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Long term heavy metal removal by a constructed wetland treating rainfall runoff from a motorway

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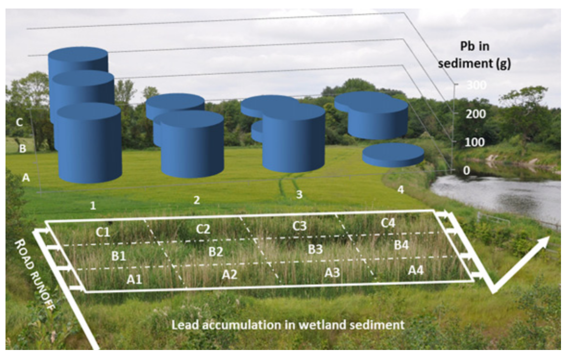
HIGHLIGHTS

- Long-term accumulation of metals in constructed wetland receiving road runoff quantified
- Average removal efficiencies over 9 years: 5% (Cd), 60% (Cu), 31% (Pb) and 86% (Zn)
- Negligible uptake of heavy metals by living vegetation
- Strong relationships between Cr, Pb, Zn and Cu in spatial distribution
- Importance of organic matter with respect to metal removal mechanisms

GRAPHICAL ABSTRACT



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ABSTRACT

The accumulation of heavy metals (Cd, Cr, Cu, Ni, Pb, Zn) in the sediment and plants growing in a constructed wetland used to treat highway runoff in Ireland has been quantified after 6 and 9 year periods of operation. The spatial distribution of the metals' deposition showed strong evidence of flow channelling through the wetland with a strong correlation between the spatial accumulation, particularly for Cr, Cu, Pb and Zn with most of these metals deposited towards the front of the wetland in the sediment. Highest accumulation in the wetland was for Zn, followed by Cu, Pb, Cr, Ni and Cd. The study also quantified that an almost negligible mass of metals had accumulated in the vegetation compared to the sediment. However, an apparent increase in metal accumulation with time may be linked to the cumulative annual production and deposition of organic matter, indicating the importance of the vegetation as an integral part of the treatment process. Based on the measured accumulation and projected runoff loads over the 9 year period, the apparent removal efficiencies were 5% (Cd), 60% (Cu), 31% (Pb) and 86% (Zn). This equates to accumulation rates of 0.1 (Cd), 15.6 (Cu), 11.6 (Pb) and 88.3 (Zn) g per m^2 highway drained per year.

1. Introduction

Constructed wetlands are used throughout the world for the treatment of different types of wastewater, such as domestic, agricultural, industrial, mine water etc. (Kadlec and Wallace, 2009; Martinez-Guerra et al., 2015). The use of these passive vegetation based treatment systems for the removal of pollutants from highway runoff has also grown over the years. Typical pollutants in highway runoff include hydrocarbons, nutrients, PAHs and heavy metals (Sansalone and Buchberger, 1997; Hvitved-Jacobsen et al., 2010). Heavy metals in road runoff tend to be associated with particulate matter, particularly in first flush loads with a preference being shown for the finer particles

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(Barbosa and Hvitved-Jacobsen, 1999; Zhao et al., 2010). The surface water (streams, rivers) into which typically highway runoff is diverted needs to be protected from heavy metals due to their impact on biodiversity, with Hg, Cd, Pb, As, Cu, Zn, Sn, and Cr being particularly ecologically toxic (Ali et al., 2013). Unlike organic substances, heavy metals are essentially non-biodegradable and therefore accumulate in the environment.

Within constructed wetlands physical treatment of pollutants occurs as a result of decreasing flow velocities in the wetland which promotes sedimentation, evaporation, adsorption, and filtration. Biological processes include decomposition, plant uptake and removal of nutrients, plus biological transformation and degradation (Schutes et al., 1999; Kadlec and Wallace, 2009; Rezania et al., 2016). The treatment of pollutants by the plants in the wetland and their rhizosphere microorganisms (i.e. phytoremediation) can include processes including the stabilizing of pollutants in the soil (phytostabilization), the accumulation of pollutants in the plants tissues (phytoextraction) the biodegradation of organic pollutants by microbes in their rhizosphere (rhizodegradation) or by their own enzymatic activities (phytodegradation) and the gaseous exhaust of certain pollutants from the plant tissue (phytovolatilization) (Pilon-Smits, 2005). Several full-scale trials using a variety of different constructed wetland configurations to intercept and treat road runoff have been reported using: vertical flow reed beds (Cheng et al., 2002; Lee and Scholz, 2007), horizontal flow subsurface reed beds (Bulc and Slak, 2003; Revitt et al., 2004) and surface flow constructed wetlands (Walker and Hurl, 2002; Adhikari et al., 2011; Di Luca et al., 2011), with some more recent studies investigating the use of floating macrophytes (Headley and Tanner, 2012; Borne et al., 2013, 2014). Such studies have demonstrated that the wetlands can promote efficient flood attenuation, reduction of peak discharges and overall enhancement of the water quality with respect to hydrocarbons, solids and heavy metals (Malaviya and Singh, 2012). However, the removal efficiencies of typical heavy metals associated with road runoff (i.e. Cu, Zn Cd, Ni and Pb) have been mixed - some studies reporting almost no removal and others up to 90%. The removal of heavy metals has been attributed to processes such as sedimentation, filtration by plants, adsorption, biological assimilation, decomposition, chemical transformation and volatilisation (Barbosa and Hvitved-Jacobsen, 1999; Walker and Hurl, 2002). Contemporary thoughts provide different conclusions as to the ability of plants to efficiently uptake heavy metals (Adams et al., 2012). For example, Pilon-Smits (2005) does not consider *Phragmites* or *Typha* to be high accumulators as opposed to Rai (2012) and Ben Salem et al. (2014) who consider both species (along with many other species of wetland plants) to be suitable due to their extensive rhizosphere which provides an enriched culture zone for biodegradation, as well as the wetland sediment zone provides reducing condition conducive to metal removal pathway. The chemical composition and sorption properties of soil influence the mobility and bioavailability of metals to plant uptake (Kłos et al., 2012; Ali et al., 2013) with Cd, Ni, Zn, As, Se, Cu considered readily bioavailable, Co, Mn and Fe moderately bioavailable and Pb, Cr, and U least bioavailable (Prasad, 2003). However, most studies on constructed wetlands treating road runoff investigating the relative importance of the different heavy metal removal mechanisms have found sedimentation to be the dominant process compared to macrophyte uptake (Lung and Light, 1996; Mays and Edwards, 2001; Walker and Hurl, 2002). In addition, where studies have found any accumulation of heavy metals in plants, the consensus seems to be that little accumulation occurs in the above ground matter (stems and leaves) with most accumulation in the roots (Cheng et al., 2002; Ben Salem et al., 2014). It is clear therefore that more research is still needed to better understand the complex interactions between contaminants, soil, plant roots, and microorganisms in the rhizosphere (Williams, 2002; Vangronsveld et al., 2009).

There are now >2.57 million vehicles in Ireland (DTTS, 2015), with traffic numbers on many roads exceeding the 30 000 vehicles per day threshold limit for implication of mitigation methods as set down in Irish and UK guidance (Highways Agency, 2006; National Roads Authority, 2008). The EU Water Framework Directive (EC, 2000) is also applying pressure for the control of discharges to any receiving water whether ground or surface. Hence, the use of constructed wetlands as a possible mitigation method to treat highway runoff in Ireland was tested with the first wetland established in 2005. The free surface wetland in this study was intensively researched for the first 2 years and then has been periodically checked over the subsequent years to present (Gill et al., 2014). The objective of this study was to quantify the accumulation of heavy metals associated with highway runoff within the constructed wetland after being in operation for 6 and 9 years and assess the relative importance of different attenuation processes.

2. Materials and methods

2.1. Constructed wetland experimental site

The plan area of the free surface constructed wetland was 12.7 m wide by 18.2 m length (with a cross sectional slope of 1%); the depth of the wetland was maintained at approximately 0.3 m. The drainage area of highway that the constructed wetland was designed to treat was a straight length of 480 m westbound and 500 m of eastbound carriageway, each with a total width of 11.6 m. The road, a motorway linking the towns of Kildare and Portlaoise in the east of Ireland, was surfaced with hot rolled asphalt and a 90° sloping kerb face, 90 mm deep. The road drainage system was a kerb and gulley construction with the surface runoff discharging into a piped drainage system via on-line trapped gullies leading to a 375 mm diameter inlet pipe into the wetland. This pipe then diverted to four 100 mm diameter pipe inlets at equal spacing across the width of the wetland to ensure even flow distribution (see Fig. 1). These inlets discharged onto stone gabions to prevent localised scouring and channelling. The outlet pipes were adjustable T-pieces used as weirs to control the water level within the wetland through which the effluent discharges into the River Barrow. A compacted clay liner was used for the base and sides with a permeability of $K < 1 \times 10^{-9}$ m/s measured in-situ using a Double Ring Infiltrometer. A nominal 100 mm layer of local topsoil was added on top. The wetland first received flows in December 2004 (although vegetation was not planted until 5th May 2005). The north half of the wetland was planted with 500 *Phragmites australis* and the south half with 500 *Typha latifolia* - approximately four plants per square metre, as detailed in Fig. 1 and shown in Fig. 2.

Since opening the motorway has had an average traffic count from data from induction loops beneath the road of over 32,000 vehicles per day over the 9 year period (with 9.8% on average as HGV) which has been slowly increasing (see Table S.1 in Supplemental information).

2.2. Storm event sampling and analysis

Rainfall was continuously measured at the site using an ISCO 674 0.1 mm tipping bucket rain gauge. Flow into the wetland was measured with a low profile ISCO 750 area velocity flow module placed within the inlet pipe to measure the average velocity and depth of the flow in the pipe. The outflow was measured using a V-notch weir system with an ISCO 730 bubble module (to measure the depth). These devices were connected to two ISCO 6712 automatic samplers which were programmed to take sequential 300 ml samples of the runoff and effluent on a flow based criteria, normally one sample after every 3 m^3 of runoff had passed the inlet flow meter. During 2005 six major storm events were captured and fully sampled. The samples of runoff were collected at both the inlet and outlet and transported immediately back to the laboratory where they were analysed for a number of parameters commonly found in highway runoff, including total suspended solids, total organic carbon, total phosphorus and four heavy metals, Zn, Cd, Cu and Pb respectively (see Section 2.4).