OPTIMISATION OF CONVENTIONAL BIOFILTRATION OF GASEOUS EMISSIONS WITH LOW METHANE CONCENTRATIONS

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ABSTRACT

Methane is one the most problematic greenhouse gas (GHG) present in industrial gas emissions such as those derived from coal mines venting, landfills, livestock facilities and wastewater treatment plants (WWTPs), among others. All those emissions are characterised by large flows and low methane contents (usually below 1.5 % vv^{-1}).

Up to now, different works have demonstrated the feasibility of methane biological abatement by the use of biofiltration, being most of them focussed on gaseous streams with inlet methane concentrations higher than 1% vv^{-1}. However, since lower concentrations are usually found in a wide variety of industrial emissions, the aim of this study was to evaluate the conventional biofiltration at very low methane concentration, i.e. 0.2% vv^{-1}.

A biofilter for the methane treatment (BF) was packed with a mixture of wood pine bark chips, perlite and compost. The BF was working for more than 250 days with no significant pressure drop problems. The influence of different parameters was assessed: i) the waterings; ii) nitrogen source (ammonia vs. nitrate); iii) empty bed residence time (EBRT) and iv) inlet loads (IL). All these operational parameters were studied in terms on their influence on methane removal efficiencies (RE) as well as elimination capacities (EC).

Our results indicate a strong influence of the waterings periods, with an optimum value of 255 mL day^{-1} carried out during one watering. Regarding nitrogen source, the best REs were obtained with ammonia instead of nitrate (average of 44.3% vs 30% of RE reached with nitrate). On the other hand, ammonia can lead to pH instability due to acidification.

In the case of the influence of EBRT, a range of values from 2.0 to 10 .0 min were tested reaching the best performance when 8 minutes was applied (RE 34 %). Concerning to the IL, it was varied from 9.7 up to 85.4 g CH_4 m^{-3} h^{-1} which was carried out by increasing the inlet methane concentration maintaining an EBRT of 4.7 min. Under these conditions the maximum RE (26%) was obtained at an inlet load of 17.2 g CH_4 m^{-3} h^{-1} but the highest EC (20.4 g CH_4 m^{-3} h^{-1}) was obtained at the highest inlet concentration. It demonstrated concentration was the key parameter to determine the maximum degradation in the biofilter.

Keywords: methane diffuse emissions, biofilter, nitrogen source, optimization, gas abatement, greenhouse gases.

1. Introduction

Methane is one of the most problematic greenhouse gases (GHG) emitted as anthropogenic source due to its high global warming potential (20 -25 times higher than CO_2). There are numerous industrial activities which involved methane emissions. Mostly of them present above 30% of methane concentration being this concentration economically viable for obtaining energy (Humer and Lechner, 1999). Other industrial activities present lower concentrations of methane which must be treated to avoid the uncontrollable release of methane to the atmosphere.
Special attention have been focused on facilities which emit large flows with very low concentrations of methane (below 1.5 % vv^{-1}). These kind of emissions, known as methane diffuse

emission, are produced in facilities such as landfills, livestock, coal mine or wastewater treatment plants (WWTP), among other.

In this sense, (Di Bella et al., 2011) measured a methane flux of 12.9 T CH_4 m^{-2} $year^{-1}$ in a municipal landfill during one year of sampling. Likewise, Daelman et al. (2013) determined a mass flux between 79 – 153 T CH_4 $year^{-1}$ in a conventional wastewater treatment plant. Moreover, concentration measured in different industrial processes demonstrated the importance of the diffuse emissions. Thus, Girard et al. (2011) measured concentrations from 250 to 4300 ppm in the venting effluents from piggery industries. Regarding WWTPs, recent works reported significant methane releases from most of the units, although always at low concentrations, below 3000 ppm (0.3% vv^{-1}) (Lorenzo-Toja et al., 2015)

Despite these low concentrations emitted, diffuse emissions must be treated since there are many sources which release this kind of greenhouse gases in an uncontrolled way which can lead to a significant increase in the overall contribution into the atmosphere. For this purpose, biological approaches are quite interesting for the treatment of this kind of emissions. In this study, a biofilter was employed due to its simplicity, the low costs involved (Devinny et al., 1998; Le Cloirec et al., 2001)and also the removal efficiencies (RE) rates reported by many authors.

The aim of present work was to evaluate the biofiltration of the lowest range of methane diffuse emissions reported for industrial activities (0.2 % vv^{-1}) in a conventional biofilter packed with low cost packaging material. The process was assessed in terms of removal efficiency (RE) and elimination capacity (EC) in a long-term study where additionally it was evaluated the influence of watering and the type and concentration of the nitrogen source concentration.

2. Materials and methods

The biofilter for the methane treatment is made of glass with a total volume of 13.4 L, inner diameter of 0.1 m and a total length of 1.7 m. It is constituted by three independent modules with three sampling points at different heights. The biofilter was packed with 5.85 L of a mixture of wood pine bark chips, perlite and compost.

A synthetic gaseous stream was fed containing air and pure methane (Carburos Metalicos, Spain), being this mixture controlled by means of mass flow controllers (Bronkhorst High-Tech BBV, The Netherlands). The inlet flow applied corresponded to an empty bed residence time (EBRT) of 4.4 min. The inlet concentrations (IC) were maintained in a range of 1305 to 2472 ppmv CH_4, corresponding to inlet loads of 11.7 to 22.2 g CH_4 m^{-3} h^{-1}.

3. Results and discussion

The biofilter (BF) was working throughout 250 days in which it was assessed the water addition and nitrogen source influence in first term and afterwards it was optimized the EBRT and IL supplied.

Water addition

Watering is one of the most important parameter to sustain microbiological activity and to spread nutrient among the bacteria (Scheutz, , et al., 2009). The control of water content in the biofilter is essential to maintain optimal conditions for biomass. Nevertheless, methane has a very low Henry constant what means a very low solubility in aqueous phase. Therefore, the aim of watering assay was to balance the need of water to maintain a proper biological activity as well as to minimize mass transfer limitations by not supplying more water than needed for biological purpose. Thus, it was varied from six to one the number of watering per day (equivalent to flow rates of 1980 and 330 ml $d^{-1,}$ respectively) increasing the RE while the watering frequency was reduced reaching the higher efficiencies (RE= 51% and EC = 7.8 ± 2.3 g CH_4 m^{-3} h^{-1}) when only one watering was carried out. Furthermore, watering flow rates were 0.45, 6.0, 160, 320 and 627 mL d^{-1} which were maintained each during more than one week. Results indicated that better results (EC = 7.5 ± 0.2 gCH_4 m^{-3} h^{-1}) were obtained when it was supplied 320 mL d^{-1} (corresponding to 0.056 $L_{Liquid\ phase}$ L_{bed}^{-1} d^{-1}) and the EC diminished significantly at different volume. As a result, only one watering per day with a volume between 250 -320 mL d^{-1} was employed throughout the whole study.

Nitrogen source

The type of nitrogen source determines the possible stability of the reaction in a long-term operation. The by-products production, pH variations and C/N ratio were essential factors in a microbial community and it must be controlled in the biofilter to achieve higher performances. So, ammonia (NH_4^+) and nitrate (NO_3^-) was compared as nitrogen source. Both have been reported as inhibitors at high concentrations through different pathways (Hütsch et al., 1993; Scheutz et al., 2009) hence the initial concentration of nitrogen was kept around 0.1 g N L^{-1} for both nitrogen sources. During short-term tests (two weeks) the performance of the biofilter with N-NH_4^+ was higher than with N-NO_3^- as nitrogen source (44.3 – 30 % of RE respectively). To demonstrate the inhibitory effect of ammonia concentration as nitrogen source, different concentrations (0.13 – 1 g N-NH_4+ L^{-1}) were tested achieving the highest performance with 0.5 g N-NH_4^+ L^{-1}. As a result, 0.5 g N-NH_4^+ L^{-1} was employed in the long-term operation. After few days using ammonia as nitrogen source an important acidification in the biofilter occurred leading a decrease in RE. A decrease of ammonia concentration and an addition of buffer solutions was not enough to control pH being a change of nitrogen source to nitrate (NO_3^-) the solution for restoring the stability. The use of a 0.2 g N-NO_3^- L^{-1} presented no problems of stability and consequently it was employed as nitrogen source during the long-term operation and optimization tests.

Optimization tests

EBRT and IL optimization tests were carried out to enhance the biofiltration performance and to find out the limits of the system to treat methane diffuse emissions.

In the case of EBRT assay, a range of values from 2.0 up to 10.0 minutes were obtained by terms of decreasing IL were tested. The best RE was achieved when 8 minutes was applied (RE = 34%) decreasing the performance of the biofilter for shorter EBRT (Figure 1b). Despite of the highest values obtained, optimum EBRT was around 4 - 6 minutes since the RE was quite high (24 – 29 %) at these times and it is more adjusted to a real scale biofilter operating in an industrial process. EC trend to decrease according to the increase in the EBRT fluctuating 2 g CH_4 $m^{-3}h^{-1}$ during the whole assay. (Figure 1 b)

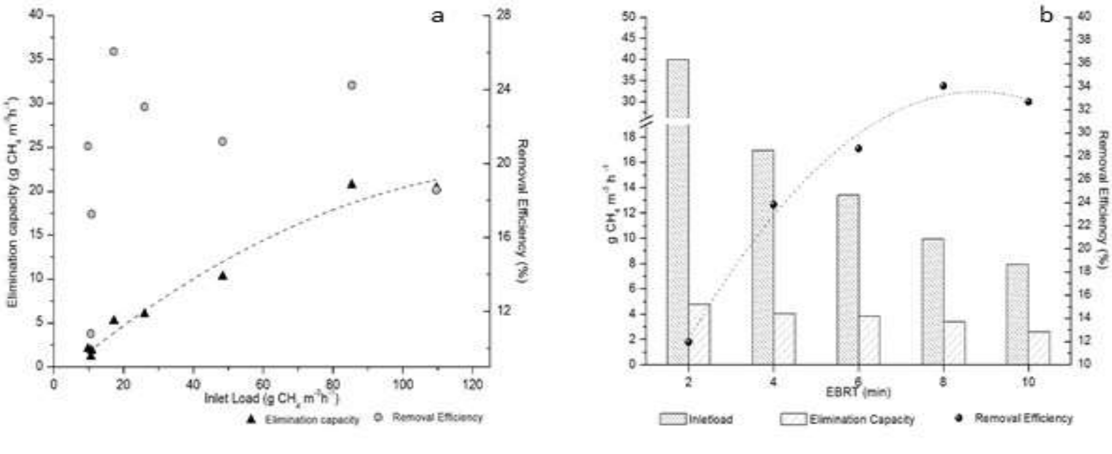

Figure 1: EBRT and IL assay results

Regarding IL, the load supplied varied from 9.7 to 85.4 g CH_4 $m^{-3}h^{-1}$ which were obtained increasing the inlet concentration from 0.11 to 1.3 % vv^{-1} and keeping constant an EBRT at 4.7 minutes. Results obtained depict a maximum RE (26 %) at an IL of 17.2 g CH_4 m^{-3} h^{-1} (0.2% vv^{-1}) remaining constant RE above 18 % for higher IL. On the other hand, EC raised as IL increased what means a direct correlation between the increase of concentration provided and the removal capability of the system. Indeed, loads above 90 g CH_4 $m^{-3}h^{-1}$ (1% vv^{-1}) trend to top out the EC reaching the limit of methanotrophic bacteria consume. Therefore, low solubility of methane in aqueous phase might be a limitation in higher concentrations the concentration applied was the key parameter to enhance the biofiltration.

4. Conclusion

Biofiltration of methane diffuse emissions in a conventional biofilter has been carried out for more than 250 days. Watering assay fixed in one watering of nutrient solution of 0.056 $L_{Liquid\ phase}\ L_{bed}^{-1}\ d^{-1}$ was found as the most adequate to achieve the best RE. In the same way, nitrate as nitrogen source has demonstrated better stability in a long term operation. EBRT around 4 - 6 minutes represents a balance between high performance and a realistic value to the industrial processes. IL assays show a direct correlation between inlet concentration and the elimination capacity of the system, showing that the biological treatment of methane diffuse emissions (0.2 % vv^{-1}) is strongly limited by the low methane available for bacterial consumption in the liquid phase, according to kinetic affinities reported for methanotrophs.

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