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AN ASSESSMENT OF HEAVY METALS POLLUTION POTENTIAL OF ROAD SEDIMENT DERIVED FROM A SUBURBAN ROAD NETWORK UNDER DIFFERENT WEATHER CONDITIONS

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Abstract

The quantification of heavy metals in dry and wet weather derived (runoff and snowfall) sediment from roads, under three different environmental conditions, has indeed the potential to allow improved understanding on urban diffuse pollution. With that aim, the reported study determined the concentrations of Cd, Cu, Pb, Ni, Cr and Zn in road sediment, which are reported to be found in elevated level in the road-traffic environment, and associated pollution levels at four different sites, representing a typical suburban road lay-out, as part of a 10 months field and laboratory studies. The heavy metal concentrations in sediments are determined by strong nitric acid digestion and atomic absorption spectrometry. The results reveal that road sediment contain significant amounts of heavy metals and their concentrations vary with sediment type and size fraction (wet weather, snow and dry sediments), and between sampling sites according to the site specific attributes rather than traffic volume alone. The metal concentrations among three different environmental conditions are found in the order of rainfall-runoff, snow and dry sediment associated with smaller size fractions, in general. Contamination assessment suggests that road sediment may likely pose a moderate to considerable level of ecological risk to the nearby water environment which need attention while designing sustainable urban drainage for roads.

Key words: Atomic absorption spectrometry, Edinburgh, heavy metals, pollution, road lay-out, road sediment

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1. Introduction

In recent years, urban areas have experienced severe environmental pollution. Roads, an integral part of any urban development, only share a small percentage of urban land uses, however they often generate various types of pollutants, among which heavy metals are very common (Barrett et al., 1993; Furumai et al., 2001; Manohar and Kavuri, 2017). Heavy metals are important environmental pollutants and are regarded as potential hazards to human health and to natural ecosystems (Hlihor et al., 2009; Marcovecchio and Ferrer, 2005). The heavy metals derived from roads come from diverse sources (Banerjee, 2003). From the study of Irish et al. (1995), atmospheric deposition, input from traffic, carriageway breakup and the surrounding land uses are found to be the key sources of pollution from roads. Aspects of heavy metals and their quantification in road sediment are often studied to illustrate associated pollution level. However, their concentrations in road sediment of different forms such as dry, wet weather and snow, have been found to be highly variable and dependent on a wide range of factors including location, traffic volume, extent of dry spells, frequencies of street sweeping, the nature

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of road surfaces, and methods used to quantify metal concentrations (Backstrom et al., 2003; Bilos et al., 2001; Drapper et al., 2000; Helmreich et al., 2010; Li et al., 2017; Manno et al., 2006; Pal et al., 2011; Sutherland et al., 2001). For example, considering location aspect, Charlsworth et al. (2003) and Ewen et al. (2009) reported that inner roads emit more pollution compared to motorways due to traffic congestion and traffic movement patterns. Similar suggestion was also evident from Mahbub et al. (2010). In addition, sediment particle size fractions also play a key role in the variability of heavy metal concentrations by mass and previous studies have found higher metal content with decreasing particle size fractions, as noted by Roger et al. (1998), Deletic and Orr (2005), Lau and Stenstrom (2005), Robertson and Taylor (2007), Kim and Sansalone, (2008), Sansalone et al. (2010) and Zafra et al. (2011).

Once deposited, road sediment could be transported from the road surfaces to nearby water bodies by different cleansing events comprised of mechanical (street sweeping) and natural (air, snow and rain) events, and thus in turn make a significant contribution to the local environmental pollution. Many studies in different parts of the world have characterised road sediments. For example, studies carried out in Australia and New Zealand (Ball et al., 1998; Brown and Peake, 2006; Drapper et al., 2000; Mosley and Peake, 2001), America (Brezonik and Sansalone et al., 1996; Lee et al., 2004; Sartor et al., 1974; Sartor and Boyd, 1972; Stadelmann, 2002; Sutherland, 2003), Asia (Kim et al., 1998; Lee et al., 2002), and Europe (Backstrom et al., 2003; Cabtree et al., 2006; Cabtree et al., 2008; Deletic and Orr, 2005; Ewen et al., 2009; Farm, 2002; Helmreich et al., 2010; Pal et al., 2011; Robertson and Taylor, 2007; Westerlund et al., 2003; Zafra et al., 2008). These studies have highlighted significant intrinsic variability in data collected in different countries, or even in different cities within a country, which justify the need for collecting local data.

While a few of the above mentioned studies of heavy metal concentrations in dry and wet weather sediment have been undertaken in the UK, only limited information is available for heavy metals on dry road sediment for Edinburgh (from an earlier study by present authors (Pal et al., 2011). Moreover, there is no previous data on the quantification of heavy metals and associated contamination levels in dry, wet weather and snow derived sediment from roads in the UK. In particular, heavy metal emission patterns associated with road lay-out conditioned different traffic movement patterns along a suburban road network is not found to be exclusively focused in the literature. In accordance with European Union Water Framework Directive (EU WFD), recent road drainage practice for new roads has seen the introduction of sustainable urban drainage systems (SUDS) for roads, replacing or modifying conventional road drainage and aimed towards addressing the water quality and water quantity issues, as a long-term robust option for urban pollution management (Heal et al., 2009). It is therefore useful to have information on pollutant concentrations that may inform road drainage designs and maintenance in the context of SUDS for roads.

Within this context, the present research aims to study heavy metal concentrations in road sediment collected during dry and wet weather spells for a suburban road network using Riccarton Campus road network of Heriot Watt University, and their distribution in different size fractions of dry, runoff and snow derived sediment. Furthermore, heavy metal pollution levels associated with the dry, wet weather and snow derived sediment are calculated and discussed in relation to the potential ecological risk to aquatic organisms. The metals in focus are Cd, Cr, Cu, Ni, Pb and Zn, which are often found in high concentrations in the road traffic environment and are toxic and persistent nature (e.g. Ball et al., 1998; Cabtree et al., 2008; Gan et al., 2008). It is hoped that the findings may inform heavy metal emission patterns associated with traffic movement patterns conditioned by road lay-outs commonly found in sub urban roads and assist water professionals to develop an effective strategy for controlling pollutants with the aim of managing urban diffuse pollution.

2. Material and methods

2.1. Study area

Edinburgh, a city in the southeast of Scotland, lies on the east coast of Scotland's central belt, alongside the Firth of Forth. It has a temperate maritime climate (moderate summer and mild winter), the annual average temperature range is about 0.9 – 12.2 ^{0}C and the annual average precipitation is about 668 mm distributed fairly evenly throughout the year. Heriot Watt University's Riccarton Campus (latitude: 55.91^0 and longitude: -3.31^0) is located southwest of Edinburgh city and has been developed during the last 40 years with good ecological and environmental perspectives. Because it has been observed that road runoff goes directly or indirectly into the Murray Burn, which is a small stream running across the campus, there is a risk of it being contaminated by traffic related pollutants. Fig. 1 illustrates a map showing the study area.

Road sediments during dry, rainfall and snow events were collected from four sites with different road lay-outs: a road bend (RB), a section of road with active speed control measures (RSC), a road intersection (RI) and a straight section (SR). As mentioned earlier, to address the objectives set for this study the sampling sites were selected to cover different traffic movement patterns conditioned by road lay-outs and to consider other attributes, such as road surface condition, presence of road paints, surrounding land use.

The road surface condition can be defined as good except for the RSC site in which increased degradation of road surface materials and paints was observed.

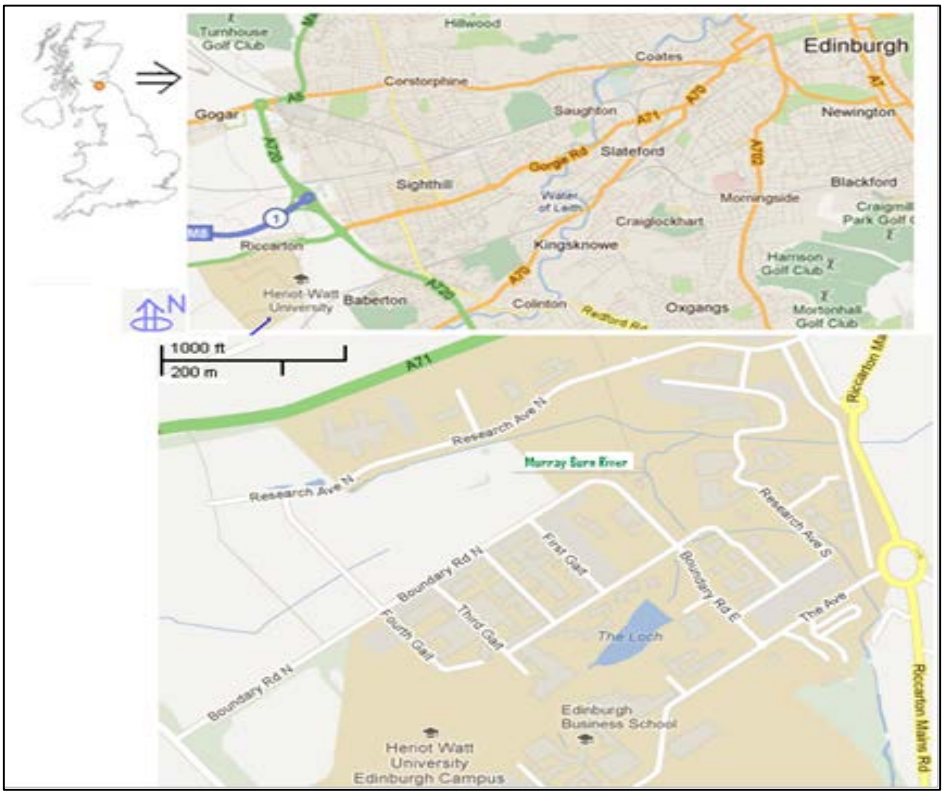

Fig. 1. Google map showing the study area

Table 1. Site details on Riccarton Campus road network, Heriot Watt University, Edinburgh

Site description	***Painted lines***	***Road condition***	***Road furniture near sampling points***	***Traffic flow type passing sampling points***	***Traffic load (vph)***	***Surrounding areas near sampling points***
Road Bend (RB)	Yellow	Good	Gully/lamp post	Braking	200	AA, FPS
Road with speed controls (RSC)	White/ yellow	Poor	Lamp post/ signs/ pedestrian crossing with barriers	Braking	250	OB, CP, FPS
Road Intersection (RI)	White/ yellow	Fair	Traffic lights/ gully	Stop-start	285	OB, MR. FP
Straight Road (SR)	Yellow	Good	Sign posts/barriers	Steady speed	650	CP, MR, FP

Note: vph = vehicle per hour, AA = Agricultural area, CP = Car park, OB = Office buildings, FP = Foot path, FPS= Foot path separated by grass strips, MR = nearby main roads linked with campus roads, SH =Student halls.

Double yellow lines are painted along the edge of the road at all the sites and additional white marks are found at the RSC and RI sites (for pedestrian crossing area and for the stop line).It should also be noted that the RI and SR sites are very close to an entry and exit point of the campus. The roads are used by a variety of personal and commercial vehicles and some are parts of bus routes. Each of the sampling sites had 2 lanes of traffic with an asphalt road surface, with low traffic densities measured as 200, 250, 285 and 650 vehicles per hour for the RB, RSC, RI and SR sites, respectively. A summary of the study sites is presented in Table 1. The surrounding land area is devoted to campus buildings, some farm land, foot paths and pavements (see Table 1). Based on the traffic movement pattern at the sites during the sampling period, primarily two types of 'stop-start' activities are found. Firstly, at the RB and RSC sites, almost all traffic was observed to undergo braking as it passed through. Secondly, at the RI site, the traffic movement pattern was found to be more controlled in terms of 'stop and start' activities and therefore, a lesser degree of braking occurred than at the previous type, while the traffic movement pattern was consistent with a steady speed at the SR site. Mechanical street-sweeping and gully cleaning occur typically twice a year on the campus by the local council.

2.2. Sample collection

2.2.1. Dry sediment sampling

Road deposited sediment (RDS) was collected from the sites during a range of dry spells (between 1 to 14 days) after any significant rainfall events or any mechanical street cleaning. A total of 25 samples from each site were collected over a 10 month period

between March and December 2010. Sampling plots comprised a 1 m^2 road surface area near the curb and samples were collected by repeated sweeping using brushes as outlined by Charlesworth et al. (2003), Kim et al. (1998) and Robertson and Taylor (2007). During sampling, care was taken to minimise sweeping pressure so that (artificial) detachment of road material particles could be avoided. Details of the sample collection technique and strategies can be found in Pal et al. (2011) and Pal (2012). In addition, four other sites were selected (on the studied road) network which carry no traffic, for measuring local background values for heavy metals. These sites were located no less than 200 m north-west of the closest site (RB) to avoid any significant influence from other sampling sites.

2.2.2. Runoff sediment sampling

Twelve different rain events were monitored during the study period with the aim of capturing monthly and seasonal variations in sediment concentrations in response to wide range of hydrological aspects such as lower intensity longer duration rainfall and vice versa; antecedent dry days from 1 to 20 days Data for the rain events were collected using a tipping bucket gauge which was installed on a building roof and located approximately 1000 m from the runoff sampling sites. Each site has road gullies to collect road runoff from the road surface during rainfall periods. Preliminary investigation suggested that each gully drains approximately 10 m long and 3 m wide road surface area. The gullies at the experimental sites were modified to intercept storm water inflow during runoff events. A trapezoidal plastic catch tray was designed to guide the road runoff as it left the road surface, to be collected in a 5 L bucket hanging inside the existing gully pot.

Different experimental set-ups were evaluated with the aim of optimising runoff collection while avoiding leakage, and designs were modified accordingly. Teflon tape was used to minimise the joint gap between bucket and gully pot grid. However, complete control of leakage could not be met in some circumstances. Nonetheless, this simple approach was found to be effective for runoff collection from the road surface with little modification of the existing road drainage systems. Runoff stored in the bucket was collected after each event as soon as was practicable and was transported back to the laboratory for further analysis. Particles attached to the catch tray were washed into the bucket by using distilled water to minimise cross contamination. The details of the runoff monitoring and collection procedures followed for this study can be found in the Caltrans Guidance Manual: Storm water Monitoring Protocols (Caltrans Guidance Manual, 2000) and Pal (2012).

2.2.3. Snow sampling

Snow samples from five snow events during November and December 2010 were collected from every sampling site by plastic scoops. The samples were taken from the whole vertical profile including dust deposition as outlined by Hricko et al. (1993) and Krčmová et al. (2009) Sampling was undertaken at the end of the snow event and a total sample of about 2 kg from each site was transferred into plastic bags and transported back to the laboratory.

2.3. Laboratory analyses

The RDS samples were dried upon collection at room temperature for 24 hours and then sieved through a 1 mm mesh to remove large particles and other debris, such as plants, leaves, litter. Runoff sediment was kept in an oven at 105 ^{0}C for 24 hours to obtain dry sample. Snow samples were allowed to melt at room temperature first and then similar steps were followed as for the runoff samples to obtain dry samples. All the sediment samples were then separated into three groups using standard sieves: integrated (whole sample irrespective of size fractions), sediment size fractions 250-63 µm (coarse) and sediment size fractions below 63 µm (fine) for metal analysis. Sediment samples were wet-sieved through 250 and 63 µm aperture metal sieves to separate them into fine and coarse fractions (Charlesworth et al., 2003). Wet sieving has an advantage over dry sieving of better accuracy. More details can be found in Deletic and Orr (2005), Huang et al. (2009), Karlsson et al. (2010) and Pal (2012). Moreover, the 63 µm size has some importance for metal concentrations by mass, as suggested by various researchers (e.g. Charlesworth et al., 2003; Pal et al., 2012; Sutherland, 2003; Zafra et al., 2008). Wet-sieved samples were then collected in a pan and left to dry for 24 hours prior to being oven dried at 105 °C for another 24 hours to obtain a dried sample for the metal analysis.

There are numerous techniques of sediment bound metal extraction procedures available in literature and are found to be varied significantly. A rigorous analysis of using different strength of reagents (0.05 M EDTA, 0.5 M HCl, concentrated HNO_3) for metal extraction from road sediment can be found in Sutherland et al. (2001). Within this context, selected samples for this study were initially tested using HNO_3, aqua regia (a mix of HCl-HNO_3 in a ratio of 3:1 by volume) and HCl-HNO_3-$HClO_4$. The results obtained did not exhibit significant differences (Kruskal – Wallis test; p >0.05) due to the different extractants used. As popular use of concentrated HNO_3 as a metal extractant was already evident in previous research, even though it is not a best alternative (Sutherland et al., 2001), to keep the metal extraction procedure consistent and logistically efficient, concentrated HNO_3 was used as metal extractant from sediment for this study.

Briefly, a small portion of oven dried sediment (0.50 g) was placed in a glass beaker and treated with 10 ml of strong HNO_3 (Anal R 70%)for acid digestion. The samples were then heated, cooled and filtered to a volumetric flask made up to 50 mL using deionised water. A Perkin Elmer 200 atomic absorption spectrometer (AAS) was calibrated for the range of 0-

2.5 ppm for all the metals analysed for this study using standard solutions which were made up from stock solutions of 1,000 ppm. Calibration curves for metal solutions were maintained with high precision (R^2 = 0.99). Standard laboratory quality control/quality assurance procedures were followed, which are illustrated in Caltrans Guidance Manual (2000). The mean of triplicate analyses was recorded as the metal concentration for all samples, for data quality purposes. Good precision of the whole procedure (less than 5% relative standard deviation) was achieved. The mean of triplicate analyses was recorded as the metal concentration for all samples, for data quality purposes. Moreover, the accuracy of the test results was determined by analysing reference material using the standard solutions prepared for metal analysis.

A portion of certified reference material MESS-3 (NRC Canada Certified Marine Sediment Reference Material) was tested three times for total metal analysis of Cd, Cr, Cu, Ni, Pb and Zn by using the Perkin Elmer 200 AAS analyser. The mean of the metal concentrations from three tests was then compared with reference values as presented in Table 2. As seen in Table 2, the results revealed that the mean value for each metal was very similar to their respective reference values and hence, the laboratory analysis was deemed to be accurate. Blank samples were tested repeatedly to avoid cross contamination between samples. De-ionised water was used throughout for cleaning apparatus, preparing solutions and other related purposes. Further details of the analytical methods followed can be found elsewhere (Deletic and Orr, 2005; Pal, 2012; Robertson and Taylor, 2007).

2.4. Heavy metal pollution assessment

Transforming heavy metal concentration levels into a single index value is often preferred to derive better understanding and decision making tools in environmental pollution research. In this regard, the degree of contamination (*CD*) and potential ecological risk (*PERI*) indices were estimated and are presented in Table 3. It may be debatable to use these indices, as they were primarily made for the assessment of aquatic toxicity of natural sediment. However, due to the robustness of the indices, they have previously been used (with necessary modification) to assess metal contamination for road sediment (e.g. Huang et al., 2009; Shi et al., 2010; Yu et al., 2003). The *CD* and *PERI* are defined by the Eqs. (1-4) as shown below:

$$C_f^i = \frac{C_s^i}{C_b^i} \quad (1)$$

where C_f^i is the single metal pollution index for metal i; C_s^i is the concentration of metal *i* in the samples; C_b^i is the reference value for metal *i*. For this study, C_b^i was taken to be the local background value from road sites which carry no traffic.

$$CD = \sum_{i=1}^{n} C_f^i \quad (2)$$

where *n* is the total number of metals (n=6 for this study). Categories of *CD* were adjusted and classified with modification after Duong and Lee (2011), as seen in Table 3.

$$E_r^i = T_r^i \times C_f^i \quad (3)$$

E_r^i is the monomial potential ecological risk factor and T_r^i is the metal toxicity factor. According to Hakanson (1980), the toxic values for each metal are in the order of Zn = 1 < Cr = 2 < Cu = Ni = Pb = 5 < As =10 < Cd = 30 < Hg = 40.

$$PERI = \sum_{i=1}^{n} E_r^i \quad (4)$$

where *PERI* is the potential ecological risk caused by the overall contamination, signifying the sensitivity of the biological community to the toxic substances. As the number of pollutants considered in this study are different to Hakanson's study (As and Hg are not being used here), an adjustment of the indices was made and categories were also classified accordingly, as seen in Table 3. A similar approach of adjustment of the indices was also used by Duong and Lee (2011) and Zhu et al. (2008) in their studies.

3. Results and discussion

3.1. Heavy metal concentrations in road sediment

The descriptive statistics of heavy metal concentrations investigated in dry and wet weather (runoff and snow) derived sediment are presented in Table 4. The background concentrations of the metals from Riccarton Campus are also shown along with the regional background values for Scotland (Appleton, 1995). The local background values for all the metals except Pb are found very similar to the regional background values.

The mean concentrations (mg kg^{-1}) for all the heavy metals except Ni show elevated concentrations compared to their local background values (Table 4) in dry, runoff and snow derived sediments, suggesting that the road sediments are highly contaminated by heavy metals related to the road-traffic inputs in addition to atmospheric deposition (Pal et al., 2011).

The mean concentrations reported here for heavy metals in dry and runoff sediment are found consistent with previous studies in the UK (Charlesworth et al., 2003; Crabtree et al., 2006; Crabtree et al., 2008; Deletic and Orr, 2005; Pal et al., 2011; Robertson and Taylor, 2007), and other parts of the world (Ball et al., 1998; Brezonik and Stadelmann, 2002; Drapper et al., 2000; Gan et al., 2008; Han et al., 2006; Kim et al., 1998; Sutherland, 2003; Zafra et al., 2008). The metal concentrations in snow sediments for this study were found lower than previously reported values in Swedish and German studies (Hallberg et al., 2007; Helmreich et al., 2010; Westerlund et al., 2003).

Table 2. Heavy metal concentrations for standard reference sample MESS-3 from the laboratory testing compared with certified values

	Concentration of metals in MESS-3 (mg/l)		
Metal	***Laboratory test (n = 3)***	***MESS-3 certified value***	***% difference***
Cd	0.24 ± 0.02	0.24 ± 0.01	+0.55
Cr	106 ± 2.84	105 ± 4.00	+1.37
Cu	34 ± 2.18	33.9 ± 1.60	+0.45
Ni	47.9 ± 3.28	46.9 ± 2.20	+2.07
Pb	21.8 ± 0.40	21.1 ± 0.70	+3.28
Zn	158.4 ±9.60	159 ± 8.00	-0.40

Table 3. Categorization of standards for C_f^i, CD, E_r^i and PERI

C_f^i	***CD***	***Degree of pollution***	E_r^i	***PERI***	***Levels of potential ecological risk of the aquatic environment***
C_f^i< 3	CD<6	Low/Unpolluted	E^i_r <30	PERI<75	Low
3≤ C_f^i<6	6≤CD<12	Moderate	30≤ E^i_r <60	75≤PERI<150	Moderate
6≤ C_f^i<9	12≤CD<24	Considerable	60≤ E^i_r <120	150≤PERI<300	Considerable
C_f^i>9	CD>24	High	120≤ E^i_r <240	PERI>300	High

C^i_f *is the single metal pollution index; CD is the degree of contamination;* E_r^i *is the monomial potential ecological risk factor; PERI is the potential ecological risk index*

Table 4. Descriptive statistics of heavy metal concentrations (mg/kg) in road sediments (averaged over sites)

Metal	***Dry sediment (n=120)***		***Runoff sediment (n=48)***		***Snow sediment (n=20)***		***Local background mean [a] (n=4)***	***Regional background mean [b]***
	Range	***Mean ± SD***	***Range***	***Mean ± SD***	***Range***	***Mean ± SD***		
Cd	0-4.00	1.00 ±0.80	0.60-1.75	1.00 ± 0.26	0.90-2.00	1.32 ±0.3	1	1.4
Cr	4-76	17 ± 11	35-120	64 ± 20	15-28	21 ± 4	8	n.a.
Cu	20-220	67 ± 41	79-206	134 ± 30	59-125	87 ± 19	44	46
Ni	3-33	12 ± 6	20-50	33 ± 7	13-27	21 ± 3	20	n.a.
Pb	6-621	92 ± 101	30-140	69 ± 24	61-138	88 ±22	28	115
Zn	99-460	212 ± 88	165-280	205 ± 27	185-292	233 ± 34	107	120

n - Number of samples analysed; SD – Standard deviation; a - Metal concentrations were measured in RDS from road sites which carry no traffic; [b]*Appleton (1995).*

The difference may be linked to a relatively longer winter spell with persistent snow coverage in Swedish studies compared to a rather short span of snow and quick melting in the Edinburgh study. Nonetheless, there is no previous study in the UK on snow sediment to compare directly with the presented results. The distribution of the highest mean concentration of individual metals was found as follows: 1.32 and 233 mg kg^{-1} for Cd and Zn, respectively (in snow sediment); 64, 134 and 33 mg kg^{-1} for Cr, Cu and Ni, respectively (in runoff sediment); 92 mg kg^{-1} for Pb (in dry sediment).

On average, Cr and Cu concentrations in the runoff sediment were found much higher compared to their concentrations in dry and snow derived sediments, while Cd, Ni and Pb exhibited little variation, and Zn showed even less variation among the three categories of sediment. These findings suggest that rainfall runoff derived sediment carry the highest load by mass for most of the metals, followed by the snow and dry sediment. Fig. 2 illustrates heavy metal concentrations of Cd, Cu, Pb and Zn in different weather conditions.

The plots are made by averaging over the sites and number of samples collected in a particular month for the dry, runoff and snow derived sediment during the study period of March 2010 to December 2010. Though in the UK rainfall is occurred in each month, rainfalls with very low intensity and duration were overlooked due to their less runoff potential.

As seen in Fig. 2, the most polluted month is July followed by June and August compared to the least polluted month of April followed by March with the exception for Pb over the study period. Point to be noted that Pb was found to be associated with greater sediment sizes derived from the disintegration of yellow paint in the study area and, hence was not observed to be carried out by rainfall induced runoff derived sediment (Deletic and Orr, 2005; Pal, 2012).

It is seen that rainfall occurred between July and August carried the highest pollutant load that were found to follow larger dry days between the rainfall events. The temporal distribution pattern obtained for this study is also in accordance with Robertson and Taylor (2007) which dealt with analysis of dry road sediment collected from Manchester City in the UK.

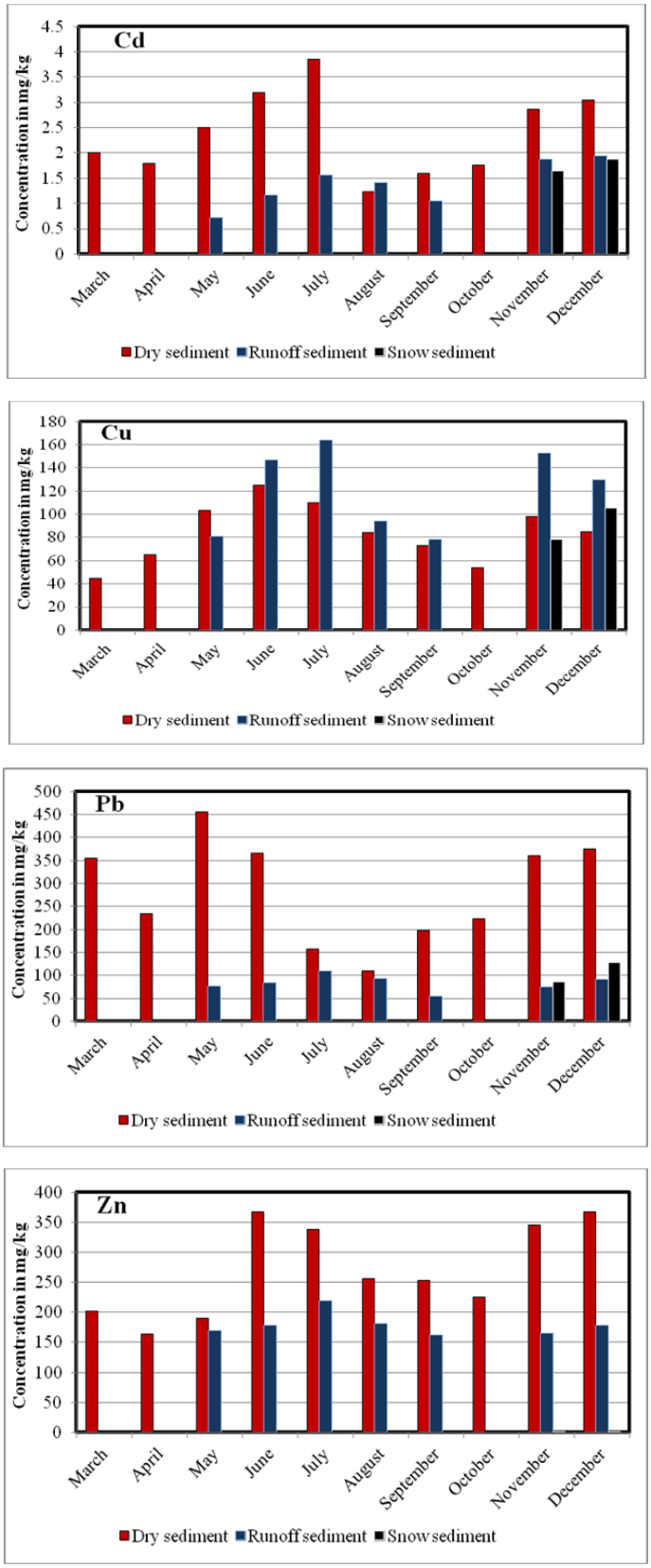

Fig. 2. Temporal distribution of mean monthly Cd, Cu, Pb and Zn (averaged over sites) concentration over the study period

At a glance, it is seen that rainfall induced runoff during June and July and snow derived runoff during November to December carried the sediment bound pollutants from the roads to the nearby water bodies, which needs site control measures in relation to sustainable urban drainage system to lessen metal pollution risks.

3.2. Site and size specific metal concentrations

The distribution of metals on the different particle sizes of road sediments are of particular importance to best management practice regarding road drainage. The influence and importance of the particle size distributions on sediment associated metal concentrations are well documented from previous studies (e.g. Robertson and Taylor, 2007; Sansalone et al., 2010). An earlier study on RDS using the same road network by the present authors also revealed that the greater metal concentrations were found with decreasing particle size fractions (Pal et al., 2011). Only heavy metals found at levels 150% higher than their respective local background values (based on Table 4), which is indicative of anthropogenic input (traffic) in addition to atmospheric deposition, were selected for further analysis.

The distributions of heavy metal concentrations for each of the two particle size fractions of road sediment for all four sampling sites were determined and are presented in Table 5. As seen in Table 5, on average, the highest concentrations for most of the metals (across the sampling sites for both particle size fractions) were found in the order of runoff sediment>snow sediment >dry sediment. In general, sediment size<63 µm usually contains approximately 2 to 4 times higher concentrations than the size fraction 250-63 µm for most of the metals across the sampling sites. For example, particle size<63 µm contains the highest mean concentrations for Zn and Cu with the value of 572 and 320 mg kg^{-1}, respectively, compared to their concentrations of 237 and 140 mg kg^{-1}, respectively, in the size fraction 250-63 µm (for the runoff sediment at the RSC site). Similarly for Cr, the highest mean of 153 mg/kg in the<63 µm size fraction compared to 56 mg/kg in the 250-63 µm size fraction (in the runoff sediment at the SR site). In contrast, the highest mean concentration of 250 mg kg^{-1} for Pb was in the 250-63 µm size fraction (in the dry sediment at the RSC site) compared to 195 mg/kg in the<63 µm size fraction (in the runoff sediment at the RSC site). This is consistent with the vast majority of previously published work (e.g. Ball et al., 1998; Crabtree et al., 2008; Drapper et al., 2000; Deletic and Orr, 2005; Han et al., 2006; Kim et al., 1998; Sutherland, 2003; Robertson and Taylor, 2007; Zafra et al., 2008; Westerlund and Viklander, 2006).

Furthermore, although the traffic volumes at the RB, RSC and RI sites are about one third to one half that for the SR site, the higher metal concentrations were not found in accordance with the higher traffic volume. The variations across the sites appear to be primarily due to site- specific attributes, such as road lay-out, road surface condition and presence of road paint rather than due to traffic volume alone. Taking Pb as an example, significantly higher Pb concentrations at the RB and RSC sites in dry road sediment were found compared to other sites. The previous use of Pb in fuel was phased out more than a decade ago; the only other likely source of Pb may be linked to the road paint particles, such as double yellow lines at these sites, based on the suggestion reported by Deletic and Orr (2005).

Table 5. Site and grain size specific mean metal concentrations (mg/kg) in road sediment

Site (Traffic volume)	***Category***	***Particle size fraction 250-63 µm***				***Particle size fraction<63 µm***			
		Cr	Cu	Pb	Zn	Cr	Cu	Pb	Zn
Road bend, RB (200 VPH)	Dry Sediment	11	35	90	128	58	94	135	522
	Runoff sediment	42	103	71	202	130	254	125	494
	Snow sediment	19	72	64	167	79	184	109	418
Road with speed controls, RSC (250 VPH)	Dry Sediment	23	34	250	134	26	102	157	426
	Runoff sediment	51	140	132	237	149	320	195	572
	Snow sediment	22	82	118	209	109	234	112	521
Road intersection, RI (285 VPH)	Dry Sediment	7	45	34	121	21	114	115	287
	Runoff sediment	45	127	59	242	126	292	111	555
	Snow sediment	24	87	46	199	59	178	87	415
Straight road, SR (650 VPH)	Dry Sediment	12	65	49	217	21	142	137	518
	Runoff sediment	56	120	71	247	135	312	120	571
	Snow sediment	19	106	57	219	95	212	110	487

Table 6. Comparison of mean heavy metal concentrations with published guide line values

Metal	***Dry sediment (n=120)***	***Runoff sediment (n=48)***	***Snow sediment (n=20)***	***Dutch sediment quality guideline****	***Canadian sediment quality guideline****	***UK rural river sediment quality*****
Cd	**1.00**	**1.00**	***1.32***	1	0.60	3.98
Cr	17	***64***	21	62	37.3	41.81
Cu	***67***	***134***	***87***	20	35.7	48.88
Ni	12	**33**	**21**	16	n.a.	67.54
Pb	***92***	***69***	***88***	40	35	440.50
Zn	***212***	***205***	***233***	147	123	682.91

*Data measured in mg kg^{-1}; Data in bold indicate metal concentrations exceed or equal to at least one of the two guidelines, while bold and italic indicate concentrations exceed both guidelines, as shown in the table; *(de Deckere et al., 2011); **Sediment Quality Guidelines for the protection of aquatic life (CCME, 1999); ***Neal and Robson (2000).*

In contrast, elevated concentrations of Pb in particular for the size fractions < 63 µm at the other sites (in particularly absence of double yellow lines), suggests that other traffic related inputs, such as wheel bearings, car paints, vehicle exhaust are likely sources of Pb in the road-traffic environment (Ewen et al., 2009; Napier et al., 2008). Similarly, the other heavy metals studied here are also found to be linked with the road-traffic emission literature. Briefly, Zn and Ni is primarily linked with tyre wear; while Cd, Cr and Cu are linked with brake wear and exhaust emissions (Charlesworth et al., 2003; Ward et al., 2004; Zanders, 2005). To relate these sources to the present study it can be noted that as traffic passed through all the above sites (except straight road section), it experienced frequent acceleration and deceleration, which may have an influence on larger metal concentrations, as also reported by Ewen et al. (2009), for traffic undergoing 'stop-start' activities. This is true for the RB and RSC sites in particular (almost all traffic was observed to undergo braking during the sampling period) compared to the RI site (controlled braking) and the SR site (steady speed), as discussed in section 3.1.

3.3. Contamination Assessment

3.3.1. Road Sediment Quality Assessment

Heavy metal concentrations (combined all sites and both size fractions) in dry, runoff and snow derived sediments (taken from Table 4) were compared with several trigger concentrations (used to monitor hazard assessment relating to sediment quality), see Table 6. In the absence of particular guidelines for road sediment in the UK, these alternatives are often used to gain an impression of how hazardous the road sediment could be in relation to ecological aspects, as evident from their previous used in literature (e.g. Heal et al., 2006; Shi et al., 2010). The mean suspended sediment associated metal concentrations from rural rivers in the UK are also shown alongside. Although the three sets of guidelines give rather inconsistent metal concentration values, a few similarities are found in the order of metal concentrations. For example, for both triggers, the highest concentration is for Zn followed by Cr, while the lowest is for Cd, and Pb and Cu in the Dutch guideline are found in reverse order in the Canadian guideline. The UK rural river sediment quality values are consistent with the highest and lowest being for Zn and Cd, respectively. The order for the other metals is Pb, Cu and Cr.

As seen in Table 6, except for Cr and Ni, all metal concentrations in dry, runoff and snow derived sediments exceed or equal their guideline values in the Dutch and Canadian standards. Considering the exceptions, Cr in runoff sediment and Ni in both runoff and snow sediment were found higher than their values in both guidelines. Cr in runoff and Cu in all different categories of road sediments from this study were well above their typical values in UK rural river sediment, while the other metal concentrations were found to be significantly lower in the road sediment than in the river sediment. Taking the different categories of road sediments into account, all the metals in runoff sediment were found to exceed the guideline values, indicating runoff as the most dominant medium of pollutant transport from roads in the study area followed by the snow precipitation (except Cr) and the dry weather derived sediment (except Cr and Ni).

3.3.2. Pollution indices for heavy metals

Transforming heavy metal concentration levels into a single index value is often preferred to gain better understanding and decision making tools in environmental pollution research. In this regard, the degree of contamination (*CD*) and potential ecological risk (*PERI*) indices were estimated and are presented in Table 7. It may debateable whether the methods used to calculate *CD* and *PERI* proposed by Hakanson (1980) should be used for different sediment particle sizes, however the intention here is to inform readers regarding the influence of the sediment size fractions on overall pollution levels.

Table 7. Pollution indices (Degree of contamination, *CD* and Potential ecological risk index, *PERI*) for the heavy metals associated with road sediment

Site	***Category***	***Sediment size fractions 250-63 µm***		***Sediment size fractions<63 µm***		***Integrated sediment [a]***	
		CD	*PERI*	*CD*	*PERI*	*CD*	*PERI*
Road bend (RB)	Dry Sediment	8	78	23	178	14	95
	Runoff sediment	14	68	36	160	16	75
	Snow sediment	10	70	25	116	11	72
Road with speed controls (RSC)	Dry Sediment	15	84	17	241	16	100
	Runoff sediment	19	81	44	204	20	94
	Snow sediment	13	79	31	129	16	92
Road intersection (RI)	Dry Sediment	5	66	15	92	8	75
	Runoff sediment	15	64	38	173	17	80
	Snow sediment	11	69	21	105	14	79
Straight road (SR)	Dry Sediment	9	86	18	124	10	80
	Runoff sediment	17	73	43	196	20	92
	Snow sediment	11	80	29	131	13	86

a - Irrespective of sediment fraction sizes

The last two columns contain these indices for integrated samples (defined as sediment irrespective of fractions), were derived by integrating the individual size fractions data (weighted average of *CD* and *PERI* in the 250-63 µm and <63 µm size fractions), which are shown in columns 3–6.

Based on the integrated samples (second last column in Table 7), the degree of contamination caused by metals in road sediments falls between 8 and 20, indicates low to moderate degree of contamination. *CD*≥12, suggesting a moderate degree of contamination, was found at the RSC site for all three categories of road sediments. For the other 3 sites 1 of the 3 sediment categories fell below the moderate contamination level. The *CD* for the snow derived sediment at the RB site and for the dry sediment at the SR site are only marginally below 12, while only for the dry sediment at the RI site was the level significantly below moderate.

Similarly, *PERI* (last column in Table 7) which introduces a metal's toxicity for aquatic species, ranges from 72 to 100, indicating low to moderate levels of ecological risk across the sites, and was found to be generally consistent with the pattern of *CD*. *PERI* ≥75, suggesting a moderate level of ecological risk, was found for all sediment categories for all the sites with the only exception being at the RB site for snow sediment. However, even this exception was only marginally below the moderate ecological risk level mark. Comparing the different road sediment for the pollution indices, the runoff sediment was found to be the most contaminated followed by dry and snow sediment. In contrast to the integrated sediment (irrespective of size fractions), both the *CD* and *PERI* in the sediment size fraction <63 µm (usually considered as the most chemically active) contain as high as 1.5 to 3, thus in turn lifting the pollution level to the next hierarchy of ecological risk (columns 5 and 6 in Table 7), as seen in Table 1. For example, at all sampling sites except the RI, *CD*≥24, suggesting a considerable level of contamination for both runoff and snow sediment. Conversely, *CD* associated with the larger sediment size fraction (250-63 µm), shown in column 3, displays a similar level of contamination as seen in the integrated sediment samples, but with a different magnitude. Taking potential ecological risk index into account in the sediment size fraction < 63, *PERI*≥150, suggesting considerable ecological risk needing immediate attention, was found at all the sites for the runoff sediment (column 6 in Table 7). A similar level of ecological risk was also found at the RB and RSC sites for the dry road sediment, while none of the snow sediment at the sites showed such a high risk. The lowest ecological risk for snow sediment was because of the lower concentration of the most toxic metals Cd (toxic factor = 30) compared to other two sediment categories. Furthermore, *PERI* associated with larger sediment size fraction (250-63 µm), shown in column 4, displays a low to moderate level of ecological risk as found for the integrated sediment, but with a different magnitude.

It is therefore clear that pollution indices based on integrated sediment (irrespective of size fractions) may underestimate the level of ecological risk. However, as the local background values of metals (derived from integrated sediment) were also used for different sediment size fractions, the probable higher pollution indices for the finer sediment particles could be biased. Therefore, these pollution indices derived for finer sediment fractions need to be used with caution. In future work related to this, local background values for metals in different size fractions could be used to derive the pollution indices, which may eliminate the present limitations.

4. Conclusions

This study characterises heavy metal concentrations in dry, wet weather (runoff and snow) derived sediment at four different sampling sites, representing typical road lay-outs on the Riccarton Campus road network at Heriot Watt University in Edinburgh. In addition, pollution indices derived from heavy metal concentrations were assessed. The following conclusions are drawn from this study:

- Road sediments on Riccarton Campus are highly contaminated by heavy metals compared to their local background values, signifying an anthropogenic input most likely from the road-traffic environment.
- The smaller sediment size fractions (<63 µm) contain approximately 2 to 4 times greater metal concentrations for the metals except Pb, compared to the larger sediment size fractions (250-63 µm) for all the sampling sites.
- The pollutant concentrations for most of the metals are significantly higher in runoff sediment followed by the snow and dry sediment for all the sampling sites.
- The assessment of heavy metal contamination in the study area indicates that there is a low to moderate level of risk associated with road sediments; among which runoff is the most dominant medium of pollution followed by the snow. However, pollution levels could potentially reach considerable levels of ecological risk (largest being associated with <63 µm size fractions) when the influence of sediment particle size fractions are considered.
- The highest pollutant concentrations and associated contamination across the sampling sites were found in the order of the road with speed control > straight road > road intersection > road bend sites. This suggests that site specific attributes are the primary factor (e.g. road lay-outs, road condition) for the difference in metal concentrations and pollution levels compared to traffic volume alone.

It is hoped that the findings may inform heavy metal emission patterns associated with traffic movement patterns conditioned by road lay-outs commonly found in sub urban roads and assist water professionals to develop an effective strategy for controlling pollutants with the aim of managing urban diffuse pollution. In this line, future work related to this, local background values for metals in different

size fractions could be used to derive the pollution indices, which may eliminate the present limitations.

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