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Mitigating off-gas emissions in the biological nitrogen removal via nitrite process treating anaerobic effluents

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ABSTRACT

A sequencing batch reactor treating the reject water produced from the dewatering of the anaerobic co-digestate of waste activated sludge and the organic fraction of municipal solid waste was monitored in order to assess the off-gas emissions that were produced. The gaseous emissions of nitrous oxide (N₂O), carbon dioxide (CO₂), methane (CH₄), ammonia (NH₃) and methyl mercaptan (CH₃SH) produced during the biological nitrogen removal via nitrite route were monitored. During experimental period 1 a volumetric nitrogen loading rate (vNLR) of 1.08 kg N/m³ d was applied, while in period 2 the vNLR was 0.81 kg N/m³ d. Nitrous oxide emissions decreased from 1.49% of the influent nitrogen load in period 1 to 0.24% in period 2. The higher dissolved oxygen (DO) concentration and the lower accumulation of nitrite in the reactor resulted in significantly lower nitrous oxide emissions in period 2. Some methane (5.8% of influent COD load) was emitted during period 1, as the lower DO concentration created micro-anaerobic conditions within the sludge flocs.

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1. Introduction

Greenhouse gas (GHG) emissions are associated with a wide spectrum of anthropogenic activities, including mining (Norgate and Haque, 2010), manufacturing, biofuel production (Reijnders and Huijbregts, 2008), municipal solid waste management (Braschel and Posch, 2013) and wastewater (Cakir and Stenstrom, 2005) and manure treatment (Riano and Garcia-Gonzalez, in press). During the operation of a wastewater treatment plant (WWTP) nitrous oxide (N₂O), carbon dioxide (CO₂), methane (CH₄), nitric oxide (NO) and other gases can be emitted. Nitrous oxide is of particular environmental concern, since it has a global warming potential that is 298 times higher than that of CO₂. In terms of CO₂ equivalents (eq.), nitrous oxide contributes by 7.9% to the total anthropogenic GHG emissions. The global production of N₂O emissions from WWTPs treating human sewage in 1990 corresponded to approximately 3.2%

of the total estimated anthropogenic nitrous oxide emissions in the world (IPCC, 2001). The nitrous oxide emissions from wastewater management are estimated to contribute by 26% to the total greenhouse gas (GHG) emissions of the water chain. In order to estimate the nitrous oxide emissions from WWTPs, the policy makers refer to the guidelines developed from the Intergovernmental Panel on Climate Change (Kampschreur et al., 2009). The IPCC decreased the standard N₂O emission factor from 1% to 0.5% of the influent nitrogen load of the WWTP (IPCC, 2006). In countries having advanced, centralized WWTPs, a lower emission factor is applied by IPCC for direct emissions from WWTPs; this is 3.2 g N/capita year and corresponds to ~0.035% nitrous oxide emissions of the influent nitrogen load of a WWTP (Kampschreur et al., 2009). On the other hand, methane is mainly emitted from livestock farming activities (166 Mt CO₂-eq. in the EU per year), from landfills, composting units and WWTPs (95 Mt CO₂-eq. in the EU per year) (European Environment Agency, 2011). It is estimated that N₂O emissions from wastewater correspond to 100 Mt CO₂-eq. for 2010, while CH₄ emissions to 630 Mt CO₂-eq. (Monni et al., 2006). Emissions occur in the sludge treatment line and should be considered (Houillon and Joliet, 2005).

Biological nutrient removal (BNR) from wastewater is an effective approach for the prevention of eutrophication in water

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Abbreviations

A_{ch}	area of the emitting surface covered by the chamber (m^2)	NOB	nitrite oxidizing bacteria
AMO	ammonia monooxygenase	OFMSW	organic fraction municipal solid waste
AOB	ammonium oxidizing bacteria	OLR	organic loading rate ($kgCOD/m^3 d$)
A_{SBR}	surface area of the pilot SBR (m^2)	ORP	oxidation reduction potential
BNR	biological nutrient removal	PE	population equivalent
C_i	gas concentration at time t_i (mg/m^3)	PLC	programmable logic controller
C_0	gas concentration at time t_0 (mg/m^3)	SBR	sequencing batch reactor
COD	chemical oxygen demand (mg/L)	sCOD	soluble chemical oxygen demand (mg/L)
DO	dissolved oxygen (mg/L)	t_i	time at the end of the linear range of gas measurement (h)
E_M	amount of the emitted gas per cycle ($mg/cycle$)	t_0	time at the beginning of the linear range of gas measurement (h)
Eq.	equivalents	$t_{n+1} - t_n$	time interval between two consecutive measurements of the gas emissions within the SBR cycle (h)
E_R	emission rate of the gas ($mg/m^2 h$)	TKN	total Kjeldahl nitrogen (mgN/L)
FA	free ammonia (mgN/L)	TS	total solids (g/L)
FNA	free nitrous acid (mgN/L)	TVS	total volatile solids (g/L)
FL	fermentation liquid	TSS	total suspended solids (mg/L)
F/M	food to microorganism ratio ($kg COD/kg MLVSS d$)	V_{ch}	volume of the measuring chamber (m^3)
HRT	hydraulic retention time (d)	VFAs	volatile fatty acids
GHG	greenhouse gas	vNLR	volumetric nitrogen loading rate ($kgN/m^3 d$)
IPCC	Intergovernmental Panel on Climate Change	VSS	volatile suspended solids (mg/L)
MLSS	mixed liquor suspended solids (g/L)	WAS	waste activated sludge
MLVSS	mixed liquor volatile suspended solids (g/L)	WWTP	wastewater treatment plant
NH_2OH	Hydroxylamine	Y_H	biomass yield ($g VSS/g COD$)

recipients. However, many practical design and operating decisions in WWTPs (including BNR processes) have considerable impact on the overall environmental performance, in particular concerning the GHG emissions (Keller and Hartley, 2003). The vast majority of nitrous oxide emissions in WWTPs occur in the biological treatment processes. At the level of a BNR treatment plant, the N_2O impact can reach up to 83% of the operational CO_2 footprint (Desloover et al., 2011). The contribution of BNR to the total anthropogenic nitrous oxide (N_2O) emissions can be up to 10.2%, if the treatment of both manure and sewage is considered (Desloover et al., 2012). N_2O emissions during BNR occur during the biological processes of nitrification and denitrification (Fig. 1). During the nitrification process, N_2O can be formed via two routes: the first pathway is as a by-product of the incomplete oxidation of hydroxylamine (NH_2OH) to nitrite. Hydroxylamine is formed through the oxidation of ammonium by ammonium oxidizing bacteria (AOB) using the enzyme ammonia monooxygenase (AMO) to catalyze the reaction. NH_2OH is then oxidized to nitrite, a biochemical reaction which produces nitrous oxide. The second pathway of N_2O formation is attributed to the process known as nitrifier denitrification. In this biochemical process, nitrite is used as electron acceptor instead of oxygen; this can occur during nitrification under limiting dissolved oxygen (DO) conditions. The third pathway occurs during the anoxic operation of the reactor. In this process, NO and N_2O are produced as process intermediates of nitrate (NO_3^-) reduction to

gaseous nitrogen (N_2). This is the stage in which N_2O is also consumed as it is reduced to N_2 (Ni et al., 2011). N_2O can be produced during the anoxic reaction and subsequently be stripped to the gas phase in an aerated compartment. As N_2O has a relatively high solubility in water, stripping is not very fast (Kampschreur et al., 2009).

Desloover et al. (2012) performed an overview of the quantified N_2O emissions from full scale BNR plants that apply the conventional nitrification/denitrification and advanced nitrification/anoxic ammonium oxidation (anammox) and nitrification/denitrification processes. The authors concluded that nitrification is the bioprocess that mainly contributes to N_2O emissions. Full-scale measurements also point to nitrite as a factor in N_2O production (Ahn et al., 2010). Taking into account the much higher greenhouse gas impact of N_2O compared to CO_2 , it is necessary to determine whether nitrogen removal bioprocesses based on nitrite accumulation are systematically greater contributors of N_2O than conventional nitrification processes (IPCC, 2001; Ahn et al., 2011). N_2O emission rates can vary considerably due to the differences in the wastewater composition, the applied treatment process, the operating parameters and the environmental conditions. The most important parameters that affect the N_2O emissions include the DO concentration, the nitrite in the mixed liquor and the chemical oxygen demand to total Kjeldahl nitrogen (COD/TKN) ratio during denitrification (Kampschreur et al., 2009).

The novel technologies that are introduced should not only remove/recover nutrients at a low cost, but should also be characterized by a low carbon footprint. Nitrification/denitrification is increasingly being applied, particularly for the treatment of nitrogenous effluents, as it has lower aeration and organic carbon source requirements than conventional nitrification/denitrification. However, such processes can result in significant N_2O emissions (Kampschreur et al., 2008). Furthermore, the use of sequencing batch reactors (SBRs), particularly when combined with the treatment of highly nitrogenous effluents such as the reject water of anaerobic stabilization processes can enhance nitrous oxide emissions (Desloover et al., 2012). The implementation of

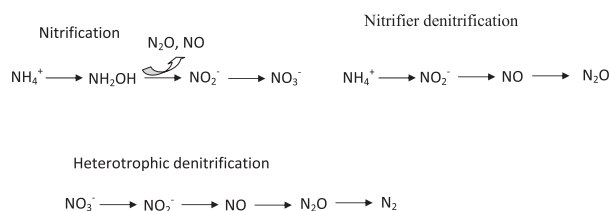


Fig. 1. Simplified representation of biochemical processes responsible for nitrous oxide production during nitrification and denitrification.

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