Dissolved methane in rising main sewer systems: field measurements and simple model development for estimating greenhouse gas emissions

Jeff Foley, Zhiguo Yuan and Paul Lant

ABSTRACT

At present, the potential generation of methane in wastewater collection systems is ignored under international greenhouse gas (GHG) accounting protocols, despite recent reports of substantial dissolved methane formation in sewers. This suggests that the current national GHG inventories for wastewater handling systems are likely to be underestimated for some situations. This study presents a new catalogue of field data on methane formation in rising main sewerage systems and proposes an empirically-fitted, theoretical model to predict dissolved methane concentrations, based upon the independent variables of pipeline geometry (i.e. surface area to volume ratio, A/V) and hydraulic retention time (HRT). Systems with longer HRT and/or larger A/V ratios are shown to have higher dissolved methane concentrations. This simple predictive model provides a means for water authorities to estimate the methane emissions from other pressurised sewerage systems of similar characteristics.

Key words | greenhouse gas emissions, methane, model, rising mains, sewers

INTRODUCTION

The United Nations Framework Convention on Climate Change is the globally recognised basis for collective action on the reduction of anthropogenic greenhouse gas emissions (UNFCCC 2007). One of the key obligations for signatory countries under the UNFCCC is the compilation of an annual national greenhouse gas (GHG) inventory, covering four general sectors (energy; industrial processes; agriculture, forestry and other land use; and waste). Emissions of methane and nitrous oxide from wastewater treatment and discharge are reported under the waste sector (IPCC 2006b). However, the plentiful supply of readily biodegradable carbon and the presence of anaerobic biofilms, particularly in fully surcharged rising mains, suggests that there is potential for the formation of methane in the raw sewage, and this is presently unaccounted in national GHG inventories (Foley et al. 2008).

Rising main sewers are anaerobic environments where both sulphate reduction and methanogenesis occur simultaneously using the organic material in wastewater as electron donors. However, most of the previous research on rising main modelling focused only on sulphide formation. Pomeroy (1959) and Thistlethwayte (1972) proposed empirical equations to predict sulphide formation based on wastewater organic strength, pipe characteristics and flow conditions. More recently, the WATS (Wastewater Aerobic/A Anaerobic Transformations in Sewers) model was developed to provide a more detailed description of the carbon and sulphur transformations in sewers (Hvitved-Jacobsen et al. 2000; Yongsiri et al. 2003; doi: 10.2166/wst.2009.718)
Abdul-Talib et al. 2005; Nielsen et al. 2005a,b; 2006). Successful attempts have also been completed to unify the traditional transport and conversion models for sewers with the Activated Sludge Model No.3 (ASM3) (Henze et al. 2000; Huisman et al. 2003). Most recently, Sharma et al. (2008) developed a comprehensive model to describe the dynamics of sulphide production in sewer systems. However, none of these models considered methane formation in rising mains. In their investigations of a pilot plant pressure sewer, Tanaka & Hvitved-Jacobsen (2002) did consider methane formation, but assumed it to be negligible.

Recently, Guisasola et al. (2008) showed a positive correlation between dissolved methane concentration and hydraulic retention time (HRT) in fully-surcharged, pressurised rising mains on the sub-tropical Gold Coast, Australia. Dissolved methane concentrations were shown to increase along the length of the sewer, which is reflective of increasing HRT, the plug-flow nature of fluid transport in fully surcharged pipelines and the methanogenic biofilm activity on the inner pipe surface. These authors showed a loss of 60–70 mg L\(^{-1}\) soluble chemical oxygen demand (COD) in laboratory-scale sewer reactors, of which 72% was demonstrated to be utilised by methanogens. These direct measurements of dissolved methane in full-scale rising mains (and corroborated by laboratory-scale reactors) confirmed that substantial generation potential of methane can take place in anaerobic rising mains.

The purpose of this study was to develop a theoretically robust, yet easily accessible model that related methane formation in rising mains to the independent variables of pipeline geometry (i.e. surface area to volume ratio) and hydraulic retention time. This functional model is proposed for water authorities to estimate the direct emissions of methane from similar pressurised rising main sewer systems, and hence address an acknowledged uncertainty in the IPCC GHG inventory guidelines for wastewater systems.

MATERIALS AND METHODS

Field sampling site

Field samples were obtained from along a single rising main (CO16) of the Gold Coast area (Queensland, Australia), as shown in Figure 1. Samples were collected at four locations: 1) at the CO16 pump station wet well; 2) a pressurised sampling point 500 m downstream; 3) a pressurised sampling point 1,100 m downstream; and 4) a pressurised sampling point 1,900 m downstream of the CO16 pump station. At 1,350 m (i.e. intersection of Amity Rd and Foxwell Rd), a second rising main (CO19) discharges into the larger CO16 rising main. Therefore, the samples collected at the 1,900 m sample point include the sewage from both rising mains. Samples from the first three locations were collected hourly between 5:00 a.m. and 8:00 a.m. to cover a wide range of HRTs, as the morning peak of raw wastewater entered the pump station’s collection wet well. Samples at the fourth location (1,900 m) were collected at 5:00 a.m. and 8:00 a.m. only.

The CO16 rising main had an internal pipe diameter of 300 mm (surface area to volume ratio, \(A/V = 13.3 \text{ m}^2 \text{ m}^{-3}\)), a total daily pumped flow of 707.4 m\(^3\), with 33 pump start events (typically 4–6 min in duration). The volume of wastewater pumped into the pipe during each pump run was calculated from the physical dimensions of the CO16 wet well and measured stop/start water levels.

The 1,700 m long CO19 rising main had an internal pipe diameter of 250 mm (surface area to volume ratio, \(A/V = 16.0 \text{ m}^2 \text{ m}^{-3}\)), a total daily pumped flow of 92.6 m\(^3\) and 34 pump start events (typically 2–3 min in duration). The volume of wastewater pumped into the pipe during each pump run was calculated from the physical dimensions of the CO19 wet well and measured stop/start water levels.

The HRT of a wastewater sample was defined as the time spent by that particular “slug” in the rising main up until the time of sampling. This was calculated from the operational data of the single pump stations feeding the CO16 and CO19 rising mains respectively (i.e. pump start/stop times recorded at 15 s intervals by the on-line supervisory control and data acquisition (SCADA) system), the pipeline geometry, and assuming ideal plug flow conditions. The HRT for the first three sample points ranged from 0.7 to 8.7 h (refer Table 1), depending on the time of day, incoming flowrate of raw sewage to the pump station wet well and the associated frequency of pumping events.

It was not possible to determine the HRT of the samples taken at the 1,900 m location, based on the pump stations’ run data alone, since the intersection of the two rising mains
nullified the assumption of ideal plug flow conditions. Accurate determination of the instantaneous HRT in this connected rising main system would require a tracer study, or more detailed network modelling than was possible under this study.

**Sample collection and analysis**

Dissolved methane was sampled and analysed using the methodology described by Alberto et al. (2000) and Guisasola et al. (2008). Sewage (containing dissolved methane) was sampled into freshly vacuumed BD Vacutainer® (BD Bioscience #367895) tubes using a hypodermic needle and 5 mL plastic syringe, attached directly to the pressurised rising main via a flexible hose (refer Figure 2). This procedure avoided any contact of the wastewater with atmosphere and possible oxygen interference. The Vacutainer tubes were weighed before and after sampling to determine the sample volume collected and mixed overnight in a shaker to allow equilibration of gas and liquid phases. Most of the methane (~97% at 25°C) was transferred to the gas phase in this process (Alberto et al. 2000). The methane concentration in the gas phase of the tubes was measured using a Shimadzu GC-9A Gas Chromatograph equipped with a flame ionization detector (FID). The concentration of methane in the initial liquid phase was then calculated using mass balance and Henry's law. The measurement error of this procedure is estimated to be ± 4.8% (t-disb., α = 0.05).

Samples were also collected for COD, soluble COD, and volatile fatty acids (VFAs). Where necessary, samples were immediately filtered using 0.22 μm syringe filters, acid-preserved and kept on ice before analysis. Temperature, pH, redox and salinity were also recorded for all field samples.

VFAs were measured by gas chromatography using a Perkin Elmer Autosystem equipped with a polar capillary column DB-FFAP and a FID. COD was measured using the colorimetric method described in APHA (1995) using commercial Lovibond tubes in a range of 0–150 mgCOD L⁻¹.
RESULTS AND DISCUSSION

The field measurements collected from rising main CO16 are shown in Table 1. By comparison, the saturation concentration for a 100% methane gas phase at 1 atm and 25°C is 21.5 mg CH$_4$ L$^{-1}$ (Perry et al. 1997). Also shown in Table 1 are field measurements previously reported for another Gold Coast rising main, UC09 (Guisasola et al. 2008). Table 1 highlights the increase in dissolved methane concentration along the length of the rising main, with a concomitant decrease in pH. This was also reflected in the accumulation of VFAs and indicates the onset of increasingly anaerobic conditions along the length of the rising main. This result corresponds well with the theory and results presented in Guisasola et al. (2008), which suggested that anaerobic methanogenesis increases with HRT.

The results also illustrate the persistence of dissolved methane in fresh raw sewage, even when the system is open to the atmosphere. The measurements taken at the CO16 pump station wet well all recorded concentrations of between 0.0010–0.0019 kg CH$_4$ m$^{-3}$ (i.e. 1.0–1.9 mg CH$_4$ L$^{-1}$), regardless of the time of sampling. Such results have not been previously reported and indicate that even fresh domestic sewage, such as that received directly into the wet well, has some small amount of methanogenic activity. Hence methane exchange was occurring continuously between the liquid phase and the gas phase. The sample collected at 5:00 a.m. was taken from sewage that had been accumulating in the wet well for up to 90 min (i.e. previous pumping event at 3:35 a.m.). Whereas the samples collected at 6:00 a.m., 7:00 a.m. and 8:00 a.m. were taken from sewage that had been accumulating in the wet well for

<table>
<thead>
<tr>
<th>Location</th>
<th>Time</th>
<th>Temp. (°C)</th>
<th>pH</th>
<th>HRT (h)</th>
<th>VFAs (kg m$^{-3}$)</th>
<th>Dissolved CH$_4$ (kg m$^{-3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CO16 P/Station</td>
<td>05:05</td>
<td>22.9</td>
<td>7.00</td>
<td>0.0</td>
<td>0.0444</td>
<td>0.00125</td>
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<td>05:25</td>
<td>22.9</td>
<td>6.83</td>
<td>5.4</td>
<td>0.0570</td>
<td>0.00567</td>
</tr>
<tr>
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<td>22.5</td>
<td>6.69</td>
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<td>0.00658</td>
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<td>6.70</td>
<td>–</td>
<td>0.0566</td>
<td>0.00884</td>
</tr>
<tr>
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<td>06:06</td>
<td>23.1</td>
<td>6.94</td>
<td>0.0</td>
<td>0.0389</td>
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</tr>
<tr>
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<td>06:18</td>
<td>24.1</td>
<td>6.77</td>
<td>0.8</td>
<td>0.0431</td>
<td>0.00458</td>
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<td>06:26</td>
<td>24.1</td>
<td>6.66</td>
<td>7.3</td>
<td>0.0571</td>
<td>0.00567</td>
</tr>
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<td>7.17</td>
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<td>0.0487</td>
<td>0.00192</td>
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<td>1.6</td>
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<td>7.13</td>
<td>3.8</td>
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<td>4.2</td>
<td>–</td>
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<tr>
<td>UC09–828 m</td>
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<td>27.5</td>
<td>7.31</td>
<td>4.4</td>
<td>–</td>
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<tr>
<td>UC09–828 m</td>
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<td>7.25</td>
<td>4.6</td>
<td>–</td>
<td>0.00547</td>
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<td>26.4</td>
<td>7.17</td>
<td>4.6</td>
<td>–</td>
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<tr>
<td>UC09–828 m</td>
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<td>7.15</td>
<td>4.6</td>
<td>–</td>
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<td>26</td>
<td>7.10</td>
<td>3.9</td>
<td>–</td>
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<tr>
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<td>7.15</td>
<td>3.1</td>
<td>–</td>
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only up to 11, 4 and 17 min respectively. Yet, the difference in dissolved methane concentration between these hourly samples was minimal. This indicates that there was continual methanogenic activity occurring upstream and within the wet well sediments and biofilms, and continual exchange of methane between the liquid and gas phase in the wet well. The dissolved methane concentration of 1.0–1.9 mg CH$_4$ L$^{-1}$ appears to represent a stable equilibrium between the rate of methane generation in the liquid phase and the rate of methane exchange to the gas phase. Quantifying the methane emissions from these open environments (e.g. gravity sewers and collection wet wells) requires further characterisation of the liquid-to-gas phase mass transfer coefficient, and certainly warrants further investigation.

The CO16 rising main is a total of 4,880 m in length, and at its end discharges into the PA9 pump station, where it mixes with another gravity sewer. The results from Table 1 suggest that dissolved methane would continue to accumulate along the full length of the rising main. At the point of discharge into the pump station, the wastewater could be subject to a high degree of turbulence as the CO16 wastewater mixes with the joining gravity sewer flows, depending on the structure design and prevailing flow conditions. Under these energetic conditions, it is possible that the much of the accumulated dissolved methane is stripped from the liquid phase wastewater into the sewer headspace, from where it ultimately leaks to the atmosphere. Similar stripping processes are also possible at air relief valves along the rising main, at the inlet works of a treatment plant, or at other points of discharge and turbulence in the sewer system. However, it is also possible that a substantial fraction of the dissolved methane is consumed via aerobic methane oxidation or anaerobic methane-driven denitrification. These possibilities warrant further investigation under full-scale conditions.

**MODEL DEVELOPMENT**

**Theoretical development**

The CO16 rising main can be assumed to approximate an ideal plug flow reactor. At each pumping event, a new “slug” of wastewater is pushed into the pipeline and this moves along the pipeline with each subsequent pumping event. For the purposes of this model, ideal conditions were assumed, whereby negligible longitudinal dispersion of the plug occurs over time. As the slug moves along the pipeline, it is exposed to the methanogenic biofilm on the pipe wall. The length of exposure of each wastewater slug to the pipeline biofilm reactor depends upon the HRT of the slug, or the frequency of pumping events, which changes throughout the day. From first principles then, the mass of methane generated in the wastewater slug can be expressed as:

$$M_{CH_4} = \text{Rate} \times A \times \text{HRT}$$  

(1)

where:

- $M_{CH_4}$: mass of dissolved methane (kg);
- Rate: specific rate of methanogenic activity of the biofilm (kg m$^{-2}$ h$^{-1}$);
- $A$: surface area of the biofilm (m$^2$); and
- HRT: hydraulic retention time of the particular slug (h).

Dividing through by the pipe volume to determine the dissolved methane concentration yields:

$$C_{CH_4} = \frac{M_{CH_4}}{V} = \frac{\text{Rate} \times A \times \text{HRT}}{V}$$  

(2)

Figure 2 | Collection of dissolved methane sample directly from the rising main into an airtight syringe.
where:

\[ C_{CH_4} \quad \text{concentration of dissolved methane (kg m}^{-3}) \]
\[ V \quad \text{volume of pipe (m}^3 \text{)} \text{; and} \]
\[ A/V \quad \text{surface area to volume ratio of pipe (m}^{-1} \text{)} \]

The physical expression of Equation (2) is illustrated in Figure 3, where dissolved methane concentration (y-axis) is plotted against the lumped parameter \([A/V \times \text{HRT}] \) (x-axis). By Equation (2), the gradient of this curve describes the methanogenic activity rate of the biofilm in units of kg m\(^{-2}\) h\(^{-1}\). This relationship is shown in Figure 3, with the measured data from the CO16 rising main in this study and the previously reported dissolved methane concentrations from the UC09 rising main. The two data points from the 1,900 m sample point on CO16 were excluded because of the difficulty in accurately calculating their HRT.

**Empirical model fitting**

In Figure 3, a simple linear regression model has been applied to the CO16 and UC09 field data for the purposes of extrapolation and application to other rising main systems:

\[ C_{CH_4} = 5.24 \times 10^{-5} \left[ \frac{A}{V \times \text{HRT}} \right] + 0.0015 \]  

(3)

where,

- \(5.24 \times 10^{-5} \) kg m\(^{-2}\) h\(^{-1}\) equals the rate of methanogenic activity of the pipeline biofilm (i.e. 52.4 mg m\(^{-2}\) h\(^{-1}\)), empirically derived using a sum of least squares fitting algorithm in MS Excel; and
- \(0.0015 \) kg m\(^{-3}\) equals the average residual concentration of dissolved methane (i.e. 1.5 mg L\(^{-1}\)) measured in raw wastewater samples, at the CO16 pump station wet well.

This model formulation provides a robust and easily accessible means for water authorities to estimate the methane emissions resulting from other pressurised rising mains, that have similar characteristics to the systems described in this study (i.e. temperature, organic matter composition). Only two easily determined independent variables are required. Firstly, rising main geometry (i.e. \(A/V\) ratio) is generally well catalogued by water authorities in corporate databases. Secondly, the average hydraulic retention time of a particular rising main can be easily calculated from SCADA system data on flowrates and/or pump run times and nominal pumping capacities. Whilst it has been demonstrated in this study that rising main HRT has a marked diurnal profile, for the purposes of annual GHG inventory reporting, an average HRT is considered to be an acceptable approximation.
Calculation of methane emissions from a rising main

As an example, consider a 300 mm diameter rising main that has an average flow of 7,000 m$^3$ d$^{-1}$ and HRT of 4.0 h, before discharging into vented headworks at a wastewater treatment plant. For the purposes of this example, it was assumed that all the dissolved methane was stripped to atmosphere upon discharge from the rising main into the vented headworks. Applying Equation (3), multiplying up to a yearly inventory and converting to UNFCCC carbon dioxide equivalents (i.e. 21 kg CO$_2$-e per kg CH$_4$) yielded an annual GHG inventory of 230 tCO$_2$-e from the rising main.

As a comparison, the total GHG emissions from a typical biological nutrient removal wastewater treatment plant receiving 7,000 m$^3$ d$^{-1}$ would be in the order of 2,500–3,800 tCO$_2$-e per year (de Haas et al. 2009). Therefore in this example, the methane emissions from the inlet rising main accounted for an additional 6–9% of the annual GHG inventory for the plant. Larger rising main sewerage networks, with longer retention times, could be expected to contribute a higher percentage to the annual GHG inventory for a water authority.

In the authors’ experience, rising main pipelines are typically in the range of 150–600 mm diameter, with nominal design HRTs of less than eight hours. Therefore, it is expected that the proposed model and the range of $[A/V \times \text{HRT}]$ values shown in Figure 3 (i.e. 0–200 h m$^{-1}$) will cover the majority of rising main configurations in practice.

Comparison with state-of-the-art model

A recent publication by Guisasola et al. (2009) has extended upon their previous work, and proposed a comprehensive model that characterises the biological and physicochemical processes in sewers. This extended model accounts for the competitive interactions of sulfate-reducing bacteria (SRBs) and methanogenic archaea (MAs) in sewers. The model development is based primarily upon lab-scale experimental data, and partly validated using the UC09 field data previously reported (Guisasola et al. 2008).

The dissolved methane concentrations from this complex model were overlaid with the CO16 and UC09 field data in Figure 3. Note that the “Guisasola Model” has been modified by increasing all values by 0.0015 kg m$^{-3}$ (i.e. 1.5 mg L$^{-1}$) to account for the influent dissolved methane concentration in the raw wastewater. The data points from the Guisasola Model were developed by modelling a large combination of theoretical $A/V$ and HRT combinations. It is therefore interesting to note that there was very little scatter in the y-direction for these different $A/V$–HRT combinations. This suggests that the use of the lumped parameter $[A/V \times \text{HRT}]$ in the simplified model proposed by this study is a valid approximation for GHG inventory purposes.

In the region of $[A/V \times \text{HRT}] = 50–100$ h m$^{-1}$, there was a very good fit between the CO16-UC09 field data, the proposed model and the complex Guisasola Model. However at lower values, the field data clearly suggests that both models underestimate the dissolved methane concentration. This is believed to be caused by competitive interaction between SRBs and MAs for early fermentation products. Further field investigations are required to determine the true kinetics that operate at these very short residence times.

Finally, the biofilm methanogenic activity rate (i.e. slope of the line) calculated for the proposed model was shown to be a good fit with the complex Guisasola Model. This suggests that for the rising main systems investigated in this study, the proposed model has valid predictive power beyond the limited range of HRT conditions tested in the field. This good fit between the measured field data, the comprehensive dynamic biological and physicochemical model and the simplified model proposed in this study, suggests that whilst an empirical approach has been useful in this instance, there is indeed some fundamental validity to the linear regression in Figure 3.

CONCLUSIONS

At present, the potential generation of methane in wastewater collection systems is ignored under the UNFCCC/IPCC greenhouse gas inventory guidelines, as it is “not believed to be a significant source of methane”. The field data presented in this paper suggest that for the operational rising mains investigated, this is a false assumption.
Dissolved methane concentrations were measured at four different longitudinal locations, and at four different times during a typical morning peak event, and were shown to be dependent upon the pipeline $A/V$ ratio and the instantaneous HRT of the associated wastewater slug. This relationship is consistent with first principles reaction theory and with earlier data and findings by other authors (Guisasola et al. 2008; Guisasola et al. 2009).

Based on this theoretical relationship with two independent variables, an empirically derived model is proposed from the field data for the estimation of dissolved methane concentrations in other rising main sewerage systems, that have similar operational characteristics (i.e. temperature, organic matter composition). The empirically derived model of this study was shown to fit well with the more theoretically grounded biological and physicochemical sewer processes model, proposed by Guisasola et al. (2009).

This study addresses the widely recognised gap in the current international GHG accounting guidelines for methane emissions from pressurised rising main sewerage systems. For the particular systems investigated, it provides a robust, empirically fitted, theoretical model for the water authority to easily estimate the dissolved methane concentrations in their rising main networks, and hence the upper limit of methane emissions to the atmosphere. Future studies should address the equally important potential for methane generation and emission from gravity sewers with an open headspace.

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