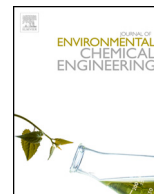




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## Municipal sewer networks as sources of nitrous oxide, methane and hydrogen sulphide emissions: A review and case studies

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### ABSTRACT

Sewers are known as longitudinal reactors where gases such as methane, nitrous oxide and hydrogen sulphide can be produced. However, gaseous emissions have been mainly assessed in wastewater treatment plants (WWTP). This article presents a critical review of studies that quantify the generation of these gases in sewers and to identify the existing research gaps. Differences in sampling methods and site selection, as well as a limited number of studies, result in incoherent comparisons. To address some of these gaps, sampling campaigns were conducted in two Spanish cities. Results showed that wet wells were the most important sources of gases with concentrations up to 321 µg CH<sub>4</sub> L<sub>air</sub><sup>-1</sup> and 6.8 µg N<sub>2</sub>O L<sub>air</sub><sup>-1</sup>. Regarding emission factors, in the case of Calafell, the estimated annual emissions were 18.6 kg CH<sub>4</sub> year<sup>-1</sup> and 0.3 kg H<sub>2</sub>S year<sup>-1</sup> in summer and 3.8 kg CH<sub>4</sub> year<sup>-1</sup> and 0.5 kg H<sub>2</sub>S year<sup>-1</sup> in winter. About Betanzos, these values were 24.6 kg CH<sub>4</sub> year<sup>-1</sup> and 0.5 kg N<sub>2</sub>O year<sup>-1</sup> in summer and 10 kg CH<sub>4</sub> year<sup>-1</sup> in winter. The summer campaign resulted in greater gas concentration than in the winter season for both cities, suggesting that temperature is a key parameter. We conclude that gas emissions from sewers are significant compared to that of WWTPs resulting in an important contribution to the carbon footprint. Further work needs to be done to assess the gas production along the entire sewer networks, which can result in very different emission factors depending on the sewer components.

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### Introduction

Currently more than 50% of the world's population resides in urban areas and is expected to increase to 70% by 2050 [1]. In this

framework, greenhouse gas (GHG) emissions coming from urban areas are major topics because of the processes leading to their production. In general, several studies have surveyed the environmental effects of GHG emissions at different scales and proposed some mathematical models related to transport and energy production [2–5]. However, because of urban intensification, more infrastructures will be required, in particular the ones related to water supply and sanitation. Hence, the sustainable management of the entire urban water cycle is a key point that needs to be addressed in order to meet the water needs of the citizens. Furthermore, an efficient water cycle management can potentially contribute to the local climate action plans for reduction of GHG emissions.

The urban water cycle includes water abstraction, drinking water treatment, water transport and distribution, water use, sewerage and rainwater transport and wastewater treatment. Due to the wastewater degradation and energy requirements, each phase contributes to the urban carbon footprint, but most of the attention has been placed on direct emissions from wastewater treatment plants (WWTPs). WWTPs have been recognised as a significant source of gaseous compounds [6], where the biological treatments result in GHG emissions of carbon dioxide ( $\text{CO}_2$ ), methane ( $\text{CH}_4$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ), as well as hydrogen sulphide ( $\text{H}_2\text{S}$ ), amongst many others.

The Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) has established guidelines for quantifying GHG emissions in WWTPs [7]. Based on these methodologies, the European Commission [8] reported that 9% and 3% of the world's  $\text{CH}_4$  and  $\text{N}_2\text{O}$  emissions figures come from WWTPs, respectively. However, there is a high degree of uncertainty in these figures given the rapid growth and urbanisation in developing countries. Furthermore, these values could be underestimated since they ignore the emissions taking place in sewer networks transporting the wastewater to the WWTPs. Sewer networks act as biological plug-flow reactors with high hydraulic retention time (HRT), which can be equal or higher than that at the WWTP depending on the configuration of the network [9]. Depending on the type of sewer (gravity or pressurised) or the type of constructive element, GHG such as  $\text{CO}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{CH}_4$ , as well as  $\text{H}_2\text{S}$  can result from aerobic, anoxic or anaerobic environments [10]. (Supporting information 1) For a more detailed description of the sewer elements and biological processes leading to emissions, we recommend [9,11,12].

Emissions of  $\text{CH}_4$  and  $\text{H}_2\text{S}$  from sewers have received a lot more attention than  $\text{N}_2\text{O}$  and  $\text{CO}_2$  given their safety, toxicity, and corrosion issues [13–19]. According to the IPCC,  $\text{CH}_4$  has a Global Warming Potential (GWP) 25 times higher than  $\text{CO}_2$  in a 100-year time horizon [20].  $\text{H}_2\text{S}$  has a Human Toxicity Potential of 0.22 kg of 1,4-dichlorobenzene (1,4-DB) eq.  $\text{kg}^{-1}$   $\text{H}_2\text{S}$  [21], and additionally, it can oxidise to sulphuric acid, resulting in the deterioration of sewer network elements [22], especially concrete pipes [23].

However,  $\text{N}_2\text{O}$  could be considered of higher concern than  $\text{CH}_4$  in terms of GWP, being 298 times greater than that of  $\text{CO}_2$  [20] and having a lifetime of 120 years in the atmosphere [24]. In addition, this gas is also a source of NO and  $\text{NO}_2$ , which participate in catalytic cycles that deplete ozone [20]. Given the Ozone Depletion Potential (ODP) and GWP of  $\text{N}_2\text{O}$ , it is important to determine its contribution to the carbon footprint of sewers and, therefore, to the entire urban water cycle. In spite of its importance, to the best of our knowledge very few studies have been specifically dedicated to determine  $\text{N}_2\text{O}$  production in sewer networks [25–27] compared to the existent literature on the production of this gas in WWTP [28–32].

In this sense, the purpose of this paper is to offer a critical overview of the published studies regarding  $\text{CH}_4$ ,  $\text{H}_2\text{S}$  and  $\text{N}_2\text{O}$  generation in sewer networks. The aim is to determine the

research gaps that need to be explored, in particular regarding the type of sewer system and the main parameters related to the quantification of these particular gases. In order to address some of the research gaps, two case studies of the sewer networks of two medium-sized Spanish cities (Betanzos in Atlantic area and Calafell in Mediterranean area) are presented, where the generation of  $\text{CH}_4$ ,  $\text{H}_2\text{S}$  and  $\text{N}_2\text{O}$  were quantified. These case studies serve to analyse the contribution of sewers to the urban water cycle footprint. Furthermore, in view of the literature review and the results of the case studies presented, we make several suggestions to guide future efforts in quantifying GHG emissions from sewer networks.

## Recent studies on GHG and $\text{H}_2\text{S}$ production in sewers

A compilation of previous literature quantifying  $\text{CH}_4$ ,  $\text{H}_2\text{S}$  and  $\text{N}_2\text{O}$  in sewer networks is presented in Table 1. Studies that focused on WWTP but provide a quantification of the emissions at the influent of the WWTP were also included. When possible, and based on data provided by the authors, we have converted the emission factors to  $\mu\text{g L}^{-1}$  (given in parentheses in Table 1), to facilitate comparison amongst studies. Comparison of the reported emission factors is not straight forward because data are reported in diverse units, different analytical and sampling methods are employed, and sampling conditions are not always reported. Calculations for unit conversions are provided in Supporting Information 2. The fourth column provides the sewer component considered (rising or gravity sewer, manhole, wet well or pumping station, influent to WWTP) and identifies which studies are conducted using data generated in the laboratory or in field sampling campaigns. A description of the methods used, the sampling conditions, and the main findings is also given. The latter are further discussed in the following paragraphs.

Emissions are often reported in different units and some assumptions were made in order to convert the values to  $\mu\text{g L}^{-1}$  since the studies did not provide all the data required for calculations. For example, Debruyne et al. [25] reported 23  $\mu\text{g}$  of  $\text{N}_2\text{O}$  per gram of suspended solids. In order to relate this result to 1 L of wastewater, a range of 270–550 mg of suspended solids per litre was applied [33] and the final converted values were 6.1–12.7  $\mu\text{g}$  of  $\text{N}_2\text{O L}^{-1}$ . In contrast, Clemens and Haas [26] gave an emission factor of 3.5 g of  $\text{N}_2\text{O}$  per person per year, and data on the population of Bayreuth served by the WWTP at the time of the study was not reported as it was a key parameter in determining the concentration. In other cases, an emission rate at the entrance of the WWTP was provided. For instance, Wang et al. [34] reported 4.34–6.82 g of  $\text{CH}_4 \text{ m}^{-2} \text{ day}^{-1}$ . According to data found in the study, there was an emitting surface of 45  $\text{m}^2$  and a daily wastewater production of 450  $\text{m}^3 \text{ day}^{-1}$  was assumed. At the end, a  $\text{CH}_4$  concentration of 434–682  $\mu\text{g L}^{-1}$  was obtained. To consult all the conversions see Supporting information 2.

As shown in Table 1, several authors have studied  $\text{CH}_4$  formation in sewers. Methane concentrations for wet wells at pumping stations previous to WWTP ranged from 200  $\mu\text{g L}^{-1}$  in Gold Coast (Australia) [35] to 3300,000  $\mu\text{g L}^{-1}$  in DeKalb County (Georgia, United States) [36]. In the case of rising mains, most studies were conducted in the Gold Coast (Australia), and methane concentrations ranged from 2800 to 30,000  $\mu\text{g L}^{-1}$  [16,28,37]. Nevertheless, the authors do not specifically determine which conditions were present in each sampling site (i.e., aerobic, anaerobic or anoxic) and a relationship between aeration and gas formation cannot be established.

Some of these studies further determined that dissolved oxygen inhibits methane formation [17,34,37], whereas a neutral pH (7.0–7.2) resulted to be optimal for methanogens to produce  $\text{CH}_4$  [34]. Other factors reported to influence  $\text{CH}_4$  formation are: biofilm area-to-liquid volume (A/V) ratio, HRT, amount of nitrite and

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