



## Phosphorus accumulation and stability in sediments of surface-flow constructed wetlands



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### ABSTRACT

Surface-flow constructed wetlands (CWs) are systems that replicate the nutrient reduction processes of natural wetlands and are considered suitable measures to reduce the nutrient losses from agricultural drainage discharge to surface waters. The present study aimed to investigate the biogeochemical stability of phosphorus (P) in three CWs. Intact soil cores were collected from each CW zone and included the soil and the sediment layer (average 28 mm thickness) formed by deposition of the incoming sediments with the water flow. Soil and sediment layer were analysed geochemically, including bulk density, pH, and total contents of carbon, nitrogen, P as well as different extracts of iron (Fe), aluminium and P. In situ measurements of soil redox potential, dissolved oxygen concentration and temperature were carried out in the CWs during 16 months. Grab water samples were collected across the CWs for determination of P species, total Fe, sulphate and pH. Soil core analysis revealed that the major P storage in the CWs was attributed to retention of Fe-bound P in the sediment layer. The Fe and total P (TP) retention differed significantly between CWs ( $11\text{--}63\text{ g Fe m}^{-2}\text{ yr}^{-1}$  and  $1\text{--}9\text{ g TP m}^{-2}\text{ yr}^{-1}$ ). Amorphous Fe oxides constituted the major P sorbent fraction in the soil and sediments of all CWs, and the higher P sorption capacity in the sediment layer demonstrated the continuous supply of P sorbents to this layer. The stability of Fe-bound P in the anaerobic sediments seemed to be controlled by the high molar ratios of bicarbonate-dithionite extractable Fe ( $\text{Fe}_{\text{BD}}$ ) to associated P, which varied from 21 to 49, and the presence of an aerobic sediment-water interface.

### 1. Introduction

Surface-flow constructed wetlands (CWs) for the treatment of agricultural drainage water are considered an environmentally friendly (Baker, 1992; Koskiahio et al., 2003) and cost-effective (Gachango et al., 2015) alternative to minimize the load of nutrients to downstream surface waters. One of their main functions is to mitigate the loss of phosphorus (P) to the environment, which is a worldwide concern (Heathwaite et al., 2000; Withers et al., 2000; Withers and Haygarth, 2007). Phosphorus is usually the limiting nutrient in freshwater systems, which can lead to eutrophication under high loads (Correll, 1998; Schindler, 1977) and cause severe changes to the ecosystem (Khan and Ansari, 2005).

Several studies have demonstrated that CWs may work as P sinks (Braskerud, 2002; Johannesson et al., 2011; Kynkäänniemi et al., 2013; Maynard et al., 2009a), but can also become P sources (Kovacic et al.,

2000; Tanner et al., 2005; Tanner and Sukias, 2011). This calls for a better understanding of the P biogeochemical cycle in CWs. They receive P from agricultural drainage water as both particulate and dissolved P in different forms (Grant et al., 1996). Thus, P retention occurs through a number of mechanisms, including sedimentation of particulate P, soil adsorption and plant uptake of dissolved P, and accretion of new sediments (Kadlec, 2005). The organic P stored in the plants is generally considered a short-term storage, as part of the P taken up by plants can return to the sediments through litter fall (Reddy et al., 1995). The CW sediments, thus, represent the main storage of P, resulting from the continuous sedimentation of particulate P, sorption of dissolved P forms and accumulation of organic P (Kadlec and Wallace, 2008). However, sediment-bound P can be transformed into dissolved forms through different biogeochemical processes (James et al., 2002; Uusitalo et al., 2003). The characterization of different P forms in the sediments allows an assessment of the potential for P mobilization

**Abbreviations:** BD, bicarbonate-dithionite; CBD, citrate-bicarbonate-dithionite; CW, surface-flow constructed wetland; ox, oxalate; SWI, sediment-water interface; DO, dissolved oxygen concentration; Eh, soil redox potential converted to standard hydrogen electrode potential;  $\text{Fe}_{\text{tot}}$ , total iron; DRP, dissolved reactive phosphorus

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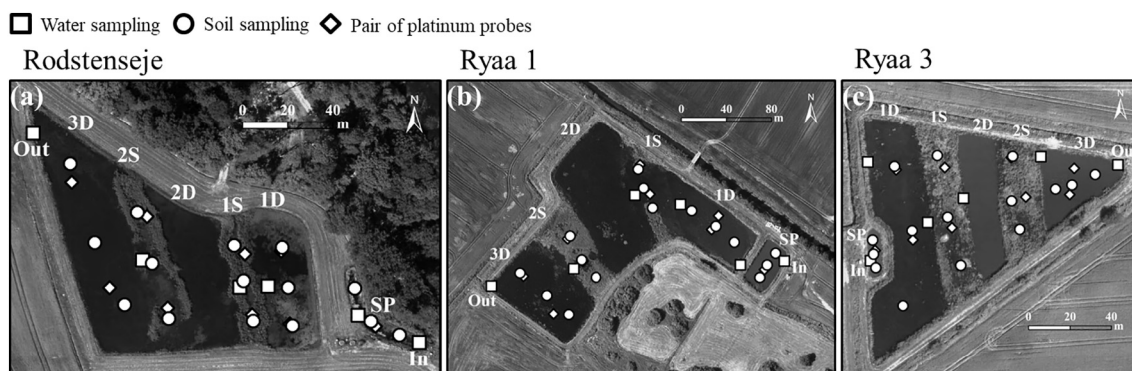


Fig. 1. Ortho photos of the surface-flow constructed wetlands Rodstenseje (a), Ryaa 1 (b) and Ryaa 3 (c), showing the sedimentation pond (SP), deep (1D, 2D and 3D) and shallow (1S and 2S) zones. The spots of water and soil samplings as well as soil redox potential measurements are marked with symbols. In and Out: inlet and outlet.

under anaerobic conditions (Forsmann and Kjaergaard, 2014; Hoffmann et al., 2009).

Phosphorus sorption capacity in soils has generally been attributed to the content of amorphous iron (Fe) and aluminium (Al) oxides (Dunne et al., 2005; Reddy et al., 1998; Richardson, 1985). Under certain conditions, biogeochemical processes in anaerobic CW sediments may favour the reduction of ferric Fe(III) to ferrous Fe(II), with consequent release of dissolved Fe and P to the pore water (Heiberg et al., 2010; Kjaergaard et al., 2012; Scalenghe et al., 2002). It is documented that P bound to redox sensitive Fe constitutes a potential source of P diffusing across the sediment-water interface (SWI) into the water phase (Fisher and Reddy, 2001; Pant and Reddy, 2003). Phosphorus release rates from CW sediments are controlled by the concentration differences across the SWI (Dunne et al., 2006; Reddy et al., 1999). Several studies have related P release, or pore water P concentrations, to soil geochemical parameters, e.g. the degree of P saturation (Hooda et al., 2000), molar ratios of total Fe to P (Geurts et al., 2008; Jensen et al., 1992), molar ratios of amorphous Fe oxides to P bound to amorphous Fe and Al oxides (Forsmann and Kjaergaard, 2014; Surridge et al., 2012) and molar ratios of the most redox sensitive Fe fraction to the associated P (Forsmann and Kjaergaard, 2014; Jensen and Thamdrup, 1993; Zak et al., 2010).

In addition, seasonal variations in soil redox conditions may regulate the biogeochemical transformation of Fe-bound P. The availability of carbon sources (Kjaergaard et al., 2012), temperature (Zhou et al., 2010) and the supply of terminal electron acceptors, i.e.  $O_2$ ,  $NO_3^-$  and  $SO_4^{2-}$  (Kjaergaard et al., 2012; Lamers et al., 2002; Shenker et al., 2005), affect the soil redox conditions. Vegetation can also have an effect through the addition of carbon to the sediments and oxygen supply into the rhizosphere (Stottmeister et al., 2003). Thus, predicting the net effect of P biogeochemical cycling in CWs can be rather difficult.

The present study aimed to evaluate the stability of P in the soil and sediment layer of three CWs after ~3–5 years of operation. It was hypothesized that the stability depends on the P saturation indexes and redox stability of P sorbents, as well as the seasonal variations in soil redox conditions. The specific objectives were (i) to determine the spatial variation in geochemical characteristics, P forms and P saturation indexes in the soil and sediment layer across different CW zones, from the basin receiving the inlet water to the last zone discharging the water into the outlet pipe, (ii) to estimate the total P (TP) accumulation in the sediment layer based on soil core samplings, and (iii) to investigate the spatio-temporal variation in redox dynamics and subsequent Fe and P dynamics.

## 2. Materials and methods

### 2.1. Sites description

Three Danish CWs receiving agricultural drainage water were selected for this study. The CW Rodstenseje is situated in a clay moraine landscape in the Norsminde Fjord catchment (55°57'23" N, 10°09'32" E). Rodstenseje was constructed in a clay loam field in the autumn 2010 and operation started in March 2011. It has a surface area of 8950 m<sup>2</sup> (1.1% of the drainage catchment area) and receives drainage water from an upland clay loam field of approximately 80 ha. A monitoring period from August 2013 to July 2016 (Mendes et al., 2018) revealed that the monthly average air temperature ranged from 0.1 to 19.8 °C and that annual precipitation ranged from 0.7 to 0.9 m yr<sup>-1</sup>. In addition, annual hydraulic loading rates ranged from 9 to 28 m yr<sup>-1</sup>, while hydraulic residence time varied from a minimum of 4 days during the winter discharge (October to the end of March) to > 100 days during the summer discharge (April to the end of September).

The CWs Ryaa 1 and Ryaa 3 are both situated in the Littorina lowland plateau area in the Limfjorden catchment (57°12'55" N, 10°00'13" E and 57°08'34" N, 9°40'5" E). These CWs receive agricultural drainage water from pumped drainage catchments representing yoldia sand (Ryaa 1) and yoldia clay (Ryaa 3), and discharge the outlet water into the river Ryaa. Both CWs were constructed in spring 2013 and operation started in May (Ryaa 1) and July (Ryaa 3) 2013. The drainage catchment areas of Ryaa 1 and Ryaa 3 are 230 and 80 ha, and their surface areas are 21,190 and 8480 m<sup>2</sup> (0.9 and 1.1% of the drainage catchment area), respectively. The monitoring period from August 2013 to July 2016 (Mendes et al., 2018) revealed that monthly air temperature and annual precipitation ranged from -0.5–19.2 °C and 0.9–1.5 m yr<sup>-1</sup> in Ryaa 1, while in Ryaa 3 these were -0.5–19.4 °C and 0.8–1.3 m yr<sup>-1</sup>. In addition, the annual hydraulic loading rates in Ryaa 1 and Ryaa 3 ranged from 52 to 66 m yr<sup>-1</sup> and from 29 to 37 m yr<sup>-1</sup>, respectively, while hydraulic residence time varied from 1 to 12 days in Ryaa 1 and from 2 to > 100 days in Ryaa 3.

### 2.2. Design of the CWs

The CWs were constructed with the same principal design, consisting of a sedimentation pond (SP, ~1 m deep) followed by a second basin compartment with a sequence of three deep zones (1D, 2D and 3D, ~1 m deep) intersected by two shallow zones (1S and 2S, ~0.3 m deep), with some variations in compartment volumes (Fig. 1). Drainage discharge flows into the SP and is further distributed to the second basin compartment by either one (Rodstenseje), two (Ryaa 1) or three (Ryaa 3) connecting pipes. The shallow zones were planted with *Typha latifolia* and *Phragmites australis* during the construction of the wetlands. The area of each CW zone was measured in ArcGIS® software with GPS

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