9 for S oxidation and NO_3-N reduction (see Eqs. 1-8 in the SI); h) when the dissolved oxygen (DO) concentration is > 1.6 mg O_2/L, denitrification is completely inhibited	a) SO_4^{2-} reduction, especially in SO_4^{2-} rich industrial wastewater; b) use of wastewater rich in organic compounds; c) high concentrations of SO_4^{2-} inhibit SRB activity; d) elevated levels of heavy metals may reduce or terminate SRB activity	a) anaerobic oxidation of NH_4-N without carbon addition; b) SO_4^{2-} reduction in wastewater; c) knowledge of microorganisms, mechanisms and their metabolic pathway is still limited; d) temperature, DO and pH would influence its practical applications; e) inhibition of sulfammox activity due to S^{2-} accumulation; f) with a high concentration of NO_3-N, SO_4^{2-} concentration may increase due to autotrophic denitrification

Table 2. Process conditions and observed NO_3^- utilization rates during S-dependent autotrophic denitrification in different types of reactors

Reactor type	Electron donor	Temperature	pH	S-compound	Initial NO_3^-N concentration	SO_4^{2-} production	Denitrification rate	References
		(°C)	(-)	(mg S/L)	(mg N/L)	(mg S/L)	(kg N/m3/d)	
Fluidized-bed reactor	$S_2O_3^{2-}$	20-30	7	184-2260	100-1230	150-320	1.24-3.25	Zou et al.., 2016
Fluidized-bed reactor	S^0	28-30	7.2-9	na	25-75	100-600	0.07-0.2	Sahinkaya and Dursun 2015
Fluidized-bed reactor	S^0/ $S_2O_3^{2-}$	20	6.8-8.2	na	20-700	na	2.53-3.37	Kim et al.., 2004
Packed-bed reactor	S^0	28-30	6-8	na	50-75	200-600	0.07-0.1	Sahinkaya and Kilic, 2014a
Packed-bed reactor	S^0	10-26	6-8	na	30-60	191-483	0.03-0.24	Sahinkaya et al.., 2014b
Packed-bed reactor	S^0	15.2-29	6.7-8.4	592.42-5924.17	20-25	640	0.2	Kimura et al. 2002
Packed-bed reactor	S^0	20-25	8.3-8.7	na	60-251	na	0.27-0.87	Koenig and Liu 2002
Packed-bed reactor	S^0	20-25	na	na	60-400	na	0.48-0.77	Koenig and Liu 2001
Up-flow continuous reactor	S^{2-}	29-31	7	160-1000	30.4-169.6	na	0.15-0.61	Jing et al. 2010
Up-flow anaerobic sludge blanket	S^{2-}	30	7.5	0.62[a]	0.33[a]	na	0.09-0.31	Yang et al. 2016
Up-flow column reactor	S^0	30	7.3	na	20	6.15 - 7.92[b]	0.22	Zhu et al.., 2019
Vertical fixed-bed reactors	S^{2-}	30	7-7.5	49.3	20	20	na	Moraes et al. 2012

[a] $kg/m^3/d$

[b] $g/g\ N_{removal}$

Table. 3. Reactor types, operational (environmental) conditions, influent S concentrations and efficiency of heterotrophic SO_4^{2-} reduction and sulfammox

Reactor type	Electron donor	Temperature (°C)	pH	SO_4^{2-} concentration (mg/L)	SO_4^{2-} removal efficiency or rate	References
HETEROTROPHIC SULFATE REDUCTION						
Gas lift reactor	Carbon monoxide	30	7	250–1000	62.5-97.5%	Sinharoy et al.., 2020a
Moving bed biofilm reactor	Carbon monoxide	30	7	250–1000	67.1–95.2%	Sinharoy et al.., 2019a
Batch	Succinic acid, yeast extract	22	-	1700	169 mg SO_4^{2-}/L/d	Virpiranta et al.., 2019
		16			98 mg SO_4^{2-}/L/d	
		6			13-42 mg SO_4^{2-}/L/d	
Batch	Methanol	5	7	-	26.7 mg SO_4^{2-}/L/d	Nielsen et al., 2019
Packed bed reactor	Ethylene glycol	30	7	250–1000	4.1 mg SO_4^{2-}/L/d	Kumar et al.., 2018
Inverse fluidized bed reactor	Scourer	30	7	700	34 mg SO_4^{2-}/gVSS/d	Reyes-Alvarado et al.., 2018
	Cork				6.1 mg SO_4^{2-}/gVSS/d	
Packed bed reactor	Molasses	4-8	6.5-7.1	287-548.2	0-22 mg SO_4^{2-}/L/d	Nielsen et al., 2018
Batch	Crab shell	30	7	721-738	6-9 mg SO_4^{2-}/gVSS/d	Reyes-Alvarado et al.., 2017
	Potato			764-766	22-34 mg SO_4^{2-}/gVSS/d	
	Filter paper			752-823	50-65 mg SO_4^{2-}/gVSS/d	
Fluidized-bed reactor	Glycerol	23	5.5-8.5	2000-3000	167 mg SO_4^{2-}/gVSS/d	Bertolino et al.., 2014
Stirred tank reactor	Hydrogen + carbon dioxide	30	6.95-7.05	-	3400 mg SO_4^{2-}/L/d	Sáez-Navarrete et al.., 2012
Fluidized-bed reactor	Ethanol	35	7.5	-	211 mg SO_4^{2-}/gVSS/d	Nevatalo et al.., 2010
	Ethanol + lactate				2016 mg SO_4^{2-}/gVSS/d	
Gas lift reactor	Hydrogen	30-35	7-7.5	5000–30000	7080 kg SO_4^{2-}/d	Van Houten et al.., 2009
Anaerobic filter	Ethanol, spent manure	6	2.5-4.3	900	961-1345 mg SO_4^{2-}/L/d	Tsukamoto et al., 2004
	Methanol, spent manure				1057-1441 mg SO_4^{2-}/L/d	
SULFAMMOX						
Upflow anaerobic sludge bed reactor	Ammonium nitrogen	35	7.9-8.3	80	8.18 mg S/L/d	Qin et al., 2021
Circulating flow completely anaerobic reactor	Ammonium nitrogen	30	8.1-8.6	88	2-27%	Zhang et al., 2020
				223	2-27%	
				154	18-64%	
Self-designed circulating flow reactor	Ammonium nitrogen	35	8.1-8.3	183	approx. 40%	Zhang et al., 2019a
				216	approx. 0%	

				116	approx. 30%	
				100	approx. 45%	
Self-designed circulating flow reactor	Ammonium nitrogen	30	8.1-8.6	90	approx. 30%	Zhang et al., 2019b
				170	approx. 30%	
				360	approx. 5%	
Sequencing batch reactor	Ammonium nitrogen	-	-	261	19%	Prachakittikul et al., 2016
Batch	Ammonium nitrogen	30	8.5	163	40%	Cai et al., 2010
Upflow anaerobic sludge blanket reactor	Ammonium nitrogen	35	7.5–8.5	240	30%	Yang et al., 2009
Non-woven rotating biological contactor	Ammonium nitrogen	35	8–8.2	-	-	Liu et al., 2008

Table 4. Technologies for integrated S, N, COD removal and the observed removal efficiencies for S, N and COD

Process	Reactor type	S removal efficiency	N removal efficiency (N form)	COD removal efficiency	References
Sulfate reduction, Autotrophic denitrification and Nitrification Integrated (SANI)	Up-flow anaerobic sludge bed, an anoxic filter, an aerobic filter	16-68 mg S^{2-}/L	74% (TN)	95%	Wang et al.., 2009b
SANI	Up-flow sludge bed reactor, an anoxic reactor and an aerobic reactor	98 % S^{2-}	55% (TN)	87%	Lu et al.., 2012
SANI	Up-flow anaerobic sludge bed, an anoxic filterand an aerobic filter	97 % S^{2-}	74% (TN)	97%	Lu et al., 2009
SANI	Sulfate-reducing up-flow sludge bed	75% SO_4^{2-}	-	90%	Hao et al., 2013
SANI	Sulfate-reducing up-flow sludge bed	72% SO_4^{2-}	-	82%	Hao et al., 2015
Flue gas desulfurization - Sulfate reduction, Autotrophic denitrification and Nitrification Integrated (FGD-SANI)	Sulfite-reducing upflow anaerobic sludge bed	~54 % S^{2-}	~98% (TN)	94%	Jiang et al.., 2013
Mixed Denitrification - Sulfate reduction, Autotrophic denitrification and Nitrification Integrated (MD-SANI)	Sulfate/sulfite reducing up-flow sludge bed and anoxic up-flow sludge bed	-	100% (NO_3^--N)	80%	Qian et al.., 2015a
MD-SANI	Sulfur-reducing upflow sludge bed and the anoxic upflow sludge bed	~100% SO_3^{2-}	100% (TN)	81%	Qian et al.., 2015b
Sulfate reduction, denitrification/anammox and partial nitrification (SRDAPN)	Laboratory scale up-flow anaerobic-anoxic biological filter reactor	400-500 mg S^{2-}/d	79% (TN)	500-2300 mg/d	Kosugi et al., 2019
Partial Nitrification/Anammox and S-dependent autotrophic Denitrification (PNASD)	PN/A reactor and an elemental sulfur-supported packed bed autotrophic denitrification	-	97% (TN)	-	Dasgupta et al., 2017
PNASD		~100% S^{2-}	84% (TN)	-	Yuan et al.., 2020

	Single reactor under mainstream conditions				
Anammox and S-dependent autotrophic Denitrification (ASD)	Expanded granular sludge bed	90-100% $S_2O_3^{2-}$	98% (TN)	-	Sun et al.., 2018
ASD	Up-flow anaerobic sludge blanket reactor	99.6% S^{2-}, 330 mg S^{2-} /L	88% (TN), 252 mg NH_4^+-N/L	-	Guo et al.., 2016
S-dependent autotrophic Partial Denitrification and Anammox (SPDA)	Up-flow anaerobic sludge blanket reactor	~100% $S_2O_3^{2-}$	>90% (TN)	-	Wu et al., 2019
SPDA	Up-flow anaerobic sludge blanket reactor	70% S^0	90% (NO_2^--N)	-	Liu et al.., 2017
Sulfammox/Anammox (SA) with COD	Moving Bed Biofilm Reactor	10% SO_4^{2-}	30% (NH_4^+-N)	-	Rikmann et al.., 2016

Table 5. Overview of the reported mechanistic models linking C, S and N transformations

No.	Reactor type	Substrate	Influent concentrations			Model structure				References
			Organic (mg COD/L)	S^{2-} (mg S^{2-}-S/L)	NO^{3-}-N (mg -N/L)	No. of processes	No. of components	No. of parameters	S and N involved processes	
1	Bench-scale EGSB reactor	Synthetic wastewater	200-800	200-800	75-275	7	10	18	Hydrolysis: Particulate N → Organic N Ammonification: Organic N → NH_4^+-N Heterotrophic: NO_3^--N → N_2 Autotrophic: NO_3^--N → N_2	Wang et al., 2010
2	Bench-scale EGSB reactor	Synthetic wastewater	275-2300 mg C/L	156-1490	100-800	6	8	31	Autotrophic: S^{2-}→S_0 → SO_4^{2-} Autotrophic: NO_3^--N → NO_2^--N →N_2 Heterotrophic: NO_3^--N → NO_2^--N → N_2	Xu et al., 2014
3	Bench-scale SBR	Synthetic wastewater	-	194 145	321 202	4	5	9	Autotrophic: S^{2-} → S_0 → SO_4^{2-} Autotrophic: NO_3^--N → NO_2^--N → N_2	Xu et al., 2016
4	Bench-scale EGSB reactor	Synthetic wastewater	2700	1000 mg SO_4^{2-}-S/L	200-700	14	15	38	Autotrophic: S^{2-} → S_0 Autotrophic: NO_3^--N → NO_2^--N Heterotrophic: NO_3^--N → NO_2^--N → N_2 Heterotrophic: SO_4^{2-} → S^{2-}	Xu et al., 2017
5	MBfR	Anaerobic digestion liq-uor	50-100	30	50-1000	18	17	60	Autotrophic: NH_4^+-N → NO_2^--N → NO_3^--N Autotrophic: NH_4^+-N, NO_2^--N → N_2, NO_3^-N Heterotrophic: NO_3^--N → N_2 Autotrophic: S^{2-} → S^0 → SO_4^{2-} Autotrophic:CH_4 → CO_2	Chen et al., 2016
6	Coastal upwelling system	Sea water	-	0.1 mmol S/m^3	0.1 mmol N/m^3	9	14	46	Autotrophic: NH_4^+-N → NO_2^--N → NO_3^--N Heterotrophic: NO_3^--N → NO_2^--N → N_2 Heterotrophic: SO_4^{2-} → S^{2-} Autotrophic: S^{2-} → SO_4^{2-}	Azhar et al., 2014

SBR: sequencing batch reactor, EGSB: expanded granular sludge bed, MBfR: membrane biofilm reactor.

Supporting information (SI)

Sulfur-dependent autotrophic denitrification (S1)

Mechanism of the process

The process of autotrophic sulfur (S)-dependent denitrification is used for the treatment of domestic and industrial wastewater (Shao et al., 2010), landfill leachate (Koenig and Liu, 1996), groundwater (Wan et al., 2009) and salt water (Wang et al., 2009). Process reactions are illustrated by the equations 1-8 (Huang et al., 2019, Cui et al., 2019, Lin et al., 2018, Pokorna and Zabranska, 2015, Sun and Nemati 2012):

$$5S^{2-} + 2NO_3^- + 12H^+ \rightarrow 5S^0 + N_2 + 6H_2O \quad (1)$$

$$5S^{2-} + 8NO_3^- + 8H^+ \rightarrow 5SO_4^{2-} + 4N_2 + 4H_2O \quad (2)$$

$$3S^{2-} + 2NO_2^- + 8H^+ \rightarrow 3S^0 + N_2 + 4H_2O \quad (3)$$

$$3S^{2-} + 8NO_2^- + 8H^+ \rightarrow 3SO_4^{2-} + 4N_2 + 4H_2O \quad (4)$$

$$5S^0 + 6NO_3^- + 2H_2O \rightarrow 5SO_4^{2-} + 3N_2 + 4H^+ \quad (5)$$

$$3S^0 + 6NO_2^- \rightarrow 3SO_4^{2-} + 3N_2 \quad (6)$$

$$5S_2O_3^{2-} + 8NO_3^- + H_2O \rightarrow 10SO_4^{2-} + 4N_2 + 2H^+ \quad (7)$$

$$3S_2O_3^{2-} + 7NO_2^- + H_2O \rightarrow 6SO_4^{2-} + 3{,}5N_2 + 2H^+ \quad (8)$$

Compared to heterotrophic denitrification, this process has many advantages, such as no addition of organic substrate, reduction of biomass (the biomass yield coefficient is 0,15-0,57 g for autotrophic biomass production, and 0,71–1,2 g for heterotrophic biomass production per 1 g of denitrified NO_3^--N and NO_2^--N) and a decrease in N_2O emissions (Huang et al., 2019, Sun and Nemati, 2012, Yang et al., 2016).

Thus, the process of autotrophic S-dependent denitrification can be used successfully in major carbon-deficient wastewater treatment systems (Cui et al., 2019). This also explains the use of this process for biogas desulfurization, cleaning of crude oil tanks before acidification, and anti-corrosion treatment of sewage systems (Qian et al., 2015, Park et al., 2014). To date, most of the research done on the S-dependent autotrophic denitrification process has been carried out with flocculent sludge (Cui et al., 2019).

Zhou et al. (2016) indicated that $S_2O_3^{2-}$ is more efficient in S-dependent autotrophic denitrification than S^0 or S^{2-}. Similarly, Park et al. (2015) obtained NO_3^--N removal efficiencies of 96.5% for $S_2O_3^{2-}$, 64.1% for S^{2-}, 58.1% for persulfide (pyrite) and 38.8% for S^0. Thus, it has frequently been shown that $S_2O_3^{2-}$ is a suitable S source for the described process. Cardoso et al. (2006) found that the specific rate of nitrate reduction for $S_2O_3^{2-}$ was 4.6 and 9.5 times higher than for H_2S and S^0, respectively.

When S compounds are transformed, intermediate compounds can also be involved in the process. An example is participation of S^0, resulting from oxidation of S^{2-}, in S-dependent autotrophic denitrification (Xu et al., 2016). Fan et al. (2021) described the effect of intermediates, such as S^{2-}, acid volatile sulfide (AVS), and S^0, in autotrophic denitrification with $S_2O_3^{2-}$ as an electron acceptor. When S^{2-}, $S_2O_3^{2-}$, AVS, and S^0 coexisted in the autotrophic process of denitrification, their preferences were as follows: $S^{2-} > S_2O_3^{2-} > AVS \approx S^0$.

Environmental factors influencing the process performance

Temperature

The optimal temperature for most SOB remains under mesophilic conditions, i.e. in the range of 25-35°C (Fajardo et al., 2014, Shao et al., 2010). When the temperature dropped from 20-25°C to 5-10°C, the efficiency of N removal decreased from about 99% to 50% (Zhou et al., 2011). However, there were also studies showing the denitrification rate of approximately 3.3

kg $N/m^3 \cdot d$ at 3°C in a fluidized-bed reactor with the HRT of 1h during 98 days (Di Capua et al., 2017).

pH

The optimal pH for this process has been shown to be in the range of 6.8-8.2 and a decrease in the activity has been reported at pH <6.5 and >9 (Chen et al., 2016a, Vidal et al., 2002). The optimal pH values for reduction of NO_2-N and NO_3-N were 7.0 and 8.5, respectively, using S^0 as an electron donor (Chen et al., 2018).

Previous research

Jing et al. (2010) performed the S-dependent autotrophic denitrification process with S^{2-} as an electron donor in a 1.3 L up-flow continuous reactor fed with synthetic wastewater. When the NO_3^--N concentration increased from 30.4 mg N/L to 169.6 mg N/L, the volumetric NO_3^--N removal rate increased from 0.15 kg $N/m^3/d$ to 0.61 kg $N/m^3/d$. However, when the NO_3^--N concentration increased again to 189.7 mg N/L, the volumetric NO_3^--N removal rate decreased to 0.59 kg $N/m^3/d$. On the contrary, when the influent concentration of S^{2-} increased from 160 mg/L to 1000 mg/L, the volumetric S^{2-} removal rate increased from 0.78 kg $/m^3/d$ to 4.57 kg $/m^3/d$ and the efficiency of S^{2-} removal was >90%.

Similarly, in the experiments of Kim et al. (2004) with two FBRs, the effect of NO_3^--N concentration on the process was explicitly observed. The feed of FBR-1 was prepared from treated domestic wastewater, while pre-treated leachate from a municipal landfill was used in FBR-2. FBR-1 was operated for 358 days with NO_3^--N of 20 mg N/L, and FBR-2 for 342 days with NO_3^--N of 700 mg N/L. The maximum denitrification rates in FBR-1 and FBR-2 were respectively 2.53 kg $N/m^3/d$ (denitrification efficiency 91.7%) and 3.37 kg $N/m^3/d$ (denitrification efficiency 83%).

Zou et al. (2016) investigated the S-dependent autotrophic denitrification process in two FBRs operated at different temperatures, i.e. 20 and 30°C, for 200 days. Oxidation of $S_2O_3^{2-}$ was particularly unstable until day 54, and then $S_2O_3^{2-}$ remained below the detection limit in both FBRs, and SO_4^{2-} concentration increased sharply to approximately 3700 and 3200 mg/L on days 38 and 33 in FBR-1 and FBR-2, respectively. The temperature had no significant effect in the course of the process and the denitrification rate remained at the level of 1.24-3.25 kg $N/m^3/d$. Autotrophic denitrification and denitritation with $S_2O_3^{2-}$ as electron donors were thus effectively maintained in the two FBRs.

In the study by Sahinkaya and Dursun (2015), the influent NO_3^--N concentration increased from 25 to 75 mg N/L in a FBR with S^0 as an electron donor. The start-up period was relatively short as almost complete NO_3^--N and NO_2^--N reduction was achieved within one week. However, the process efficiency decreased during the long-term operation, which resulted in an increase in NO_3^--N and NO_2^--N concentrations. The denitrification rate ranged from 0.07 to 0.2 kg $N/m^3/d$. SO_4^{2-} was produced and reached the level of 100-600 mg S/L in accordance with the process stoichiometry.

Moraes et al. (2012) investigated S-dependent autotrophic denitrification involving two electron acceptors – NO_3^--N and NO_2^--N in the absence of S^{2-}, in excess of S^{2-} and according to the stoichiometric equation of the S-dependent autotrophic denitrification process in vertical FBRs. The results showed that sulfur intermediate compounds (S^0) were mainly formed when excess of S^{2-} was used, especially for NO_3^--N. Moreover, NO_2^--N was more readily consumed than NO_3^--N, and higher concentrations of S^{2-} led to greater formation of S intermediates. The observed NO_3^--N removal efficiencies were 60-15%, 25.5-98.5% and 84%, respectively, in the absence of S^{2-}, in excess of S^{2-} and in concentration according to the stoichiometric equation of the S-dependent autotrophic denitrification process.

In the study by Sahinkaya and Kilic (2014a), two parallel column bioreactors were operated under autotrophic and heterotrophic conditions. For S-dependent autotrophic denitrification, a packed bed reactor was used. The simultaneous removal of NO_3^--N and chromate (VI) was achieved under both autotrophic and heterotrophic conditions. Cr (VI) concentrations up to 0.5 mg/L did not adversely affect the autotrophic denitrification efficiency and higher concentrations reduced the denitrification potential of the column. During the entire period, production of SO_4^{2-} ranged from 200 to 600 mg S/L and the denitrification rate was 0.07-0.1 kg N/m^3/d.

Sahinkaya et al. (2014b) performed S-dependent autotrophic denitrification under the temperature falling from 26 to 10°C. Three identical pilot-scale column bioreactors were installed, different S to limestone ratios (1/1–3/1) were used and the results were compared under different loading conditions during the long-term operation. Complete denitrification was achieved until the NO_3^--N loading was 10 mg N/L/h. When the temperature dropped to 10°C in winter at the load of 18 mg N/L/h, the denitrification efficiency decreased to 60-70% and the bioreactor with the S/L ratio of 1/1 showed slightly better performance. Throughout the study, the denitrification rate was 0.03-0.24 kg N/m^3/d.

Yang et al. (2016) conducted S-dependent autotrophic denitrification in an up-flow anaerobic sludge blanket reactor operated continuously for 600 days. The nitrogen removal efficiency of 94% and complete removal of S^{2-} were achieved. The denitrification rate was 0.09-0.31 kg N/m^3/d with the HRT of 5 h, and the influent NO_3^--N and S^{2-} loads were 0.33 kg-N/m^3/d and 0.62 kg S/m^3/d, respectively.

Sulfate reduction (S2)

Mechanism of the process

Due to the formation of wastewater streams rich in SO_4^{2-}resulting from various anthropogenic activities, there is a growing interest in the SO_4^{2-} reduction process using SRB. Although high concentrations of SO_4^{2-} do not pose a direct threat to the environment and health, they disrupt the natural S cycle. This imbalance can lead to H_2S formation, metal corrosion, and SO_x emissions (Abel et al., 2015). H_2S is often undesirable in wastewater treatment, but SO_4^{2-} reduction may be beneficial due to the use of S^{2-} for heavy metal removal by precipitation, autotrophic denitrification or autotrophic phosphorus removal (Rubio -Rincón et al., 2017).

Although SO_4^{2-} is the main electron acceptor for SRB, they can also use S compounds such as $S_2O_3^{2-}$, SO_3^{2-}, S^0. When SO_4^{2-} is the electron acceptor, the process takes place in two stages - first SO_4^{2-} is reduced to SO_3^{2-} and then to S^{2-} (Muyzer and Stams, 2008). When SO_4^{2-} decomposes into S^{2-}, some of the decomposed S^{2-} leaves the reactor along with the biogas as H_2S gas and the remaining H_2S present in the reactor as total dissolved sulfide (TDS). The components of TDS in the aquatic environment are S^{2-}, HS- and H_2S (aq) (Samarathunga and Rathnasiri, 2019).

Organic electron donors are used for the biological reduction of SO_4^{2-}, which at the same time provide a carbon source for the SRB, as well as inorganic ones that require supplementation with a carbon source, e.g. CO_2 (Sinharoy et al., 2020a). Some SO_4^{2-} rich wastewater also contains high concentrations of organic compounds that can be used by SRB to reduce SO_4^{2-}. When they contain no organic compounds, compounds such as sugars (glucose and sucrose) (Barber and Stuckey, 2000), alcohols (methanol and ethanol) (Kaksonen et al., 2003), short-chain fatty acids (acetate, propionate, butyrate, lactate, pyruvate, malate) (Kiran et al., 2017) and aromatics (benzoate, phenol) can support the biological reduction of SO_4^{2-} (Liamleam and Annachhatre, 2007).

Lactate, and especially sodium lactate, is the most common electron donor in the biological reduction of SO_4^{2-} with SRB. It can be degraded by a wider range of SRB compared to methanol. However, lactate is more expensive compared to methanol, ethanol or acetate, making this solution on an industrial scale not economical.

Reduction of SO_4^{2-} with lactate (9) (Virpiranta et al., 2019):

$$2CH_3CHOHCOO^- + SO_4^{2-} \rightarrow 2CH_3COO^- + 2HCO_3^- + H_2S \quad (9)$$

Alternative methanol is the cheapest carbon source and is therefore widely used as an electron donor in biological processes. Compared to other carbon sources, its complete oxidation to CO_2 is ensured, while e.g. lactate or ethanol are oxidized only to acetate (Rubio-Rincón et al., 2017), which reduces treatment costs due to the smaller amount of carbon source needed to reduce all SO_4^{2-}. However, it has been reported that methanogens compete with SRB for methanol under mesophilic and thermophilic conditions (Kaksonen and Puhakka, 2007). However, adjusting factors such as pH, temperature, S^{2-} concentration and metal concentration can limit the growth of methanogens in the presence of methanol (Tsukamoto et al., 2004).

Also, ethanol and succinate are widely used as a carbon and electron source as it is a relatively economical option (Virpiranta et al., 2019).

Ethanol reduction occurs in two reactions (10-11), the latter of which is based on acetate reduction (Virpiranta et al., 2019).

Reduction of SO_4^{2-} with ethanol (Virpiranta et al., 2019):

$$2CH_3CH_2OH + SO_4^{2-} \rightarrow 2CH_3COO^- + H_2S + 2H_2O \quad (10)$$

$$CH_3COO^- + SO_4^{2-} + H^+ \rightarrow 2HCO_3^- + H_2S \quad (11)$$

Reduction of SO_4^{2-} with succinate (12) (Virpiranta et al., 2019):

$$4(CH_2)_2(COO^-)_2 + 3SO_4^{2-} + 6H^+ \rightarrow 4CH_3COO^- + 4HCO_3^- + 3H_2S + 4CO_2 \quad (12)$$

H_2, CO and CH_4 can also be used as electron donors, as well as their mixtures, which reduce the cost of the process. Van Houten et al. (1996) have indicated that H_2 is the best electron donor for SO_4^{2-} reduction when working on a large scale. Often, however, CO_2 is required to achieve high process efficiency.

Reduction of SO_4^{2-} with H_2 (13) (Sinharoy et al., 2020b):

$$4H_2 + SO_4^{2-} + H^+ \rightarrow HS^- + H_2O \quad (13)$$

There are SRBs that use CO as a carbon and energy source, however, for most of them, CO is toxic. However, this toxicity can be mitigated by the use of mixed culture systems or the use of a layered structure of biomass (Sipma et al., 2006).

Reduction of SO_4^{2-} with CO (14) (Sinharoy et al., 2020b):

$$4CO + SO_4^{2-} + 4H_2O \rightarrow 4HCO_3^- + HS^- + 3H^+ \quad (14)$$

The biological reduction of SO_4^{2-} can also be combined with the anaerobic oxidation of CH_4. CH_4 can provide 4 electrons, which is twice as much as H_2, so less gas volume is required to achieve the same SO_4^{2-} reduction efficiency. However, CH_4 can only be used by a limited number of microorganisms, which is a major disadvantage of this substrate.

Reduction of SO_4^{2-} with CH_4 (15) (Sinharoy et al., 2020b):

$$CH_4 + SO_4^{2-} + 4H_2O \rightarrow HCO_3^- + HS^- + H_2O \quad (15)$$

Electron donors were also investigated as manure, sawdust, wood shavings, lignocellulosic materials (Reyes-Alvadro et al., 2018), agricultural residues (Chang et al., 2000), sewage sludge, whey or dairy sewage (Wolicka, 2008) and organic sewage (Das et al., 2013), which unfortunately cause secondary environmental pollution.

Sun et al. (2018) proposed a conceptual model for the reduction of SO_4^{2-} and S^0 in the presence or absence of HS^-. When there is no HS^- in the system, the production of HS^- is mainly through the reduction of SO_4^{2-}, while when HS^- is present, polysulfide is formed by the reaction of SO with HS^-. As the concentration of HS^- increases, more polysulfide is formed which stimulates indirect S^0 reduction. When HS^- exceeds a certain level, the indirect reduction of S^0 with polysulfide plays a major role in the production of HS^-, and the reduction of SO_4^{2-} is almost completely stopped. This is important from the point of view of the process economy, as the reduction of S^0 requires ¼ of the organic matter needed to reduce SO_4^{2-}, according to the (16) and (17) reactions (Sun et al., 2018):

$$CH_3COO^- + 4S^0 + 2H_2O \rightarrow 2CO_2 + 4HS^- + 3H^+ \quad (16)$$

$$CH_3COO^- + SO_4^{2-} \rightarrow 2HCO_3^- + HS^- \quad (17)$$

In a study by Sun et al. (2018), an effective S^0 reduction was observed in the sulfidogenic bioreactor even at 1300 mg S/L of SO_4^{2-} in the inlet. 98.5% of HS^- was obtained as a result of S^0 reduction, and SO_4^{2-} was barely consumed in the bioreactor.

Environmental factors influencing the proces performance

Temperature

SRBs are able to tolerate temperatures ranging from -5°C to 75°C and readily adapt to temperature changes (Cocos et al., 2002). However, the optimal temperature for most SRBs is in the narrow range of 28-30°C (Virpiranta et al., 2019), although there are strains for which the optimum temperature is below 20°C (Knoblauch et al., 1999). Most of the bacteria that tolerate cold temperatures are mesophilic (rather than psychrophilic) strains that can grow under such conditions, termed psychrotolerant bacteria or psychotrophs. For example, some *Desulfobulbus* strains can grow at 6-10°C (Kharrat et al., 2017, Virpiranta et al., 2019).

Studies have also been conducted in the range of even lower 4-8°C (Nielsen et al., 2018), which, however, has extended the adaptation time to such extreme conditions. The presence of SRB was revealed by the presence of the *Deltaproteobacteria* and *Clostridia*. The study showed a change in the composition of SRB taxa over time - an increase in the relative abundance of *Deltaproteobacteria* and a decrease in the relative abundance of members of the *Clostridia*.

pH

Most of the studies on SO_4^{2-} reduction were carried out at pH in the range of 7-7.6, which is close to the optimal value for SRB. Pagnanelli et al. (2012) found the optimal pH value of 7.6 for the SRB, while Bratkova et al. (2013) observed the maximum SO_4^{2-} reduction rate at pH 7.25.

Previous research

Sinharoy et al. (2019, 2020a) desulphurized wastewater containing SO_4^{2-} using carbon monoxide in a moving bed biofilm reactor (2019) and in a gas lift reactor (2020a). The effect of HRT on the process was investigated and it was found that at the HRT of 72, 48 and 24 h, SO_4^{2-} removal was 93.5%, 91.9% and 80.1%, respectively, and CO use was 85% throughout the study (Sinharoy et al., 2019). These results were improved in a subsequent study (Sinharoy et al., 2020a). At the HRT of 72 h, the reduction of SO_4^{2-} and the use of CO were 97% and 89%, respectively (2020a). The best results in terms of SO_4^{2-} reduction (> 80%) were obtained for low SO_4^{2-} loading and high CO loading conditions. The results for both types of reactors were slightly better for the gas lift reactor.

Batch studies of Virpiranta et al. (2019) were performed in sealed vials and incubated at 3 different temperatures of 6°C, 16°C and 22°C for cold acclimatization and characterization of SRB consortia enriched from a sample of arctic sediments. Postgate

medium was supplemented with lactate, ethanol or succinic acid and the resulting consortia grew with lactate and succinic acid, but not ethanol. The SO_4^{2-} reduction rates at 22°C were 169 mg/L/d, at 16°C - 98 mg/L/d, and for 6°C the rates ranged from 13 to 42 mg/L/d. Temperature had a significant effect on the activity of SRB. However, it is useful to be able to acclimatize SRB to low temperatures due to the versatile applicability of the process.

Nielsen et al. (2019) conducted batch tests to check various carbon sources for SO_4^{2-} reduction. Simple organic carbon sources (methanol and ethylene glycol) and complex organic carbon sources (potato oil, brewing residues, peat and straw) were used to support SRB growth. After 162 days, all bioreactors showed a decrease in both total organic carbon and SO_4^{2-} concentration at 5°C. However, a long acclimation period (98–112 days) was required. The use of methanol and ethylene glycol resulted in SO_4^{2-} reduction by 71.2% and 36.9%, respectively. The decrease in SO_4^{2-} concentrations was limited to 13.8 and 5.3%, respectively, when using peat and straw.

Reyes-Alvarado et al. (2017, 2018) used electron donors, such as potato, filter paper, crab shell (2017) and natural scourer and cork (2018), in their research. Maintaining SO_4^{2-} concentration close to 700-800 mg/L in both studies, the following SO_4^{2-} removal rates were obtained: 6-9, 22-34, 50-65, 34, 6.1 mg SO_4^{2-}/gVSS/d for crab shell, potato, filter paper, scourer, cork, respectively. It was shown that the natural properties of the carbohydrate-based polymers limit the hydrolysis-fermentation step and thus the SO_4^{2-} reduction rate.

Sulfammox process (S3)

Mechanism of the process

A novel sulfammox biological process has recently been described in which NH_4^+-N is oxidized to N_2 and SO_4^{2-} plays the role of an electron acceptor and is reduced to S^0 under anaerobic conditions. Sulfammox was first reported by Fdz-Polanco et al. (2001b) in a

granular activated carbon anaerobic fluidized bed reactor treating vinasse from an ethanol distillery of sugar beet molasses. The sulfammox process can be most generally represented by the following equation (18):

$$SO_4^{2-} + 2NH_4^+ \rightarrow S^0 + N_2 + 4H_2O \quad (18)$$

In the follow-up studies, Liu et al. (2008) and Yang et al. (2009) identified SO_4^{2-} as a potential electron acceptor as it was the feed component. The mechanism of the sulfammox process is not fully understood yet - it turns out that there are exceptions to the described process. In addition to N_2 and S^0, sulfammox can lead to the formation of NO_2^--N and NO_3^--N as well as S^{2-}. Moreover, there are two different assumptions about the actual course of the sulfammox process. The first assumes that NH_4^+-N is partially oxidized to NO_2^--N with SO_4^{2-} and some of the NO_2^--N produced is reduced to N_2 by S^{2-} and then NO_2^--N and NH_4^+-N are converted to N_2. The second is that NH_4^+-N is partially oxidized to NO_2^--N by SO_4^{2-} and then NH_4^+-N is oxidized to N_2 by NO_2^--N (Yang et al., 2009, Liu et al., 2008).

Even though COD is not required for the sulfammox process (Zhang et al., 2009), the experiments were performed either without COD addition (Bi et al., 2020, Zhang et al. 2019a,b, Prachakittikul et al., 2016, Cai et al., 2010) or with COD addition (Wang et al., 2017, Rikmann et al., 2016, Fdz-Polanco et al., 2001a,b). When COD is present in wastewater, the sulfammox process can be coupled with subsequent heterotrophic denitrification (Zhang et al. 2019b).

The differences in the NH_4^+-N/SO_4^{2-} ratio in the conducted studies with respect to the stoichiometric ratio may result from the activity of ammonia oxidizing bacteria (AOB), which oxidizes NH_4^+-N, and the reduction of SO_4^{2-} by organic compounds, as described by Bi et al. (2020). In this way, they questioned the presence of the sulfammox process, so it is important to conduct more detailed research.

Environmental factors influencing the process performance

Temperature

The sulfammox process has been carried out at the temperatures in the range of 15-55°C (Zhang et al., 2019a,b, Cai et al., 2010, Yang et al., 2009, Zhao et al., 2006) with the optimal value in the range of 25-35°C (Cai et al., 2010). However, the sulfammox process could also be maintained at lower temperatures, e.g. 20°C (Rikmann et al. 2016) and 14-15°C (Wu et al. 2020). In the latter case, the NH_4^+-N and overall SO_4^{2-} removal efficiencies still remained high, i.e. 98.5% and 52.8%, respectively.

pH

Sulfammox studies have mainly been carried out in the pH range of 7.5-8.6, specifically 7.5-8.5 (Yang et al., 2009), 8-8.2 (Liu et al., 2008) 8.1-8.3 (Zhang et al., 2019a), 8.1-8.6 (Zhang et al., 2019b) with the best value of 8.5 (Cai et al., 2010). The efficiency of NH_4^+-N and SO_4^{2-} removal decreased when the pH was < 7.5 and > 9.5 (Cai et al., 2010).

Previous research

Qin et al. (2021) conducted a sulfammox process in an upflow anaerobic sludge bed reactor for more than one year. The influent NH_4^+-N concentration was 70 mg N/L and the NO_2^-: NO_3^-: SO_4^{2-} molar ratio were 1:0.2:0.2, 0.5: 0.1: 0.3 and 0: 0:0.5, respectively, in stages I, II and III. The NH_4^+-N and SO_4^{2-} removal rates were 31 mg N/L/d and 8.18 mg S/L/d, respectively. The excessive conversion of NH_4^+-N in stage III was mainly attributed to the sulfammox reaction due to the high removal rate ratio of NH_4^+-N: SO_4^{2-} (8.67: 1), accumulation of S^{2-} and decreased pH, as well as a changed structure of the microbial communities.

The results of Zhang et al. (2019a) in a circulating flow reactor showed that the efficiency of NH_4^+-N oxidation and SO_4^{2-} reduction increased in the presence of NO_2^--N and

306 NO_3^--N. Nitrogen has been converted by nitrification, denitrification and conventional
307 anammox, simultaneously with the sulfammox process. The SO_4^{2-} removal efficiency reached
308 the maximum of 45%.

309 In the batch tests of Cai et al. (2010), reduction of SO_4^{2-} (40%) and NH_4^+-N (44%)
310 was considered exclusively due to sulfammox. Similarly, Yang et al. (2009) successfully
311 performed sulfammox in an upflow anaerobic sludge blanket reactor. Only sulfammox was
312 assumed to be responsible for the reduction of NH_4^+-N and SO_4^{2-} with the SO_4^{2-} removal
313 efficiency of 30%.

314 ***Modeling N, S and C transformations in wastewater treatment systems (S4)***
315 Koenig and Liu (2001) established a half-order kinetic model for biofilms to describe
316 autotrophic denitrification by *Thiobacillus denitrificans* in an upflow S packed-bed reactor
317 fed with synthetic wastewater. The half-order reaction rate constants for autotrophic
318 denitrification using S^0 were approximately one order of magnitude lower than those of
319 heterotrophic denitrification. However, the model was validated with pure substrate and pure
320 bacteria in a biofilm under autotrophic conditions, which largely limited its application to
321 other complex systems.

322 An et al. (2011) investigated the kinetics of two-step heterotrophic denitrification
323 (reduction of NO_3^--N to NO_2^--N, and subsequently to N oxides and N_2 gas) using an oil
324 reservoir culture, which was capable of functioning under both autotrophic and heterotrophic
325 conditions. The developed kinetic model predicted the experimental results of batch and
326 continuous systems in terms of simultaneous removal of S^{2-}, NO_3^--N, NO_2^--N and organic
327 compounds.

328 Wang et al (2010) proposed a kinetic model to monitor a denitrifying S^{2-} removal
329 (DSR) process with the ASM1 as a core model. By establishing inhibition and switch
330 functions, the competition between autotrophic and heterotrophic denitrifiers was described,

including the effect of S^{2-} inhibition on heterotrophic denitrification. The calibrated model was used to quantify the impact of the influent C/S ratio and S^{2-} levels on the performance of a bench-scale EGSB reactor. Model predictions indicated that the DSR reactor would operate efficiently when the influent C/S ratio was kept in the range of 0.5-3.0, and the S^{2-} concentration remained below 1000 mg S/L.

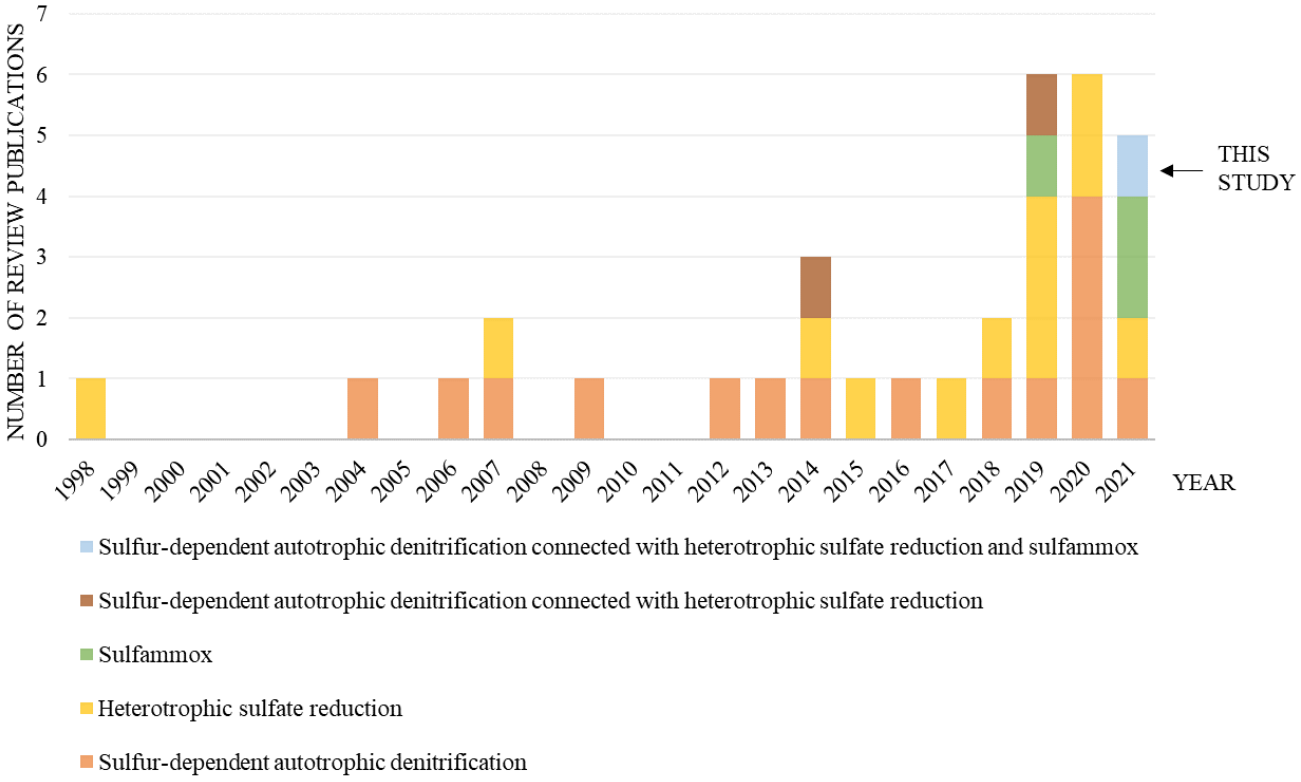
Xu et al (2014) developed a model to describe simultaneous removal of S^{2-}, NO_3^--N and acetate (sole organic substrate) under denitrifying S^{2-} removal conditions in a continuous flow reactor. The kinetic parameters were estimated via data fitting while considering the effects of initial S^{2-} concentration, S^{2-}/ NO_3^--N ratio and acetate/ NO_3^--N ratio. The proposed model accurately described the performance of DSR over wide ranges of the parameters. Model predictions suggested that the adjustment of HRT would be an efficient way to mitigate high S^{2-} loadings. Despite accurate predictions for a pure substrate (acetate), the model might not be applicable for actual industrial wastewater with complex characteristics.

Xu et al (2016) developed an autotrophic denitrification kinetic model to describe S^{2-} oxidation and NO_2^--N removal in a bench-scale sequencing batch reactor (SBR). The model parameters were estimated by data fitting from two studies with different combinations of S^{2-}, S^0, SO_4^{2-}, NO_3^--N and NO_2^--N. The final products of S^{2-} oxidation (S^0 and SO_4^{2-}) and their concentrations could be accurately predicted, providing a strategy to control the effluent SO_4^{2-} concentration or recover S^0 as the main end product from S^{2-} oxidation.

Xu et al (2017) further developed a complex model for C-N-S removal by combining the ASMs and ADM1 (Anaerobic Digestion Model No.1), extended with oxygen/NO_3^--N driven S^{2-} oxidation processes. The proposed model was also capable of simulating S relevant processes, such as SO_4^{2-} reduction, S^{2-} oxidation and denitrifying S^{2-} removal process. Due to some simplifications in the model structure and parameter uncertainty, that model would not yet serve as a precise and quantitative tool in various full-scale applications.

Mechanistic models may also be useful in understanding the mechanisms of sulfammox and its interactions with co-existing biochemical processes. However, due to the complexity of the interactions, it still lacks research reports about the model development for the sulfammox process. Chen et al (2016b) established a S-involved anammox model to simulate the coexistence of AOB, nitrate oxidizing bacteria (NOB), anaerobic ammonia oxidizing bacteria (AAOB), denitrification anaerobic methane oxidation (DAMO) bacteria, and SOB in a membrane biofilm reactor (MBfR). The model described removal of NH_4^+-N, dissolved CH_4, and S^{2-} from sidestream anaerobic sludge digestion liquors. However, other potential processes, such as endogenous heterotrophic denitrification, S-dependent autotrophic denitrification and SO_4^{2-}/S reduction, were not incorporated in that model, and S^{2-} inhibition was also neglected.

Azhar et al (2014) modeled the coupled N and S cycles in a coastal upwelling system. In the S cycle, S^{2-}-driven denitrification, SO_4^{2-} reduction, and S^{2-} oxidation by NO_3^--N (chemolithoautotrophic NO_3^--N reduction) were considered. However, as the experimental study was conducted in a natural costal water column and sediment, the reaction formulations, process rates and kinetic parameters might not be directly applicable in wastewater treatment systems. Moreover, some reactions, such as nitrogen fixation and remineralization, might be neglected in those systems.



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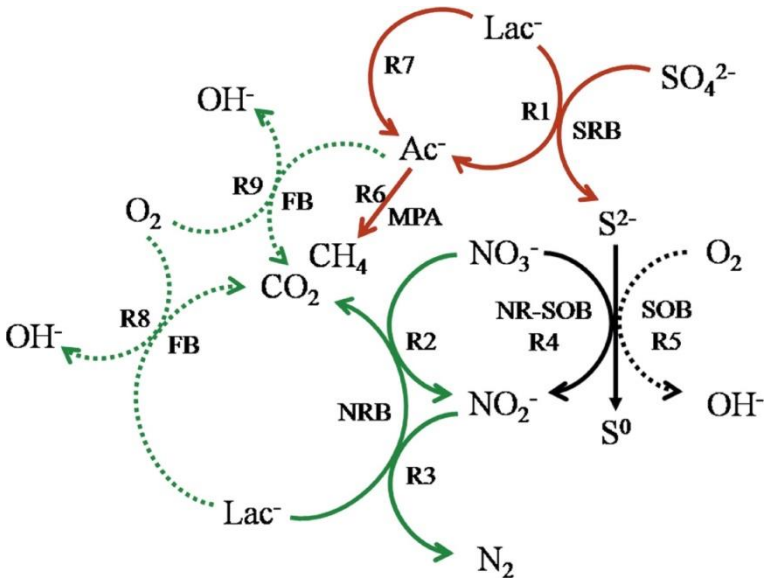
380 Figure S1. Number of review papers on S-dependent biochemical processes, including
381 autotrophic denitrification, heterotrophic sulfate reduction and sulfammox

382



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384 Figure S2. Simplified representation of the biochemical process associated with C, N and S
385 conversions (Xu et al., 2017).

386

387 Table S1. Issues incorporated in the review articles on the N-S-C cycles in wastewater treatment

Process/System	Issue	Cui et al., 2019	Tian and Yu, 2020	Hu et al., 2020	Ma et al., 2020	Wang et al., 2020	Park and Yoo, 2009	Lin et al., 2018	Wu et al., 2021	Show et al., 2013	Costa et al., 2020	Serrano et al.., 2019	Lens et al.., 1998	Xu and Chen, 2020	Kumar et al., 2021	Hao et al.., 2014	van den Brand et al., 2015	Rios-Del Toro and Cervantes,	Liu et al., 2021	Grubba et al.., 2020	This study
S - DEPENDENT AUTOTROPHIC DENITRIFICATION	recent advances in S-dependent auotrophic denitrification	✔	✔	✔	✔	✔	✔	✔	✔	✔											updated
	potential use of S-dependent autorophic denitrification	✔	✔	✔	✔	✔	✔	✔	✔	✔						✔	✔				updated
	microbiological reactions and microorganisms involved	✔		✔	✔	✔	✔	✔	✔	✔											✔
	factors influencing the operation of S-dependent auotrophic denitrification	✔		✔	✔	✔			✔	✔							✔				✔
	comparison of greenhouse gas emissions from autotrophic and heterotrophic denitrification	✔				✔			✔							✔					
	reactor configurations									✔											✔
	comparison of different electron donors (S compounds) for autotrophic denitrification	✔	✔	✔	✔	✔	✔	✔	✔	✔											✔
	cost comparison of reduced-S species and organic substrate as an electron donor	✔				✔	✔														
	recent works on the hydrogen and/or iron- and/or methane- and/or carbon- S-dependent autotrophic denitrification	✔	✔		✔	✔	✔		✔												
	mixotrophic denitrification		✔				✔		✔	✔											✔
	natural occurrence of S-dependent autorophic denitrification			✔																	

	autotrophic denitrification in constructed wetlands		✔															
	S^0 reclamation and energy harvest			✔		✔	✔											
	S cycle combined with C and/or N and/or P cycles			✔		✔	✔	✔						✔				
	environmental effects			✔			✔											
	comparison of biofilm reactors						✔											
	modeling description							✔										
	prospects, limitations and future research needs	✔	✔	✔	✔	✔	✔	✔										updated
HETEROTROPHIC SULFATE REDUCTION	recent reports on SO_4^{2-} removal efficiency												✔	✔				updated
	application of Sulfate Reducing Bacteria (SRB)								✔	✔	✔	✔	✔	✔	✔			✔
	SRB characteristics and metabolism								✔	✔	✔	✔		✔				✔
	competition of SRB for substrates										✔							✔
	SRB biomass source									✔		✔						
	strategies to improve SRB tolerance to heavy metals											✔						
	factors influencing the SRB metabolism									✔	✔	✔	✔	✔				✔
	sulfate and sulfide removal techniques										✔			✔				✔
	bioreactors configurations								✔				✔					✔
	electron donors characteristics								✔	✔		✔		✔				✔
	electron acceptors characteristics									✔				✔				
	SRB biomass disposal methods											✔						
	SRB-test monitoring									✔								
	reporting data in SRB-test									✔								
	emissions of SO_4^{2-}-contaminated waters										✔							

	characteristics and effects of water pollution by SO_4^{2-}			✓								
	treatment of acid mine drainage (AMD)	✓					✓					
	heavy metal removal	✓			✓	✓	✓					
	methods for enhancement of SRB activity				✓							✓
	sulphide forms in aqueous medium at different pH values	✓		✓								
	prospects, limitations and future research needs	✓	✓	✓	✓	✓	✓					updated
SULFAMMOX	recent advances in sulfammox process							✓	✓	✓		updated
	characteristics of sulfammox							✓	✓	✓		✓
	potential use of sulfammox process							✓	✓	✓		updated
	characteristics of NH_4-N anaerobic oxidation processes with different electron acceptors							✓				
	spontaneity and oxidation-reduction potential									✓		
	functional bacteria								✓	✓		✓
	possible interactions with other bacteria								✓	✓		✓
	bacteria responsible for the specific N and S transformations								✓	✓		✓
	factors influencing the sulfammox process								✓	✓		✓
	the occurrence of sulfammox in the environment							✓				✓
	source of SO_4^{2-}									✓		
	reactors types							✓	✓	✓		✓
	co-existence of anammox, S-dependent autotrophic denitrification and sulfammox								✓	✓		✓

	prospects, limitations and future research needs			✔	✔	✔	✔
SANI PROCESS	SANI process characteristics	✔	✔				✔
	diagram of the SANI reactor system	✔	✔				✔
	SANI key-parameters		✔				✔
	recent advances in SANI process						✔
	FGD-SANI and MD-SANI characteristics						✔
	diagram of the FGD-SANI and MD-SANI reactors system						✔
	FGD-SANI and MD-SANI key-parameters						✔
S CYCE WITH ANAMMOX	Sulfate Reduction, Denitrification/Anammox and Partial Nitrification (SRDAPN) characteristics						✔
	Partial Nitrification/Anammox and S-dependent autotrophic Denitrification (PNASD) characteristics						✔
	Anammox and S-dependent autotrophic Denitrification (ASD) characteristics						✔
	S-dependent autotrophic Partial Denitrification/Anammox (SPDA) characteristics						✔
	diagram of SRDAPN, PNASD, ASD nad SPDA						✔
	recent advances in SRDAPN, PNASD, ASD nad SPDA						✔
	SRDAPN, PNASD, ASD nad SPDA key-parameters						✔

SYSTEMS WITH SULFAMMOX	Sulfammox/Anammox (SA) characteristics	✔
	Sulfammox - S-dependent autotrophic Denitrification (SSD) characteristics	✔
	Sulfammox – Anammox - S-dependent autotrophic denitrification (SASD) characteristics	✔
	diagram of the SA, SSD and SASD	✔
	recent advances in SA, SSD and SASD	✔
	SA, SSD and SASD key parameters	✔

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