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Greenhouse gas emission in constructed wetlands for wastewater treatment: A review

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ABSTRACT

A literature analysis of 158 papers published in international peer-reviewed journals indexed by the Thomson Reuters Web of Knowledge from 1994 to 2013 showed that CO_2—C emission was significantly lower in free water surface (FWS) constructed wetlands (CW) than in subsurface flow (SF) CWs (median values from 95.8 to 137.0 mg m^{-2} h^{-1}, respectively). In vertical subsurface flow (VSSF) CWs the CH_4—C emission was significantly lower than in horizontal subsurface flow (HSSF) CWs (median values 3.0, 6.4, and 4.0 mg m^{-2} h^{-1}, respectively). There were no significant differences in N_2O—N emission in various CW types (median for FWS, VSSF and HSSF CWs: 0.09, 0.12, and 0.13 mg m^{-2} h^{-1} correspondingly).

The highest value of emission factor (EF) of CH_4 ((CH_4—C/inflow TOC_{in}) * 100%) was found for FWS CWs (median 18.0%), followed by HSSF CWs (3.8%), and VSSF CWs (1.28%). Median values of N_2O EFs ((N_2O—N/inflow TN_{in}) * 100%) differed significantly in all three CW types: 0.34% for HSSF, 0.11% for FWS, and 0.018% for VSSF CWs.

We found a significant correlation between TOC_{in} and CH_4—C emission and between the TN_{in} and N_2O—N emission values for all of the types of CWs we studied.

Hybrid CWs (e.g., the subsequent combination of VSSF, HSSF and FWS CWs) are beneficial from the point of view of both water purification and minimization of greenhouse gas (GHG) emissions. Likewise, intermittent loading in VSSF CWs and macrophyte harvesting in HSSF and FWS CWs can mitigate GHG emissions.

1. Introduction

Constructed wetlands (CW) are engineered wetland systems that have been designed and constructed to utilize natural processes in treating wastewater (Vymazal et al., 1998). Constructed wetlands are used to improve the quality of wastewater from point and nonpoint sources of water pollution, including domestic, industrial and municipal wastewater, stormwater runoff, farm wastewater, collated runoff from agricultural land and landfill leachate (Kadlec and Knight, 1996; Kadlec and Wallace, 2008).

The main types of CWs are: free water surface (FWS) or surface flow, vertical subsurface flow (VSSF) and horizontal subsurface flow (HSSF) CWs (Vymazal, 2007, 2011). In addition to wastewater treatment, the CWs provide several ecosystem services such as provisional (food, energy, fibers), regulating (carbon (C) sequestration, climate regulating, flood control), supporting (biodiversity,

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nutrient cycling) and cultural (recreational, educational) services (Mitsch and Gosselink, 2007).

Free water surface CWs are shallow and low flow velocity wetlands which have areas of open water and floating, submerged and/or emergent plants (Kadlec and Wallace, 2008). FWS CWs are very effective in the removal of organics through microbial degradation and the removal of suspended solids through filtration and sedimentation (Vymazal et al., 1998). The removal of nitrogen (N) and phosphorus (P) can be sustainable, but depends on inflow concentration, the chemical form of nitrogen, water temperature, the season, organic carbon availability, substrate material and dissolved oxygen concentration (Vymazal, 2011). The FWS wetlands are mostly used for the tertiary treatment of domestic and municipal wastewater, mine drainage waters, and for stormwater and agricultural runoff (Kadlec and Knight, 1996; Kadlec and Wallace, 2008).

In *horizontal subsurface flow CWs*, the wastewater is fed in at the inlet and flows slowly through the porous medium under the surface of the bed planted with emergent vegetation to the outlet, where it is collected before leaving via a water level control structure (Vymazal et al., 1998). During passage the wastewater comes into contact with a network of aerobic, anoxic and anaerobic zones. Most of the bed is anoxic/anaerobic due to the permanent saturation of the beds. The aerobic zones occur around roots and rhizomes that leak oxygen into the substrate (Brix, 1987). The most important properties of macrophytes planted in HSSF CWs are filtration bed insulation during the winter, substrate for the growth of attached bacteria, oxygen release to the rhizosphere, nutrient uptake and storage, C sequestration and root exudates with antimicrobial properties (Brix, 1997; Vymazal and Kröpfelova, 2008). HSSF CWs are commonly sealed with a liner to prevent seepage and to ensure controllable outflow, and are mostly used for secondary treatment of domestic and municipal wastewater (Vymazal and Kröpfelova, 2008). Organic compounds are degraded by bacteria under aerobic and anaerobic conditions. It has been shown that the oxygen transport capacity in these systems is insufficient to ensure aerobic decomposition and that anaerobic processes play an important role in HSSF CWs (Vymazal and Kröpfelova, 2008). Suspended solids settle into micropockets in the filtration bed or are filtered out. Removal of ammonia-N is limited by the lack of oxygen and hence nitrification in the filtration media. The HSSF CWs do, however, provide suitable conditions for denitrification (Vymazal and Kröpfelova, 2008). Removal of P is usually low unless special media with high sorption capacity are used. The selection of filtration material is also very important for the longevity of the system, because media that are too fine will clog the system, and surface runoff will occur (Vohla et al., 2011).

Vertical subsurface flow CWs comprise a flat bed of graded gravel topped with sand or other porous filter materials planted with macrophytes. In contrast to HSSF CWs, VSSF CWs are fed intermittently with large batches, thus flooding the surface. Wastewater then percolates down through the bed and is collected by a drainage network at the bottom. The bed drains completely, which allows air to refill the bed. The VSSF CWs provide greater oxygen transfer into the bed, thus producing a nitrified (high NO_3^-) effluent (Cooper et al., 1996; Cooper, 2005). On the other hand, VSSF CWs do not provide suitable conditions for denitrification to complete conversion to gaseous nitrogen forms which escape to the atmosphere. Removal of organics and suspended solids is high (Vymazal and Kröpfelova, 2008). As compared to HSSF CWs, which need 5–6 m^2 per population equivalent (PE), vertical flow systems require less land, usually 1–3 m^2 PE^{-1} (Cooper, 2005).

Both VSSF and HSSF CWs with the ability to insulate the surface of the bed are capable of operation under colder conditions than are FWS systems (Mander and Jenssen, 2003).

Various types of CWs are usually combined (i.e., hybrid or combined systems) in order to achieve higher removal efficiency, especially for nitrogen. The design commonly consists of two stages: several parallel VSSF beds followed by two or three HSSF beds in series (Vymazal, 2007). The VSSF wetland is intended to remove organics and suspended solids and to provide nitrification, while denitrification and the further removal of organics and suspended solids occur in the HSSF wetland. When aquatic macrophyte production is the main practical function of a wetland system, the VSSF–HSSF bed complex can be followed by a larger FWS wetland (Maddison et al., 2009).

As a bias of the water purification, the CWs for wastewater treatment have been found to be sources of greenhouse gases (GHG). Carbon dioxide (CO_2) emission has been measured in few full-scale CWs (Mander et al., 2003, 2005a,b, 2008; Teiter and Mander, 2005; Liikanen et al., 2006; Ström et al., 2006; Garcia et al., 2007; Picek et al., 2007; Van der Zaag et al., 2010), and C balance has been compiled in only one HSSF CW based on the long-term direct measurement of C in inflow and outflow, accumulation in filter material (sand), microbes, above ground and below ground plant biomass, and the emission of CO_2 and CH_4 (Mander et al., 2008). On the other hand, there are more measurements of CH_4 and N_2O emission from full-scale CWs: CH_4 by Tanner et al. (1997), Xue et al. (1999), Johansson et al. (2004), and Chiemchaisri et al. (2008); N_2O by Fey et al. (1999) and Johansson et al. (2003); and both CH_4 and N_2O by Tai et al. (2002), Wild et al. (2001), Mander et al. (2003, 2005a,b, 2008, 2011), Stadmark and Leonardson (2005), Teiter and Mander (2005), Liikanen et al. (2006), Søvik et al. (2006), Gui et al. (2007), Picek et al. (2007), Søvik and Kløve (2007), Ström et al. (2006), Liu et al. (2009), and Van der Zaag et al. (2010).

Recent research has shown that N_2O can be produced through a number of different pathways, both chemical and biochemical, during nitrification (stepwise conversion of ammonia to nitrate) and denitrification (stepwise conversion of nitrate to nitrogen gas; Colliver and Stephenson, 2000). Under aerobic conditions in a nitrifying wastewater treatment system, N_2O production through nitrifier denitrification has been identified as the predominant production pathway (Wunderlin et al., 2013; Aboobakar et al., 2013). Similarly, research from soil science has shown that in well-aerated, moist conditions (soil water filled pore space at 40–60%), N_2O can be emitted during nitrification (Robertson and Tiedje, 1987; Mosier, 1998; Mosier et al., 1998) by ammonia-oxidizing bacteria during the oxidation of hydroxylamine (NH_2OH) to nitrite (NO_2^-) (Arp and Stein, 2003), and also via reducing NO_2^- to N_2O and N_2 under aerobic conditions by nitrifier denitrification (Goreau et al., 1980; Wrage et al., 2001).

Denitrification, as the microbial reduction of NO_3—N to NO_2—N and further to gaseous forms of NO, N_2O and N_2 (Knowles, 1982), has been found in numerous studies to be a significant process in nitrogen removal in treatment wetlands (Bachand and Horne, 2000a,b; Spieles and Mitsch, 2000; Hernandez and Mitsch, 2006, 2007; Batson et al., 2012). Denitrification rates in soils are influenced by nitrate availability, carbon availability, temperature and pH (Mitsch and Gosselink, 2007). The last step of denitrification, i.e., the conversion of N_2O to N_2, is very sensitive to oxygen and redox status, and disruption of this step results in incomplete denitrification and N_2O emissions (Colliver and Stephenson, 2000). The relative contribution to N_2O emissions from a treatment system will depend on the environmental conditions that are generated and maintained throughout the pollutant transformation processes. Both denitrification and methane formation depend on the oxygen and redox status of the soil or sediment, which changes in both spatial and temporal contexts. In this relation, the variability of fluxes of both N_2O and CH_4 is high (Willison et al., 1998; Teiter and Mander, 2005).