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The impact of a pulsing water table on wastewater purification and greenhouse gas emission in a horizontal subsurface flow constructed wetland

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ABSTRACT

Artificial pulsing of water table is often used in horizontal subsurface flow (HSSF) constructed wetlands (CW) to enhance removal of BOD, COD, NH_4 and total N utilization by bacteria. In 2008–2010 and during the experimental session in October–November 2012 we studied the impact of a fluctuating water table on the water purification efficiency and GHG emissions in the 216 m^2 HSSF bed of a hybrid CW commissioned in 2002 in Paistu, Estonia. For comparison, water purification data from a previous study (Öövel et al., 2007) have been used. Isotopologue studies have been conducted in order to distinguish between denitrification and nitrification as source processes of N_2O.

Short-term (one month) and short-range (up to 35 cm) fluctuations in the water table enhanced CO_2 and N_2O emission whereas the median values varied from 0.5 to 246 μg CH_4-C $m^{-2}\,h^{-1}$ and 1.5 to 8.3 μg N_2O-N $m^{-2}\,h^{-1}$, respectively. There was a significant ($p < 0.05$) negative correlation between water table depth and CO_2 ($R^2 = 0.53$) and N_2O ($R^2 = 0.35$) emissions. Due to the plant cover disturbance in the outflow section of the HSSF bed, CH_4 emission did not show any significant difference between the high and low water table phase, and the correlation between water table depth and CH_4 emission was positive.

The impact of pulsing water table on water purification was minor: high water table enhanced the TOC removal and inhibited the TN removal, whereas the TP concentrations showed remarkable variations but no significant differences between the inflow and outflow values. Likewise, pH and dissolved O_2 values, and NH_4^+-N removal were not significantly influenced by water table manipulations.

The value of $\delta^{15}N^{bulk}N_2O$, $\delta^{18}O$-N_2O and site preference (SP) of N_2O in water samples varied from −2 to 32 ‰, between 41 and 78 ‰, and from 15 to 41 ‰, respectively. There was a significant positive correlation between the $\delta^{18}O$-N_2O vs SP N_2O values ($R^2 = 0.77$). This correlation, the range of isotopologue values and relatively low N_2O emission corroborate that the main source of N_2O fluxes in the studied HSSF CW bed is denitrification.

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1. Introduction

Due to increasing human impact on the global environment, the emission of the main greenhouse gases (GHG) carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) has shown rapid growth, being the main factor in recent global warming (Crowley, 2000). N_2O, which has an atmospheric lifetime of about 120 years, a global warming potential (GWP) of 296 relative to CO_2 over a 100-year time horizon, and is responsible for about 6% of anticipated warming (IPCC, 2007), is increasing in the atmosphere at a rate of about 0.3% $year^{-1}$ (Lashof and Ahuja, 1990). Methane in the atmosphere has a lifetime of 8.4 years on a 100-year time horizon. The GWP of CH_4 is 25 relative to CO_2, and CH_4 is responsible for about 20% of anticipated warming (IPCC, 2007).

Artificial fluctuations of water table are often used in constructed wetlands (CW). In particular, in vertical subsurface flow (VSSF) CWs intermittent loading increases the water purification efficiency (Kadlec and Knight, 1996; Langergraber

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et al., 2008). In created riverine wetlands (CRW), pulsing regimes significantly enhance sedimentation (Nahlik and Mitsch, 2008) and nutrient removal (Fink and Mitsch, 2007). Water level fluctuations are also used in horizontal subsurface flow (HSSF) CWs to enhance the removal of BOD, COD, NH_4^+-N and TN (Vymazal and Masa, 2003)

Little is known of the impact of fluctuating water table on GHG emissions from CWs – the bias of water purification in CWs (Mander et al., 2014a,b,b). In all cases, however, the pulsing hydrological regime clearly decreases methane emission. This has been found for FWS CWs and CRWs (Altor and Mitsch, 2006, 2008). Regarding N_2O emission, some studies show enhanced N_2O release from VSSF CWs due to pulsing hydrology (Jia et al., 2011; Mander et al., 2011). Some other studies on created riverine wetlands, however, suggest that pulsing hydrology slightly decreases N_2O emission (Hernández and Mitsch, 2006, 2007). Mander et al. (2011) have found that the lower water table level in the HSSF bed of a hybrid CW caused a significant increase in CO_2 and N_2O emission and a decrease in CH_4 emission. However, no systematic experiments have been performed in HSSF CWs to study the impact of a fluctuating water table.

Nitrous oxide emission from CWs can range from 90 to 130 $\mu g\ N_2O\text{-}N\ m^{-2}\ h^{-1}$ (median values) whereas no significant differences have been found between different types of CWs (Mander et al., 2014b). N_2O can be produced through a number of different pathways, both chemical and biochemical, during denitrification (stepwise conversion of nitrate to nitrogen gas; Reddy and DeLaune, 2008) and nitrification (Robertson and Tiedje, 1987) by ammonia-oxidizing bacteria during the oxidation of hydroxylamine (NH_2OH) to nitrite (NO_2^-) (Arp and Stein, 2003), by ammonia-oxidizing archaea (Stieglmeier et al., 2014), and also by reducing NO_2^- to N_2O and N_2 under aerobic conditions by nitrifier denitrification (Wrage et al., 2001). 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).

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, 1992), has been found in numerous studies to be a leading process in nitrogen removal in CWs (Spieles and Mitsch, 2000; Hernández and Mitsch, 2006, 2007). Denitrification rates in wetlands are influenced by nitrate availability, carbon availability, water depth, temperature, plant species and pH (Mitsch and Gosselink, 2007). The last step of denitrification, i.e., the conversion of N_2O to N_2, which depends on the abundance and expression of the *nosZ* gene in sediments (Garcia-Lledo et al., 2011; Ligi et al., 2014a,b), is very sensitive, and disruption of this step results in incomplete denitrification and N_2O emissions (Colliver and Stephenson, 2000).

Apportioning N_2O to its source processes is still a challenging task (Well et al., 2012). Several researchers suggest that the information obtained from measuring the intramolecular distribution of ^{15}N on the central (α) and the end (β) position of the linear N_2O molecule (i.e., the site preference of N_2O) is crucial for a better understanding of the apportioning of N_2O between nitrification and denitrification, but also to estimate N_2O reduction (Toyoda and Yoshida, 1999; Toyoda et al., 2011; Sutka et al., 2006). The N_2O site-specific ^{15}N signatures from bacterial denitrification and the NH_2OH-to-N_2O pathway of nitrification have been shown to be clearly different, making this signature a potential tool for N_2O source identification (Toyoda and Yoshida, 1999; Bol et al., 2003; Sutka et al., 2006; Well et al., 2005; Cardenas et al., 2007; Jinuntuya-Nortman et al., 2008). The majority of studies have been dedicated to the analysis of the $\delta^{15}N$ and $\delta^{18}O$ isotopomers ($\delta^{15}N_\alpha$ and $\delta^{15}N_\beta$) values of produced N_2O (Bol et al., 2003; Cardenas et al., 2007; Jinuntuya-Nortman et al., 2008; Toyoda et al., 2011), while there are only a limited number of studies dedicated to the analysis of dissolved N_2O in groundwater (Well et al., 2005, 2012; Koba et al., 2009).

The main objectives of the study are: (1) to estimate the effect of short-term water table fluctuations on the water purification efficiency and GHG emissions in the HSSF bed of a hybrid CW treating domestic wastewater, (2) to determine the source process of N_2O using stable isotope signatures of N_2O, and (3) to evaluate, first time for CWs, site-specific $\delta^{15}N$ of N_2O (SP) as an indicator of N_2O from denitrification in water-saturated ecosystems.

We hypothesize that (a) due to the change in the redox conditions in the filter bed, water table lowering will decrease CH_4 emission but increase gaseous losses of CO_2 and N_2O, and (b) the main source of N_2O before the water manipulation experiment started was denitrification.

2. Materials and methods

2.1. Study sites

The Paistu hybrid wetland system (constructed in 2002; 58°14′30.62″N, 25°35′341.77″E) treats the wastewater of 140 people (about 64 PE) and consists of a two-chamber VSSF filter bed (12 m × 18 m) and a 216 m^2 HSSF filter bed. The latter has a depth of 0.9 m and is filled with 2–4 mm of light-weight aggregates (LWA) and covered with reed (*Phragmites communis*) and few nettles (*Urtica dioica*) (see Öövel et al. (2007) for a detailed description).

During the previous study on performance of the Paistu hybrid wetland system, 18 water samples, once per month from October 2003 to October 2005 have been taken. Months with almost zero inflow (July and September) have been skipped. The water table in the HSSF bed varied from −5 to −40 cm, according to inflow hydraulic load (4.1–17.7 $m^3\ d^{-1}$). The whole system demonstrated an outstanding purification effect: for BOD_7 the average purification efficiency is 91%; for total suspended solids (TSS) 78%, for total P (TP) 89%, for total N (TN) 63%, and for NH_4-N 77% (Öövel et al., 2007).

In our recent paper we use data on HSSF bed from 2003–2005 study whereas all the later analyses from 2008–2010 and 2012 have been made in the HSSF bed too.

2.2. Experimental set up

The Paistu CW treats schoolhouse wastewater which, as is typical for schoolhouses, shows significant changes in discharge on both diurnal and annual scales: it is 7.4 $m^3\ d^{-1}$ on average, and fluctuates from 0 (at night and in the summer vacation period from June to September) to 17.7 $m^3\ d^{-1}$ (Öövel et al., 2007). Due to changing loadings (higher from September to May and lower in the school vacation from June to August), water table depth in the HSSF bed varied from 0 to −60 cm during the GHG sampling session in 2008–2010. Likewise, in 2012, before the short-term manipulation the water table rose from −35 cm at the beginning of July to −20 cm in the middle of September. This was the second part of the school vacation and the beginning of a new school semester, and thus, regarding the following experimental manipulation, we call this a stabilization period (Fig. 2). On September 26th we first artificially changed the water level, changing the height of the stand-pipe in the outflow well. In the following two months, we raised and lowered the water level three times, resulting in water depth variation of between +4 and −25 cm (Fig. 2).

The estimated hydraulic loading rate (HLR) to the HSSF bed during the stabilization period was approximately 20 $mm\ d^{-1}$ and varied between 25 and 28 $mm\ d^{-1}$ and 22–52 $mm\ d^{-1}$ during the high and low water table phases, respectively.

Precipitation data originate from the Viljandi weather station (58°22′40″N, 25°36′01″E). During the stabilization period (1st

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