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Indirect sulfur reduction via polysulfide contributes to serious odor problem in a sewer receiving nitrate dosage

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ABSTRACT

Nitrate dosing is commonly used to control hydrogen sulfide production in sewer systems. However, quick rebound of the sulfide concentration after nitrate depletion has been observed and results in more serious odor and corrosion problem. To investigate the mechanism of sulfide regeneration in the nitrate-free period, a laboratory-scale sewer reactor was run for 30 days to simulate sulfide production and oxidation with intermittent nitrate addition. The results show that nitrate addition substantially reduced the sulfide concentration, but the produced elemental sulfur was then quickly reduced back to sulfide in nitrate-free periods. This induced more and more sulfide production in the sewer reactor. Elemental sulfur and polysulfide reductions were found in the sewage in nitrate-free periods, showing their contributions to the sulfide regeneration. Through batch tests, polysulfide was confirmed as the key intermediate for accelerating sulfur reduction during the nitrate-free period in the sewer. Sulfide production rates significantly increased by 65% and 59% in the presences of tetrasulfide and sulfur with sulfide, respectively, at the beginning of the test. While polysulfide formation was prevented by the ferrous chloride addition, the sulfur reduction rate remarkably decreased from 12.8 mgS/L-h to 1.8 mgS/L-h. This indicates that direct sulfur reduction was significantly slower than the indirect sulfur reduction via polysulfide; the latter process could be the cause for the quick rebound of the sulfide concentration in the sewer with intermittent nitrate dosing. Thus, the pathways of sulfur transformations in a sewer, both in the presence and absence of nitrate, were proposed. Microbial community analysis results reveal that some common sulfate-reducing bacteria (SRB) genera in sewer sediment were possible sulfur reducers. According to this finding, the effect and strategy of nitrate dosing for hydrogen sulfide control in sewers should be re-evaluated and re-considered.

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1. Introduction

In anaerobic conditions, sulfate-reducing bacteria (SRB) facilitate the production of sulfide in sewer systems, resulting in sewer corrosion and odor problems. In large cities, this problem is getting worse because the hydraulic retention time (HRT) is getting longer due to sewer network expansion. Many strategies have therefore been exploited to prevent or diminish sulfide formation, during which the addition of nitrate has been proven to be effective and has been widely used for sulfide control in sewers (Jiang et al., 2009; Mohanakrishnan et al., 2009; Yang et al., 2005; Zhang et al., 2008). Various nitrate salts (i.e., ammonia and calcium nitrate) coupled with different concentrations (10–40 mg N/L) have been successfully tested as being able to suppress sulfide concentrations to 0.2–3 mg S/L over the recent seven decades (Mohanakrishnan et al., 2009; Saracevic et al., 2007). Mathioudakis et al. (2006) reported that the use of nitrate of 6.9, 15 and 27.7 Kg for a period of 4–8 h in a 6.7 km combined sewer resulted in 84, 98 and 99% removal efficiencies of sulfide, respectively. No sulfide was discharged when nitrate was not completely consumed. The mechanism involved is the biological oxidation of sulfide to elemental sulfur or sulfate by nitrate-reducing sulfide-oxidizing bacteria (NR-SOB), as described in Eqs. (1) and (2) (Li et al., 2009).

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$$5S^{2-} + 2NO_3^- + 12H^+ \rightarrow 5S^0 + N_2 + 6H_2O \quad \Delta G^0 = -995kJ/reaction \tag{1}$$

$$5S^0 + 6NO_3^- + 2H_2O \rightarrow 5SO_4^{2-} + 3N_2 + 4H^+ \quad \Delta G^0 = -2738kJ/reaction \tag{2}$$

However, to avoid increasing the nitrogen loading to the downstream wastewater treatment plant and control chemical cost, nitrate dosing frequency and dosage are generally limited (Liu et al., 2015c). The dosed nitrate in sewage is not only consumed by sulfide oxidation but also through heterotrophic denitrification, resulting in a high nitrate consumption rate in sewers (Jiang et al., 2011). Therefore, in intervals between nitrate dosing events or in a downstream pipeline, nitrate is generally depleted. Some studies have reported that nitrate depletion in sewers enhanced the sulfide generation rate and caused more serious odor problems (Jiang et al., 2009, 2013c; Mohanakrishnan et al., 2009).

After investigations, Jiang et al. (2009) claimed that this resulted from the quick elemental sulfur reduction in sewer sediment and biofilm, with the sulfur in sediment/biofilm coming from anoxic sulfide oxidation. In the presence of nitrate, most of the sulfide in the sewer was oxidized into elemental sulfur, which was deposited in sewer sediment and biofilm (Jiang et al., 2009). This finding was also reported by Yang et al. (2005), who found that elemental sulfur was the main end-product in sewers.

However, elemental sulfur is characterized by low water solubility (5 μg/L at 25 °C), which could lead to low bioavailability. Only a limited proportion of SRB in sediment/biofilm can directly contact and grow with the deposited elemental sulfur. This means that the contribution of direct sulfur reduction in sediment/biofilm to sulfide regeneration in sewers should be limited. However, on the contrary, the sulfide production rate due to elemental sulfur reduction was found to be as high as 5.4 mg S/m^2-h in the lab-scale sewer receiving nitrate dosage (Jiang et al., 2009). Thus, a new question arises: how does a fast sulfur reduction happen in sewers?

In previous studies, polysulfide (S_n^{2-}) has been reported as the intermediate of sulfur reduction (Blumentals et al., 1990; Boyd and Druschel, 2013; Ringel et al., 1996). In ambient conditions, the S_8 ring of elemental sulfur is cleaved by a nucleophilic attack of the HS^- anion in the presence of sulfide, which results in the formation of polysulfide (see Eq. (3)) (Rabus et al., 2013). Polysulfide is a carrier of zero valent sulfur atoms but is soluble and highly bioavailable (Hedderich et al., 1998; Schauder and Müller, 1993). Thus, it can be inferred that the soluble polysulfide can diffuse into deeper layers of sediment/biofilm and be utilized by sulfur reducers. This indicates that the formation of polysulfide in sewers could possibly enhance the bioavailability of elemental sulfur and increase the sulfide production rate after the depletion of nitrate.

$$HS^- + (n-1)S^0 \rightarrow S_n^{2-} + H^+ \tag{3}$$

To validate this hypothesis, a lab-scale sewer reactor was established and fed with synthetic sewage (Fig. 1). We aimed to study the mechanism of sulfide regeneration after nitrate depletion; thus, an intermittent nitrate dosing strategy was applied to this sewer reactor after it was stable. Batch tests were performed to investigate the role of polysulfide in sulfur reduction. Microbial communities were also analyzed by using an Illumina MiSeq sequencer to identify the metabolically active microbial communities.

2. Material and methods

2.1. Experimental sewer reactor set-up and operation

The lab-scale system was an airtight box culvert reactor (10 cm × 10 cm × 10 cm) with a volume of 1 L, as shown in Fig. 1. A square lightweight expanded polystyrene of 1 cm depth was used as biofilm carriers to support biofilm growth. The sharp of the biofilm carriers fitted to the reactor's configuration. The total volume of the carriers was approximately 100 ml, accounting for 10% of the reactor volume. The seeding sludge in this work was collected from a 1.5 km long sewer located at the Hong Kong University of Science and Technology (HKUST), Hong Kong, China. The details of this sewer were introduced in our previous study (Jiang et al., 2007). The depth of sediment in the reactor was 2 cm. To avoid fluctuations of sulfide production rate due to varied organic matters and sulfate concentrations in the influent wastewater, synthetic domestic wastewater was used in this study and prepared every two days according to our previous research (Jiang et al., 2013b). It consisted of 122 mg/L glucose, 61 mg/L yeast extract, 163 mg/L sodium acetate, 115 mg/L NH_4Cl, 4.5 mg/L KH_2PO_4, 12 mg/L K_2HPO_4, 927 mg/L Na_2SO_4 and 2 mL of a trace element concentrated solution. Afterwards, the synthetic domestic wastewater was stored in a 4 °C refrigerator and slowly pumped into the sewer reactor to obtain: 297 ± 20 mg/L chemical oxygen demand (COD), 209 ± 14 mg/L SO_4^{2-}-S. Four inlets were distributed in parallel to simulate uniform flow in the sewer. Sewage was pumped into the sewer reactor from inlets and partially recycled back from outlet to inlets at an internal recycling ratio of 10:1 to provide a surface flow rate of 0.3 cm/min. Thus, the hydraulic retention time (HRT) in the sewer reactor was 10 h. The temperature of the sewer reactor was kept at 25 °C by using a water bath.

The sewer reactor was operated for 14 days before adding nitrate to provide anaerobic conditions for the development of sulfate-reducing sediment/biofilm. Concentrated sodium nitrate solution (2 g/L) was prepared weekly, stored in a 4 °C refrigerator and then periodically fed into the sewer reactor by a timer-controlled peristaltic pump for 12 h in one day to provide 40 mg N/L nitrate in the sewage. Nitrate addition was performed for 16 days.

Water samples were collected at the end of each nitrate exposure event and nitrate free time. The concentrations of nitrate, nitrite, sulfate, sulfide, COD and elemental sulfur in sewage were measured. Meanwhile, the concentration of polysulfide was also analyzed.

After the 30-days experiment, sewage samples were intensively collected bihourly to study the mechanism of sulfide regeneration. The variations of nitrate, nitrite, sulfate, thiosulfate, sulfide, elemental sulfur and polysulfide concentrations over 12 h after nitrate dosing were analyzed.

2.2. Batch tests: role of polysulfide in sulfur reduction

To investigate the mechanism of sulfur reduction, two sets of batch test were conducted in this study.

Batch test set I was performed to study whether polysulfide stimulates sulfur reduction in a sewer environment. In batch test set I, sulfur, polysulfide and sulfur with sulfide were added to each batch test reactor with sewer sediment and synthetic sewage. Detailed information regarding these batch tests is presented in Table 1. The sediments taken from the HKUST sewer were evenly distributed into each 300-ml flask (72 ± 7 mg VSS/L) (VSS: volatile suspended solids). Disodium tetrasulfide (Na_2S_4, analytical grade, 98%) was used as the polysulfide in the batch test. Bio-sulfur from a biological sulfide-oxidizing reactor in our lab was used as elemental

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