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Relationship between the removal of organic matter and the production of methane in subsurface flow constructed wetlands designed for wastewater treatment

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ABSTRACT

The aim of this study was to evaluate the relationship between the organic matter removal and methane production in wetlands with a horizontal subsurface flow (HSSF) treating wastewater using *Phragmites australis* (Phr) and *Schoenoplectus californicus* (Sch). Four HSSF systems with a superficial area of 4.5 m^2 a water table depth of 0.4 m that were planted with Phr and Sch were evaluated. For the evaluation, each HSSF was divided into three transversal zones (A, B, and C). The operation was controlled for the hydraulic loading (19.8–27.8 mm d^{-1}), hydraulic retention time (3–7 d) and organic loading rates (3.3–4.2 g biological oxygen demand—BOD_5 m^{-2} d^{-1}). The removal efficiencies for the organic matter (BOD_5 and chemical oxygen demand (COD)) and solids were evaluated in each HSSF during the 420 days of operation. To evaluate the methane produced in the HSSFs, maximum methanogenic activity assays from the biomass of three zones (A, B and C) along the HSSF were performed. The results show that the HSSF planted with Phr and Sch presented removal efficiencies of 70–80% BOD_5, more than 60% for the COD and 70–95% of solids during the monitoring period. However, the methane biomass activity shows similar average methane production for HSSF-Phr and HSSF-Sch, although a mild difference was noted between Zone A (700 mL CH_4 g volatile suspended solid—VSS^{-1}), Zone B (1035 mL CH_4 g VSS^{-1}) and Zone C (368 mL CH_4 g VSS^{-1}). Moreover, mass balance determined that HSSF-Phr and HSSF-Sch are able to degrade 13.74 g COD d^{-1}. Finally, the average methane production for HSSF-Phr and HSSF-Sch was 1455 ± 482 mg CH_4 m^{-2} d^{-1} and 1305 ± 27 mg CH_4 m^{-2} d^{-1}, respectively.

1. Introduction

Constructed wetlands (CW) are proven, effective wastewater treatment systems (Vymazal and Kröpfelová, 2008; García et al., 2010; Vera et al., 2011). In particular, the use of HSSF in wastewater treatment has shown solids elimination efficiencies above 90% (Caselles-Osorio et al., 2007) and organic matter (OM) measured as a COD and BOD_5 reductions in the range of 67–84% and 78–94%, respectively (Vymazal and Kröpfelová, 2011). In this way, the concentrations of COD and BOD_5 in the effluent treated by HSSF have values lower than 60 mg L^{-1} and 20 mg L^{-1}, respectively (Vera et al., 2011; Rojas et al., 2013; Vymazal and Kröpfelová, 2008). The main OM removal processes in HSSF correspond to anaerobic microbiological processes (greater than 94.7%) because of the continuous saturation of the bed and applied organic load (3–15 g BOD_5 m^{-2} d^{-1}) (Marecos do Monte and Albuquerque, 2010; Vasudevan et al., 2011). Consequently, anoxic–anaerobic conditions prevail in HSSF with a redox potential in the range of −100 mV to −500 mV and with dissolved oxygen concentrations below 2 mg L^{-1} (Vasudevan et al., 2011; Vymazal, and Kröpfelová, 2008). Additionally, these processes can be influenced by the design parameters of the HSSF, including the water depth (depth of the water table), length/width ratio and type of granular medium (gravel) (Aguirre et al., 2005; García et al., 2005). In the HSSF designed with a hydraulic piston flow, the OM contained in the wastewater decreases by 50%, principally in the section of the entrance of HSSF (Tanner et al., 1998; García et al., 2007; Caselles-Osorio et al., 2007). This occurs because the particulate fraction precipitates and the soluble fraction is removed, although they are influenced by the wastewater path in the HSSF (Kadlec and Wallace, 2009; Ávila et al., 2013).

However, the depth of the water table (greater than 30 cm) in the HSSF generates conditions that allow the development of anaerobic microbiological activity (greater than 90%)

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(García et al., 2004; Aguirre et al., 2005). Specifically, HSSFs have been shown to produce methane emissions between 0 and 16,760 mg CH_4 m^{-2} d^{-1} (Johansson et al., 2004; Wang et al., 2008). HSSF is also known to emit methane into the atmosphere, corresponding to the net result of methane production (methanogenesis) and consumption (oxidation). This emission rate is affected by a number of factors, such as the redox status, spatial characteristics, seasonality, organic load applied, pretreatment system, depth of the HSSF, and macrophyte species used (Grünfeld and Brix, 1999; Wang et al., 2013; Nadarajah et al., 2007; Enright et al., 2009; Samsó and García, 2013).

Particularly, the amount and type of vegetation plays an important role in methane production, oxidation and transport to the atmosphere (Grünfeld and Brix, 1999; Van bodegom et al., 2001; Inamori et al., 2007). Grünfeld and Brix (1999) found that the presence of *Phragmites australis* attenuates the rate of methane emission by 64% when comparing wetlands with plants (882.2 mg CH_4 m^{-2} d^{-1}) to wetlands without plants (1363.4 mg CH_4 m^{-2} d^{-1}). Moreover, a lower water surface height (22 cm of deep) in a wetland with plants increased the methane oxidation by 28% at the interface of the anoxic and oxic sites of the roots of plants.

Even with multiple studies reporting *in situ* emissions of methane (0–16,760 mg CH_4 m^{-2} d^{-1}), CO_2 (1464–3360 mg CO_2-C m^{-2} d^{-1}), N_2O (−5.5 to 32.7 mg N_2O m^{-2} d^{-1}) and N_2 (4.08–3120 mg N_2-N m^{-2} d^{-1}) (Grünfeld and Brix, 1999; Van bodegom et al., 2001; Teiter and Mander, 2005; Wang et al., 2008, 2013), minimal information is available regarding the methane produced by the anaerobic biomass associated with the gravel of the HSSF and its relation with the removal of OM.

Therefore, the aim of this study was to evaluate the relationship between the removal of OM and the production of methane in wetlands with a horizontal subsurface flow (HSSF) treating wastewater using *Phragmites australis* and *Schoenoplectus californicus*.

2. Material and methods

2.1. HSSF constructed wetlands

The wetland system is located in Hualqui (36°59′26.93″ south latitude and 72°56′47.23″ west longitude), Biobío Region, Chile. The influent into the HSSF corresponds to the wastewater from a treatment plant that serves a rural community of 20,000 inhabitants. The wastewater was extracted after pre-treatment (chamber with 40 mm bars and a sand trap) and transported to a homogenizing tank (650 L). The HSSF was then fed by gravity (Vera et al., 2014).

The HSSF system consists of four wetland horizontal subsurface flow units. Each HSSF unit has an area of 4.5 m^2, a total volume of 1.28 m^3, a length/width ratio of 2, and a water table of 0.4 m in depth. The support medium was gravel (19–25 mm), with an average height of 0.57 m. Fig. 1 shows the HSSF system: units HSSF-Phr1 and HSSF-Phr2 were planted with the *Phragmites australis* macrophyte species, meanwhile units HSSF-Sch1 and HSSF-Sch2 were planted with the *Schoenoplectus californicus* macrophyte species. In each HSSF, three gravel samplers were installed in each zone: in Zone A (initial zone), 0.65 m from the entrance point; in Zone B (middle zone), 1.4 m from the entrance point; and in Zone C (output zone), 2.25 m from the wetland entrance point (Fig. 1b(i)). The superficial area of each zone is 1.5 m^2 (Rojas et al., 2013).

Table 1 shows the operational and climatic parameters for all of the HSSF units. The hydraulic loading (HL) varied between 19.8 and 27.8 mm d^{-1}, and the hydraulic retention time (HRT) varied between 3 and 7 days. The organic loading rate (OLR) was between 3.3 and 4.2 g BOD_5 m^{-2} d^{-1}. The evapotranspiration rate (ET) presented maximum variations between winter (1.1 L m^{-2} d^{-1}) and summer (4.0 L m^{-2} d^{-1}). Marked seasonal trends of rainfall were observed, with maximum rainfall in the fall and winter (2.2–4.0 L m^{-2} d^{-1}) and minimum rainfall of 0.6 L m^{-2} d^{-1} in the spring and summer.

2.2. Sampling strategy

The system was implemented in July 2011. The HSSFs were operated for 420 days, and the first 85 days corresponded to a stabilization period. The sampling was performed in the spring (S), summer (Sm), fall (F) and winter (W) over the total operation period, as indicated in Table 1.

At three points in each HSSF, the following measurements were monitored *in situ*: (a) temperature, (b) pH and (c) oxidation reduction potential (ORP). The dissolved oxygen (DO) was measured only at one point (middle) because of the small variations. Fig. 1b(i) displays the *in situ* monitoring points. The parameters are presented as the average between the fall and winter (fall/winter) and spring and summer (spring/summer). A physicochemical characterization of the HSSF wastewater influent and effluent samples was performed in each season; this analysis determined the COD, BOD_5, total suspended solids (TSS), and volatile suspended solids (VSS). *In situ* and physicochemical characterizations were performed every 15 days (Vera et al., 2014).

2.3. Methanogenic activity

2.3.1. Inoculum

The anaerobic biomass used in the methanogenic activity essays corresponded to the biomass attached to the gravel of each HSSF. The biomass that adhered to the gravel was suspended in a saline solution (0.9% NaCl) and then sonicated for 3 min. The samples were then vortexed for 30 s. The leachate is allowed to decant to analyze the TSS and VSS (Morato et al., 2005). A sample of the granular material was withdrawn from Zones A, B and C (Fig. 1b(i)). The sampling was performed between 300 and 400 days (seasons fall/winter).

2.3.2. Specifies methanogenic activity tests$_{(maximum)}$ (SMA_m)

To determine the methanogenic activity, the extracted biomass was transferred from the CW to 100 mL reactors. A mix of volatile fatty acids (VFA) (2.0 g L^{-1} acetic acid, 0.5 g L^{-1} propionic acid and 0.5 g L^{-1} butyric acid) and nutrients was subsequently added as described by Soto et al. (1993). Na_2S was added to each reactor to generate the anaerobic conditions (Soto et al., 1993). Nitrogen gas was then bubbled (for 3 min) in each reactor to remove air from the headspace. The assays were performed at 30 °C. Methane production was measured by volumetric displacement method. This method is based on quantifying the amount of methane produced by the use of a displacing substance, such as NaOH (2.5%). The NaOH reacts with the biogas precipitating the CO_2, allowing just the methane measurement (Soto et al., 1993), as shown in Fig. 1b(ii).

Specific methanogenic activity$_{maximum}$, SMA_m (g COD g VSS^{-1} d^{-1}), was determined according to Eq. (1) (Soto et al., 1993).

$$SMA = \frac{R}{f \times V \times [VS]} \quad (1)$$

where R is the methane production rate (mL CH_4 d^{-1}), f is the conversion factor of CH_4 to g COD (388 mL CH_4 g COD^{-1}), V is the volume of the reactor (0.1 L) and [VS] is the biomass concentration (g VSS L^{-1}).