



Influence of carbon to nitrogen ratio on nitrous oxide emission in an Integrated Fixed Film Activated Sludge Membrane BioReactor plant

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ABSTRACT

In this study a University of Cape Town (UCT) Integrated Fixed Film Activated Sludge (IFAS) Membrane BioReactor (MBR) wastewater treatment plant was monitored in terms of nitrous oxide (N₂O) emissions. The short term effect on the N₂O emission due to the influent carbon-to-nitrogen (C/N) ratio variation (C/N ratios of 2, 5 and 10 gCOD/gN) was evaluated. Since in a previous study, the effect of the C/N ratio was studied in the same system without biofilm (UCT-MBR configuration) the main aim here was to investigate the role of biofilms on N₂O emissions. Under all the investigated C/N ratios, the N₂O fluxes and the average emission factors were lower than that of previous studies with no biofilm presence. The total average N₂O emission was 0.5% of the influent nitrogen with biofilm (IFAS system) and 3.5% without biofilm. This result emphasizes the potential role of the biofilms in attenuating the N₂O emissions especially in the case of stress conditions (i.e., low C/N influent ratios). An increase of N₂O flux from the anoxic reactor (till 28 mgN₂O m⁻²h⁻¹) occurred at the lowest influent C/N tested (2 gCOD/gN - phase III). At C/N equal to 2 gCOD/gN the anoxic reactor was the main source of N₂O, contributing 45% of all produced N₂O. This result was attributed to an incomplete denitrification caused by a lack of organic carbon and a slight increase of dissolved oxygen concentration.

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1. Introduction

During the last ten years, the interest towards greenhouse gas (GHG) emission from wastewater treatment plants (WWTPs) has increased considerably with the aim to design and operate plants that have the minimum environmental impact (Mannina et al., 2016a). In addition to the “traditional pollutants” such as organics compounds (indirectly measured by chemical oxygen demand - COD, biochemical oxygen demand - BOD₅), nitrogen - N or phosphorus - P, WWTPs can also emit GHGs (i.e., carbon dioxide - CO₂, methane - CH₄ and nitrous oxide - N₂O). GHGs can be emitted as a direct consequence of the biological processes (Mannina et al., 2016a) or indirectly due to the power requirements (Papa et al., 2016). Among the GHG emitted from WWTPs, N₂O has caused the greatest interest among scientist and researchers due to its high

global warming potential (GWP) (298 times higher than that of CO₂) (IPCC et al., 2013). N₂O from WWTPs is mainly produced by the biological nitrogen removal (BNR) processes through nitrification and subsequent denitrification both from autotrophic and heterotrophic bacteria (Kampschreur et al., 2009).

Ammonia oxidizing bacteria (AOB) have been identified as responsible of producing N₂O due to: i. the reduction of NO₂ as terminal electron acceptor to N₂O (AOB denitrification) (Kim et al., 2010); ii. the incomplete oxidation of hydroxylamine (NH₂OH) to NO₂ (Law et al., 2012). The predominance of AOB pathway to the other has not yet been demonstrated (Pocquet et al., 2016).

WWTP operating factors (such as, dissolved oxygen, C/N ratio, sludge retention time - SRT and temperature) may strongly influence the N₂O emission (Kampschreur et al., 2009). Stenström et al. (2014) have found that decreasing the oxygen concentration during nitrification lead to the increase of N₂O emission during the denitrification; similar results were also found by Frison et al. (2015). Recently, Mannina et al. (2017a) reported the effect of the influent C/N ratio on N₂O emissions from the same UCT- MBR pilot plant

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List of abbreviations

AOB	ammonia oxidizing bacteria	NH ₄ Cl	ammonium chloride
ATU	Allylthiourea	NH ₄ -N	ammonium nitrogen
b _H	endogenous decay coefficient	NO ₂ -N	nitrite nitrogen
BNR	biological nutrient removal	NO ₃ -N	nitrate nitrogen
BOD	biochemical oxygen demand	NOB	Nitrite Oxidizing Bacteria
BOD ₅	biochemical oxygen demand in five days	NO _x -N _{in}	influent nitrite and nitrate concentration
C	carbon	NO _x -N _{out}	permeate nitrite and nitrate concentration
C ₃ H ₈ O ₃	glycerol	ODR	oxygen depletion reactor
CAS	conventional activated sludge	OHO	ordinary heterotrophic organism
CH ₃ COONa	sodium acetate	OP	ortho-P
CH ₄	methane	OUR	Oxygen Uptake Rate
CIP	clean in place	P	phosphorus
CO ₂	carbon dioxide	PAOs	phosphate accumulating organisms
COD	chemical oxygen demand	PHA	poly-β-hydroxyalkanoates
COD _{in}	influent COD	P _{IN}	influent orthophosphate concentration
COD _{out}	permeate COD	O ₄ -P	phosphate
COD _{SUP}	supernatant COD	P _{OUT}	permeate orthophosphate concentration
COD _{TOT}	total COD	Q _{airi}	volumetric gas flux
DO	dissolved oxygen	SBR	sequential batch reactors
DPAOs	denitrifying phosphate accumulating organisms	SNDPR	simultaneous nitrification, denitrification, and phosphorus removal
ECD	Electron capture detector	SRT	Sludge Retention Time
FA	free ammonia	TKN	Total Kjeldahl Nitrogen
GC	Gas Chromatograph	TN	total nitrogen
GHG	greenhouse gas	TP	total phosphorus
GWP	global warming potential	TSS	total suspended solid
HRT	hydraulic retention time	UCT	University Cape Town
IFAS	Integrated Fixed Film Activated Sludge	VSS	volatile suspended solids
K ₂ HPO ₄	dipotassium hydrogen phosphate	WWTPs	wastewater treatment plants
MBBR	moving bed biofilm bioreactors	η _P TOT	TP removal efficiency
MBR	Membrane BioReactor	η _{BIO}	biological COD removal efficiency
N	nitrogen	η _{denit}	denitrification efficiency
N ₂ O	nitrous oxide	η _{nit}	nitrification efficiency
N _{assimilation}	assimilated nitrogen	η _P	OP removal efficiency
NH ₂ OH	hydroxylamine	η _{TOT}	total COD removal efficiency
NH ₄ ⁺ -N _{in}	influent nitrogen ammonia concentration	η _N total	total nitrogen removal efficiency
NH ₄ ⁺ -N _{out}	permeate nitrogen ammonia concentration	μ _{H,max}	heterotrophic growth rate

here discussed and, they found the highest N₂O emissions at the lowest investigated C/N ratio (5 gCOD/gN).

The variability of the influent features and of the used process configuration make the weight that each operating factor has on the total N₂O emission unknown. Such a fact leads, as a consequence, to a huge variability of the N₂O emission factors (with respect to the influent nitrogen load) available in the literature: 0.01–1.8% (Ahn et al., 2010; Rodriguez-Caballero et al., 2015), 0.036% (Aboobakar et al., 2013), 0.04–11% (Daelman et al., 2015).

Further, the existing difficulties in capturing and measuring the real overall N₂O produced from WWTPs (Marques et al., 2016) make data more case study specific and difficult to transfer to other systems (Mannina et al., 2016a).

Despite the knowledge on the N₂O emission from WWTP has considerably increased, it has been mostly acquired on conventional activated sludge systems (CAS) (Todt and Dörsch, 2016). Therefore, current knowledge may not directly be transferred into innovative systems such as MBR or moving bed biofilm bioreactors (MBBR or hybrid biological systems such as Integrated Fixed Film Activated Sludge – IFAS) (Mannina et al., 2011). MBR systems as well as IFAS systems are characterized by specific peculiarities that would strongly influence the N₂O production/emission. For

example, the intensive aeration for fouling mitigation in MBR can promote the N₂O stripping. Therefore, further studies to better understand how the peculiarities of MBR or IFAS systems influence the N₂O emissions are required.

With this regard, only few studies have been reported in literature. Todt and Dörsch (2016) found that the AOB denitrification and the incomplete heterotrophic denitrification as the main sources of N₂O production in biofilm systems. These two N₂O formation pathways occur mainly in hypoxic or anaerobic zone of biofilm, where the low O₂ concentration and the presence of NO₂ favor the N₂O formation.

Lo et al. (2010) demonstrated that from the hybrid system the production of N₂O was significantly higher (21.2% of the influent nitrogen) than that of pure biofilm systems (0.5% of the influent nitrogen) and suspended biomass systems (4.2% of the influent nitrogen).

Very recently, Kinh et al. (2017) have demonstrated that membrane-aerated biofilm reactor (MABR) provides lower N₂O emission than conventional biofilm reactor (CBR). Specifically, N₂O emission factor was 0.0058 ± 0.0005% in the MABR and 0.72 ± 0.13% in the CBR.

Studies on the quantification of N₂O from biofilm systems, such

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