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Greenhouse gases from sequential batch membrane bioreactors: A pilot plant case study



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ABSTRACT

The paper reports the results of nitrous oxide (N₂O) emissions from aerobic and anoxic tank of a Sequential Batch Membrane Bioreactor (SB-MBR) pilot plant. The influence of salinity variation on N₂O emission was analyzed by gradually increasing the inlet salt concentration from 0 to 10 g NaCl L⁻¹.

The observed results showed that the N₂O concentration of the gaseous samples was strongly influenced by the salt concentration. This result was likely related to a worsening of the nitrification activity due to the effect of salinity on autotrophic bacteria. Dissolved oxygen concentration and salinity were found to be the key factors affecting N₂O concentration in the gaseous samples withdrawn from the anoxic tank. Despite the fact that the N₂O concentration in the anoxic tank was higher than in the aerobic one, it was found that the aerobic tank emitted around 25 times more N₂O than the anoxic one.

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1. Introduction

During the last years the main goal of wastewater treatment plants (WWTPs) has become broader than simply to meet effluent standards for receiving water body protection: many efforts were spent with the aim to include reduction/control of greenhouse gases (GHGs) emission [1,2]. Indeed, WWTPs can produce GHGs such as nitrous oxide (N₂O), carbon dioxide (CO₂) and methane (CH₄) [2]. GHG emissions from a WWTP can have a triple nature: direct, indirect internal and indirect external [3]. Direct emissions are mainly related to the biological processes (biomass respiration; biological nitrogen removal, etc); indirect internal emissions are associated with the electric consumptions and indirect external emissions are related to the other sources not directly controlled inside the WWTP (e.g., disposal of the excess sludge). Among the GHGs produced in a WWTP, N₂O plays a key role in terms of climate change. Indeed, the global warming potential (GWP) of N₂O is 298 times higher than that one of CO₂ based on a time horizon of 100 years [4,5]. It is therefore crucial to identify potential anthropogenic sources of N₂O in order to evaluate their relevance in the global N₂O budget. The IPCC reports [6] have established that N₂O emissions from WWTP account for approximately 3% of

the total anthropogenic sources. Furthermore, the global N₂O emissions from WWTP are expected to increase by approximately 13% between 2005 and 2020 [2]. Thus, it is imperative to better understand the core mechanisms connected with the N₂O production and emission in WWTPs and identify the main operating conditions affecting its formation.

During the last years, several efforts have been spent by the scientific community (*inter alia* Law et al. [2]; Kampschreur et al. [7]) with the aim to better understand processes and key operating factors promoting the N₂O emission in WWTPs.

The processes associated with biological nitrogen removal have been reported to be the key source of N₂O emission [2,7,8]. Indeed, N₂O can be produced during both nitrification (only by the ammonia oxidizing bacteria – AOB) and denitrification processes (during the NH₂OH pathways of nitrifier denitrification and/or the heterotrophic denitrification pathway). In detail, N₂O is an intermediate of the heterotrophic denitrification but it can also be produced during the ammonia oxidation process (nitrification) [9]. However, N₂O formation mechanisms have not been completely elucidated yet [2,10]. Contrasting opinions have been reported in the technical literature regarding the prevalent pathway in N₂O formation [2]. Furthermore, large variations in terms of measured N₂O emissions have been reported for different WWTPs ranging between 0.01% and 1.8%, and in some cases even higher than 10% referring to the N-loading rate [7,11–14]. These variations have been mainly ascribed to the different operational conditions (e.g.,

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dissolved oxygen (DO) concentration, C/N-ratio, pH level, ammonium (NH_4^+) loading rate, and NO_2^- accumulation) applied to the plants and also in different zones of the same plant [15,16]. Therefore, the fixed emission factors, applied to estimate N_2O emissions from WWTPs, as suggested by the IPCC [6,17], may drive to erroneous quantifications. Indeed, fixed emission factors do not take into account the interrelationship between the process configurations and operating conditions. Therefore, the understanding of the key operating conditions or plant configurations affecting or promoting the N_2O emissions is imperative for the reduction of N_2O emissions.

Kampschreur et al. [7] identified the main operational conditions that promote the N_2O production during the nitrogen removal process (e.g. low DO or nitrite accumulation due to the presence of toxic compound in the aerobic reactor or the low ratio C/N required for the denitrification). Zhao et al. [8] found that high salinity or salinity variations could also promote N_2O production during nitrification. Several authors have previously studied the role of salinity on N_2O emissions [8,18–20].

In particular, Tsuneda and co-workers [18] found that an increase in salt concentration strongly influenced the N_2O emission in a conventional pre-denitrification plant due to direct inhibition of N_2O acceptor oxidoreductase activity. Furthermore, Tsuneda et al. [18] found that the high concentration of DO, transported from the oxic tank to the anoxic one through the recycling stream, promotes the N_2O formation in the anoxic tank due to the high salinity.

Shang et al. [19] investigated the effect of salt on the nitrification process comparing two plants treating ordinary municipal wastewater and saline wastewater and found that high N_2O production was related to the saline wastewater treatment and that a rapid salinity increase led to the greatest N_2O production. Therefore, they suggested to avoid huge fluctuation of salinity.

Zhao et al. [20] investigated the effect of salinity stress under various COD/N ratios on the N_2O production during the denitrification process in the presence of different terminal electron acceptors. Zhao and co-workers [20] found that the salinity was the factor that most influenced N_2O production, whilst the COD/N ratio was the sub-important one. Specifically, when the salinity was high (20 g NaCl L^{-1}), N_2O accumulated at various COD/N ratios and in the presence of different terminal electron acceptors.

Very recently, Zhao et al. [8] have investigated the salinity effect on N_2O production pathway during nitrification. They found that saline shocks led to nitrite accumulation, thus enhancing the N_2O production that can be reduced by pre-acclimatizing the activated sludge to the saline wastewater.

The literature review reported above, shows that there is a worldwide high interest in the N_2O emissions from WWTPs, as well as all factors that affect the N_2O production.

In this study, a sequencing batch reactor (SBR) was used. It is reason to suspect that the changing operating conditions of a SBR may make this type of WWTP prone to high N_2O emissions. Indeed, transient conditions in terms of DO, typical of the SBRs, should imply the increase of N_2O emission [16].

Rodriguez-Caballero et al. [21] have recently demonstrated that SBRs operated with long aerated phases provide the largest N_2O emissions. Stenström et al. [22] studied N_2O production under various C/N-ratio and DO in full-scale SBR treating digester supernatant founding that reduced DO concentrations during nitrification ($<1.0\text{--}1.5 \text{ mg L}^{-1}$) enhanced N_2O formation. Furthermore, Stenström et al. [22] pointed out that the N_2O formed in the water phase during denitrification accumulates in the water volume until aeration starts and thereafter it is quickly stripped off. Rapid changes in operating conditions, for instance lowering the DO set point from 2.0 to 1.9 mg L^{-1} , resulted in an increase in N_2O emitted in the off-gas during nitrification by 65.6%.

To our knowledge, there has not been any study reporting the role of salinity in N_2O emissions from sequencing batch membrane bioreactors (SB-MBRs) despite their worldwide application for the treatment of wastewater. Therefore, the novelty of the present study consists in the investigation of N_2O emissions from a SBR pilot plant equipped with a membrane module (MBR) for the solid/liquid separation. The SB-MBR pilot plant was fed with domestic wastewater and was subjected to a gradual salinity increase (addition of NaCl to yield concentrations from 0 to 10 g NaCl L^{-1}), carried out at moderate steps (2 g NaCl L^{-1}). The main aim of the study was to gain insight about the short term effect of this gradual salinity increase on N_2O emission both from oxic and anoxic tanks. The present study is part of a wider research project focused on the use of a non-specialized bacterial consortium for the treatment of saline wastewater contaminated by hydrocarbons [32,33]. The present paper reports the results of a part of the study aimed at analyzing the effect of salinity up to 10 g NaCl L^{-1} , that was recognized to represent a sort of threshold value, beyond which a significant impact on bacteria might occur [27].

2. Materials and methods

2.1. SB-MBR pilot plant

The SB-MBR pilot plant consisted of two reactors in-series, one anoxic (volume 45 L) and one aerobic (volume 224 L), according to a pre-denitrification scheme (Fig. 1). An ultrafiltration hollow fiber membrane module (Zenon Zeeweed, ZW10) was installed into a separate aerated compartment (volume 50 L) while an oxygen depletion reactor (ODR) was placed in the recycling line in order to ensure anoxic conditions inside the anoxic reactor despite the intensive aeration in the aerobic tank. The aerobic, anoxic and MBR reactors were equipped with specific covers that guaranteed the gas accumulation in the headspace.

The SB-MBR pilot plant was fed discontinuously with real domestic wastewater (stored in a feeding tank of 320 L volume) according to fill-draw-batch operation approach. More in detail, 40 L of wastewater (V_{IN}) (previously mixed inside the mixing tank with salt, in order to meet the design salinity concentration) were cyclically fed in, whereas the permeate was extracted at 20 L h^{-1} (Q_{OUT}). Each cycle had the duration of 3 h that were split into 1 h of biological reaction and 2 h of MBR filtration. During the biological reaction time the permeate extraction pump was turned out, thus Q_{OUT} was equal to zero. During the cycle, 80 L h^{-1} (Q_{R1}) were continuously pumped from the aerobic to the MBR tank. Furthermore, a recycling activate sludge stream (Q_{RAS}), equal to 80 L h^{-1} during the reaction period and to 60 L h^{-1} ($Q_{\text{R1}} - Q_{\text{OUT}}$) during the filtration phase, was recycled from the MBR to the anoxic tank via the ODR tank. The SB-MBR pilot plant was operated for 3 months without sludge withdrawals (indefinite sludge retention time – SRT). The main wastewater characteristics as well as pilot plant operational parameters are summarized in Table 1. The experimental campaign was divided into six phases each characterized by a specific salt concentration from 0 up to 10 g NaCl L^{-1} . The NaCl concentration in the influent was increased at step of 2 g NaCl L^{-1} on a weekly basis. The Phase VI had a duration of 26 days.

2.2. Gas sampling

During pilot plant operation, both liquid and gaseous samples were withdrawn from the aerobic and anoxic tanks and analyzed to evaluate the N_2O concentration. Furthermore, in order to quantify the N_2O flux emitted from both the aerobic and anoxic compartment, the gas flow rate (Q_{gas}) was indirectly measured by using a hot wire anemometer.

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