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Dissolved methane in the influent of three Australian wastewater treatment plants fed by gravity sewers

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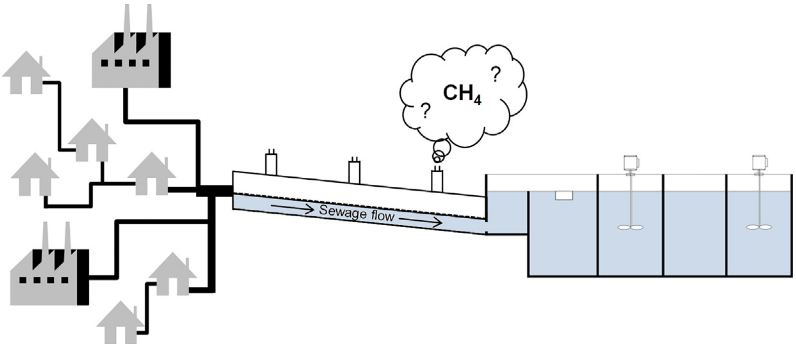
HIGHLIGHTS

- Raw sewage from municipal gravity sewers was surveyed for dissolved methane (CH_4).
- Sewered wastewater contained moderate levels of dissolved CH_4 (approx. 1 mg L^{-1}).
- Wastewater CH_4 levels correlated negatively with daily sewage flow rate.
- Emissions of up to 62 g CH4 person −1 y−1 estimated for gravity sewage entering WWTP
- Contrary to current IPCC consensus, gravity sewers are a source of $CH_{4.}$

GRAPHICAL ABSTRACT



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ABSTRACT

Methane (CH_4) is an important anthropogenic greenhouse gas and a by-product of urban sewage management. In recent years and contrary to international (IPCC) consensus, pressurised (anaerobic) sewers were identified as important CH_4 sources, yet relatively little remains known regarding the role of gravity sewers in CH_4 production and conveyance. Here we provide the results of a nine month study assessing dissolved CH_4 levels in the raw influent of three large Australian wastewater treatment plants (WWTPs) fed by gravity sewers. Similar to recent international research and contrary to IPCC guidance, results show that gravity sewered wastewater contains moderate levels of CH_4 ($\approx$1 mg L^{-1}). Dissolved CH_4 concentration correlated negatively with daily sewage flow rate (i.e. inversely proportional to sewer hydraulic residence time), with daily CH_4 mass loads on average some two-fold greater under low flow (dry weather) conditions. Along with sewage hydraulic residence time, sewer sediments are thought to interact with sewage flow rate and are considered to play a key role in gravity sewer CH_4 production. A per capita load of 78 g CH_4 person^{-1} y^{-1} is offered for gravity sewered wastewater entering WWTPs, with a corresponding emission estimate of up to 62 g CH_4 person^{-1} y^{-1}, assuming 80% water-to-air transfer of inflowing CH_4 in WWTPs with combined preliminary–primary plus secondary treatment. Results here support the emerging consensus view that hydraulic operation (i.e. gravity versus pressurised, sewage flow rate) is a key factor in determining sewer CH_4 production, with gravity sewer segments likely to play a dominant role in total CH_4 production potential for large metropolitan sewer networks. Further work is warranted to assess the scale and temporal dynamics of CH_4 production in gravity sewers elsewhere, with more work needed to

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adequately capture and assess the scale of diffuse sewer network CH_4 emissions from sprawling urban settlements globally.

1. Introduction

With a carbon dioxide (CO_2)-equivalent 100-year global warming potential of 28 (non-climate–carbon feedback) and the second-largest radiative forcing after CO_2 (IPCC, 2013) methane (CH_4) is an important anthropogenic greenhouse gas (GHG). Present day anthropogenic CH_4 emissions equal or exceed those from natural sources and at around 1800 ppb in 2011, global atmospheric CH_4 has increased 2.5-fold relative to pre-industrial levels. Despite a decade of near stability in global atmospheric CH_4 concentration during the 1990s, CH_4 levels are once again increasing (IPCC, 2013) and while total global CH_4 emissions are relatively well defined, the magnitude and dynamics of many individual CH_4 sources, including wastewater, remain poorly characterised (Dlugokencky et al., 2011).

CH_4 is an unavoidable by-product of municipal wastewater collection and treatment, and occurs during anaerobic microbial metabolism of organic substrates. If not intentionally captured and flared, CH_4 emissions can occur from a range of processes including anaerobic reactors (digesters, lagoons, septic systems), overloaded aerobic systems, open sewers and receiving environments (Doorn et al., 2006). Globally, the rate of CH_4 emissions from wastewater management practices has increased in recent decades as a result of an expanding and increasingly urbanised population (US EPA, 2006; Bogner et al., 2007). In the year 2000, CH_4 emissions from global wastewater management were estimated to represent some 5–7% of the total anthropogenic CH_4 source (US EPA, 2006; Denman et al., 2007); however, considerable uncertainties and gaps in emissions accounting methods remain (Doorn et al., 2006).

While open sewers throughout the developing world are a long-accepted and likely significant source of CH_4 (Doorn and Liles, 1999; Doorn et al., 2006), the extent to which enclosed municipal sewers in developed countries emit CH_4 has been much less clear. Current IPCC Guidelines for GHG inventories indicate that closed and underground sewers are "…not believed to be a significant source of CH_4" (Doorn et al., 2006; p. 6.8). Contrastingly, other IPCC authors stated around the same time that "Substantial emissions of CH_4 and N_2O can occur during wastewater transport in closed sewers…" (Bogner et al., 2007; p. 589). Remarkably, no referenced basis was provided in support of either claim.

Despite the early recognition by Czepiel et al. (1993) of appreciable CH_4 generation in the "influent lines" of a domestic wastewater treatment plant (WWTP), there was little research interest in sewer CH_4 during the ensuing 15 years. Since then it has emerged that underground sewers do emit significant amounts of CH_4 (Guisasola et al., 2008; Foley et al., 2009), with this limited work focusing on likely CH_4-producing 'hotspots' throughout sewer networks (i.e. anaerobic regions such as pressure mains, rising mains and pump station wet wells). In addition to these largely anaerobic zones, Foley et al. (2009) presented data from a limited field sampling campaign (1–2 days) which indicated that untreated domestic sewage exposed to the atmosphere was also methanogenic.

While this research provided evidence of CH_4 formation and anecdotal evidence of its persistence in gravity sewers, there remains a lack of information regarding the extent of CH_4 production and/or persistence in gravity sewer networks. Daelman et al. (2012), for example, stated that "…methane formation in sewer systems can be substantial, but actual quantities of methane entering a WWTP have as yet not been reported". At the same time, Willis et al. (2012), after detecting CH_4 emissions from a series of predominantly gravity-fed pumping stations, stressed that "…gravity sewer CH_4 emissions could be significant and warrant further research." Initial efforts to begin filling this gap were reported by Ren et al. (2013) who, following a four month study, reported relatively low levels of CH_4 in the raw influent of three Chinese WWTPs. Subsequent work has also reported on CH_4 emissions from sewer maintenance holes, pumping stations and sewer sediments (Chaosakul et al., 2014; Liu et al., 2014; Liu et al., 2015a).

Accordingly, this research sought to provide new information on the levels and dynamics of CH_4 in gravity sewered wastewater and to better understand the contributing factors by correlating CH_4 data with key system parameters. The raw influent of three large, primarily domestic Australian metropolitan WWTPs was routinely sampled over a nine month period and analysed for dissolved CH_4 to estimate the extent of CH_4 production and/or persistence in gravity sewered wastewater. Such an extended investigation of raw sewage CH_4 levels does not exist in the literature and as such, new information is provided on the conveyance and temporal dynamics of this important GHG in gravity sewers.

2. Material and methods

2.1. Study sites and sampling protocol

Untreated wastewater was collected from the raw influent at three large WWTPs (Plants A, B and C) in the state of New South Wales, Australia during the period December 2011 to August 2012 (Australian summer–winter). These three WWTPs service a combined equivalent population of three million people; key details of the WWTPs, their catchments and influent sewage are provided in Table 1. All three WWTPs provide primary level treatment prior to offshore wastewater discharge to the adjacent continental shelf. Wastewater to all three WWTPs is of medium strength (Henze and Comeau, 2008) and mainly domestic in origin (≥70%), with Plant A receiving largely domestic wastewater, Plant B receiving ≈30% industrial wastewater and Plant C intermediary between Plants A and C. While there are some three hundred pumping stations throughout the sewer networks servicing the three WWTPs (Table 1), wastewater flow to each coastal plant is primarily by gravity. Detailed information on sewer gradients was unavailable from the managing water utility; however, literature data indicates that the lower reaches of the Plant B network are relatively flat (≈0.05%) (Henry, 1939), while the main trunk sewer supplying Plant C is steeper (1.2%) (Wang et al., 2012). The hydraulic residence time (HRT) of sewer networks supplying each WWTP varies considerably with incident weather conditions, but under dry weather flows is nominally 5 h for Plant A, 21 h for Plant B and 15 h for Plant B.

During the nine month study, samples were collected on random weekdays a total of 11 times each for Plants A and B, with 12 sampling intervals for Plant C (see Short et al. (2014) and supplementary data Table S1 for more details). At each sampling interval, duplicate wastewater samples were drawn from the raw WWTP influent stream prior to any treatment interventions and as such represent final sewer network effluent. Bubble-free grab samples for dissolved CH_4 analysis were collected without headspace in 300 mL borosilicate glass bottles and hermetically-sealed with chlorobutyl/45 rubber septa-containing caps (Wheaton Science Products). Due care was taken to minimise the atmospheric exposure of sampled wastewater during sampling. Parallel grab samples (500 mL; Nalgene®) were also collected for conventional water quality analyses. To investigate possible sub-daily patterns in CH_4 dynamics, a limited one-off sampling program was also carried out at