Hydrogen Sulfide Generation and Control in Sewer Systems with Drop Structures and Pump Stations

by

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Abstract

The build-up of hydrogen sulfide in sewer systems can lead to odor nuisances and corrosion of concrete sewers. The City of Edmonton, Alberta, Canada, like many other cities worldwide, is facing sewer odor nuisance and pipe corrosion issues. Field studies were carried out in the sanitary sewer trunk with drops and pump station in Steinhauer area and in the sewer trunk with drops in Bonnie Doon area to identify causes of H_2S in both areas and the effect of drops on H_2S release so as to develop proper odor control strategies. In addition, four bioreactors imitating force mains of pump stations were operated to evaluate sulfide generation rate. Ferric and nitrate were added into two of the four bioreactors to investigate their effects on sulfide control. Sulfide formation rates in the force mains were also evaluated in the field and continuous nitrate dosing was applied in the wet well of the Big Lake pump station to investigate its effectiveness on sulfide control.

In Steinhauer area, relatively high concentrations of H_2S were detected at the beginning and the end of the trunk with odor complaints. At the beginning of the trunk, sulfide emission was mainly caused by the increased stripping effect of the drop structures. The pump operation at the end of the trunk led to the long hydraulic retention time (HRT) of the sewage and the subsequent sulfide generation in the trunk and wet well. The calibrated sulfide generation models by using field measurements in the trunk were applied to assess the proposed mitigation strategy, pump operation optimization, and this strategy was found to be able to eliminate sulfide generation in the study trunk sewer.

In Bonnie Doon area, the liquid sulfide concentration in the upstream trunk was low (less than 1.0 mg/L), and no H_2S gas was detected in the head space. However, high H_2S gas concentration was detected in the middle reach of the trunk due to the stripping effect of the three drops (2.7 m, 5.2 m, and 2.0 m) along the trunk. The released H_2S at drops was transported to downstream trunk and tributaries in the sewer system and caused odor concerns at these locations. These drops played an important role on H_2S release, and the overall H_2S mass transfer coefficient at drops was much higher than that in normal gravity sewers. The overall oxygen and H_2S mass transfer coefficient were estimated to be around 200 h^{-1} and 300 h^{-1} at first two drops, respectively.

Sulfide controlled by ferric in the lab-scale bioreactor was only via chemical oxidation and precipitation and total sulfate reducing bacteria (SRB) population did not change. The heterotrophic nitrate reducing bacteria (hNRB), *Thauera*, outcompeted SRB for organic matter as electron donor as the dominant bacteria when nitrate was added at the beginning of the pump cycle. The sulfide generation was controlled by hNRB activities. A cost-effective nitrate dosing strategy is proposed to add the nitrate at the end of the pump cycle instead of at the beginning of the pump cycle which can save up to 75% nitrate dosage. The co-existence of sulfide and nitrate stimulated the development of nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB), *Sulfurovum*, which was responsible for sulfide removal in the presence of nitrate. Both ferric and nitrate did not have long-lasting inhibitory/toxic effect on sulfate reduction.

The sulfide production rates at the force mains of four pump stations were estimated to be 0.08~0.15 g/m^2h under 20 °C. The 1/2-order biofilm kinetics of DCOD was used to model the sulfide generation rate in the force main and the modelled rate constant for field trial was around 0.006. In the discharge manhole of the Big Lake pump station, sulfide concentration was measured at around 20 mg/L while the H_2S gas concentration reached up to 400~500 ppm. A continuous dosing of 85 mg/L nitrate in the pump wet well could completely suppress the sulfide generation in the force main. An optimized dosing strategy is to add the nitrate at a location close to the point of sulfide control rather than in the wet well and the dosing rate should be proportional to the HRT of the wastewater in the between the point of nitrate addition and the end of the force main.

Preface

This thesis is composed of my original work, and contains no material previously published or written by another person except where due reference has been made in the text. I have clearly stated the contribution by others to jointly authored works that I have included in my thesis.

The field measurements described in Chapter 3, 4, 5 and 7 of this thesis were planned by myself and executed by myself with assistance from colleagues. The data analysis discussed in this thesis is my original work with direction from my supervisor, Professor D. Z. Zhu.

Chapter 4 of this thesis has been published as Yang, Z., Zhu, D.Z., Yu, T., Edwini-Bonsu, S. and Liu, Y. (2019). "Case Study of Sulfide Generation and Emission in Sanitary Sewer with Drop Structures and Pump Station". *Journal of Water Science and Technology*, 79 (9), 1685-1694. I was responsible for the data collection and analysis and writing the manuscript. Zhu D.Z was the supervisory author and was involved in the concept formation and manuscript composition and editing. Edwini-Bonsu S. assisted with data collection and contributed to manuscript edits along with Yu T. and Liu Y..

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Chapter 1 Introduction

1.1 Background

Sewer odor problems have been widely reported in cities worldwide such as San Francisco, Los Angeles, and Edmonton (Ganigue et al. 2011, Vollertsen et al. 2015). The City of Edmonton has been suffering from sewer odor problem for many years. It receives close to 1000 sewer odor complaints every year, and the mapping of these complaint locations indicates city-wide sewer odor issues (**Figure 1-1**) (Pan et al. 2020). Research is urgently needed in order to find out the specific reasons for sewer odor problems in Edmonton and corresponding mitigation strategies. What makes the Edmonton's sewer system unique is the deep trunk lines (up to 50 m below ground), which result in over 900 pump stations and 800 drop structures in its sanitary sewer systems. The pump stations and drop structures are potential sites for sulfide generation and release, respectively. These locations could be the main causes of the odor problems in Edmonton.

Both problems of sewer odor and pipe corrosion are essentially related to sewer gases (mainly H_2S). When anaerobic conditions prevail in a sewer system, sulfate can be reduced to sulfide by sulfate-reducing bacteria (SRB) residing in biofilms on the pipe walls and in the sediments (Hvitved-Jacobsen et al. 2013). A number of factors can influence the production of sulfide in sewer network. The sulfide generation rate is mainly controlled by the factors involving dissolved oxygen (DO), sulfate, readily biodegradable organic matters, pH and temperature. DO in sewage determines the condition (aerobic, anoxic or anaerobic) for corresponding transformation processes of different types of organic matters, and sulfate reduction primarily takes place under anaerobic condition (Gutierrez et al. 2008, Mohanakrishnan et al. 2009b, Shypanski et al. 2018). Sulfate and biodegradable organic matters are two main substrates utilized by SRB for sulfate reduction process. Temperature and pH have an effect on SRB activity and H_2S solubility and dissociation (Sharma et al. 2014). In addition, the total amount of sulfide generated is affected by hydraulic characteristics of sewers, such as the flow rate, flow velocity, pipe diameters, which control the residence time and area-to-volume ratio of sewer pipe and finally affect the level of sulfide formation in sewage. Therefore, large and long gravity pipes with low velocity and force main and wet well of pump station operated intermittently could be potential sites for sulfide formation in sewer system. Identifying the sulfide generation rates at these sites is essential to develop mitigation strategies such as chemical dosing, or optimized pump operation.

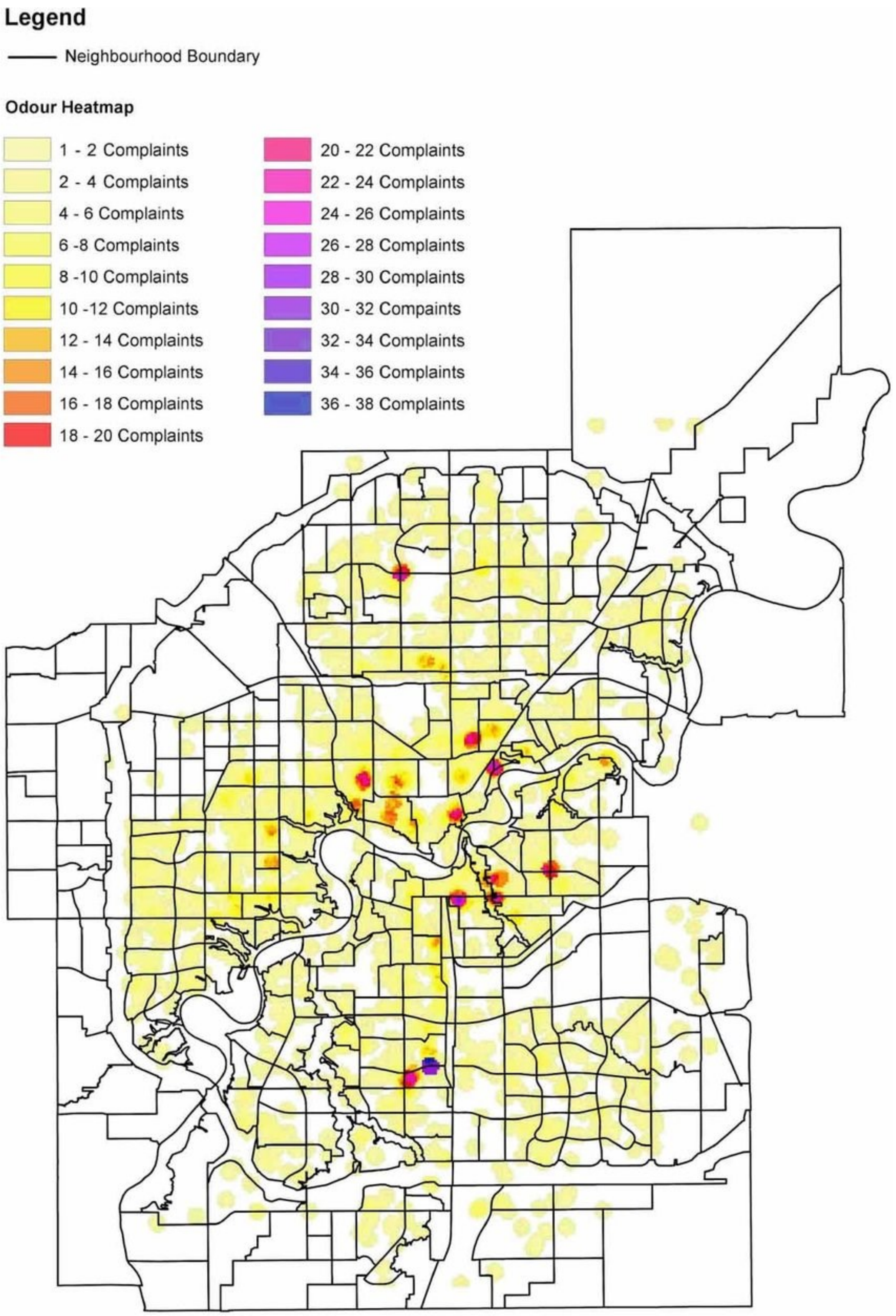


Figure 1-1 Sewer odor density map in Edmonton, Alberta, Canada (Pan et al. 2020)

H_2S is generated under anaerobic conditions when sulfate in domestic wastewater is reduced to sulfide by sulfate-reducing bacteria (SRB). SRB exist in abundance in sewer systems and reside in biofilms on the pipe walls and in the bottom sediment of sewers. The generated H_2S diffuses out of biofilms and sediments, and then dissolves in sewage water. The dissolved H_2S in the water phase diffuses and is released into the air in the sewer headspace above the sewage water level, particularly under turbulent flow found in drop structures. The mass transfer between water and air can be significantly increased by phenomena such as splashing droplets and entrainment of air in the water phase at sewer drops (Ma et al. 2019, Qian et al. 2017). Such structures can therefore promote H_2S stripping and oxygen reaeration (Beceiro et al. 2017, Jung et al. 2017). H_2S in the sewer air will escape via any openings (e.g. manholes, vent pipes, pump stations) to the ground, causing sewer odor complaints. Meanwhile, sulfide oxidizing bacteria (SOB) in the sewer headspace can oxidize H_2S to sulfuric acid, which reacts with corrodible concrete pipe walls and causes pipe corrosion. The effect of drops on H_2S stripping in the sewer system needs to be investigated.

A number of chemical treatments are currently used by the wastewater industries worldwide, to prevent or control sulfide generation in sewer systems (Park et al. 2014, Zhang et al. 2008). These chemicals can be divided into following categories: nitrate that establish anoxic conditions and suppress sulfate reduction, oxidizing chemicals (H_2O_2, NaClO, $KMnO_4$, oxygen/air) that oxidize sulfide, iron salts (Fe^{2+} or Fe^{3+}) that precipitate sulfide, basic chemicals ($Mg(OH)_2$, NaOH, $Ca(OH)_2$) that elevate sewage pH and shift the chemical equilibrium from more volatile H_2S towards more non-volatile species (S^{2-}, HS^-). These strategies effectively prevent sulfide accumulation in wastewater. However, these chemicals are costly and optimizing the dosing strategy to reduce the demand for is crucial.

Addition of nitrate has been proven to be effective to control sulfide production. A few mechanisms of addition of nitrate for sulfide control have been proposed (Zhang et al. 2008): (1) nitrate addition induces the heterotrophic nitrate reduction process and the SRB is supressed by the interspecies competition between heterotrophic nitrate reducing bacteria (hNRB) and SRB for common carbon source (Hubert and Voordouw 2007); (2) addition of nitrate favors biological oxidation of sulfide to elemental sulfur or sulfate by means of nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB) such as *Thiomicrospira denitrificans*, some strains of *Thiomicrospira sp.*, *Thiobacillus sp.*, and *Arcobacter sp.*(Li et al. 2009); (3) the nitrite accumulation as an intermediate of the nitrate reduction process can inhibit SRB activities (Jenneman et al. 1986). However, the exact control mechanism needs to be investigated and the cost-effective nitrate dosing strategy in force main needs to be found out in an effort to save nitrate demand according to the control mechanism.

Sulfide precipitation by addition of iron has been extensively used for sulfide control in sewers. The method relies on the ability of dissolved sulfide to form insoluble iron sulfides, thereby preventing emission of gaseous hydrogen sulfide into the sewer atmosphere. Iron salts may be added either as ferrous or ferric salts. Previous studies on the effect of ferric salt dosing on sulfide was performed in sewer biofilms (Ganigué et al. 2018, Zhang et al. 2009), in which the addition of ferric chloride was found to significantly reduce sulfide production in the anaerobic sewer biofilm of force main by more than 50%. This indicates ferric addition may inhibit the SRB in the biofilm which can potentially reduce the ferric dosage for sulfide control in force main which may be a potential cost-effective chemical for sulfide control in force main.

The Steinhauer and Bonnie Doon areas where numerous odor complaints were reported were selected as the field study areas. The Steinhauer area features large drop structures at the beginning of the trunk and Duggan pump station at the end of the trunk. The Bonnie Doon area has a large number of drops at the trunk and laterals in the sewer network. These are typical sites for potential H_2S problem. In particular, the long force mains of the pump station with long hydraulic retention time (HRT) are the most significant sites for sulfide formation. Therefore, four pump stations which have different length of force mains were selected as representative examples, and the field experimental investigations were performed to evaluate the sulfide formation rates in the force mains of these pump stations. Nitrate that is relatively non-toxic and easy to operate was added at the Big Lake pump station with a high sulfide concentration to control the sulfide generation in this force main. Nitrate and ferric addition were investigated in the lab-scale reactors mimicking force mains prior to practical application in Big Lake pump station.

1.2 Objectives of the research

1) Identifying the hotspots of odor issue and corresponding causes in Steinhauer area including both the trunk and pump station. Modelling the H_2S generation and emission in this area and assessing potential mitigation strategies on H_2S control at this area.

2) Investigating the generation of sulfide in the water phase and transport of H_2S in the air phase along the trunk and laterals in Bonnie Doon area, then evaluating the effect of drop structures on reaeration and H_2S release into air phase in the trunk and finally proposing the control strategies.
3) Quantifying the sulfide generation rates in the lab-scale reactors and investigating the effect of ferric and nitrate on sulfide control in force main and finding out the cost-effective dosing strategies for nitrate addition.
4) Evaluating the sulfide formation rates in the force mains of four pump stations (Blackburne, Kaskitayo, Twin Brooks and Big Lake pump station) and investigating the effectiveness of nitrate dosing to control the sulfide generation in the force main of Big Lake pump station.

1.3 Organization of the thesis

This thesis is organized into eight chapters.

Chapter 2 presents a comprehensive literature review highly relevant to the thesis topic.

Chapter 3 describes an overview of the lab and field studies, including the lab setup, analysis method, measurement procedure and instrumentation.

Chapter 4 presents the field study at Steinhauer area. The reasons for H_2S problem in the area was identified and its generation and emission were simulated by WATS and SeweX models. The application of optimization of Duggan pump station operations as control strategies was modelled.

Chapter 5 presents the field study at Bonnie Doon area. The causes for H_2S problem in this area was identified. The effect of drop structures on H_2S release was estimated and its transportation in the sewer systems was identified.

Chapter 6 presents the lab study in the lab-scale bioreactors imitating force mains. The sulfide generation rates in the bioreactors were quantified. The effect of ferric and nitrate on sulfide control and corresponding control mechanism was investigated. The cost-effective nitrate dosing strategy in force mains was identified.

Chapter 7 presents the field study of evaluation of the sulfide generation rates in the force mains of four pump stations (Blackburne, Kaskitayo, Twin Brooks and Big Lake pump station) and the use of nitrate dosing to control the sulfide generation in the force main of Big Lake pump station.

The sulfide generation rate in the four force mains were identified. The nitrate dosage in the force main of Big Lake pump station was identified and cost-effective nitrate dosing strategy was proposed.

Chapter 8 summarizes the significant outcomes of this work as well as the recommendations for future research.

Chapter 2 Literature Review

2.1 Sulfur cycle and related transformation processes in sewers

As is presented in **Figure 2-1**, a number of processes proceeding in a multiphase system of the biofilm, the sewer deposits, the water phase, and at the air–solid surfaces under aerobic and anaerobic conditions attribute to the sulfur cycle in sewer networks. The sulfide is formed in the biofilm on the pipe wall where is anaerobic and then is transferred into the water phase. If the water is aerobic, the sulfide can be oxidized into sulfite and sulfate. Part of sulfide in the water can form H_2S. Some of H_2S gas could be transferred into the air phase to produce H_2S gas in the headspace of the sewer. H_2S gas could further be emitted into city atmosphere by ventilation to cause the odor complaints from nearby areas. Some of H_2S in the sewer headspace could be absorbed on the crown of the pipe and be oxidized into sulfuric acid which causes the corrosion of the pipe.

1) Aerobic transformation processes

Aerobic condition always occurs in gravity sewer resulting in oxygen transfer between atmosphere and wastewater through reaeration process or the beginning of the pressure pipe where oxygen is brought from upstream pipe. Aeration process often limits the transformations and is therefore a key process. Significant changes in the wastewater quality, the reduction of biodegradable substrate and production of biomass, could happen. Above all is the heterotrophic growth of suspended biomass and growth-related oxygen consumption in both sewage and biofilm phases, which intake the readily degradable organic substance as substrates. Secondly, a fraction of the readily degradable organic is also used by the biomass for maintenance energy requirement. Finally, the aerobic hydrolysis process both in water and biofilm phases could transform particulate substrates to readily biodegradable substrates which is important for the growth of biomass and below microbial transformations.

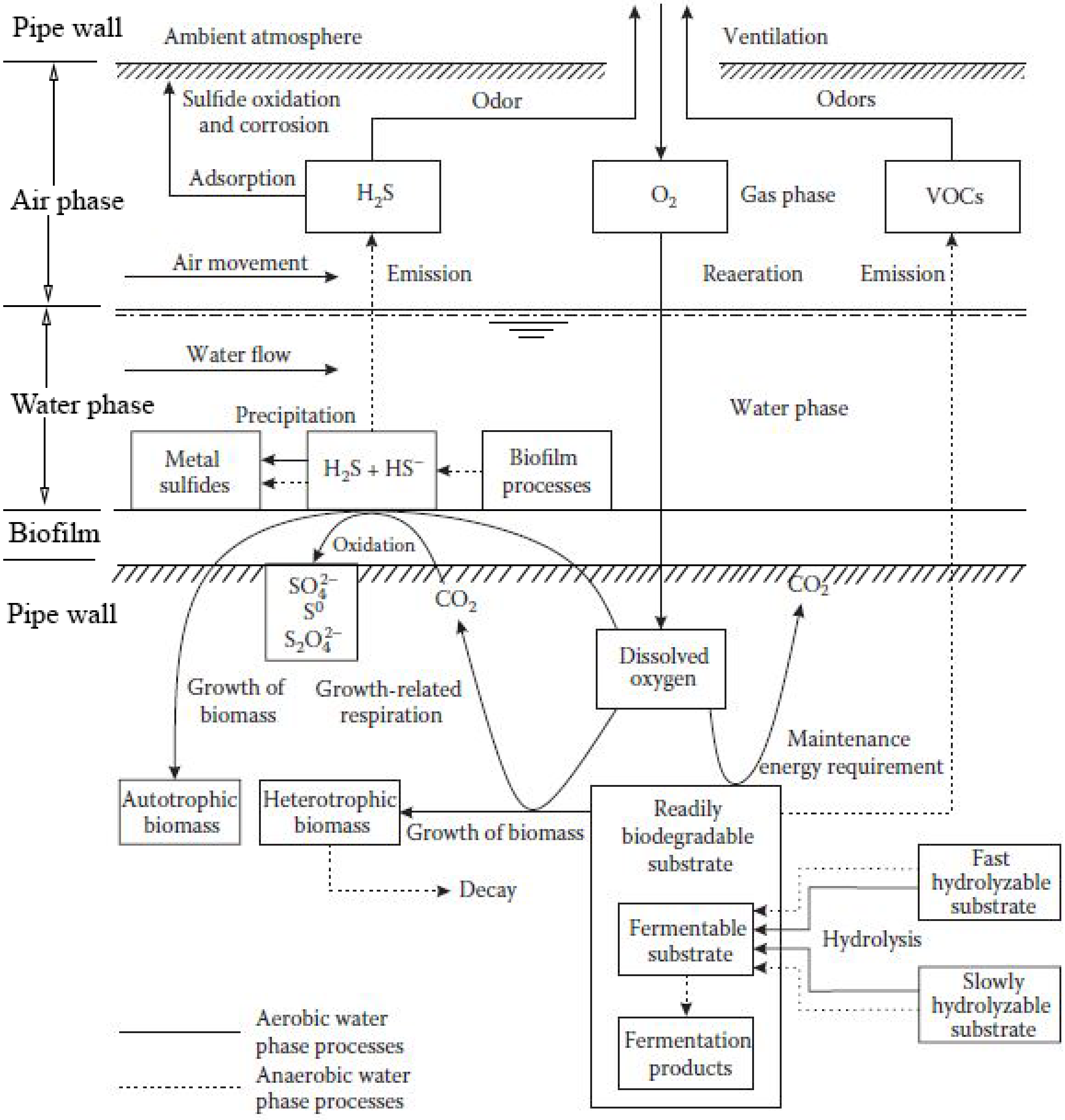


Figure 2-1 Microbial transformations of organic carbon and sulfur in sewers (Hvitved-Jacobsen et al. 2013)

2) Anaerobic transformation processes and sulfide production

Anaerobic conditions exist in pressure mains and long and slow-flowing gravity sewers where dissolved oxygen (DO) concentration in aerobic wastewater is typically fast depleted, depending on the level of the DO aeration and the aerobic respiration rate of the wastewater. Anaerobic microbial-induced transformations of wastewater generally occur when DO is absent. These include the anaerobic hydrolysis process converting slowly biodegradable into readily biodegradable organics like amino acids, sugars, and long chain fatty acids etc. and fermentation process transforming readily degradable substances into alcohols and volatile fatty acids (VFA) in both water and biofilm phase that primarily determine the quality and the changes in the composition of organic matter. Sulfate-reducing bacteria (SRB) can utilize a wide range of organic matter produced by hydrolysis and fermentation processes in wastewater. Besides, the decay of biomass can happen if the biomass is exposed to long-term anaerobic conditions.

Anaerobic conditions are also required for the formation of sulfide. There are basically two types of processes for production of sulfide: sulfate reduction and degradation of sulfur-containing organic matter. The anaerobic, sulfate-reducing bacteria (SRB) grows slowly and, therefore, are subject to being washed out of the sewer system if they occur in the water phase. As a consequence, sulfate reduction primarily takes place in the biofilm covering the submerged sections of sewer walls and the sediments. The generated sulfide then diffuses into the bulk water phase. Since H_2S is a weak acid (pKa1 = 7 and pKa2 = 13 at 25 ℃), sulfide may be present in aqueous solutions in the form of S^{2-}, HS^- and H_2S depending on pH. S^{2-} is generally neglected except at very high pH values, and H_2S is predominant below 7 (Churchill and Elmer 1999). The production of sulfide in sewage is the result of anaerobic respiration of sulfates by SRB. In sewers, multiple species of SRB have been identified, including *Desulfovibrio*, *Desulfobacter*, *Desulfobulbus*, *Desulfomicrobium* and *Desulfotomaculum* (Mohanakrishnan et al. 2009a, Mohanakrishnan et al. 2009b, Okabe et al. 1999).

3) Emission of hydrogen sulfide

The mass transfer of H_2S from the liquid phase is the key process of H_2S emission. Only the molecular form (H_2S) in water phase can be emitted. The emission of hydrogen sulfide from wastewater phase to sewer atmosphere causes the odor problems in manholes, vent pipes and other places in contact with air. The temperature, wastewater quality, sewer characteristics and hydraulics can have been proved to have an effect on the sulfide transfer (Lahav et al. 2006, Yongsiri et al. 2004a, 2005). The turbulence of the wastewater flow created by such as hydraulic drops, line bends, pipe size changes, areas of dynamic slope changes, junction structures, etc. along the sewer enhances the transfer of H_2S from the water phase into the sewer atmosphere. In addition to H_2S, the production of volatile organic compounds (VOCs) under anaerobic conditions also contribute to the odors (Hvitved-Jacobsen et al. 2013).

4) Chemical and biological oxidation of sulfide

The oxidation of sulfide mainly occurs in gravity sewers where oxygen is available. Sulfide produced in deep anaerobic zone of the sewer biofilm or in an upstream part of sewer network transports into the aerobic biofilm and water parts and could be oxidized to elemental sulfur (S^0), thiosulfate ($S_2O_3^{2-}$), sulfite (SO_3^{2-}) and sulfate (SO_4^{2-}), both chemically and biologically (Nielsen et al. 2004, Wilmot et al. 1988). Hydrogen sulfide emitted to the sewer atmosphere and following oxidized are especially significant in sewers where corrosion occurs at the crown as a direct result of sulfuric acid produced by bacterial oxidation of hydrogen sulfide. The genus of *Thyobacillus* bacteria results in the production of sulfuric acid. *Thiobacillus thioparus*, *Thiobacillus denitrificans*, *Thiodoxidans* and *Thiobacillus concretivorus* are reported capable of oxidizing sulfur, sulfide, and thiosulfates (Click and Reed 1975, Meyer and Ledbetter 1970, Santry 1963).

5) Metal precipitation of sulfide

Another process that removes sulfide from the water phase is precipitation by metals. Several metals (primarily iron, zinc, and copper) react readily with dissolved sulfide to produce insoluble metal sulfides (White et al. 1997). Municipal wastewater contains metals, typically iron and zinc, in the range of a few hundred micrograms per litre (Hvitved-Jacobsen et al. 2013, Nielsen et al. 2005b). Metal sulfide precipitation is not therefore mainly of importance when the metal concentration in the wastewater is low. However, inputs of industrial wastewater may significantly add to the metal concentration of the wastewater, thereby increasing the importance of sulfide precipitation.

2.2 Factors affecting the formation of sulfide

The generation of hydrogen sulfide is affected by several factors

1) Dissolved Oxygen (DO)

The presence of DO in wastewater is significant because oxygen will chemically and biologically react with hydrogen sulfide, principally forming sulfate. DO in sewage determines the condition (aerobic or anaerobic) for corresponding transformation processes. Sulfide is only formed under anaerobic condition.

2) Sulfate

Sulfate is typically found in all types of wastewater. At dissolved sulfate concentrations higher than 5–15 $gS.m^{-3}$ and a thin biofilm, the sulfide formation is not limited by sulfate (Nielsen et al. 1998).

3) Biodegradable organic matter

Wastewater with a high BOM rapidly takes up available dissolved oxygen and can create anaerobic conditions that allow the formation of sulfide. BOM could be utilized by the SRB for growth. Several specific organics, e.g., formate, lactate and ethanol, have been identified as particularly suitable substrates for SRB (Nielsen and Hvitved-Jacobsen 1988).

4) Temperature

Temperature is of importance with regard to the presence of H_2S in sewer systems, because it affects the solubility of H_2S, the equilibrium constants for dissociation of H_2S and the rate of biochemical reaction under both aerobic and anaerobic condition. Sulfide generation increases at higher temperatures because of greater bacterial activity and the increased diffusion of nutrient into the slime layer. Because diffusion of substrate into biofilms or sediments is typically limiting sulfide formation, the temperature coefficient is about 1.03 (Nielsen et al. 1998).

5) pH

The pH affects chemical reactions and chemical equilibrium of H_2S. The optimal pH for the growth of the SRB mainly lies between 5.5 and 9 (Gutierrez et al. 2009). Sharma et al. (2014) revealed that the sewer biofilm has the highest sulfate reduction rate at around neutral pH (6.5–7.5), and the activity decreased when pH is higher or lower.

Besides, the generated dissolved sulfide species (H_2S, HS^- and S^{2-}) in stream is controlled by pH. It is observed that H_2S is dominant at lower pH levels and as the pH is increased above 8, HS^- is dominant and when pH is more than 12, S^{2-} becomes prevalent. This is of great importance in terms of the air-water mass transfer characteristics.

6) Area-to-volume ratio of sewer pipe

The actual concentration of dissolved sulfide in the wastewater can be directly related to the total area of the biofilm and sediment, which is determined by the flow area to full sewer volume (A/V) ratio of a sewer system (Hvitved-Jacobsen et al. 2013).

7) Flow velocity

The flow velocity will determine the thickness of the slime layer. At flow velocity higher than 0.8-1.0 m/s, the corresponding biofilm is rather thin, typically 100-300 μm (Gutierrez-Padilla 2007). At higher velocities, the biofilm resistance to mass transfer is reduced due to the rather thin thickness of the diffusional boundary layer. Totally, a high flow velocity will normally reduce the potential for sulfide formation.

8) Anaerobic residence time

Anaerobic residence time will affect the level of sulfide formation in wastewater. The anaerobic residence time should typically exceed 0.5–2 hours before sulfide build up is significant (Hvitved-Jacobsen et al. 2002).

2.3 Sulfur cycle modelling

1) Sulfide generation

In order to predict the sulfide formation in sewer network, considerable attention was paid to the modelling of sulfide formation in the last decades and a few empirical models developed for the evaluation of H_2S generation in force mains were summarized (Carrera et al. 2016). **Table 2-1** includes three to six central parameters characterizing the nature of the sewer system and the most important processes relevant for the occurrence of sulfide. They are established for raw water with high organic matter content (500–1,000 mg.L^{-1} of total COD) and high sulfate concentration (above 4–5 mg S-SO_4^{-2}.L^{-1}) (Equations 2–5). Equation 7 was obtained with field data with mean total COD of 72 mg.L^{-1}, DO 3.3 mg.L^{-1}, and Reynolds number 240,000 (Kitagawa et al. 1998). Equations 3 and 4 account for the origin of the wastewater.

Table 2-1 Empirical equations for sulfide production in force mains

NO.	Flux at 20 °C (F_{sp}(20 °C) ($gS.m^{-2}.h^{-1}$)	References
1	$0.5 \cdot 10^{-3} u \cdot BOD^{0.8} C^{0.4}_{SO_4^{-2},L} \times 1.039^{(T\text{-}20)}$	Thistlethwayte (1972)
2	$0.228 \cdot 10^{-3} COD \times 1.07^{(T\text{-}20)}$	Boon and Lister (1975)
3	$M' \cdot BOD \times 1.07^{(T\text{-}20)}$	Pomeroy and Parkhurst (1977)
4	$\varepsilon \times (COD - 50)^{0.5} \times 1.07^{(T\text{-}20)}$	Hvitved-Jacobsen et al. (1998)
5	$1.52 \times 10^{-2} COD \times \left(\frac{1+0.004D}{D}\right)$	Boon (1995)
6	$0.0025 \times 10^{-3} COD^{2.1} \times 1.07^{(T-20)}$	Elmaleh et al. (1998)

Where ε is the rate constants, u is mean flow velocity ($m.s^{-1}$), M' is coefficient ($gS.gO_2^{-1}.m.h^{-1}$), D is pipe diameter (m), T is wastewater temperature (K or °C).

Compared to pressure mains, the sulfur cycle in partly filled gravity sewer pipes is more complex. Sulfide in water phase results in reduction in three major processes: sulfide transfer to sewer atmosphere, oxidation of sulfide and precipitation. Pomeroy and Parkhurst (1977) firstly proposed dominantly empirical equation for sulfide prediction in gravity pipe, which accounts for two terms: the first term indicated the sulfide buildup and the second term stands for the sinks for sulfide in the water phase that are primarily caused by oxidation in the water phase, emission into the sewer atmosphere, and, also to some extent, precipitation. Until recently, (Hvitved-Jacobsen et al. (1998) developed the Wastewater Aerobic/Anaerobic Transformations in Sewers (WATS) model, which describes both the anaerobic and aerobic processes involving multiple carbon and sulfur species and was a major step forward. Sharma et al. (2014) proposed a dynamic sewer model (SeweX) taking into account the hydraulics and the biochemical transformation processes. The two typical models are more or less the same except the sulfide generation rate which is expressed by using readily biodegradable organic matter. The generation rate is described by following 1/2-order kinetics in WATS model (equation (1.1)), but double Monod kinetics in SeweX model by using VFA and fermentable COD (FCOD) as substrates (equation (1.2) and (1.3)).

$$r = k_{H_2S}(S_F + S_A + X_{S1})^{0.5} \frac{Ko}{So + Ko} \frac{A}{V} \alpha_S^{(T-20)} \tag{1.1}$$

$$r_1 = k_{H_2S} \frac{(S_{FCOD} + S_{VFA})}{K_{Sf} + (S_{FCOD} + S_{VFA})} \frac{S_{O_2}}{K_{O_2} + S_{O_2}} \frac{S_{VFA}}{(S_{FCOD} + S_{VFA})} \frac{A}{V} \alpha_S^{(T-20)} \tag{1.2}$$

$$r_2 = k_{H_2S} \frac{(S_{FCOD} + S_{VFA})}{K_{Sf} + (S_{FCOD} + S_{VFA})} \frac{S_{O_2}}{K_{O_2} + S_{O_2}} \frac{S_{FCOD}}{(S_{FCOD} + S_{VFA})} \frac{A}{V} \alpha_S^{(T-20)} \tag{1.3}$$

where, r is biofilm surface formation rate for sulfide (g S m^{-3} h^{-1}); k_{H2S} is the rate constant for sulfide production (g S (g COD)$^{-0.5}$$m^{-2}h^{-1}$), S_F is fermentable substrate (g COD m^{-3}); S_A is fermentation products (g COD m^{-3}); X_{S1} is fast hydrolysable substrate (g COD m^{-3}); S_{VFA} is volatile fatty acids(g COD m^{-3});T is temperature; K_{O2} is saturation constant for DO (g O_2 m^{-3}); S_{O2} is DO (gO_2 m^{-3}); and A/V is ratio of biofilm area to wastewater volume (m^{-1}).

2) Sulfide emission

Both sulfide and oxygen transfer between air and water could happen in gravity sewer. The two-film theory considering molecular diffusion through stagnant liquid and gas films is the traditional way of understanding mass transfer across the air–water boundary. The following relation is frequently used for describing the emission of H_2S and reaeration of oxygen processes:

J_A=K_{La}($C_{A,L}$-($C_{A,G}$/H_A)) (1.4)

where J_A is volumetric flux rate of component A [moles (total moles)$^{-1}$ s^{-1} m^{-3}]; K_L is overall liquid transfer rate coefficient (m.h^{-1}); $C_{A,L}$ is concentration of species A in liquid phase (mol.L^{-1}); $C_{A,G}$ is concentration of species A in gas phase (mol.L^{-1}); H_A is Henry's law constant for A [atm (mole fraction) $^{-1}$]

The determination of the overall mass transfer coefficient, K_{La} is crucial in this respect. Several factors such as water quality, temperature and interface thickness, may affect K_{La}. The methods to determine the K_{La} for H_2S is to make a connection between the mass transfer coefficients of O_2 and H_2S (equation (1.5)), since measurement of mass transfer coefficient for O_2 is much easier than for H_2S and empirical equations for prediction of the air-water mass transfer coefficient for oxygen have been reported (Jensen 1994, Parkhurst and Pomeroy 1972). n is about 1 in a slow-flowing sewer and that it approaches 0.5 in turbulent conditions (Hvitved-Jacobsen et al. 2013).

The key parameter, mass transfer coefficient for oxygen ($K_La_{,O2}$.), can be determined from a few empirical expressions that have been proposed according to enormous reaeration studies, as shown in **Table 2-2**.

$$(K_{L,H2S}/K_{L,O2})=(D_{L,H2S}/D_{L,O2})^n \qquad (1.5)$$

where, $D_{L,H2S}$ is molecular diffusion coefficient of species H_2S, $D_{L,O2}$ is molecular diffusion coefficient of species O_2.

Table 2-2 Empirical expressions proposed for the determination of the overall oxygen transfer coefficient in gravity sewers

No.	Expressions for K_{La} (20) (h^{-1})	References
1	$0.121(u\cdot s)^{0.408}d_m^{-0.66}$	Krenkel and Orlob (1962)
2	$0.00925u^{0.67}d_m^{-1.85}$	Owens et al. (1964)
3	$0.96(1+0.17Fr^2)(s\cdot u)^{3/8}d_m^{-1}$	Parkhurst and Pomeroy (1972)
4	$B\cdot u\cdot s$	Tsivoglou and Neal (1976)
5	$0.4u\left(\frac{d_m}{R}\right)^{0.613}d_m^{-1}$	Taghizadeh-Nasser (1986)
6	$0.86(1+0.20Fr^2)(s\cdot u)^{3/8}d_m^{-1}$	Jensen (1994)
7	$0.66(1+0.20Fr^2)(s\cdot u)^{3/8}d_m^{-1}$	Huisman et al. (1999)

where, $Fr= u(gd_m)^{-0.5}$ is the Froude number; u is the mean velocity of flow (m s^{-1}); g is the gravitational acceleration (m s^{-2}); d is the hydraulic mean depth (m); s is the slope; B is the coefficient given as a function of water quality and intensity of mixing.

In addition to the two-film theory, other researchers investigated the relation between mass transfer and hydrodynamic parameters for the determination of the H_2S emission rate by empirical or theoretical equations. Pomeroy and Parkhurst (1977) proposed equation (1.6) for the prediction of sulfide formation and the second term represents the sulfide transfer in the sewer atmosphere from liquid phase. Matos and De Sousa (1992) also proposed an equation to calculate the flux of H_2S from steam surface in sewer systems, which includes the turbulence intensity. Yongsiri et al. (2004a, 2004b, 2005) did a series of lab experiments on how parameters affected H_2S mass transfer. Equation (1.7) was proposed and then was applied in partly filled gravity sewer. A similar equation (1.8) was proposed by Lahav et al. (2006). The effect of temperature, wastewater quality (β), sewer characteristics and hydraulics (s, u, dm) was considered in these two equations.

$$Fsp = M'BOD \cdot 1.07^{(T-20)} - N(su)^{3/8} d_m^{-1} R C_{S^{2-},L} \tag{1.6}$$

where, Fsp is biofilm area net formation rate of sulfide (g S m^{-2} h^{-1}); A/V is area/volume ratio (m^{-1}); $C_{S^{2-},L}$ is sulfide concentration (g S m^{-3}); M′ is empirical rate constant; T is temperature (°C); N is empirical sulfide loss coefficient; s is slope (m m^{-1}); u is flow velocity (m s^{-1}); d_m is hydraulic mean depth of the water phase (m); R is hydraulic radius (m); P is wetted pipe-wall perimeter (m); b is pipe width at the water surface (m).

$$\frac{dC_{H_2S,L}}{dt} = \alpha(0.86f) \cdot \left[\frac{0.86 + (1 + 0.2F^2 (su)^{3/8}}{d_m} \right] \times \left[C_{H_2S,L} - \frac{\beta C_{H_2S,G}}{fH'_{H_2S}} \right] 1.034^{(T-20)} \tag{1.7}$$

where, $C_{H2S,L}$ is dissolved sulfide concentration in water phase (g S m^{-3}); $C_{H2S,G}$ is H_2S concentration in air phase (g S m^{-3}); f is $H_2S(aq)$ fraction relative to total sulfide; $H_{H2S,G}$ is nondimensional Henry’s law constant for H_2S; α is correction factor for overall mass-transfer coefficient in wastewater; β is correction factor for saturation concentration in wastewater; F is Froude number: $u/\sqrt{gD_m}$,where g is gravitational acceleration and D_m is hydraulic mean depth.

$$\frac{dC_s}{dt} = 8 \cdot 10^{-7} \sqrt{\frac{\gamma su}{\mu}} \frac{w}{A_{CS}} \times a \times 1.024^{(T-20)} \left(\frac{C_S}{1 + 10^{pH-pKa1} + 10^{2pH-pKa1-pKa2}} - P_{H_2S} K_H \right) \tag{1.8}$$

where, C_S is the total sulfide concentration in the aqueous phase (mg/L); w is the flow surface width (m); A_{cs} is the cross-sectional area (m^2); K_H is the Henry's constant (mol L^{-1} atm^{-1}); P_{H2S} is the partial pressure of $H_2S(g)$ in the sewer atmosphere (atm).

The air–water mass transfer at sewer drops is complex because of the turbulence at drop structures that considerably increases the air–water oxygen transfer (reaeration). The construction of the drop structure will, for example, affect reaeration and stripping. Furthermore, it has been shown that the mass transfer at drop structures is the largest at low flow rates because of large residence times and significant interfacial area (Chanson 2004). More research is still required in order to obtain a better description of mass transfer in hydraulic structures (e.g. gravity sewers, junctions or waterfalls).

3) Sulfide oxidation

Numerous researches have contributed to the sulfide oxidation kinetics both in water and biofilm phase. The results demonstrated that sulfide was oxidized both chemically and biologically in water phase that can be formulated by power functions. Nielsen et al. (2003, 2006) report typical values of n1 (both n1c and n1b) ranging from 0.8 to 1.0 (equation (1.9)) and those for n2 (both n2c and n2b) ranging from 0.1 to 0.2 for chemical oxidation (equation (1.10)). Unlike in the water phase, the oxidation on biofilm is mainly biological oxidation following the half-order kinetics n (equation (1.11)) (Nielsen et al. 2005a).

$$r_{S(-II),che\text{-}ox,w} = k_{S(-II)chem\text{-}oxi}\left(S_{O_2}\right)^{n1c}\left(S_{S(-II)}\right)^{n2c} \tag{1.9}$$

$$r_{S(-II),bio\text{-}ox,w} = k_{S(-II)bio\text{-}oxi}\left(S_{O_2}\right)^{n1b}\left(S_{S(-II)}\right)^{n2b} \tag{1.10}$$

$$r_{S(-II),ox,f} = k_{S(-II)bio\text{-}oxi} S_{O_2}^{0.5} S_{S(-II)}^{0.5} \frac{A}{V} \tag{1.11}$$

where, $r_{S(-II)}$ is rate of sulfide oxidation (g S m^{-3} day^{-1}); $k_{S(-II)}$ is rate constant (unit depends on the values of n1 and n2); S_S is concentration of dissolved sulfide (g m^{-3}); S_{O2} is concentration of DO (g m^{-3}); n = n1 + n2 = reaction order (unit dependent on n1 and n2).

The H_2S gas in the gravity sewer also can be adsorbed and oxidized on the sewer walls which causes the corrosion of the pipe. This reaction rate is described by the following function (Æsøy et al. 2002).

$$r_{H_2S} = -k_{S(-II)gas,bio\text{-}oxi} \frac{pH_2S}{K_{pH_2S} + pH_2S} \frac{A_c}{V_g} \tag{1.12}$$

where, ks(-II) is the estimated rate constant for adsorption and oxidation of H_2S to the moist concrete surface (g S m^{-2} h^{-1}). The ratio, A_c/V_g, represents the proportion of concrete surface to volume of sewer atmosphere.

4) Aerobic and Anaerobic transformation of organic matters

The transformation of the different forms of organics: particulate organics, soluble organics, fermentable substrates and fermentation products, is important for the formation of sulfide, since the SRB consume the readily biodegradable organics as electron donor for respiration. The double-Monod equation is used for description of the most processes for organic transformation. Under the aerobic, such water quality changes involve consumption of readily biodegradable organics by biomass in water phase described as equation (1.13), which is demonstrated by Bjerre et al. (1995, 1998) based on lab and field experiments with wastewater under gravity sewer conditions and by biofilm on the pipe wall which is formulated by the half-order kinetics for DO biofilm surface removal rates (equation (1.14)) (Nielsen et al. 1998). In addition to growth, the maintenance energy requirement, nongrowth-related consumption of substrate of suspended biomass, also needs to be considered. This process is formulated as equation (1.15). On the other hand, the aerobic hydrolysis of particulate substrates to biodegradable organic matters in both biofilm and water phase is described by following concept of Activated Sludge Model No. 1 (equation (1.16)) (Henze et al. 1987).

$$r_{grw} = \mu_H \frac{(S_{FCOD} + S_{VFA})}{K_{Sw} + (S_{FCOD} + S_{VFA})} \frac{S_{O_2}}{K_{O_2} + S_{O_2}} X_{Bw} \alpha_w^{(T-20)} \qquad (1.13)$$

where, r_{grw} is growth rate of heterotrophic biomass in suspension (g COD m^{-3} day^{-1}); μ_H is maximum specific growth rate (day^{-1}); S_{FCOD} is readily (fermentable) biodegradable substrate (g COD m^{-3}); S_{VFA} is volatile fatty acids (g COD m^{-3}); K_{Sw} is saturation constant for readily biodegradable substrate (g COD m^{-3}); S_{O2} is DO concentration in bulk water phase (g O_2 m^{-3}); K_{O2} is saturation constant for DO (g O_2 m^{-3}); X_{Bw} is heterotrophic biomass concentration in the water phase (g COD m^{-3}); α_w is temperature coefficient for the water phase process;

$$r_{grf} = k_{1/2} \sqrt{S_{O_2}} \frac{Y_{Hf}}{(1 - Y_{Hf})} \frac{A}{V} \frac{(S_{FCOD} + S_{VFA})}{K_{Sf} + (S_{FCOD} + S_{VFA})} \alpha_f^{(T-20)} \qquad (1.14)$$

where, r_{grf} is growth rate of heterotrophic biomass in a biofilm (g COD m^{-3} day^{-1}), $k_{1/2}$ is 1/2-order rate constant (g $O_2^{0.5}m^{-0.5}$ day^{-1}); Y_{Hf} is biofilm yield constant [g COD, biomass (g COD, substrate)–1]; K_{Sf} is saturation constant for readily biodegradable substrate in biofilm (g COD m^{-}

^{3}); A/V is wetted sewer pipe surface area divided by the water volume (m^{-1}); α_f is temperature coefficient for the biofilm process ; T is temperature (°C).

$$r_{maint} = -q_m \frac{S_{O_2}}{K_{O_2} + S_{O_2}} X_{Bw} \alpha_w^{(T-20)} \tag{1.15}$$

$$r_{hydr} = -k_h \frac{X_{SBCOD} + X_{Bw}}{K_X + X_{SBCOD} + X_{Bw}} \frac{S_{O_2}}{K_{O_2} + S_{O_2}} (X_{Bw} + \varepsilon X_{Bf} \frac{A}{V}) \alpha_w^{(T-20)} \tag{1.16}$$

where, r_{hydr} is rate of anaerobic hydrolysis (g COD m^{-3} day^{-1}); k_h is hydrolysis rate constant (day^{-1}); X_{SBCOD} is slowly biodegradable COD(g COD m^{-3}); X_{Bw} is heterotrophic biomass concentration in the water phase (g COD m^{-3}); ε is relative efficiency constant for hydrolysis of the biofilm biomass; X_{Bf} is heterotrophic biomass in the biofilm (g COD m^{-2}); K_{O2} is saturation constant for DO (g O_2 m^{-3}); S_{O2} is DO concentration in bulk water phase (g O_2 m^{-3}).

The anaerobic hydrolysis and fermentation process resulting in the increase of the biodegradable substrates. The kinetic of anaerobic hydrolysis is similar to aerobic hydrolysis, as described by equation (1.17). The fermentation is formulated based on Monod kinetics (equation (1.18)).

$$r_{hydr,ana} = -\eta_{h,ana} = k_h \frac{X_{SBCOD} + X_{Bw}}{K_X + X_{SBCOD} + X_{Bw}} \frac{S_{O_2}}{K_{O_2} + S_{O_2}} (X_{Bw} + \varepsilon X_{Bf} \frac{A}{V}) \alpha^{(T-20)} \tag{1.17}$$

where, $r_{hydr,ana}$ is rate of anaerobic hydrolysis (g COD m^{-3} day^{-1}); $\eta_{h,ana}$ is efficiency constant for anaerobic hydrolysis relative to aerobic hydrolysis; k_h is hydrolysis rate constant (day^{-1}); X_{SBCOD} is slowly biodegradable COD (g COD m^{-3}) ; X_{Bw} is heterotrophic biomass concentration in the water phase (g COD m^{-3}); ε is relative efficiency constant for hydrolysis of the biofilm biomass; X_{Bf} is heterotrophic biomass in the biofilm (g COD m^{-2}); K_{O2} is saturation constant for DO (g O_2 m^{-3}); S_{O2} is DO concentration in bulk water phase (g O_2 m^{-3}); α is temperature coefficient .

$$r_{fer} = -q_{fer} \frac{S_{FCOD}}{K_{fer} + S_{FCOD}} \frac{S_{O_2}}{K_{O_2} + S_{O_2}} (X_{Bw} + \varepsilon X_{Bf} \frac{A}{V}) \alpha^{(T-20)} \tag{1.18}$$

where, r_{fer} is fermentation rate (g COD m^{-3} day^{-1}); q_{ferm} is fermentation rate constant (day^{-1}); S_{FCOD} is fermentable substrate (g COD m^{-3}); K_{fer} is saturation constant for fermentation (g COD m^{-3}).

2.4 Hydrogen sulfide control and mitigation methods in sewer systems

According to H_2S generation processes in sewer, the available methods used for odor control can be simply illustrated in **Figure 2-2**.

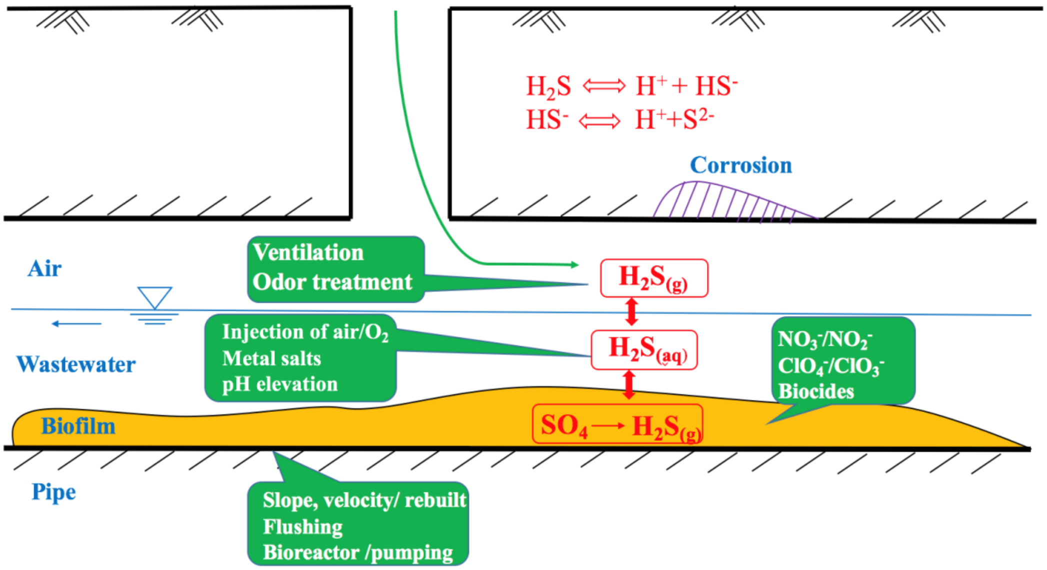


Figure 2-2 Odor processes and control methods in a sewer pipe

1) Gas phase technologies

Ventilation and odor treatment**:** ventilation (natural and forced) can be applied for the elimination of gaseous H_2S in sewer atmosphere. The ventilation system is considered to use either slotted manhole covers or ventilation stacks or mechanical means (fan) in an attempt to release odors and maintain adequate oxygen levels within the sewer system by air flow. The released odor could be further removed by technologies including chemical scrubbing, activated carbon adsorption, biofiltration and biotrickling filtration for odor abatement (Herrygers et al. 2000). This technology is effective in places where sulfides are generated locally. But beds tend to degrade with time and require close maintenance.

2) Liquid phase technologies

Air or oxygen injection: the addition of air or oxygen to create aerobic conditions to oxidize sulfide to sulfate (SO_4^{2-}) has been widely used. Oxygen injection is often an attractive option as it is relatively inexpensive and targets rising mains, where SRB activity is highest. Air or oxygen injection is mainly used in medium and high flow systems (pipes with average dry weather flow (ADWF) between 1 and 5 ML/d) with large pipes (diameters larger than 0.3 m) (Ganigue et al. 2011). Oxygen injection to sewers can further stimulated SRB growth and increased SRB activity in downstream biofilms due to increased availability of sulfate at these locations as the result of aerobic conditions upstream. Oxygen does not have long-lasting inhibitory effects on sulfide production by SRB, with SRB activity resuming immediately after oxygen depletion (Gutierrez et al. 2008).

Precipitation of formed sulfide with metal salts: another widely used strategy for H_2S mitigation in sewage is the addition of metal salt to remove dissolved sulfide from the wastewater, thereby decreasing the amount of sulfide available for release to the sewer atmosphere. Because of its effectiveness in controlling the dissolved sulfide concentration, iron has been widely used to control sulfide buildup in sewer networks (Nielsen et al. 2005b). Iron salts of chloride, sulfate or nitrate are being added to wastewater either in ferric or ferrous forms. Ferrous ions (Fe^{2+}) precipitate sulfide by forming highly insoluble metallic sulfide precipitates. On the other hand, Ferric ions (Fe^{3+}) oxidize sulfide to elemental sulfur while being reduced into (Fe^{2+}), which precipitates with sulfide to form ferrous sulfide precipitants. Field studies indicated that a mixture of ferrous and ferric iron salts is more effective than either salt alone in controlling the dissolved sulfide concentration (Padival et al. 1995). Iron salts are preferentially used in medium and large systems (flows larger than 1 ML/d). But, addition of iron salts may add undesirable anions to the wastewater as well as cause unsolicited flocculation and settling in the sewer. Besides, it may precipitate with phosphorous compounds thus demand will increase beyond stoichiometry.

pH elevation: pH elevation is another strategy to control H_2S in sewer. Elevation of pH above 8.5 by addition of strong base will shift the equilibrium of the dissolved sulfide towards the non-volatile species (S^{2-}, HS^{-}). Magnesium hydroxide can be applied to sewer networks for this purpose. Continuous addition of magnesium hydroxide can achieve pH increase to 8.5-9. The maximum pH achievable by $Mg(OH)_2$ dosing is approximately 9.0 due to its limited solubility in water, a level that would not endanger the performance of the downstream biological wastewater treatment plants. But it is effective only locally, since pH is bound to decrease. An alternative sulfide control strategy also involving manipulating pH is the pH shock strategy by adding sodium hydroxide. Sodium hydroxide is only added intermittently and for a short period (hours) to reach pH 11-13, which can inactivate SRB in sewer biofilms thus leading to suppressed sulfide production. The regrowth of the sulfide-producing bacteria starts 1-3 days after a pH shock and is completed within 5-7 days after the pH shock (Gutierrez et al. 2014). Compared to pH elevation using $Mg(OH)_2$, the pH shock strategy has several disadvantages. The wastewater of the high pH section has to be isolated at the wastewater treatment plant (WWTP). In addition, the high pH can also cause other problems like carbonate precipitation, sludge generation and ammonia gas release (Hvitved-Jacobsen et al. 2013). But it is relatively cheap as dosing is not continuous.

Addition of oxidizing chemicals: when hydrogen peroxide (H_2O_2) or ozone (O_3) is added to wastewater, it oxidizes dissolved sulfide and decomposes oxygen, thus keeping conditions aerobic. Potassium permanganate ($KMnO_4$) and Chlorine will oxidize sulfide to sulfate.

General disadvantages for all agents are that they are toxic to transportation and maintenance personnel and expensive to purchase and handle. The effectiveness of chemical oxidizers is frequently low because of their reactions with other components in sewage

3) Biofilm phase technologies

Addition of nitrate: adding nitrate (NO_3^-) to an anaerobic wastewater system will establish anoxic conditions due to the increased presence of nitrate as an electron acceptor for the microbial process. In fact, nitrate addition stimulated the activity of nitrate reducing, sulfide oxidizing bacteria (NR-SOB) that appeared to be primarily responsible for the prevention of sulfide build up in the wastewater in the presence of nitrate (Gutierrez et al. 2014). NR-SOB oxidize sulfide biologically coupled with the reduction of nitrate thus achieving sulfide control. Nitrate is an expensive chemical, usually dosed as $NaNO_3$ or $Ca(NO_3)_2$. It is also preferentially used in small systems (flows lower than 1 ML/d). Longer retention time is required for sulfide oxidation. Addition of nitrate is a preventative measure rather than a sulfide removal method. Hydrogen sulfide will regenerate upon cessation of NO_3^- dosage. It has possible negative effect on wastewater treatment plant because of nitrate load and can increase sulfate concentrations in downstream sections of sewer networks, leading to the enrichment of SRB populations in these region

Inhabitation of SRB populations with biocides: the use of microbial inhibitors such as formaldehyde (Zhang et al. 2008) or nitrite (Jiang et al. 2009) have also been proposed, successfully demonstrated at laboratory and field-scale systems and in the near future may represent a more cost-effective method for sulfide mitigation. Free nitrous acid (FNA) was recently revealed to be a strong biocide for microbes in anaerobic biofilm, achieving approximately 1-log (90%) inactivation at a concentration of 0.2–0.3 mg HNO_2-N/L with an exposure time longer than 6 h. Particularly, hydrogen peroxide, in combination with FNA, was found to enhance the microbial inactivation by 1-log, in comparison with FNA dosing alone (Jiang and Yuan 2013).

4) Technologies for pipes

Mechanical cleaning to remove sewer biofilms: a complete way for elimination of sulfide generation is to flush the pipe wall to remove both biofilm on the pipe wall and the sediments in the bottom of pipe at regular intervals. So there are no sites for sulfide generation. But this method can be expensive.

Improve the hydraulic design: the hydraulic mean depth, the hydraulic radius, the wastewater flow velocity, and the slope of the sewer pipe are important factors, which can affect the reaeration rate and thereby affect anaerobic-related problems. Furthermore, if sewage flows too slowly, sediment within the sewage settles out and deposits within the pipe. These deposits provide an ideal environment for an anaerobic slime layer where hydrogen sulfide is produced. A slow wastewater flow velocity may lead to an increase of the biofilm thickness and the occurrence of a permanent sediment layer. It is essential to increase the pipe slope to increase the velocity of the flows.

There are turbulent cascades or waterfalls within sewer system, where sewers contain significant odorous compounds from upstream anaerobic activity, these odorants are largely off-gassed under these cascade-waterfall conditions. This creates strong foul air concentrations within these structures and add to the odor level emitted from them at various locations. Hydraulic structure modification would be required to minimize this turbulence and allow for more quiescent flow regimes.

2.5 Effect of chemical dosing on microbial community of biofilm

The microbial community exists in biofilms on the drainage pipe wall plays a fundamental role in the transformations of sulfur, especially SRB, sulfate oxidation bacteria (SOB) and methanogenic archaea (MA). Mohanakrishnan et al. (2009b) revealed that the biofilm structure and activity varied considerably along the length of rising mains, both with regard to general bacterial populations and sulfate reducers. Sun et al. (2014) found that sulfide was mainly produced in the outer layer of the biofilm, between the depths of 0 and 300 μm. In contrast, MA mainly inhabited the inner layer of the biofilm. Auguet et al. (2015a) demonstrated that H_2S emission was notably high during early stages of biofilm development, CH4 emissions increased after biofilm maturation, coinciding with an establishment of a methanogenic community better adapted to sewer conditions. In sewers, multiple species of SRB have been identified, including *Desulfovibrio, Desulfobacter, Desulfobulbus, Desulfomicrobium and Desulfotomaculum* (Mohanakrishnan et al. 2009a, Mohanakrishnan et al. 2009b, Okabe et al. 1999). Detection of the presence of SRB may provide one way of diagnosing potential problematic areas as well as giving early opportunity for implementing the most promising remediation techniques.

Addition of chemical is an important and effective way to control sulfide, which has impacts on the composition of microbial biofilms communities. Nitrate and nitrite are common chemicals dosed in sewer for reduction of sulfide and methane. Mohanakrishnan et al. (2009a) studied the impact of nitrate addition on the microbial activities of anaerobic sewer biofilm and found nitrate was not toxic or inhibitory to SRB activity and did not affect the dominant SRB populations in the biofilm. The molecular methods were used to investigate the impacts and microbial activities related to the nitrate and nitrite addition (Auguet et al. 2015b). As the nitrate was added, a complete abatement of H_2S generated, with a fraction transformed to elemental sulfur (S^0) and methane discharged was reduced to 50%. Meanwhile, an increase of microorganisms of the genera *Simplicispira, Comamonas, Azonexus* and *Thauera* was detected. For addition of nitrite, both sulfide and methane emission reduced. The decrease of sequences affiliated with classes *Clostridia, Deltaproteobacteria* and *Synergistia* and the increase of sequences affiliated with classes *Betaproteobacteria* and *Gammaproteobacteria* during nitrite addition showed an important role of these groups in nitrite, methane and sulfur species transformation were observed.

Chapter 3 Materials and Methods

3.1 Field work overview

The Steinhauer and Bonnie Doon areas where numerous odor complaints were reported were selected as the field study areas. The Steinhauer area characterizes the large drop structures at the beginning of the trunk and Duggan pump station at the end of the trunk. The Bonnie Doon area has lots of drops at both the trunk and laterals in the sewer networks. These are typical sites for potential H_2S problem. In particular, the long force mains of the pump station with long hydraulic retention time (HRT) are the most significant site for sulfide formation. Several pump stations including the Blackburne, Kaskitayo, Twin Brooks and Big Lake pump stations were investigated to evaluate the sulfide formation in the force mains. Finally, the force main at Big Lake pump station with the most severe H_2S problem was chosen to apply nitrate dosing strategy to control the odor problem associated with sulfide.

The field monitoring and sampling program in the trunk was conducted in May 2017 in Steinhauer area. Sewage samples were grabbed from the trunk line on May 15^{th} and 16^{th}, 2017 at downstream manhole to upstream manhole. At Duggan pump station, two rounds of field work were performed. Sewage samples were collected hourly for 24 hours on May 15th, 2017. H_2S gas was continuously monitored in the air space of the pump well by Odalog for one week. Before this field work, a preliminary field work was also performed at the pump station on December 15th, 2016. The H_2S gas concentration and air pressure at the sewer headspace of the trunk and laterals were monitored for one month.

The field work was also conducted in Bonnie Doon area, a neighbourhood in south-central Edmonton, Alberta, Canada. Two rounds of field monitoring were implemented in the area in 2017 and 2018. Water samples were obtained from the manholes in the trunk and all the laterals on Nov. 29 – 30, 2017 and Sept. 27^{th}, 2018. The H_2S gas concentration and air pressure at the sewer headspace of the trunk and laterals were monitored for one month.

Four pump stations (Blackburne Pump Station, Kaskitayo Pump Station, Twin Brooks Pump Station and Big Lake Pump Station) were monitored in the field. One sampling campaign was conducted at Blackburne, Kaskitayo and Twin Brooks pump stations. The monitoring was conducted on Oct. 30^{th}, 2018. The water samples were taken from pump wells and force main discharge manhole of these three pump stations once. The Odalogs were installed at pump wells and force main discharge manholes of these three pump stations for continuously monitoring the H_2S gas in the air phase for two weeks. Five sampling campaign were carried out in the Big Lake pump station. Prior to the nitrate dosing, two measurement campaigns were conducted to monitor the sulfide generation in the force main on Sept. 08th and Sept. 11th, 2020 for 5 hours and 24 hours, respectively. Then 48~54% calcium nitrate ($Ca(NO_3)_2$) (by weight) solution was continuously injected into the wet well by pump at a constant flow rate above the inlet of the wet well inflow starting from Sept. 12th, 2020. Two monitoring campaigns were conducted during dosing phase on Sept. 25th and Oct. 09th, 2020 for 24 hours at 7 L/h and 15 L/h dosing rate, respectively. The dosage was halted on Oct.10th, 2020 and the last campaign was performed on Oct.16th, 2020 during recovery phase. Each campaign involved sampling wastewater by pumping event at both the wet well and discharge manhole. Odalogs were installed at both wet well and discharge manhole to continuously detect the H_2S gas.

The H_2S gas concentration was continuously monitored by Odalog (App-Tek, Queensland, Australia) and air pressure was also continuously monitored by SmartReader (ACR Systems, Vancouver, British Columbia, Canada) in the field. At each sampling location, DO, pH, ORP and temperature of sewage were firstly measured on site by portable pH meter, DO and ORP meter. Sewage samples for determination of total and dissolved sulfide (TS and DS) were preserved on site. The samples for total sulfide determination were preserved by addition of 2N zinc acetate (2 mL/L) and 6N sodium hydroxide (2 mL/L) solution to the sampling bottles before each sampling. Dissolved and suspended sulfide were separated by addition of aluminum chloride to the sewage samples, which produced an aluminum hydroxide floc that trapped suspended sulfide. The sample was then settled, and the dissolved sulfide concentration of the clear supernatant was taken which was preserve with 2N zinc acetate. All the field sewage samples were stored in an ice box during transportation to the laboratory and were kept refrigerated till analysis was done.

3.2 Lab setup

As shown in **Figure 3-1,** four identical airtight laboratory scale reactors (R1, R2, R3 and R4) were running in parallel which mimicked force mains at pump stations. These reactors were made of Plexiglas. Each cylindrical reactor had a valid volume of 3.5 L with a diameter of 15.5 cm and height of 20 cm. Four rods were placed inside each reactor and 40 plastic Kaldnes carriers (circular, 2.5 cm diameter, 1.0 cm height) were fixed on the rods in each reactor, which had a total surface area of 1178 cm^2. Biofilm grew on the walls of the reactors and on the plastic Kaldnes carriers and each reactor had biofilm area of 2529 cm^2. The biofilm area/volume (A/V) ratio was 72 m^2/m^3. The use of carriers allowed easy retrieval of biofilm sample for microbial community analysis. The sediment collected from Edmonton's sewer system was inoculated into the reactors at the start-up and synthetic water imitating the wastewater quality in Edmonton's sewer system was used in this experiment (**Table 3-1**). Its composition was based on the recommendations of Liang et al. (2016) (**Table 3-2**). The reactors were fed with synthetic water through a peristaltic pump under the room temperature (20 ℃) from a storage tank. The reactors were completely sealed and covered with aluminum foil to avoid exposing the sewage and biofilm to light. Each reactor lid was equipped with a small container of 70 mL filled with the same wastewater as in the reactors, so as to prevent any vacuum and air entry during wastewater displacement. Each reactor consisted of two pumping events per day and resulted in a retention time of 12 h. Every feed pumping event lasted 6 min, delivering 5.0 L fresh sewage into the reactor to replace the whole volume of the wastewater in the reactor. To ensure homogeneous distribution in reactors, gentle mixing (100 rpm) was provided with magnetic stirrers (Fisherbrand, Canada) under each reactor.

Table 3-1 Synthetic water characteristics

NH_4-N (mg/L)	TP (mg/L)	COD (mg/L)	Sulfate (mg S/L)	pH	ORP (mV)	T (℃)
40±4	20±2	300±20	50±5	7±0.2	-150±10	20±1

Table 3-2 Composition of basic wastewater used in the reactors

Substance	Concentration	Trace element	Concentration
Glucose	129 mg/L	H_3BO_3	0.4 mg/L
Sodium acetate	164 mg/L	$CuSO_4\cdot 5H_2O$	0.1 mg/L
Yeast extract	117 mg/L	KI	0.16 mg/L
NH_4Cl	156 mg/L	$MnSO_4\cdot H_2O$	0.5 mg/L
Na_2SO_4	150 mg/L	$ZnSO_4\cdot 7H_2O$	0.3 mg/L

KH_2PO_4	26 mg/L	$CoCl_2 \cdot 6H_2O$	0.4 mg/L
K_2HPO_4	85 mg/L		
Cystine	700 µg/L		
Methionine	900 µg/L		

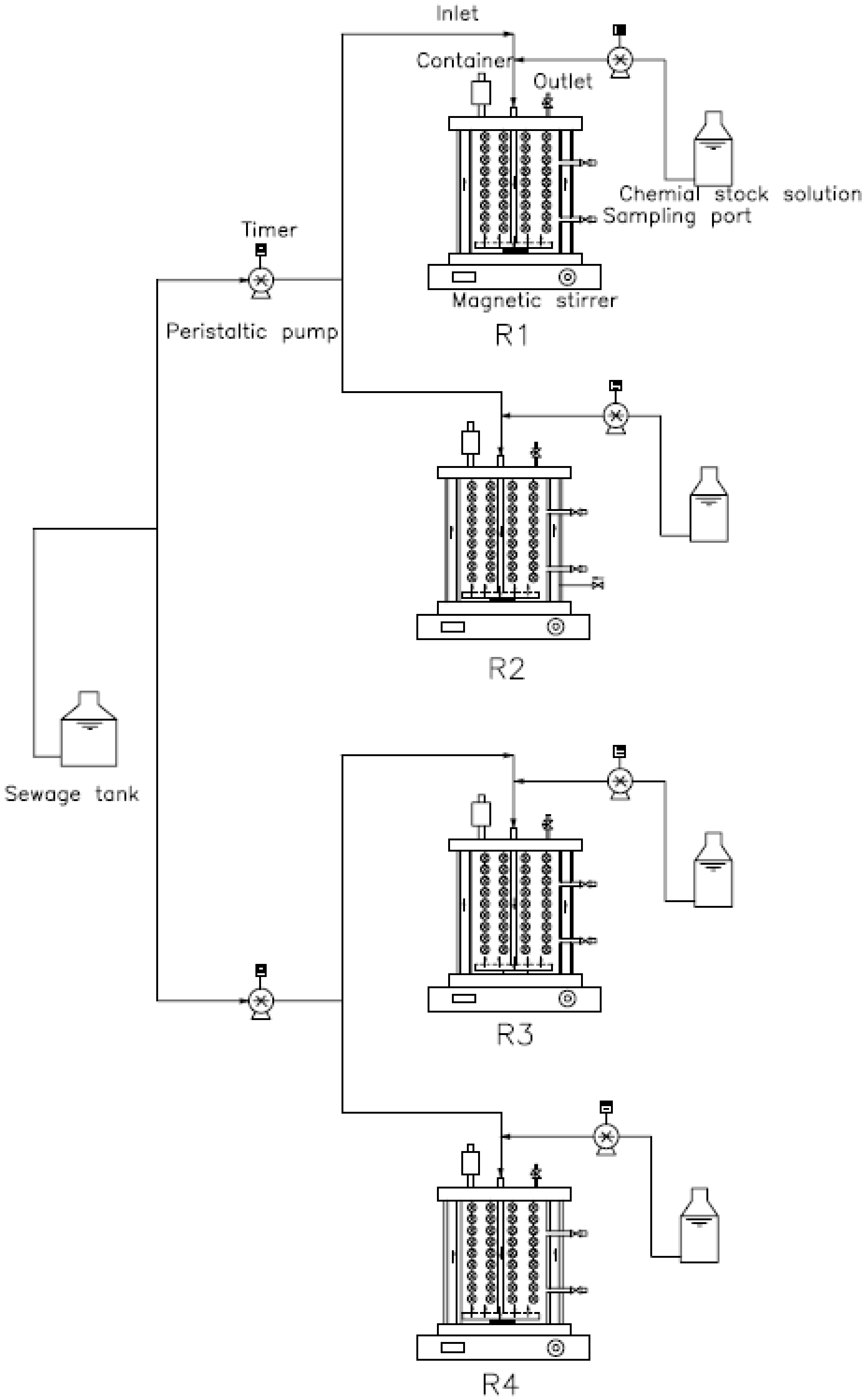


Figure 3-1 Schematic of the lab-scale reactors

The reactors were operated for eight months to develop mature anaerobic biofilm and establish steady-state performance in terms of sulfide production (phase 1). Sulfide was produced due to the biofilm activities. Sulfide generation rates in R1, R2, R3 and R4 were measured at the end of phase 1. Then ferric chloride and sodium nitrate started being added in the R1 and R2 for three months and 6 months (three months for each nitrate dosing strategy), respectively (phase 2). During the dosing phase, ferric was injected by the peristaltic pump from ferric stock solution right after each 6-minute pumping event and then stayed in the R1 for 12 h. Nitrate was added into the R2 with two different strategies. The first strategy was to add it at the beginning of each pump cycle (0 h). Like ferric addition, nitrate was injected from nitrate stock solution right after each 6-minute pumping event and then stayed in the R2 for 12 h. The nitrate was added to prevent the formation of the sulfide along the 12 h pump cycle for the first nitrate dosing strategy. Unlike adding nitrate at the beginning of the pump cycle (0 h), the nitrate was added at the end of pump cycle for the second nitrate dosing strategy. No nitrate was added into R2 after pumping event and the wastewater stayed in R2 for a whole pump cycle (12 h). After the 12 h pump cycle, there was 50 mg/L sulfide generated, 0 mg/L sulfate and 170 mg/L DCOD in the wastewater according to phase 1 results. Then the nitrate was added in R2 at this moment (12 h). To achieve this, instead of running the R2 for 12 h, the modified synthetic wastewater by adjusting ingredient which stimulated the effluent after 12 h pump cycle in R2 was used in this case. Nitrate was injected in R2 after pumping the modified synthetic wastewater in R2 and stayed in the reactor for 12 h. The HRT set to 12 h again was just for the convenience of operation. The nitrate was added to biologically remove the formed sulfide during first 12 pump cycle in the wastewater by autotrophic denitrification process. The appropriate HRT required for the complete removal of the generated sulfide was identified. Wastewater samples were collected at the end of each pumping cycle. The concentration of dissolved sulfide, sulfate, NH_4-N, NO_3-N, TN, TP, dissolved COD (DCOD), ferric, ferrous, nitrate and nitrite in the wastewater samples were measured.

Batch tests were carried out regularly to measure the change of related parameters over a whole pumping cycle (12 h) when the reactor was stabilized under certain conditions. Wastewater samples were taken hourly after pumping event, for the analysis of dissolved sulfide, sulfate, dissolved COD (DCOD), nitrite, nitrate and iron. In phase 1, the sulfate-reducing bacteria activity under anaerobic condition was measured as the sulfide production rate. The sulfide generation rates were calculated by the change of sulfide concentration measured within one hour divided by total biofilm area of each reactor.

3.3 Chemical and microbial community analysis

The water samples were analyzed for the determination of total and dissolved sulfide (TS and DS), sulfate, total, dissolved and soluble COD (TCOD, DCOD and SCOD), volatile fatty acid (VFA), nitrate (NO_3), ammonia (NH_4), total nitrogen (TN), total phosphorus (TP), total suspended solids (TSS) and volatile suspended solids (VSS). Total and dissolved sulfide were analyzed according to the methylene blue method (APHA et al., 2017). The sulfate, total, dissolved and soluble COD, nitrate, ammonia, TN and TP in wastewater samples were analyzed according to Standard Methods (APHA et al., 2017). DCOD was measured firstly by filtering the water samples through 0.45 µm membrane. The soluble COD (SCOD) was measured using the approach proposed by Mamais et al. (1993). The method involves removal by flocculation and precipitation (precipitating by $Zn(OH)_2$ at pH 10.5) of colloidal matter that normally passes through 0.45 µm membrane filters. All the parameters analyzed in our lab were measured twice (variation was less than 5%). The water samples for the determination of VFA, SS and VSS were sent to a commercial lab (CARO, Edmonton. Canada) for analysis according to Standard Methods (APHA et al., 2017).

At the end of each phase, 4 mL biofilm samples were collected for the microbial community analysis. All the biofilm samples were analyzed through 16s rRNA for identification of microbial community. The total genomic DNA was extracted from the collected biofilm and sediment samples by using the MO BIO PowerSoil DNA isolation kit (QIAGEN, Toronto, CA) according to the manufacturer's instructions. Extracted DNA was stored at -80 °C until further processing. The 16S rRNA gene fragments were amplified from the extracted total DNA with a primer set (515F/806R) targeting the V4 hypervariable region of both the *Bacteria* and *Archaea* domains (Caporaso et al. 2012). The sequencing was performed on Illumina HiSeq2500 platform at BGI Genomics Inc., Shenzhen, China. The raw data were filtered to eliminate the adapter pollution and low quality to obtain clean reads, then paired-end reads with overlap were merged to tags. And tags were clustered to Operational Taxonomic Units (OUT) at 97% sequence similarity. Taxonomic ranks were assigned to OTU representative sequence using Ribosomal Database Project (RDP) Naive Bayesian Classifier v.2.2. At last, alpha diversity, beta diversity and the different species screening were analyzed based on OTU and taxonomic ranks.

Chapter 4 Sulfide Generation and Emission in a Sanitary Sewer with Drop Structures and Pump Station in Steinhauer Area

4.1 Introduction

The buildup of sulfide in sewer systems and subsequent emission of hydrogen sulfide (H_2S) can cause sewer corrosion and sewer odor-related problems (Sun et al. 2015). Many large cities in warm climates like San Francisco and Australia, have occasionally experienced odor nuisances from its sewer system and odor complaints are not uncommon (Ganigue et al. 2011, Vollertsen et al. 2015). San Francisco, for example, encountered frequent odor nuisance at the eastern coast along the San Francisco Bay with the extended sewer system (Vollertsen et al. 2015). Unlike them, the City of Edmonton in cold climate also has experienced chronic sewer corrosion and sewer odor problems. During past ten years, thousands of complaints from residents have been recorded by the City of Edmonton, indicating a city-wide odor problem (Guo et al. 2018). What makes the Edmonton's sewer system unique is the deep trunk lines (more than 20 m deep), which result in over 900 pump stations and 800 drop structures in its sanitary sewer systems. The pump stations and drop structures are potential sites for sulfide generation and release, respectively. They could be the main causes of the odor problems in Edmonton. Research is urgently needed in order to find out the specific reasons for sewer odor problems in Edmonton and corresponding mitigation strategies.

When anaerobic conditions prevail in a sewer system, sulfate can be reduced to sulfide by sulfate-reducing bacteria (SRB) residing in biofilms on the pipe walls and in the sediments (Liu et al. 2015b). A number of factors can influence the production of sulfide in sewer network. The sulfide generation rate is mainly controlled by the factors involving dissolved oxygen (DO), sulfate, readily biodegradable organic matters, pH and temperature. DO in sewage determines the condition (aerobic, anoxic or anaerobic) for corresponding transformation processes of different types of organic matters, and sulfate reduction primarily takes place under anaerobic condition (García et al. 2017, Liu et al. 2015a, Shypanski et al. 2018). Sulfate and biodegradable organic matters are two main substrates utilized by SRB for sulfate reduction process (Nielsen and Hvitved-Jacobsen 1988). Temperature and pH have an effect on SRB activity and H_2S solubility and dissociation (Nielsen et al. 1998, Sharma et al. 2014). In addition, the total amount of sulfide generated is affected by hydraulic characteristics of sewers, such as the flow rate, flow velocity, pipe diameters, which control the residence time and area-to-volume ratio of sewer pipe and finally affect the level of sulfide formation in sewage (Li et al. 2019). Therefore, large and long gravity pipes with low velocity and pump station operated intermittently would be potential sites for sulfide formation.

When the sewage has certain amount of sulfide, the molecular form of sulfide, H_2S, can emit from water phase to sewer headspace and leak into manholes, vent pipes, atmosphere and even buildings. From an engineering point of view, H_2S can, therefore, be used as an indicator for odors found in sewer networks. The temperature, sewage quality, sewer characteristics and hydraulics have been proved to have an effect on the H_2S transfer (Carrera et al. 2016, 2017). The turbulence of the sewage flow created by hydraulic features such as drops, line bends, pipe size changes, areas of dynamic slope changes, junction structures, along the sewer can significantly enhance the transfer process. Matias et al. (2014) evaluated the influence of free-fall drops on the release of hydrogen sulfide gas in a lab experiment. Their results showed that the maximum concentration of H_2S released from the bulk sewage into the sewer air space increased with the drop height and could reach 500 ppm. Drop structures have also been reported to cause a large amount of air entrainment and the pressurization of the headspace in manholes, which then accelerates the release of H_2S from the sewer systems (Zhang et al. 2015). The air pressure at key locations is an indicator for the potential for off-gassing to the atmosphere, which can cause odor complaints.

The modelling of sulfide generation provides an efficient method to diagnose the area with increased risk of odor issue and to simulate future development applying mitigation strategies such as chemical dosing, or optimized pump operation. Significant efforts have been devoted to the prediction of sulfide generation in sewers. A few empirical models have been developed for the evaluation of sulfide generation rate (Thistlethwayte 1972; Boon & Lister 1975; Pomeroy & Parkhurst 1977; Elmaleh *et al.* 1998; Nielsen *et al.* 1998). These empirical equations introduce several key parameters such as biological chemical demand (BOD), chemical oxygen demand (COD), sulfate and temperature. Recently, two advanced models known as WATS (Hvitved-Jacobsen *et al.* 2013) and SeweX (Sharma et al. 2008) were developed. They describe both the anaerobic and aerobic processes involving multiple carbon and sulfur species. The key difference between the two models is in the sulfide generation rate, which is expressed by using readily biodegradable organic matter. The generation rate is described by following 1/2-order kinetics in

WATS model, but double Monod kinetics in SeweX model by using volatile fatty acids (VFA) and fermentable COD (FCOD) as substrates.

This study focuses on the Steinhauer area in the City of Edmonton, Alberta, Canada, which is one of areas receiving significant odor complaints. The sewer system in the area is comprised of a major deep sanitary trunk with two drop structures at the beginning of the trunk and a pump station at the end of the trunk. The objectives of the field study were to firstly identify the hotspots of odor issue and corresponding causes in this area including both the trunk and pump station, then to model the H_2S generation and assess potential mitigation strategies.

4.2 Methodology

4.2.1 Sewer system information

The study trunk sewer system is located in Steinhauer, Edmonton, Alberta, Canada. The trunk is approximately 2.7 km in length from the upstream point to the downstream point just before the pump station, Duggan pump station, as shown in **Figure 4-1**. The average flow in the trunk was about 0.150 m^3/s. There are 10 manholes along the trunk (T1~T10, here T denotes the manholes along the trunk), six of which (T1, T6, T7, T8, T9, T10) also serve as the intersection of the trunk and lateral pipes. At the beginning of the trunk, two main tributaries (T1-L2 and T1-L3) with flow rate of 0.065 m^3/s and 0.039 m^3/s transport sewage into the trunk.

The Duggan pump station is located at the end of the trunk and operates intermittently by two pumps. In a typical day, the pumps run for 4~5 cycles (**Figure 4-2**). For each cycle, the pumps stop for 2~4 hours and work for 2~4 hours. When the sewage level in the wet well is above 6.00 m, both pumps start to work, and both stop working when the sewage level drops below 5.00 m. The wet well at Duggan pump station is a cylinder with a diameter of 4.876 m. The invert height of the inlet pipe is 4.59 m above the bottom of the wet well (**Figure 4-3**). When the sewage level is at a low level of 5.00 m, both T10 and T9 are submerged by sewage. When the sewage level is at a high level of 6.00 m, sewage can back up to reach T7.

(a)


Duggan pump station
Whitemud Dr NW
Whitemud Park
111 St NW
43 Ave NW
T10-L1
T10-L2
T10
110 St NW
RIDEAU PARK
T9-L1
T9
41 Ave NW
40 Ave NW
108 St NW
107 St NW
112a St NW
T8-L1
T8
105 St NW
Duggan Park
37 Ave NW
T7
T7-L1
T7-L2
36 Ave NW
109 St NW
35a Ave NW
T6-L1
35 Ave NW
T6
T4
34 Ave NW
T5
T3
T6-L2
T2
32a Ave NW
31 Ave NW
T1-L2
Saddleback Rd NW
T1-L3
T1
29 Ave NW
112 St NW

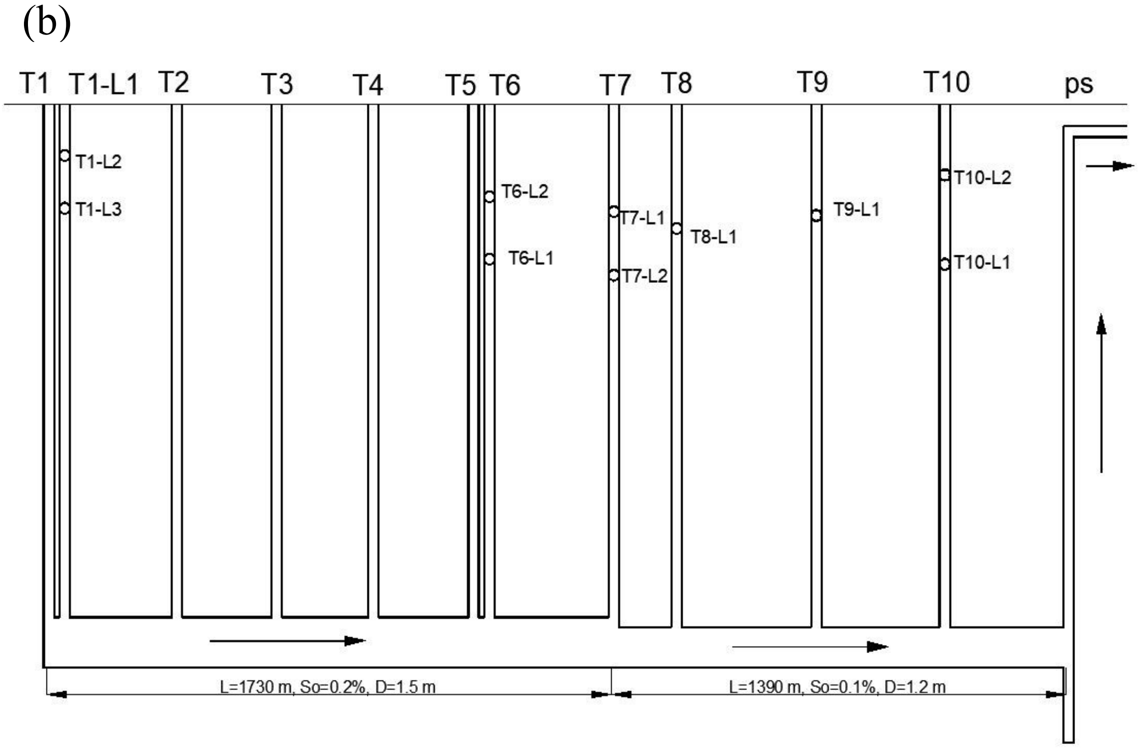


Figure 4-1 Study sewer system in Steinhauer area. (a) Plan view with the manholes on the trunk sewer indicated as T1 to T10. The sewage flows from T1 to T10; (b) Profile view with L indicating the locations where laterals entering the trunk sewer

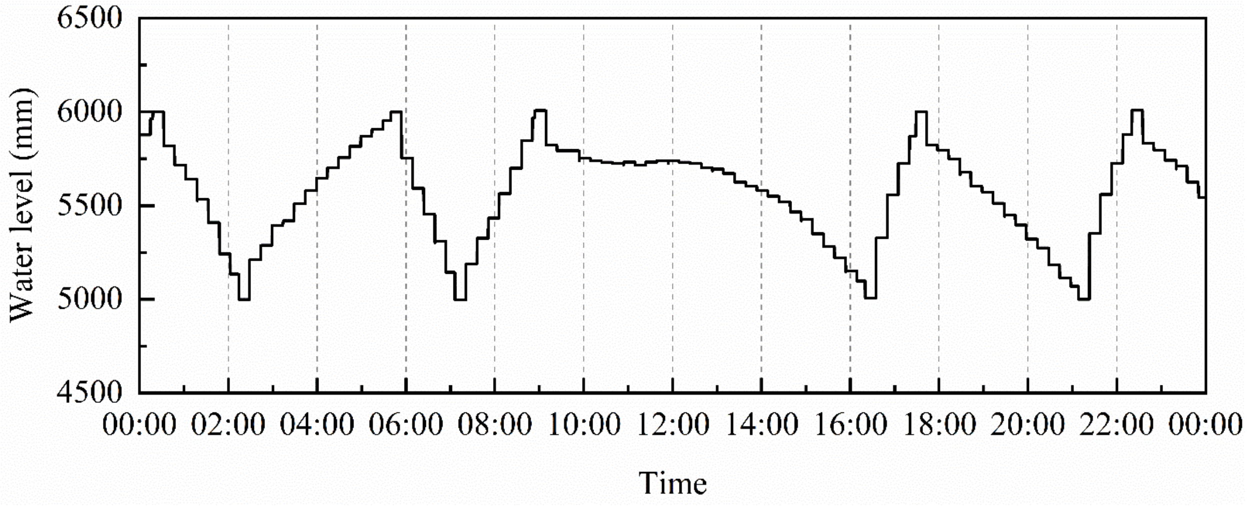


Figure 4-2 Variation of water level at Duggan pump station

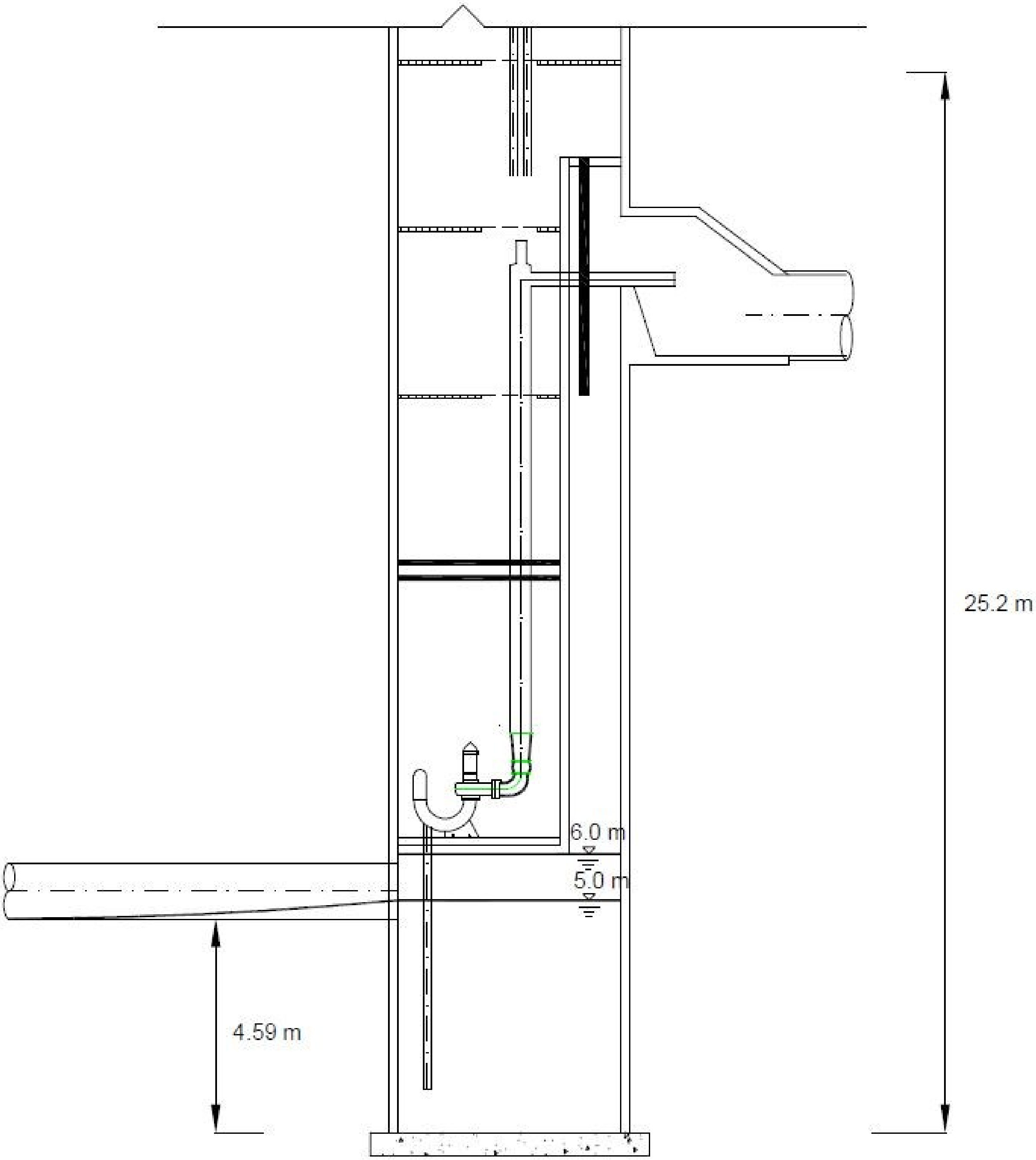


Figure 4-3 Profile of wet well at Duggan pump station

4.2.2 Field program

The field monitoring and sampling program in the trunk was conducted from May 15th to 24th, 2017. Sewage samples were grabbed from the trunk line on May 15th and 16th, 2017 at downstream manhole T10 to upstream manhole T1. The manholes included T1, T2, T3, T5, T8, T10, T1-L2, T1-L3, T8-L1, T9-L1, T6-L1, T6-L2, T7-L1, T7-L2, T10-L1 and T10-L2. The H_2S gas concentration was continuously monitored by Odalog (App-Tek, Queensland, Australia) for one week at T1, T2, T3, T4, T5, T8 and T10. Air pressure was also continuously monitored for one week by SmartReader (ACR Systems, Vancouver, British Columbia, Canada) at T1, T2, T4, T5, T8, and T10. At Duggan pump station, two rounds of field work were performed. Sewage samples were collected hourly for 24 hours on May 15th, 2017 and H_2S gas was continuously monitored in the air space of the pump well by Odalog for one week. Before this field work, a preliminary field work was also performed at the pump station on December 15th, 2016.

4.2.3 Modeling

Both WATS and SeweX model describing the anaerobic and aerobic carbon and sulfur transformation processes were applied which involves key chemical and biological processes and their kinetic expressions proposed by Hvitved-Jacobsen *et al.* (2013) and Sharma *et al.* (2008). As H_2S was detected during 12:00~24:00, the sulfide generation was simulated at peak time (around 22:00) when the flow rate and H_2S were high. Typical hydraulic characteristics of the trunk are shown in **Table 4-1**. The average flow rate and pipe information (diameter, length and slope) are obtained from the profile of Mike Urban. In terms of upstream part (from T1~T7), the flow velocity of each individual pipe is calculated by using Manning's equation. With regard to downstream part (T7~T10), the velocity is calculated by pipe length divided by retention time. The average retention time is around 4 h which is roughly estimated by pump operation. The trunk was modelled as plug flow from one manhole to the next manhole. Using the data obtained from the field work as initial input values for manhole T1 and then process it downstream until T10 by applying all relevant kinetic expressions, other values previously established in these two models were used for most model parameters like rate, yield and saturation constant. The evolution of these key values like VFA, DO, DS and H_2S was obtained by doing mass balance of each parameter along the trunk. A few main parameters like sulfide production rate constant were adjusted to fit the measured H_2S gas concentration. The calibrated key parameter of sulfide production rate constant for WATS model and maximum sulfide production rate for SeweX model were 0.05g S/(m^2h) and 10g S/(m^2d), which is within the range of literature values.

Table 4-1 Hydraulic characteristics of the trunk

Section	D (mm)	L (m)	Q (m^3/s)	S_0 (%)	V (m/s)	T (min)
T1~T2	1500	392	0.104	0.1900	0.84	0
T2~T3	1500	305	0.105	0.2370	0.90	8
T3~T4	1500	296	0.106	0.1740	0.80	13
T4~T5	1500	306	0.107	0.2400	0.90	20
T5(T6) ~T7	1500	429	0.124	0.1240	0.74	25
T7~T8	1200	192	0.126	0.1320	0.79	35
T8~T9	1200	427	0.141	0.1000	0.12	150
T9~T10	1200	393	0.142	0.0120	0.12	275
T10~PS	1200	379	0.151	0.1880	0.12	275

4.3 Results and Discussions

4.3.1 General sewage quality at Steinhauer area

To evaluate the sewage quality in this area, sewage samples from the trunk and pump station were collected for analysis. The general sewage quality is shown in **Table 4-3**. The TN was around 56.8 mg/L in which the NH_4^+ was dominant (36.1 mg/L). The nitrate NO_3^- level was very low at about 0.98 mg/L. The total COD (TCOD) was about 618 mg/L. The TP was high at about 18.7 mg/L. The sewage temperature was around 15.8 ℃ and pH was 7.8 as the field work was conducted in spring time (May 15th and 16th, 2017).

Table 4-2 General sewage quality in the Steinhauer area (May 15th and 16th, 2017)

Location	Time	NH_4-N (mg/L)	NO_3-N (mg/L)	TN (mg/L)	TP (mg/L)	TCOD (mg/L)	pH	T (°C)
T1		33.3	1.8	53.4	24.5	673	8.1	15.2
T2		37.0	1.4	56.2	23.0	660	8.1	13.6
T3		28.8	0.7	40.7	15.7	564	8.0	15.0
T4		65.3	2.6	92.9	17.2	610	8.1	13.2
T5		50.1	0.6	67.0	22.4	503	8.6	15.3
T8		37.2	1.0	70.4	21.2	616	8.2	14.5
T10		25.3	0.8	48.9	18.5	768	8.3	17.5
PS	9:30	36.3	0.8	55.3	16.3	667	7.4	17.8
	13:30	47.1	0.7	73.1	23.0	600	7.9	17.8
	17:30	34.1	1.0	60.1	21.7	714	7.3	17.2
	19:30	30.0	0.6	46.0	16.4	668	7.3	16.7
	0:30	25.4	0.6	41.9	14.2	645	7.6	16.2
	4:30	25.8	0.6	38.9	12.5	426	7.4	15.8
	8:30	30.4	0.6	51.3	15.1	541	7.4	15.8
Average		**36.1**	**1.0**	**56.8**	**18.7**	**618**	**7.4**	**15.8**

4.3.2 Identification of the hotspots for H_2S emission in the trunk

The air pressure is shown in **Figure 4-4**. The T2 was pressurized with high pressure following a diurnal change. The air pressure decreased in the early hours of the morning (22:00 to 5:00) from the around 900 Pa to the minimum value of 200 Pa. Then it started to increase until 10:00~12:00 with a maximum value around 900 Pa. From 12:00 to 22:00, it kept at high level in the range of 500~900 Pa with some fluctuations. The T8 was also pressurized with high pressure, but it oscillated between 0 to 800 Pa. Unlike T2 and T8, the air pressure at T10 was rather low (50 Pa) all the time. T1~T6 followed the same air pressure pattern as T2, T7 followed the same air pressure pattern as T8 and T9 followed the same air pressure pattern as T10 (not shown).

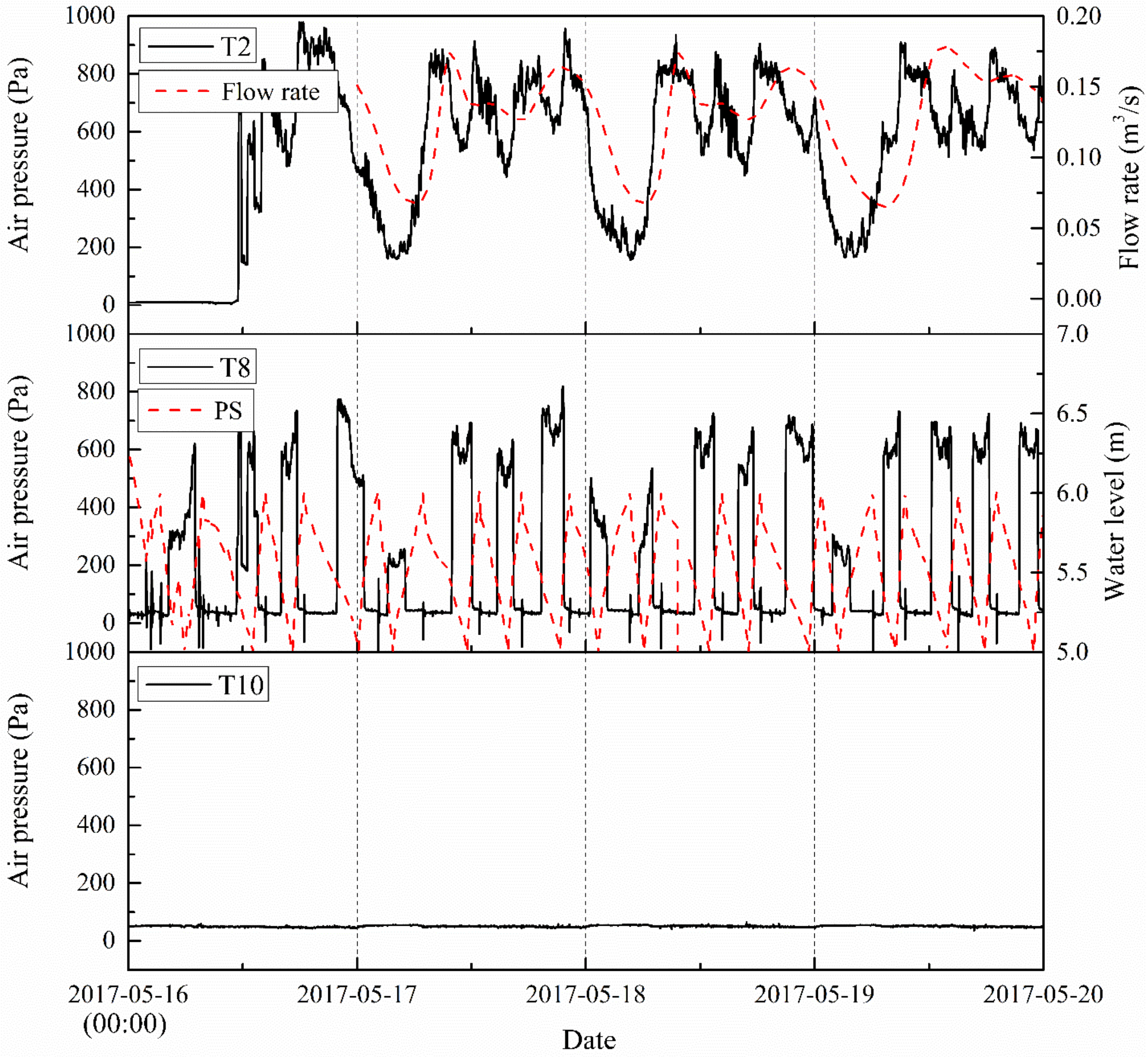


Figure 4-4 Measured diurnal pattern of air pressure and flow rate in the trunk and corresponding sewage level at pump station

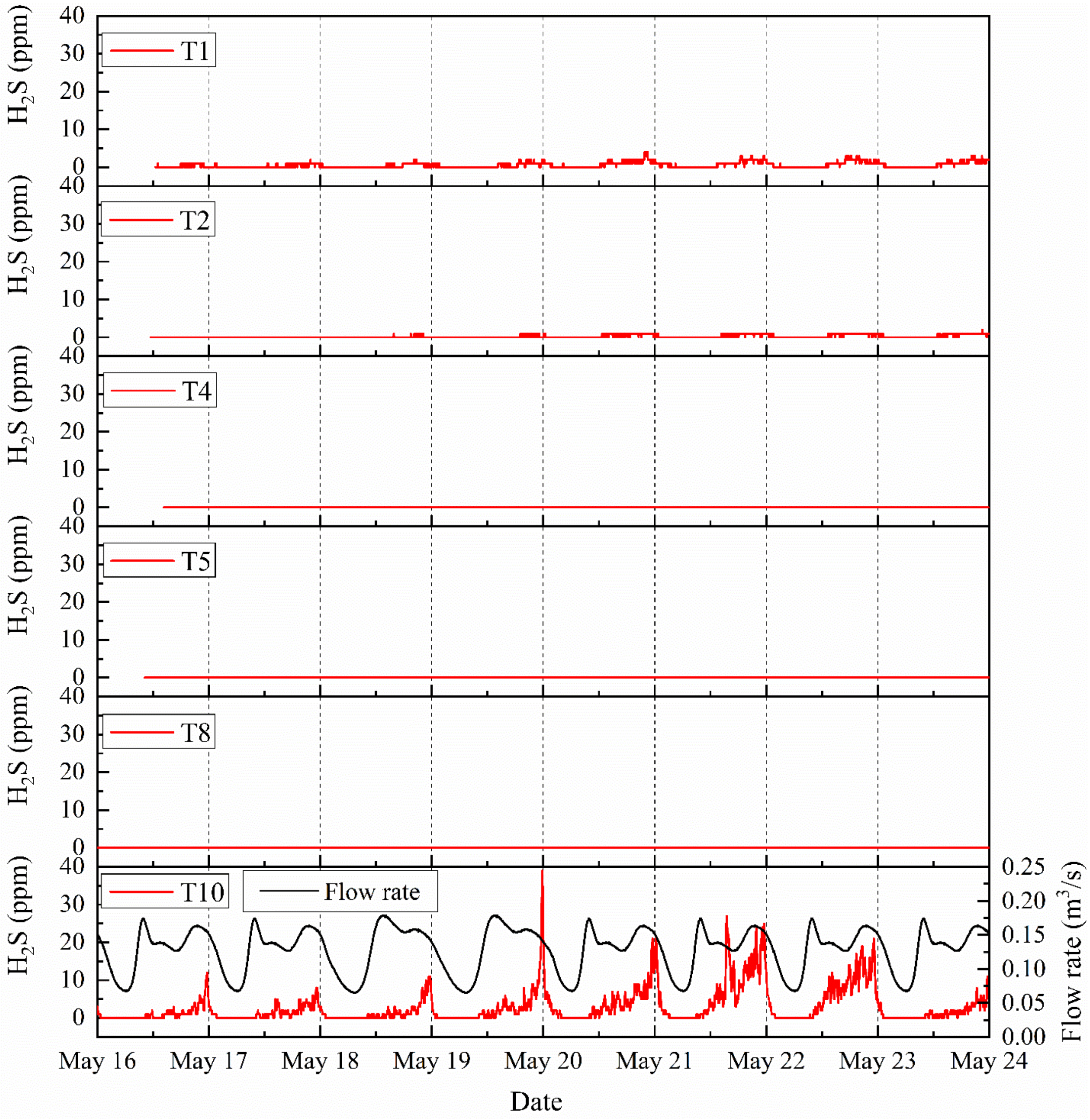


Figure 4-5 Measured diurnal pattern of H$_2$S gas in the trunk

The high pressure in the trunk was caused by the drop structures at site T1. The sewage from two main tributaries T1-L2 and T2-L3 dropped into T1 with a drop height of 22 m and 25m, respectively. The drop structures dramatically increased air drag since the sewage was broken into small liquid drops. As a result, a large amount of air was dragged into trunk and caused the pressurization of the trunk (T1~T6) (Ma et al. 2016). From the air pressure variation of T2, shown in **Figure 4-4**, it was clear that the air pressure responded to the change in the sewage flow rate.

With regard to the downstream trunk (T7~T10), the air pressure was controlled by the pump operation, as T7 and T8 were intermittently submerged by the sewage backed up from the pump wet well. When the sewage level at the pump well was above 5.4 m, the trunk at T8 ran full with sewage and the pressure dropped to 50 Pa until sewage level decreased below 5.4 m (**Figure 4-4**). The pressure oscillation followed the pump operation cycles of 4~5 cycles per day. The trunk at T9 and T10 were permanently submerged by sewage, thus the measured air pressure did not respond to the air pressure in the trunk and was low all the time. Therefore, the trunk can be divided into two parts: upstream part (before T7) of normal gravity pipe and downstream part (after T7) serving part of the storage volume for pump station. The section between T7 and T8 was intermittently submerged depending on sewage level of the pump well.

The continuously monitored H_2S concentration is shown in **Figure 4-5**. H_2S was detected at locations of T1, T2 and T10 over 12:00~24:00. The increase in flow corresponded to an increase in H_2S, reaching maximum values at around 00:00. The corresponding maximum values were in the range of 3~7 ppm, 2 ppm and 10~40 ppm. The stripping of H_2S in the sewage receiving from T1-L2 and T1-L3 resulted in H_2S gas at the headspace of T1 and T2 and the formation of sulfide locally in the sewage led to a higher H_2S level at T9 and T10, which is further discussed in the next section. Meantime, the large air pressure inside the sewer headspace accelerated the emission of H_2S gas to ambient atmosphere and led to odor complaints.

4.3.3 Sulfide generation in the trunk

As shown in **Table 4-3**, the DS concentration at locations of T1-L2, T1, T2, T3, T5, T6, T8-L1 and T9-L1 were low, all close to zero. By comparison, T1-L3, T9, T10, T6-L1, T6-L2, T7-L1, T7-L2, T10-L1 and T10-L2 had higher DS concentration (>0.1 mg/L). These levels were not so significant, because most of the sewage samples were taken in the morning and H_2S only detected in the afternoon, as shown in **Figure 4-5**.

The T1 received sewage from T1-L2 and T1-L3. At T1-L3, DS (0.43 mg/L) could be released into the air space through the enhanced mass transfer when the sewage broke up into small liquid drops. After the stripping of H_2S gas, the DS at T1 was close to zero (0.02 mg/L). The stripped H_2S at T1 was transported downstream, but part of H_2S could be absorbed and oxidized on the moist pipe and manhole wall with a portion emitted to the atmosphere. So H_2S gas concentration dropped at

T2 and there was no H_2S existing from T3 to T8. The falling liquid drops also increased mass transfer between sewage and air which resulted in high DO at T1 and T2 with a concentration of 8.8 mg/L and 9.2 mg/L. Thus, drop structures in a sewer network can promote both H_2S stripping and sewage reaeration, and their effects on H_2S generation and transport need to be carefully considered. From T2 to T6, the sewage was under a high dissolved oxygen level (8 mg/L), together with a short retention time (0.62 h), no significant amount of sulfide could not be generated.

The DS at T8 and T10 was 0.12 and 0.11 mg/L, respectively. The average DO in all samples was about 5 mg/L, since samples were not taken in the afternoon (12:00~24:00) when H_2S gas was present. Most of the sulfide could be oxidized by the high DO, so the DS concentration was not significant in the sewage samples. As for downstream part, T7~T10 were submerged by sewage for a long time which served as sulfide production sites. DO was depleted for the degradation of easily degraded organic substances with the increased flow rate in the afternoon (12:00~24:00). A high sulfide concentration could be formed in the sewage and then transferred into the air phase, which made T9 and T10 as hotspots downstream. The tributaries of T6-L1, T6-L2, T7-L1, T7-L2, T10-L1 and T10-L2 where the flow rate was small compared to the trunk could not affect the trunk so dramatically. But the DS generated upstream could also make the surrounding area as potential hotspots.

The formation of sulfide in sewers is controlled by a few factors. The temperature did not change significantly during the field study period. The pH was in the range of 7.5~8.0. All SRB are characterized by using of sulfate as a terminal electron acceptor during anaerobic respiration. The sulfate concentration in the sewage was 30~70 mg S/L. This concentration level is not likely the limiting factor as has been shown by many researchers unless its concentration is below 10 mg/L (Nielsen & Hvitved-Jacobsen 1988). At this high sulfate concentration, the sulfide production rate is believed to be limited by SCOD. It is a well-known fact that SRB consume readily biodegradable organic matters, which consist of FCOD and fermentation product VFA (Hvitved-Jacobsen et al. 2013). The average TCOD and SCOD were 600 mg/L and 150 mg/L, respectively. The average VFA was 20 mg/L and an increase in DS with high VFA could be seen in Table1. A longer retention time was responsible for higher dissolved sulfide and VFA. DS concentration at downstream locations was higher than that at upstream locations. Similar trend was also observed for VFA.

Table 4-3 Sewage characteristics in the trunk sewer (mg/L) (May 15th and 16th, 2017)

Location	Sampling Date	Sampling Time	TS mg/L	DS mg/L	Sulfate mg S/L	TCOD mg/L	SCOD mg/L	VFA mg/L	TSS mg/L	VSS mg/L	T (℃)	DO mg/L	pH
T1-L2	May 16th	13:42	1.04	0.43	60.5	523	127	3.8	248	220	14.7	5.9	8.1
T1-L3	May 16th	12:38	0.55	0.05	48.0	600	167	13.3	318	260	15.2	5.5	8.3
T1	May 16th	12:13	1.00	0.05	31.0	673	146	9.9	800	376	15.2	8.8	8.1
T2	May 16th	11:26	1.86	0.03	38.0	660	146	0.22	_	_	13.6	9.2	8.1
T3	May 16th	1:25	0.65	0.04	70.5	564	144	18.8	_	_	15.0	6.5	8.0
T5	May 16th	8:48	0.50	0.01	61.0	503	121	0.22	206	184	15.3	5.2	8.6
T8	May 15th	11:41	0.81	0.12	59.5	616	151	24.8	202	168	14.5	3.7	8.2
T10	May 15th	10:16	0.73	0.11	51.5	768	273	62.5	208	164	17.5	5.6	8.3
T6-L1	May 15th	13:45	3.50	1.46	35.5	1185	435	109.8	_	_	14.7	4.6	7.4
T6-L2	May 15th	14:00	1.22	0.71	64.5	674	220	50.9	175	163	15.3	3.6	7.5
T7-L1	May 15th	12:23	0.58	0.24	48.5	487	116	_	_	_	18.0	4.0	7.9
T7-L2	May 15th	12:12	0.94	0.48	67.0	532	213	_	_	_	16.4	3.9	7.6
T8-L1	May 15th	11:34	1.05	0.02	33.5	997	219	68.7	436	268	15.8	5.4	7.8
T9-L1	May 15th	10:58	0.21	0.04	410	285	76	_	_	_	12.0	6.2	8.1
T10-L1	May 15th	10:37	0.87	0.13	26.0	1690	1180	6.6	112	96	15.2	6.7	8.0
T10-L2	May 15th	10:47	0.88	0.20	67.5	844	269	71.3	278	240	17.7	3.5	7.4

_: not applicable

4.3.4 Simulation of sulfide generation in the trunk and optimization of pump operation

The model results are shown in **Figure 4-6(a)**, the DO was depleted gradually to zero from T1 to T8 as the increase of the retention time. When the DO existed, the formed DS would be oxidized chemically and biologically. So the DS increased slowly from T1 to T7 and aerobic bacteria utilized the VFA for aerobic respiration resulting in the drop of the VFA. From the T8, the fermentation process under the anaerobic condition led to the rise of the VFA and sulfide generation rate increased as the increase of VFA. The sulfide formation was sensitive to VFA. The long retention time caused the formation of high level of DS at T8 (2.6 mg/L), T9 (4.8 mg/L) and T10 (4.8 mg/L). The simulated maximum sulfide generation rate in this area is 0.005 g/m^2h. By using the two-film theory and considering the loss of H_2S due to oxidation at concrete surface, the simulated H_2S gas was zero from T1 to T7, but was 5 ppm, 14 ppm and 14 ppm at T8, T9 and T10. Both WATS and SeweX models gave similar results and an acceptable correlation between predicted and observed H_2S gas concentrations can be seen, except T1. T1 did not agree with the model predictions mainly because the release of H_2S gas at drop structures was not included in the models.

As the hotspots of T9 and T10 were caused by pump operation, one solution for this was to optimize its operation to reduce the retention time and avoid the trunk running full. The developments of DS in sewage phase and H_2S gas at headspace were simulated after applying optimized pump operation where the trunk is no longer act as the pump storage and the entire sewer trunk has normal gravity pipe flow. Using the calibrated WATS and SeweX model, the simulated results are shown in **Figure 4-6(b)**. The generated DS and H_2S at T9 and T10 are now low, less than 1mg/L and 1 ppm, respectively. Thus both T9 and T10 will no longer become the hotspots, and the proposed pump operation optimization is effective in preventing H_2S generation in the trunk.

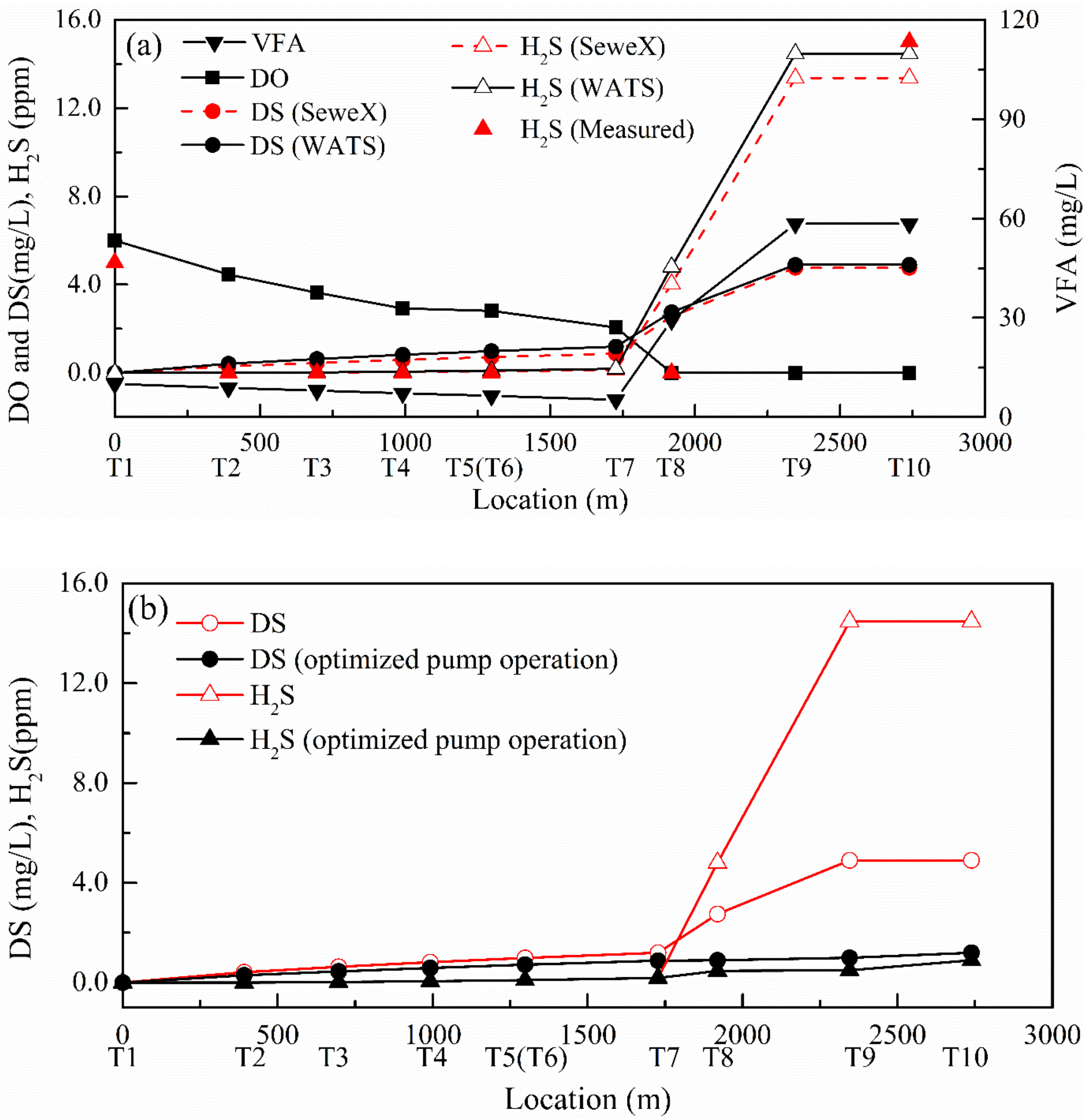
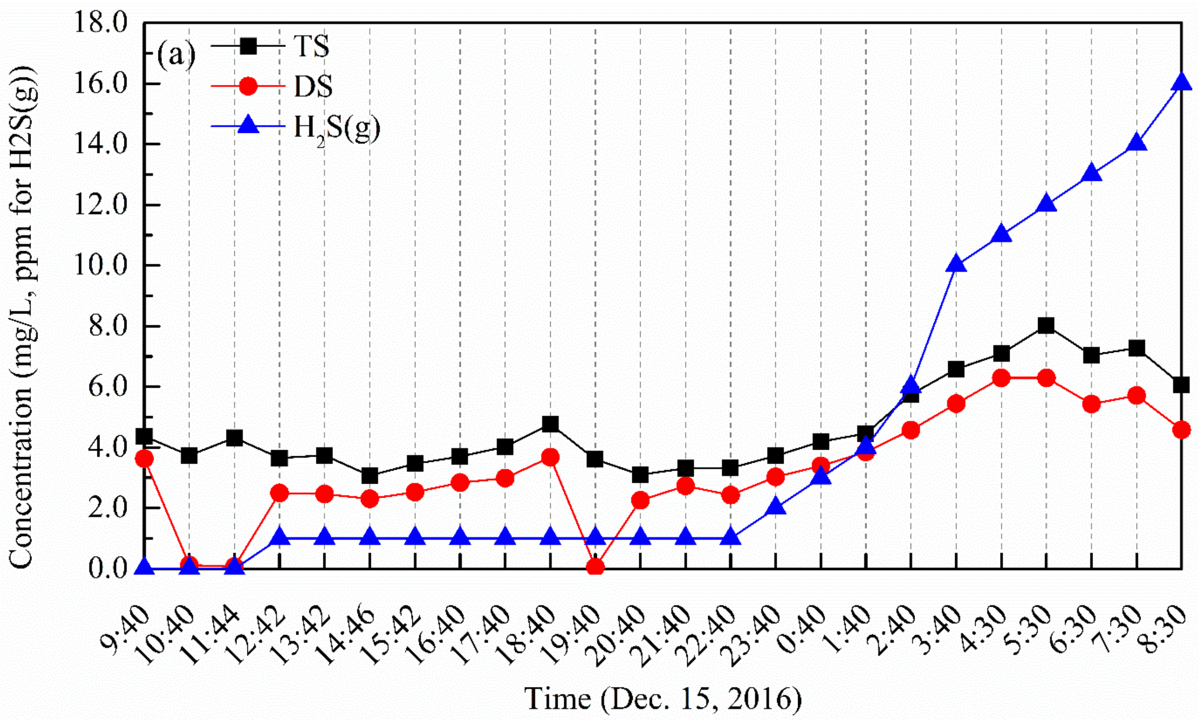
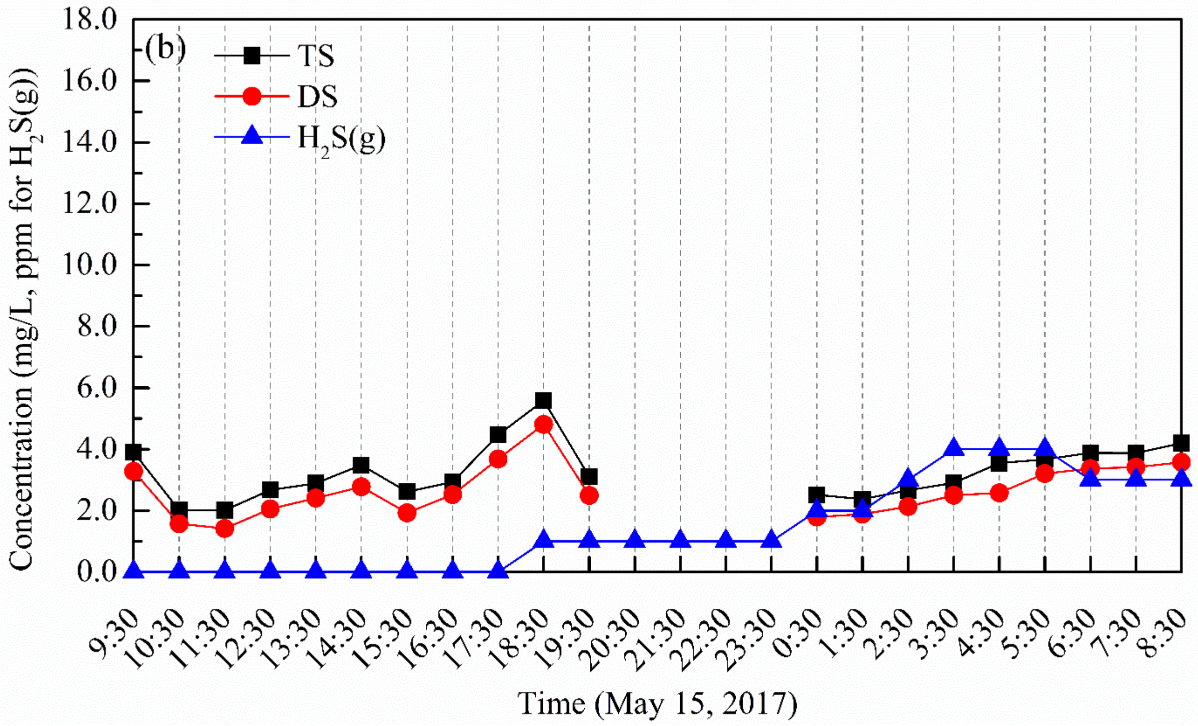


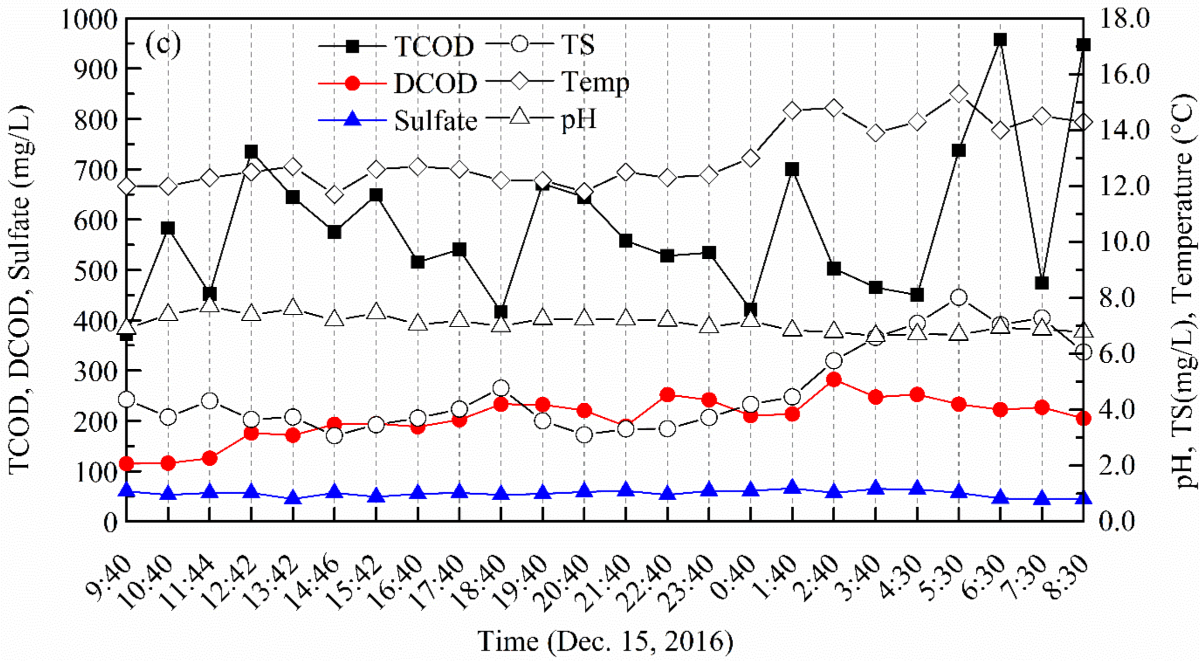
Figure 4-6 Simulated results in the trunk before (a) and after (b) by applying optimized pump operation

4.3.5 Sulfide generation at the pump station

The sewage samples were collected hourly from the pump well and Odalog was installed in the air space of the pump well to continuously monitor H_2S gas concentration on Dec. 15th, 2016 and May 15th, 2017. As shown in **Figure 4-7(a) and (b)**, the TS and DS levels in water samples were around 4.0 mg/L and 2.5 mg/L, respectively, over the time period of 9:40 to 22:40 on Dec.15th, 2016. Then they started to rise to the maximum of 8.0 mg/L and 6.0 mg/L, respectively, from 22:40 to 5:30 followed by a decrease to 6.60 mg/L and 4.58 mg/L between 5:30 and 8:30. The DS value at 10:40, 11:40 and 17:40 were not reliable due to the inappropriate sample preservation that can be ignored. Meanwhile, the corresponding H_2S in gas phase was about 0~1 mg/L from 9:40 to 22:40, when DS was relatively low. Then it increased with the increase of the DS until the end of the study cycle (8:30) when the concentration of H_2S was the highest at 16 ppm. On May 15th, 2017, the TS and DS had the similar trend. Four water samples lost from 17:30 to 00:30 due to the breakdown of the operation pump. The corresponding H_2S in gas phase was about 0~1 mg/L from 9:40 to 23:30 and then it increased with the increase of the DS until the end of the study cycle (8:30) during which the maximum was 4 ppm.




(b)
TS
DS
$H_2S(g)$
Concentration (mg/L, ppm for $H_2S(g)$)
18.0
16.0
14.0
12.0
10.0
8.0
6.0
4.0
2.0
0.0
9:30
10:30
11:30
12:30
13:30
14:30
15:30
16:30
17:30
18:30
19:30
20:30
21:30
22:30
23:30
0:30
1:30
2:30
3:30
4:30
5:30
6:30
7:30
8:30
Time (May 15, 2017)


(c)
TCOD
DCOD
Sulfate
TS
Temp
pH
TCOD, DCOD, Sulfate (mg/L)
pH, TS(mg/L), Temperature (°C)
1000
900
800
700
600
500
400
300
200
100
0
18.0
16.0
14.0
12.0
10.0
8.0
6.0
4.0
2.0
0.0
9:40
10:40
11:44
12:42
13:42
14:46
15:42
16:40
17:40
18:40
19:40
20:40
21:40
22:40
23:40
0:40
1:40
2:40
3:40
4:30
5:30
6:30
7:30
8:30
Time (Dec. 15, 2016)

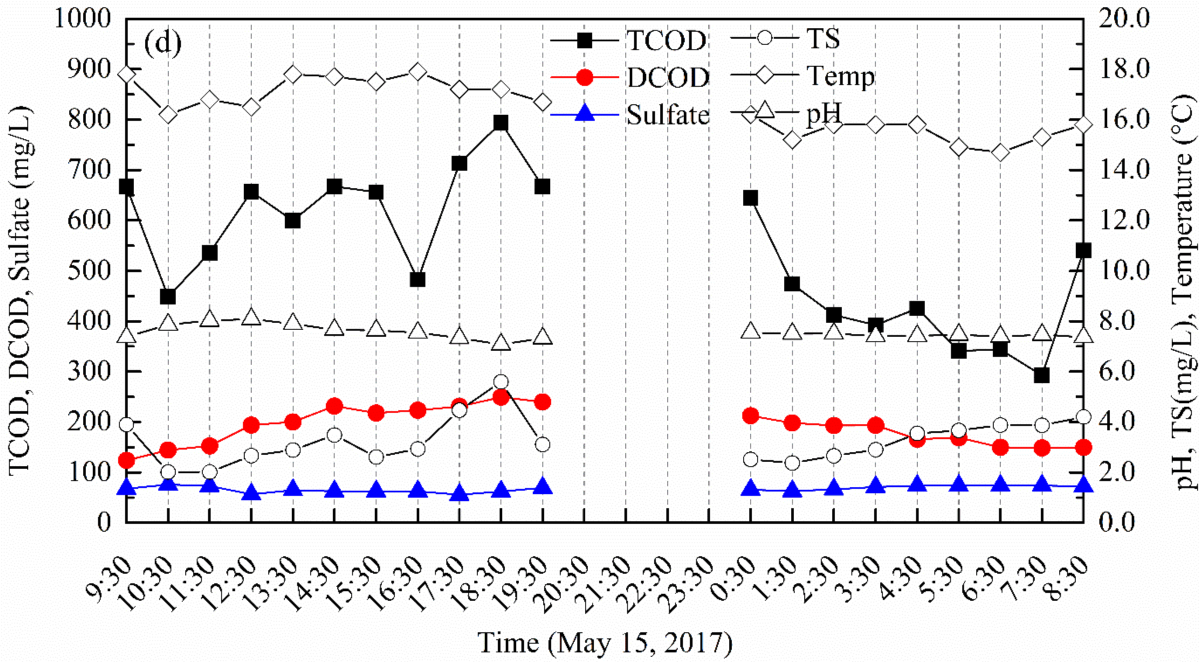
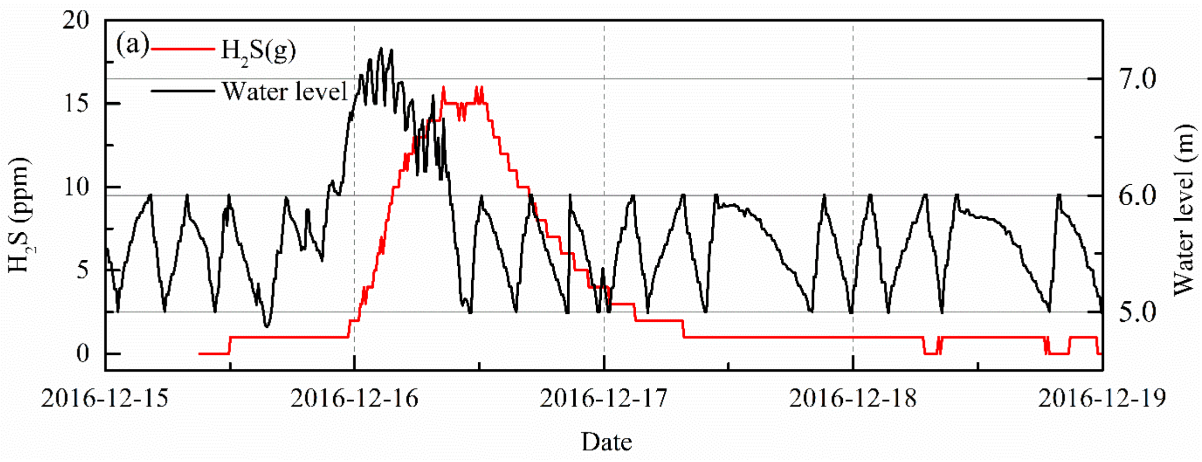


Figure 4-7 Diurnal pattern of TS, DS and H_2S gas at pump station on Dec. 15, 2016 (a) and May 15, 2017 (b) and affecting factors on Dec. 15, 2016 (c) and May 15, 2017 (d)

The main factors associated with the formation of sulfide in the pump station are shown in **Figure 4-7(c) and (d)**. The temperature of sewage was 12~15 ℃ for the first time (Dec.15th, 2016) and it increased to 15~18 ℃ for the second time (May 15th, 2017). The pH decreased during the cycle on Dec.15th, 2016, especially close to the end of the cycle, which may be due to the transformation of anaerobic processes and the generation of high sulfide. The sulfate in the sewage was somewhat constant at about 50 mg/L and 60 mg/L for two times. The total COD (TCOD) fluctuated significantly in the range of 400 ~900 mg/L and 300 ~800 mg/L during the two field periods. The DCOD presented the same growing trend as the TS in both cases and its average number was 200 mg/L and 190 mg/L. As the limiting substrate for sulfate reduction reaction, when the DCOD increases, the generation rate increases. The pump station was a potential site for sulphide generation.

The pump operation conditions determine the retention time and the length of the trunk submerged by sewage. **Figure 4-8** shows the monitoring gaseous H_2S along with water level at the pump well from Dec. 15th to 18th, 2016 and May 15th to 18th, 2017. The high H_2S only occurred on the days when the sewage samples were taken. For other time, it was only 0~1 mg/L. Generally, the pumps should operate when sewage level in the wet well is above 6.00 m. But for the two specific days (Dec.15th, 2016 and May 15th, 2017), the sewage kept rising beyond 6 m to a maximum level of 7.2 m and 6.4 m which dramatically increased the retention time at both pump wet well and downstream trunk. Besides, more downstream trunk was served as the site for sulfide formation. The H_2S gas concentration increased with the rise of water level with a maximum level of 16 ppm and 4 ppm when the sewage level was more than 6 m (at around 21:00 for both cases). It turned out that the pumps broke down and did not start working after 21:00, which caused the rise of sewage level above 6.0. The pump operation conditions had effect on the sulfide generation at the pump wet well. Even though the pumps did not work properly after 21:00, the results before 21:00 also could reflect the normal conditions at the pump station. From results of two rounds of field work before 21:00, the DS level was in the range of 2~3 mg/L and H_2S gas was 0~1 ppm. A few studies investigated the sulfide formation in pump station. A sewage pump station located in the Gold Coast, Australia with retention time 3~7 h, where the average DS generated was about 7.4 mg/L and corresponding average H_2S was 36.5 ppm (Jiang et al. 2013). The values are much higher due to the high temperature (26.3 °C). The sewage temperature in Denmark is 14.4 °C which is similar in Edmonton. On average, the total sulfide concentration increased by 2.53 mg/L during approximately 4 hours of anaerobic transport in the force main of pump station (Vollertsen et al. 2015). The gas-phase concentrations of H_2S were generally below the detection limit of the gas detectors (1 ppm) and never exceeded 5 ppm for extended periods which was in agreement with the results in Edmonton. The elevated temperature is favorable for sulfide buildup, but in cold climate regions, relative high dissolved sulfide (>2 mg/L) also could be formed under long retention time, likely more than 3 hours.



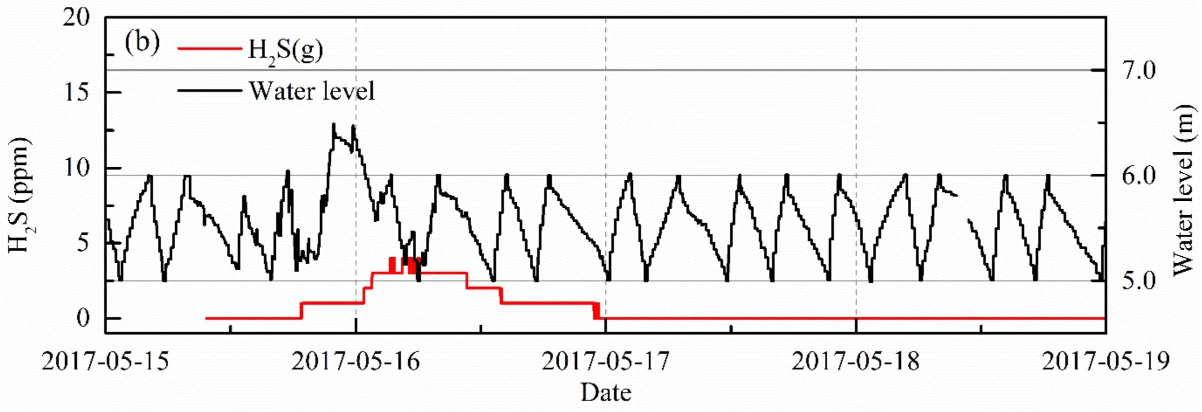


Figure 4-8 H_2S gas and sewage level at the pump station during the two field periods (a) December 2016, (b) May 2017.

4.4 Conclusions

A field study was performed in a deep trunk sewer in the Steinhauer area in Edmonton, Canada, on the sewer system with two drop structures and a pump station to identify the causes and assess potential mitigation strategies for odor complaints in this area. The main findings are:

(1) The results demonstrated that the locations of T1, T2, T9 and T10 in the trunk were the hotspots along the trunk (T1~T10). The T1 received sewage from T1-L3 containing DS which was released through the drop structure with drop height of 25 m to make T1 and T2 as hotspots, even though the DS was low in the water phase. The downstream part was served as additional storage volume for pump station and was submerged by sewage. The long retention time (4h) and large biofilm area to volume at T9 and T10 allowed the formation of high DS as they were submerged by sewage all the time. The sulfide generated locally was emitted into the air phase as H_2S gas to make them as hotspots. Therefore, the retrofitting of current drop structures is needed to minimize its stripping effect. How H_2S is released at a drop structures with significant H_2S emission rate due to the high turbulence occurring at waterfalls needs to be further investigated. For future sewer system design, the drop structures should be avoided and pump station design should aim to not leave back flows in the trunk.

(2) The WATS and SeweX models for H_2S prediction were calibrated by field data. The H_2S gas buildup in the trunk predicted by both models correlated well with measured H_2S gas concentration. The calibrated model was applied to simulate the optimized pump operation

situation where downstream pipe was no longer served as part of storage volume for pump station. The simulated results revealed that downstream of the trunk would not be hotspots if applying optimized operation. The results at pump station showed that pump station was a potential site for sulfide generation and the generated TS and DS were about 4.0 mg/L and 3.0 mg/L. Less frequent pumping results in a substantial sulfide generation at pump station and increasing pumping frequency as much is essential to reduce sulfide generation.

Chapter 5 H_2S Release and Transport in a Combined Trunk Sewer with Drops in Bonnie Doon Area

5.1 Introduction

The City of Edmonton, Alberta, Canada, like many other cities worldwide, is facing sewer odor nuisance and pipe corrosion issues. The main reason for the odor complaints and pipe corrosion problems is hydrogen sulfide (H_2S) generated in sewer systems (Hvitved-Jacobsen et al. 2013). When dissolved oxygen and nitrates are depleted as bacteria metabolize organic material in sewage, sulfate (SO_4^{2-}) is utilized and reduced to sulfide (S^{2-}) by sulfate reducing bacteria (SRB). The SRB mainly belong to eight genera: *Desulfovibrio*, *Desulfotomaculumn*, *Desulfomonas*, *Desulfobulbus*, *Desulfobacter*, *Desulfococuus*, *Desulfosarcina* and *Desulfobacterium* (Visser 1995). The dissolved H_2S in sewage can be released into the sewer headspace above the sewage and transported by the air flow, causing sewer odor complaints when it emits to the community. Meanwhile, the H_2S in the sewer headspace can be absorbed by the moist concrete surface in the pipe and oxidized to sulfuric acid (H_2SO_4) by sulfur oxidizing bacteria (SOB) with the presence of oxygen and cause corrosion problem (Gomez-Alvarez et al. 2012). The *Thiobacillus* was found to be the key community members of SOB (Hernandez et al. 2002). Besides, *Thiothrix*, *Thiomonas intermedia*, *Halothiobacillus neapolitanus*, *Acidiphilium acidophilum*, and *Acidithiobacillus thiooxidans* were also found to be SOB responsible for concrete corrosion (Okabe et al. 2007).

What makes Edmonton's sewer system special is the large number of drop structures (over 800) due to the rather deep trunks (up to 50 m below the ground) in the sewer network. Sewer structures such as junctions, manholes, bends, weirs, and drops may give rise to increased turbulence compared with the hydraulic conditions that exist under normal flow conditions in a sewer pipe. In particular, at sewer falls and drops, the mass transfer between water and air is significantly increased caused by phenomena such as splashing droplets and entrainment of air in the water phase (Ma et al. 2016, Qian et al. 2017). Such structures can therefore promote H_2S stripping and oxygen reaeration (Beceiro et al. 2017, Jung et al. 2017). Matias et al. (2014) evaluated the influence of free-fall drops on the release of H_2S gas in a laboratory experiment and reported that the maximum H_2S concentration in the air phase can reach 500 ppm with 0.9 m drop and 10 mg/L sulfide in the liquid phase. Then further study was carried out to establish empirical relationships between the mass transfer of oxygen and physical parameters of drop structures (Matias et al.

2017). But these results are likely specific to particular structures and flow conditions, and their applicability in real sewer systems need to be tested.

The emission of H_2S to the ground is directly related to the dynamics and transport of H_2S and air movement in the headspace along the system, in particular the pressurization due to the drop structures (Qian et al. 2018, Wang et al. 2012). The air movement could transport the sewer air from one location in the network to another. Once the H_2S gas is released at certain drop structures in real sewer systems, it can move with the sewer air to other locations and lead to more locations of odor complaints. It is essential to conduct the field work in real sewer network to identify not only the generation of H_2S in the sewage, but also the stripping of the H_2S gas from sewage and its subsequent transport in the sewer network.

The Bonnie Doon area in Edmonton, Alberta, where the odor problem is prevalent, is chosen as the study area for field work. The study focuses on the sulfide emission and transport along the trunk together with the laterals. The objectives of this study are firstly to investigate the generation of sulfide in the water phase and transport of H_2S in the air phase along the trunk and relevant laterals in this area, then to evaluate the effect of drop structures on reaeration and H_2S release into air phase in the trunk and finally, identify the functioning bacteria in the biofilm for sulfide generation and H_2S gas oxidation.

5.2 Methodology

5.2.1 Field study

The field work was conducted in Bonnie Doon area, a neighbourhood in south-central Edmonton, Alberta, Canada. The sewer system in the area is a combined system which carries both sewage and stormwater runoff. This study only focuses on the dry weather case. The main trunk and corresponding laterals that are connected to the trunk is shown in **Figure 5-1(a) and (b)**. The main trunk is connected to 10 manholes in the study area, which are numbered as T1 to T10, with an average flow rate from 1.257 m^3/s at T1 to 1.521 m^3/s at T10. There are 14 laterals (L1-L14) contributing to the trunk. Among them L8 and L12 are major laterals which has the flow rate of 0.240 m^3/s and 0.290 m^3/s, respectively. The flow rate of the rest of laterals was relatively low (less than 0.02 m^3/s) compared to that in the trunk. In the trunk, there are three drops with a drop height of 2.7 m, 5.2 m and 2.0 m, as shown in **Figure 5-1**, which is located at the manholes T3-1, T4 and T5, respectively.

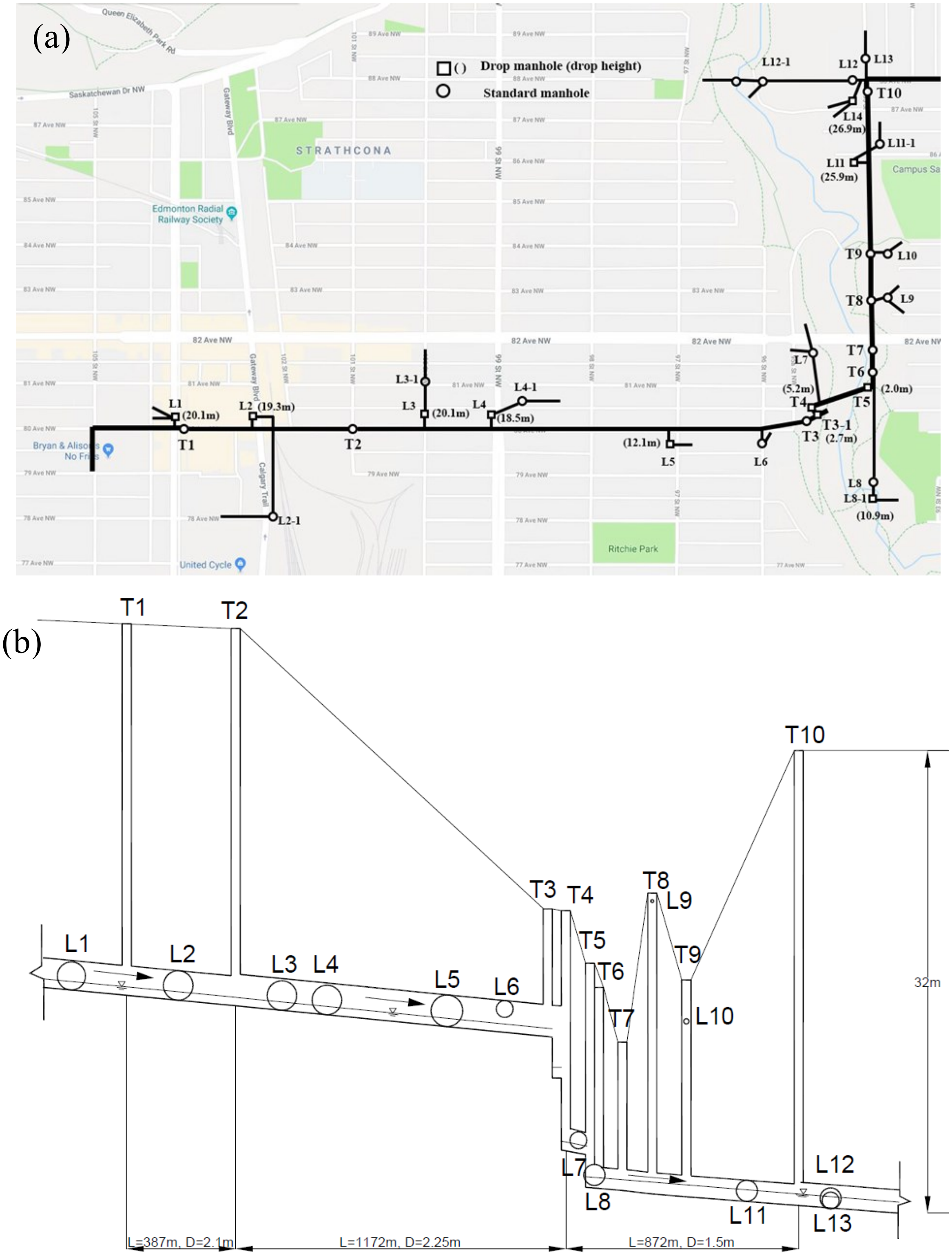


Figure 5-1 Study sewer system in Bonnie Doon area. (a) Plan view with the manholes on the trunk sewer indicated as T1 to T10. The sewage flows from T1 to T10; (b) Profile view with L indicating the locations where laterals entering the trunk sewer.

Two rounds of field monitoring were implemented in the area in 2017 and 2018. Water samples were obtained from the manholes in the trunk (T1 to T10) and main laterals (L8 and L12) for the first round on Nov. 29 – 30, 2017. The H_2S gas concentration and air pressure at the sewer headspace of the trunk (T1 to T10) and laterals (L1, L3, L4, L5, L6, L8, L8-1, L9, L10, L12, L13) were monitored continuously by Odalog and SmartReader from Nov. 29th to Dec. 6th, 2017 and Dec. 13th to Dec. 20th, 2017. Odalog was directly suspended 2 meters using a steel cable below the manhole cover. For the second round, water samples obtained from manholes in the trunk (T2 to T7, T9, T10) and main laterals (L8 and L12) on Sept. 27th, 2018. Besides, water samples were also taken from manholes (L2-1, L3-1, L4-1, L6, L7, L10, L11-1) before drops in the laterals on Sept. 19th, 2018. Odalog and SmartReader were installed in the trunk (T1~T7, T9~T10) and laterals (L1, L2~L14) from Sept. 18th to 25th, 2018 and Sept. 27th to Oct. 4th, 2018. All water samples were collected in planned locations. The wastewater samples from the trunk (T1~T10) and main laterals (L8 and L12) were analyzed to evaluate the general water characteristics in this area, as is shown in **Table 5-1**. The biofilm samples were collected in the first round by scraping using a spoon from the manhole wall 5 m above the bottom at the manholes T1 and T2, and from sewer wall above the water level interface at T1 and L8. For sediment collection, the sediment was quickly grabbed at the bottom of the trunk at T1. All the biofilm and sediment samples were analyzed through 16s rRNA for identification of microbial community.

5.2.2 Estimating overall transfer coefficient (K_La) at drops

The mass transfer rate across liquid-gas interface is based on the two-film theory (Lewis and Whitman 1924) and is expressed as:

$$\frac{dC}{dt} = -K_L a(C - C_S) \tag{5.1}$$

where

C is the concentration in the liquid phase (g/m^3)

C_S is the saturation concentration of a particular gas in the liquid phase (g/m^3)

K_La is the overall mass transfer coefficient at (h^{-1})

The average rate of the concentration change (or mass transfer rate) at a drop can be estimated as

$$\frac{dC}{dt} = \frac{C_u - C_d}{t_f + t_w} \tag{5.2}$$

where

C_u is the concentration at the upstream of the sewer drop (g/m^3)

C_d is the concentration at the downstream of the sewer drop (g/m^3)

t_f is the falling time (h)

t_w is the residence time of the tailwater (h)

The falling time is calculated according to free fall formula (Cooper 1935):

$$t_f = \sqrt{\frac{2H}{g}} / 3600 \tag{5.3}$$

where

H is the drop height (m)

g is the gravity acceleration constant 9.81 m/s^2

The residence time t_w is estimated using the distance between the two drops divided by the flow velocity, which stands for the travel time of turbulent sewage between two drops. Note that the inclusion of this residence time here is to account for the additional mass transfer occurred after the impingement of the falling sewage on the bottom and the enhanced turbulence during the transport in the trunk. The determination of this residence time is somewhat uncertain.

From equations (4.1) and (4.2), the overall mass transfer coefficient K_La at drops is expressed as

$$K_L a = -\frac{C_u - C_d}{(t_f + t_w)(C_u - C_s)} \tag{5.4}$$

5.3 Results and Discussion

5.3.1 Sulfide and H_2S in the trunk

The water samples from the trunk and major laterals were analyzed for the sulfide (TS and DS) and sulfide-related parameters and the results were presented in **Table 5-2**. The total sulfide (TS) and dissolved sulfide (DS) at T1 were 1.43 mg/L and 0.42 mg/L, respectively, in the first round which was formed at the upstream section of the trunk. The DS concentration in the two major laterals, L8 and L12, was low in the first round (0.00 and 0.31 mg/L) and zero in the second round. Therefore, only the sulfide built up at the upstream sections of the trunk mainly contributed the sulfide in the trunk.

The evolution of sulfide along the trunk can be seen in **Figure 5-2**. Both rounds of monitoring results show a consistent trend. The sulfide (TS and DS) increased from T1 to T3, while the dissolved oxygen (DO) slightly dropped. After T3, the sulfide fell all the way until the end of the trunk (T10) except for T8 in the first round and DO increased along the trunk until T9, then decreased again from T9 to T10. Since the retention time between T7 and T8 was rather short (0.9 min), which did not allow the formation of sulfide. The abnormal rise at T8 resulted from the diurnal variation. On the other hand, H_2S in the air phase along the trunk is shown in **Figure 5-3**. H_2S was present at manholes of T3, T4, T5, T6, T7, T8 and T9. H_2S gas was detected over the day with a diurnal variation and the highest value occurred around 12:00 AM. However, H_2S gas in the manholes of T1, T2 and T10 was zero. The air temperature was 13~15°C in the first round and the air temperature ranged from 9 °C to 13 °C in the second round (**Figure 5-3**).

As shown in **Table 5-2**, in the first round, DS concentration rose from 0.42 mg/L to 0.83 mg/L and DO concentration decreased from 1.5 mg/L to 1.2 mg/L from T1 to T3. The retention time along the nearly 1500 m trunk distance (T1-T3) allowed the generation of sulfide by sulfate-reducing bacteria (SRB) in the biofilm on the pipe wall and then transferred into water phase under the low DO level. The drop of DO was because of the activity of aerobic bacteria. At the beginning of the trunk, the flow at T1 and T2 was under normal flow conditions where the flow velocity was around 1.6 m/s. The DS in the sewage was low (less than 1.0 mg/L) and not much H_2S gas can be transferred into air phase. The transferred H_2S gas from the sewage can be completely absorbed and oxidized on the moist sewer and manhole wall in the deep trunk (around 26 m deep). There was no H_2S at manholes T1 and T2. However, it should be mentioned that H_2S was detected at 1 ppm and 1~2 ppm at T1 and T2 in the first 10 minutes when the manhole covers were opened to install Odalog and SmartReader. More sewer air moved to the top of the uncovered manholes which brought more H_2S gas to the top. The H_2S could not be completely removed by adsorption process. This phenomenon indicated that conditions close to equilibrium of H_2S between air and water phase rarely exist in real sewer networks because of adsorption and oxidation processes at sewer and manhole walls.

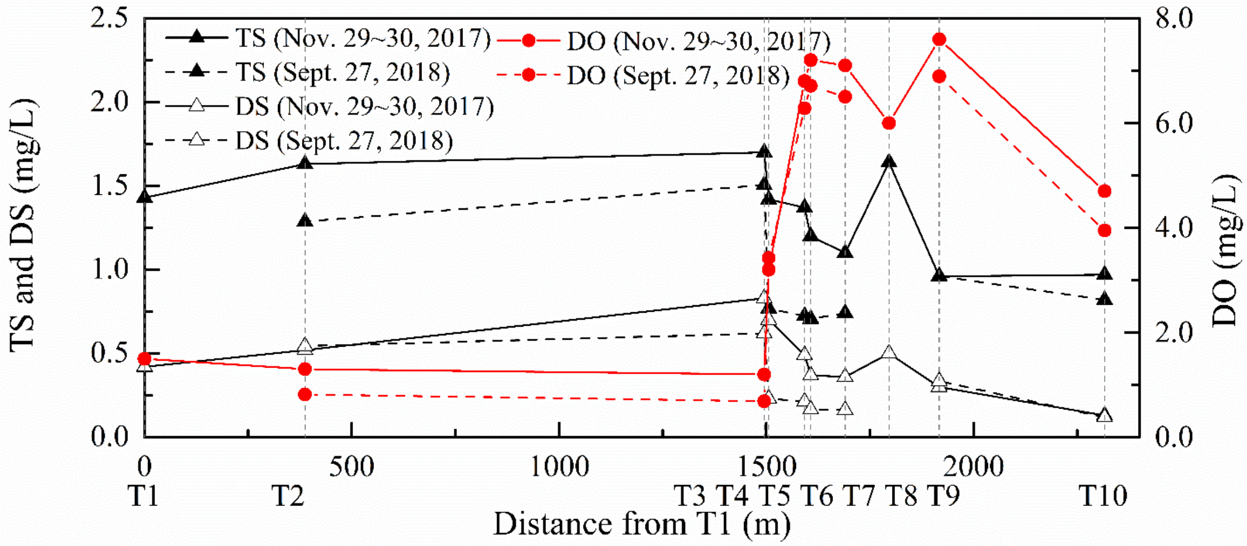


Figure 5-2 Variation of Total Sulphide (TS), Dissolved Sulphide (DS), and Dissolved Oxygen (DO) along the trunk

From T3 to T4, the concentration of TS and DS decreased to 1.42 mg/L and 0.70 mg/L, respectively, and DO increased to 3.2 mg/L at T4. The retention time from T3 to T4 was also too short (0.8 min) to generate sulfide. And the trunk sewer drops at T3-1 (2.7 m) and T4 (5.2 m) gave rise to the increased turbulence of the flow which accelerated mass transfer of both sulfide and oxygen between water and air phase. The release of H_2S gas and reaeration of oxygen led to the decrease in sulfide and increase in oxygen in the sewage. Similarly, the trunk sewer drop at T5 with a drop height of 2.0 m further released the sulfide and reaerated oxygen in the sewage. The TS and DS dropped to 1.20 mg/L and 0.37 mg/L, respectively, and the DO increased to 6.8 mg/L at T5. The release of H_2S by these drops resulted in the high H_2S gas concentration at T3, T4 and T5. The average H_2S gas was about 15 ppm at these locations.

Because of the high DO level in the sewage after reaeration process at these drop structures, part of sulfide was oxidized. In addition, the sulfide kept being stripped into the air along the trunk (T6~T10). Sulfide concentrations in the liquid phase were observed to decrease as the wastewater continued downstream. The TS and DS at T10 decreased to 0.82 mg/L and 0.13 mg/L respectively.

By section T9 DO decrease was observed (from 7.6 mg/L at T9 to 4.6 mg/L at T10) likely due to bacterial respiration. The released H_2S in the air phase at drops was transported downstream and dropped because the process of adsorption, and oxidation of H_2S on the moist pipe and manhole wall was dramatic. So the H_2S gas concentration kept decreasing from T5 to T10 (**Figure 5-3**). The average H_2S gas concentration was 10 ppm, 3 ppm, 5 ppm, 1.5 ppm and 0 ppm at T6, T7, T8, T9 and T10, respectively. The H_2S at T8 was higher than that at T7 likely because the air pressure at T8 (24 Pa) was higher than that at T7 (12 Pa) (**Figure 5-4**) and more H_2S gas could be moved to the top the manhole T8.

The second round of field work gave the similar results as the first round (**Figure 5-2** and **Figure 5-3**). The sulfide generated from the upstream sections of the trunk was not high and part of sulfide was built up at the upstream trunk (T1-T3) under low DO concentrations. No H_2S was detected at the manholes under normal gravity pipe flow (T1-T2) since the sulfide in the sewage was low (DS less than 1 mg/L). However, high H_2S gas was detected at the drops (T3-T5) under the same low sulfide concentration in the sewage and the released H_2S could be transported downstream to make them as hotspots (T6-T9). The trunk sewer drops played an important role on H_2S release resulting in relatively high H_2S gas concentration despite the rather low sulfide concentration in the sewage.

Table 5-1 Mean wastewater characteristics based on the samples obtained in trunk (T1~T10) and main laterals (L8 and L12)

Date	NH_4-N	NO_3-N	TN	TP	TCOD	DCOD	Sulfate	TSS	VSS	VFA	pH	T
	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg/L)	(mg S/L)	(mg/L)	(mg/L)	(mg/L)		(°C)
Nov. 2017	49	0.89	67	24	748	260	53	276	220	78	7.5	12.9
Sept. 2018	44	0.43	64	22	620	227	58	185	157	52	8.0	14.8

Table 5-2 Water quality parameters in the trunk sewer and main laterals

Locations	Sampling#1 Nov. 29, 30*, 2018	Sampling#2 Sept. 27, 2018	TS#1	TS#2	DS#1	DS#2	Sulfate#1	Sulfate#2	DO#1	DO#2
	Time	Time	(mg/L)		(mg/L)		(mg S/L)		(mg/L)	
T1	10:55 AM	N/A	1.43	N/A	0.42	N/A	40	N/A	1.5	N/A
T2	2:28 PM*	11:40 AM	1.63	1.29	0.52	0.54	130	47	1.3	0.8
T3	3:57 PM	1:38 PM	1.70	1.51	0.83	0.62	52	45	1.2	0.7
T4	1:33 PM*	9:43 AM	1.42	0.77	0.70	0.23	39	46	3.2	3.4
T5	12:20 PM*	10:46 AM	1.37	1.32	0.49	0.21	64	61	6.8	6.3
T6	2:55 PM	11:08 AM	1.20	0.87	0.37	0.17	66	46	7.2	6.7
T7	3:30 PM	3:21 PM	1.10	1.14	0.36	0.29	51	157	7.1	6.5
T8	2:11 PM	N/A	1.64	N/A	0.50	N/A	37	N/A	6.0	N/A
T9	1:50 PM	11:45 AM	0.96	1.36	0.30	0.74	48	41	7.6	6.9
T10	12:47 PM	1:44 PM	0.97	0.82	0.13	0.12	38	42	4.7	4.0
L8	12:00 PM*	2:21 PM	1.16	0.79	0.00	0.04	54	68	8.5	7.3
L12	12:00 PM	1:28 PM	0.74	0.17	0.31	0.00	24	32	1.2	7.3

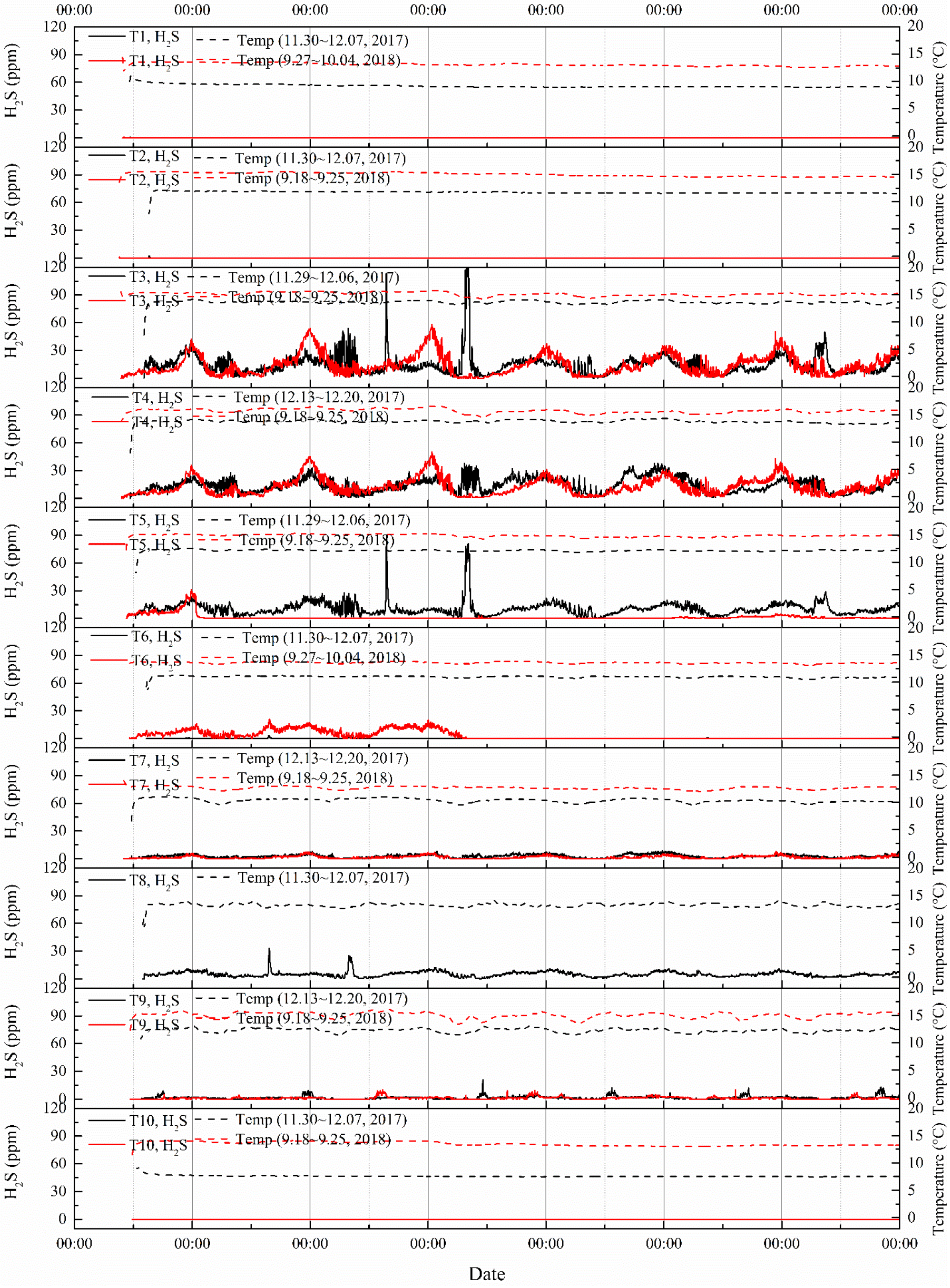


Figure 5-3 Monitored H_2S gas concentration and air temperature in the trunk from T1 to T10

5.3.2 H_2S and oxygen transfer at drops

As shown in **Figure 5-3**, the H_2S and O_2 concentration in the water phase at the upstream (C_u) and downstream (Cd) of each drop based on the field measurements of the oxygen (C_{O2}) and dissolved sulfide concentration in the sewage (shown in **Table 5-2**). The H_2S gas (C_{H2S}) in the sewage then was determined by dissociate equilibrium between dissolved sulfide and H_2S gas in the sewage. H_2S gas concentration in the sewer air was measured in the field (**Figure 5-3**) and oxygen concentration in the sewer air is assumed to be 21%. Then the saturation concentration of H_2S ($C_{S(H2S)}$) and oxygen ($C_{s(O2)}$) in the sewage were determined by Henry's law with H_2S and O_2 concentration in the sewer air. The total stripping time (t) at drops consists of falling time (t_f) and residence time (t_w). t_f is the falling time at drop. The residence time t_w accounts for the additional mass transfer occurred after the impingement of the falling sewage on the bottom and the enhanced turbulence during the transport in the trunk which is estimated using the distance between the two drops divided by the flow velocity. Finally, the overall oxygen and H_2S mass transfer coefficient ($K_La_{(O2)}$ and $K_La_{(H2S)}$) at drops in the trunk were estimated using Eq. 5.4, and the results are shown in **Table 5-3**.

The overall oxygen and H_2S mass transfer coefficient was around 200 h^{-1} and 300 h^{-1} with drop height 2.7 m at T3-1 followed by drop height 5.2 m at T4. The oxygen in the sewage increased by approximately 3 mg/L and half of the H_2S (0.05 mg/L) in the sewage was released into air phase which resulted in 15 ppm H_2S gas in the sewage headspace. The overall oxygen and H_2S mass transfer coefficient was around 70 h^{-1} and 54 h^{-1} with drop height 2.0 m at T5. Combining this additional drop effect, another 3 mg/L O_2 was added into the sewage after T5 and more H_2S was stripped into air phase to almost reach equilibrium concentration in the sewage. In particular, the H_2S transfer coefficient at T5 during the second field work was positive which indicated much more H_2S entered into air phase and the H_2S in the liquid phase was lower than that in the air phase after the first two drops (T3-1 and T3), no H_2S was released into air phase at T5.

Compared to normal gravity pipe, the coefficient at the drops was relative high. The oxygen transfer coefficient was in the range of 3~50 h^{-1} in gravity sewers with different slope and flow rate (Lahav et al. 2006). Yongsiri et al. (2004) demonstrated that a constant ratio of $(K_La)_{H2S}$ to $(K_La)_{O2}$ was 0.86 ± 0.08 at 20°C in gravity sewers, which was independent of the degree of mixing. So the H_2S overall mass transfer coefficient in the normal gravity sewer is also in the same range as O_2 transfer coefficient. In addition, the overall mass transfer coefficient dramatically increased with the drop height. The number from T3-1 to T4 was much higher than that at T5. Drop height has a very significant influence on reaeration and H_2S stripping efficiencies at drop structures. Matias et al. (2017) studied the effect of the characteristic of drops on reaeration and the O_2 overall mass transfer coefficient ranged from 10 h^{-1} to 90 h^{-1} with different tailwater depth and drop height. The highest number was 88.7 h^{-1} with the lowest tailwater (0.06 m) and highest drop height (1.16m). The drops in the field are in larger scale in terms of drop height, pipe size, flow rate and tailwater and overall mass transfer coefficient is larger compared to that lab study results.

Table 5-3 Calculated $(K_La)_{O2}$ and $(K_La)_{H2S}$ at drops from the field investigation（#1 and #2 indicate two field measurements in 2017 and 2018, respectively）

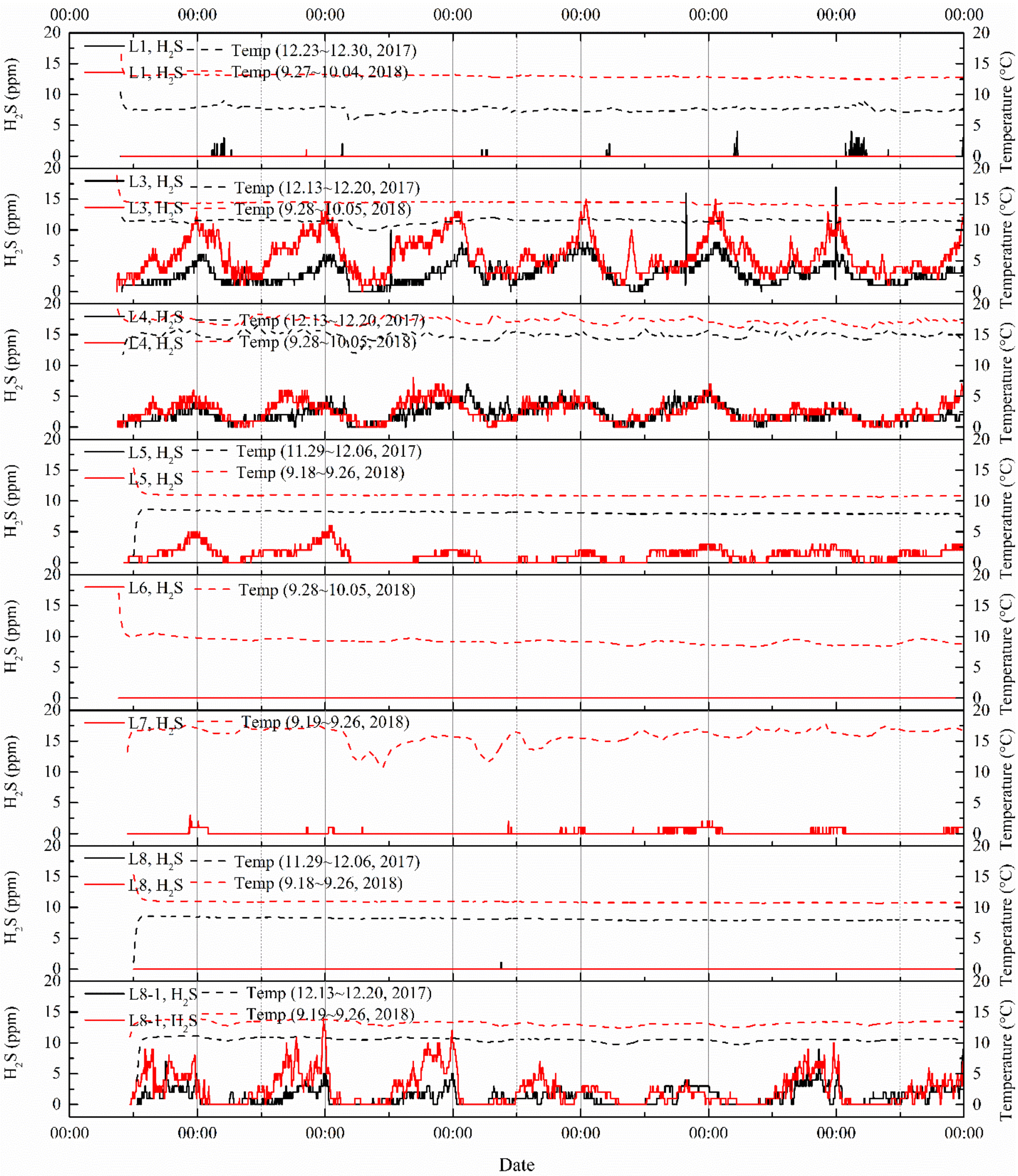
Loc.	Drop (m)	t_f (h)	t_w (h)		C_u #1 (mg/L)	C_u #2 (mg/L)	C_d #1 (mg/L)	C_d #2 (mg/L)	K_La #1 (h^{-1})	K_La #2 (h^{-1})
T3-1	2.7	2.1×10^{-4}	1.2×10^{-4}	O_2	1.15	0.69	N/A	N/A	N/A	N/A
				H_2S	0.114	0.070	N/A	N/A	N/A	N/A
T4	5.2	2.9×10^{-4}	12×10^{-4}	O_2	N/A	N/A	3.23	3.42	190	234
				H_2S	N/A	N/A	0.078	0.018	180	431
T5	2.0	1.8×10^{-4}	N/A	O_2	3.23	3.42	6.75	6.28	68	58
				H_2S	0.078	0.018	0.055	0.012	54	23

5.3.3 H_2S in the laterals

The concentration of H_2S gas in the headspace of the laterals connected to the trunk was continuously monitored and the results are shown in **Figure 5-4**. H_2S existed at the manholes of L1, L3, L4, L5, L7, L8-1, L9 and L10. No H_2S gas was detected at L6, L8, L11, L12-1 and L13. The formed DS along the major lateral L8 was stripped into the air phase by the drop (10.9 m) at T8-L1 to make it as location of sewer odor concern. The DS concentration in the sewage at L8 was zero after release (**Table 5-2**), so the H_2S in the air phase was also zero. Liquid samples were taken from the upstream manhole of these laterals to identify where the detected H_2S came from. The results are shown in **Table 5-4**. The DS in all the sampling manholes was close to zero under high DO concentration (>4 mg/L). The H_2S detected in those laterals was transported to these locations from the trunk. The air pressure in the trunk and laterals were continuously monitored for two weeks and the average value at each location was calculated, as is shown in **Figure 5-4**. The air pressure in the trunk was higher than that in the neighboring laterals. The air pressure in the trunk T1~T3 was around 60 Pa and the air pressure in laterals of L1, L3 and L4 was 59 Pa, 0.4 Pa and 46 Pa. Likely T9 had the air pressure of 0 Pa, while its nearby lateral L10, had air pressure of -4 Pa. The air pressure difference between the trunk and the laterals could push the air into the laterals which brought the H_2S into these laterals to make them locations of odor concerns.

Table 5-4 TS, DS and DO of water samples from laterals (2018.09.19)

Locations	Time	TS (mg/L)	DS (mg/L)	DO (mg/L)
L2-1	9:05 AM	0.28	0.05	5.6
L3-1	9:30 AM	0.57	0.06	4.2
L4-1	9:50 AM	0.02	0.00	5.6
L5-1	10:10 AM	no water available		
L6	10:25 AM	0.31	0.00	4.5
L7	10:50 AM	0.58	0.04	6.7
L10	1:30 PM	0.51	0.01	6.2
L11-1	12:50 PM	0.71	0.07	5.4



00:00
H$_2$S (ppm)
Temperature (°C)
L1, H$_2$S – Temp (12.23~12.30, 2017)
L1, H$_2$S – Temp (9.27~10.04, 2018)
L3, H$_2$S – Temp (12.13~12.20, 2017)
L3, H$_2$S – Temp (9.28~10.05, 2018)
L4, H$_2$S – Temp (12.13~12.20, 2017)
L4, H$_2$S – Temp (9.28~10.05, 2018)
L5, H$_2$S – Temp (11.29~12.06, 2017)
L5, H$_2$S – Temp (9.18~9.26, 2018)
L6, H$_2$S – Temp (9.28~10.05, 2018)
L7, H$_2$S – Temp (9.19~9.26, 2018)
L8, H$_2$S – Temp (11.29~12.06, 2017)
L8, H$_2$S – Temp (9.18~9.26, 2018)
L8-1, H$_2$S – Temp (12.13~12.20, 2017)
L8-1, H$_2$S – Temp (9.19~9.26, 2018)
0
5
10
15
20
Date

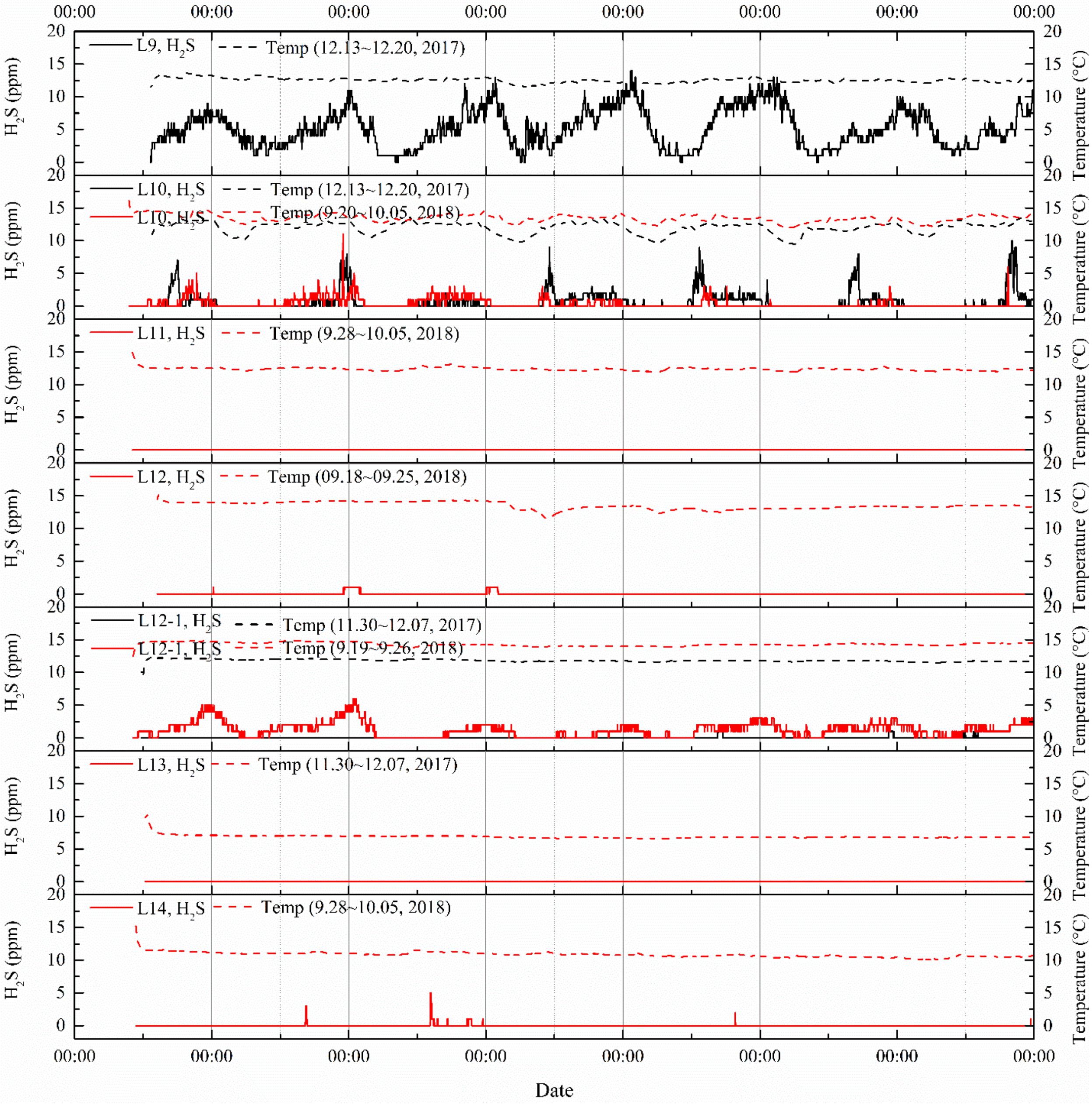


Figure 5-4 Monitored H_2S gas concentration and air temperature in the laterals

5.3.4 Microbial community structures in biofilms

The microbial community of the biofilm samples at different locations in the sewer system was analyzed via Illumina MiSeq sequencing. **Figure 5-5** presents the relative abundance of the microbial community at the genus level. *Desulfomicriobium*, a known SRB was identified in the biofilm on sewer pipe wall at T1, L8 and in the sediment at T1 with relative abundance of 0.3%, 0.97% and 0.01%. *Desulfomicriobium* belongs to the family of *Desulfomicrobiaceae,* and most of these species use pyruvate and lactate to reduce sulfate to sulfide (Watanabe et al. 2017). *Thiobacillus*, known as SOB, were identified as dominant species (relative abundance >5%) in both manhole wall samples at T1 and T2 with abundance of 14.2% and 6.51%. *Thiobacillus* are reported to be responsible for both producing sulfuric acid and deteriorating concrete in sewer systems (Wei et al. 2010). In terms of other samples, *Pseudomonas* (33.5%) was dominant genus in sewer biofilm sample at T1. The predominant bacterial population in sewer biofilm sample at L8 was comprised of *Arcobacter* (7.3%) and *Paludibacter* (5.6%) as well as *Acidovorax* (5.3%) and *Faecalibacterium* (6.6%) in sewer biofilm sample at T1. *Pseudomonas* are widespread in nature (water and soil) and can produce a wide range of extracellular polymeric substances which are involved in attachment processes and biofilm formation (van Delden 2004). *Arcobacter and Faecalibacterium* species are highly abundant in raw sewage as pathogens (Fisher et al. 2014). The genera *Paludibacter* is strictly anaerobic and chemoorganotrophic, which is able to utilize melibiose, glycogen and soluble starch as growth substrates (Gronow et al. 2011). *Acidovorax* genus is affiliated to class of *Comamonadaceae,* which is isolated in activated sludge as denitrifiers (Heylen et al. 2008).

Only one SRB genera (*Desulfomicrobium*) was identified in low relative abundance in the sewer wall biofilm and sediment samples at T1 and T2. The low proportions and diversity of SRB indicate aerobic operational environment under low oxygen with low concentrations of H_2S in the sewage system. Most of bacteria in sewer manhole biofilm were unclassified and *Thiobacillus* represented one of the dominant microbial communities, which have been implicated in the corrosion of concrete surfaces. So *Thiobacillus* is the major key community member contributing to sewer pipe corrosion in this area. There are severe manhole corrosion issues in the trunk especially at location of T3-T5 where H_2S gas concentration was high. The rest of the genera previously recognized to comprise a dramatic portion of biofilm/sediment microbial community in the sewer were present (Gao et al. 2016, Liang et al. 2016).

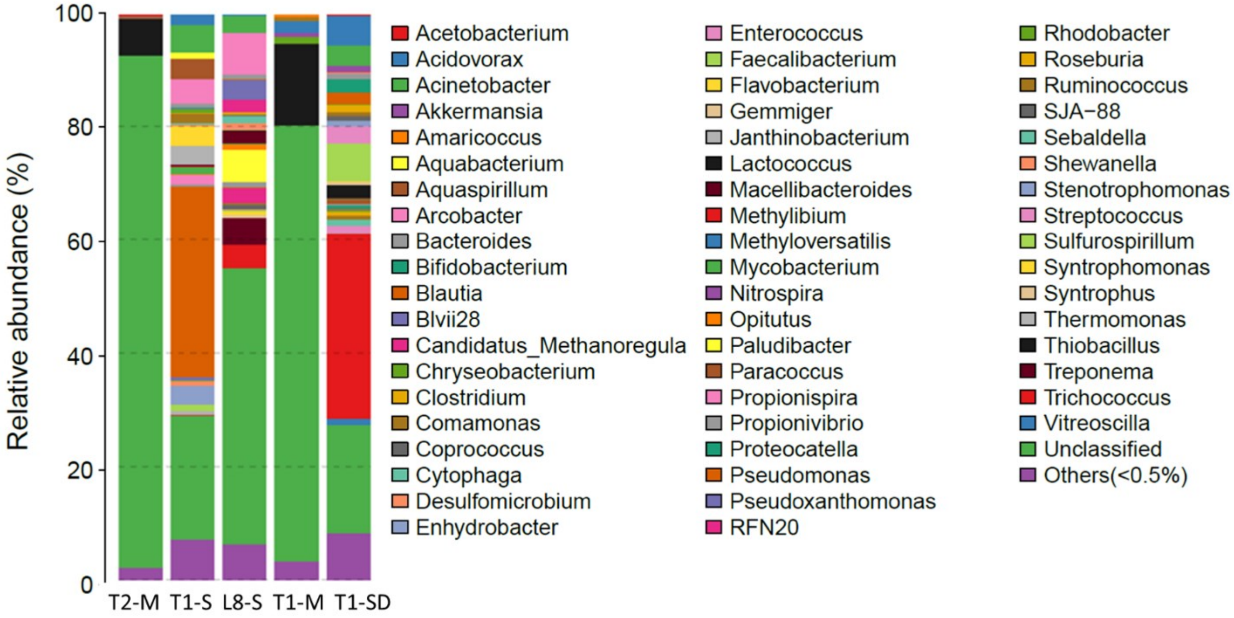


Figure 5-5 Relative abundance of microbial communities at genus level in biofilms at different locations (M indicates manhole wall, S indicates sewer wall, SD indicates sewer sediment)

5.4 Conclusions

Two rounds of field work were carried out in Bonnie Doon area, Edmonton, Alberta, Canada, to study the generation, release and transport of hydrogen sulphide in the trunk sewers with three drops.

The results from both rounds were similar: the sulfide generated from the upstream sections of the trunk was not high (DS less than 0.5 mg/L) and part of sulfide could be built up in the upstream trunk (T1-T3) under low DO level (less than 1.5 mg/L). No H_2S was detected in the manholes under normal gravity pipe flow (T1-T3) since the sulfide in the sewage was low (DS less than 1 mg/L). However, high H_2S gas concentration was detected at the drops (T3-T5) with high turbulence under the same low sulfide concentration in the sewage with average H_2S of 15 ppm. The released H_2S at drops was transported downstream to make them as the locations of odor concerns (T6-T9) for sewer odor complaints. The average H_2S gas concentration was measured at 10 ppm, 3 ppm, 5 ppm and 1.5 ppm at T6, T7, T8 and T9, respectively. The drops in the sewer system played an important role on H_2S release and rather low sulfide could still result in high H_2S gas concentrations locally. The overall oxygen and H_2S mass transfer coefficient at drops was estimated to be much higher than that in normal gravity sewers and the overall mass transfer coefficient dramatically increased with the drop height. The overall H_2S mass transfer coefficient was around 300 h^{-1} with drop height 2.7 m at T3-1 followed by drop height 5.2 m at T4 and 54 h^{-1} with drop height 2.0 m at T5. The H_2S detected in those laterals was transported to these locations from trunk due to the air movement in the sewer network which made some of them as hotspots. *Desulfomicrobium* was identified as the only SRB in sewer wall biofilm, but not in richness, *Thiobacillus* was the main populations contributing to sewer pipe corrosion in this area, which was predominant in biofilm on the manhole wall. According to the field results, two odor control strategies could be applied in this case, either retrofitting the three drops in the trunk to reducing the stripping effect of these drops or removing the sulfide in the sewage in the trunk right before these three drops by adding chemicals like nitrate, ferric/ferrous and hydrogen peroxide et..

Chapter 6 Effect of Ferric and Nitrate on Hydrogen Sulfide Control in Lab-Scale Reactors

6.1 Introduction

Hydrogen sulfide, produced by sulfate-reducing bacteria (SRB) in sewers under anaerobic conditions, can lead to sewer odors, corrosion and health hazards. Pump stations are commonly used in sewer systems, for example, in the City of Edmonton, Alberta, Canada there are over 90 pump stations in its sanitary sewer system. The long force mains at most of the pump stations have long hydraulic retention times (HRT) which can lead to the formation of sulfide in the force mains. As SRB are slow growers, they are mostly abundant in biofilms at pipe wall. Biofilms on the pipe wall of force mains provide an ideal environment for sulfide production as it is always submerged in wastewater containing high organic matters and a limited amount of oxygen. Multiple species of SRB have been identified at different environment, including *Desulfovibrio*, *Desulfobacter*, *Desulfobulbus*, *Desulfomicrobium* and *Desulfotomaculum* (Castro et al. 2000, Okabe et al. 1999).

A number of chemical treatments are currently used by wastewater industries worldwide, to prevent or control sulfide generation in sewer systems (Park et al. 2014, Zhang et al. 2008). These chemicals can be divided into following categories: nitrate that provides electron acceptor and suppresses sulfate reduction, oxidizing chemicals (H_2O_2, NaClO, $KMnO_4$, oxygen/air) that oxidize sulfide, iron salts (Fe^{2+} or Fe^{3+}) that precipitate sulfide, basic chemicals ($Mg(OH)_2$, NaOH, $Ca(OH)_2$) that elevate sewage pH and shift the chemical equilibrium from more volatile H_2S towards more non-volatile species (S^{2-}, HS^-). These strategies effectively prevent sulfide formation in wastewater. Selecting effective and economical chemicals in order to remove sulfide is crucial.

Addition of nitrate has been proven to be effective to control sulfide production. A few mechanisms of addition of nitrate for sulfide control have been proposed: (1) nitrate addition induces the heterotrophic nitrate reduction process and the SRB is suppressed by the interspecies competition between heterotrophic nitrate reducing bacteria (hNRB) and SRB for common carbon source (Hubert and Voordouw 2007); (2) addition of nitrate favors biological oxidation of sulfide to elemental sulfur or sulfate by means of nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB) such as *Thiomicrospira denitrificans*, some strains of *Thiomicrospira sp.*, *Thiobacillus sp.*, and

Arcobacter sp. (Gieg et al. 2011, Li et al. 2009); (3) the nitrite accumulation as an intermediate of the nitrate reduction process can inhibit SRB activities (Fida et al. 2016, Greene et al. 2003).

To date, the petroleum industry has also significantly contributed to our understanding of sulfide control with nitrate at laboratory or reactor scale (Gieg et al. 2011). Diverse 16S rRNA gene sequences affiliating with hNRB such as *Marinobacter*, *Marinobacterium*, and *Halomonas* spp. have also been detected in oilfield enrichment cultures with nitrate control (Al-Tamimi and Mehdi 2017). Microbial control of biogenic production of hydrogen sulfide in oil fields was studied and NR-SOB oxidizing sulfide in the presence of sufficient nitrate led to its complete removal from the environment (Nemati et al. 2001). The effect of nitrate addition in an artificial souring experiment, using diluted crude oil as substrate and electron donor was examined and the addition of nitrate resulted in the significant growth of the nitrate-reducing bacteria (NRB) *Thalassospira* sp. (Fan et al. 2020). Studies performed on oil reservoirs with regard to the sulfide control with nitrate injection have pointed out the highly significant role of hNRB and NR-SOB in the microbial community. How the nitrate achieves sulfide control in sewers and the involving microbial community need to be investigated. Besides, previous studies in sewers indicated that continuous dosing of nitrate in the force main was required to achieve completely sulfide control which implied the high demand of nitrate (Jiang et al. 2013, Mohanakrishnan et al. 2009a). More work is needed to develop cost-effective nitrate dosing strategy in an effort to save nitrate demand according to nitrate control mechanism.

Sulfide precipitation by addition of iron have been extensively used for sulfide control in sewers. The method relies on the ability of dissolved sulfide (H_2S and HS^-) to form insoluble iron sulfides, thereby preventing emission of gaseous hydrogen sulfide into the sewer atmosphere. Iron salts may be added either as ferrous or ferric salts. The required ratio between iron salts and sulfide is usually high and all these ratios are much higher than the stoichiometric requirement. This indicates a very high dosage of iron salts (Firer et al. 2008, Padival et al. 1995). However, several recent studies on the effect of ferric salt dosing on sulfide were performed in sewer biofilms (Ganigué et al. 2018, Zhang et al. 2009), in which the addition of ferric chloride was found to reduce sulfide production in the anaerobic sewer biofilm of force main by more than 50%. This indicates ferric addition may inhibit the SRB in the biofilm which can potentially reduce the ferric dosage for sulfide control in force main. Ferric may be a promising cost-effective chemical for sulfide control.

Four sewer biofilm reactors were constructed in this study to mimic force mains conditions. Four reactors were running for 8 months to reach steady state. Ferric and nitrate were then injected in the reactors. The objective of this study was (1) to quantify the sulfide generation rates in the lab-scale reactors; (2) to identify the inhibitory effect of ferric salt addition on SRB in biofilm of the reactor; (3) to investigate the effectiveness of nitrate dosing for controlling sulfide in force main and identify the cost-effective dosing strategy; (4) to characterize the control mechanism of sulfide production by identifying the shift of microbial community by ferric and nitrate addition.

6.2 Methodology

See section 3.3

6.3 Results and Discussion

6.3.1 Sulfide generation rate in lab-scale bioreactors

Four bioreactors imitating force mains had been running for 8 months to develop mature and black biofilm and reach stabilization. **Figure 6-1** shows the bioreactors R1 and R2 were stabilized in the last month of 8-month startup period (R3 and R4 were the same, but not shown in the figure). During the steady period, all the 50 mg/L sulfate was reduced into sulfide by SRB after 12 h hydraulic retention time in four reactors by utilizing the DCOD. The DCOD was consumed by 130 ~180 mg/L. The batch tests were undertaken in four bioreactors to identify the sulfide generation rate (**Figure 6-2 (a) and (b)**) and the results indicated four bioreactors had varying sulfide generation rate even though they were running under the same condition. R1 had the highest generation rate, it only took 5 h to completely convert 50 mg/L sulfate to 50 mg/L sulfide. And the effluent DCOD was around 140 mg/L. While R2 had the lowest generation rate among them, the reduction of 50 mg/L sulfate took over the 12 h period and the effluent DCOD was around 190 mg/L. The generation rate of R3 and R4 were in between.

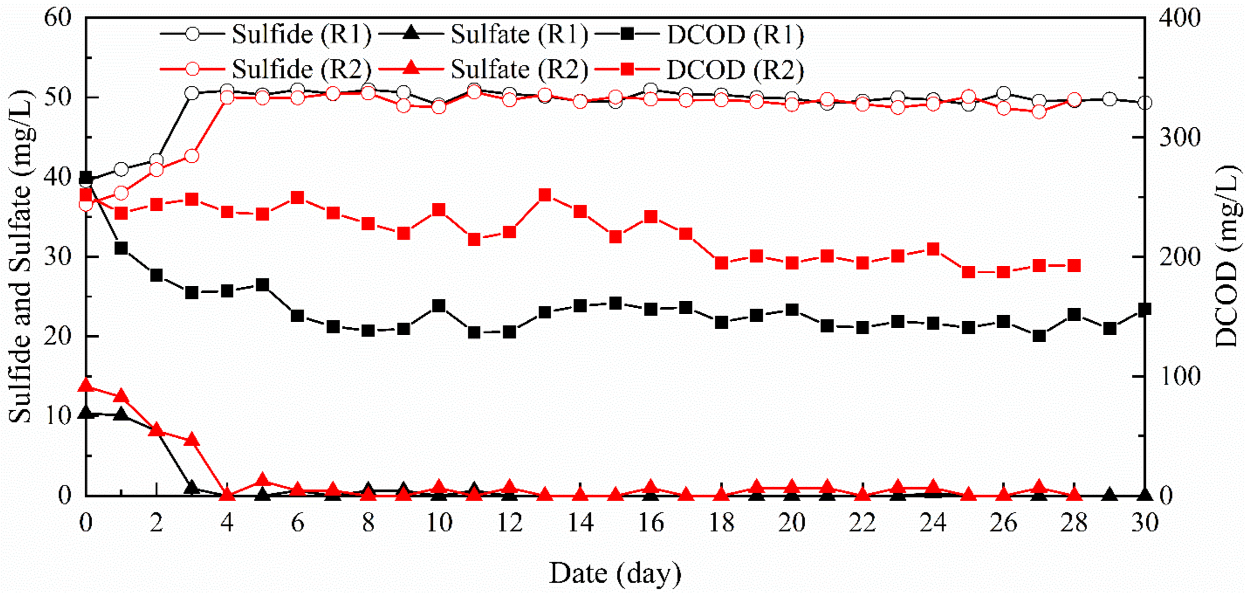


Figure 6-1 Effluent sulfide, DCOD and sulfate in bioreactor R1 and R2 after 12 h pump cycle at the last month of start-up phase

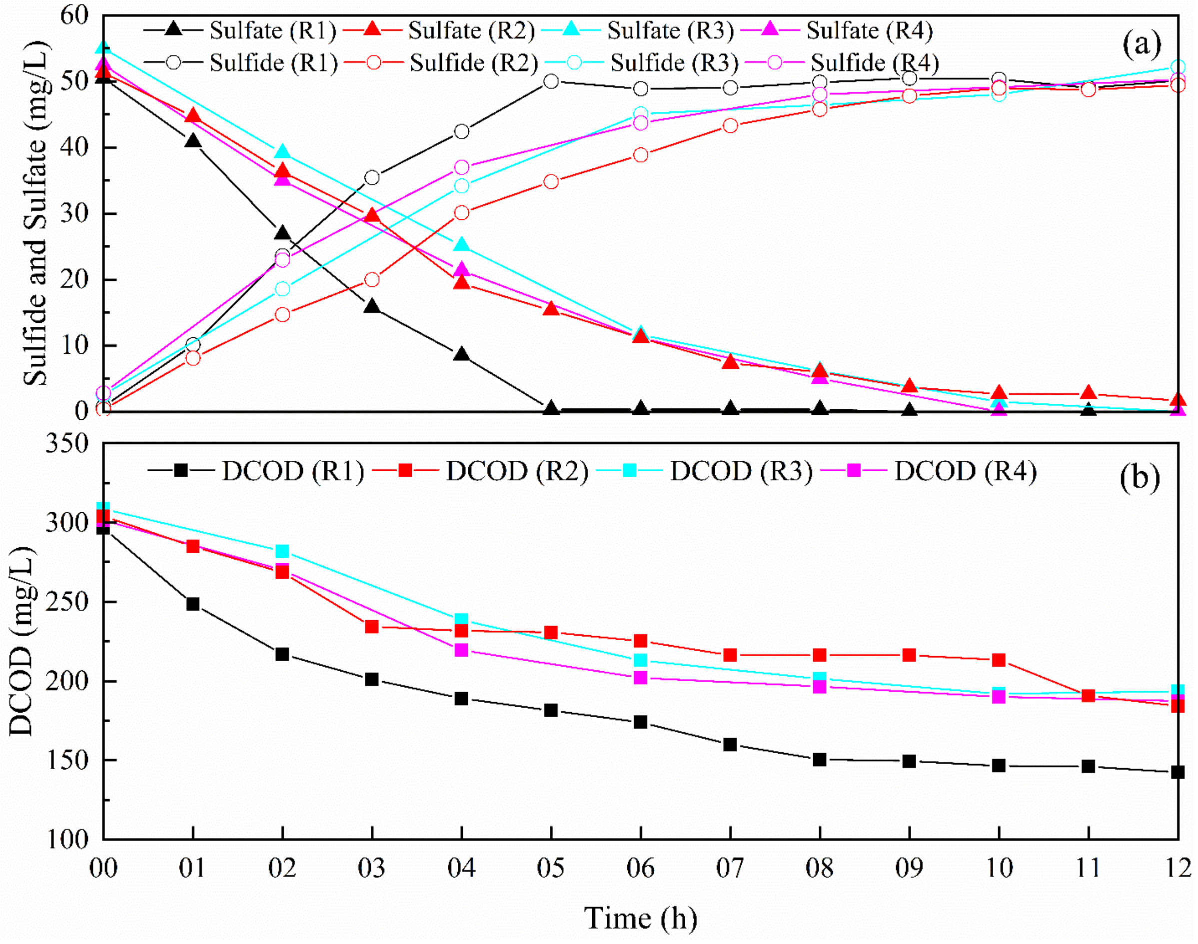


Figure 6-2 Change of sulfide, sulfate (a) and DCOD (b) in the batch test in four bioreactors during 12 h pump cycle

Sulfate and DCOD are the electron acceptor and donor for SRB reduction process. In order to determine which one is limiting sulfide formation, the sulfate concentration in R1 was increased to 90 mg/L. As shown in **Figure 6-3**, 80 mg/L sulfide was formed when initial sulfate concentration was 90 mg/L and the effluent DCOD was around 150 mg/L. The sulfide generation rate under 90 mg/L sulfate was close to that under 50 mg/L sulfate in the first 6 h, which suggests that sulfate was not limiting sulfate formation (**Figure 6-4**). After 6 h, the sulfate was used up under initial sulfate concentration of 50 mg/L but sulfide continued to be generated under initial sulfate of 90 mg/L until the end of the cycle.

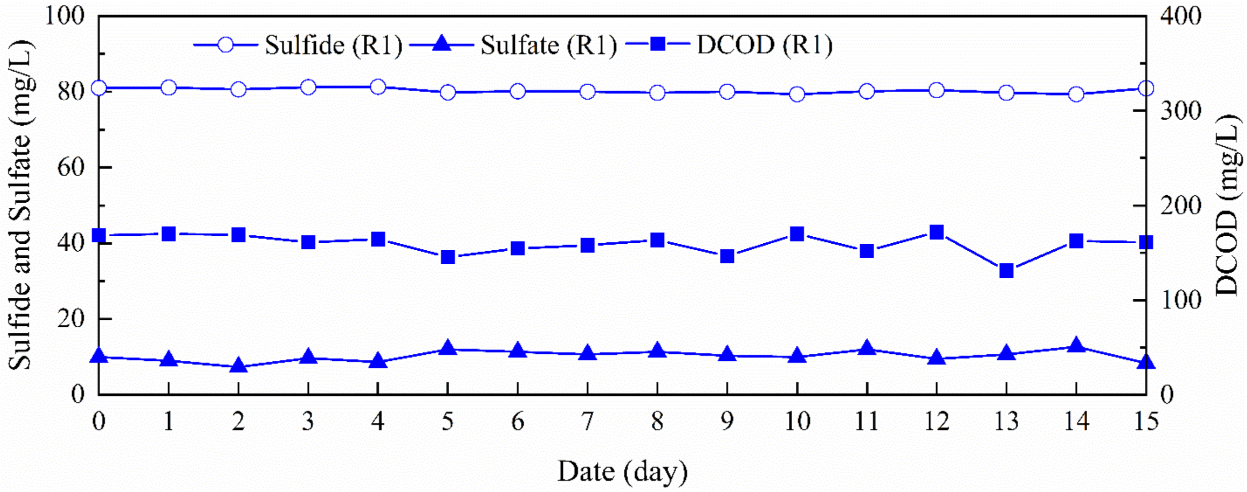


Figure 6-3 Effluent sulfide, DCOD and sulfate in bioreactor R1 after 12 h pump cycle with 90 mg/L sulfate in influent

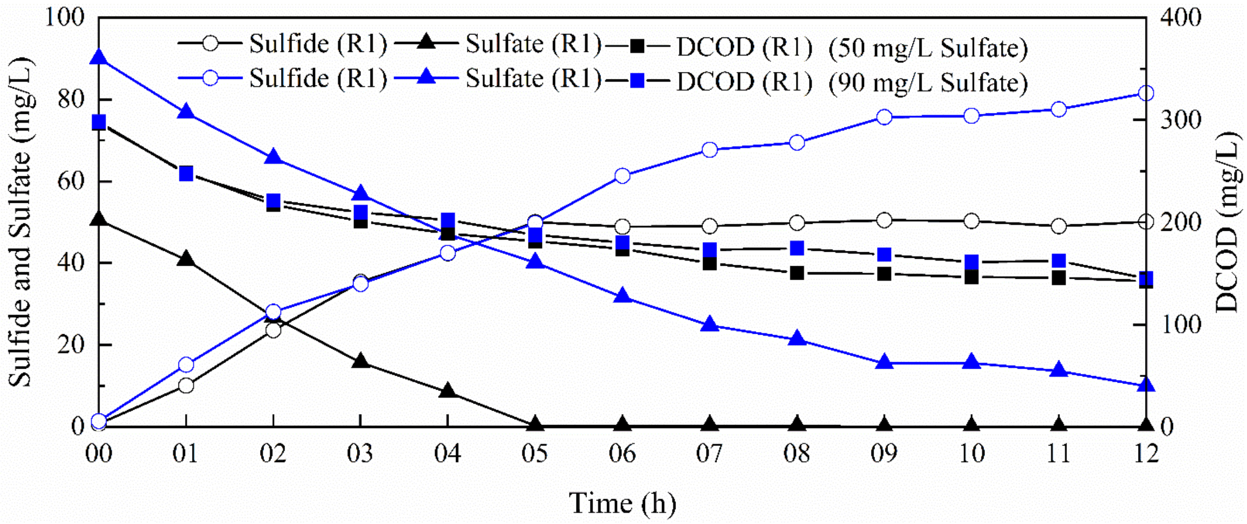


Figure 6-4 Change of sulfide, sulfate and DCOD in the batch test in bioreactor R1 under 50 mg/L and 90 mg/L sulfate in influent during one 12 h pump cycle

The four reactors running under identical conditions had different sulfide generation rates. The potential production of sulfide depends on the thickness of the biofilm. The biofilm thickness of four reactors varied which led to different amount of SRB in each reactor. The potential production of sulfide depends on the anaerobic thickness of the biofilm. The biofilm thickness can vary over time due to the detachment and regrowth processes. Sulfate is typically found in all types of municipal wastewater and experiment results show that sulfate is not limiting sulfide formation when it is larger than 5–15 mg/L. So biodegradable organic matter is limiting factor for the sulfate-reducing bacteria.

The sulfide generation rate in four bioreactors with different DCOD in batch test was calculated, as shown in **Figure 6-5**. Nielsen and Hvitved-Jacobsen (1988) proposed that 1/2-order biofilm kinetics with regard to DCOD is suitable to describe the sulfide formation in force main (**Equation (6.1)**), while Sharma et al. (2008) proposed to use Monod kinetics with saturate constant (k). With around 5 mg DCOD/L saturation constant (k) in Sharma et al.'s study, the k value was insignificant compared with DCOD concentration. The Monod kinetics can be described by zero order kinetics with regard to DCOD. Mitchell (Mitchell 1971) demonstrated the biofilm kinetic can be described by zero order for full penetrated biofilm and half order for a partly penetrated biofilm. Applying both equations to fit the measured generation rate points in four bioreactors, the half order kinetics had better fit than Monod kinetics especially when the DCOD was less than 200 mg/L. It indicated that the partly penetrated biofilm with half kinetics was more suitable to describe the sulfide generation rates in the bioreactors. As is seen in **Figure 6-5**, the rate constant for R1 is 0.009 and for R2, R3 and R4 is around 0.006 for half order biofilm kinetics. The modelled rate constant was in the range of 0.005~0.011, higher than the value (0.003~0.006) proposed by Hvitved-Jacobsen et al. (Hvitved-Jacobsen et al. 2013). These numbers are useful to determine the sulfide generation rate in Edmonton's force mains.

$$r_a = a(DCOD-50)^{0.5}1.03^{T-20} \tag{6.1}$$

where, a is an empirical rate constant (g S g $O_2^{-0.5}m^{-0.5}h^{-1}$)

DCOD is dissolved COD (g O_2 m^{-3})

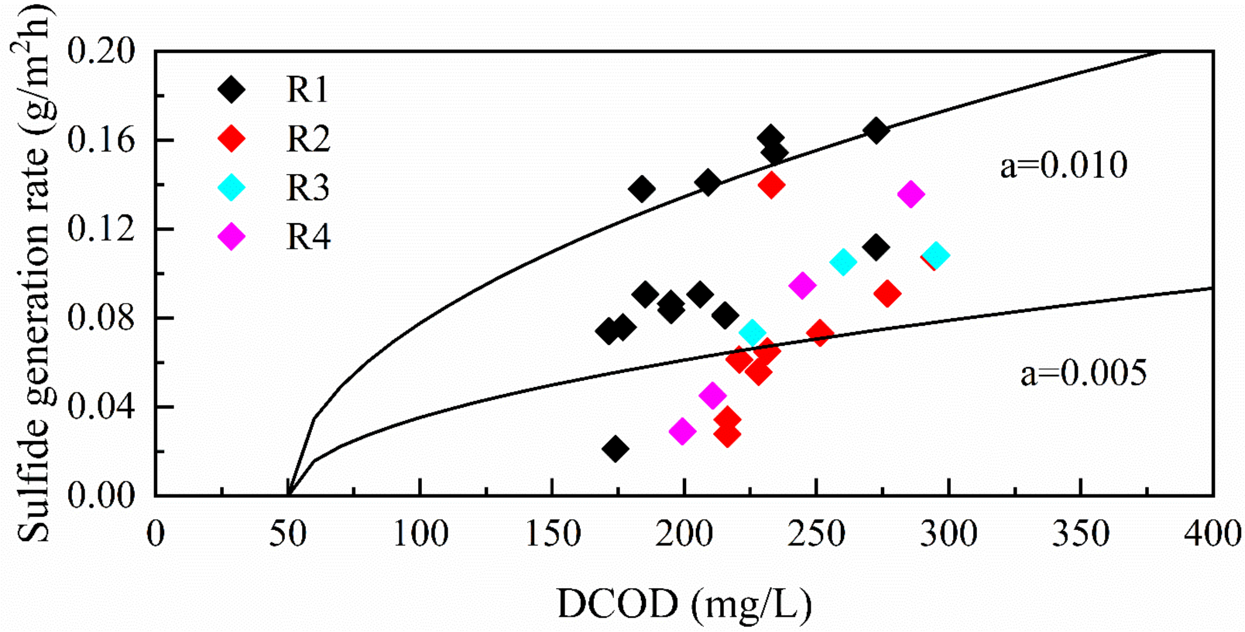


Figure 6-5 Sulfide formation rate versus DCOD in the four bioreactors

6.3.2 Effect of ferric on sulfide control

Ferric was added in R1 to control sulfide generation. The results indicate that there was still around 15 mg/L sulfide formed under 50 mg/L ferric dosage (**Figure 6-6)**. The corresponding effluent sulfate and DCOD was 0 mg/L and 65 mg/L, respectively. Changing ferric dosage to 85 mg/L, sulfide was completely removed with 0 mg/L. The corresponding effluent sulfate and DCOD was 0 mg/L and 50 mg/L, respectively. The remaining Fe was 1~10 mg/L. Batch tests in R1 was performed with ferric addition (**Figure 6-7 (a) and (b)**). The 50 mg/L ferric was depleted in the first 3 hours and the sulfide started to form from 3 h upon the depletion of iron salt until the end of the cycle. When the ferric dosage was 85 mg/L, the iron salt was quickly dropped in the first hour to 20 mg/L and then decreased slowly to 10 mg/L until the 10th h. Finally, the iron salt maintained at 10 mg/L after 10 h until the end of the cycle. In both cases, sulfate dropped to 0 mg/L at 6 h which was the same as pre-dosing phase. The DCOD decreased significantly in the first hour and then decrease gradually until the end of the 12-h cycle.

The ferric effectively suppressed the hydrogen sulfide formation. Hydrogen sulfide control by ferric dosage was mainly due to chemical sulfide oxidation and precipitation. The addition of iron had no effect on the formation rate of sulfide. The sulfide generation still occurred by reducing the sulfate under ferric dosing. It took 6 h to completely reduce the sulfate, which was the same as pre-dosing phase. The ferric ions were chemically reduced to ferrous, which can subsequently precipitate sulfide to generate black FeS precipitate. In addition to reacting with sulfide, the ferric could also react with organic substance in wastewater which increased the amount of ferric required to suppress the hydrogen sulfide generation in the force mains. Therefore, the DCOD quickly dropped in the first hour. The ferric addition in R1 was ceased after three months, as shown in **Figure 6-8**. Subsequently, sulfide concentration rapidly jumped back to pre-dosing level after 2 days. Ferric did not have long-lasting inhibitory/toxic effect on sulfate reduction and did not shift the major SRB populations in the biofilm of force mains. In order to completely control the sulfide generation in force mains, continuous dosing of ferric is required.

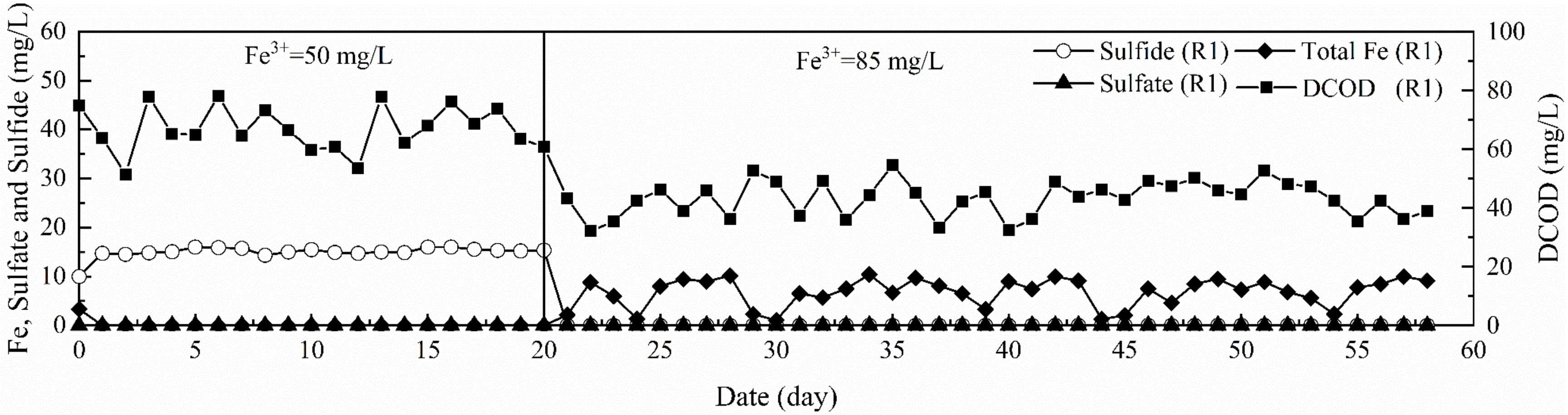


Figure 6-6 Effluent sulfide, sulfate DCOD and total Fe in bioreactor R1 after 12 h pump cycle with different ferric dosage

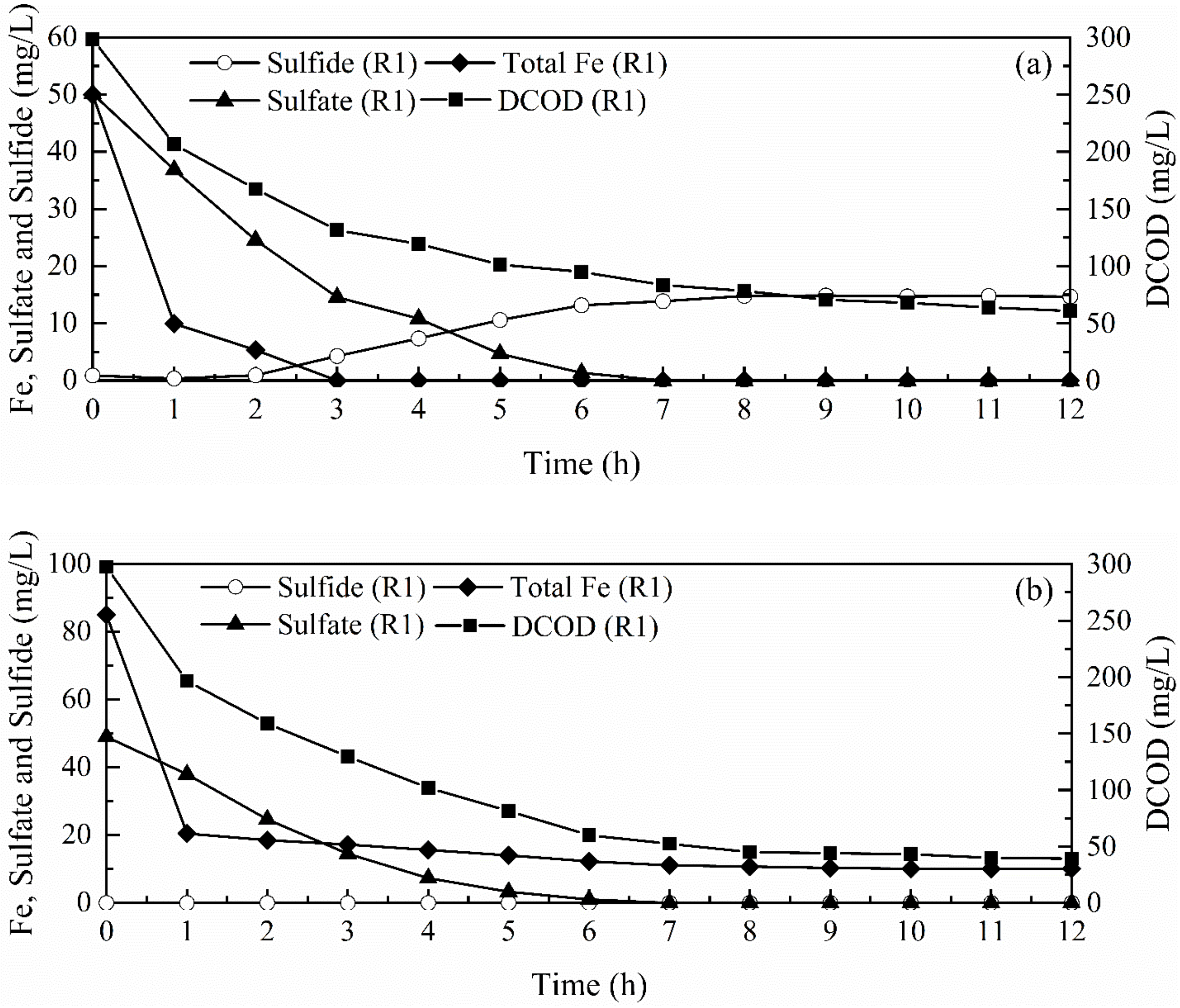


Figure 6-7 Change of sulfide, sulfate, DCOD and total Fe in the batch test in bioreactor R1 under 50 mg/L (a) and 85 mg/L (b) ferric dosage

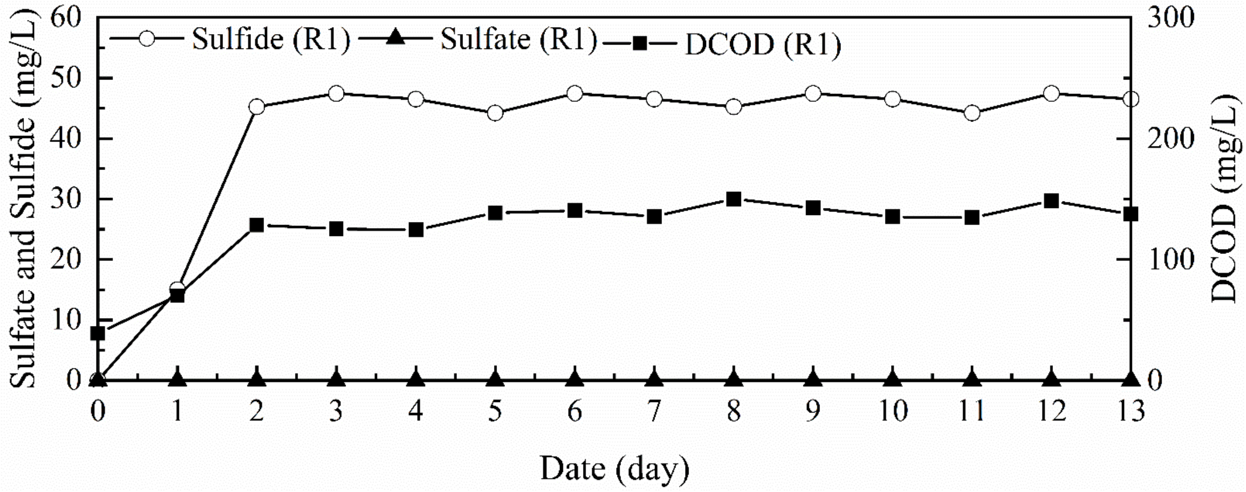


Figure 6-8 Recovery of the sulfide generation in R1 after stopping ferric dosing

6.3.3 Effect of nitrate on sulfide control and nitrate dosing optimization

Nitrate was injected in R2 to control sulfide generation. **Figure 6-9 (a)** shows that there was still around 20 mg/L sulfide formed under 67 mg/L nitrate dosage and the residue sulfate was 30 mg/L. Adjusting nitrate dosage to 134 mg/L, sulfide was completely suppressed and the sulfate was 50 mg/L same as influent. However, nitrate was overdosed and there was approximately 30 mg/L in excess. Adjusting the dosage twice, the optimal nitrate dosage for R2 was found at around 100 mg/L. Under this dosage, no sulfide being formed and remaining nitrate was 1~6 mg/L. The DCOD dramatically decreased with the addition of nitrate (**Figure 6-9 (b)**). As more nitrate was added, more DCOD was consumed which implies that the high DCOD in wastewater can result in a need for greater concentrations of nitrate for sulfide control. The effluent DCOD was 80 mg/L and 35 mg/L when the nitrate dosage was 67 mg/L and 100 mg/L. Batch tests in R2 was undertaken under 67 mg/L and 100 mg/L nitrate dosage (**Figure 6-10 (a) and Figure 6-10 (b)**). The 67 mg/L nitrate was almost depleted in the 1st h and the sulfide started to form from 2nd h when the nitrate was depleted until the end of the cycle. When the dosage was 100 mg/L, the nitrate was quickly consumed in the first 3 hours and then dropped slowly to 6 mg/L at 9th h. Finally, the nitrate maintained at 6 mg/L after 9 h until the end of the cycle. Nitrite was only detected when nitrate was present and no nitrite accumulated. The DCOD decreased significantly while nitrate was consumed, which was proportional to the amount of nitrate consumed. When the DCOD was around 50 mg/L, no nitrate was consumed at all. It seems the denitrification process terminates when the DCOD is lower than 50 mg/L.

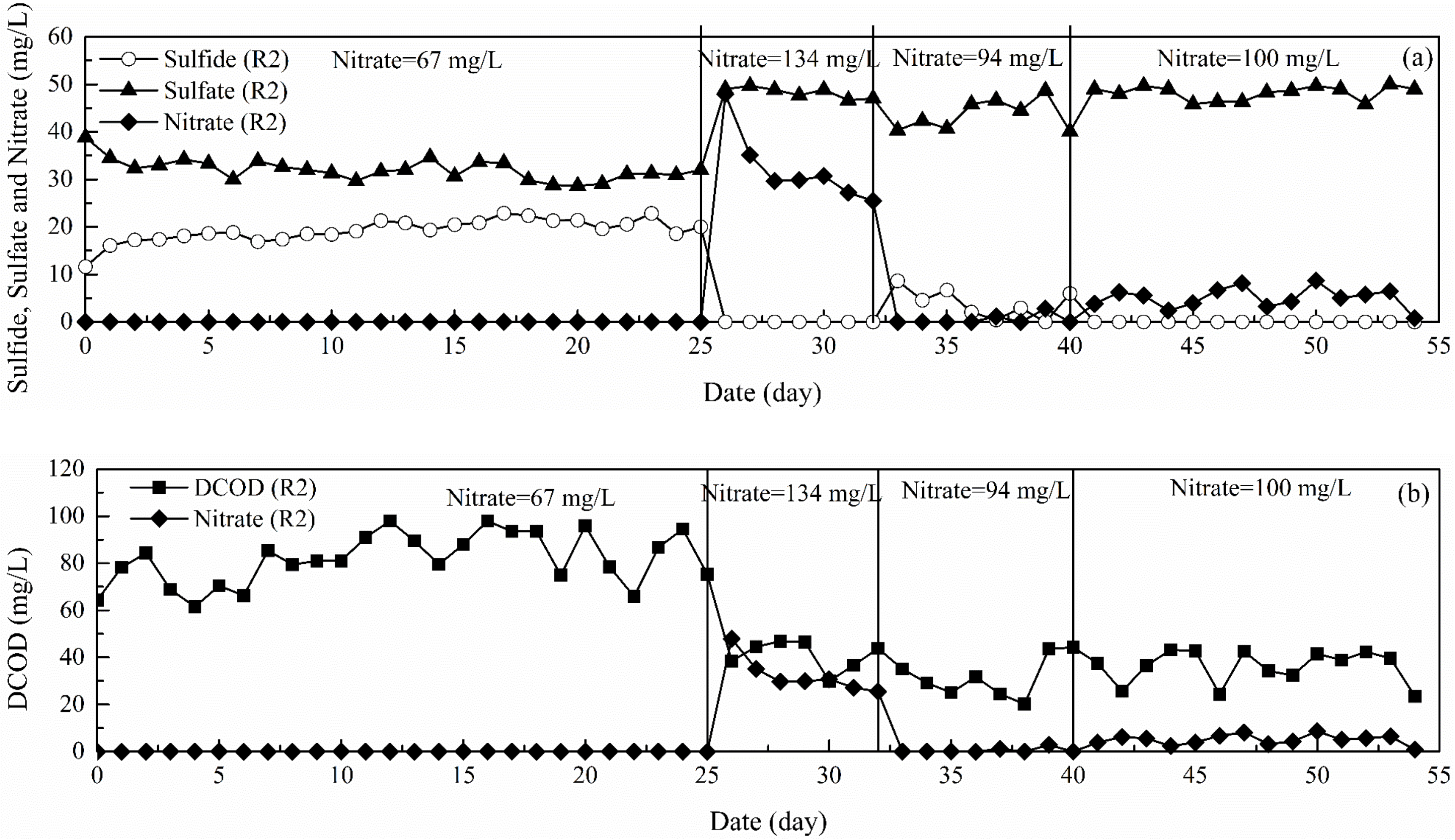


Figure 6-9 Effluent sulfide, sulfate nitrate (a) and DCOD (b) and in bioreactor R2 after 12 h pump cycle with different nitrate dosage

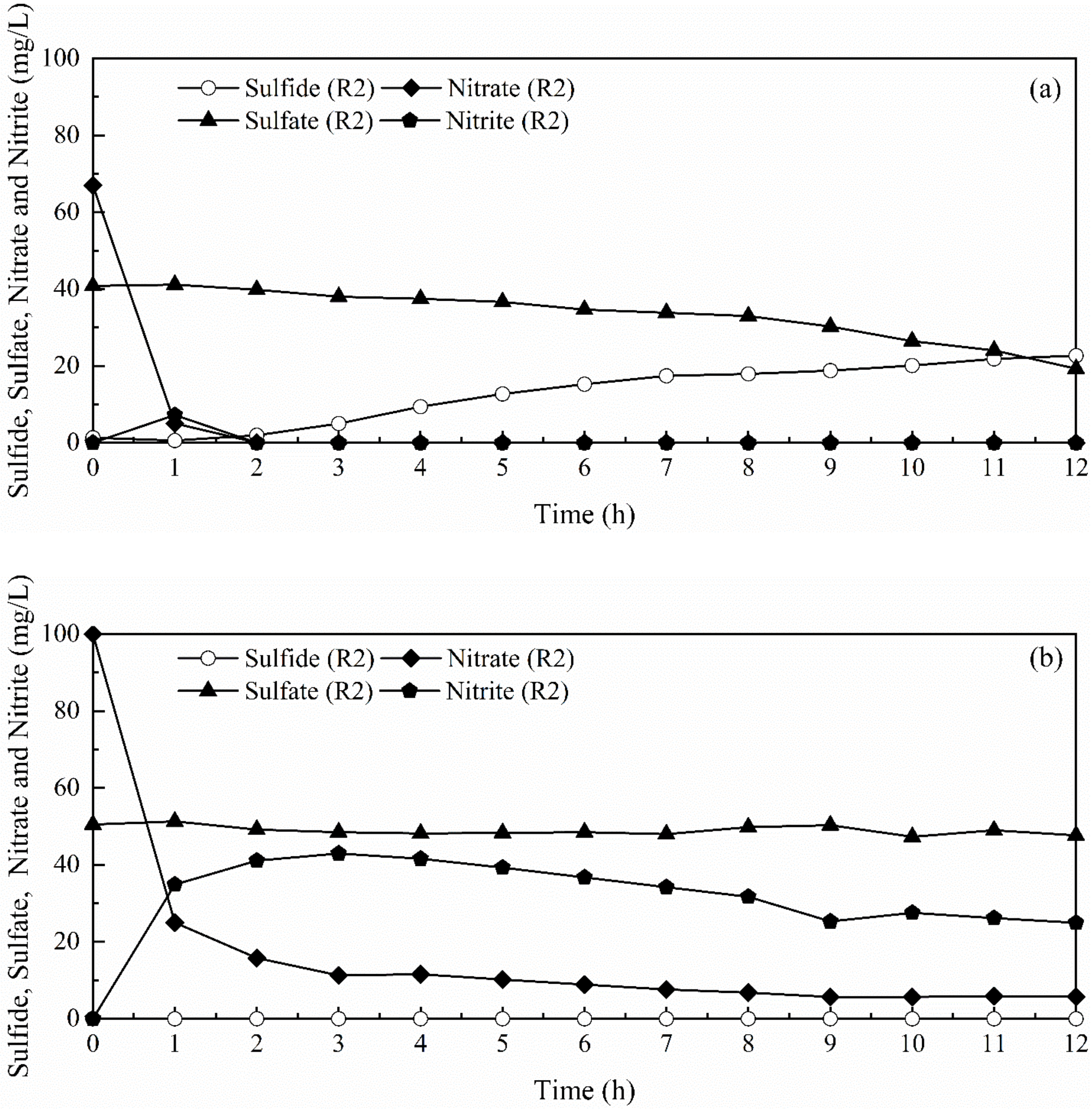


Figure 6-10 Change of sulfide, sulfate, nitrate and nitrite in the batch test in bioreactor R2 under (a) 67 mg/L and (b) 100 mg/L nitrate dosage

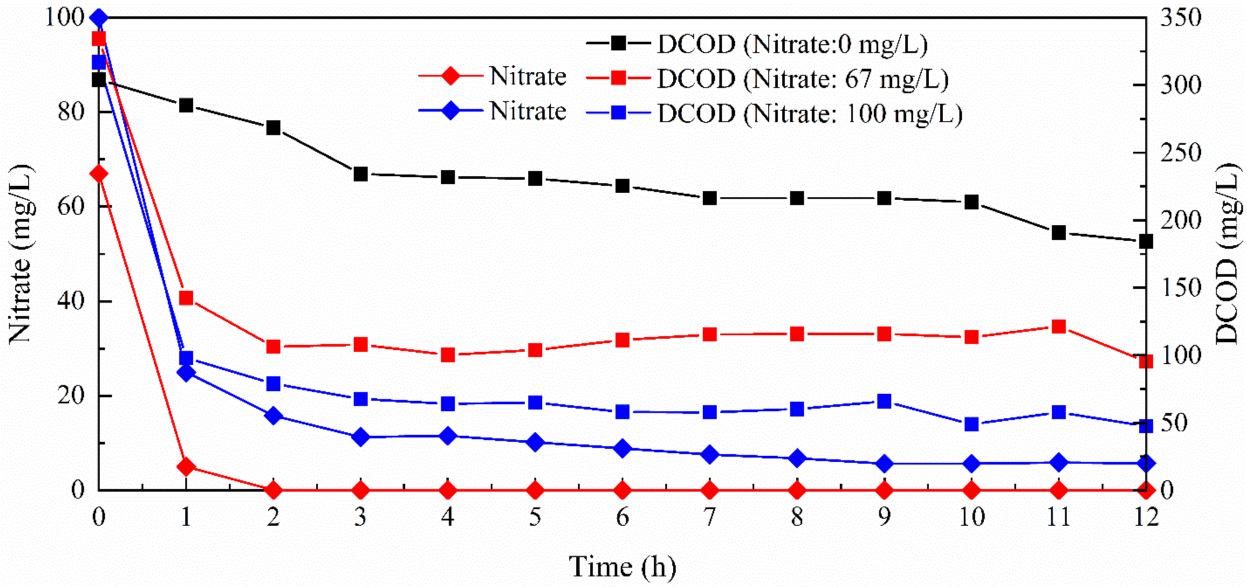


Figure 6-11 Change of nitrate and DCOD in the batch test in bioreactor R2 under 0 mg/L, 67 mg/L and 100 mg/L nitrate dosage

Sulfide control was achieved through nitrate addition. However, sulfide was observed to accumulate upon depletion of nitrate. Nitrite did not accumulate beyond 10 mg/L during the process. The proposed mechanism for nitrite accumulation as an intermediate of the nitrate reduction process can inhibit SRB activities was not observed in the bioreactors. Our observations support the conclusion that nitrate addition can induce the development of heterotrophic nitrate reducing bacteria (hNRB) (Zhang et al. 2018). The SRB can then expected to be suppressed by the interspecies competition between heterotrophic hNRB and SRB for common carbon (organic electron donors). Nitrate was mainly consumed for heterotrophic denitrification process by utilizing DCOD. Mathioudakis and Aivasidis (2009) demonstrated that heterotrophic denitrification in force main can be well described by half order kinetics. Heterotrophic denitrification depends on the availability of DCOD and the denitrification terminates when the DCOD is lower than 50 mg/L (**Figure 6-11**). So modified half order kinetics with respect to DCOD is proposed to predict the heterotrophic denitrification rate in the force main (**Equation (6.2)**). The modelled empirical rate constant b was around 0.05 (**Figure 6-12**).

$$r_b = b(\mathrm{DCOD}-50)^{0.5} 1.05^{T-20} \tag{6.2}$$

where, b is empirical rate constant (g N g $O_2^{-0.5} m^{-0.5} h^{-1}$)

DCOD is dissolved COD (g O_2 m^{-3})

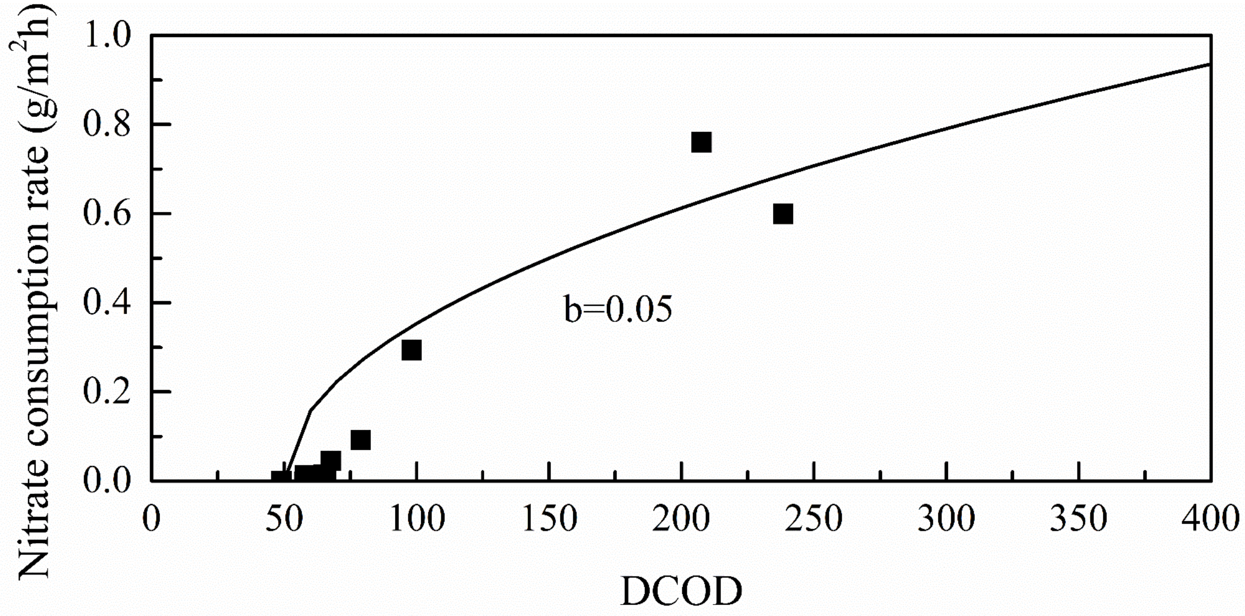


Figure 6-12 Nitrate consumption rate versus DCOD in bioreactor R2

The nitrate dosing in R2 was terminated after three months, as shown in **Figure 6-13**. Sulfide rapidly recovered after 4 days, reaching 60% of the initial level only after 1 day. Nitrate did not have long-lasting inhibitory/toxic effect on sulfate reduction.

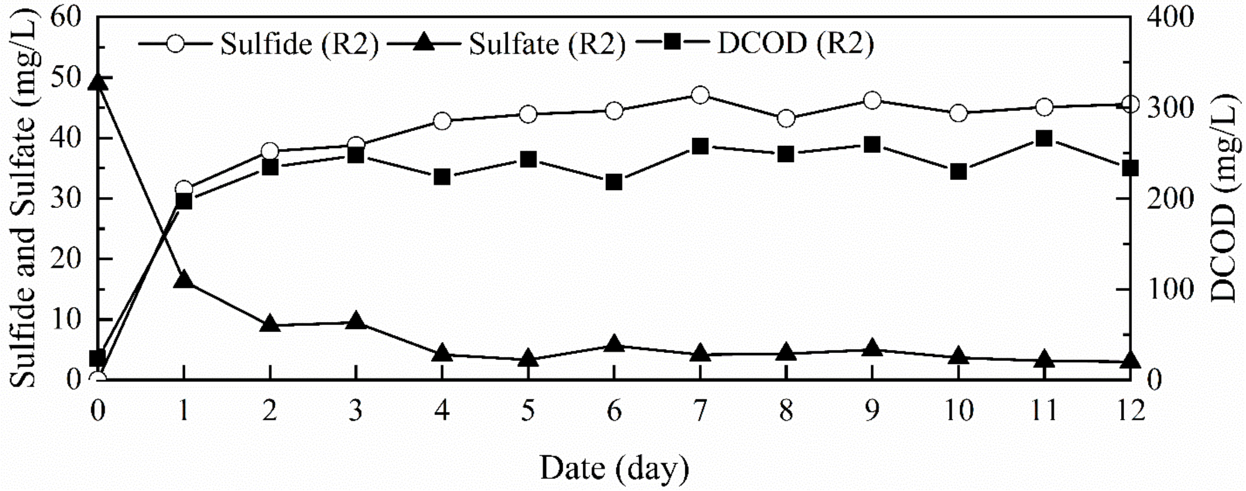


Figure 6-13 Recovery of the sulfide generation in R2 after stopping nitrate dosing

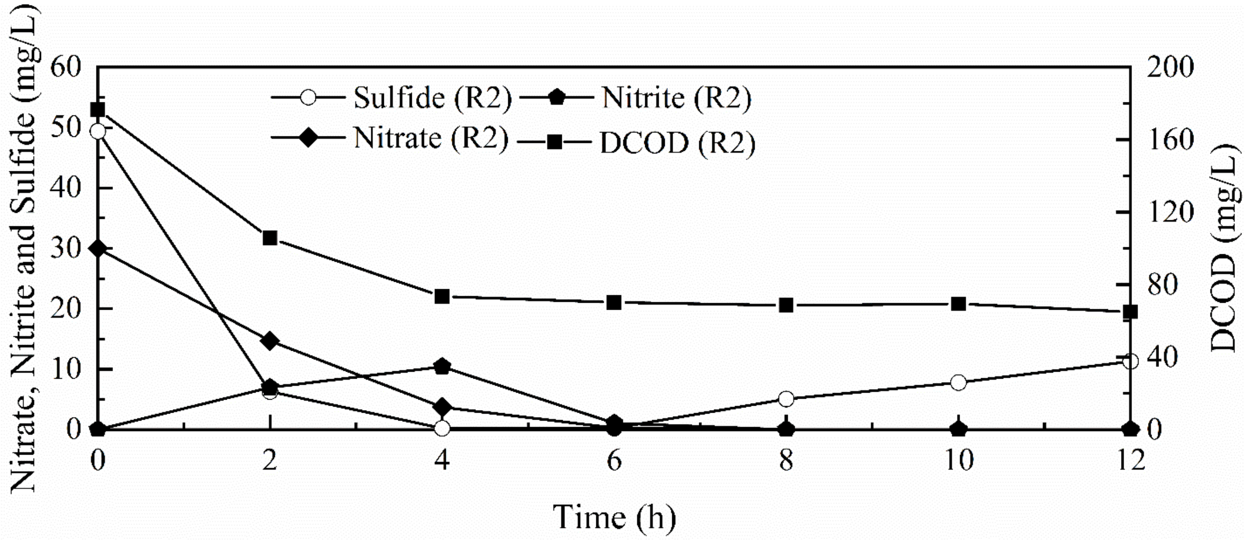


Figure 6-14 Change of sulfide, sulfate, nitrate and nitrite in the batch test in bioreactor R2 with nitrate dosage at end of the pump cycle instead of at the beginning of the pump cycle

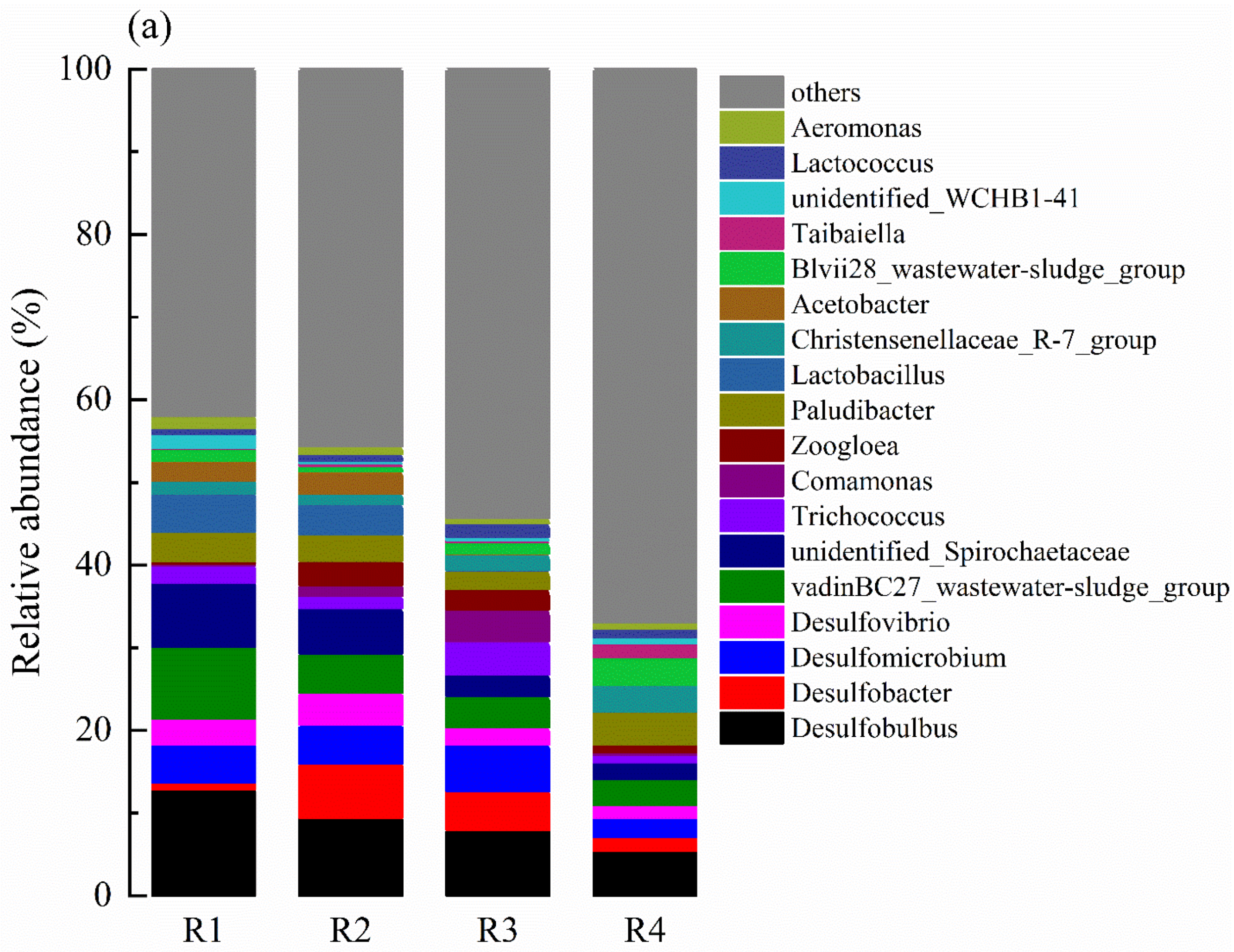
From a practical point of view, it is important to reduce the demand for nitrate to control the sulfide generation in force mains, since nitrate is a relatively expensive chemical to use for sulfide control. The nitrate is normally dosed at wet wells. However, the main limitation of this approach is that nitrate must be continuously kept through the pipeline, otherwise H_2S build-up resumes immediately after the depletion of the dosed nitrate. This implies very high costs in chemicals, since the presence of nitrate has to be ensured along the sewer pipeline. The addition of nitrate at a point close to the end of the pipe may be a better dosing strategy.

When nitrate was added into the R2 at the end of the 12 h pump cycle, the batch test demonstrates the formed 50 mg/L sulfide within the 12 h in the reactor could be biologically oxidized in presence of nitrate to 0 mg/L after 4 h HRT in the reactor (**Figure 6-14**). Only around 25 mg/L nitrate was consumed during the 4-hour period which was only one fifth of that (100 mg/L) when the nitrate was added at the beginning of the pipe (**Figure 6-10 (b)**). The nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB) prevailed when sulfide and nitrate was co-existing. NR-SOB can remove the existing sulfide by oxidizing it to sulfate and elemental sulfur (S°), using nitrate as the electron acceptor to produce primarily nitrite and N_2 (Garcia-de-Lomas et al. 2007, García de Lomas et al. 2006). The autotrophic denitrification process was dominant in the R2. The HRT which allowed the complete oxidation of sulfide generated was shorter (4 h) than that during the whole pump cycle (12 h) when dosing nitrate at the end of pump cycle. Therefore, the total amount of nitrate required to control the sulfide in the force main was considerably reduced. Nitrate consumption was reduced by about 75% while still ensuring complete suppression of sulfide. Adding nitrate at the downstream section of the force main is potentially a highly cost-effective dosing strategy. With this strategy, sulfide formation was still happening at the upstream section of the pipe not being exposed to nitrate with 8 h HRT but was immediately consumed as soon as passing through the downstream sections with 4 h HRT where nitrate was dosed. The nitrate dosing at downstream of the force main while ensuring having adequate HRT to completely biologically oxidized formed sulfide is a cost-effective dosing strategy for sulfide control. The disadvantage of this dosing strategy is that the upstream section of the force main (8 h) which allows the formation of sulfide still has the corrosion potential due to the high H_2S generated at this section. In addition, the implementation of this dosing strategy in the field needs to be further investigated.

6.3.4 Variation in the microbial community structure

Prior to chemical dosing, the microbial community of the biofilm in four reactors were analyzed, as shown in **Figure 6-15 (a)**. The major genus in four reactors were the same, which was dominated by SRB consisting of *Desulfobulbus*, *Desulfobacter*, *Desulfomicrobium* and *Desulfovibrio*. The relative abundance of total SRB was 21.4%, 24.5%, 20.4% and 11% in reactor R1, R2, R3 and R4, respectively. In addition to SRB, the rest of the major genus were responsible for organic matter transformation. *vadinBC27_wastewater-sludge_group* was reported to be in syntrophic association with hydrogenotrophic methanogens for degrading amino acids (Li et al. 2015). *Trichococcus* has been found to utilize a variety of sugars, sugar alcohols, and polysaccharides. Fermentation of glucose by strains of this genus results in the production of lactate, formate, acetate, ethanol, and CO_2 (Rainey 2015). *Comamonas* has been shown to be capable of catabolizing a wide range of organic substrates, including amino acids, carboxylic acids, steroids and aromatic compounds (Wu et al. 2018). *Zoogloea* has long been considered the typical activated sludge bacterium responsible for the formation of biofilm (Rosselló-Mora et al. 1995). The genera *Paludibacter* is strictly anaerobic and chemoorganotrophic, which can utilize melibiose, glycogen and soluble starch as growth substrates (Gronow et al. 2011).

The microbial community structure shifted after the addition of ferric and nitrate, as seen in **Figure 6-15 (b)**. After ferric treatment (R1(Fe)), the abundance of three SRB genera (*Desulfobulbus*, *Desulfomicrobium* and *Desulfovibrio*) decreased (less than 1%), while the relative abundance of *Desulfobacter* increased from 0.7% to 21.8 %. However, the relative abundances of total SRB were close (21.4% and 21.8%). The addition of ferric did not change the total population of SRB. After nitrate addition, all four SRB genera (*Desulfobulbus*, *Desulfobacter*, *Desulfomicrobium* and *Desulfovibrio*) dropped and only *Desulfobacter* remained as major genera. When nitrate was added at the beginning of pump cycle (R2 (NO_3)1), the relative abundance of total SRB dropped from 24.5% to 3.4%. The addition of nitrate induced the development of *Thauera* which was the most dominant genera in the biofilm with a relative abundance of 41.2 %. *Thauera* is heterotrophic nitrate reducing bacteria (hNRB), which can use organic carbon as electron donors to reduce nitrate or nitrite to N_2 (Etchebehere and Tiedje 2005). *Thauera*, which competed with SRB for degradable organic electron donors and thus potentially prevented SRB metabolism in presence of nitrate. Autotrophic denitrification prevailed in the reactor when nitrate was dosed at the end of pump cycle (R2 (NO_3)2) and the relative abundance of total SRB dropped from 24.5% to 1.8%. The co-existence of sulfide and nitrate stimulates autotrophic denitrification. *Sulfurovum* was dominating the microbial community and the relative abundance was 16.2%. *Sulfurovum*, which is nitrate reducing sulfide-oxidizing bacteria (NR-SOB), was the main contributors to sulfide oxidation. *Sulfurovum* genera has been demonstrated to grow chemolithoautotrophically with S^{2-}, in addition to S^0 and thiosulfate as electron donors and nitrate as electron acceptors (Rodriguez-Mora et al. 2016). *Sulfurovum* was responsible for sulfide removal when sulfide and nitrate were present simultaneously. The rest of the major genera identified in the biofilm after nitrate and ferric addition, *Macellibacteroides*, *Arcobacter*, *Dechloromonas* and *Paludibacter*, were the key functional genera of the anaerobic environment (Achenbach et al. 2001, Gronow et al. 2011, Jabari et al. 2012).


(a)
Relative abundance (%)
0
20
40
60
80
100
R1
R2
R3
R4
others
Aeromonas
Lactococcus
unidentified_WCHB1-41
Taibaiella
Blvii28_wastewater-sludge_group
Acetobacter
Christensenellaceae_R-7_group
Lactobacillus
Paludibacter
Zoogloea
Comamonas
Trichococcus
unidentified_Spirochaetaceae
vadinBC27_wastewater-sludge_group
Desulfovibrio
Desulfomicrobium
Desulfobacter
Desulfobulbus

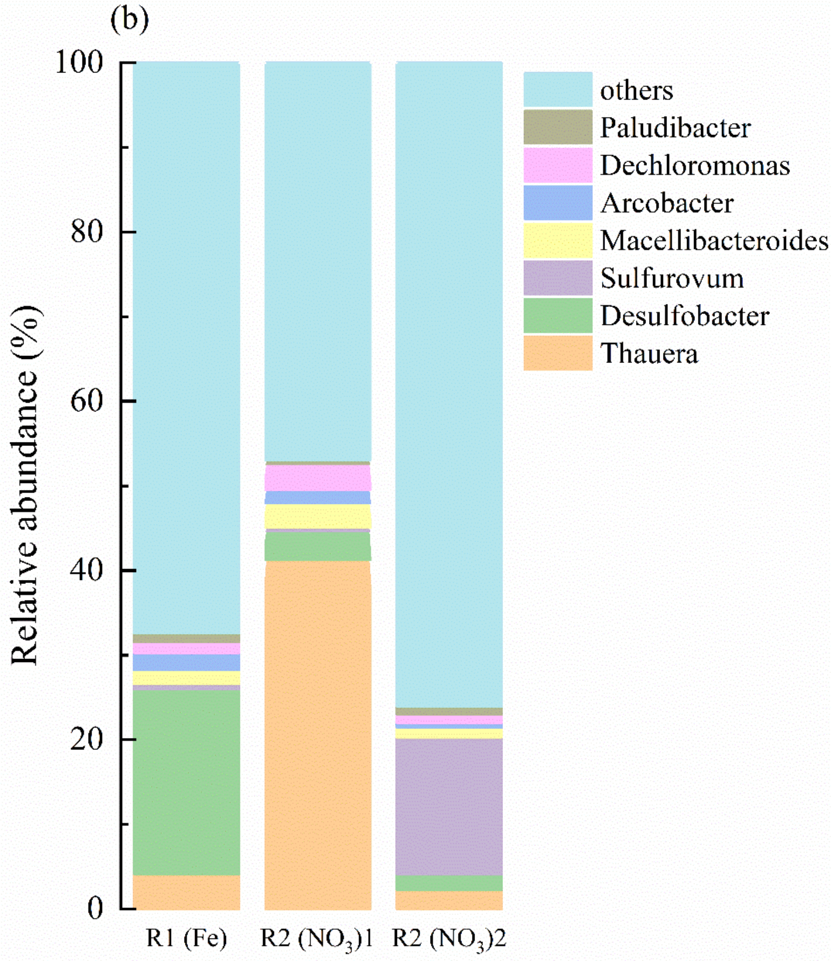


Figure 6-15 Relative abundance of each major genus in the biofilm samples: (a) before chemical dosing, (b) after chemical dosing. Genera that occurred at <1% abundance in all samples are defined as ‘‘Others”

6.4 Conclusions

The effects of nitrate and ferric on sulfide control in force mains were both investigated in the lab using 4 bioreactors. The following conclusions are drawn:

(1) Lab study demonstrates that half order kinetics regarding DCOD was capable of predicting the sulfide formation in the bioreactors and the modelled empirical rate constant was in the range of 0.005~0.0011. The numbers were useful to predict the sulfide generation rates in force mains.

(2) Lab study shows ferric could effectively control the sulfide generation in the force main. However, it did not inhibit the sulfide generation rate in the bioreactor. Hydrogen sulfide was controlled with the ferric salt addition only via chemical oxidation and precipitation and total SRB population did not change.

(3) Nitrate addition at the beginning of pump cycle induced the development of *Thauera* as hNRB. The hNRB outcompeted SRB for organic matter as electron donor as the dominant bacteria resulted the suppression of sulfide generation. The amount of nitrate required for sulfide control depended on heterotrophic denitrification rate and the heterotrophic denitrification in the bioreactor could be well described by half order kinetics with regard to DCOD concentration. A modified empirical equation was provided which was able to predict the denitrification rate in the biofilm. The modelled empirical rate constant b was around 0.05. The developed equation could be a very useful tool for design of nitrate dosing strategy. Nitrate did not have the long-lasting inhibitory/toxic effect on sulfate reduction.

(4) The cost-effective dosing strategy for nitrate dosing in the force main was to add the nitrate at the end of pump cycle instead of at the beginning of pump cycle which could save up to about 75% nitrate dosage. The co-existence of sulfide and nitrate stimulates autotrophic denitrification. *Sulfurovum*, which is nitrate-reducing sulfide-oxidizing bacteria (NR-SOB), was the main contributor to sulfide oxidation resulting in sulfide removal in the reactor.

Chapter 7 Sulfide Generation in Force Mains and its Control Using Nitrate Dosing

7.1 Introduction

The presence of anaerobic conditions in sewer systems especially in force mains results in significant production of sulfide by sulfate reducing bacteria (SRB) present in the fixed biofilm phases (Nielsen et al. 1998). The design of deep trunks may bring new challenges to sewer management. Compared to the normal sewer systems in other cities, the deep trunk systems in the City of Edmonton, Alberta, Canada have led to a large number of pump stations (Guo et al. 2018). The long hydraulic retention time of sewage in the long force main facilitates the development of anaerobic conditions, which result in the production of hydrogen sulfide (H_2S) by sulfate-reducing bacteria (SRB). High hydrogen sulfide in force mains can lead to numerous problems, such as sewer corrosion, odor nuisance and health hazards to sewer workers (Ganigue et al. 2011, Hvitved-Jacobsen et al. 2013).

The variations in sewage quality, flowrate, temperature and hydraulic retention time (HRT), or even a storm event, can significantly affect sulfide production and H_2S emission in force mains (Nielsen and Hvitved-Jacobsen 1988, Shypanski et al. 2018). Kitagawa et al. (1998) revealed that the biofilm sulfide generation rate in a 1.8 km, 100 mm diameter force main ranged from 0.115 to 0.22 g/m^2h (based on the pipe wall surface area) with wastewater temperature from 15 ~27 °C and BOD from 100 to 350 mg/L. The biofilm generation rate in two force mains with 4.6 km and 1.63 km length was found to vary between 0.005 and 0.05 g/m^2h between 4 and 12 °C with volatile fatty acid (VFA) from 5 to 50 mg/L (Hvitved-Jacobsen et al. 1995). Tanaka et al. (2000) also investigated the sulfide generation rate in the biofilm of the force mains in Oga city in Japan and found out it was in the range of 0.067~0.166 g/m^2h at 20 °C with DCOD from 100 to 300 mg//L. Pikaar et al. (2019) found that the gaseous hydrogen sulfide concentrations at the discharge manhole of a 1 km force main (diameter: 150 mm) with HRT ranging between 3 and 7 h was 173 ppm. These sulfide generation rates were site-specific and varied with different biodegradable organic matter concentration and temperature. The half order biofilm kinetics with regard to DCOD have been proposed to predict the sulfide generation rates in the lab. It is essential to validate its feasibility in the actual force mains by conducing the field study.

Addition of a favorable electron acceptor like nitrate has been widely used to control odors and sulfide production in many environments, including sewage systems and oil reservoirs (Jiang et al. 2009, Jiang et al. 2013, Xue and Voordouw 2015). Different nitrate salts (e.g. sodium and calcium nitrate) and different dosing concentrations have been tried. Nitrate additions of 40 mg-N/L to wastewater successfully decreased sulfide concentrations from 10~20 mg/L to below 2~3 mg/L in a 5.0 km long force main (Saracevic et al. 2007). Nitrate additions of 10 mg-N/L to wastewater successfully decreased sulfide concentrations from 4.2 mg/ L to 0.2 mg/ L in a 2.4 km long force main (Bentzen et al. 1995). Rodríguez-Gómez et al. (2005) found that 5 mg-N/L of nitrate in sewage was capable of reducing sulfide production effectively in a 61 km long gravity sewer transporting reclaimed water. The nitrate dosage for control the sulfide in the force main in the field relies on the wastewater quality, temperature and HRT. Addition of nitrate for odor control favor its use among other chemicals due to its no need for special occupational training in comparison to other chemicals like hydrogen peroxide or potassium dichromate. Despite this fact, few studies until now have presented the effectiveness of nitrate use for odor control under field conditions, especially in terms of nitrate dosing optimization. Further studies are required to confirm the practical and financial feasibility of nitrate dosing strategy in the field.

In Edmonton, nearly 90 pump stations were constructed in the sewer systems and the odor and corrosion problems associated with hydrogen sulfide generated in these force mains have been a long-recognized concern (Yang et al. 2019). In this study, the four pump stations which have different length of force mains were selected as representative examples, and the field experimental investigations were performed to evaluate the sulfide formation rates in the force mains of these pump stations. Nitrate that is relatively non-toxic and easy to operate was added at one pump station with a high sulfide concentration to control the sulfide generation in this force main. The effectiveness of nitrate dosing was investigated.

7.2 Methodology

7.2.1 Field monitoring at four pump stations

The total sulfide generation increases with the HRT in a force main. Four pump stations (Blackburne, Kaskitayo, Twin Brooks and Big Lake Pump Station) in Edmonton, Alberta, Canada with different length of force mains and different flow rates which resulted in different HRT, were chosen to be monitored in the field. The sulfide generation rates in the four force mains were evaluated. These four pump stations run intermittently by cycles, as shown in **Figure 7-1**. The four pump stations receive sanitary sewage from residential areas and the flow rates are small from 1:00 AM ~ 6:00 AM when the retention times are long.

The Blackburne pump station is located at 111 Street and Blackburn Drive. The pump starts to work when the water level in the pump well is higher than 2.4 m and stops to work when the water level is lower than 1.34 m (**Figure 7-1 (a)**). The rectangular pump station (PS) wet well is 3.0 m×1.17 m. Each pumping event delivers about 3.72 m^3 of wastewater into the force main pipe. The force main pipe leading from Blackburne PS to a discharge manhole is 671 m with a diameter of 250 mm. The retention time in the force main is around 5~9 h (**Table 7-1**).

The Kaskitayo pump station is located at 125 Street and 29a Avenue. The pump starts to work when the water level in the pump well is higher than 1.98 m and stops to work when the water level is lower than 0.91 m (**Figure 7-1 (b)**). The circular PS wet well is 1.93 m in diameter. Each pumping event delivers about 3.13 m^3 of wastewater into the force main pipe. The force main pipe leading from Kaskitayo PS to a discharge manhole is 989 m with a diameter of 200 mm. The retention time in the force main is around 2~5 h (**Table 7-1**).

The Twin Brooks station is located at 113 Street and 10a Avenue. The pump starts to work when the water level in the pump well is higher than 2.29 m and stops to work when the water level is lower than 1.18 m (**Figure 7-1 (c)**). The rectangular PS wet well is 2.6 m×1.59 m. Each pumping event delivers about 4.59 m^3 of wastewater into the force main pipe. The force main pipe leading from Kaskitayo PS to a discharge manhole is 989 m with a diameter of 200 mm, among which only 22 m serves as storage volume (with negative pipe slope). The retention time in the force main is around 0.08~0.5 h (**Table 7-1**).

The Big Lake pump station is located at 205 Street and 131 Avenue, as shown in **Figure 7-3**. The pump starts to work when the water level in the pump well is higher than 2.65 m and stops working when the water level is lower than 1.2 m (**Figure 7-1 (d)**). The PS wet well was one section of the circular structure with 9 m diameter and 2.9 m height. Each pumping event delivers about 26.5 m^3 of wastewater into the force main pipe. The force main pipe leading from the Big Lake PS to a discharge manhole (MH461065) is 4764 m with a diameter of 350 mm, which is located at 199 Street and 111 Avenue. The storage volume for force main is about 458 m^3 and corresponding biofilm area is 5238 m^2. The biofilm area/volume (A/V) ratio was 11.4 m^2/m^3. The retention time in the force main is around 16~20 h (**Table 7-1**).

The sulfide generation rates in the force mains of three pump station (Blackburne, Kaskitayo and Twin Brooks) were firstly investigated by conducting the field work on Oct. 30th, 2018. The wastewater samples were taken from the PS wet wells at Twin Brooks (9:33 AM), Blackburne (10:37 AM), and Kaskitayo (12:05 PM) pump stations. The water samples were taken from discharge manholes at 11:32 AM, 1:05 PM, 10:08 AM of these three pump stations when the retention time was about 5.7 h (Blackburne PS), 2.3 h (Kaskitayo PS) and 0.3 h (Twin Brooks PS) in the force main. Additionally, the 5-hour and 12-hour wastewater sampling programs were conducted at the Big Lake pump station on Sept. 08th and Sept. 11th, 2020 at both wet well and discharge manhole and nitrate was dosed at the Big Lake pump station as detailed later.

The pH, temperature, dissolved oxygen (DO) and oxidation and reduction potential (ORP) of the water samples were measured on site. The samples for total and dissolved sulfide (TS and DS) analysis were preserved on site. All the field wastewater samples were stored in an ice box during transportation to the laboratory and were kept refrigerated till analysis was done. The Odalogs (App-Tek, Queensland, Australia) were installed at pump wells and force main discharge manholes of these four pump stations to continuously monitor the H_2S concentration in the air phase. The sulfide generation rates (r_a) in the force mains of these four pump stations were calculated. Since the wastewater temperature were different at the four pump stations, the calculated sulfide generation rates were converted to that under 20 °C ($r_{a(20)}$) for comparison reason.

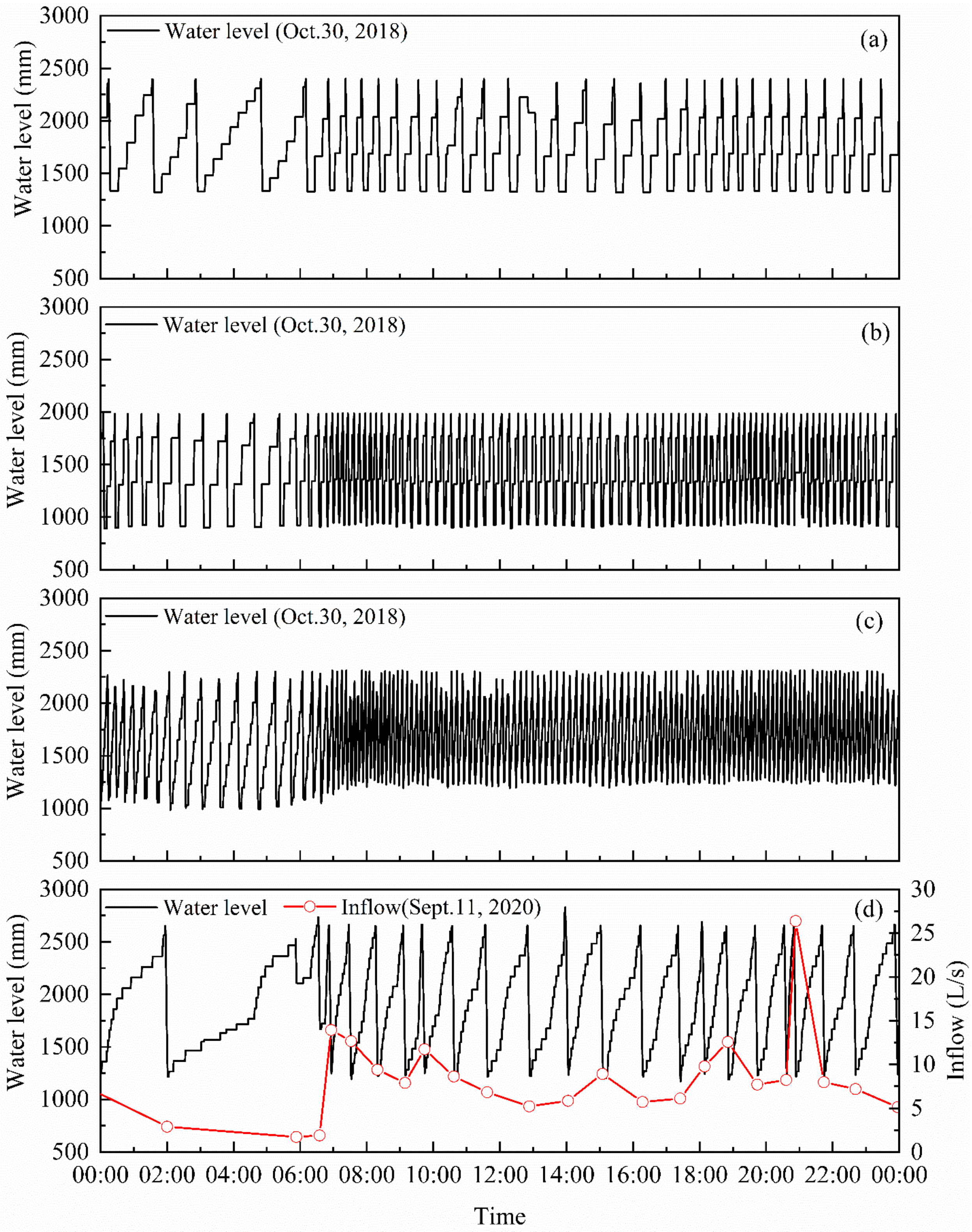


Figure 7-1 Water level at the wet wells of the pump stations: (a) Black burne PS, (b) Kaskitayo PS, (c) Twin Brook PS and (d) Big Lake PS

Table 7-1 Force main characteristics of the four pump stations

PS	Total length (m)	Storage length (m)	Diameter (mm)	Sewer volume (m^3)	Discharge volume (m^3/event)	Pumping events	Retention time (h)
Blackburne	671	671	250	33	3.72	9	5~9
Kaskitayo	989	989	200	31	3.13	10	2~5
Twin Brook	1327	22	350	2	4.59	1	0.08~0.5
Big Lake	4764	4764	350	458	20	19	16~20

7.2.2 Field nitrate dosing at the Big Lake pump station

A field dosing at the Big Lake pump station was conducted to investigate nitrate dosing as a means for sulfide control in a real force main. Five sampling campaign were carried out, as shown in **Figure 7-2** from Sept. 11th to Oct. 16th, 2020. First two sampling programs were carried out to identify baseline value before nitrate dosing. Then the next two sampling programs were performed under low and high dosage to investigate the effect of different nitrate dosage on sulfide control in the force main. The sampling program under low nitrate dosage was carried out two weeks after starting nitrate dosing when it was stabilized. The sampling under high nitrate dosage was conducted at the end of the one-month dosing phase. Finally, the recovery of sulfide generation in the force main after applying nitrate dosing for one month was investigated by conducting the last sampling program one week after ceasing nitrate dosing.

Prior to the nitrate dosing, two measurement campaigns were conducted to monitor the sulfide generation in the force main on Sept. 08th for 5 hours (10:00 AM to 3:00 PM) and Sept. 11th, 2020 for 24 hours (4:00 AM to 4:00 AM). The first sampling campaign was to test run to make sure the sampling process worked well before the second 24-hour intense sampling. Then 48~54% calcium nitrate ($Ca(NO_3)_2$) (by weight) solution which resulted in 125±3 g N/L calcium nitrate was continuously injected into the wet well by a pump at a constant flow rate above the inlet of the wet well inflow starting from Sept. 12th, 2020. The inflow varies from 2 L/s to 28 L/s as shown in **Figure 7-1 (d)**. The concentration of the calcium nitrate in sewage depended on the inflow coming into the wet well. Two monitoring campaigns were conducted during dosing phase on Sept. 25th and Oct. 09th, 2020 for 24 hours (4:00 AM to 4:00 AM) at 7 L/h and 15 L/h dosing rate, respectively. The dosage was halted on Oct.10th, 2020 and the last campaign was performed on Oct.16th, 2020 during recovery phase. Each campaign involved sampling wastewater by pumping event at both the wet well and discharge manhole. Odalogs were installed at both wet well and discharge manhole to continuously detect the H_2S gas concentration for the entire field sampling period. Before reaching to the 15 L/h, the dosing rate was gradually increased three time (5 L/h, 7 L/h, 12 L/h) to avoid overdosing.

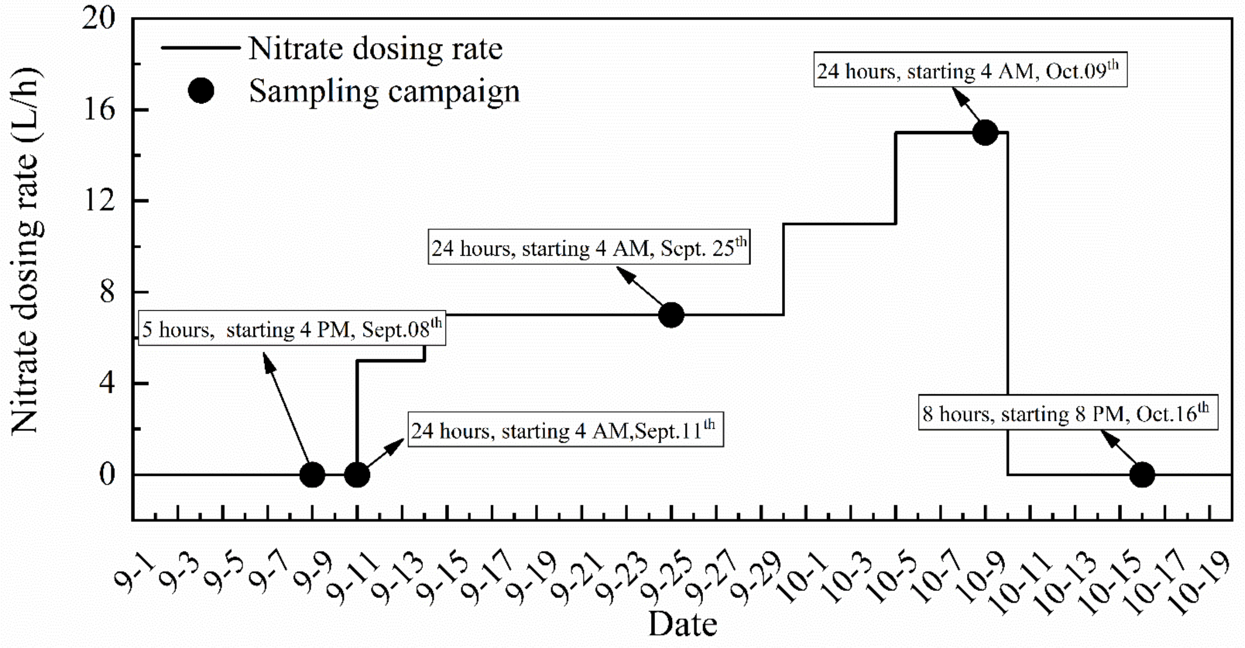


Figure 7-2 Overall field work campaign at the Big Lake pump station with nitrate dosing. Black circles indicate the days and durations of the field liquid sampling

The water samples were taken from the wet well from a hose connected from the prime valve on pump 1 to the top level of the dry well (**Figure 7-3**). A peristaltic pump unit was set up at the discharge manhole on the study test day for sampling at discharge manhole (**Figure 7-3**). The water samples were grabbed during the pumping event at a specified time duration. When a pump cycle started, the valve was turned on to release wastewater at wet well and the peristaltic pump was turned on to sample wastewater samples at discharge manhole at the same time.

Figure 7-3 Location of Big Lake pump station and its discharge manhole

7.3 Results and Discussion

7.3.1 Evaluation of sulfide formation in force mains

1) Sulfide formation at Blackburne, Kaskitayo and Twin Brooks force mains

Field monitoring was implemented at force mains of three pump station to identify the sulfide generation rate. As shown in **Table 7-2**, the temperature of the wastewater samples was 14~16℃ and pH was 8~9. The DO at wet wells of three pump stations and discharge manhole of Twin Brooks PS was higher than 2 mg/L. The DO at discharge manholes of the Blackburne, Kaskiyato and Twin Brooks PS was low at 0.94 mg/L, 1.29 mg/L and 2.43 mg/L, respectively. The TS and DS at all these wet wells was less than 1 mg/L and 0.5 mg/L, which was low. With regard to the discharge manholes, the highest TS and DS were detected at Blackburne PS with 8.64 mg/L and 6.53 mg/L, respectively. At the Kaskitayo PS, the TS and DS were 6.76 mg/L and 4.58 mg/L, respectively. The TS and DS at Twin Brooks PS were 1.35 mg/L and 0.69 mg/L, respectively. The sulfate at discharge manhole was lower than that in wet wells which indicates the sulfate was utilized to generate sulfide in force mains.

The H_2S gas concentration was consistent with sulfide concentration in the water samples (**Figure 7-4**). The H_2S gas was only detected in these pump wells at specific short period of time over a day with a low level, 0~4 ppm (Blackburne PS), 0~1 ppm (Kaskitayo PS) and 0~3 ppm (Twin Brooks PS). In terms of discharge manholes, high H_2S was detected at Blackburne and Kaskitayo PS but no H_2S was detected at Twin Brook PS. The average H_2S gas at Blackburne PS was around 70 ppm with peak value around 200 ppm. The average H_2S gas at Blackburne PS was around 10 ppm with peak value around 20 ppm.

The high sulfide and H_2S was measured in the discharge manholes, not in wet wells. This suggests that the sulfide in the discharge manholes was mainly generated in the force mains. According to field sampling results (**Table 7-3**), the sulfide generation rate was calculated by the difference of sulfide in the wet well and discharge manhole divided by HRT and pip wall surface area. The calculated sulfide generation rate per biofilm area in the force mains was in the range of 0.08~0.13 g S/ m^2 h under 15 °C. The generation rate was 0.09~0.15 g S/ m^2 h under 20 °C after temperature correction. The generation rate was 0.09~0.15 g S/ m^2 h under 20 °C after temperature correction. The HRT of sewage in the three force mains are 5~9 h, 2~5 h and 0.08~0.5 h which could lead to 5.6~10 mg/L, 2.8~7 mg/L and 0.06~0.4 mg/L DS in the sewage at discharge manholes of the force main, respectively. The sulfide in the water and H_2S in the air phase formed in the force main under the long HRT were pushed to the discharge manholes when the pumps started working, which resulted in both high sulfide and H_2S gas at Blackburn and Kaskitayo PS. The HRT in the force main of Twin Brooks PS was too short to generate high sulfide. Observed sulfide production rates are of the same order of magnitude as measured in other investigations (0.067~0.166 g/m^2h) (Hvitved-Jacobsen et al. 1995, Kitagawa et al. 1998, Tanaka et al. 2000).

Table 7-2 Water sample measurements in the three pump stations

Pump station	Location	Time	pH	DO (mg/L)	T (°C)	TS (mg/L)	DS (mg/L)	Sulfate (mg S/L)
Blackburn PS	Wet well	10:37 AM	8.86	2.41	14.1	0.94	0.34	48
	Discharge manhole	11:32 AM	8.18	0.91	15.0	8.64	6.53	44
Kaskitayo PS	Wet well	12:05 PM	8.52	3.16	15.8	0.80	0.41	47
	Discharge manhole	1:05 PM	8.11	1.29	14.7	6.76	4.58	44
Twin Brooks PS	Wet well	9:33 AM	8.89	5.17	14.2	0.93	0.38	47
	Dishcage manhole	10:08 AM	8.98	2.43	14.0	1.35	0.69	43

Table 7-3 Sulfide generation rate (r_a) in the force main at three pump stations

Force main	HRT (h)	TS_{in} (mg/L)	TS_{out} (mg/L)	V (m^3)	Biofilm area (m^2)	T (°C)	r_a (gS/h m^2)	$r_{a(20)}$ (gS/h m^2)
Blackburn PS	5.7	0.94	8.64	33	527	15.0	0.08	0.09
Kaskitayo PS	2.3	0.80	6.76	31	621	14.7	0.13	0.15
Twin Brooks PS	0.3	0.93	1.35	2	24	14.0	0.12	0.14

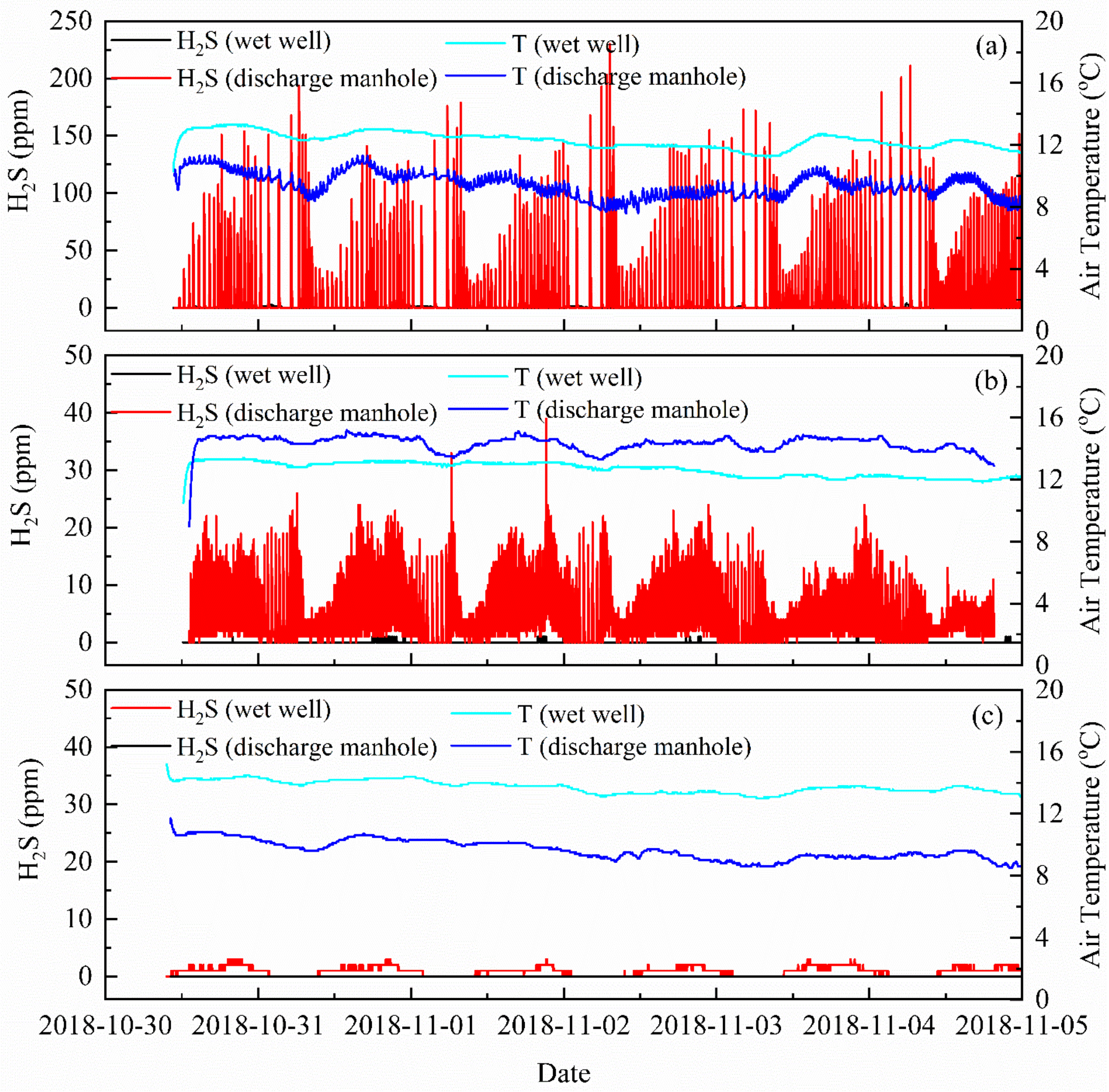


Figure 7-4 H$_2$S gas in the air phase of three pump stations ((a) Blackburn PS (b) Kaskatayo PS (c) Twin Brooks PS)

2) Sulfide formation at the Big Lake force main

Prior to nitrate dosing, 5-hour and 24-hour sampling campaigns were performed at Big Lake pump station by pumping event. The wastewater samples from both the wet well and discharge manhole at different time were analyzed to evaluate the general water characteristics in Big Lake, as is shown in **Table 7-4**. The wastewater quality reflected the general domestic wastewater quality. As shown in **Figure 7-5**, there were around 2.3 mg/L DS, 2.8 mg/L TS and 40 mg/L sulfate detected in the wet well, while 19.4 mg/L DS, 20 mg/L TS and 20 mg/L sulfate were found in discharge manhole. The around 19.5 mg/L DS could result in about 300~500 ppm H_2S gas at discharge manhole (**Figure 7-6**).

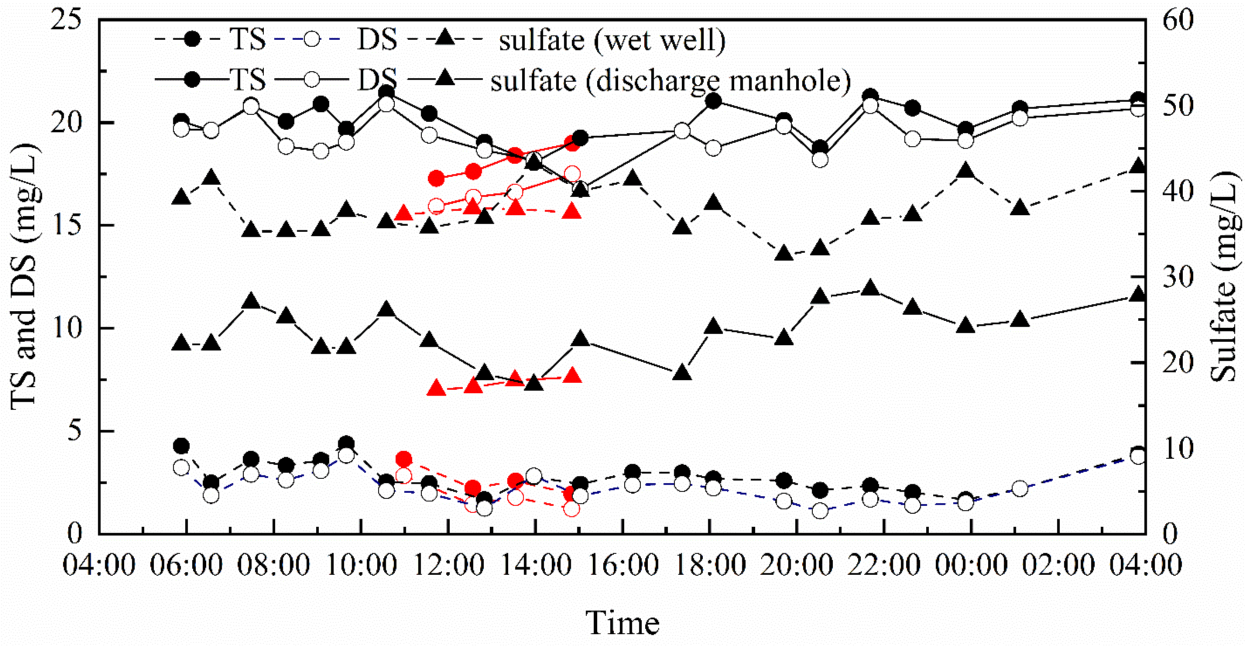


Figure 7-5 The 5-hour (red ones, Sept. 08th, 2020) and 24-hour (black ones, Sept. 11th, 2020) variation of sulfide (TS and DS) and sulfate and at the wet well and the discharge manhole.

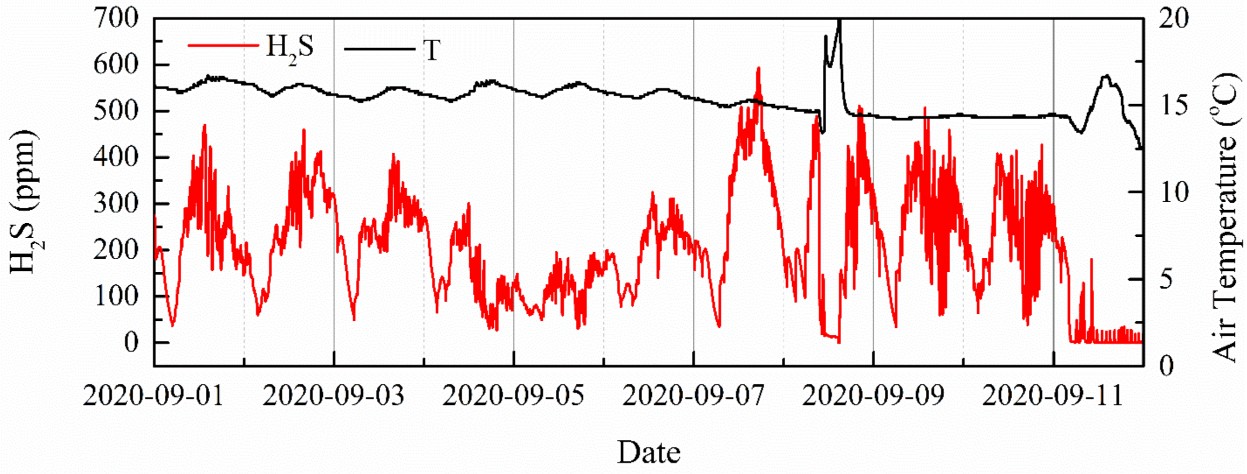


Figure 7-6 H_2S gas in the air phase at the discharge manhole

Table 7-4 Mean wastewater characteristics in the Big Lake pump station

NH_4-N (mg/L)	NO_3-N (mg/L)	TN (mg/L)	TP (mg/L)	TCOD (mg/L)	DCOD (mg/L)	pH	T (°C)
49	0.5	78	24	900	300	7.5	19

Approximately 17 mg/L sulfide was generated in the force main. The sulfide production rate is believed to be limited by DCOD. The TCOD in the wet well (900 mg/L) was higher than that in discharge manhole (750 mg/L), and DCOD was close at both locations, around 300 mg/L (**Figure 7-7 (a)**). The pH was 7.4~8.5 in the wet well and 7.0~8.0 in the discharge manhole. In terms of ORP and DO (**Figure 7-7 (b)**), ORP was in the range of -20 ~ -40 mV (standard hydrogen electrode (SHE)) in wet well and -130 ~ -160 mV (SHE) in the discharge manhole in the first trial. The value was -110 ~ -170 mV (SHE) in the wet well and -200 ~ -230 mV (SHE) in the discharge manhole in the second field trial. DO was 2~5 mg/L in the wet well and 2~4 mg/L in the discharge manhole. The DO measured in discharge manhole could reflect DO in force main, since DO was introduced into sewage when it was quickly discharged into discharge manhole in the event of pumping. The temperature and pH in wet well were also higher than that in discharge manhole (**Figure 7-7 (c)**). The temperature was 18~21°C in the wet well and 16~19 °C in the discharge manhole. It varied during the whole day.

The sulfide was generated in the force main during the 14.5~19.5 h hydraulic retention time (HRT). The H_2S gas was released into air phase from the sewage containing sulfide as the sewage was discharged at the discharge manhole. DCOD was consumed as substrate for biomass growth and as electron donor for the sulfate-reducing bacteria (SRB) in the biofilm of force main, the TCOD dropped. The anaerobic hydrolysis and fermentation processes could transform the particulate COD into dissolved COD, so the DCOD could maintain at around 300 mg/L in the force main. The temperature of the sewage is affected by air temperature. Thus, it was higher at noon and lower at night which had the same trend as daily air temperature. The pH decreased in the force main probably due to the anaerobic activity. As for the ORP, sulfide formation occurs when the ORP is between -50 to -250 mV (standard hydrogen electrode (SHE)) (Leung 1998).

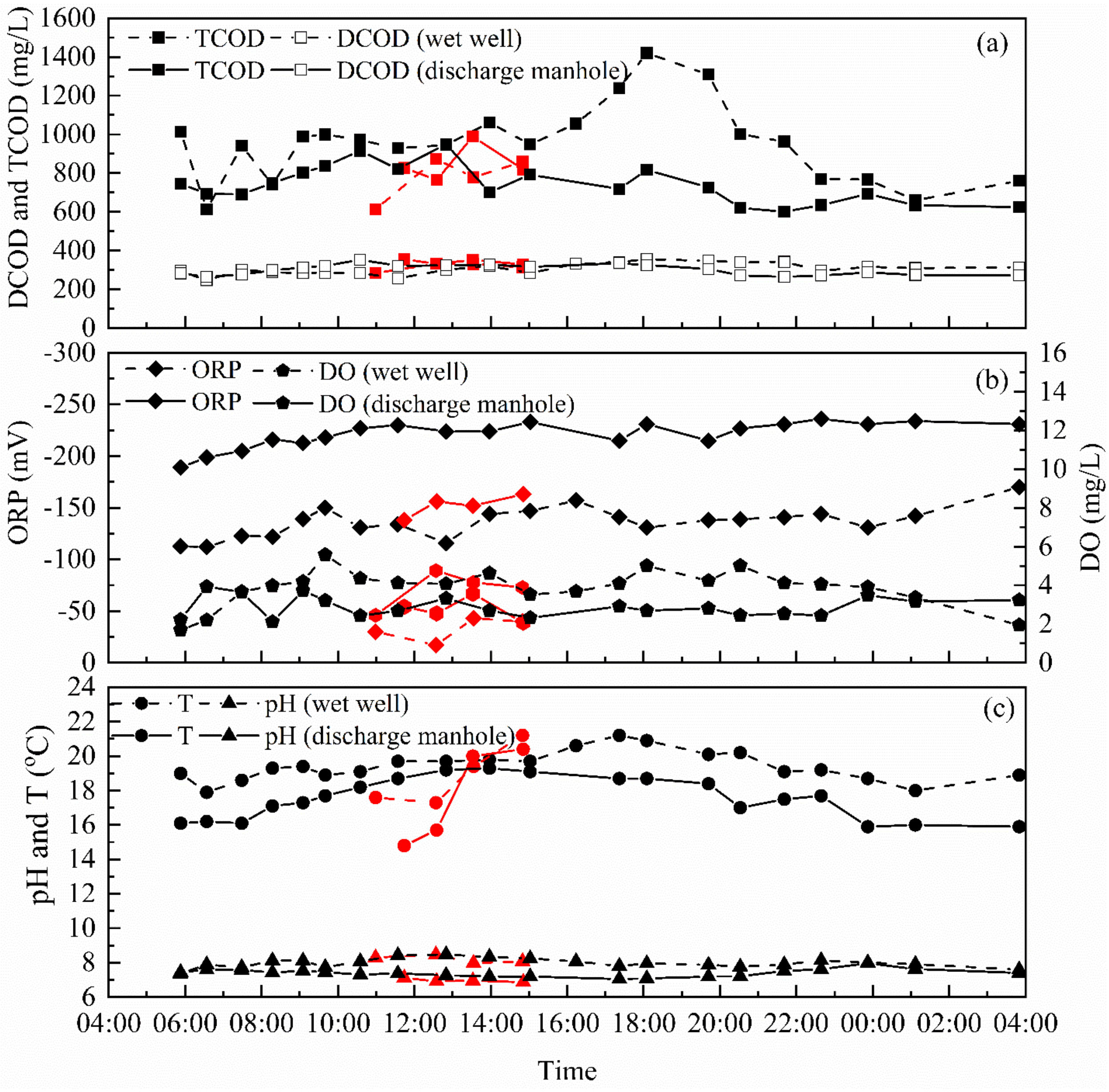


Figure 7-7 The 5-hour (red ones, Sept. 08th, 2020) and 24-hour (black ones, Sept. 11th, 2020) variation of (a) TCOD and DCOD, (b) ORP and DO, (c) T and pH at the wet well and the discharge manhole

As seen in **Figure 7-8**, the HRT varied from 14.5 h and 19.5 h in the force main and the resulting sulfide generation rate was in the range of 0.07~0.11 g/m^{2}h. The sulfide generation rate was converted to the value at 20 °C. The generation rate was 0.08~0.12 g/m^{2}h. The 1/2-order biofilm kinetics of DCOD has been proposed to model the sulfide generation rate in force main. Applying this with field trial date (**Figure 7-9**) and the modelled rate constant for field trial was around 0.006, which was within the range of lab studies (0.005~0.010). Therefore, the 1/2-order biofilm kinetics can be used to predict the sulfide generation rate in force main and the corresponding rate constant is around 0.006.

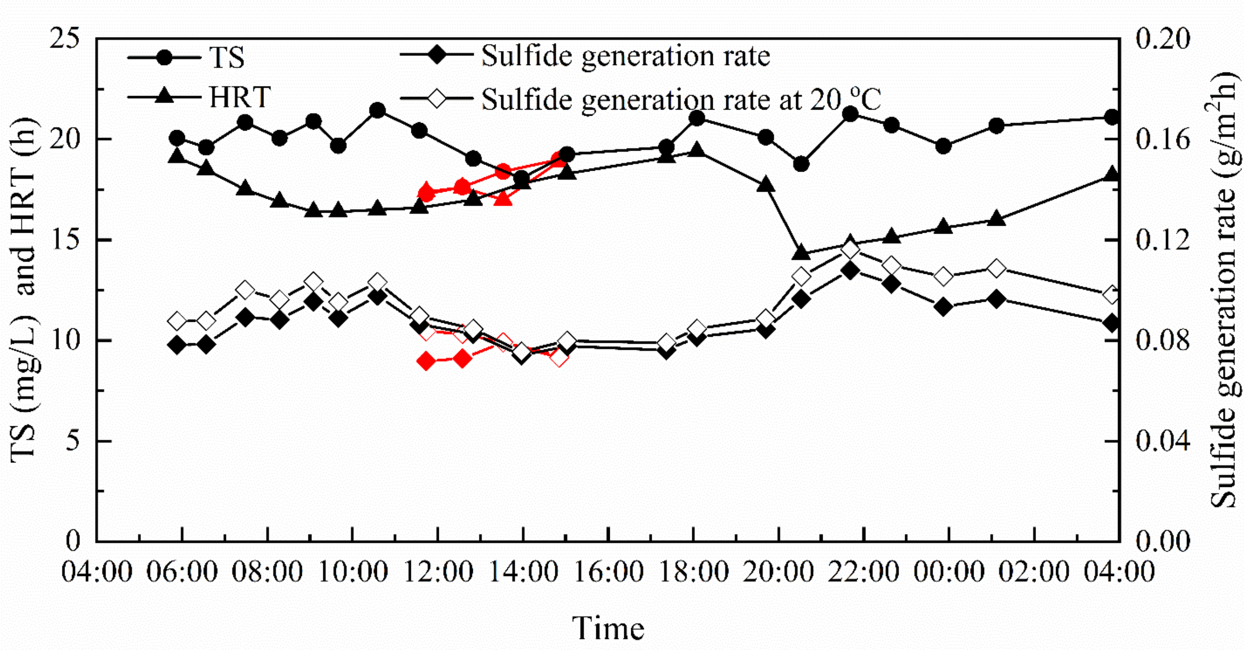


Figure 7-8 Variation of HRT and sulfide generation rate in the force main (red ones: first field trial on Sept. 08th, 2020, black ones: second field trial on Sept. 11th, 2020) of the Big Lake pump station

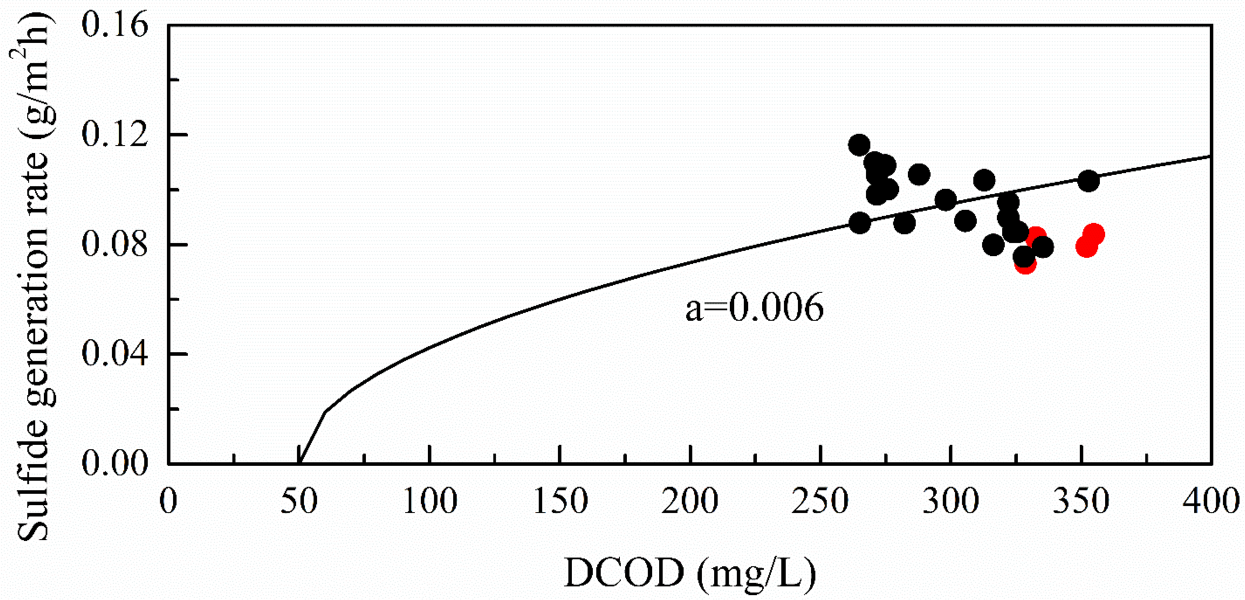


Figure 7-9 Sulfide formation rates versus DCOD in field trial of the Big Lake pump station

7.3.2 Effect of nitrate on sulfide control

Calcium nitrate was injected in the wet well to control sulfide in the force main at a constant flow rate of 5 L/h, 7 L/h, 11 L/h and 15 L/h and 24-hour field sampling campaign was carried out under 7 L/h and 15 L/h. The nitrate in wet well was calculate and measured (**Figure 7-10**). The calculated nitrate value based on dosing rate was higher than measured value. The calculated nitrate was around 20~30 mg/L and 40~80 mg/L during day time under 7 L/h and 15 L/h nitrate dosing rate. However, the measured nitrate was 2~10 mg/L and 20~50 mg/L under 7 L/h and 15 L/h nitrate dosing rate during day time. And the nitrite was detected in the wet well with 2~10 mg/L and 5~10 mg/L under 7 L/h and 15 L/h during day time. These numbers were 2~4 times higher in midnight than that during day time. There were several abnormal points for measured nitrate concentration under 7 L/h nitrate dosing rate. That was because the injected nitrated in the wet well was not mixed well due to lots of FOG (fats, oils and grease) floating on the water surface. The mixing was improved after cleanup of FOG under 15 L/h nitrate dosing rate.

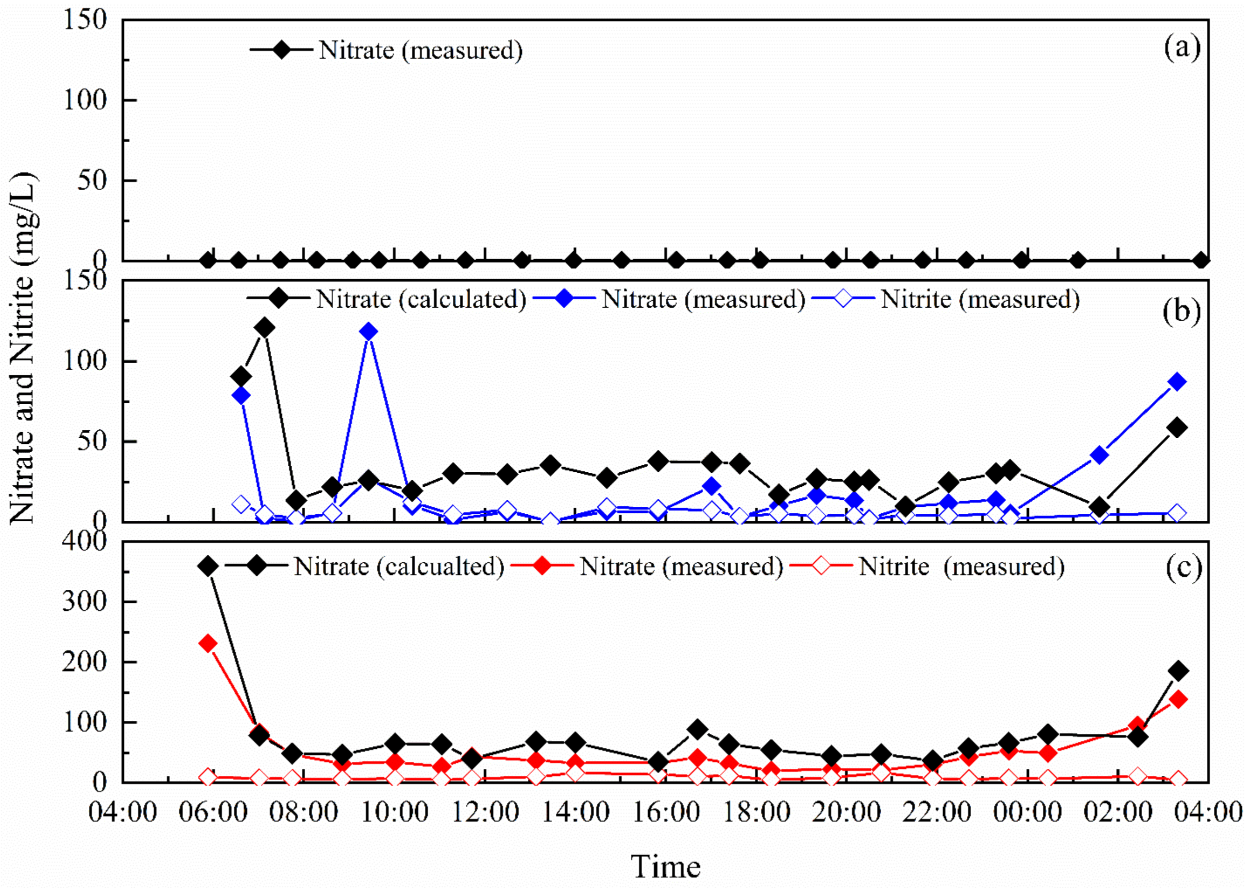


Figure 7-10 Calculated nitrate concentration and measured nitrate and nitrite concentration in the wet well under different nitrate dosing rate: (a) 0 L/h, Sept.08th, 2020, (b) 7L/h, Sept.25th, 2020, (c) 15L/h: Oct. 16th, 2020

The sulfide (TS and DS) did not change after nitrate addition except for midnight when the sulfide decreased below 1.0 mg/L (**Figure 7-11**). As shown in **Figure 7-12**, there were 20~50 ppm H_2S gas detected in the wet well under the DS concentration of 1~4 mg/L. And the H_2S gas at midnight dropped to 0 ppm after nitrate addition. Both the TCOD and DCOD decreased after nitrate addition (**Figure 7-13 (a)**). The TCOD was 730 mg/L and 690 mg/L under 7 L/h and 15 L/h nitrate dosing rate, being compared to 960 mg/L baseline value. The DCOD was around 230 mg/L at both dosing flow rate and the baseline value was 300 mg/L. The ORP and DO did not change for three sampling campaigns (**Figure 7-13 (b)**). The ORP was -170~-100 mV (SHE) and DO was 2~5 mg/L. The DO was lower at midnight than that during day time. The pH did not vary for three sampling campaigns and temperature decreased about 1°C and 2 °C for the last two campaigns (**Figure 7-13 (c)**).

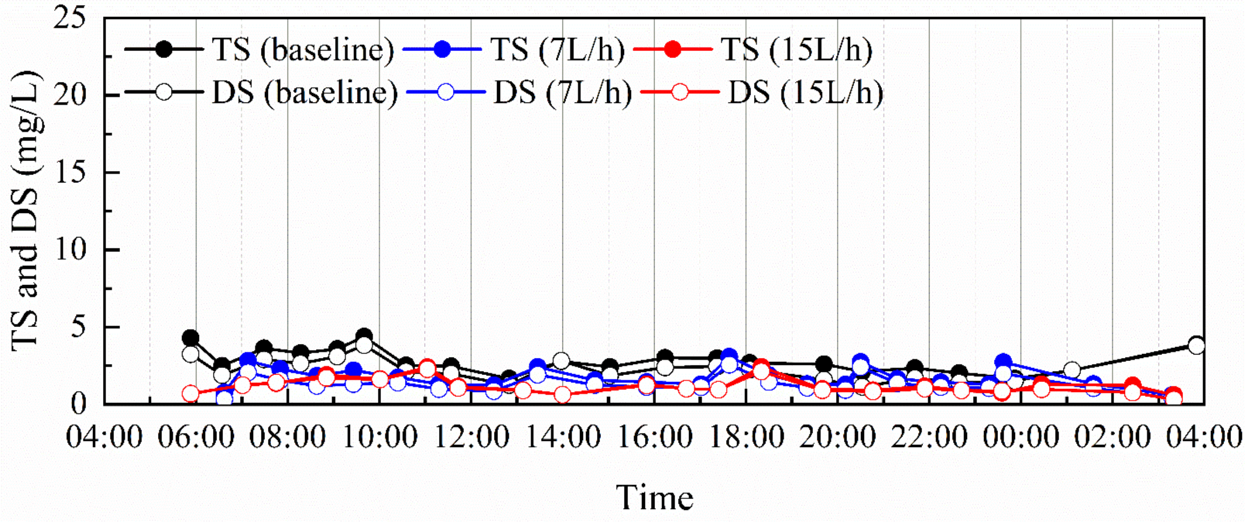


Figure 7-11 The 24-hour variation of sulfide (TS and DS) in the wet well under different nitrate dosing rate (7L/h: Sept. 25th, 2020, 15 L/h: Oct. 09th, 2020)

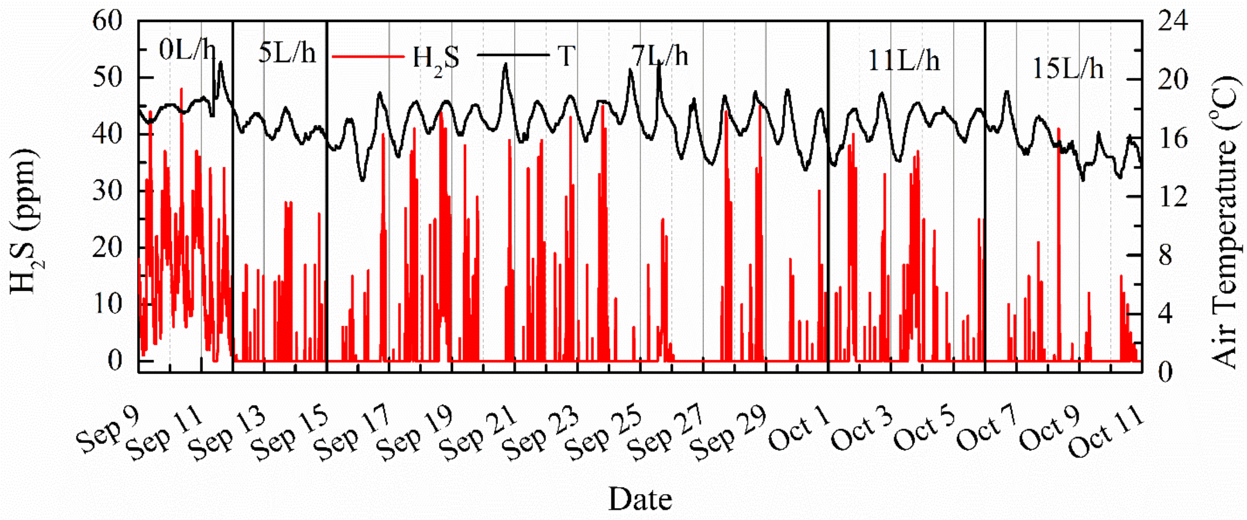


Figure 7-12 H_2S in the wet well under different nitrate dosing rate in the Big Lake pump station

Since the nitrate was added in the wet well at a constant flow rate, the nitrate concentration in the sewage depended on the inflow rate into the wet well. The nitrate concentration was high in midnight when the inflow was small and low during daytime when the inflow rate was high. Nitrite was detected in wet well which revealed that denitrification process occurred in wet well by using nitrate and DCOD. Therefore, TCOD and DCOD dropped. Even though there was DO present, denitrification still happened due to the stratification of the DO in the biofilm on the wet well wall.

The deep layer of the biofilm was under anoxic condition where denitrification could happen. The value of sulfide and H2S gas did not change after nitrate addition except for in the midnight, the sulfide in the wet well actually came from upstream sewers. The HRT in the wet well in day time was short which was now enough to remove the sulfide from upstream after nitrate addition. However, the longer HRT in the wet well in the midnight when the inflow was small allowed the NR-SOB to oxidize the sulfide from upstream in presence of nitrate.

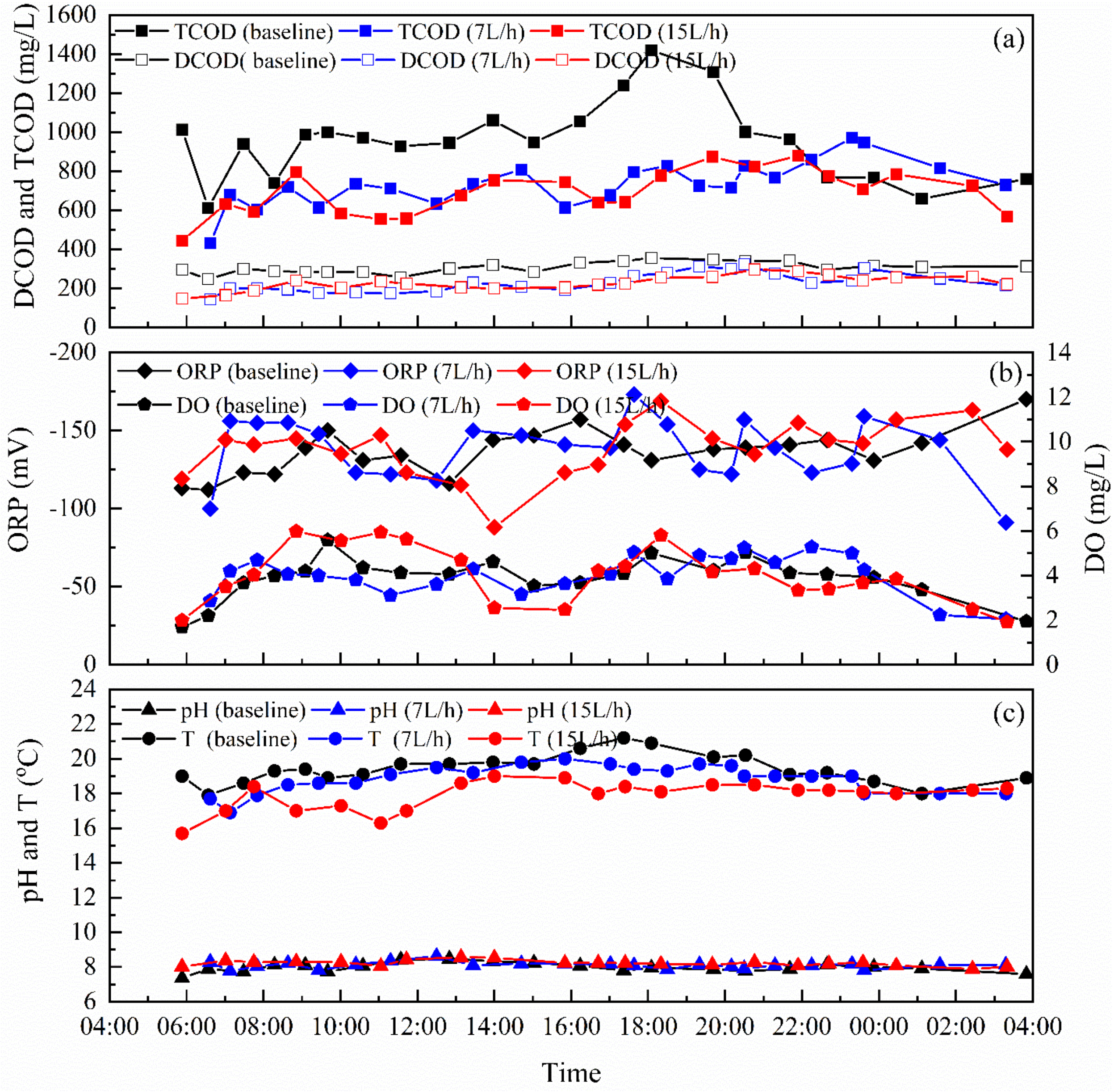
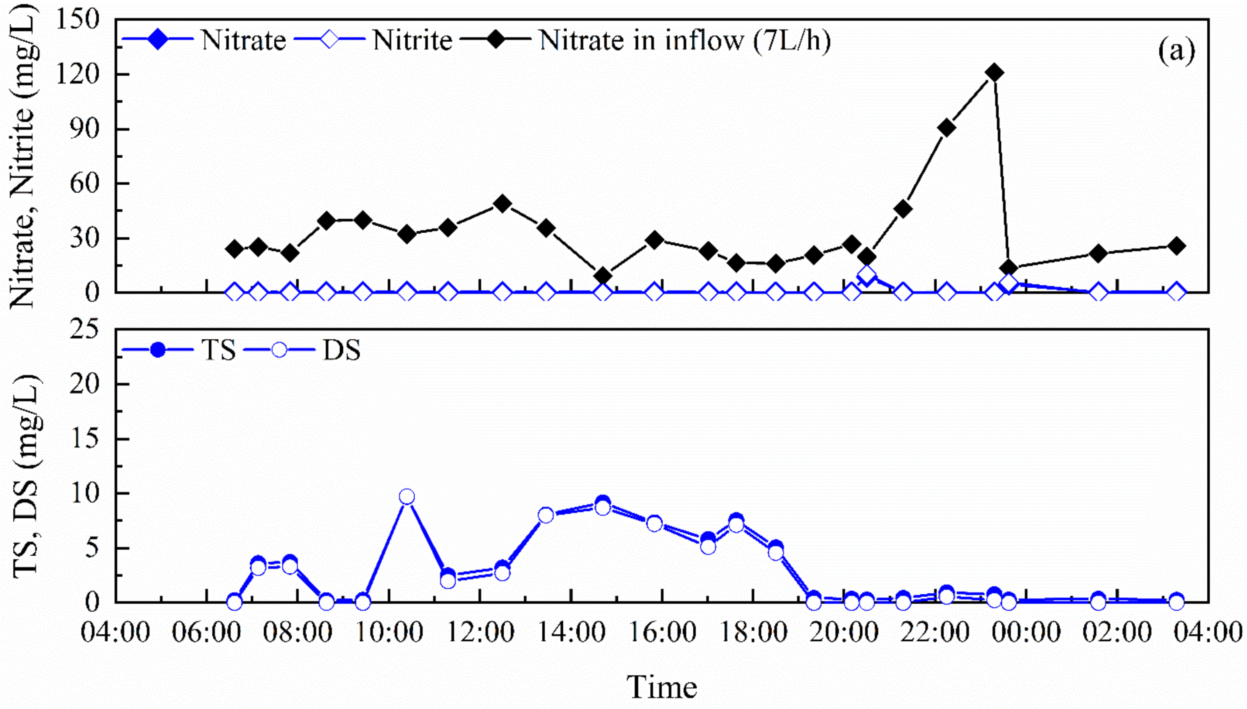


Figure 7-13 The 24-hour profile of (a) TCOD and DCOD, (b) ORP and DO, (c) T and pH in the wet well under different nitrate dosing rate (7L/h: Sept. 25th, 2020, 15 L/h: Oct. 09th, 2020)

With regard to discharge manhole, when the nitrate dosing rate was 7 L/h, the nitrate in inflow was 10~50 mg/L from 4:00 to 19:00 and the sulfide decreased to around 3~10 mg/L (**Figure 7-14 (a)**). However, the sulfide was 0 mg/L from 19:00 to 4:00 during which there was two points of high nitrate concentration: 90 and 120 mg/L. No remaining nitrate was detected during the whole day at discharge manhole except for 20:00 to 2:00 when the remaining nitrate and nitrite were both 5~10 mg/L. When the nitrate dosing rate was 15 L/h, the sulfide was only detected from 12:00 to 18:00 with the sulfide value 0.7~5 mg/L (**Figure 7-14(b)**). The nitrate concentration in inflow was 20~85 mg/L during the day except at 20:00 when the value was 360 mg/L. And the residue nitrate and nitrite were detected from 16:00 to 21:00 with nitrate and nitrite value of 2~47 mg/L and 2~26 mg/L.



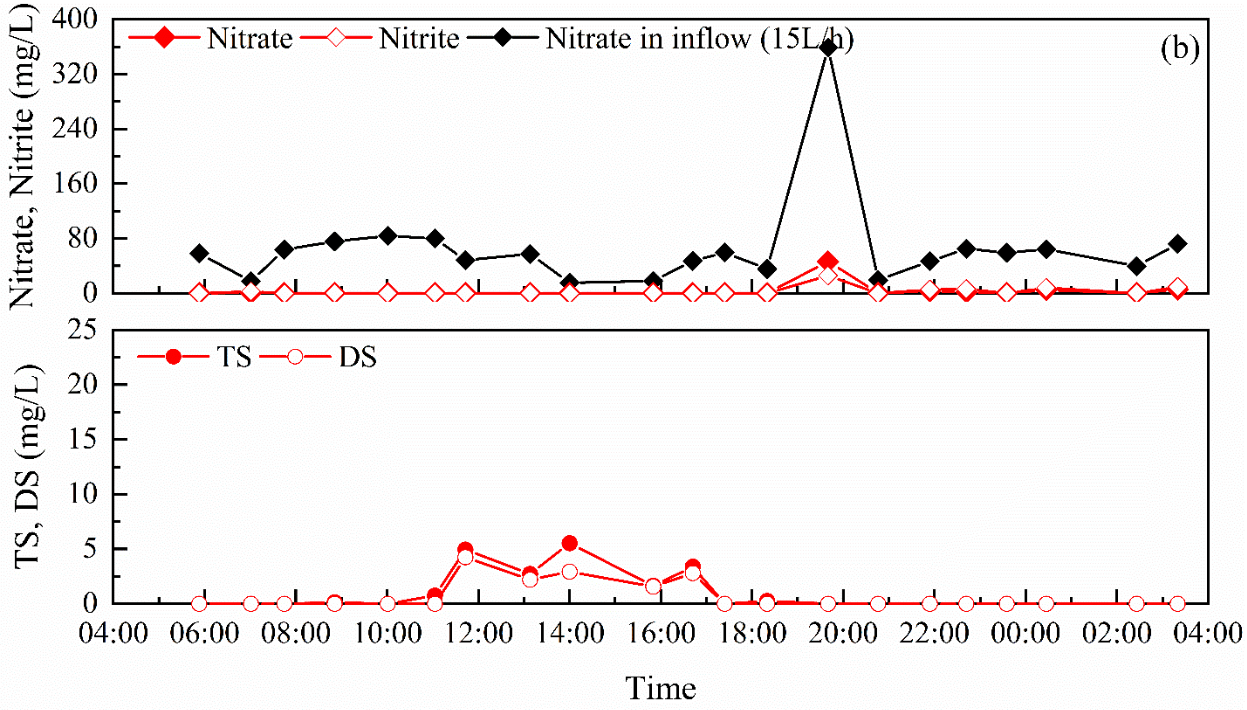


Figure 7-14 Concentration of nitrate and nitrite and sulfide in the discharge manhole: (a) 7L/h nitrate dosing rate (Sept. 25th, 2020), (b) 15L/h nitrate dosing rate (Oct. 09th, 2020)

The H_2S gas dropped from 300~400 ppm to 100~200 ppm, 50~100 ppm, 30~50 ppm and 0~20 ppm with the dosage of 5 L/h, 7 L/h, 11 L/h and 15 L/h, respectively **(Figure 7-15**). The H_2S dropped to zero at midnight after nitrate dosing. When the dosing rate was 15 h/L. No H_2S detected during the whole day except the period from 12:00 to 18:00 with H_2S value of 10~20 ppm. There was a sharp drop in TCOD and DCOD after nitrate addition. The TCOD was 641 mg/L and 490 mg/L under 7 L/h and 15 L/h nitrate dosing rate with respect to 740 mg/L baseline value (**Figure 7-16 (a)**). The DCOD was around 202 mg/L and 150 mg/L under 7 L/h and 15 L/h nitrate dosing rate and the baseline value was 300 mg/L. The ORP during some period surged from -200 mV to 10~-50 mV (SHE) after nitrate addition (**Figure 7-16 (b)**). The temperature of first two sampling campaign was close (around 17 °C) and it decreased 2 °C at the third sampling campaign (**Figure 7-16 (c)**). The pH rose from 7.4 to 8.3 and 8.6 with 7 L/h and 15 L/h nitrate dosing rate.

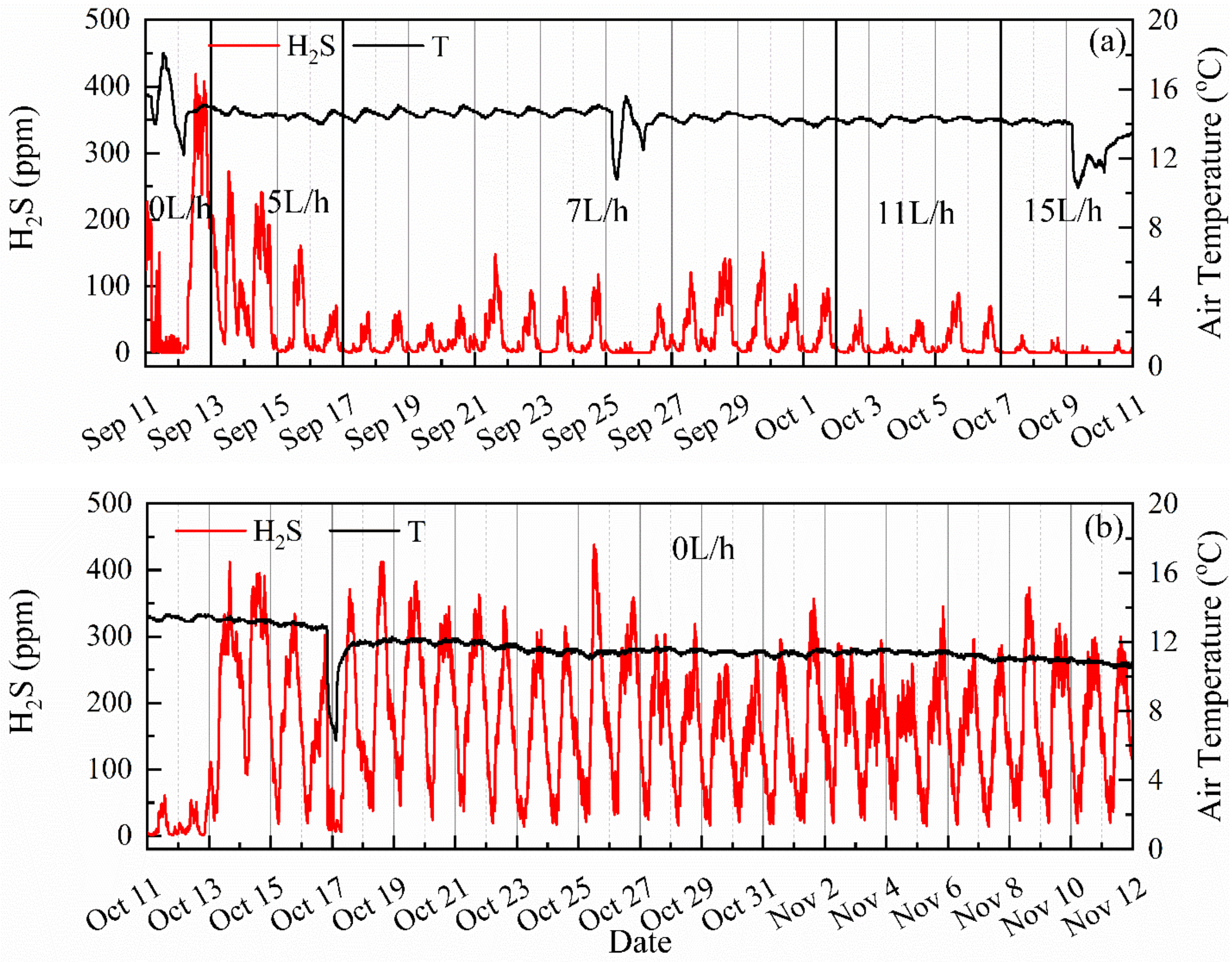


Figure 7-15 H_2S gas concentration in the discharge manhole under different nitrate dosing rate: (a) dosing phase, (b) recovery phase

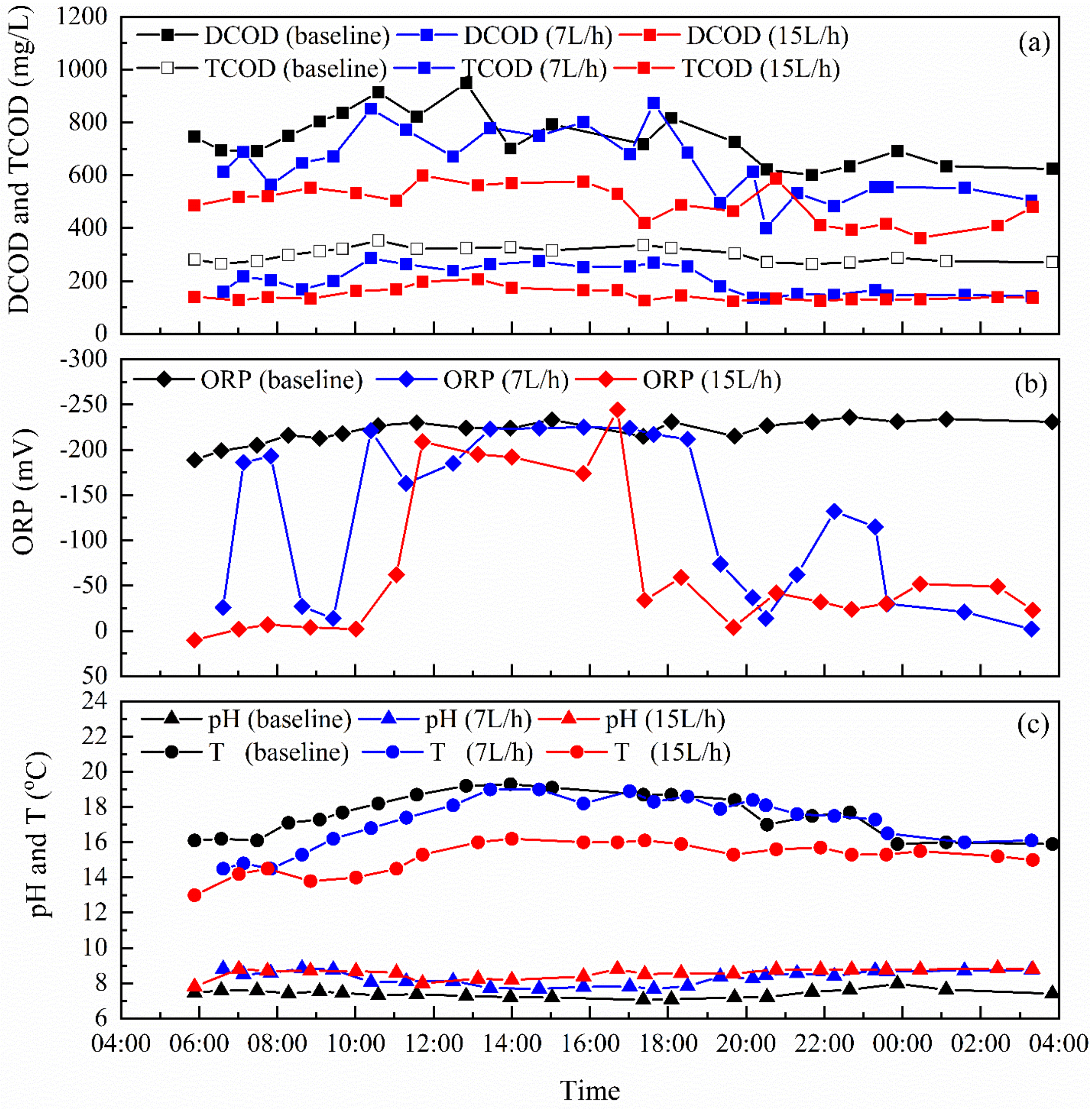


Figure 7-16 The 24-hour variation of (a) TCOD and DCOD, (b) ORP, (c) T and pH in the discharge manhole under different nitrate dosing rate (7L/h: Sept. 25th, 2020, 15 L/h: Oct. 09th, 2020)

The addition of nitrate effectively suppressed the sulfide generation in the force main. When the nitrate dosing rate was 7 L/h, the dosage (10~50 mg/L) was not enough to completely suppress the sulfide generation for most of the time. However, the high nitrate concentration (90 and 120 mg/L) coming from midnight when the flow rate was low which diffused along the force main completely suppressed the sulfide generation from 19:00 to 4:00. When the nitrate dosing rate was 15 L/h, the general dosage of 85 mg/L was the appropriate dosage for the force main from 4:00~11:00 when the sulfide was zero and there was no residue nitrate. And it was overdosed from 16:00 to 21:00 due to 360 mg/L nitrate at 20:00 coming from midnight in wet well. However, the dosage was underdosed from 11:00 to 16:00 during which there were two low nitrate dosage with concentration of around 20 mg/L. The nitrate addition induces the development of heterotrophic nitrate reducing bacteria (hNRB). The hNRB was able to utilize the DCOD to reduce nitrate to nitrogen gas. The denitrification process of hNRB suppressed the SRB for competing the carbon source. Thus, the TCOD and DCOD dropped with nitrate addition. The alkalinity generated during denitrification process, so pH increased with nitrate addition. The denitrification process generally happened under the ORP ranging from 50 mV to -50 mV (SHE). Therefore, the ORP has increased to 10~-50 mV during some period when the sulfide generation was completely suppressed and no sulfide was detected.

The nitrate addition in wet well was terminated on Oct. 10, 2020. As seen in the **Figure 7-15 (b)**, the nitrate quickly rebounded to pre-dosing level (300~400 ppm) at discharge manhole after only two days. The 8-hour sampling was conducted on Oct. 16, one week after cessation of nitrate dosage. **Table 7-5** indicates that the sulfide returned to pre-dosing level too, around 16 mg/L**.** It was a little lower than baseline value (20 mg/L) due to the lower temperature. The temperature decreased from 17.5 ℃ to 13.3 ℃. All the other water parameters returned to pre-dosing phase. Nitrate did not have long-lasting inhibitory/toxic effect on sulfate reduction and did not shift the major SRB populations in the biofilm of force main.

Table 7-5 Water quality at wet well and discharge manhole one week after termination of nitrate dosing (Oct.16th 8:00 PM to Oct.17th 4:00 AM)

Location	Time	pH	DO	T	ORP	TS	DS	Sulfate	TCOD	DCOD
			mg/L	°C	mV	mg/L	mg/L	mg/L	mg/L	mg/L
wet well	10:27 PM	8.3	4.0	16.9	-122	1.4	0.8	41.5	345	735
	11:21 PM	8.2	4.1	18.1	-145	1.5	1.0	44.8	324	755
	12:35 AM	7.9	4.5	17.7	-139	1.6	0.0	46.3	344	788
	3:00 AM	Sampling hose got frozen								
Discharge manhole	8:38 PM	7.6	3.1	13.1	-214	17.6	16.5	27.7	277	681
	9:25 PM	7.6	4.2	11.5	-212	17.2	16.8	25.6	282	786
	10:27 PM	7.7	3.9	13.6	-217	16.6	15.3	25.3	277	613
	11:21 PM	7.6	2.5	14.0	-228	16.9	16.3	28.3	283	676
	12:35 AM	Sampling hose got frozen								
	3:00 AM	7.6	2.5	14.5	-225	17.2	16.6	28.6	292	733

7.3.3 Discussion on optimization of nitrate dosing in force mains

Both the dosing location and dosing rate are important when dosing nitrate for sulfide control in force mains. Part of nitrate could be consumed at the wet well. Injecting nitrate directly into the beginning of the force main rather than in the wet well is better. If applicable, the best dosing location is at downstream section of force main while securing enough HRT for sewer biofilm to oxidize the generated sulfide from upstream section according to our lab study. The generated sulfide from the upstream section of the force main could be removed when it is passing the downstream section of the force main. The coexistence of sulfide and nitrate in the downstream section can stimulate the development of nitrate-reducing and sulfide-oxidizing bacteria (NR-SOB) which is responsible for sulfide removal. The nitrate-reducing, sulfide-oxidizing bacteria (NR-SOB) prevailed when sulfide and nitrate was co-existing. NR-SOB can remove the existing sulfide by oxidizing it to sulfate, using nitrate as the electron acceptor to produce primarily nitrite and N_2. The autotrophic denitrification process was dominant. The HRT which allowed the complete oxidation of sulfide generated reduced was shorter than that along the whole pipe section when dosing nitrate at the downstream section of the force main. Therefore, the amount of nitrate required to completely control the sulfide generation can be reduced significantly by dosing at the downstream section of the force main. However, the expected HRT which allows the complete oxidation of the sulfide formed in the upstream section of the force main needs to be investigated in the field which determines the exactly dosing location in the force main.

In terms of nitrate dosing rate, the nitrate was heavily overdosed at midnight when the inflow rate was small and the dosage was underdosed during the peak hour when the inflow rate was large. Therefore, the continuous injection of nitrate in the wet well at a constant flow rate was not an optimal dosing rate. When dosing at the wet well or at the beginning of the force main, flow-paced dosing rate aims to achieve the constant nitrate concentration in sewage is better than constant dosing rate. If the HRT in the force main varies considerably, the dosing nitrate concentration should be proportional to the HRT other than dosing at a constant nitrate concentration. For the dosing location at downstream section, the dosing rate should be designed that the amount of nitrate added to a sewage slug is proportional to the expected HRT of this slug in the remaining section of the pipe.

7.4 Conclusions

The effect of nitrate on sulfide control in the force main was investigated in field study. The following conclusions are drawn:

(1) According to field sampling results, the sulfide generation rate per biofilm area in the force mains of Blackburne, Kaskitayo and Twin Brooks pump station was around 0.09~0.15 g / m^2h under 20 °C. The observed sulfide production rates are of the same order of magnitude as measured in other investigations. The sulfide generation rate at the force main of the Big Lake pump station was 0.08~0.12 g/m^2h under 20 °C. There was around 17 mg/L sulfide generation in the force main of the Big Lake with 14.5 h~19.5 h retention time. There was approximately 3 mg/L sulfide found in the wet well of Big Lake pump station which came from upstream sources. This resulted in 30~50 ppm H_2S gas at wet well. Therefore, 20 mg/L sulfide was detected in discharge manhole in total which led to 400~500 ppm H_2S gas at discharge manhole. Applying the 1/2-order biofilm kinetics of DCOD to model the

sulfide generation rate in the force main in the field trial and the modelled rate constant for field trial was around 0.006, which was in the agreement with our lab study (in Chapter 6).

(2) The production of sulfide in the force main was successfully controlled when the nitrate was added. When the nitrate dosing rate was 7 L/h, the nitrate was underdose with the general dosage from 10 to50 mg/L) during the whole day except for 19:00 to 4:00. The nitrate was overdosed during 19:00 to 4:00 due to the high nitrate concentration (90 and 120 mg/L) coming from midnight when the flow rate was low When the nitrate dosing rate was 15 L/h, the general dosage of 85 mg/L was the proper dosage for the force main from 4:00~11:00 when there was no excess of nitrate. But it was overdosed from 16:00 to 21:00 due to 360 mg/L high nitrate at 20:00 coming from midnight in wet well and underdosed from 11:00 to 16:00during which there were two low nitrate dosage with concentration of around 20 mg/L. The nitrate addition induces the development of heterotrophic nitrate reducing bacteria (hNRB). The hNRB was able to utilize the DCOD to reduce nitrate to nitrogen gas. The denitrification process of hNRB suppressed the SRB for competing the carbon source. The TCOD and DCOD dropped with nitrate addition. The alkalinity generated during denitrification process and pH increased with nitrate addition. The denitrification process generally happened under the ORP ranging from 50 mV to -50 mV. Therefore, the ORP has increased to 10~-50 mV during some period when the sulfide generation was completely suppressed and no sulfide was detected.

(3) The continuous doing of nitrate at a constant flow rate in the wet well was not an optimal nitrate dosing strategy, because part of the nitrate could be consumed in the wet well and the nitrate was dramatically overdosed at midnight when the flow rate was low. Both dosing location and dosing rate are important for nitrate dosing strategy. It is better that the nitrate is added directly into the beginning of the force main rather than in the wet well. If at all tenable, the best dosing location is at downstream section of force main while securing enough HRT for sewer biofilm to oxidize the generated sulfide upstream. The amount of nitrate required to completely control the sulfide generation can be reduced significantly by dosing at the downstream section of the force main. When dosing at the wet well and at the beginning of the force main, flow-paced dosing rate aims to achieve the same nitrate

concentration in sewage is better than constant dosing rate. If the HRT in the force main varies considerably, the dosing nitrate concentration should be proportional to the HRT other than dosing at a constant nitrate concentration. For the dosing location at downstream section, the dosing rate should be designed that the amount of nitrate added to a sewage slug is proportional to the expected HRT of this slug in the remaining section of the pipe.

Chapter 8 Summary and Recommendations

8.1 Summary of research outcomes

The cause of H_2S problem in Steinhauer area was investigated through field study and modelling. The main findings are as follows:

- The H_2S at upstream of the trunk was caused by drop structure at the beginning of the trunk. The drop structure dramatically release the H_2S gas in the air phase and led to the pressurization of the trunk. The problem can be addressed by drop structure retrofitting of the drop structure.
- The H_2S at downstream of the trunk was due to the long HRT of sewage at wet well of the pump station and downstream of the trunk.
- The WATS and SeweX models for H_2S generation were calibrated by field data. The models were used to simulate the H_2S generation by applying optimized pump operation as control strategy. The results show that there was no H_2S gas present if applying pump operation optimization.

The effect of drop structures on H_2S emission and transport in Bonnie Doon area was investigated through field study. The main findings are as follows:

- The high H_2S gas concentration was detected in the middle reach of the trunk due to the stripping effect of the three drops (2.7 m, 5.2 m and 2.0 m) along the trunk.
- The overall H_2S mass transfer coefficient was around 300 h^{-1} with drop height 2.7 m followed by drop height 5.2 m and 54 h^{-1} with drop height 2.0 m.
- The released H_2S gas at these drops was transported to downstream of the trunk and some laterals made some of them as hotspots in the sewer networks.

The effects of nitrate and ferric on sulfide control in force mains were both investigated in the lab using 4 bioreactors. The key findings are:

- Lab study demonstrates that half order kinetics regarding DCOD was capable of predicting the sulfide formation in the bioreactors and the modelled empirical rate constant was in the range of 0.005~0.0011. The numbers were useful to predict the sulfide generation rates in force mains.

- Lab study shows ferric could effectively control the sulfide generation in the force main. However, it did not inhibit the sulfide generation rate in the bioreactor. Hydrogen sulfide was controlled with the ferric salt addition only via chemical oxidation and precipitation and total SRB population did not change.

- Nitrate addition at the beginning of pump cycle induced the development of *Thauera* as hNRB. The hNRB outcompeted SRB for organic matter as electron donor as the dominant bacteria resulted the suppression of sulfide generation. The amount of nitrate required for sulfide control depended on heterotrophic denitrification rate and the heterotrophic denitrification in the bioreactor could be well described by half order kinetics with regard to DCOD concentration. A modified empirical equation was provided which was able to predict the denitrification rate in the biofilm. The modelled empirical rate constant b was around 0.05. The developed equation could be a very useful tool for design of nitrate dosing strategy. Nitrate did not have the long-lasting inhibitory/toxic effect on sulfate reduction.

- The cost-effective dosing strategy for nitrate dosing in the force main was to add the nitrate at the end of pump cycle instead of at the beginning of pump cycle which could save up to about 75% nitrate dosage. The co-existence of sulfide and nitrate stimulates autotrophic denitrification. *Sulfurovum*, which is nitrate-reducing sulfide-oxidizing bacteria (NR-SOB), was the main contributor to sulfide oxidation resulting in sulfide removal in the reactor.

The effect of nitrate on sulfide control in the force main was investigated in field study. The following conclusions are drawn:

(1) According to field sampling results, the sulfide generation rate per biofilm area in the force mains of Blackburne, Kaskitayo and Twin Brooks pump station was around 0.09~0.15 g / m^2h under 20 °C. The observed sulfide production rates are of the same order of magnitude as measured in other investigations. The sulfide generation rate at the force main of the Big Lake pump station was 0.08~0.12 g/m^2h under 20 °C. There was around 17 mg/L sulfide generation in the force main of the Big Lake with 14.5 h~19.5 h retention time. There was approximately 3 mg/L sulfide found in the wet well of Big Lake pump station which came

from upstream sources. This resulted in 30~50 ppm H_2S gas at wet well. Therefore, 20 mg/L sulfide was detected in discharge manhole which led to 400~500 ppm H_2S gas at discharge manhole. Applying the 1/2-order biofilm kinetics of DCOD to model the sulfide generation rate in the force main in the field trial and the modelled rate constant for field trial was around 0.006, which was in the agreement with our lab study (in Chapter 6).

(2) The production of sulfide in the force main was successfully controlled when the nitrate was added. When the nitrate dosing rate was 7 L/h, the nitrate was underdose with the general dosage from 10 to50 mg/L) during the whole day except for 19:00 to 4:00. The nitrate was overdosed during 19:00 to 4:00 due to the high nitrate concentration (90 and 120 mg/L) coming from midnight when the flow rate was low When the nitrate dosing rate was 15 L/h, the general dosage of 85 mg/L was the proper dosage for the force main from 4:00~11:00 when there was no excess of nitrate. But it was overdosed from 16:00 to 21:00 due to 360 mg/L high nitrate at 20:00 coming from midnight in wet well and underdosed from 11:00 to 16:00during which there were two low nitrate dosage with concentration of around 20 mg/L. The nitrate addition induces the development of heterotrophic nitrate reducing bacteria (hNRB). The hNRB was able to utilize the DCOD to reduce nitrate to nitrogen gas. The denitrification process of hNRB suppressed the SRB for competing the carbon source. The TCOD and DCOD dropped with nitrate addition. The alkalinity generated during denitrification process and pH increased with nitrate addition. The denitrification process generally happened under the ORP ranging from 50 mV to -50 mV. Therefore, the ORP has increased to 10~-50 mV during some period when the sulfide generation was completely suppressed and no sulfide was detected.

(3) The continuous doing of nitrate at a constant flow rate in the wet well was not an optimal nitrate dosing strategy, because part of the nitrate could be consumed in the wet well and the nitrate was dramatically overdosed at midnight when the flow rate was low. Both dosing location and dosing rate are important for nitrate dosing strategy. It is better that the nitrate is added directly into the beginning of the force main rather than in the wet well. If at all tenable, the best dosing location is at downstream section of force main while securing enough HRT for sewer biofilm to oxidize the generated sulfide upstream. The amount of nitrate required to

completely control the sulfide generation can be reduced significantly by dosing at the downstream section of the force main. When dosing at the wet well and at the beginning of the force main, flow-paced dosing rate aims to achieve the same nitrate concentration in sewage is better than constant dosing rate. If the HRT in the force main varies considerably, the dosing nitrate concentration should be proportional to the HRT other than dosing at a constant nitrate concentration. For the dosing location at downstream section, the dosing rate should be designed that the amount of nitrate added to a sewage slug is proportional to the expected HRT of this slug in the remaining section of the pipe.

8.2 Recommendations for future research

During the whole period of my PhD, many research challenges have been identified that entail further research. Some of the recommendations for the future research are summarized below:

- The main problem of current hydrogen sulfide emission control technologies is the cost. The demand for the current different chemicals to control hydrogen sulfide is rather high, so it is costly. To optimize the chemical dosing is essential to reduce the dosage for controlling hydrogen sulfide emission in sewer systems. The optimized nitrate dosing strategy was developed. However, further studies are required to demonstrate the practical and financial feasibility of nitrate optimization in the field. The new chemicals that can inhibit or kill SRB efficiently while do not have environmental impacts could be cost-effective which needs to be explored in future studies.
- The effect of drops on hydrogen sulfide release is dramatic, but accurate methods or empirical expressions to evaluate the gas stripping rate at those particular sites are still missing. Little information is known about H_2S emissions under highly turbulent conditions, which is the key to understand and predict local odor and corrosion problems encountered at force main discharges, sewer drop structures and other similar sections. The approach for the prediction of H_2S gas emission under specific highly turbulent conditions needs to be developed.
- Apart from sulfide, VOSCs (volatile organic sulfide compounds) are also important sewer emissions causing sewer odor but little attention was paid to this aspect in the past. With the changed urban water management practices, the transformation of VOSCs in sewer

systems may also be affected. However, the studies of VOSC are always hindered by its complicated detection methods and reactive nature.

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