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Evaluation of process conditions triggering emissions of green-house gases from a biological wastewater treatment system

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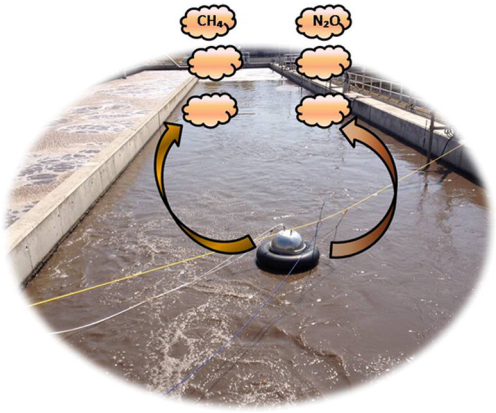
HIGHLIGHTS

- Monitoring of CH_4 and N_2O emissions from a full-scale activated sludge bioreactor
- Process perturbations leading to CH_4 and N_2O peak emissions were identified.
- Peak emissions increased severely the overall emission account of the bioreactor.
- CH_4 emissions were related with the inflow of influent and reject wastewater.
- N_2O was generated as consequence of nitrification imbalances.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, methane (CH_4) and nitrous oxide (N_2O) emission dynamics of a plug–flow bioreactor located in a municipal full-scale wastewater treatment plant were monitored during a period of 10 weeks. In general, CH_4 and N_2O gas emissions from the bioreactor accounted for 0.016% of the influent chemical oxygen demand (COD) and 0.116% of the influent total Kjeldahl nitrogen (TKN) respectively. In order to identify the emission patterns in the different zones, the bioreactor was divided in six different sampling sites and the gas collection hood was placed for a period of 2–3 days in each of these sites. This sampling strategy also allowed the identification of different process perturbations leading to CH_4 or N_2O peak emissions. CH_4 emissions mainly occurred in the first aerated site, and were mostly related with the influent and reject wastewater flows entering the bioreactor. On the other hand, N_2O emissions were given along all the aerated parts of the bioreactor and were strongly dependant on the occurrence of process disturbances such as periods of no aeration or nitrification instability. Dissolved CH_4 and N_2O concentrations were monitored in the bioreactor and in other parts of the plant, as a contribution for the better understanding of the transport of these greenhouse gases across the different stages of the treatment system.

1. Introduction

Carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) are the most important green-house gases (GHG) related to wastewater treatment processes. Due to the impact of these gases on the global

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climate, emissions from any industrial process should be understood, quantified and minimized. CH_4 and N_2O have a global warming potential which is about 25 and 265 times larger than the one attributed to CO_2 respectively, in a 100-year scope (IPCC, 2013). When assessing direct GHG emissions from wastewater treatment systems, only CH_4 and N_2O are considered, since CO_2 is assumed to originate from biogenic material and therefore it is excluded from greenhouse gas inventories (IPCC, 2013). Furthermore, there is very limited data on CH_4 and N_2O emissions from full-scale wastewater treatment systems, and the process conditions that trigger gas production and emission in WWTPs are still under investigation.

During collection and treatment of wastewater, anaerobic conditions may occur resulting in CH_4 production. In wastewater treatment systems, CH_4 is produced as a consequence of anaerobic processes, such as sludge digestion. Sewer systems have also been proven to be a source of CH_4 from which it can be transferred and released (Guisasola et al., 2008; Sudarjanto et al., 2014). Agitation and aeration during wastewater treatment facilitates CH_4 stripping to the atmosphere. To the best of our knowledge, there is only one published study reporting on-line CH_4 emissions from a domestic WWTP (Daelman et al., 2012). These authors reported that 1.13% of the influent chemical oxygen demand (COD) of the WWTP of Kralingseveer (Netherlands) was emitted as CH_4. About three quarters of these emissions were originated during primary and secondary sludge digestion. In that specific case, the CH_4-related footprint of the sludge digester was larger than the CO_2 emissions that were avoided by using biogas for energy generation (Daelman et al., 2012), putting into context the impact that uncontrolled CH_4 emissions can have on the overall carbon footprint of wastewater treatment processes. Previous research by other authors (Czepiel et al., 1993; Wang et al., 2011) also focused on measuring CH_4 emissions from WWTPs (with no anaerobic sludge digestion). These studies reported values between 0.08 and 0.16% of the influent organic load (biological oxygen demand (BOD)) or COD being emitted as CH_4. In both studies however, the analyses were based on grab samples not fully representative of the dynamics of the WWTPs and thus, the relevance of these results is more limited for comparative purposes.

N_2O production in wastewater treatment systems is generally attributed to nitrification and denitrification processes, with the first one being considered the source of the majority of emissions in full-scale plants (Colliver and Stephenson, 2000). Several parameters affecting N_2O production and emissions have been identified and evaluated. Low dissolved oxygen (DO) concentrations can lead to N_2O production during nitrification due to the activation of the nitrifier denitrification process (Tallec et al., 2006; Kampschreur et al., 2008; Pijuan et al., 2014). High nitrite (NO_2^-) concentrations have also been reported to be responsible for N_2O production by ammonia oxidizing bacteria (AOB) in nitrifying systems (Colliver and Stephenson, 2000). In denitrification stages, relatively high NO_2^- or free nitrous acid concentrations have been reported to lead to N_2O accumulation due to the inhibition of the last step of the denitrifying reaction (Schulthess et al., 1994; Zhou et al., 2008). Similarly, low COD/N ratios and high DO concentrations are known to increase N_2O emissions as a result of impaired denitrification performance (Schulthess et al., 1994; Otte et al., 1996). Some studies have also shown that transient conditions in terms of DO (oxic/anoxic), ammonium (NH_4^+) concentration (shock loading) or NO_2^- concentrations (accumulation) are of great importance and can generally generate N_2O emission events (Tallec et al., 2006; Kampschreur et al., 2008; Rodriguez-Caballero and Pijuan, 2013).

In the last years, data on N_2O emissions from full-scale WWTPs has been collected in different countries. In general, the methodology utilized for quantifying the emissions can be in itself a source of variability. The latest published results in terms of N_2O emissions have been obtained through online measurements, including the present study, which facilitates the evaluation of temporal patterns and dynamics. Ahn et al. (2010) presented N_2O emissions data from 12 different WWTPs located in the United States with results ranging from 0.01 to 1.8% of the influent total Kjeldahl nitrogen (TKN). More recently, Aboobakar et al. (2013) reported 0.036% of the total nitrogen (TN) load being released as N_2O in a full-scale nitrifying WWTP. On the other hand, long-term research performed by Daelman et al. (2013) delivered values as high as 2.3% of the incoming N being released as N_2O, representing three quarters of the carbon footprint of the WWTP under study. These last reports have contributed to put into perspective the high relevance of N_2O emissions from wastewater treatment facilities.

The high variability of green-house gas emissions reported in full-scale studies has spread the general idea of these emissions being strongly bounded to specific configurations and operating conditions applied (Law et al., 2012). To identify the most important operating conditions affecting the emission of these gases is the key to develop mitigation strategies to reduce fugitive N_2O and CH_4 emissions in WWTPs. This study presents the CH_4 and N_2O gas emission dynamics at different stages of an activated sludge plug–flow reactor of a municipal WWTP conducting biological nutrient removal. The main goal was to identify and evaluate the key operational conditions implemented in the plant that originated the majority of the uncontrolled CH_4 and N_2O emissions during the monitoring period.

2. Material and methods

2.1. Wastewater treatment process description

The monitoring site chosen in this study was the WWTP of the city of Granollers, near Barcelona (Spain). This plant treats the domestic wastewater of 112,000 population equivalents (P.E.). After primary treatment and settling, the wastewater is treated biologically in two parallel and identical plug–flow reactors where chemical oxygen demand (COD) and nitrogen removal is performed. Then, the wastewater flows into a secondary settler before being released into the environment. Excess sludge is anaerobically digested in order to produce biogas for electricity generation. A flow of reject wastewater (produced in the anaerobic digester sludge dewatering process) is regularly released at the inlet of the plant for its treatment. The configuration layout of Granollers WWTP is shown in Fig. 1.

2.2. CH_4 and N_2O monitoring and calculations

2.2.1. Gas emissions and emission factors

All the gas emission data recorded during this study was registered in aerated and non-aerated zones of one of the activated sludge lanes at Granollers WWTP. Wastewater is entering the first anoxic zone and it is transported across the reactor passing through an aerated zone, followed by a short anoxic zone (where mixed liquor is recirculated to the first anoxic zone) to finally end in another aerated zone before exiting the treatment lane. For the purposes of this study, the first anoxic zone is divided into two sites (Site 1 and Site 2) with the first one being more turbulent due to the flow of wastewater entering the bioreactor. The main aerated zone is also divided into 2 sites (Site 3 and Site 4) corresponding to the zones where two different air diffusers are present and independently controlled. The two last monitoring sites correspond to the second anoxic zone (from where mixed liquor is internally recirculated, Site 5) and the final aerated part of the bioreactor (Site 6). See Fig. 1 for locations of the monitoring sites.

CH_4 and N_2O emissions were monitored on-line for 48–72 h per week during ten weeks between June and October, 2013. Measurements of CH_4 and N_2O gases were performed using a commercial gas collection hood (AC'SCENT® Flux Hood) connected via gas tubing to a commercial gas analyser (VA-3000, Horiba, Japan) equipped with a sample conditioning system (series CSS, M&C Tech group). Off-gas was collected continuously (at 0.5 L/min) from the reactor headspace and concentration data was logged every 15 s. Oxygen concentration was also analysed using the same analyser for estimation of the Q_{gas}