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Reduction of nitrous oxide emissions from biological nutrient removal processes by thermal decomposition

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ABSTRACT

During the last decade municipal wastewater treatment plants have been regulated with increasingly stringent nutrient removal requirements including nitrogen. Typically biological treatment processes are employed to meet these limits. Although the nitrogen in the wastewater stream is reduced, certain steps in the biological processes allow for the release of gaseous nitrous oxide (N_2O), a greenhouse gas (GHG). A comprehensive study was conducted to investigate the potential to mitigate N_2O emissions from biological nutrient removal (BNR) processes by means of thermal decomposition. The study examined using the off gases from the biological process, instead of ambient air, as the oxidant gas for the combustion of biomethane. A detailed analysis was done to examine the concentration of N_2O and 58 other gases that exited the combustion process. The analysis was based on the assumption that the exhaust gases were in chemical equilibrium since the residence time in the combustor is sufficiently longer than the chemical characteristics. For all inlet N_2O concentrations the outlet concentrations were close to zero. Additionally, the emission of hydrogen sulfide (H_2S) and ten commonly occurring volatile organic compounds (VOCs) were also examined as a means of odor control for biological secondary treatment processes or as potential emissions from an anaerobic reactor of a BNR process. The sulfur released from the H_2S formed sulfur dioxide (SO_2) and eight of the ten VOCs were destroyed.

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1. Introduction and historical background

Nitrous oxide (N_2O) is a naturally occurring greenhouse gas (GHG) that has a direct effect on global warming. In the United States, N_2O accounts for an estimated 5.2% of the total greenhouse gas emissions. Although a small percentage, it is important because N_2O is approximately 300 times more potent than carbon dioxide (CO_2) as a greenhouse gas (United States EPA, 2008). Approximately 8% of the total 5.2% N_2O emissions are from wastewater treatment facilities (EPA, 2008). In addition to its role in global warming, nitrous oxide is also involved in the destruction of ozone in the upper atmosphere. Nitrous oxide released into the troposphere enters the stratosphere by diffusion where it is either destroyed by photolysis or reacts with a single oxygen atom. The result of either yields nitric oxide which enters the ozone destruction cycle (Kramlich and Linak, 1994).

The emission of N_2O results from the biochemical transformation of different nitrogen species to the inert nitrogen gas (N_2). The production of nitrous oxide from biological nitrogen removal systems, by both nitrification and denitrification of wastewater, is well documented, (Ahn et al., 2010; Kampschreur et al., 2009; Schalk-Otte et al., 2000; Garrido et al., 1998; Czepiel et al., 1995; Von Schulthess et al., 1995, and Thörn and Sörensson, 1996). Despite the production of gaseous nitrous oxide emissions during biological treatment of wastewater, more stringent nitrogen effluent standards have been applied to wastewater treatment plants in recent years and the result has been an increase in the use of BNR systems. This trend is likely to continue; however, the concern over greenhouse gas emissions and global climate change has led to an interest in ways to minimize or eliminate them. The high volume, low N_2O concentration in the emissions has resulted in a number of attempts to reduce the emissions at the source through process control (Von Schulthess et al., 1994; Béline and Martinez, 2002) as well methods that employ gas bioscrubbers (Frutos et al., 2015).

The thermodynamics and kinetics regarding the thermal decomposition of nitrous oxide have been studied in detail

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(Ramsperger and Waddington, 1931; Friedman and Bigeleisen, 1953; Loirat et al., 1985). The primary steps in the decomposition of N_2O are:

$$N_2O + M \rightarrow N_2 + O + M \quad (1)$$

$$N_2O + O \rightarrow N_2 + O_2 \quad (2)$$

$$N_2O + O \rightarrow NO + NO \quad (3)$$

M −other species needed to extract energy from the endothermic reactions.

Reaction 1 occurs at a temperature of approximately 569 K (296 °C). The predominant reaction products of N_2O decomposition are generally N_2 and O_2. Thermodynamics dictates that reaction 3 will not be significant unless temperatures exceed approximately 1073 K (800 °C) (Lipkea et al., 1973).

Application of this phenomenon was first applied in World War II with the injection of nitrous oxide into internal combustion (IC) engines, as a means to increase power output in aircraft engines and later, in the 1950s, in high performance racing automobiles (Langfield, 2006). More recently, it has been utilized as a means of reducing industrial emissions. Thermal decomposition of nitrous oxide has been implemented by the nylon industry to reduce the unavoidable N_2O emissions from the manufacture of adipic acid (Reimer et al., 1994; Shimizu et al., 2000). However, that application involves high flow rate gas streams with high concentrations, while the emissions from the biological processes are high flow and low concentration gas streams.

2. Objectives

Given the ease of thermal decomposition within combustion processes, the first objective of this research is to demonstrate that the fugitive N_2O emissions of the biological processes could be thermally destroyed when introduced into the air intake of the facility's existing combustion processes. The net effect is therefore, a reduction in the quantity of harmful emissions. By using the emissions (off gases) from the biological reactors as the inlet oxidant (i.e. air) gas stream to an existing combustion process, the nitrous oxide is removed by thermal decomposition. A conceptual layout is depicted in Fig. 1. The biological process is outlined in blue and the combustion process, the focus of this study, is outlined in red. In this study, only the aerobic process as shown in Fig. 1 was considered; however, the emissions from both the anoxic and anaerobic reactors could combined with the emissions from the aerobic reactor and directed to the inlet of the combustion process. In practice the off gases from all of the biological processes would likely be collected in one system. Although a municipal treatment plant was considered for this study, the application is also relevant to an industrial plant as well.

The idea is to take the off gases from the biological process, which contains air, carbon dioxide and other emissions, and send them to the combustion process as the oxidant gas stream for combustion with biomethane. The temperature of 569 K (296 °C) at which thermal decomposition occurs is well below the 2000K combustion temperature of fossil fuels and biomethane. The oxygen from the N_2O is consumed as an oxidant and the inert nitrogen gas is released to the atmosphere.

2.1. Process to combustion process

The second objective is to determine the effect of using the gaseous emissions as an oxidant on the production or reduction of thermal NO_x. Since the combustion temperature is approximately 2000K, reaction 3 (above) indicates the formation of nitrogen oxides, primarily nitric oxide (NO) by the oxidation of atmospheric nitrogen and the nitrous oxide in the off gas. As the formation of these oxides is a function of flame temperature they are termed thermal NO_x (Turns, 1996; Warnatz et al., 2001). The oxidant, which consists of both air and N_2O, will have a greater amount of N_2 available, from the decomposition of N_2O than if the oxidant were only air. The replacement of 1 mol of air with 1 mol of nitrous oxide, in the oxidant gas stream, increases the amount of nitrogen by

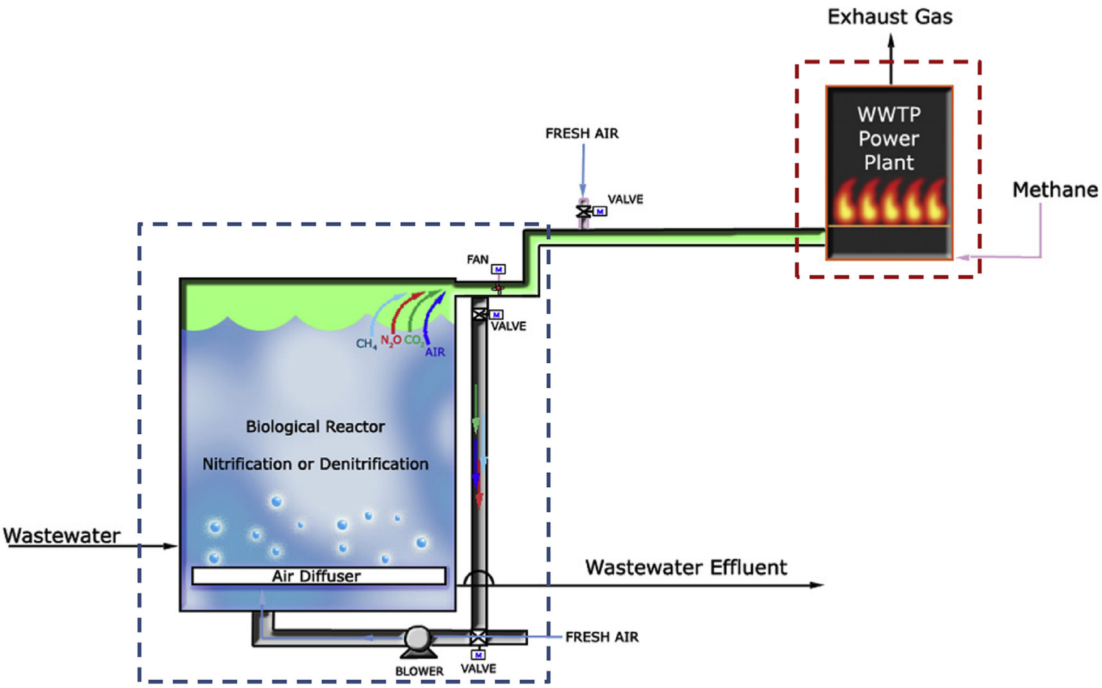

Fig. 1. Flow schematic of gaseous emissions from the aerobic reactor of a BNR.

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