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"Gheorghe Asachi" Technical University of Iasi, Romania

SPATIAL VARIABILITY OF HEAVY METAL POLLUTION POTENTIAL FROM AN URBAN ROAD NETWORK

Sudip K. Pal[1]*, Steve G. Wallis[2], Scott Arthur[2]

[1]Department of Civil Engineering, Chittagong University of Engineering & Technology, Chittagong-4349, Bangladesh
[2]School of the Built-Environment, Heriot-Watt University, Riccarton, Edinburgh, EH14 4AS

Abstract

This study investigated the spatial variability of heavy metal emission patterns and associated measures of contamination on Riccarton Campus at Heriot-Watt University in Edinburgh, Scotland. Road deposited sediments were collected from 12-different sites representing typical urban/sub-urban road layout over a 10 month period covering seasonal variations. The heavy metal concentrations of the road sediments collected from different sites of a road were determined by strong nitric acid digestion and atomic absorption spectrometry. The contamination levels of the heavy metals in the road deposited sediments were assessed by the accumulation index, the degree of contamination and the ecological risk index. The outcomes of the investigation showed highly site-specific heavy metal emissions that primarily varied with road lay-out, with also influences from road surface condition, surrounding land use and traffic volume. The degree of contamination and the associated ecological risk index revealed that bus stops, a road bend, a road with speed control measures and a road intersection site were the pollutant hot-spot areas among all the sites that may likely pose moderate to considerable levels of pollution to the nearby water environment.

Key words: atomic absorption spectrometry, Edinburgh, metal contamination, road deposited sediments, heavy metal

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1. Introduction

Roads are an integral part of any development in urban areas, and although roads comprise a small share of urban land uses, they often generate various types of pollutants, among which heavy metals and hydrocarbons are very common. The pollutants derived from roads are found from diverse sources among which atmospheric deposition, input from traffic and road surfaces are often key (Irish et al., 1995; Manohar and Kavuri, 2017). Their concentrations in road deposited sediment (RDS) and in road runoff have been found to be highly variable and dependent on a wide range of factors including location, traffic volume, extent of dry spells, frequency of street sweeping and the nature of road surfaces. Conventional road drainage through underground pipe systems is primarily designed to cope with water quantity, overlooking water quality issues. However, water quality has become increasingly important, due to pollution from road runoff impacting on the terrestrial and aquatic environments of nearby water courses or groundwater (Charlesworth et al., 2003; Karim et al., 2014; Robertson and Taylor, 2007; Virad et al., 2004).

Sustainable urban drainage systems (SUDS) for roads, replacing or modifying existing designs, are increasingly seen as long-term robust options. However the integration of roads and SUDS has revealed technical difficulties in the collection of runoff from road surfaces and perceived conflicts between SUDS design requirements and current highway management practices. Nonetheless, site control, among other treatment options, is often suggested by experts for dealing with road runoff, with the advantage of leaving moderately clean water to

* Author to whom all correspondence should be addressed: e-mail: sudiphwu@gmail.com; Phone: +88(0) 31 714948; Fax: +88(0) 31 714948

flow to regional controls, which are often designed for amenity and urban biodiversity benefits.

For site control measures, it is essential to identify the priority locations, based on pollution levels along the road network, to select the most appropriate treatment systems. However, only a limited study exist highlighting site specific variability (Ewen et al., 2009; Hjortenkrans et al., 2006; Pal et al., 2012). Furthermore, it has been revealed that irrespective of traffic volume particular aspects of road lay-out such as, road bends, speed control measures, intersections, roundabouts etc. may have a significant influence on how and where metals are dispersed (Ewen et al., 2009; Hjortenkrans et al., 2006; Pal et al., 2011).

Based on the above background, this paper investigates the spatial variability of heavy metal pollution using the Riccarton Campus road network of Heriot-Watt University as a study site. The specific aim of this study is to identify hot-spot areas based on metal concentrations and associated pollution levels. The metals in focus were Cd, Cr, Cu, Zn, Pb and Ni which are frequently found in high amounts in road traffic environments and are treated as priority pollutants for ecological risk (Karim et al., 2014). It is hoped that the identification of pollutant hot-spots may aid the design of appropriate SUDS tools for source/site control measures which in turn reduce pollution load for regional control.

2. Methodology

2.1. Site selection

Heriot-Watt University's Riccarton Campus (latitude: 55.91^0 and longitude: -3.31^0) is located southwest of Edinburgh city. 12 different sites were selected to represent typical road lay-outs. The roads are used by a variety of personal and commercial vehicles and some are parts of bus routes. The details of the sites are presented in Table 1.

2.2. Sampling RDS for metal analysis

RDS was collected from the sites shown in Table 1 over a 10 month period for different dry spells (between 1 to 14 days) after any significant rainfall events or any mechanical street cleanings. The number of samples collected from each site varied from 3 to 25. Sampling plots comprising a 1 m^2 road surface area for both positions were initially cleaned by repeated sweeping using brushes. During sampling, care was taken to minimise sweeping pressure so that (artificial) detachment of road material particles could be avoided. Details of sample collection technique and strategies can be found in Pal (2012).

2.3. Laboratory analysis

The samples were dried 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 etc. A small portion of RDS (1.0 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 (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 and 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 in Pal (2012).

2.4. Heavy metal pollution assessment

A modification of the degree of contamination (CD) and the potential ecological risk index (RI) suggested by Hakanson (1980) were applied to assess the RDS heavy metal contamination. The CD and RI are defined by the equations, as seen in Eqs. (1) to (4):

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

where C_f^i is the single metal pollution index; 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 (Table 2).

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

where n is the total number of metals (n=6 for this study).

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

Site No.	*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*	*No. of sample (n)*
1	Roundabout (G-4)	White/Red	Good	Lamp post/ signs/ gully	Braking	200	AA, CP, OB, FP	4
2	Straight Road (G-4)	Yellow	Good	None	Steady speed	200	AA, FPS	4
3	Road Bend	Yellow	Good	Gully/lamp post	Braking	200	AA, FPS	25
4	Straight Road (RAN)	Yellow	Good	Lamp post/signs	Steady speed	200	OB, FPS	4
5	Bus Stop (RAN)	Red/ blue/yellow	Good	Bus shelters/ sign post	Braking	230	OB, CP, FP	3
6	Road with speed controls	White/ yellow	Poor	Lamp post/ signs/ pedestrian crossing with barriers/gully	Braking	250	OB, CP, FPS	25
7	Straight Road (RP)	Yellow	Good	Sign post	Steady speed	265	OB, CP, FPS	4
8	Road Intersection	White/yellow	Fair	Traffic lights/ gully	Braking	285	OB, MR. FP	25
9	Straight Road (Avenue)	Yellow	Good	Sign posts	Steady speed	650	CP, MR, FP	25
10	Roundabout (Avenue)	White	Good	Road barriers/ sign post	Braking	650	OB, FPS	4
11	Bus stop (Avenue)	Red/ blue/yellow	Good	Sign post	Braking	550	SH, OB, FP	3
12	Car Park	White	Fair	Lamp post	Accelerating	--	--	10

G = Gait No., RAN= Research Avenue North, RP = Research Park, 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

Four categories of CD were used with modification after Duong and Lee (2011).

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

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.

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

where *RI* is the potential ecological risk caused by the overall contamination, signifying the sensitivity of the biological community to the toxic substances in the overall contamination. As the number of pollutants considered in this study are different to Hakanson's study, (As and Hg are not taken into account for present study) an adjustment of the indices was made and categories were also classified accordingly (Duong and Lee, 2011; Zhu et al., 2008).

3. Results and discussions

3.1. Total RDS metal concentration

The mean metal concentrations of the six metals (Zn, Cu, Cd, Cr, Ni and Pb) in RDS from the 12 study sites are presented in Table 2. The background concentrations of the metals from Riccarton Campus are also shown along with regional background values for Scotland (Appleton, 1995). The local background values for all the metals except Pb are similar to the regional background values. The accumulation index (AI), which is the ratio of the mean to the local background mean, is also presented in Table 2.

As seen in Table 2, considering all the sites the highest mean concentration of metals are found in the order of 287, 170, 99, 31, 24 and 7 mg/kg for Zn, Pb, Cu, Cr, Ni and Cd, respectively. The mean concentrations reported here are very similar to the concentrations reported previously for residential catchments in the UK (Charlesworth et al., 2003; Deletic and Orr, 2005; Robertson and Taylor, 2007) and other parts of the world (Karim et al., 2014; Kim et al., 1998; Sutherland, 2003).

Similarly, the highest accumulation index of metals Cd, Pb, Cr, Zn and Cu were approximately 7, 6.1, 3.9, 2.7 and 2.3, respectively, for all sites. These ratios are also found to be consistent with other studies (e.g. Hjortenkrans et al., 2006; Zhu et al., 2008). Note that the accumulation index for Ni was either significantly less than 1 or only marginally greater than 1 for all sites. The probable sources of heavy metals in the RDS at the different sites are discussed below.

Table 2. Mean metal concentrations (mg kg^{-1}) in RDS on road network in study area

Site	***1 (4)***	***2 (4)***	***3 (25)***	***4 (4)***	***5 (3)***	***6 (25)***	***7 (4)***	***8 (25)***	***9 (25)***	***10 (4)***	***11 (3)***	***12 (10)***	***Local background mean[a]***	***Regional background mean[b]***
Zn	225	160	252	133	205	268	213	195	287	110	167	209	107	120
AI	**2.1**	**1.5**	**2.4**	1.2	**1.9**	**2.5**	**2.0**	**1.8**	**2.7**	1.0	**1.6**	**1.9**		
Cu	67	40	89	41	74	97	59	99	88	31	69	59	44	46
AI	**1.5**	0.9	**2.0**	0.9	**1.7**	**2.2**	1.3	**2.3**	**2.0**	0.7	**1.5**	1.3		
Cd	1.3	0.7	1.5	0.7	7	1.5	1.3	2.2	1.9	0.4	6	1	1	1.4
AI	1.3	0.7	**1.5**	0.7	**7**	**1.5**	1.3	**2.2**	**1.9**	0.4	**6**	1		
Cr	6	18	19	31	28	20	25	14	15	12	10	4	8	n.a.
AI	0.8	**2.3**	**2.4**	**3.9**	**3.5**	**2.5**	**3.2**	**1.8**	**1.9**	**1.5**	1.3	0.5		
Ni	6	5	20	6	5	18	9	11	19	23	24	4	20	n.a.
AI	0.3	0.3	1	0.3	0.3	0.9	0.5	0.6	1.0	1.2	1.2	0.2		
Pb	35	77	125	112	66	170	68	53	69	35	23	43	28	115
AI	1.3	**2.8**	**5**	**4**	**2.4**	**6.1**	**2.4**	**1.9**	**2.5**	1.3	0.8	**1.5**		

Note: Value in brackets under site represents number of sample analysed; AI: Accumulation index - number ≥1.5 in bold to identify sites where metal from anthropogenic sources may likely be key; a metal concentrations were measured in RDS from road sites which carry no traffic; b Appleton, 1995

Cd, which is primarily linked to exhaust emissions, shows much higher concentrations at both bus stop sites indicating exhaust emissions from buses as a likely source with a possible influence from the painted road surface. Similarly, significant Pb concentrations at the road with speed control measures (Site 6) and the road bend (Site 3) is likely due to double yellow lines (Deletic and Orr, 2005), while, in contrast, moderate Pb concentrations at other sites (in absence of yellow lines e.g. Site 9) suggest traffic as a likely source (Ewen et al., 2009). Higher Cr, Zn and Cu concentrations across the sampling sites are related with tyre wear, wear and tear of brake pads and linings, body rust etc (Hjortenkrans et al., 2006; Zhu et al., 2011; Zhao et al., 2017), while relatively low concentrations of Ni may be linked with geological background of the study site (DeMiguel et al., 1997).

Furthermore, Table 2 reveals that metal concentrations show site specific variability across the road network. Among the 12 different sites a road bend (Site 3), a bus stop (Site 5), a road with speed control measures (Site 6), a road intersection (Site 8) and a straight road (Site 9) show AI ≥ 1.5 for all metals except Ni, suggesting that RDS for these sites are highly contaminated by heavy metals related to road-traffic in addition to atmospheric deposition. Based on the average value of AI for all metals (except Ni) the order of the contamination is found as bus stop (Site 5: AI = 3.3) > a road with speed control measure (Site 6: AI = 3.0) > a road bend (Site 3: AI = 2.7) > a straight road (Site 9: AI = 2.2) > a road intersection (Site 8: AI = 2.0). It is interesting to note that traffic counts of 230, 250, 200, 650 and 285 vph for the above sites, respectively, (Table 1) and higher concentrations of metals were not found to be correlated in this study. Nonetheless, all the above sites except a straight road (Site 9) are somewhat related with acceleration and deceleration where traffic often undergoes 'stop-start' activities that are likely linked to metal emissions (Ewen et al., 2009; Pal et al., 2011).

Apart from these five sites (Site 3, 5, 6, 8 and 9) all other sites show average AI ≤ 1.5 suggesting that RDS in these sites are less contaminated by heavy metals and are very unlikely to pose any threats to nearby environments at this stage. However, it is clear that traffic volume alone is not sufficient to explain the data unless site specific attributes (e.g. road lay-out, road surface condition and surrounding land use) are taken into consideration. Similar suggestions were also noted by Irish et al. (1985), Barrett et al. (1993) and Nishimiyimana et al. (2016).

3.2. Assessment of heavy metal pollution

Heavy metal concentrations for all the sites were further assessed for possible pollution linked to RDS being a direct threat to the nearby water environment. To better assess this, the degree of contamination (CD) and the associated potential ecological risk index (RI) suggested by Hakanson (1980) were estimated, illustrating the overall pollution caused by heavy metals, as shown in Figs. 1 and 2, respectively. As seen in Fig. 1, except the roundabout (Site 10) all the sites fall between moderate to considerable degree of contamination. CD ≥ 12, suggesting considerable degree of contamination, were found in the order of 17.5 for a bus stop (Site 5) followed by 16, 14, 12.5 and 12 for a road with speed control measures (Site 6), a road bend (Site 3), a bus stop (Site 11) and a straight road (Site 9), respectively. RDS from both bus stop sites indicate very high contamination by Cd, considerable contamination by Cr, and moderate contamination by Zn and Cu. Similarly, Sites 3 and 6 are found contaminated mainly with Pb followed by Cr, Zn and Cu. Sites 4 and 9 touch the upper limit of moderate degree of contamination dominated mainly by Pb along with an influence of Cr, Zn and Cu.

The degree of contamination caused by heavy metals for this study is found to be consistent with previous studies of RDS elsewhere (Zhu et al., 2008; Duong and Lee, 2011). Fig. 2 illustrates the potential ecological risk (*RI*) caused by heavy metals at different sites in the study area. *RI* values show a large spatial variability, ranging from approximately 25 to 240 across the sites.

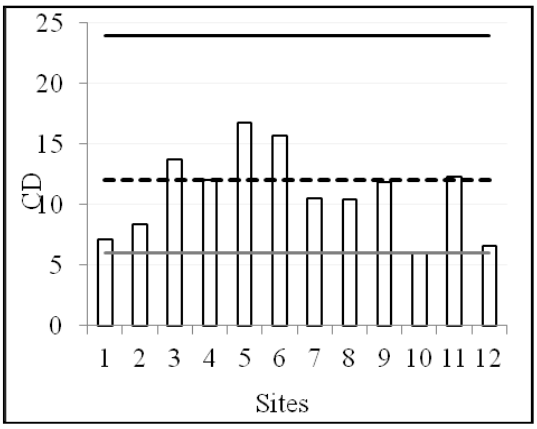

Fig. 1. Histogram of degree of contamination in RDS. Grey, dotted and solid lines represent upper limits of low, moderate and considerable degree of contamination, respectively

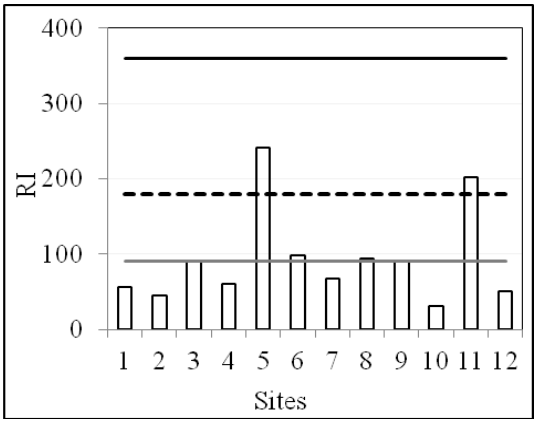

Fig. 2. Histogram of potential ecological risk in RDS. Grey, dotted and solid lines represent upper limits of low, moderate and considerable levels of risk, respectively

The values are consistent with previous studies carried out by Zhu et al. (2008) and Duong and Lee (2011). The assessment of ecological risk for metals in RDS for all sites as shown in Fig. 2 reveals that both the bus stop sites (Site 5: RI = 240 and Site 11: RI = 200) have heavy metal emissions that may have moderate to considerable levels of ecological impact if transported to the nearby water courses. Both sites are highly dominated by Cd (toxic factor =30) as mentioned earlier. However, as the number of samples analysed for both the bus stop sites (n=3) are very low, pollution levels for these sites should be used with caution. Furthermore, from Fig. 2 Sites 3, 6, 8 and 9 may also pose low to moderate level of risk (RI = 90-100) primarily by Pb and Cu (toxic factor, Pb = Cu = 5) along with Cd and Zn (toxic factor = 1). Metal contamination for all other sites (1, 2, 4, 7, 10 and 12) falls well below the low level of ecological risk.

4. Conclusions

Spatial variations of heavy metal emission patterns were found across 12 sites on the Riccarton Campus road network. Site specific attributes were found to be drivers of the difference in heavy metal concentrations in the RDS: primarily road lay-out, with other influences from road surface condition, surrounding land use and traffic volume. The accumulation index, degree of contamination and potential ecological risk associated with heavy metal pollution were further assessed to identify 'hot-spot' sites. Based on the results, a road bend (Site 3), bus stops (Site 5 and 11), a road with speed control measures (site 6), a road intersection (Site 8) and a straight road (Site 9) were identified as pollutant hot-spot sites that are very likely to pose a risk to the urban ecological system in the study area.

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