Accumulation of heavy metals in a constructed wetland treating road runoff

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Abstract

The long-term performance of a constructed wetland treating highway runoff has been studied with respect to heavy metal removal in a temperate, maritime Irish climate. The accumulation of heavy metals in both the sediment and the plants growing in the wetland have been quantified over a 6 year period of operation as well as the spatial distribution of the metals' deposition. Based on the measured accumulation and projected runoff loads over a 6 year period, the removal efficiencies were 7% (Cd), 60% (Cu), 20% (Pb) and 73% (Zn), values which are much less than the apparent removal efficiencies for the system determined from monitoring the inlet and outlet of discrete storm events which were 95% (Cd), 88% (Cu), 86% (Pb) and 95% (Zn). The study also quantified that an almost negligible mass metals had accumulated in the vegetation compared to the sediment. There was a strong correlation between the spatial accumulation of Cu, Pb and Zn with most of these metals deposited at front of the wetland in the sediment. Finally, although the wetland was initially planted with *Typha latifolia* over one half and *Phragmites australis* the other half, after 6 years of operation the *Phragmites* had spread to colonise almost all of the wetland.

Keywords

- Constructed wetland;
- Highway runoff;
- Heavy metals;
- Phytoremediation

1. Introduction

Constructed wetlands are increasingly being installed as systems to treat highway runoff. Typical pollutants in highway runoff include hydrocarbons, nutrients, PAHs and heavy metals (Sansalone and Buchberger, 1997; Hvitved-Jacobsen et al., 2010). The most problematic heavy metals with regards to ecological toxicity are Hg, Cd, Pb, As, Cu, Zn, Sn, and Cr (Ali et al., 2013), although Cu and Zn are also essential trace elements. Heavy metals in road runoff tend to be associated with fine particulate matter, particularly in first flush loads (Barbosa and Hvitved-Jacobsen, 1999; Zhao et al., 2010). The streams/rivers into which typically highway runoff is diverted needs to be protected from heavy metals due to their impact on biodiversity, particularly since they are essentially non-biodegradable.

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Constructed wetlands provide the environment for a variety of different attenuation processes for treatment of highway runoff. Physical treatment 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 (Kadlec and Wallace, 2009). Several full-scale trials using a variety of different constructed wetland configurations to intercept and treat road runoff have been reported, e.g. Cheng et al. (2002), Walker and Hurl (2002), Bulc and Slak (2003), Revitt et al. (2004), Adhikari et al. (2011), Headley and Tanner (2012), etc. 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. Removal efficiencies reported of typical heavy metals associated with road runoff (i.e. Cu, Zn Cd, Ni and Pb) however, have been mixed with some studies reporting almost no removal and others up to 90%. Most studies investigating the relative importance of the different heavy metal removal mechanisms have found that sedimentation seemed to

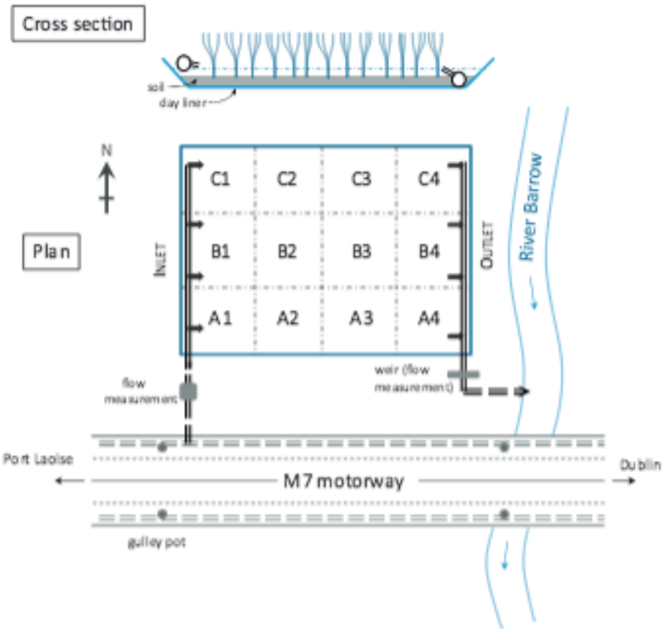


Fig. 1. Plan and cross section schematic of wetland system (with reference cell numbers).

be the dominant process compared to macrophyte uptake (Lung and Light, 1996; Mays and Edwards, 2001; Walker and Hurl, 2002). More research is still needed however, to better understand the complex interactions between contaminants, soil, plant roots, and microorganisms in the rhizosphere (Williams, 2002; Vangronsveld et al., 2009).

The construction of motorways in Ireland intensified in the last decade with traffic numbers on many roads now exceeding the 30,000 vehicles per day threshold for implication of mitigation methods as set down in Irish and UK guidance (NRA, 2008). The EU Water Framework Directive legislation 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 as a pilot trial in 2005. This wetland was intensively researched for the first year and then has been periodically checked over the subsequent 6 years to present.

2. Materials and methods

2.1. Constructed wetland experimental site

The constructed wetland was built adjacent to a new motorway linking the towns of Kildare and Portlaoise in the east of Ireland. The plan area dimensions of the base were 12.7 m wide by 18.2 m length with a cross sectional slope of 1%. The depth of the wetland was no greater than 0.4 m at any point. The runoff drainage area of highway for the wetland was a 980 m straight length of hot rolled asphalt carriageway (total width 11.6 m) with a standard kerb and gulley construction, the surface runoff discharging into a piped drainage system via on-line gully traps installed at 20 m intervals. The runoff entered the wetland via four equally spaced 100 mm diameter pipe inlets across the inlet width to ensure even flow distribution (see Fig. 1). The outlet pipes at the same spacings had adjustable T-pieces as weirs to control the water level within the wetland (at approximately 0.3 m depth). A compacted clay base was laid to produce a relatively impermeable layer ($K < 1 \times 10^{-9}$ m/s) with a nominal 100 mm layer of local topsoil was added on top. The wetland first received flows in December 2004 and vegetation was planted on 5th May 2005. The north half of the wetland was planted with 500 *Phragmites australis* and the south half with 500 *Typha latifolia* – approximately 4 plants/m^2. Since opening the motorway has had an average traffic count of just over 30,000 vehicles per day over the 6 year period (with 10.5% on average as HGV) which has been slowly increasing.

2.2. Storm event sampling and analysis

The site was installed with an ISCO 674 0.1 mm tipping bucket rain gauge and ISCO 750 area velocity flow module to measure the flow into the wetland. A V-notch weir system with an ISCO 730 bubble module to measure the depth was installed at the outflow and a relationship between depth and flow over the V-notch was established. These devices were connected to two ISCO 6712 automatic samplers which took sequential 300 ml samples of the runoff and effluent on a flow based criteria. Between summer and autumn 2005 six major storm events were captured and fully sampled.

Storm runoff samples were analysed for total suspended solids as well as priority heavy metal pollutants (Cd, Cu, Pb and Zn) commonly found in urban runoff (Eriksson et al., 2007). The water quality analysis was carried out in the laboratory using acid digestion followed by analysis with a Varian Liberty AX sequential ICP-AES machine (see Supplementary Information for details). For 5 out of the 6 storms the storm runoff samples for heavy metals were also fractionated into their particulate and dissolved forms by filtering the sample through 0.45 μm filter paper. The discrete sample results were converted to EMC values using the mid-point volume method (Charbeneau and Barrett, 1998).

After this intensive period flow monitoring in 2006, instrumentation was removed (except the rain gauge) and the site periodically visited over the next 6 years where small adjustments were made to ensure the inlet and outlet pipes were level (using a Trimble 4700 GPS system).

2.3. Sampling and analysis of sediment and vegetation

After 6 years of operation the amount of heavy metals and suspended solids that had accumulated in the wetland were calculated by sampling sediment and vegetation in the summer of 2011. The wetland was divided into 12 cells of equal area (each 19.3 m^2) as referenced in Fig. 1. Two samples of sediment were taken from each cell (total of 24 sediments samples) using a tube augur. Samples of the fresh topsoil that was used to fill the wetland during construction were also analysed to determine the initial concentration of heavy metals in the wetland. Each sediment sample was then analysed for suspended solids concentration and heavy metal (Cd, Cr, Cu, Ni, Pb and Zn) concentration in the laboratory using acid digestion to extract the metals followed by ICP-AES analysis. For each sediment sample the depth of sediment to the clay liner was measured and the density of the sediment also calculated. The pH of the water and redox of the water as well as in the sediment was also measured in each cell at the time of sampling in situ with a field probe (YSI 556MPS).

Two complete plants (stem and roots) were carefully dug up and taken from each cell (total of 24 plant samples). In addition, the number of plants in a 1 m^2 quadrat within each cell was counted to give a representative density from which the total number in each cell was calculated. By the summer 2011 the *P. australis* had colonised all but one square (A4) of the wetland with the *T. latifolia* only still growing in a small area near the outlet zone. The plant samples were taken immediately to the laboratory for analysis

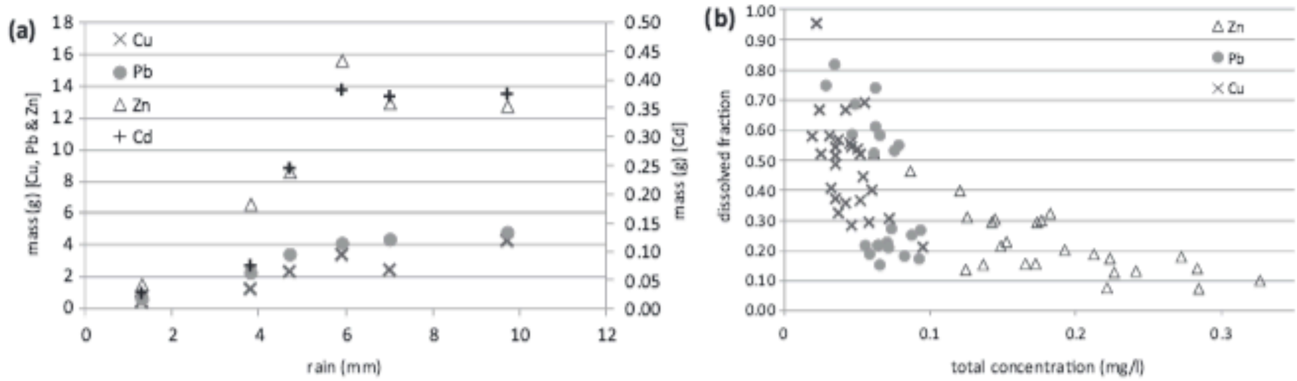


Fig. 2. Heavy metals highway runoff into wetland: (a) relationship to storm rainfall and (b) dissolved fraction.

where they were thoroughly washed in distilled water to remove any clay/sediment before being separated into three different parts – roots, stem and leaf – and cut into 2 cm segment samples. They were then each weighed and recorded and placed in an oven at 105 °C to establish the dry mass. They were then ground down and acid digested as per the method detailed above for the sediment samples before being analysed using ICP-AES.

3. Results

3.1. Storm runoff and wetland treatment performance

The hydrological characteristics of the 6 storm events intensively monitored during 2005 showed that the constructed wetland performed well hydraulically, buffering the inflow to the system from the highway and substantially reducing the peak flow by over 90% (see Supplemental Information). The nominal hydraulic retention times (HRT) for the wetland during these periods varied from between just 21 min to 8.7 h (not far off the recommended design criteria for 1 in 10 year storm design event (Schutes et al., 1999)). However, much of the runoff was detained by the wetland for a number of days and then was discharged by subsequent rainfall events. The storms produced representative events for the temperate maritime climate in Ireland with antecedent dry days for the storms varying from 0 to 4 days – such relatively low intensity events are very common with over 90% of storms over the 6 year monitoring period following periods of rain at least 48 h before hand.

A summary of the event mean concentrations for both total and dissolved fraction is given in the Supplemental Information. The individual flow weighted samples revealed a high correlation between all metal species and TSS in the influent as well as metals removed. The relationship between the heavy metal loads into the wetland and against rainfall per storm in Fig. 2a shows that the higher the volume of rainfall, the more metals delivered to the wetland up to 7 mm at which point there seems to be a tapering off indicating that most wash off had occurred by that point. Interestingly, however there was no discernible relationship between highway runoff and rainfall intensity, which may be due to the relatively low range of rainfall intensities experienced and short antecedent dry conditions.

The analysis of the flow based influent samples for their dissolved fractions of Cu, Pb and Zn (Fig. 2b) indicated an inverse relationship between concentration and dissolved fraction (i.e. more of the metals were associated with the particulate fraction at higher concentrations). The Zn load in the highway runoff was more associated with particulate form, whereas Cu and Pb demonstrated a broader spectrum up to much higher dissolved fractions. Interestingly, Pb in the runoff seemed to be grouped into two distinct sets, one with a high particulate fraction, the other with a high dissolved fraction, perhaps indicating two different mechanisms (or sources) of deposition onto the highway.

3.2. Heavy metal accumulation in sediment and vegetation

3.2.1. Sediment

The amount of heavy metals in the wetland after 6 years of operation is shown in Fig. 3. Cu, Pb and Zn show much higher deposition in the sediment at front of the system (i.e. cells 1) with a very high correlation between where they accumulated. Conversely Cd showed little relationship to most of the other metals. Statistical analysis on the location of accumulation showed the highest levels at the front of wetland (with cell A1 accounting on average for 26.4% of metal accumulation) with levels diminishing with distance through the wetland as well as evidence of channelling through the A cells and a hydraulic dead-zone in the area of cells B3 and C3. The concentrations of the heavy metals in the sediment samples were much higher compared to the topsoil that was originally used to fill to wetland indicating significant accumulation over 6 years of operation. In general the concentrations were highest for Zn (reflecting it being highest in the highway runoff) with mean concentrations in the sediment as follows: 0.91 (Cd), 17.0 (Cr), 62.1 (Cu), 20.1 (Ni), 37.5 (Pb) and 269.3 (Zn) mg/kg.

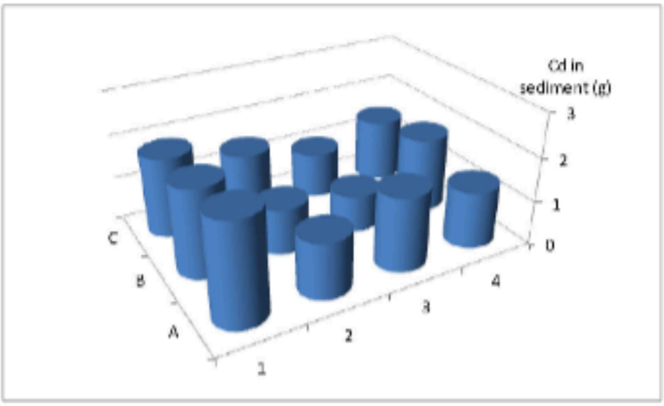
3.2.2. Vegetation

The spatial pattern of heavy metals in the vegetation (net roots, leaves and stems) is shown in Fig. 3. The fraction of the overall mass of heavy metals accumulated in the wetland within the vegetation was very small: 0.73% (Cd), 1.35% (Cr), 2.07% (Cu), 1.23% (Ni), 0.55% (Pb) and 0.61% (Zn). The concentrations of metals within the leaf, stem and root parts of the plant samples taken from each cell (see Supplemental Information) revealed that almost no Cd was being transported to either the leaves or stems of the plants. Cu revealed the highest concentrations in the leaves compared with the other heavy metals, although only elevated in cells A2 and A3 although Zn showed the highest concentrations most consistently across all the cells in the leaves and stems. Levels for Cr, Ni and Pb are broadly similar between the leaves and stem concentrations. The concentrations of heavy metals found in the roots of the plants were 4–5 times higher on average compared to the stem and leaf results for all metals with the noticeable exception of Pb (which was more than 20 times higher). Cu and then Zn revealed the highest translocation factors (Padmavathiamma and Li, 2007) which can be attributed to them being essential trace metals. In contrast to the above ground samples concentrations of Cd were measured in all of the root samples. However, in general the concentrations of the different heavy metals in the leaves, stems and roots were insignificant when compared with the sediment levels. Note, all results represent analysis on *P. australis* with the exception of the *T. latifolia* located in cell A4 which revealed metal concentrations within the same spectrum as the other cells in similar positions in the wetland. A cross correlation between the different metals in the plants showed a reasonable correlation between most metals (with the noticeable exception of Cu) but no significant correlations were found between sediment concentration and concentration in the plants.

3.3. Comparison between heavy metal accumulation and predicted performance

An analysis of the predicted runoff loads into the wetland using the data collected from the intensely monitored storm events in

Cd sediment



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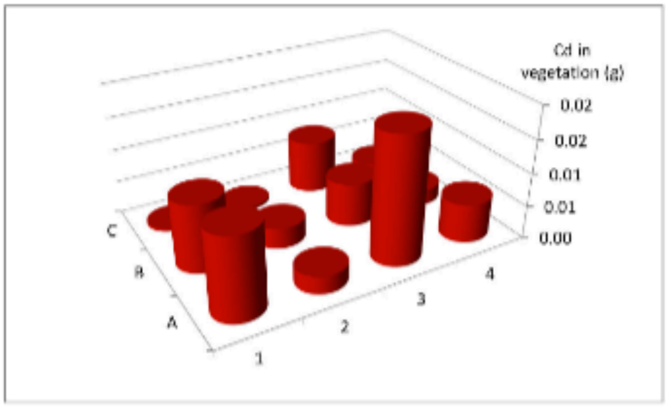
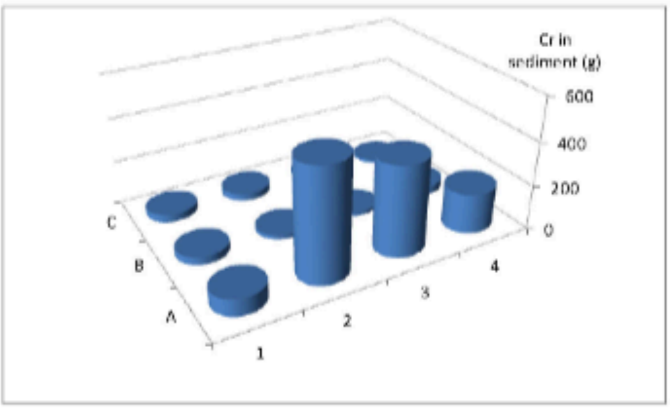


Cr sediment



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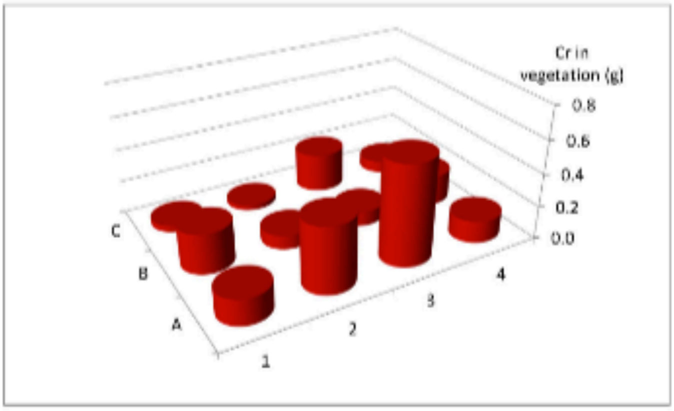
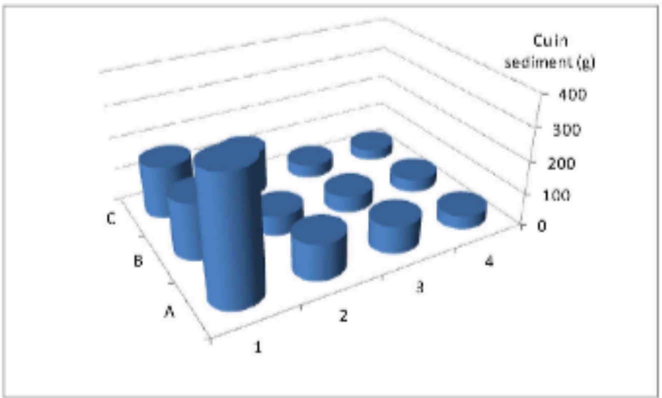


Cu sediment



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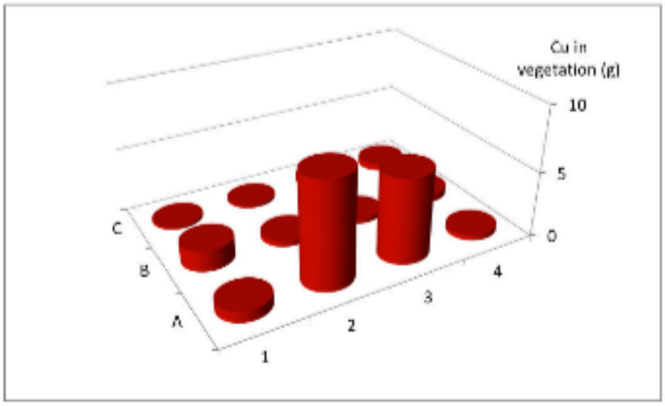

Table 1
Mass of metals accumulated in CW and total removed vs. predicted in runoff (g).

	Cd	Cr	Cu	Ni	Pb	Zn
A1	2.33	76.8	382.5	65.3	202.5	1530.8
A2	1.28	28.7	120.2	32.4	71.6	514.0
A3	1.72	22.9	91.8	34.9	60.9	421.6
A4	1.31	14.4	41.2	28.3	28.3	182.8
B1	2.04	37.3	178.5	50.4	92.6	658.5
B2	1.04	20.2	61.4	22.4	37.7	271.4
B3	0.85	16.5	52.9	19.1	32.2	220.9
B4	1.72	38.6	40.9	44.4	33.7	215.5
C1	1.85	35.1	165.4	44.2	94.5	697.6
C2	1.46	32.6	136.1	34.9	77.1	597.1
C3	1.05	15.7	42.5	22.0	30.5	202.2
C4	1.44	13.3	35.5	22.0	26.9	195.6
Total in wetland (g)	18.09	352.1	1348.8	420.4	788.6	5707.9
Total removed (g)	10.55	179.0	1043.8	164.3	494.0	4593.7
Predicted (g)	142.3	–	1731.8	–	2492.8	6294.4

inlet and outlet of discrete storm events which were 95% (Cd), 88% (Cu), 86% (Pb) and 95% (Zn). In particular, much less Cd and Pb appeared to accumulate than would have been predicted from the early monitoring results, indicating either a reduction in treatment efficiency over time or possible remobilisation of these metals from the sediments. Interestingly a review of 17 constructed wetlands treating urban runoff in Kadlec and Wallace (2009) showed on average higher removal efficiencies for Cd and Pb (71% and 74%) than for Cu and Zn (49% and 60%). Other studies have reported varying removal efficiencies: e.g. Revitt et al. (2004) reported median Cu and Pb removal efficiencies of 40–75% across highway runoff storm events in the UK; Walker and Hurl (2002) found 57% (Zn), 71% (Pb) and 48% (Cu) removal efficiencies in Australia; and Terzakis et al. (2008) showed removal efficiencies of 23% (Cu), 33% (Ni), 61% (Pb) and 59% (Zn) in Crete. Most of these studies, however, were based on comparing inlet and outlet samples during single peak storm events which, as discussed above, is debatable as to whether such a methodology provides an accurate indication of overall long term removal efficiency for such vegetated systems. The concentrations of metals for both total and dissolved fractions in the road runoff storm profiles broadly matched other sites being monitored in Ireland (Bruen et al., 2006) as well as elsewhere (Barbosa and Hvitved-Jacobsen, 1999; Terzakis et al., 2008; Borne et al., 2013) with the highest overall metal concentrations in the road runoff associated with the particulate form and a trend of lower dissolved fractions with higher flows.

Contemporary thoughts provide different conclusions as to the ability of plants to efficiently decontaminate heavy metals (Adams et al., 2012; Rai, 2012). The fraction of overall load in the living vegetation during the summer of 2011 was 0.73% (Cd), 1.35% (Cr), 2.07% (Cu), 1.23% (Ni), 0.55% (Pb) and 0.61% (Zn) and therefore negligible compared to accumulation in the sediment which concurs with several other studies on road runoff (Mays and Edwards, 2001; Walker and Hurl, 2002; Pilon-Smits, 2005; Lee and Scholz, 2007). 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). The percentage of overall annual metal load contained in the leaves and stem varied from 0.1% for Pb up to a maximum of only 1.2% for Cu. The comparison of the different parts of the plant clearly showed that the metal concentrations are much higher in the roots than above ground shoots and leaves by an average factor of 4.8 (Cr), 6.3 (Cu), 6.6 (Ni), 21.0 (Pb) and 5.0 (Zn), again similar to other studies (Cheng et al., 2002; Di Luca et al., 2011). However, the total mass contained in the roots compared to the total mass in above ground vegetation revealed closer average factors of 1.56 (Cr), 0.84 (Cu), 1.89 (Ni), 7.14 (Pb) and 1.64 (Zn). There was no relationship however, between plant numbers and the heavy metal concentration in the roots. Equally, there was no relationship between the number of plants in each cell and the sediment accumulated (depth) as might have been expected ($R = -0.195$). Finally, the somewhat limited comparison between *P. australis* and *T. latifolia* showed broadly similar concentrations in leaf, stems and roots for all metals (given the position of the cell A4), matching the findings of Arroyo et al. (2013). *Typha* had clearly struggled compared to the *Phragmites* that had colonised almost the entire wetland after 6 years which has been shown to act as an invasive species in many places across the world with competitive advantages due to high rates of primary productivity (Brisson et al., 2010).

The pattern of accumulation across the wetland in general revealed lower concentrations of metals in the sediment with length through the wetland (inlet to outlet) which corresponds with other studies (Walker and Hurl, 2002; Di Luca et al., 2011). This could indicate that sedimentation is the primary initial removal mechanism. Cu, Pb and Zn showed much higher deposition at front of system (i.e. cells 1) with a very high correlation between them. Zn also revealed higher deposition further into wetland (despite it being in the most particulate form in runoff) which may be evidence of some remobilisation process at higher flow rates or its release in a dissolved form after wetland processes in the sediment. Despite Cu in the road runoff having a higher dissolved fraction compared to Zn and Pb (Fig. 2b) still most of its load in the sediment had accumulated at the front of system, indicating other removal mechanisms than just sedimentation. Generally reducing conditions were found in the sediment in this wetland study (down to −160 mV) with aerobic conditions in the surface water as found in other studies such as Faulwetter et al. (2009). These reduced conditions would promote anaerobic processes such as sulphate reduction, providing the sulphide which could act to precipitate the divalent metal cations.

One explanation for the more consistent apparent accumulation of Cu and Zn over time compared to Cd and Pb might be that Cu and Zn would have more interaction with the vegetation being essential trace metals. The numbers (and size) of the plants has burgeoned over the years with >24,000 full size reeds (most between 2.5 and 3 m tall) growing in summer 2011. Although the above ground plant matter was shown to contain only a relatively small fraction of metals, the annual cycle of uptake into the growing plants with subsequent die back to produce organic sediment may cumulatively over several years have a more significant impact.

5. Conclusions

Over a 6 year period the wetland has removed a considerable amount of the heavy metals associated with road runoff that otherwise would have discharged directly to the river. Based on the measured accumulation and projected runoff loads, the long term heavy metal removal efficiencies over a 6 year period were considerably lower than the apparent removal efficiencies determined from the sampling of discrete storm events which also highlighted potentially different removal/remobilisation processes between Cd and Pb as opposed to Cu and Zn in such a wetland environment. The study also quantified that an almost negligible mass of metals had accumulated in the vegetation compared to the sediment.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.ecoleng.2014.03.056.

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