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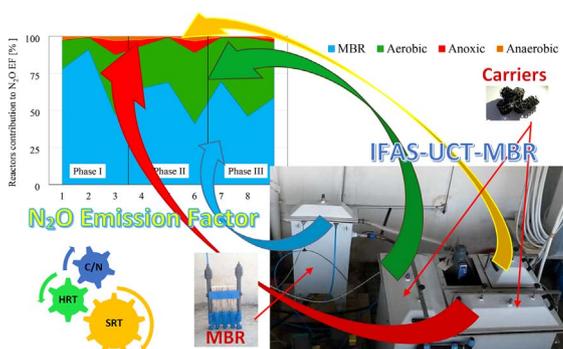
Short Communication

Nitrous oxide from integrated fixed-film activated sludge membrane bioreactor: Assessing the influence of operational variables

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GRAPHICAL ABSTRACT



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ABSTRACT

The influence of the main operational variables on N_2O emissions from an Integrated Fixed Film Activated Sludge University of Cape Town membrane Bioreactor pilot plant was studied. Nine operational cycles (total duration: 340 days) were investigated by varying the value of the mixed liquor sludge retention time (SRT) (Cycles 1–3), the feeding ratio between carbon and nitrogen (C/N) (Cycles 4–6) and simultaneously the hydraulic retention time (HRT) and the SRT (Cycles 7–9). Results show a huge variability of the N_2O concentration in liquid and off-gas samples (ranged from $10^{-1} \mu\text{g } N_2O\text{-N } L^{-1}$ to $10^3 \mu\text{g } N_2O\text{-N } L^{-1}$). The maximum N_2O concentration ($1228 \mu\text{g } N_2O\text{-N } L^{-1}$) in the off-gas samples occurred in the anoxic reactor at the lowest C/N value confirming that unbalanced C/N promotes the N_2O emission during denitrification. The aerated reactors (aerobic and MBR) have been the major N_2O emitters during all the three Phases.

1. Introduction

Wastewater treatment plants (WWTPs) can be a source of greenhouse gases (GHG) and, when designed for nitrogen removal, can emit a large amount of nitrous oxide (N_2O), representing 1.3% of the total anthropogenic N_2O emission (Kampschreur et al., 2009). N_2O has a relevant environmental interest as GHG (IPCC Working, 2013). Over the last decade, significant efforts were devoted to better understand

the key mechanisms involved in N_2O production from WWTPs and it is now widely recognised that N_2O can be produced during both nitrification and denitrification processes (Kampschreur et al., 2009; Law et al., 2012). It has also been suggested that ammonium oxidizing bacteria (AOBs) could be the main source of N_2O production in WWTPs (Wunderlin et al., 2012). Several studies have been performed on N_2O emissions from WWTPs focusing the attention on the identification of the key factors (operational conditions, influent features, etc.) mostly

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affecting the N_2O production (Kampschreur et al., 2009; Law et al., 2012; Wunderlin et al., 2012). With this regard, it has been demonstrated that the influent C/N ratio and sludge retention time (SRT) can influence significantly N_2O formation (Kampschreur et al., 2009; Li et al., 2013). Most of the existing studies on N_2O focus their attention on conventional activated sludge (CAS) systems adopting suspended biomass. In contrast, very poor information still exists on the N_2O emission from advanced treatment systems and biofilm systems (such as membrane bioreactors – MBR; moving bed biofilm reactors – MBBR; etc.). Advanced systems have specific features that make hard the direct transferability of the knowledge on N_2O emissions acquired for CAS systems. As an example, MBRs enhance specific biomass selection and intensive aeration for fouling mitigation that could promote N_2O production/stripping. However, only few studies on GHG investigations in MBR systems have yet been presented in literature to define mature the acquired knowledge (Mannina et al., 2016a; Mannina et al., 2016b; Mannina et al., 2017; Nuansawan et al., 2016) and most of the studies refer to non domestic wastewater (i.e., saline or leachate).

On the other hand, biofilm systems (MBBR or Integrated Fixed film Activated Sludge – IFAS) are characterized by high residence time which enhances the development of a nitrifying community avoiding the influence of seasonal temperature (Pal et al., 2012; Di Trapani et al., 2010, 2013; Martín-Pascual et al., 2015). Nevertheless, how the peculiar feature of IFAS (simultaneous growth of suspended and attached biomass), can influence the N_2O emission is still unknown. Indeed, as recently emphasized by Todt and Dörsch (2016) the knowledge on factors affecting N_2O emissions in biofilm systems is still limited if compared to CAS processes. Recent modelling studies on N_2O suggested that the emissions from nitrifying biofilms could be significantly higher than those from suspended growth systems under similar conditions (Sabba et al., 2015). Thus, given the spread of the MBBR and IFAS systems over the last years, there is a need of establishing a mature knowledge on N_2O emission in order to better control such systems (Schreiber et al., 2012).

The aim of the present study was to investigate the influence of different operational factors (namely SRT, carbon to nitrogen (C/N) ratio and hydraulic retention time – HRT) on the N_2O emissions from an IFAS-MBR pilot plant. The pilot plant had a University of Cape Town (UCT) configuration and was aimed at biological nutrient removal. During 340 days of operation, intensive gathering and measurement campaign have been performed collecting data on dissolved and gaseous N_2O . The wide data set adopted in this study can represent a solid support for enhancing the knowledge acquired so far on the N_2O emissions from biofilm systems.

2. Materials and methods

2.1. Pilot plant lay-out

A pilot-scale UCT-IFAS-MBR system was realized in accordance with the layout depicted in Fig. 1.

The pilot plant consisted in one anaerobic reactor (62 L with stirring device), one anoxic reactor (102 L with stirring device) and one aerobic reactor (211 L with fine bubble aeration device). The anoxic and aerobic reactors were filled with plastic suspended carriers (courtesy of Amitech s.r.l.) with 15 % and 40 % filling fraction, respectively, in order to yield a net surface area for biofilm growth of $75 \text{ m}^2 \text{ m}^{-3}$ and $205 \text{ m}^2 \text{ m}^{-3}$. The ultrafiltration membrane module (PURON® courtesy of Koch Membrane Systems, Inc.) was located inside a MBR reactor (36 L, continuously aerated). The recycle flow rate (Q_{RAS}) from the MBR to the anoxic compartment passed through an oxygen depletion reactor (ODR). Permeate was collected in a clean in place (CIP) tank; the permeate volume required for the membrane backwashing (1 min every 9 min) was withdrawn from the CIP and pumped back inside the membrane fibers. Anaerobic, anoxic, aerobic and MBR reactors were provided of funnel shaped covers in order to create a headspace for

sampling the off-gas.

2.2. Operational conditions

The experimental campaign lasted 340 days and was divided into three main phases (namely Phase I, Phase II and Phase III, respectively). During each phase, the influence of operational conditions and influent features on nitrous oxide production/emission has been investigated. More in detail, in Phase I the effect of the mixed liquor suspended solid (MLSS) SRT was investigated. In Phase II, the effect of the influent C/N ratio was analyzed. Finally, in Phase III the simultaneous effect of the HRT/SRT variation was assessed. Each experimental phase was divided into three different operation cycles (Cycles 1 – 9, respectively). Table 1 summarizes the operational parameters and influent wastewater features for each phase and cycle. The pilot plant was fed with a mixture of real and synthetic wastewater (sodium acetate and glycerol)

2.3. Analytical procedures

Samples from each reactor, influent and effluent were collected in order to analyze the main parameters necessary to investigate the biological performances of the pilot plant. Concerning nitrous oxide, samples from the headspace of each reactor, excepting the ODR, were collected 2 times per week throughout experiments. Samples from the bulk liquid of each reactor were collected with the same frequency and the dissolved nitrous oxide was extracted in accordance with the procedure proposed by Kimochi et al. (1998). Permeate samples were also collected in order to quantify the dissolved N_2O concentration discharged with the effluent flow rate. Both dissolved and headspace samples were stored in glass vials and then analyzed by means of Gas Chromatography using an Electron Capture Detector (ECD) for assessing the N_2O concentration. Furthermore, a hot wire anemometer allowed the air velocity measurement within the funnel of each reactor and thus the flux of nitrous oxide emitted from the liquid surface of each reactor was assessed. The nitrous oxide emission was assessed also in terms of Emission Factor (EF) evaluated in accordance with method proposed by Tsuneda et al. (2005). Moreover, the abundance of measured N_2O concentrations, dissolved and emitted, coupled with the detailed knowledge of the liquid flow rates passing through each reactor enabled to assess the nitrous oxide mass balance, thus highlighting the N_2O production or consumption within each reactor. For further information regarding gas sampling and extraction methods as well as the EF calculation and N_2O mass balance, the reader is addressed to literature (Mannina et al., 2016b; Mannina et al., 2017).

3. Results and discussion

For sake of completeness, a brief summary of the removal efficiency achieved during experiments is provided in Table 2.

From Table 2, it is worth noting that the total COD removal efficiency (measured in the permeate flow) was always higher than 90 %; in contrast, the biological COD removal efficiency (measured in the supernatant of MBR reactor, thus before membrane filtration) was affected by the imposed condition during experiments. Moreover, the nitrification efficiency suffered mainly during Phase II, due to the imposed C/N ratio. In addition, the reduction of the denitrification efficiency in Cycle 6 ($C/N = 2 \text{ mgCOD mg}^{-1}\text{TN}$) resulted in a low TN removal efficiency. The pattern of N_2O concentrations in the anaerobic and MBR reactors (both in liquid and gaseous samples) and in the permeate flux throughout experiments is reported in Fig. 2. By observing Fig. 2, it is possible to notice the huge variability of N_2O concentration, from $10^{-1} \mu\text{g N}_2\text{O-N L}^{-1}$ up to $10^3 \mu\text{g N}_2\text{O-N L}^{-1}$. The minimum dissolved concentration ($0.442 \mu\text{g N}_2\text{O-N L}^{-1}$) was measured in the aerobic reactor in Cycle 2 (SRT = 30 d), while the highest ($1415 \mu\text{g N}_2\text{O-N L}^{-1}$) was achieved in Cycle 7 (HRT = 30 h / SRT = 56 d). Concerning the N_2O concentration measured in the

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