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## Plant-integrated measurement of greenhouse gas emissions from a municipal wastewater treatment plant



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#### ABSTRACT

Wastewater treatment plants (WWTPs) contribute to anthropogenic greenhouse gas (GHG) emissions. Due to its spatial and temporal variation in emissions, whole plant characterization of GHG emissions from WWTPs face a number of obstacles. In this study, a tracer dispersion method was applied to quantify plant-integrated, real-time emissions of methane and nitrous oxides. Two mobile cavity ring-down spectroscopy sampling devices were used to record downwind gas concentrations emitted from a municipal WWTP situated in Copenhagen, Denmark. This plant is equipped to remove biological nitrogen and employs anaerobic digestion for sludge stabilization. Over the course of nine measurement campaigns, a wide range of emissions were detected: methane from 4.99 kg  $h^{-1}$  up to 92.3 kg  $h^{-1}$  and nitrous oxide from below the detection limit (0.37 kg  $h^{-1}$ ) up to 10.5 kg  $h^{-1}$ . High emissions were observed during periods experiencing operational problems, such as during foaming events in anaerobic digesters and during sub-optimal operation of biological nitrogen removal in the secondary treatment of wastewater. Methane emissions detected during measurement campaigns corresponded to 2.07-32.7% of the methane generated in the plant. As high as 4.27% of nitrogen entering the WWTP was emitted as nitrous oxide under the sub-optimal operation of biological treatment processes. The study shows that the unit process configuration, as well as the operation of the WWTP, determines the rate of GHG emission. The applied plant-integrated emission measurement method could be used to ease the burden of quantifying GHG emissions from WWTPs for reporting purposes and could contribute to the development of more accurate depictions of environmental performance of WWTPs.

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#### 1. Introduction

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The treatment of wastewater and sewage sludge has been identified as a source of anthropogenic greenhouse gas (GHG)

emissions. Carbon dioxide ( $CO_2$ ) is formed through aerobic microbial degradation and through combustion of organic matter. More potent GHG gases, such as methane (CH<sub>4</sub>) and nitrous oxide ( $N_2O$ ), are also emitted during wastewater and sewage sludge treatment, as CH<sub>4</sub> is generated through the

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anaerobic degradation of organics, while N<sub>2</sub>O is produced as the result of the biological removal of nitrogen (N) through enhanced nitrification and denitrification (e.g. Kampschreur et al., 2009; Law et al., 2012; Shaw and Koh, 2011). In order to formulate effective mitigation strategies against global climate change, the Intergovernmental Panel on Climate Change (IPCC) formulated a guideline in 2006 to account for GHG emissions under the United Nations Framework Convention on Climate Change. The guideline considers that carbon in wastewater to be biogenic, and thus on-site emissions of CO<sub>2</sub> are to be excluded from reporting, while CH<sub>4</sub> and N<sub>2</sub>O emissions from wastewater treatment plants (WWTPs) are included in the national inventory (Bogner et al., 2007). In addition, N<sub>2</sub>O is now listed as the leading ozone depletion substance (Ravishankara et al., 2009). For the past 15 years, measurement campaigns have been conducted to determine reliable emission factors from WWTPs. A wide range of emission rates have been reported for N2O at WWTPs. Between 0.001 and 2.59% of the N in the influent has reportedly been emitted as N<sub>2</sub>O by WWTPs operating biological nitrogen removal processes (Benckiser et al., 1996; Ahn et al., 2010). An even higher emission rate was reported by a plant equipped with sequencing N removal batch reactors (5.6% of N in influent; Sun et al., 2013) and a plant with the combination of partial nitrification and anammox process (5.1-6.6% of N in influent; Desloover et al., 2011).

Spot emission measurements using floating flux chambers, combined with an analysis of liquid samples have been the most prevalent way of measuring fugitive GHG emissions from WWTPs (Czepiel et al., 1995; Sümer et al., 1995; Sommer et al., 1998; Foley et al., 2010; Ren et al., 2012; Aboobarkar et al., 2013). While several studies have been conducted over extended time periods (e.g. one year (Sümer et al., 1995) and one-and-a-half years for (Sommer et al., 1998), the results of the measurements provide only a snapshot of these dynamic systems and may not fully capture changes in emissions over time (Daelman et al., 2013a). In a few cases, in-line continuous measurements have been performed at WWTPs equipped with extensive air collection systems (Daelman et al., 2013b, 2012; Toyoda et al., 2011). Although these measurements provide an extensive dataset and are able to capture diurnal and seasonal changes in emissions, they only cover a portion of the plant, and so physical GHG leakages from pipes and fittings, as well as incidental releases of gases outside of the ventilation system, are not captured.

Besides spot and in-line measurements, some attempts have been made to quantify whole plant-scale GHG emissions escaping from open WWTPs. Recently Yver-Kwok et al. (submitted for publication), compared regional and plantintegrated measurement technologies for CH<sub>4</sub> emissions with the floating chamber methods, confirming the efficacy of the plant-integrated method. Modrak et al. (2006) applied a vertical radial plume mapping method using an open-path Fourier transform infrared (OP-FTIR) instrumentation. They found elevated concentrations of CH<sub>4</sub> and N<sub>2</sub>O but were only able to quantify the CH<sub>4</sub> emission rate due to the background concentration of N<sub>2</sub>O and the limited sensitivity of the instrument. In order to overcome these limitations, and to provide reliable plant-integrated GHG emissions measurements for wastewater and sludge treatment processes, a



Fig. 1 – Aerial photo of the Avedøre WWTP (Map data: Google, Aerodata International Surveys). Numbers correspond to the description in the Section 2.1.

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