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Feast-famine biofilter operation for methane mitigation

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ABSTRACT

Packed bed clogging and channeling derived from the accumulation of biomass still represent technical challenges to be addressed in gas biofiltration in order to enable a more cost-effective performance under long-term operation. In the present study, multiple feast-famine strategies were assessed, for the first time, in two alternate biofilters and compared with a standard continuous biofilter using CH_4 as the model carbon source. The robustness of the biofilters towards increasing famine periods, the decrease of the irrigation frequency and air deprivation was evaluated. The alternate biofilters, where the lowest average pressure drops were recorded, exhibited higher CH_4 elimination capacities (by 27.2 ± 6.4%) and mineralizations (by 18.3 ± 8.6%) than the standard biofilter (CH_4 elimination capacities and mineralizations of 10.3 ± 3.6 g m^{-3} h^{-1} and 79.7 ± 20.8%, respectively), along with the lowest recovery period so far reported in biofiltration after pollutant supply resumption (1.5 ± 0.0 h). Metagenomics analysis revealed a significant shift in the structure of the microbial population induced by the feast-famine regimes, which favoured the occurrence of *Planctomycetes* and *Proteobacteria* phyla. Type I/II methanotroph ratios in the alternate units were 7.5 times higher than those found in the control unit, *Methylomonas* becoming the most resilient genus under feast/famine operation. The current work represents a scaled-down study that demonstrates the feasibility of applying feast-famine strategies at full-scale to increase the performance of biofilters under long-term operation and the lifespan of the packed bed.

1. Introduction

International greenhouse gas (GHG) inventories rank methane (CH_4) as the second most prevalent GHG, with a contribution of 10–16% to the annual GHG emissions worldwide and an atmospheric concentration increase of 150% in the past 250 years (IPCC, 2014; EPA, 2016). Despite the lower mass emissions of CH_4 compared to CO_2, its global warming potential is 25 times higher than that of CO_2 over a 100-y horizon, this factor increasing up to 72 in a 20-y horizon. Therefore, CH_4 contributes to ~30% of the so-called "current net climate forcing" (Solomon et al., 2007). In nature, CH_4 is mainly emitted from the anaerobic decomposition of organic matter in wetlands and oceans. However, more than 60% of the CH_4 emissions worldwide are anthropogenic. These anthropogenic CH_4-laden emissions are usually generated from livestock farming (199 million tons of CO_2-eq), landfilling, composting and wastewater treatment (125 million tons CO_2-eq), and coal mining (24 million tons CO_2-eq) (EEA, 2015). The concentration of CH_4 in these emissions varies from 0 to 0.2 gCH_4 m^{-3} for compost piles to 20–100 gCH_4 m^{-3} for old landfills. It is noteworthy that more than 55% of anthropogenic CH_4 emissions possess concentrations below the lower explosive limit of CH_4-air mixtures (5% *v/v*), which render them unsuitable for energy recovery (Estrada et al., 2014). In addition, the gradual implementation of the Kyoto protocol and the EU landfill Directive 1999/31/EC has tightened European regulations on GHGs, thus resulting in emissions with even lower CH_4 concentrations. In this context, the development of cost-efficient and sustainable technologies for the abatement of these diluted CH_4 emissions is crucial (López et al., 2013).

Biotechnologies have emerged as a cost-efficient and environmentally-friendly alternative to conventional physical-chemical technologies (Estrada et al., 2012) and showed to be promising for the abatement of methane mediated by the activity of methanotrophs (Ménard et al., 2012; Rocha-Rios et al., 2013; Estrada et al., 2014). Among biotechnologies, biofilters (BFs) have been extensively employed in the past decades for industrial off-gas control and represent the most cost-effective option for the mitigation of diffuse CH_4 emissions at full-scale, ensuring that the optimum packed bed selection, nutrient requirements and operational conditions are implemented (Gómez-Cuervo et al., 2017).

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However, despite the significant advances carried out in the past years in the field of biological CH_4 abatement, biotechnologies are still limited by the low aqueous solubility of CH_4 (dimensionless Henry's law constant of 29.5 at 25 °C and 1 atm) and the unresolved problems of conventional biofiltration such as bed clogging and channeling as a result of biomass overgrowth. In this regard, previous studies have consistently demonstrated that an excess of biomass in BFs ultimately reduces the stability and abatement performance of the target pollutant, with an associated increase in operating costs derived from bed conditioning and energy consumption for air circulation (Morgan-Sagastume et al., 2001; Devinny and Ramesh, 2005). Design and operational strategies to control biomass growth in BFs such as back- or chemical washing and mechanical bed stirring or continuous ozone addition often entail a significant down-time after treatment, complex reactor design and additional operating costs, which has limited their widespread use in off-gas biofiltration (Smith et al., 1996; Wübker et al., 1997; Covarrubias-García et al., 2017). In this context, the implementation of innovative low-cost biomass control strategies such as protozoa and mite predation (Woertz et al., 2002), reverse gas flow operation (Rojo et al., 2012), step-feed operation (Estrada et al., 2013) and the implementation of famine regimes (Dorado et al., 2012) have resulted in an efficient long-term performance in biotechnologies devoted to the treatment of VOCs, the later strategy significantly reducing the overall biomass accumulated per volume unit of the biofilter at the expenses of an additional abatement unit. To date, few studies have systematically compared the performance of alternate units operated under feast-famine regimes with that of a continuous control unit. Indeed, the recovery of CH_4 abatement performance following the suppression of pollutant supply during feast-famine regimes (microbial activity robustness) has been poorly explored in biotechnologies devoted to GHG treatment (Ferdowsi et al., 2016). In addition, to the best of our knowledge, the influence of these feast-famine regimes on the diversity and structure of the active methane-oxidizing populations has not been previously assessed.

The main aim of this work was to develop an innovative operational strategy in order to minimize biomass accumulation and sustain long-term CH_4 mitigation. Thus, a systematic evaluation of the feasibility of feast-famine strategies as a promising alternative to conventional biomass control approaches during CH_4 biofiltration, with a special focus on the influence of the extent of feast-famine periods, the frequency of irrigation and air deprivation on the recovery of pollutant abatement performance was carried out. Next-generation sequencing was used to assess the influence of feast-famine strategies on the structure and diversity of the active methanotrophic populations.

2. Materials and methods

2.1. Chemicals and nutrient solution

Methane was purchased from Abelló Linde S.A. (Barcelona, Spain) with a purity of at least 99.5%. All reagents and chemicals were purchased from Panreac® (Barcelona, Spain) with a purity of at least 99%.

Centrate wastewater from the centrifugation of anaerobically digested mixed sludge of wastewater treatment plants (WWTPs) was selected as low-cost nutrient medium for the enrichment of methanotrophs and the irrigation of the inorganic biofilters. Centrate, which is characterized by a low biodegradable fraction of organic matter and high nutrient concentrations, was monthly obtained from Valladolid WWTP (Valladolid, Spain) and stored at 4 °C prior to use. According to preliminary batch tests conducted in our lab, a 3-fold dilution was applied to centrate for the irrigation of the BFs in order to avoid inhibition of methanotrophs due to its high N-NH_4^+ concentrations. Additionally, this diluted centrate was supplemented with SO_4^{2-} (using $MgSO_4 \cdot 7H_2O$) to a final concentration of 150 mg L^{-1} (~50 mgS L^{-1}) in order to prevent any biological limitation due to the absence of S-SO_4^{2-} in the nutrient medium. S-SO_4^{2-} was not detected through HPLC-IC measurements (lower detection limit of 5 ppm SO_4^{2-}) in the raw centrate used in the present study. Based on elemental composition analyses (CHONS) carried out for methanotrophic biomass samples in our laboratory, a S content ranging from 0.5 to 1% is typically found, which highlighted the need of S-SO_4^{2-} supplementation in the diluted centrate used for biofilter irrigation. The adjusted sulfate concentration was selected according to the concentrations used in previous studies on CH_4 biodegradation (30–130 mg S L^{-1}, Whittenbury et al., 1970; López et al., 2014). The final composition of the diluted centrate is shown in Table S1 (Supporting Information).

2.2. Methanotrophic enrichment

A 10-L laboratory scale stirred tank reactor (STR) was set-up for the enrichment of methanotrophs under continuous feeding of a CH_4-laden air stream for 30 days. Fresh aerobic activated sludge from Valladolid WWTP was used as inoculum. The operational conditions of the STR were described in the Supporting Information.

2.3. Experimental set-up, inoculation, operation mode and biomass sampling

Three lab-scale BFs consisting of cylindrical PVC columns (height = 0.45 m; inner diameter = 0.08 m) were set-up (Fig. 1). Each BF was packed with 2.3 L of the impregnated Kaldnes rings used during methanotrophic enrichment and further irrigated with 150 mL of the concentrated methanotrophic biomass suspension. The BFs were maintained at a temperature of 24.7 ± 1.0 °C in a temperature-controlled room. A pure CH_4 gas stream, regulated with a mass flow controller (Aalborg™, USA), was mixed with pre-humidified air in a mixing chamber and the resulting stream was continuously fed at 8.4 L h^{-1} to the control BF (BF 1) and the BF operated under feast conditions (either BF 2 or BF 3). This operation resulted in a gas stream with an inlet CH_4 concentration of 31.0 ± 1.3 g m^{-3} (4.7 ± 0.2% *v/v*), a humidity of 96.8 ± 3.2% and an empty bed residence time (EBRT) of 17.1 min. Thus, a CH_4 inlet load (IL) of 108.7 ± 4.5 g m^{-3} h^{-1} was applied to the units under CH_4 supply. The concentrations here applied can be typically found in old landfills, coal mines and WWTPs, and remained under the lower explosive limit (<5% (*v/v*)). Since the treatment of CH_4 emissions via incineration or energy recovery are only applicable for CH_4 concentrations above 30% (*v/v*), biotechnologies such as BFs represent an environmentally-friendly platform for the treatment of these off-gases. The use of such relatively high CH_4 concentration might mediate a faster biomass accumulation, which allows evaluating the impact of feast-famine strategies on biofilter performance. On the other hand, the BF unit under famine conditions (either BF 3 or BF 2) was either fed with CH_4 free-air at 8.4 L h^{-1} or enclosed to prevent air circulation through the BF, depending on the operational stage evaluated. The succession of each feast-famine cycle occurred through alternate shifts in gas feeding between BFs 2 and 3 from CH_4-deprived (famine regime) to CH_4-supplemented (feast regime) conditions and *vice versa*. Before each change, the gas phase of the unit under famine was purged with air at 30 L h^{-1} for 0.5 h to avoid an overestimation of the CO_2 production during the first hours of the feast periods.

The BFs were initially operated for 3 days under abiotic