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Removal of organic pollutants from oak leachate in pilot scale wetland systems: How efficient are aeration and vegetation treatments?

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ABSTRACT

This study investigated the effects of aeration and/or vegetation in experimental constructed wetlands (CWs) as mesocosms on the removal of pollutants in oak wood leachate. Twelve outdoor wetland mesocosms, with randomized replicated treatment combinations of vegetation (*Phragmites australis*) and aeration was monitored during the second and third year after construction. The investigation included control tanks with no aeration and no vegetation. The parameters monitored were polyphenols (PPs), chemical oxygen demand (COD) and water colour. The reduction of COD after 28 days was approx. 50% and more than 50% of PPs, whereas only 40% of the water colour was removed. Aeration increased the effect of both COD and PP removal. The vegetation treatment had a small but significant effect on removal of COD. The vegetation + aeration treatment, as well as aeration alone, increased the removal efficiency of COD from 9.5 g m^{-3} d^{-1} in the control to 11 g m^{-3} d^{-1}. The results suggest that CWs can be used to treat stormwater contaminated by oak wood leachate. Further, it is suggested that the main processes for removal of pollutants in the leachate occur in the open-water habitat and that the hydraulic retention time is more important for removal than aeration and vegetation related processes.

1. Introduction

All over the world, wood is used as a building material and for the production of paper, textiles, fuel and bioenergy. Handling of wood material may have strong negative impacts on recipient waters depending on the species used and the scale of the operation (Zenaitis et al., 2002). Some impacts are caused by machinery, such as spillage and emissions from the handling sites (Orban et al., 2002), whereas others occur when wood is handled for biofuel (Zelikoff et al., 2002) or is processed into fractions creating hazardous particles (Kauppinen et al., 2006).

Wood leachate appears in the stormwater runoff from sites with outdoor storage of wood. In this paper, wood leachate refers to organic compounds originating once wood is exposed to water. This includes the runoff water that percolates throughout wood residues or logs and the water used for irrigation of timber.

The main problems for the recipient water bodies posed by wood handling areas are related to the high concentration of organic compounds with a low BOD/COD ratio (Tao et al., 2005), which are difficult to biodegrade (Zenaitis et al., 2002). Wood leachates contribute to brownification in recipient fresh waters, a trend in the northern hemisphere over recent decades of increasing water colour (Graneli, 2012). The effects include impaired light climate, reduced aquatic primary production, obstruction of drinking water purification and reduced recreational value of surface waters. Further, the leachates may cause anoxic conditions and are reported to have toxic effects on aquatic organisms (Hedmark and Scholz, 2008). The main problematic organic compounds include terpenes (Rupar and Sanati, 2005), resin acids (Samis et al., 1999) and tannins and lignin (T&L) (Bailey et al., 1999). The chemical composition of wood leachate is highly variable and is strongly species-specific. Leachate from oak wood has a high organic content and the colour of the water is correlated with high amounts of polyphenols (PPs) (Svensson et al., 2012). Reducing compounds are often measured in wood leachate by Folin's phenol reagent, correlated to gallic acid and then reported as T&L or

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polyphenols depending on the author (Zenaitis et al., 2002; Tao et al., 2005; Svensson et al., 2013).

Constructed wetlands (CWs) have been used for decades as a cost-efficient method to treat wastewater. Already in the 1950s, the reduction of phenolic components in CW was studied by Seidel at the Max Planck Institute in Germany (Vymazal, 2005). Recent research has been on the potential of CWs for the treatment of wood leachate (Masbough et al., 2005; Hedmark et al., 2009; Tao et al., 2006b). But it is still unclear what treatment is most appropriate. Alternative treatment options for wood leachate include soil–plant systems (Hedmark et al., 2010), trickling filters (Woodhouse and Duff, 2004), sand filters (Doig et al., 2007), peat-ash filters (Svensson et al., 2013) and ozone (Zenaitis and Duff, 2002; Zenaitis et al., 2002). The variability of leachates among wood species makes it difficult to draw general conclusions about the applicability of CWs compared with other treatment options. CW experiments seldom compare replicated treatments in balanced experimental designs, with appropriate controls (such as retention tanks without plants), which makes it difficult to draw robust conclusions based on statistics.

Treatment efficiency of CWs appear to be strongly related to retention time (e.g. Tao et al., 2006a) and other factors related to sedimentation processes (Doing et al., 2006). In principle, prolonged retention time facilitates sedimentation and biological degradation of pollutants. Microbial processes may occur in three habitats: (i) the water column (bacterioplankton); (ii) sediments (sediment bacteria); and (iii) vegetation (epiphytic bacteria). Tao and Hall (2004) showed that bacterioplankton was important for treatment effect of wood leachate in a wetland. Further, they demonstrated that sediment bacteria were responsible for nearly all uptake of organic matter, whereas the effect of epiphytic bacteria was negligible.

Emergent vegetation, such as the common reed (*P. australis*) or cattail (*Typha* spp.), is often used in CWs (Masbough et al., 2005; Tao et al., 2006a,b). Emergent plants are supposed to have several treatment effects in CWs. Plants provide surface substrates for attachment of microbes (epiphytic biofilm). Shade from the canopy reduces dissolved oxygen in the water by lowering the photosynthetic rate and increasing the respiration rate in the water column. Further, emergent species provide shelter from wind, which prevents resuspension of sediments and reduces the diffusion of oxygen from the atmosphere to the water column. In addition, many plants transport oxygen from the atmosphere to rhizomes and roots, and thereby aerate the sediment in the rhizosphere. Although several potential mechanisms suggest a functional role for emergent plants, their importance for treatment of wood leachate has been questioned (Tao et al., 2006b).

Wetlands are complex ecological systems, which require time to mature and stabilize. Therefore, evaluations of wetland functions, in treatment of wood leachate, may be biased by the short duration, and the usage of early successional stages, in most previous experiments. In CWs with emergent plants, it may take years before a fully developed root zone is established (Werker et al., 2002; Kadlec and Knight, 2008). Initially, the ecosystem changes quickly and very often algae appear as rapid colonizers before emergent vegetation closes the canopy (Kadlec and Knight, 2008). Increased spatial complexity, accumulation of litter and detritus, interact with microbiota, which causes successional changes in the ecological communities within the wetland. Dissolved oxygen is one example of a key factor that is affected by the successional stage (Hansson and Graneli, 1983; Duke, 2012).

Oxygen typically increases the degradation rate of organic matter; however, anaerobic degradation is common in wastewater treatment. Aeration is suggested to have a positive effect on degradation of wood leachate (Tao et al., 2007; Taylor et al., 1996; Zenaitis et al., 2002; Woodhouse and Duff, 2004).

Wood handling sites are common around the world and therefore, it is important to identify treatment methods to reduce pollutants transported by the water that runs off these sites.

In this study, we analyse the efficiency of CW for treatment of leachate from logs of pedunculate oak (*Quercus robur*) in an outdoor storage area. It is the first evaluation of CW for the treatment of oak leachate. More specifically, the present study assesses the effects of aeration and/or vegetation treatments on target indicators of potentially harmful organic substances in wood leachate. To do this, we constructed twelve outdoor wetland mesocosms in a randomized, replicated and fully balanced design with treatment combinations of vegetation and aeration. Water flow, temperature, pH, O_2 and target indicators were monitored over a period of two years, after a first year of vegetation establishment. Hence, one objective is to study changes in treatment efficiency during a relatively long period of maturation compared to previous studies. The target indicators were polyphenols (PPs) and compounds represented by chemical oxygen demand (COD) and water colour.

2. Material and methods

2.1. Experimental set-up

The experiment was performed in a log yard (ca. 6000 m^2) in southeast Sweden used for irrigation of oak wood and storage of oak woodchips. Most of the logs stored in the area are pedunculate oak (*Q. robur*). During storage, logs are continuously irrigated to preserve quality and prevent dry cracks. The irrigation water is recirculated in order to save extracted groundwater. Additionally, stormwater from the industrial area is collected and also used for irrigation. The area has a drainage system designed for collection and recirculation of water. Drainage water from irrigation and precipitation is collected in a collection pond (estimated 350 m^3) from which water is pumped to a second storage pond (estimated 700 m^3) and used for irrigation. Groundwater is pumped to the second pond if the water level becomes critically low. Typically, this occurs during summer periods when there is a negative water balance in the drainage area. Hence, the stormwater contains leachate from logs, planks and woodchips stored in the catchment area. Flow, chemical characterization and toxicity of the stormwater runoff from this site is described by Kaczala et al. (2011, 2012). A detailed description of the water recirculation system at the site is provided by Svensson et al. (2012).

Twelve mesocosms with four randomized treatment combinations of vegetation and aeration, and three replicates, were constructed to analyse degradation of PPs and substances represented by COD and water colour. Water from the first collection pond was used for the CW experiment with 12 mesocosm units. Each unit consisted of three tanks (Fig. 1). The mesocosms were designed as surface-flow wetlands (free water surface constructed wetlands).

Each mesocosm unit was assigned one of four treatment combinations, i.e: (1) aeration + vegetation; (2) aeration only; (3) vegetation only and; (4) a control without aeration and vegetation. The first tank (Part 1) of the mesocosm units received an aeration treatment (aeration yes/no), and the second and third tanks (Part 2) a vegetation treatment (vegetation yes/no, Fig. 1). Treatment combinations were randomly assigned to each mesocosm unit to avoid spatial influences on treatment effects. Samples were taken from three sampling points as following: 1) inflow, 2) after Part 1 and 3) after Part 2 (Fig. 1).

Each part (Part 1 and Part 2) of the mesocosm units had a volume of 600 L. The surface areas of Parts 1 and 2 were 1 and 2 m^2, respectively. Part 2 consisted of two tanks connected in series, with the inlet placed above the substrate surface and the outlet arranged