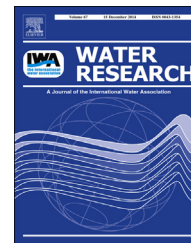


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Degradation of methanethiol in anaerobic sewers and its correlation with methanogenic activities



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ABSTRACT

Methanethiol (MT) is considered one of the predominant odorants in sewer systems. Therefore, understanding MT transformation in sewers is essential to sewer odor assessment and abatement. In this study, we investigated the degradation of MT in laboratory anaerobic sewers. Experiments were carried out in seven anaerobic sewer reactors with biofilms at different stages of development. MT degradation was found to be strongly dependent on the methanogenic activity of sewer biofilms. The MT degradation rate accelerated with the increase of methanogenic activity of sewer biofilms, resulting in MT accumulation (i.e. net production) in sewer reactors with relatively low methanogenic activities, and MT removal in reactors with higher methanogenic activities. A Monod-type kinetic expression was developed to describe MT degradation kinetics in anaerobic sewers, in which the maximum degradation rate was modeled as a function of the maximum methane production rate through a power function. It was also found that MT concentration had a linear relationship with acetate concentration, which may be used for preliminary assessment of MT presence in anaerobic sewers.

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1. Introduction

For decades, the unpleasant odors emitted from sewer systems have been a major issue for water utilities (Boon, 1995; Hvitved-Jacobsen, 2002; Sharma et al., 2008). Studies and practices dealing with this problem were, for a long time, focused on hydrogen sulfide, a well-known odorant in wastewater. However, odors in wastewater can be caused by many other compounds, such as free ammonia, volatile fatty acids (VFAs), volatile organic compounds (VOCs) and volatile organic sulfur compounds (VOSCs) (Hvitved-Jacobsen, 2002; van Leerdam et al., 2011; Zarra et al., 2008). Among these compounds, VOSCs are believed to be of particular importance due to a combination of malodorous characteristics, high volatility and low odor thresholds which are typically at

the level of parts per billion by volume (ppbv) (Cheng et al., 2005; Munoz et al., 2010; Sivret et al., 2013). At higher concentrations, i.e. >0.5–20 parts per million by volume (ppmv), VOSCs could cause health problems (Kastner et al., 2003; Lomans et al., 2002b). As a result, VOSCs should be considered in the design and assessment of odor abatement systems (Sivret et al., 2013; Wang et al., 2014a, 2014b).

Methanethiol (MT) is a typical VOSC and has a putrid smell like rotten cabbage. The odor threshold value of MT (0.07 ppbv) is one of the lowest in the VOSC category (Feilberg et al., 2010). In addition, the volatility of MT (interpreted by the Henry's law constant which is equal to 5 kg bar/mol for solubility in water at 298.15 K) is one of the highest (Debruyne et al., 1995). Thus, MT could have a higher potential to contribute to malodor than other VOSCs, if all these compounds were present at similar concentrations. To our knowledge, the concentration

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ranges of VOSCs in sewer systems are not well-documented. However, some case studies have revealed that MT can be a predominant VOSC in wastewater and sewer gases. Hwang et al. (1995) measured that the average MT concentration in the influent of a wastewater treatment plant (WWTP) was about 3–200 times higher than other VOSCs like dimethyl sulfide (DMS), dimethyl disulfide (DMDS) and carbon disulfide (CS₂). Lasaridi et al. (2010) also found that MT was the dominant VOSC at a pump station and seven WWTPs in Greece. More recently, Wang et al. (2014a) conducted a long term VOSC monitoring program for sewers located at 18 different sites in two major Australian cities (Sydney and Melbourne). In both cities, the MT concentration (675.3–1421.1 µg/m³) in the sewer air was substantially higher than the concentrations of four other tested VOSCs (7.8 µg/m³–94.0 µg/m³). From these results, MT is likely a key odor-causing VOSC in domestic wastewater. Therefore, understanding the transformation of MT in sewer systems is critical for solving odor problems caused by VOSCs.

Our recent study (Sun et al., 2014a) found that MT concentration in anaerobic sewer wastewater changed dynamically with the hydraulic retention time (HRT). The concentration initially increased with the increase of HRT and then decreased. This indicated that MT was being both generated and degraded under anaerobic sewer conditions. The production of MT under anaerobic conditions is mainly attributed to the cleavage of sulfur containing amino acids or methylation of sulfide during the degradation of methoxylated aromatic compounds (Chasteen and Bentley, 2004; Higgins et al., 2006; Kadota and Ishida, 1972; Lomans et al., 2002b). However, the mechanism for MT degradation in anaerobic sewer systems is unclear. In the past few decades the degradation of MT has been studied in anaerobic/anoxic aquatic environments e.g. marine sediment (Kiene et al., 1986; Visscher et al., 1995), salt marsh sediment (Kiene and Capone, 1988; Kiene et al., 1986), freshwater sediment (Kiene et al., 1986; Lomans et al., 1999a, 1999c; Zinder and Brock, 1978) and anaerobically digested biosolids (Chen et al., 2005; Higgins et al., 2006; van Leerdam et al., 2006). Methanogens and/or sulfate reducing bacteria (SRB) were found to be responsible for MT degradation in these environments (Chen et al., 2005; Higgins et al., 2006; Kiene and Capone, 1988; Kiene et al., 1986; Lomans et al., 1999b, 1999c; Tanimoto and Bak, 1994; Visscher et al., 1995). These microorganisms also exist in anaerobic sewers (Guisasola et al., 2008; Sharma et al., 2008), and they could be involved in MT degradation. Since MT degradation could alleviate odor emission from sewers, an understanding of the process is essential for identification of MT emission hot spots along the sewer pipes, which would then provide guidance for odor abatement strategies in the water industry.

Therefore, the aim of this study is to understand the degradation of MT in anaerobic sewers. The study was carried out in biofilm reactors simulating anaerobic sewers at different biofilm development stages. Batch tests were conducted to investigate the degradation pathways and kinetics. A kinetics expression for MT degradation in anaerobic sewers is proposed based on the results of the batch test. To our knowledge, the present study describes for the first time the pathways and kinetics of MT degradation in anaerobic sewer systems.

2. Materials and methods

2.1. Operation of anaerobic sewer reactors

Seven cylindrical reactors (R1–R7), each with a volume of 1 L, were used in this study to grow anaerobic wastewater biofilms mimicking those in sewers (Guisasola et al., 2008). The seven reactors were operated in parallel, fed with actual domestic wastewater (Supporting Information, Fig. S1A). Biofilms were developed on the walls and inner surface of reactor lids. The wastewater was pumped into each reactor intermittently (Fig. S1B) to simulate the typical dynamic flow patterns of rising main sewers, where anaerobic wastewater biofilms grow (Sharma et al., 2008). The HRT of the wastewater in each reactor varied from 15 min to 3 h. Previous studies demonstrated that the biotransformation processes in these biofilm reactors mimic well those in real sewers (Guisasola et al., 2008; Gutierrez et al., 2009; Jiang et al., 2013, 2011).

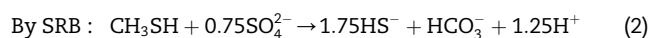
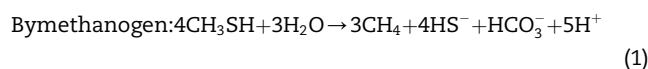
Based on the reactor performance, the seven reactors (R1–R7) were divided into two groups. R4–R7 had been operated for a minimum of two years and the biofilms had reached pseudo-steady states, indicated by stable methane and sulfide production rates (Fig. S2, SI). On the other hand, R1–R3 had been previously treated by FNA at various times for sulfide and methane control (Jiang et al., 2011). These three reactors were in the 'recovery' stage when experiments were conducted. The methane and sulfide production rates measured before the experiment are also shown in Fig. S2 (SI).

2.2. Batch tests I to correlate MT profile with methane, sulfide, DMS and acetate profiles

MT profiles in the seven reactors were monitored in 2–4 h batch tests. At the beginning of each test, fresh sewage was pumped through the reactor for 10 min to ensure complete replacement of liquid in the reactor. MT, methane, inorganic sulfide, DMS and VFA concentrations in the reactors were measured every 30 min. The methanogenic and sulfidogenic activities were described as the maximum methane production rate (MPR) and maximum sulfide production rate (SPR), respectively. These were calculated through linear regression of methane and sulfide concentration based on the methane and sulfide data in the first hour, during which there was no substrate limitation.

2.3. Batch tests II to identify MT degradation pathways

Studies in other anaerobic environments suggest that methanogens and SRB may be responsible for the consumption of MT in anaerobic sewers. The proposed reactions of MT degradation by methanogen and SRB are shown in Equation (1) and Equation (2), respectively (Finster et al., 1992; Zinder and Brock, 1978).



Based on this hypothesis, the role of methanogens and SRB on MT degradation in anaerobic sewers was investigated by

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