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## Effect of primary treatment and organic loading on methane emissions from horizontal subsurface flow constructed wetlands treating urban wastewater

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#### ABSTRACT

Methane is emitted in horizontal subsurface flow constructed wetlands (HSSF CWs) during wastewater treatment. The objective of this work was to determine the influence of primary treatment and organic loading rate on methane emissions from constructed wetlands. To this aim, methane emissions from a HSSF CW pilot plant were measured using the closed chamber method. The effect of primary treatment was addressed by comparing emissions from wetlands receiving the effluent of an anaerobic (HUSB reactor) or a conventional settler as primary treatments. Alternatively, the effect of organic loading was addressed by comparing emissions from wetlands operated under high organic loading  $(52 g)$ COD  $\rm m^{-2}$  day<sup>-1</sup>) and low organic loading (17 g COD  $\rm m^{-2}$  day<sup>-1</sup>). Results showed that methane emission rates were affected by the type of primary treatment and, to a lesser extent, by the organic loading applied. Accordingly, lower redox conditions and slightly higher organic loading of a wetland receiving the effluent of a HUSB reactor resulted in methane emissions twelve times higher than those of the wetland fed with primary settled wastewater. Moreover, systems subjected to three times higher organic loading than that recommended lead to higher methane emission rates, although high data variability resulted in no statistically significant differences.

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#### 1. Introduction

Horizontal subsurface flow constructed wetlands (HSSF CW) are natural wastewater treatment systems that represent a suitable alternative to conventional technologies. Low energy consumption and operation costs are some of the advantages of this technology that make it a viable option for the sanitation of small communities (PE < 2000) [\(García et al., 2001; Puigagut et al.,](#page--1-0) [2007\)](#page--1-0). In HSSF CWs organic matter is removed by means of physical, chemical and biological processes that occur naturally and simultaneously within the treatment bed. Although subsurface constructed wetlands are systems subjected to great spatial redox variations (especially in depth) [\(García et al., 2003](#page--1-0)) they are considered to be mainly anaerobic ([Baptista et al., 2003](#page--1-0)) and, therefore, methane emission takes place during the wastewater treatment.

Methane is among the most important gases of greenhouse effect as it has not only increased by ca. three times since pre-

industrial times but also its global warming potential is about 25 times higher than  $CO<sub>2</sub>$  ([IPCC, 2001](#page--1-0)). Methane in the atmosphere is mainly from biological origin (70–80%) and comes from the activity of methanogenic bacteria in environments where anaerobic pathways predominate. In wetlands methane is produced whenever redox conditions are below  $-200 \text{ mV}$  and only after other electron acceptors such as nitrate or sulphate have been reduced [\(Mitsch and Gosselink, 2000](#page--1-0)). Furthermore, besides redox conditions, there are other environmental and operational parameters such as temperature or organic loading that has a great impact on methane emission from wetlands ([Søvik et al.,](#page--1-0) [2006; García et al., 2010; Mander et al., 2014](#page--1-0)). Moreover, organic loading is of special concern in the context of Spain due to the large number of systems operated under high organic load conditions ([Puigagut et al., 2007](#page--1-0)). Wetlands overloading not only contributes to increase methane emissions during wastewater treatment, but has been also directly linked to one of the main operational problems associated to constructed wetlands: the clogging ([Pedescoll et al., 2011b](#page--1-0)). In order to prevent clogging in wetlands, primary treatments are applied to wastewater. Generally, physical treatments such as settlers or imhoff tanks are used. However, Corresponding author. Tel.: +34 934010898; fax: +34 934017357.<br>E-mail address: jaume.puigagut@upc.edu (J. Puigagut). Tecently other technologies are being considered as a suitable

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primary treatment for HSSF CWs, such as hydrolytic upflow sludge blanket (HUSB) reactors ([Pedescoll et al., 2011a\)](#page--1-0). Applying HUSB reactors as primary treatment for wetlands has the advantage of supplying higher biodegradable substrate to the system [\(Ligero](#page--1-0) [et al., 2001\)](#page--1-0). However, HUSB effluents are also characterized by imposing higher organic loading rates ([Barros et al., 2008\)](#page--1-0) and lower redox conditions ([Pedescoll et al., 2011a](#page--1-0)) within the wetland that, in turn, may enhance methane emissions during water treatment.

The main objective of the present study was to determine the influence of both the organic loading conditions and the type of primary treatment (conventional settling vs anaerobic treatment) on methane emissions from horizontal subsurface flow constructed wetlands (HSSF CWs). The effect of redox conditions imposed by either the type of primary treatment or the organic loading applied on plant performance is also discussed.

#### 2. Materials and methods

#### 2.1. Pilot plant

The constructed wetlands pilot plant was set up in March 2011 and was fed with urban wastewater pumped directly from the municipal sewer. Initially, wastewater was coarsely screened and pumped to a homogenization tank having a hydraulic retention time of five hours. Within the homogenization tank wastewater was kept in constant agitation to avoid solids sedimentation. After the homogenization tank, wastewater was conveyed to the primary treatment that consisted either of one HUSB reactor of 114 L of volume operated at 4 h of HRT and at 10 g of volatile solids per liter or two settlers of 14 L each that were operated in parallel at two hours of sedimentation time. After the primary treatment, wastewater was pumped to the secondary treatment. Secondary treatment consisted of three wetlands of  $0.4 \text{ m}^2$  of surface (70 cm length  $\times$  55 cm width  $\times$  35 cm depth) with a gravel matrix  $(D_{60} = 7.3; C<sub>u</sub> = 0.8)$  having an initial porosity of 40%. Water level inside the wetlands was kept at 30 cm depth (5 cm below the gravel surface). All wetlands were planted from the beginning of its operation with common reed (Phragmites australis). A sketch of the pilot plant is shown in Fig. 1.

For the purposes of this study three experimental lines were considered. The first two lines (named under low and high organic loading lines - HUSB-L and HUSB-H, respectively) consisted of two of the wetlands fed with the HUSB reactor effluents, one at



Fig. 1. Sketch of the pilot plant: (1) homogenization tank; (2) HUSB reactor; (3) pair of settlers; (4) wetland receiving HUSB effluents and operated at low organic loading (HUSB-L line); (5) wetland receiving HUSB effluents and operated at high organic loading (HUSB-H line); (6) wetland receiving settler effluents and operated at low organic loading (SET line); (7) outflow storage tanks.

 $21$  L day<sup>-1</sup> (2.6 days of hydraulic retention time) and the other at  $63$  L day<sup>-1</sup> (0.85 days of hydraulic retention time). The HUSB-L and HUSB-H lines were operated at ca. 17 and ca. 52 g COD  $m^{-2}$  day<sup>-1</sup>, respectively, which was equivalent to approximately 7 and 20 g  $BOD_5$  m<sup>-2</sup> day<sup>-1</sup>, respectively. The third wetland (named under settler line – SET line) was operated at a hydraulic loading rate of  $21L$  day<sup>-1</sup> but was fed with the conventional settler effluent. The SET line was operated at ca. 15 g  $COMm^{-2}$  day<sup>-1</sup> which was equivalent to ca.  $6$  g BOD<sub>5</sub> m<sup>-2</sup> day<sup>-1</sup>. The effect of organic loading rate on methane emissions was addressed by comparing the HUSB-L and HUSB-H lines between April and September 2013, whereas the effect of primary treatment on methane emissions was addressed by comparing the HUSB-L line and the SET line between July 2012 and July 2013. It is important to mention that the HUSB reactor was set in operation in May 2012 and the wetlands fed with HUSB effluents in the present experiment had been previously fed with settled wastewater at a hydraulic loading of 21 Lday<sup>-1</sup>.

Furthermore, each wetland had a PVC cylinder of 20 cm diameter placed at the middle of its surface that was used to implement the closed chamber for methane measurements.

#### 2.2. Methane measurements

Methane emissions were measured following the closed chamber method ([Livingston and Hutchinson, 1995\)](#page--1-0). The closed chamber employed consisted of a PVC cylindrical reservoir of ca. 4 L of effective volume, having 19 cm and 15 cm to the diameter and height, respectively. The sampling port was located at the top of the chamber and was also equipped with a thermometer (OAKTON) and a rolled vent tube (2 mm of internal diameter and 2 m long). At the end of the sampling port a two-way stopcock was disposed for sample withdrawal. The chamber was implemented with a lap-top power-adjustable 12V fan  $(0.011 \text{ m}^3 \text{ s}^{-1})$ attached to the upper part of the chamber with adhering rubber. The fan had a diameter of 120 mm with blades length of 25.4 mm.

Measurements were conducted by placing the closed chamber at the middle zone of the wetlands leaving a headspace were methane accumulated. During experiment deployment the base of the chamber was kept in contact with water to avoid methane leaching. Temperature conditions within the chamber were recorded for each experiment. Once the chamber was placed in the wetland, samples were extracted after 0, 10 and 20 min for sampling campaigns carried out in 2012 and after 0, 10, 20, 30 and 40 min for sampling campaigns carried out in 2013. Sample withdrawal was conducted with 100 mL syringes and always extracting 60 mL of air from the head space. Methane was analyzed once the experiment had finished (between 2 and 4 h after the last sample withdrawal had been carried out) by a gas chromatograph coupled to a FID detector (GC system–Agilent Technologies 7820 A). Methane emission rates were then estimated assuming linear emission patterns with minimum coefficients of determination (R-square values) of 0.85.

#### 2.3. Water quality parameters and redox potential

Water quality parameters surveyed during the experiment were COD and ammonia. Sampling was conducted at the inlet and outlet of the wetlands around the time were methane sampling campaigns were conducted. Analyses were performed according to standard methods [\(APHA-AWWA-WEF, 2005](#page--1-0)).

Redox potential was measured before, during and just after methane analysis were conducted at each sampling campaign during periods ranging between 1 and 9 days. Wetland redox status was monitored at 5 and 15 cm depth from the water table (10 and 20 cm depth from the gravel level) by means of a portable meter (Digimed TH-404) equipped with a platinum electrode

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