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Effect of process design and operating parameters on aerobic methane oxidation in municipal WWTPs

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ABSTRACT

Methane is a potent greenhouse gas and its emission from municipal wastewater treatment plants (WWTPs) should be prevented. One way to do this is to promote the biological conversion of dissolved methane over stripping in aeration tanks. In this study, the well-established Activated Sludge Model n°1 (ASM1) and Benchmark Simulation Model n°1 (BSM1) were extended to study the influence of process design and operating parameters on biological methane oxidation. The aeration function used in BSM 1 was upgraded to more accurately describe gas–liquid transfer of oxygen and methane in aeration tanks equipped with subsurface aeration. Dissolved methane could be effectively removed in an aeration tank at an aeration rate that is in agreement with optimal effluent quality. Subsurface bubble aeration proved to be better than surface aeration, while a CSTR configuration was superior to plug flow conditions in avoiding methane emissions. The conversion of methane in the activated sludge tank benefits from higher methane concentrations in the WWTP's influent. Finally, if an activated sludge tank is aerated with methane containing off-gas, a limited amount of methane is absorbed and converted in the mixed liquor. This knowledge helps to stimulate the methane oxidizing capacity of activated sludge in order to abate methane emissions from wastewater treatment to the atmosphere.

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1. Introduction

Municipal wastewater treatment entails the emission of methane (CH_4), a potent greenhouse gas with a global warming potential of 34 CO_2-equivalents (IPCC, 2013). In a long-term study on a municipal wastewater treatment plant (WWTP) near Rotterdam, the Netherlands, methane was found to make up 13.5% of the plants greenhouse gas footprint, exceeding the carbon dioxide contribution related to the plant's electricity and natural gas consumption (Daelman et al., 2013). The share of methane in the climate footprint of a WWTP near Gothenburg, Sweden, was estimated at 31% (Tumlin, 2011). In the US, wastewater treatment is the seventh most important source of methane emission (EPA, 2013),

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while globally, wastewater treatment accounts for 4–5% of the total methane emission (El-Fadel and Massoud, 2001; Conrad, 2009).

It was recently discovered that about 80% of the dissolved methane entering an aerobic activated sludge tank was converted, with the remainder being stripped (Daelman et al., 2012). Dissolved methane in sewage or in reject water entering an aerobic activated sludge tank can be biologically converted by methanotrophic bacteria (Ho et al., 2013). These bacteria use methane as their sole source of carbon and energy (Hanson and Hanson, 1996). First, methane is oxidized to methanol (CH_3OH) by particulate or soluble methane monooxygenase. Next, methanol is further oxidized to formaldehyde (HCHO), formate (HCOOH) and carbon dioxide (CO_2) by methanol dehydrogenase, formaldehyde dehydrogenase and formate dehydrogenase, respectively (Hanson and Hanson, 1996).

Intermediate metabolites of the aerobic methane oxidation pathway (e.g. methanol) can serve as electron donors for denitrifying microorganism. Methane could therefore potentially be used for biological denitrification of wastewater (Harremoës and Henze, 1971; Modin et al., 2007). However, this requires a critical supply of oxygen to the organisms: enough for oxidizing methane but not too much in order to prevent inhibition of denitrification. In a wastewater treatment plant designed for nitrogen removal, the organisms are cycled between anoxic zones, where there is no oxygen to oxidize methane to substrates for denitrification, and aerobic zones, where the high oxygen concentrations (>1 g O_2 m^{-3}) inhibits denitrification, making aerobic methane oxidation coupled to denitrification not likely to happen in wastewater treatment plants. Denitrification with methane is also possible under anoxic conditions by *Methylomirabilis oxyfera* (Ettwig et al., 2010; Kampman et al., 2012), but since the microorganisms in activated sludge are frequently exposed to high oxygen concentrations and given the low growth rate of this organism, anoxic denitrification with methane is unlikely to happen in a wastewater treatment plant. For these reasons, the present study focuses on the complete oxidation of methane to carbon dioxide. Given the frequent exposure of the microorganisms in the activated sludge to aerated conditions and the presence of nitrate in the anoxic reactors, any methanogenic activity in activated sludge tanks is deemed to be insignificant (Gray et al., 2002).

Since the global warming potential of methane is 34 times that of carbon dioxide, the oxidation of methane to carbon dioxide is beneficial to mitigate climate change. Both lab-scale and full-scale studies have been performed to establish the potential of methanotrophs to curb the emission of methane from gaseous or liquid waste streams. Bio-filtration units were tested for the removal of methane from landfill gas (Nikiema et al., 2007), animal husbandry ventilation (Melse and van der Werf, 2005), manure storage ventilation (Girard et al., 2011) and coal mine ventilation (Sly et al., 1993). As far as liquid streams are concerned, digester effluent received some attention because it can be supersaturated with methane (Pauss et al., 1990; Hartley and Lant, 2006). To avoid that the dissolved methane is emitted to the atmosphere, Hatamoto et al. (2010) and Matsuura et al. (2010) developed a biofilm reactor, while van der Ha et al. (2011) explored a co-culture of methanotrophic bacteria and microalgae to degrade methane in the effluent of anaerobic wastewater treatment plants.

To describe and predict the behaviour and performance of methanotrophic bio-filter systems, a number of models have been developed. Delhomenie et al. (2008) and Yoon et al. (2009) developed models to describe the removal of methane from gas streams using methanotrophs in biofilms, while Oldenhuis et al. (1991), Broholm et al. (1992) and Alvarez-Cohen and McCarty (1991) carried out similar studies for suspended cells. Arcangeli and Arvin (1999) modelled the co-metabolic degradation of dissolved chlorinated aliphatic hydrocarbons in a biofilm by methanotrophic bacteria. To our knowledge, no models for methane degradation in activated sludge have been reported.

The present study investigates the fate of dissolved methane in an activated sludge plant. To this end, the Activated Sludge Model n°1 (ASM1, Henze et al. (1987)) was extended with aerobic methanotrophic growth. The resulting model, ASM1m, was implemented in the Benchmark Simulation Model n°1 (BSM1, Copp (2002)). This extended plant model, termed BSM1m, was used to simulate the effect of process design and process conditions on the fate of methane in an activated sludge plant. Taking into account biological methane oxidation on the one hand and stripping of methane on the other hand, BSM1m is the first model that describes biological methane conversion in activated sludge tanks. As such, it complements existing models for the emission of nitrous oxide in estimating greenhouse gas emissions from WWTPs.

2. Materials and methods

2.1. ASM1m model description

The ASM1m model developed in the present study adds two processes to ASM1: aerobic growth and decay of methanotrophs. The two additional state variables are methane as substrate (S_{CH_4}) and methane oxidizing bacteria (X_{MOB}). Since the interest of this study is in the fate of methane in activated sludge systems, methanotrophic bacteria are singled out from the other heterotrophic organisms (X_{BH}) and are therefore described by a separate state variable, X_{MOB}, as in Arcangeli and Arvin (1999). The reaction stoichiometry and kinetics related to the growth and decay of methanotrophic biomass are summarized in Table 1.

In ASM1m, growth of methanotrophs was modelled using Monod kinetics for methane and oxygen. Monod kinetics for methane were also used in Oldenhuis et al. (1991), Alvarez-Cohen and McCarty (1991), Broholm et al. (1992), Arcangeli and Arvin (1999), Oldenhuis et al. (1991) and Yoon et al. (2009). Unlike in Yoon et al. (2009), oxygen was also considered as a limiting substrate.

Ammonia inhibition, as considered by Arcangeli and Arvin (1999), was not included in the model. The effect of the ammonium concentration on the methane oxidation rate by methanotrophs is ambiguous. A number of studies reported an inhibitory effect of ammonium (Hanson and Hanson, 1996; Begonja and Hrsak, 2001; Nyerges and Stein, 2009), others reported no effect (van der Ha et al., 2010; van der Ha et al., 2011)

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