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Effect of process parameters and operational mode on nitrous oxide emissions from a nitritation reactor treating reject wastewater

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ABSTRACT

Nitrous oxide (N_2O) and methane emissions were monitored in a continuous granular airlift nitritation reactor from ammonium-rich wastewater (reject wastewater). N_2O emissions were found to be dependent on dissolved oxygen (DO) concentration in the range of 1–4.5 mg O_2/L, increasing within this range when reducing the DO values. At higher DO concentrations, N_2O emissions remained constant at 2.2% of the N oxidized to nitrite, suggesting two different mechanisms behind N_2O production, one dependent and one independent of DO concentration. Changes on ammonium, nitrite, free ammonia and free nitrous acid concentrations did not have an effect on N_2O emissions within the concentration range tested. When operating the reactor in a sequencing batch mode under high DO concentration (>5 mg O_2/L), N_2O emissions increased one order of magnitude reaching values of 19.3 ± 7.5% of the N oxidized. Moreover, CH_4 emissions detected were due to the stripping of the soluble CH_4 that remained dissolved in the reject wastewater after anaerobic digestion. Finally, an economical and carbon footprint assessment of a theoretical scaled up of the pilot plant was conducted.

1. Introduction

Specific treatments for high ammonium (NH_4^+) streams such as reject wastewater produced in the anaerobic digester sludge dewatering process have been implemented in many wastewater treatment plants (WWTPs). Anaerobic digestion reject water is characterized by its high NH_4^+ content (500–1500 mg N/L) and its treatment is normally done via partial nitrification followed by denitrification (Hellinga et al., 1998; Mulder et al., 2001) or the combination of partial nitrification with anammox (Van Dongen et al., 2001) to reduce the operational costs. In the last few years, several studies have reported uncontrolled direct nitrous oxide (N_2O) emissions during this treatment, especially in the nitritation reactor, where conversion of NH_4^+ to nitrite (NO_2^-) occurs due to the action of ammonia oxidizing bacteria (AOB) (Kampschreur et al., 2008a; De Graaff

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Table 1 – Experimental tests conducted under continuous operation at different dissolved oxygen concentrations.

	T1	T2	T3	T4	T5	T6	T7	T8	T9	T10	T11	T12
DO (mg O_2/L)	1.1	1.2	1.5	1.6	1.6	1.7	2.0	2.4	2.5	2.6	3.2	3.2
Air flow (L/min)	13	11	14	37	11	11	13	50	11	24	13	18
pH	7.5	7.6	7.7	7.7	7.6	8.1	7.6	7.5	7.6	7.6	7.6	7.7
	T13	T14	T15	T16	T17	T18	T19	T20	T21	T22	T23	T24
DO (mg O_2/L)	3.2	3.2	3.3	3.3	4.1	4.4	4.6	4.7	4.8	5.3	6.7	7.5
Air flow (L/min)	16	15	100	27	50	100	50	50	65	100	100	50
pH	8.1	7.6	8	7.7	8.4	7.7	7.6	7.5	7.6	8.0	7.5	7.2

et al., 2010; Desloover et al., 2011; Law et al., 2012). In full-scale partial nitritation installations N_2O values ranging from 1.7 to 6.6% of the nitrogen load have been measured, which correspond to 3.4–11.2% of the NH_4^+–N oxidized emitted as N_2O since these systems operate with partial conversion of NH_4^+ to NO_2^- (Kampschreur et al., 2008a; Desloover et al., 2011). N_2O has a warming potential 265 times higher than that of CO_2 (IPCC, 2013) and in some cases might be responsible of the majority of the carbon footprint of the plant, especially in those systems with high conversion of NH_4^+ to NO_2^-.

AOB are known to be net producers of N_2O which originates via two possible pathways: a) oxidation of hydroxylamine (NH_2OH) which could be regulated by the concentration of free ammonia (Stein, 2011); b) the reduction of NO_2^- to N_2O in a process known as nitrifier denitrification (Bock et al., 1995). Low dissolved oxygen (DO) concentration levels, high NO_2^- concentrations and variation in influent NH_4^+ concentrations have been identified to promote N_2O formation (Kampschreur et al, 2009a). To this end, effective process control specifically devoted to keep desired set-points for the key parameters of operation in nitritation reactors (i.e. DO concentration, NH_4^+ concentration, pH) should be targeted.

The aim of this manuscript was to identify the DO, NH_4^+ and free ammonia concentration thresholds that originated the lowest N_2O emissions in a nitritation reactor. The novel control strategy applied in this reactor and described in Bartrolí et al. (2010) allowed the flexibility of operating at a desired DO set-point without compromising the effectiveness of the system. The control strategy also allowed to operate at full (100% conversion of NH_4^+ to NO_2^-) or partial nitritation (50% conversion of NH_4^+ to NO_2^-) depending on the subsequent denitritation step: either heterotrophic or autotrophic (anammox), respectively. A comparison in terms of treatment performance and N_2O emissions between continuous and discontinuous operation mode for the same reactor is also presented and highlights the importance of considering greenhouse gas emissions when implementing a technology. Finally, an economic and carbon footprint analysis of applying a N_2O mitigation strategy was conducted for the pilot plant and extrapolated to real facilities.

2. Materials and methods

2.1. Pilot plant

2.1.1. Continuous operation

The pilot plant consisted in a 150 L granular airlift reactor with a height to diameter ratio of 8.4. It was located in a municipal WWTP in Catalonia, Spain, and it was performing full partial nitrification of reject wastewater produced in situ during the dewatering process of the anaerobic digester sludge from the WWTP. The temperature of the reactor was kept at 30 °C by using an electric heating system and a temperature controller. The pH was maintained at 7.5 ± 0.2 through the addition of solid Na_2CO_3. DO concentration was monitored with an online DO probe (LDO luminescence sensor, Hach-Lange, Düsseldorf, Germany) and was maintained around the desired set-point (see Table 1) by changing the aeration flow-rate (from 11 to 100 L/min). The total ammonium nitrogen (TAN = NH_4^+–N/L + NH_3–N/L) concentration in the bulk liquid was monitored with an online probe (NH4D sc probe with a Cartrical cartridge, Hach Lange, Düsseldorf, Germany).

The reactor was operated with a variation of the control strategy presented in Bartrolí et al. (2010) during the period of monitoring. The variable measured for the control loop was the TAN concentration whereas the manipulated variable was the wastewater inflow rate fed to the reactor. The feedback control loop maintaining the TAN concentration in the bulk liquid allows for a maximization of the treatment capacity at any time, because the loading rate is as high as possible during the continuous operation of the reactor. The influent wastewater was added in an on/off mode controlled by the concentration of NH_4^+ present in the bulk liquid. When the NH_4^+ concentration was lower than the set-point (40 mg NH_4^+–N/L), the feeding pump was activated, until the NH_4^+ concentration was again at the set-point value. With this strategy, NH_4^+ concentration was always kept between the desired set-point ±5 mg N/L and NO_2^- concentration depended on the concentration of NH_4^+ in the influent, but always with an NH_4^+ to NO_2^- conversion higher than 92%, except for the period where partial nitritation was applied (see Fig. 1). Nitrate (NO_3^-) was hardly detected in the reactor at all times, presenting concentrations around 1–2 mg N/L in the bulk liquid (see Fig. 1). The system was controlled and monitored through a SCADA (supervisory control and data acquisition) program. For the control system, the manipulated variable was the inflow rate of the reject water. DO concentration was manipulated by changing the air flow-rate through the opening of the pneumatic valve that could be regulated continuously (i.e. via a frequency modulated solenoid valve). The air flow-rate was kept constant during each monitoring period to decrease the impact of total aeration flow-rate on the estimation of N_2O and CH_4 emissions. The DO varied during a monitoring period within a very small range (i.e. ± 0.1 of the desired set-point).

At the time of the study, the reactor had been working for more than 100 days under stable operation (see Fig. 1) and had