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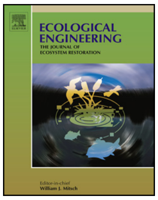

Solute transport and nitrate removal in full-scale subsurface flow constructed wetlands of various designs treating agricultural drainage water

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ABSTRACT

Subsurface flow constructed wetlands (SSF-CWs) consisting of woodchip based filter matrixes are promising measures targeting agricultural N loss via subsurface tile drains. Optimization of these systems may include the selection of appropriate hydraulic designs (i.e. horizontal and vertical flow), which may affect the solute residence time. In this study a bromide tracer experiment was performed in three full scale SSF-CWs, consisting of a woodchip-based filter matrix. Three different hydraulic designs (horizontal (H), vertical upward (V_{up}) and vertical down flow (V_{down})) and two flow rates were investigated (0.49 and 1.83 $L\,s^{-1}$). Additionally, batch experiments investigating the intra-granular diffusion into woodchips using two tracer solutes (tritium and bromide) were carried out. Non-equilibrium solute transport, including a mass exchange between a mobile and an immobile domain, was found in all SSF-CWs. The V_{up} demonstrated the most pronounced non-equilibrium and the lowest N removal rate. In contrast the largest N removal rate was observed in the V_{down}. The higher NO_3 removal rates were attributed to a longer solute residence time. Tailing of the tracer BTC indicated the influence of diffusive exchange in solute residence time, and this was further supported by the intra-granular diffusion of tracer solutes. Generally, the results suggested the vertical downwards SSF-CW as the best performing SSF-CW in terms of solute transport behaviour and N removal efficiency.

1. Introduction

Agricultural nitrogen (N) losses are recognized as the single major source of N to surface waters (Carpenter et al., 1998; Kronvang et al., 2009), driving eutrophication and consequently the degradation of water quality and loss of biodiversity (Carpenter et al., 1998). Agricultural subsurface tile drains constitute a major transport pathway (Dinners et al., 2002), connecting fields with receiving waters and thus, allowing the direct transport of NO_3-N to surface waters (Fausey et al., 1995). Promising strategies to mitigate agricultural subsurface drainage losses of N include the implementation of subsurface flow constructed wetlands (SSF-CW) containing a reactive filter media as part of the tile drainage structure or as end of pipe solutions (Schipper et al., 2010; Robertson, 2010; Blowes et al., 1994).

Nitrate reduction via denitrification is generally recognized as the dominant mechanism controlling NO_3-N removal in reactive granular woodchips based filter media (Gibert et al., 2008; Greenan et al., 2006). Results from laboratory experiments showed that woodchips based filters are characterized by a dual pore structure (Bruun et al., 2016a; Cameron and Schipper 2010; Robertson, 2010) and non-equilibrium solute transport (Bruun et al., 2016a; Cameron and Schipper, 2012; Herbert, 2011). The non-equilibrium solute transport can be conceptualized by the mobile and immobile approach (Van Genuchten and Wierenga, 1976), with convective mass transfer in the mobile domain, and mass transfer between domains restricted to diffusion (Van Genuchten and Wierenga, 1976). Non-equilibrium solute transport in woodchips media have been attributed to variations in pore water velocities and/or inter or intra-granular diffusion (Bruun et al., 2016a; Cameron and Schipper, 2012). Diffusive mass exchange into the immobile domain increase the solute residence time and can affect the reaction rate of reactive solutes (e.g. NO_3) (Bruun et al., 2016a).

In the highly porous woodchips, diffusion of solutes into the intra-granular pore space affects the transport of both non-reactive

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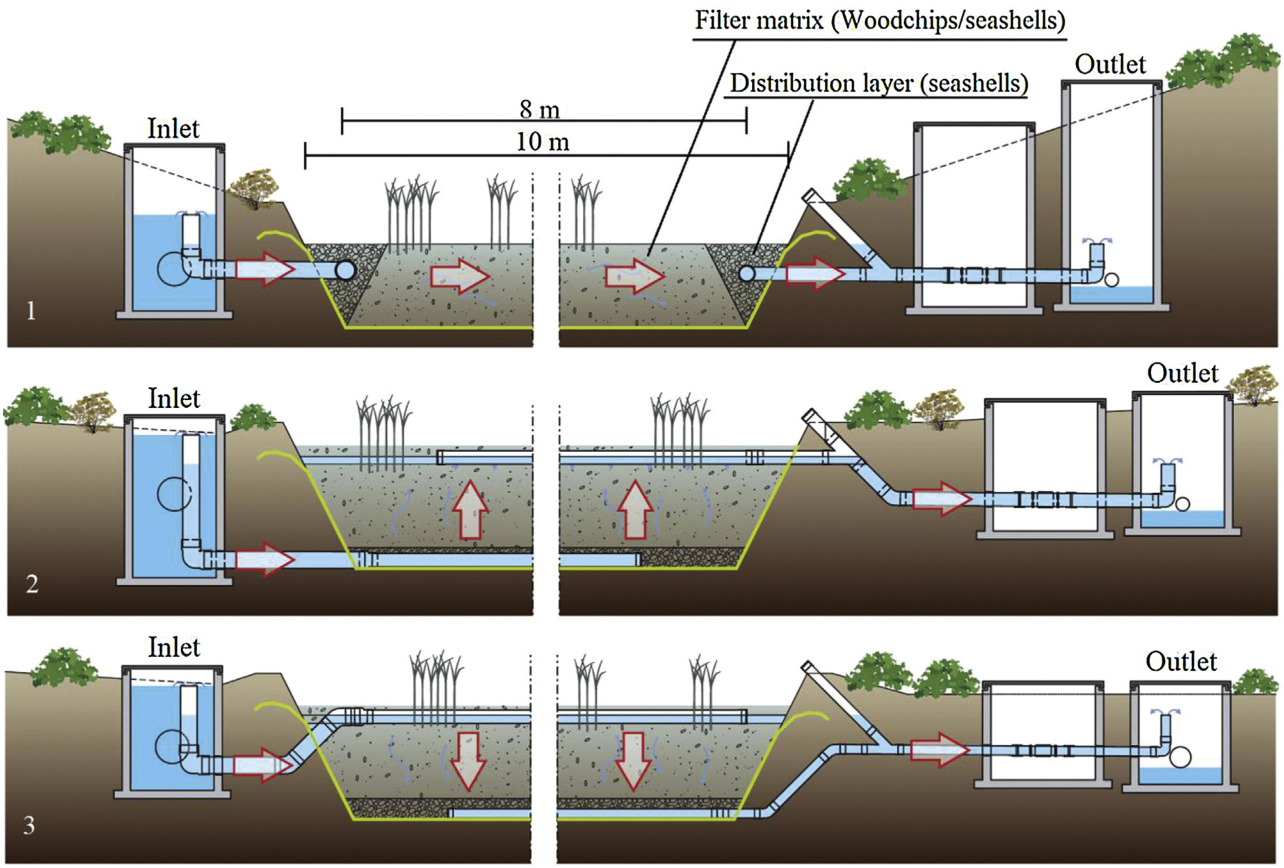

Fig. 1. Principal sketch of subsurface flow constructed wetlands (SSF-CWs) with horizontal flow (1), vertical upwards flow (2) and vertical downwards flow (3). SSF-CW components include inlet wells, perforated distributions pipes, coarse granular distribution/collection layer, woodchips:seashell filter matrix, outlet well with electromagnetic flow-meter and outlet well with effluent sampling and aeration of the effluent water.

(tracers) and reactive (e.g. NO_3) solutes. The effect of intra-granular diffusion on the solute transport of anions (e.g. NO_3 or Br), may be limited due the electrostatic repulsion of anions (anion exclusion) by negatively charged surfaces on the woodchips, resulting from the oxidation of organic matter (e.g. polyanions created under oxidation of lignin (Hui et al., 2003)). Thus, anion exclusion may affect the observed hydraulic performance of woodchip-based systems.

The hydraulic efficiency (λ) of SSF-CWs has been used to evaluate the solute transport (e.g. García et al., 2004; Masbough et al., 2005). In horizontal SSF-CWs the λ have been found to increase with the aspect ratio, which is defined at the length: width ratio (García et al., 2004). Increasing the aspect ratio by one unit, increases the normalized delay time (tracer delay time/HRT) by a factor of 1.6–1.7 and decreases the dispersion number by 0.3–0.4 (García et al., 2004), which further more increases the hydraulic efficiency (García et al., 2004).

In SSF-CWs with different hydraulic design (horizontal, vertical upward and vertical downward flow), the annual N removal efficiencies have been found to be higher when applying a horizontal flow as compared to the vertical flow hydraulic design (Hoffmann and Kjaergaard, 2015). Horizontal flow SSF filters, may provide the highest aspect ratio, whereas vertical flow SSF filters due to the dimensions have a larger cross sectional area for the infiltration water and a shorter flow path, and thus a smaller aspect ratio. However, Hoffmann and Kjaergaard (2015) did not investigate the link between the internal solute transport and the NO_3 removal.

The objective of this study was to investigate the solute transport in SSF-CWs of three hydraulic designs (horizontal, vertical upward, and vertical downward flow) and link the hydrological flow paths and the NO_3 removal efficiency.

We hypothesized that: i) the vertical flow hydraulic designs would demonstrate the most pronounced non-equilibrium solute transport, ii) the NO_3-N removal rate would be highest in the horizontal flow SSF-CW, and iii) that solutes can diffuse into the intra-granular pores of woodchips.

To test these hypothesis three woodchip-based SSF-CWs, having different hydraulic designs (horizontal (H), vertical upward (V_{up}) and vertical downward flow (V_{down})) where investigated. Bromide (Br) was used as a tracer, at two different flow rates. Additional batch experiments were conducted to investigate if intra-granular diffusion of anions occurs in woodchips.

2. Materials and methods

2.1. Field site and subsurface flow constructed wetland

Tracer experiments were conducted in 3 full-scale SSF-CW, located in Skannerup 56.214132–9.742723 N/E, Jutland (DK) as part of a tile drainage system covering an agricultural catchment of 85 ha. The SFF-CWs were constructed in 2012 by excavating 6 SSF-CWs with dimensions 10 m wide, 10 m in length and 1 m deep (Hoffmann and Kjaergaard, 2015). The excavated SSF-CWs were lined with an impermeable membrane (Junifold PE HD GEO MEMBRANE 1.0 mm; Millag, Denmark) to avoid infiltration/exfiltration of water to/from the SSF-CWs. Three different hydraulic designs allow either horizontal (H), vertical upwards (V_{up}) or vertical downwards (V_{down}) flow (Fig. 1). Drainage water was directed to each SSF-CW by individual drainage wells, supplied from the same drainage inlet pond. A distribution layer consisting of coarse Seashells size 8–32 mm (Danshells A/S, Denmark) was placed in the