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Simultaneous biological nitrous oxide abatement and wastewater treatment in a denitrifying off-gas bioscrubber

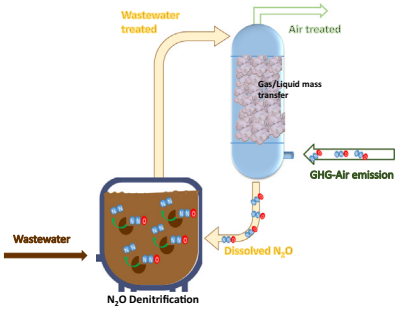
Osvaldo D. Frutos [a,b,1], Guillermo Quijano [a,1], Rebeca Pérez [a,1], Raúl Muñoz [a,*]

[a] Department of Chemical Engineering and Environmental Technology, University of Valladolid, Dr. Mergelina, s/n, Valladolid, Spain
[b] Facultad de Ciencias Agrarias, Universidad Nacional de Asunción, Campus San Lorenzo, Paraguay

HIGHLIGHTS

- The potential of an innovative anoxic bioscrubber for N_2O abatement was evaluated.
- The simultaneous N_2O abatement and wastewater treatment was feasible.
- Higher N_2O removals supported by increasing liquid recycling velocities and EBRTs.
- N_2O removal efficiencies of 92% were achieved at an EBRT of 40 min.
- Efficient organic carbon removals (85–95%) from wastewater were recorded.

GRAPHICAL ABSTRACT



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ABSTRACT

The simultaneous treatment of N_2O-laden air emissions and domestic wastewater was assessed in a novel denitrifying bioscrubber composed of a packed bed absorption column interconnected to a fixed bed reactor (FBR). The influence of liquid recycling velocities (U_L) and gas empty bed residence times (EBRTs) in the absorption column on bioscrubber's performance was evaluated using synthetic wastewater (SW) and a 100 ± 8 ppm_v N_2O air emission. Steady state N_2O removal efficiencies of 36 ± 3% concomitant with SW total organic carbon removals of 91 ± 1% were achieved at an EBRT of 3 min and at the highest U_L tested (8 m h^{-1}). The removal of dissolved N_2O by heterotrophic denitrification in the FBR constituted the main N_2O biodegradation mechanism and limited the abatement of N_2O. While the supplementation of SW with Cu^{2+} (a cofactor of the N_2O reductase) did not result in an enhancement in N_2O reduction, the increase in FBR volume supported a higher N_2O removal. The increase in EBRT up to 40 min supported an enhancement in the gas N_2O removal of up to 92%. The DGGE-sequencing analysis of FBR microbial population revealed a high microbial diversity and the abundance of denitrifying bacteria capable of reducing N_2O to N_2.

1. Introduction

Nitrous oxide (N_2O) is one of the major greenhouse gases (GHG) emitted nowadays, which contributes to climate change with a 6.2% of the total GHG emissions due to its high global warming potential (≈300 times higher than that of CO_2) [1]. N_2O is also considered the most important O_3-depleting substance emitted in this XXI century [2]. In Europe, N_2O is mainly emitted from agriculture (268300 Gg of CO_2 eq), wastewater treatment processes (12299 Gg of CO_2 eq) and adipic and nitric acid production (9682 Gg of CO_2 eq) [3]. In wastewater treatment plants (WWTPs), N_2O is mainly produced during biological nitrogen removal, with nitrifier denitrification,

* Corresponding author. Tel.: +34 983186424; fax: +34 983423013.
E-mail address: mutora@iq.uva.es (R. Muñoz).
[1] Tel.: +34 983186424; fax: +34 983423013.

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heterotrophic denitrification and hydroxylamine oxidation as the main routes of N_2O production in activated sludge processes [4]. Some authors have also reported N_2O emissions during wastewater biofiltration [5,6], where N_2O production was mainly associated to nitrification and denitrification processes. Even new microbial nitrogen removal processes such as nitritation/anammox or SHARON emit significant amounts of N_2O [7,8].

Based on the renovated and more ambitious EU objective for the reduction of the European GHG emissions by 40% in 2030 (compared to 1990 levels) [9], the minimization of N_2O emissions from wastewater treatment has become one of the main challenges of WWTP operators in this XXI century. In this regard, physical/chemical technologies such as thermal decomposition, selective catalytic reduction and selective non-catalytic reduction, typically used for industrial NOx emission abatement, could be applied as end-of-the-pipe technologies in WWTPs. However, these technologies entail the consumption of costly and/or hazardous chemicals, process operation at high temperatures and the generation of secondary pollution, which results in high operating costs and environmental impacts [10]. On the other hand, biotechnologies have been consistently shown as an environmentally friendly and low cost alternative for off-gas treatment, which exhibit a robustness and efficiency comparable to that of their physical/chemical counterparts [11]. Unfortunately, despite some works on NO/NO_2 nitrification and denitrification have been carried out [12,13], the number of studies assessing the potential of biotechnologies for N_2O abatement is scarce. This GHG is an obligate intermediate during the anoxic nitrogen reduction ($NO_3^- \rightarrow NO_2^- \rightarrow NO \rightarrow N_2O \rightarrow N_2$), which up to date has been reported as the only biological N_2O removal mechanism. Therefore, the removal of N_2O from air emissions entails the need for bioreactor configurations involving a N_2O absorption step in water followed by a N_2O reduction step under anaerobic conditions. Bioscrubbers are two-stage systems that can support the above mentioned functionalities simultaneously: the contaminant (N_2O) is transferred from the polluted air emission to a liquid phase flowing counter currently in a packed column (absorption step). The absorption column is interconnected to a stirred tank reactor where the N_2O transferred to the liquid phase in the absorption step is biologically reduced to N_2 under anoxic conditions (biotransformation step). However, the maintenance of anaerobic conditions in the denitrification tank requires the external supply of a biodegradable carbon source (e.g. methanol) to biologically deplete all O_2 present in the N_2O-laden aqueous stream, with the subsequent increase in process operating costs [14]. Therefore, innovative operational strategies based on the use of free carbon sources such as wastewater in WWTPs must be developed in order to achieve cost-effective N_2O removal processes.

The aim of this work was to evaluate the feasibility of the simultaneous N_2O abatement and wastewater treatment in a lab-scale bioscrubber as a model technology for an integrated wastewater treatment. The influence of liquid recycling velocities and gas empty bed residence times on the removal of N_2O and wastewater treatment performance was also investigated.

2. Materials and methods

2.1. Chemicals and synthetic wastewater

A 40 L calibration gas mixture of 10,000 ppm_v of N_2O in N_2 was purchased from Abelló Linde S.A. (Barcelona, Spain). A modified synthetic wastewater (SW) from Bajaj, et al. [15] was used as a model urban wastewater with the following composition (in g L^{-1} of tap water): peptone 0.16, meat extract 0.11, urea 0.03, NaCl 0.007, $CaCl_2{\cdot}2H_2O$ 0.004, $MgSO_4{\cdot}7H_2O$ 0.002, K_2HPO_4 0.028, $CuCl_2{\cdot}2H_2O$ 50×10^{-6} and glucose 0.25. The final concentrations of total organic carbon (TOC), total nitrogen (TN) and PO_4^{3-} of the SW were 256.1 ± 22.7, 54.4 ± 2.9 and 11.7 ± 3.3 mg L^{-1}, respectively. All reagents were purchased from PANREAC with a purity of +99% (Barcelona, Spain). The biodegradability of the SW was experimentally determined in independent batch assays by monitoring the TOC and TN concentrations for 14 days in three 500 mL Erlenmeyer initially filled with 99 mL of sterilized SW and 1 mL of activated sludge from Valladolid WWTP (Spain). Two non-inoculated sterilized Erlenmeyer with 100 mL of SW were used as controls to elucidate any potential carbon or nitrogen abiotic removal.

2.2. Experimental set up

A lab-scale bioscrubber was set up for the continuous abatement of a diluted air emission of N_2O and the simultaneous treatment of SW for 140 days. The experimental system was composed of a N_2O absorption column made of PVC (8.3 cm of inner diameter, 53 cm height) and packed with 2 L of Kaldnes rings (High Density Polyethylene rings of 50% porosity, diameter = 0.9 cm, Evolution Aqua, United Kingdom) interconnected with a 3 L fixed bed bioreactor (FBR) (Afora S.A., Spain). The FBR was filled with 1 L of methylotrophs-containing polyurethane foam (PUF) cubes (1 cm^3) used in a previous experiment as the packed bed of an absorption column [14]. The FBR was constructed with a 0.55 L liquid distribution chamber located at the bottom of the tank and operated with magnetic stirring at 300 rpm (Fig. 1). The experimental set-up was located in an air-conditioned room at 25 °C. Prior to inoculation, an abiotic test was performed with tap water for 4 days in order to assess any potential removal of N_2O by adsorption or photodegradation in the experimental set-up.

2.3. Bioscrubber operation

The SW was introduced at the bottom of the FBR, where it mixed with the N_2O-laden recycling liquid from the absorption column, and was further recirculated from the top of the FBR to the top of the packed bed absorption column using a peristaltic pump (Watson Marlow, UK). The N_2O-laden air emission was introduced at the bottom of the absorption column flowing upwards counter currently with the recycling liquid. The synthetic N_2O-laden air inflow was obtained by mixing 660 mL min^{-1} of air and 6.7 mL min^{-1} of the 10,000 ppm_v N_2O calibration gas mixture using a mass flow controller (Aalborg, Denmark), resulting in a gas empty bed residence time (EBRT) in the absorption column of 3 min and a mean N_2O concentration of 100 ± 8 ppm_v, which correspond to typical off-gas emissions from WWTPs. The SW was supplied to the FBR at flow rates determined by the maintenance of anoxic conditions (targeting a dissolved oxygen concentration = 0 mg L^{-1}) in the FBR. No N_2O (0.05 ± 0.06 ppm_v corresponding to the atmospheric N_2O concentration) was supplied to the inlet air for the first 18 days of operation (stage I) in order to assess any potential N_2O generation in the system as a result of wastewater treatment. During stage I, the bioscrubber was operated with a SW flow rate of 3 ± 0.1 L d^{-1} and a liquid recycling velocity (U_L) of 1 m h^{-1}. Stage II (days 19–51) was characterized by process operation at a N_2O of 100 ± 7 ppm_v, U_L of 1 m h^{-1} and a SW flow rate of 4 ± 1 L d^{-1}. U_L was increased up to 4 m h^{-1} during stage III (days 52–83) concomitantly with an increase in SW flow rate to 19 ± 1 L d^{-1}, while maintaining the inlet N_2O concentration at 104 ± 11 ppm_v. The bioscrubber was operated from day 84 to 104 (stage IV) with a U_L of 8 m h^{-1}, a SW flow rate of 36 ± 4 L d^{-1} and at 95 ± 5 ppm_v of N_2O. Similar SW flow rates and U_L were maintained during stage V (days 105–118) while maintaining the N_2O concentration at 100 ± 5 ppm_v, which was