

Assessment of heavy metals emission from traffic on road surfaces⁺

Research Article

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Received 30 September 2010; Accepted 22 November 2010

Abstract: This study aims to analyse RDS heavy metal concentrations on road deposited sediment (RDS) using Riccarton Campus of Heriot-Watt University, Edinburgh, Scotland as a study site. RDS samples were collected at two transverse positions from different sites over a 4 month period in order to describe the influence of traffic on heavy metal emissions. The heavy metal concentrations of the RDS were determined by strong nitric acid digestion and atomic absorption spectrometry. The mean concentrations for Zn, Cu, Cd, Cr, Ni, Pb and Fe were found to be 213, 57, 1, 16, 15, 118, and 13497 mg kg⁻¹ from samples near to the curb and 211, 79, 2, 15, 9, 35, and 14276 mg kg⁻¹ from samples 1 m from the curb respectively. Furthermore for both positions the highest concentrations for all metals were associated with the finer fraction (<63 μm) and stronger correlations between the metals were found further from the curb than near the curb, indicating that metals accumulating on the road surface further from the curb may likely be from the same source (traffic), while the sources of metals near the curb are more diverse.

Keywords: Edinburgh • Heavy metals • Atomic absorption spectrometry • Road deposited sediment • Particle size

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1. Introduction

Road and highway surfaces are impervious, and serve as temporary sinks for various types of pollutants that are washed off during rainfall to the surrounding environment [1-3]. Furthermore both the water quality of road runoff and the air quality near roads are directly affected by the characteristics of the sediment associated pollutants deposited on the roads. It is also noted that road traffic pollution poses an environmental threat for nearby farmland used for agricultural purposes [4-5]. Several local, national and international studies also point to the traffic sector as a major contributor of diffuse heavy metal emissions [6].

Within this context, along with the degradation of urban watercourses caused by storm runoff from roads, environmental pollution from heavy metals on roads has received much attention in the recent past. The raised awareness of traffic as one of the major contributors of diffuse pollution has driven a need for a more detailed understanding of the various traffic related pollutant sources and dispersal mechanisms [7].

Road deposited sediments (RDS) play a key role in road traffic pollution assessment and comprise a complex

mix of particulates and contaminants derived from the road traffic environment which are of serious concern for environmental regulators. The characteristics of RDS vary with a range of natural and anthropogenic factors and have been found to vary significantly between geographical regions [3].

Most of the previous RDS studies have been based on samples collected within 1 m from the curb along roads [1,8,9] or from road side soils [7]. Only a limited number of studies have looked at carriageway positions further from the curb and these found that a larger proportion of particles below 150 μm are found towards center of the road compared to near the curb where larger particles are dominant. Similarly, heavy metal concentrations in road dust were higher in the centre of the road and decreased towards the road gutter - confirming their higher association with the smaller particle size fraction [10,11].

Although some previous studies have focused on Scotland, there is virtually no information on pollution build-up and heavy metal concentrations around Edinburgh. Based on the given background and the potential role of RDS in causing toxic pollution derived from the road traffic environment, this study therefore set

⁺ Presented at the 15th International Conference on Heavy Metals in the Environment, September 19-23, 2010, Gdańsk, Poland

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the hypothesis that sampling from further towards the center of the road characterises heavy metal emissions better than sampling nearer the curb. The objectives of this study were: (1) to characterise heavy metal emissions on road surfaces for both positions of near the curb and 1 m from the curb towards the center and (2) to analyse heavy metal concentrations in RDS in order to describe emission patterns in the road traffic environment. This study is based on a road network in Riccarton Campus of Heriot-Watt University, located in south-west Edinburgh, Scotland. In the following sections, descriptions of the field work and experimental procedures are given, the results and discussions are presented and conclusions are drawn.

2. Experimental Procedure

2.1 Study area

Edinburgh, Scotland's capital lies at the eastern end of the country's central belt, along the Firth of Forth near the UK's North Sea coast. Heriot-Watt University's Riccarton Campus (latitude: 55.91° and longitude: -3.31°) is located southwest of Edinburgh city and has been developed over the past 40 years with a good ecological and environmental perspective. Continuing its successful progress as a provider of quality education, it is expanding with many infrastructure developments. The Murray Burn is a river running through the campus from west to north east. Storm water drainage along with road runoff discharges to this river largely without any major treatment or attenuation. Each of the sampling sites had 2 lanes of traffic with an asphalt road surface, with low traffic densities measured as between 285 and 650 vehicles per hour, with the surrounding land area devoted to campus buildings, some farm land, foot paths and pavements. Mechanical street-sweeping on the campus by the local council occurs typically twice a year. However, from monitoring the sites, at no time during the study period was there any evidence of significant removal of RDS by this process. No previous research study has been undertaken on road runoff regarding water quality and pollution aspects from the campus roads.

2.2 Sampling and analytical procedures

RDS samples were collected from eight sites on the campus roads and for two transverse positions (from the gutter along the curb; and from 1 m further away from the curb) to characterise the the influence of road-traffic on heavy metal emissions. Sites inspection suggested that RDS particles deposited originate primarily from the

vehicles, road surface, atmospheric deposition and the surrounding land. The sites comprise a road bend, a road with speed control measures, a road intersection/traffic lights, and a typical road section to represent a typical road lay-out. Samples were collected on between 4 and 12 antecedent dry days over a 4 month period since March 2010. Sampling plots comprising a 1 m² road surface area for both positions were initially cleaned by repeated sweeping using brushes. Then RDS samples were collected by sweeping the plots using a clean plastic dustpan and brush as outlined by [1,3,12]. Several identical dustpan and plastic brushes were used in order to minimise cross contamination between the sites. The sweeping technique was also kept consistent to avoid sample variability. During sampling, care was taken to minimise sweeping pressure so that (artificial) detachment of road material particles could be avoided. Samples were then transported back to the laboratory using self-sealing plastic bags to avoid contamination.

After collection samples were air-dried at room temperature and weighed prior to further analysis. Samples were dry-sieved using a 1 mm aperture metal sieve to remove large objects, litter, plants and leaves. Then samples were wet-sieved through a 63 µm aperture metal sieve to separate them into fractions above and below 63 µm. This size has some importance for metal concentrations as suggested by various researchers, for example [13]. Wet-sieved samples were then oven dried at 105°C overnight in preparation for the metal analysis. Fig. 1 shows the flow chart of the analytical method followed for the determination of metal concentrations. Total RDS metal concentrations for the elements Cd,

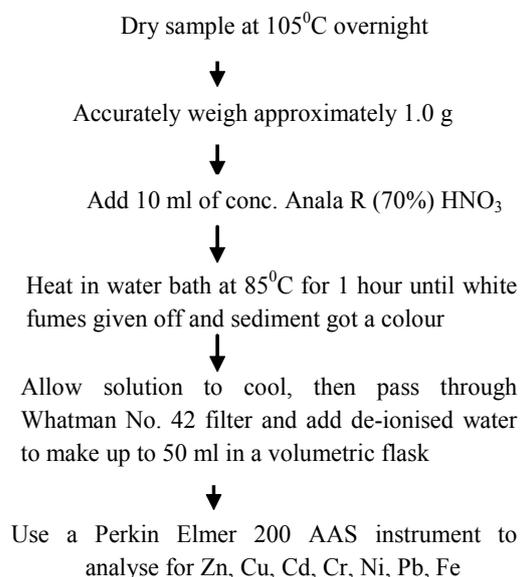


Figure 1. Analytical protocol for RDS metal determination.

Cr, Cu, Zn, Pb, Ni and Fe were determined by strong nitric acid digestion (Anala R 70%) followed by flame atomic absorption spectrometry (AAS) using a Perkin Elmer 200 AAS analyser. 1 g of sample was digested in 10 mL of concentrated nitric acid and then heated in a water bath at 85°C for 1 hour. The Perkin Elmer 200 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. Any sample that gave a reading beyond the calibrated ranges was diluted until a satisfactory reading was obtained. De-ionised water was used throughout for cleaning apparatus, preparing standard solutions and other related purposes of laboratory work in order to avoid cross contamination. Details of the analytical procedure followed can be found elsewhere [3,11].

3. Results and Discussion

3.1 Heavy metal concentration in street dust

Table 1 presents the descriptive statistics of the metal analysis for both positions for integrated samples (*i.e.*, both size fractions combined using all sampling sites). The background concentrations of the metals from Riccarton Campus are also shown along with regional background values for Scotland [14]. The local background values for all the metals except Pb are similar to the regional background values. The accumulation coefficient, which is the ratio of the mean to the local background mean, is also presented in Table 1.

The mean concentrations were found to be 213, 57, 1, 16, 15, 118, and 13497 mg kg⁻¹ near the curb, while 211, 79, 2, 15, 9, 35, and 14276 mg kg⁻¹ at 1 m from

the curb for Zn, Cu, Cd, Cr, Ni, Pb, Fe respectively. The mean concentrations reported here are very similar to the concentrations reported previously for residential catchments in the UK [1,3,11] and other parts of the world [12,13].

In general, the data presented in Table 1 suggests that the concentrations of Zn, Cr and Fe found at the two transverse positions were very similar. However, levels of Cu and Cd were respectively 1.4 and 2.0 times higher at 1 m from the curb than those near the curb. Inversely, Ni and Pb concentrations showed respectively 1.7 and 3.4 times higher levels near the curb than at 1 m from the curb.

The accumulation coefficients suggest that all metals but Ni and Fe show elevated concentrations compared to background concentrations indicating that there are some anthropogenic inputs (*e.g.* vehicular activities) on top of atmospheric deposition. Particularly, Pb followed by Cr, Zn and Cu near the curb and Cr and Zn followed by Cd, Cu and Pb at 1 m from the curb. It has been noted in the literature that all of these metals are somewhat related to traffic and are found at elevated levels in urban environments. However, the elevated Pb concentration is somewhat surprising, since Pb has been banned as an anti-knocking additive in fuel for over a decade. Additionally any residual affect from the previous use of Pb is unlikely to be associated with RDS, as RDS is characterized by the short residence times indicative of recent accumulation on road surfaces [10,15,16]. The only other likely source of Pb in RDS at this position is from particles of road paint (from double yellow lines) as noted by Deletic and Orr [11].

Table 1. Concentrations of metals (mg kg⁻¹) near curb (A) and 1 m from the curb (B) as measured between 06/03/10 and 09/07/10.

Metal	Sampling Position	Mean ± standard deviation	Median	Min	Max	Background mean ¹	Regional background mean ²	Accumulation coefficient
Zn	A	213 ± 8	192	107	457	107	120	1.99
	B	211 ± 9	172	99	460			1.97
Cu	A	57 ± 21	57	22	112	44	46	1.30
	B	79 ± 53	58	26	220			1.79
Cd	A	1 ± 0.4	1	0	2	1	1.4	0.81
	B	2 ± 0.8	2	1	4			1.84
Cr	A	16 ± 13	11	5	76	8	na	2.08
	B	15 ± 5	16	6	29			2.02
Ni	A	15 ± 6	15	6	33	20	na	0.76
	B	9 ± 2	8	3	15			0.44
Pb	A	118 ± 118	84	25	621	28	115	4.26
	B	35 ± 22	34	6	102			1.25
Fe	A	13497 ± 5164	12747	585	32176	15897	na	0.85
	B	14276 ± 3053	14502	7543	18622			0.90

¹The background concentrations were measured in RDS from roads which carry no traffic.

²Appleton, 1995

3.2 Grain-specific metal concentrations for both sampling points

For metals having an accumulation coefficient >1 , further assessment of the emission patterns for both transverse positions, but now separating the two different size fractions, was undertaken. The means (based on all sampling sites) revealed that the highest concentrations for all metals were associated with the finer fraction ($<63 \mu\text{m}$) for both positions. The only exception is Pb which shows a higher association with the $>63 \mu\text{m}$ fraction near the curb (Table 2).

This also supports the idea that the source of Pb was road markings near the curb rather than vehicles. Paired t-student tests showed that there is always a significant difference between the mean concentrations measured in the two fractions. Overall, the $<63 \mu\text{m}$ fraction contained two to four times higher concentrations of Zn, Cu, Cd, Cr and Pb than the $>63 \mu\text{m}$ fraction for both positions indicating the metals' affinity to attach to the finer fraction (Table 2).

It was also discovered that concentrations associated with the $<63 \mu\text{m}$ fraction for Zn, Cu, Cd were higher at

1 m from the curb than those near the curb, while the highest concentrations for Pb and Cr were found near the curb: particularly, the reduction was as high as 75% for Cd followed by 30% for Cu and 9% for Zn towards the curb (Table 2). Similar observations are evident from the study carried out by Harrison *et al.* [10], although the percentage changes for Cd and Cu are rather higher in the current study.

The reason behind higher concentrations for Zn, Cu and Cd at 1 m from the curb than near the curb may be vehicular emission as it was observed that usually vehicles passed directly over the 1 m from the curb position and hence any deposition of particulates, from e.g. tyres and brakes, containing heavy metals would most likely occur at this position before being transported towards the curb by vehicle induced turbulence or natural processes, such as wind, rain splatter or runoff.

3.3 Correlation between metals

To assess the influence of traffic-related sources (tyres, brakes, corrosion, exhaust fumes and pavement wear), correlations between the metals were analysed

Table 2. Concentrations of metals (mg kg^{-1}) near curb (A) and 1 m from the curb (B) in the two particle fraction sizes as measured between 06/03/10 and 09/07/10.

Metal	Sampling Position	Fraction size $<63 \mu\text{m}$		Fraction size $>63 \mu\text{m}$	
		Range	Mean \pm standard deviation	Range	Mean \pm standard deviation
Zn	A	89-720	415 \pm 146	44-381	151 \pm 65
	B	134-949	455 \pm 231	56-445	147 \pm 93
Cu	A	52-212	116 \pm 36	15-104	42 \pm 20
	B	18-794	166 \pm 169	17-137	62 \pm 41
Cd	A	0-4	1 \pm 1	0-2	1 \pm 0.5
	B	2-17	5 \pm 4	0-3	1 \pm 0.6
Cr	A	7-135	22 \pm 21	2-73	13 \pm 11
	B	0-58	17 \pm 13	4-30	14 \pm 6
Pb	A	44-338	153 \pm 66	14-669	113 \pm 140
	B	4-181	81 \pm 44	4-53	23 \pm 12

Table 3. Spearman rank correlation coefficient among metal concentrations for integrated sample*

	Zn	Cu	Cd	Cr	Ni	Pb
Position A: near curb (40 samples)						
Zn			0.617**		0.528**	0.534**
Cu				0.544**		
Cd	0.617**					
Cr		0.544**				
Ni	0.528**					0.585**
Pb	0.534**				0.585**	
Position B: 1 m from curb (25 samples)						
Zn		0.639**	0.429**	0.615**	0.745**	
Cu	0.639**		0.423**	0.481**	0.324*	
Cd	0.429**	0.423**				
Cr	0.615**	0.481**			0.455**	0.744**
Ni	0.745**	0.324*		0.455**		
Pb				0.744**		

*Significance to 0.05%; **Significance to 0.01%; blanks indicate no significant correlation.

Table 4. Comparison of observed metal concentrations with published guide line values: (A) near curb; (B) 1 m from the curb)

Group 1: Contaminants which may pose hazard to health

Contaminant		Metal concentration		Trigger concentrations (mg kg ⁻¹)		
		Size fraction >63 µm	Size fraction <63 µm	SGV ^a	SQG ^b	SQGEH ^c
Cd (B)	Mean	1	5	3*	-	-
	Max.	1-3	2-17	15**	-	-
Cr (A)	Mean	13	22	600*	250*	64*
	Max.	2-73	6-135	1000**	750**	64**
Pb (A)	Mean	112	153	500*	500*	140*
	Max.	14- 669	44-337	2000**	375**	70**

Group 2: Phytotoxic contaminants not normally hazardous to health

Contaminant		Metal concentration		Trigger concentrations (mg kg ⁻¹)		
		Size fraction >63 µm	Size fraction <63 µm	SGV ^a	SQG ^b	SQGEH ^c
Cu (B)	Mean	62	166	130	100-150	63
	Max.	17-137	18- 794			
Zn (B)	Mean	147	455	300	500-600	200-600
	Max.	56- 445	134- 949			

Concentrations exceeding trigger values are shown in bold

^a Soil Guideline values (SGV) by [17]

^b Soil Quality Guidelines (SQG) by [18]

^c Soil Quality Guidelines for Environmental Health (SQGEH) by [19]

* Open space, park, Playing ground

** any places where plants grow

for both positions. Table 3 presents Spearman's rank correlation coefficients, r , (these being used because the experimental data did not follow a Gaussian distribution). Concentrations of certain metals did correlate well with each other, and there were more statistically significant correlations between metals further from the curb than near the curb (Table 3). The most plausible explanation is that metals accumulating on the road surface farther from the curb may all be of same origin (road-traffic), while the sources of the metals along the curb are far more diverse.

3.4 Environmental significance

For metals having an accumulation coefficient >1, the highest mean (and maximum) concentration obtained from both size fractions and both sampling points (taken from Table 2) was compared with several trigger concentrations (used to monitor hazard assessment relating to soil quality), see Table 4. The highest mean concentrations were associated with size fraction <63 µm at position B (1 m from the curb) for Zn, Cu and Cd, but at position A for Cr and Pb. In addition, these highest mean concentrations for Zn, Cu and Cd exceeded the SGV trigger values, but those for Cr and Pb did not. Similar comments are also true for the maximum concentrations except for Pb where the

maximum concentration was found for the >63 µm size fraction. Note that the three sets of trigger values shown in Table 4 are rather inconsistent, but the SGV ones are used here because they are relevant to UK locations whereas the other trigger values apply to Canada.

The mean concentration for metals in street sediment near the curb are lower than the trigger concentrations (Tables 2 and 4); however they are higher than the trigger values towards the centre of the road. Therefore, an assessment of hazard associated with RDS might be misleading if sampling near the curb is used. Furthermore, concentrations may be even higher further than 1 m away from the curb so that the figures presented here may be conservative. This may have environmental consequences for all users of the campus roads and as well as for nearby farm land [4,5] and watercourses [7,8].

4. Conclusions

Monitoring of the RDS on Riccarton campus roads, Edinburgh, Scotland has shown elevated emission patterns of traffic derived heavy metals. A transverse distribution of metal concentration was observed with higher concentrations of Zn, Cu and Cd in RDS collected

at 1 m from the curb, and higher concentrations of Pb, Fe, Ni near the curb. In addition, a stronger correlation between metals was found at 1 m from the curb than near curb, indicating different sources for the metals found at the two positions. Furthermore from the assessment of metal concentrations located at 1 m from the curb, mean and maximum values for Zn, Cu and Cd were found to be well above environmental pollution trigger levels indicating potential harm to the nearby environment.

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Acknowledgements

This research was carried out under a PhD scholarship from the School of the Built-Environment, Heriot-Watt University (HWU), which is gratefully acknowledged. Sean McMenemy and Hugh Barras are thanked for technical assistant during work in Environmental Chemistry Laboratory in the School of Life Sciences at HWU.