



The role of aluminium and iron in phosphorus removal by treatment peatlands



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ABSTRACT

Wetlands are commonly used to treat phosphorus from the effluent of municipal wastewater plants after conventional treatment and wastewater from various diffuse sources, with good results. The long term phosphorous (P) retention capacity of wetland treatment systems is a key research question. This study examined phosphorus retention in wetland (peat) soil columns in order to clarify the role of aluminium (Al) and iron (Fe) concentrations in wastewater on P removal. Since Al and Fe in wastewater could be expected to increase P uptake by increasing peat sorption capacity, laboratory flow-through column experiments were run for almost 700 days in conditions replicating the natural conditions in treatment wetlands. The study set comprised 18 peat columns and five water types from different origins (municipal wastewater, peat extraction runoff, distilled water with phosphate solutions containing 0.1 or 0.4 mg PO₄³⁻ L⁻¹, and pure distilled water). To study retention of sudden P peak concentrations, a high P peak was injected into the columns after about 500 days of wastewater loading. The results clearly showed that Al and Fe in input water maintained P removal in peat soils, with Al form also affecting retention processes, and P saturation did not occur. Therefore constructed wetlands can in some cases be safely used without the risk of P saturation. Furthermore, in the high P peak test, the additional P was successfully retained in columns with accumulated metals, showing that artificial addition of Al can be used to increase P retention capacity in peat soils with low sorption capacity.

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1. Introduction

Phosphorus (P) is a major pollutant from different land uses and municipal wastewater treatment plants (Diebel et al., 2008; Son et al., 2013) and its negative impacts on aquatic environments are well understood (USEPA, 2000). Constructed wetlands (CW) are commonly used to treat wastewater and runoff from various diffuse and point sources, with good results (e.g. Silvan et al., 2004; Heal et al., 2005; Vymazal, 2011). However, the processes involved and the stability of the retained P in CWs, especially after long time loading and use, remain unclear for peat-based wetlands, which are commonly used in countries in the northern hemisphere such as Finland. Also influence of fluctuate inflow P concentration on purification performance is poorly know. This is typically a case as runoff from peat extraction, forestry or agriculture is at issue.

In CWs, P is removed from water through chemical precipitation, adsorption and absorption (Heikkinen and Ihme, 1995; Heikkinen et al., 2002; Aslan and Kapdan, 2006; Kadlec, 2006; Lee et al., 2010; Liang et al., 2010; Kõiv et al., 2010), and biological uptake by vegetation (Huttunen et al., 1996; Silvan et al., 2003) and micro-organisms (Chen et al., 2005). In treatment peatlands, where water table typically fluctuates from 0 to 30 cm above the peat surface, these processes occur in topmost “free water layer”, in the vegetation/poorly decomposed peat layer (acrotelm) or in the deeper peat layer (catotelm). Biological and physical processes take place in these two first mentioned layers, but in the deeper layer, chemical processes are dominating the removal. Since peat is organic soil, also humic substances have a role in removal processes but as far as they have no capacity to retain P, they can react with P and improve P removal as it has been discussed in some peatland studies (Heikkinen and Ihme, 1995).

Generally, different types of wetlands and sorption materials are widely studied and used (Vymazal, 2007; Vohla et al., 2010). Modified biomaterials (e.g. Carvalho et al., 2011) and industrial by-products (e.g. Klimeski et al., 2014) have been tested but their long-term performance for P removal is not well proved. Peat is

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abundant in the Boreal region offering a cost-efficient way to reduce environmental impact of diffuse and point sources.

Chemical sorption is considered to be one of the most important factors in P removal in CWs. In general, the presence of aluminium (Al), iron (Fe) and calcium (Ca) increases the P sorption capacity of filter materials (Karjalainen et al., 2003; Xu et al., 2006; Lai and Lam, 2009), especially oxalate-extractable Al and Fe (Yuan and Lavkulich, 1994; Arias et al., 2006). The importance of Fe and Al to P sorption is typically in acidic soils such as peat whereas Ca bound P is typical in alkaline conditions ($\text{pH} \geq 7$) (Reddy and DeLaune, 2008). Uptake of P by vegetation has also been noted in treatment peatlands (Huttunen et al., 1996; Uusi-Kämppe et al., 2000) and in peatland buffer areas (Silvan et al., 2004), but the role of vegetation in annual P retention differs depending on site. As biomass production is high in wetlands, accretion of peat can function as a major long-term P sink (Vymazal, 2007).

Previous studies have found that saturation of the wetland medium with P can restrict its lifetime and reduce purification efficiency (Heikkinen and Ihme, 1995; Arias et al., 2001), which can increase transport of nutrients to receiving water bodies. Due to this finite capacity to retain P, the life time of treatment peatlands has been estimated by laboratory tests to be around 20 years (Heikkinen et al., 1995). However, there is evidence that the amount of P retained in treatment peatlands can be significantly higher than the estimated maximum (Ronkanen and Kløve, 2009), indicating that the life time could exceed 20 years. However, P sorption capacity of highly loaded filter material or CW medium has not yet been studied. At the moment, we also lack a clear understanding of how metals in runoff waters and wastewaters affect long-term P retention processes in peat material.

To study P sorption capacity of filter material is well known to be complicated (Cucarella and Renman, 2009). One crucial factor is contact between filter material and the inflow water which is essential in chemical sorption (Heikkinen et al., 1995; Cucarella and Renman, 2009). Also hydraulic loading rate effect the P binding capacity of filters (Herrmann et al., 2013). This highlight the importance of keeping hydraulic and flow processes in laboratory tests as similar as possible to those in natural conditions. Considering peat as a filter material, the structure of the peat layer and its hydraulic properties (hydraulic conductivity, porosity etc.) must be maintained, as those are important for water flow. Most previous studies have used a batch test procedure, by shaking samples in artificial solutions (e.g. Heikkinen and Ihme, 1995; Gray et al., 2000; Karjalainen et al., 2003; Xu et al., 2006), or column tests for disturbed samples (Seo et al., 2005; Babatunde et al., 2009). In these studies conditions are not close to natural wetland conditions, which can affect the results obtained. There is therefore a clear need for laboratory studies on undisturbed samples. An advantage with using an artificial sample solution (such as KH_2PO_4) is that it easily provides eligible constant concentrations for the test. However, this approach does not take into account the natural chemical composition of water, which is often complex and can influence sorption processes, especially if metals are present.

In this study, column experiments were performed with undisturbed peat samples taken in the natural direction of water flow at the active flow depth in a treatment peatland treating peat extraction runoff for nearly 20 years. The flow velocity used in the experiments was set to be equal to the flow velocity in treatment peatland, in order to maintain natural flow conditions. As changes in environmental condition, such as temperature and pH, are known to influence biological processes and P sorption (e.g. Oliveira et al., 2015), these were kept constant. The column experiments were run for almost 700 days, providing a unique dataset on long-term peat retention processes. In addition, the performance of the peat columns was also studied during a sudden

high P concentration imitating extreme situation and clarifying differences between the study columns.

The main objective of the present study was to clarify P removal mechanisms in wetlands constructed on pristine peatlands. Four different influent sources were used in the columns and the P removal was analysed as regard the following research questions and hypotheses:

- i. What is the effect of wastewater and peat extraction runoff composition and the presence of chemical residues such as Al and Fe on the P removal capacity of peat? Our starting hypothesis was that Al and Fe present in the wastewater would maintain sorption in peat and prevent P saturation. We also hypothesised that P in peat extraction runoff containing Fe would be better retained than phosphate in distilled water.
- ii. What is the response of peat columns when loaded different types of water with a sudden high peak of P concentration? Our starting hypothesis in this case was that peat in columns which had high metal retention in the first experiment would retain more P.

2. Material and methods

2.1. Peat soil samples

A total of 18 undisturbed peat samples were taken from the Kompasuo treatment peatland ($65^{\circ}44'43''\text{N}$, $25^{\circ}57'80''\text{E}$) which had purified peat extraction runoff. The Kompasuo area, in the mid-boreal region of the southern aapa mire zone of Northern Finland, was drained for peat extraction in 1986–1989 and a wetland treatment is used to purify the runoff after two sedimentation ponds. Prior to sampling in 2005, the wetland had treated runoff from the extraction area of 50 ha for 18 years. The peat samples for the column experiment were taken from the top peat layer (acrotelm layer/active flow depth) of the wetland at a depth of 0.2 m. All samples were taken parallel to the natural flow direction in order to provide as natural conditions and peat structure as possible in the study. The samples were drilled directly into the column containers (\varnothing 10 cm, height 16 cm) which had a sharp lower edge in order to preserve the natural structure of the peat. The density of the peat samples ranged from 0.11 to 0.24 g cm^{-3} (mean 0.18 g cm^{-3}).

2.2. Column experiment procedure

Phosphorus and metal (Al and Fe) removal were studied by vertical columns with upward flow direction (Fig. 1). In preparation, 2.5 cm of peat soil at each end of the column was replaced with inert quartz gravel to ensure uniform water flow over the cross-sectional area. A filter paper ($0.45 \mu\text{m}$) was placed between peat and quartz to prevent clogging of peat by organic matter which is common problem in column tests. The filter paper also enabled to study removal of dissolved forms of elements which are most important forms to be transported through the peat layers and to be absorbed to the peat in the treatment peatlands. The columns were sealed at both ends and saturated with inflow water from the Kompasuo peatland before the experiments in order to keep initial conditions in the columns as similar as possible. A peristaltic pump was installed to pump one type of four test solutions namely (i) humic water, (ii) wastewater, (iii) a $0.1 \text{ mg PO}_4^{3-} \text{ L}^{-1}$ solution and (iv) a $0.4 \text{ mg PO}_4^{3-} \text{ L}^{-1}$ P solution to the inlet of three columns (Fig. 1). Two pump sets were used to pump pure Milli-Q water to the control columns with zero P concentration (v). Thus, there were three replicates for solutions (i–iv) and six replicates for Milli-Q water (v) (Table 1).

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