Remediation of heavy metal impacts in roadside corridors, Wet Tropics World Heritage Area, Australia

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Abstract

Elevated heavy metal concentrations have been detected in road sediments and runoff waters throughout the world. Significant concentrations of these metals are derived from vehicle degradation and emissions. The availability of these metals to plants and lower food-chain organisms in roadside corridors is a potential concern.

This paper examines heavy metal impact along the Kuranda Range Road in Cairns, northern Australia. The Kuranda Range Road passes through a section of Wet Tropics World Heritage Area (WTWHA) listed rainforest.

The study investigated: a) Cd, Cu, Pb, Ni and Zn concentrations in road sediments along the Kuranda Range Road; b) Cd, Cu, Pb, Ni and Zn levels in runoff waters along the Kuranda Range Road, following an extended dry spell in the region; and c) the potential of zeolites to adsorb these metals from solution and reduce their dispersal into roadside rainforest streams and over soils.

Elevated concentrations of Cu, Pb, Ni and Zn were detected in road sediment samples collected from gutters on the Kuranda Range Road between October 2002 and November 2003. In some of these samples these metal concentrations were above ANZECC Draft Guidelines (2000).

Rainwater runoff samples collected in January 2003 and February 2003 contained dissolved Cu and Zn concentrations that exceeded ANZECC Draft Guidelines (2000).

The dispersal of Cd, Cu, Ni and Zn from roadside runoff waters into surrounding environments may be controlled by filter traps that incorporate zeolites. In a laboratory experiment zeolites proved extremely effective in reducing dissolved Zn loads in contaminated waters over a period of five minutes to 24 hours. However, other results from the adsorption experiment showed that zeolites were much less effective in reducing Cd, Cu, Pb and Ni concentrations in solution.

The capacity of zeolites to adsorb Cd, Cu, Pb and Ni requires further research to validate their incorporation into filtration traps in gutters to reduce the levels of these metals in runoff waters. This research will most likely involve methods such as active stirring or some kind of agitation of solution treated with zeolites.

Ideally, zeolites may be incorporated into low-cost and low-maintenance remediation traps that can be installed in drainage points along the Kuranda Range Road to reduce the aqueous dispersal of heavy metals into sensitive roadside environments. These traps may be integrated into existing or proposed road structures that pass through environmentally sensitive landscapes.

Introduction

Road sediments typically host elevated levels of heavy metals, which may be mobilised by runoff waters (Sansalone and Buchberger 1997; Sezgin et al. 2003). Motor vehicles constitute a principal source of these heavy metals (Viklander 1998; Varrica et al. 2003).

Heavy metal contaminants in road sediments are derived from: engine and brake pad wear, (e.g. Cd, Cu, and Ni) (Viklander 1998); lubricants (e.g. Cd, Cu and Zn) (Birch and Scollen 2003); exhaust emissions, (e.g. Pb) (Gulson et al. 1981; Al-Chalabi and Hawker 2000; Sutherland et al. 2003); and tyre abrasion (e.g. Zn) (Smolders and Degryse 2002). A smaller proportion of these metals are typically derived from local rock sources.

Sediment-bound metals have been reported to be extremely mobile in runoff waters, in both the dissolved phase (Dierkes and Geiger 1999; Rose et al. 2001) and as fine, suspended particles (Sansalone and Buchberger 1997; Turner et al. 2001).

The metals Cd, Cu and Zn have been reported to have been primarily mobilised in 'first flush' waters, while Pb has been reported to show weak mobility in first flush runoff waters (Sansalone and Buchberger 1997).

The availability of heavy metals to plants as well as lower food-chain organisms in roadside corridors is a potential concern (Thomson et al. 1997; Sutherland and Tolosa 2001). Elevated concentrations of heavy metal contaminants in road runoff waters can result in their accumulation in roadside soils in toxic levels (Turner et al. 2001).

Recently, a number of approaches have been investigated to limit the dispersal of heavy metals from road runoff waters into roadside streams and soils. The implementation of retention ponds (Lee et al. 1997), filtration systems comprising organic matter (Zhou et al. 2003) and grassed embankments (Dierkes and Geiger 1999) are examples of methods that have been employed in roadside settings with varying degrees of success.

There have also been investigations into the capacities of various mineral and organic substances to sequester heavy metal mobility in contaminated roadside sediments, soils and water bodies (Alvarez-Ayuso and Garcia-Sanchez 2003).

Only few investigations into the composition and chemistry of road sediments or road dust have been conducted in Australia (Birch and Scollen 2003). There is scarce data on heavy metal distribution in sediment, soils and runoff waters in tropical North Queensland, which has experienced rapidly increasing traffic volumes in the past ten years (Table 1). Further to understanding the chemistry of heavy metal contaminants in roadside corridors in these locations, there exists a great challenge to investigate methods to effectively limit the dispersal of these contaminants into roadside corridors whilst incurring minimal disturbance to surrounding natural environments.

Aims and Approaches

This paper examines heavy metal impact along the Kuranda Range Road in Cairns, northern Australia. The Kuranda Range Road passes through a section of Wet Tropics World Heritage Area (WTWHA) listed rainforest. Proposed upgrading works on this road are likely to be accompanied by an increase in traffic volume in this region. This is likely to generate a marked elevation in metal contamination, derived from automotive sources, in soils and sediments along the Kuranda Range Road.

Specifically, the aims of this study were to: a) measure concentrations of Cd, Cu, Pb, Ni and Zn in gutter sediments along the Kuranda Range Road; b) measure the levels of Cd, Cu, Pb, Ni and Zn in rainwater runoff waters along the Kuranda Range Road following an extended dry spell in the region; and c) investigate the potential of zeolites to adsorb these metals from runoff waters and limit their dispersal into roadside rainforest streams and soils.

Materials and Methods

Sample locations

The location of the research area is shown on Figure 1. Six road sediment sample locations were selected on the Kuranda Range Road. These locations are shown on Figure 2. Locations K1, K2, K5 and K6 were targeted as representative bends on the road, whereas locations K3 and K4 were investigated as representing straight sections on the road.

Figure 1 – Location of the Kuranda Range Road, from (Forbes et al. 2002; Kuranda Map 2003)

Figure 2 – Road sediment and runoff water sample locations on the Kuranda Range Road

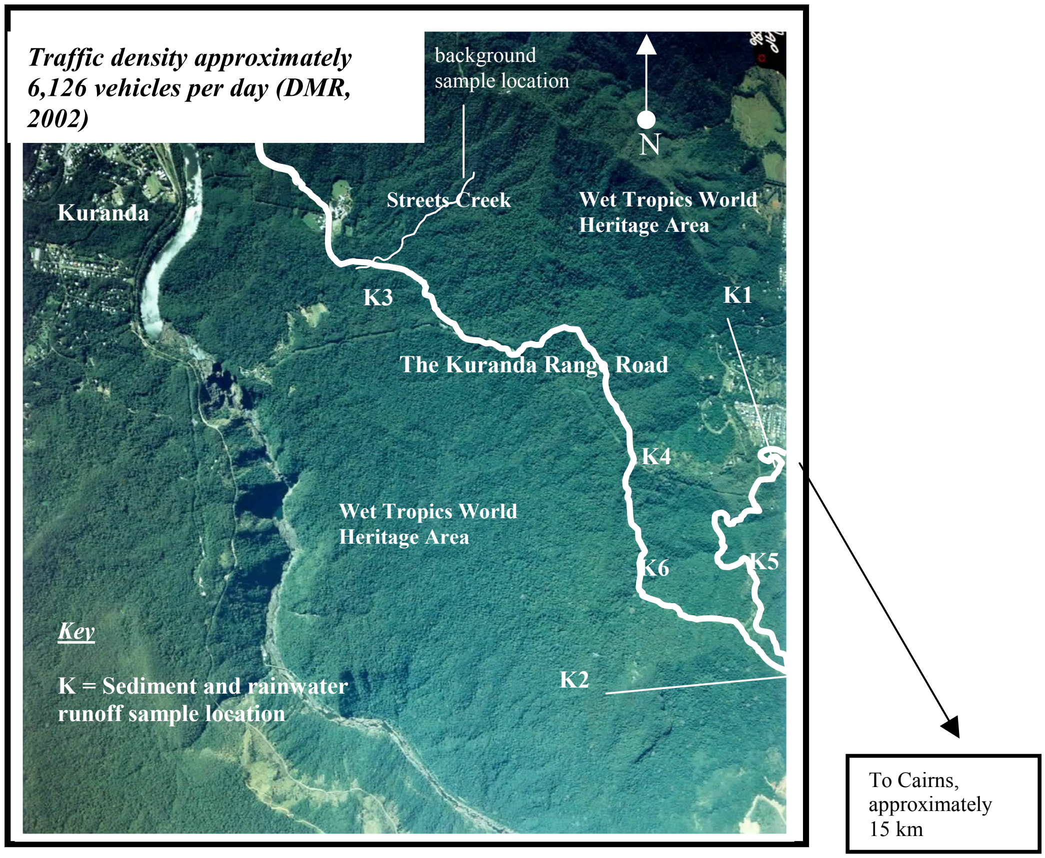


Table 1 – Daily traffic volumes on the Kuranda Range Road over the past 10 years (DMR - Traffic density data 2002)

Year	*Annual daily traffic volume (vehicles/day)*	*Growth %*
1991	4000	
1993	4905	10.3
2000	6451	3.0
2001	5912	-8.4
2002	6126	3.6

Collection and preparation of road sediments

Road sediments were swept from concrete gutters using a nylon broom and dust pan in October 2002. In one location, where concrete gutters were absent (K3), sediments were swept from the bitumen surface at the road's edge. The sediments were dried in paper bags and 75 g of sediment from each location were sieved through a 2mm nylon mesh to remove coarse debris (Sutherland and Tolosa 2001).

This procedure was repeated in August 2003 and November 2003.

All sediments were analysed for total Cd, Cu, Pb, Ni and Zn concentrations through inductively coupled plasma – mass spectrometry (ICP-MS) at Australian Laboratory Services (ALS) in Brisbane.

Composite sediment samples, comprising material collected in October 2002 and November 2003, were prepared from the remaining sediments.

Grain size distribution analysis of road sediments

Two-hundred grams of the composite sediment sample collected in October 2002 were passed through a series of sieves with sieve size intervals shown in Figure 3. The weight of sediment captured in each sieve fraction was recorded and the material was analysed for total Cd, Cu, Pb, Ni and Zn, concentrations through ICP-MS at ALS.

Collection and preparation of rainwater runoff samples

In January 2003 six plastic bottles with floating stop devices were placed in brackets in concrete drains at each of the sample locations shown on Figure 2. Due to the poor design of the brackets only one bottle, placed at location K2, managed to collect a significant volume of runoff water during a rainfall event. This water was filtered through 0.45 µm pore-size filter paper, poured into a plastic bottle rinsed with HNO_3 at pH <2 and submitted to the Advanced Analytical Centre (AAC) in Townsville for Cd, Cu, Pb, Ni and Zn concentrations through ICP-MS.

Following a redesign of the brackets, six more plastic bottles with floating stop devices were fixed into the same sampling locations, except for K5, where no bottle could be secured. Collected runoff water was analysed as described above.

The 5-minute, 1-hour and 24-hour zeolite treatment experiment

An upright section of clear plastic tubing (0.5 m length) underlain by a section of 180 µm sieve mesh and a plastic funnel was placed into a column-stand. A portion of the composite sample of road sediment collected from the Kuranda Range Road in November 2003 was placed into the clear plastic tubing. Distilled water was poured through the sediment in the column and captured in a glass beaker.

Six other beakers were prepared; three containing 50 g of clinoptilolite zeolites (acquired from Supersorb environmental, Western Australia) and three without zeolites.

The leachate collected from the column was split into six equal portions which were poured into the six beakers, so that there were three sets of beakers.

One pair of beakers (one beaker with and one beaker without zeolites) was sampled after 5 minutes of settling, another pair was sampled after one hour and the last pair was sampled after 24 hours.

Again, samples were filtered, poured into plastic bottles rinsed with HNO_3 at pH <2 and submitted to the AAC for analyses for Cd, Cu, Pb, Ni and Zn through ICP-MS.

All equipment was rinsed in 1M HCl and distilled water during the different stages of the experiment.

Results

Heavy metals in road sediments

Table 2 – Heavy metal concentrations (mg/kg) in road sediments from sample locations shown on Figure 2

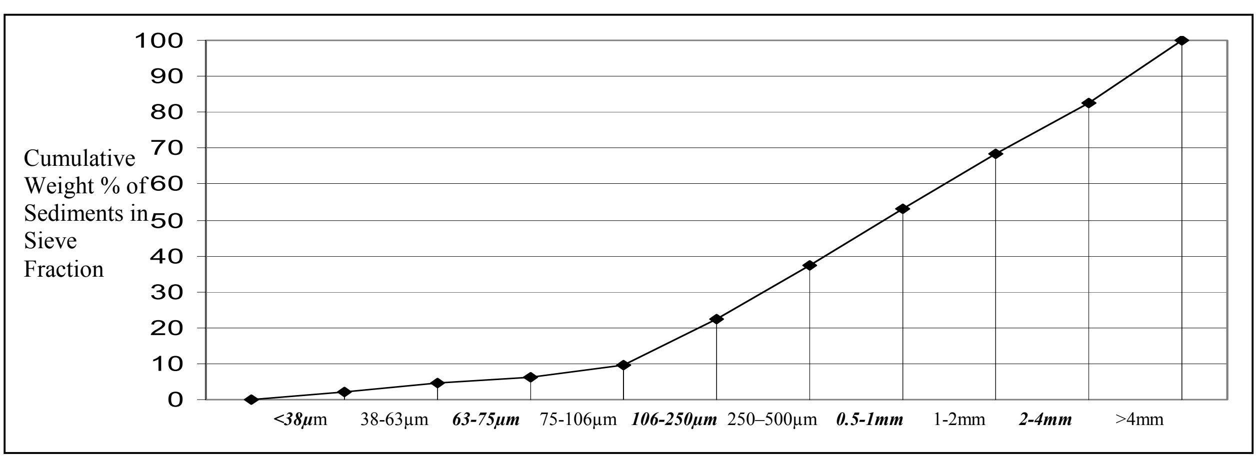
		Sample Location K1			***Sample Location K2***			***Sample Location K3***		
	Date	*Oct - 2002*	*Aug - 2003*	*Nov - 2003*	*Oct - 2002*	*Aug - 2003*	*Nov - 2003*	*Oct - 2002*	*Aug - 2003*	*Nov - 2003*
	*ANZECC Guidelines for sediments **									
Cd	1.5	0.19	0.05	0.26	0.29	0.21	0.25	0.13	0.15	0.14
Cu	65	***34***[x]	19.2	49.6	***44***[x]	40.8	46.4	***66***[x]	33.1	49.5
Pb	50	123	21.1	53.5	53.7	53	146.5	174	36.5	160.5
Ni	21	***36***[x]	34	41.2	***94***[x]	43.9	29.6	***29***[x]	42.2	43.9
Zn	200	1100	112	1155	2200	1150	2070	235	900	174

		Sample Location K4			***Sample Location K5***			***Sample Location K6***		
	Date	*Oct - 2002*	*Aug - 2003*	*Nov - 2003*	*Oct - 2002*	*Aug - 2003*	*Nov - 2003*	*Oct - 2002*	*Aug - 2003*	*Nov - 2003*
	*ANZECC Guidelines for sediments **									
Cd	1.5	0.08	0.2	0.18	0.14	0.25	0.24	0.19	0.25	0.12
Cu	65	***32***[x]	43	19.4	***31***[x]	42.1	44.9	***60***[x]	71	32.9
Pb	50	55	55.4	11.8	29.3	422	34.6	43.9	109.5	39.6
Ni	21	***38***[x]	47.6	44.1	***60***[x]	47.7	51	***75***[x]	28.7	36.4
Zn	200	357	1025	268	804	451	1410	966	1045	647

Notes: *ANZECC Guidelines for sediments ** = ANZECC Recommended Sediment Quality Draft Guidelines (2000)
Shaded values exceed ANZECC Guidelines
[x] = Analysed by X-ray fluorescence

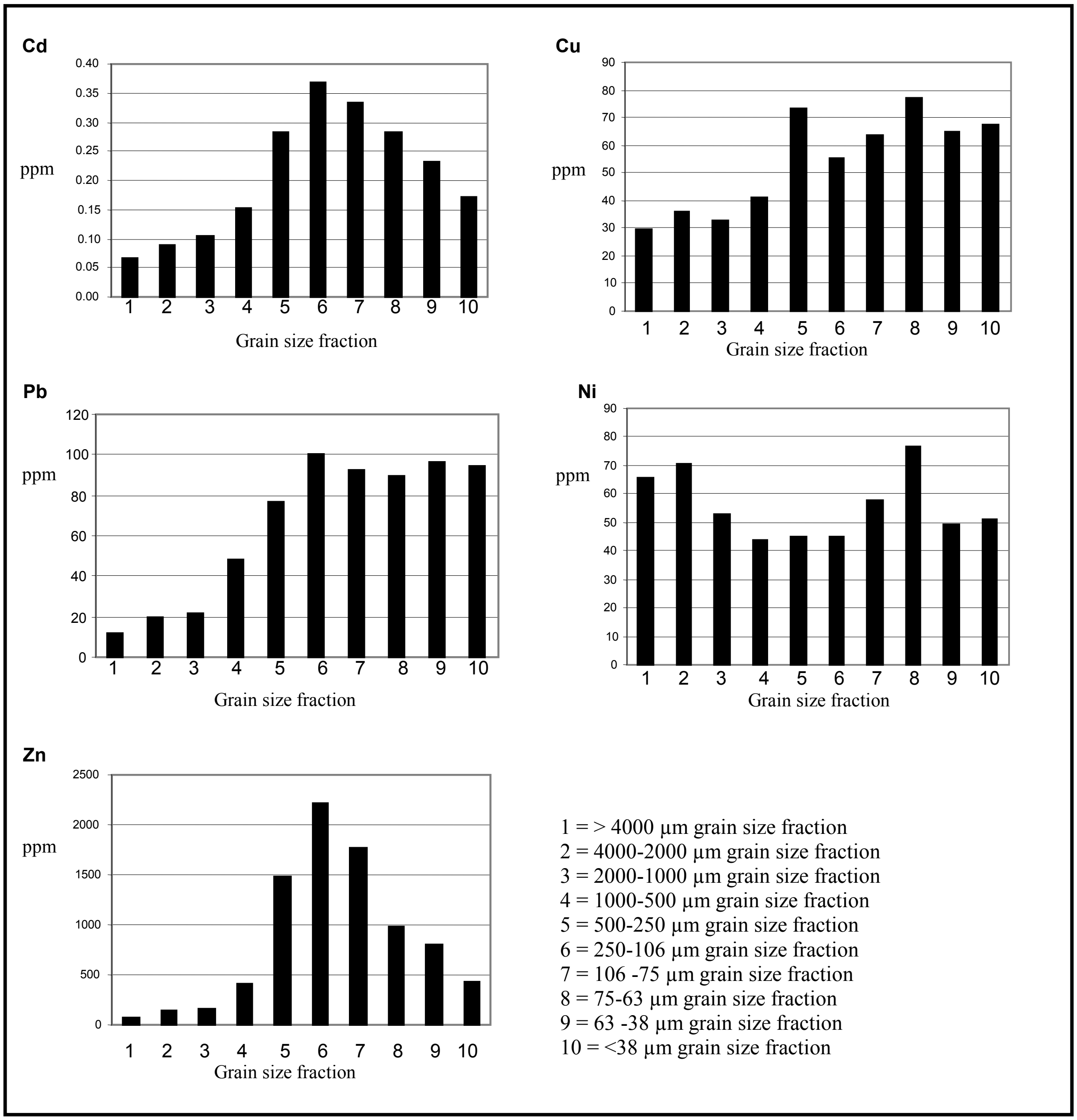
Elevated heavy metal concentrations in road sediments were detected at all sampling locations shown on Figure 2. Nickel concentrations were above ANZECC Draft Guidelines (2000) in all samples over the three sampling intervals. Zinc concentrations were above the ANZECC Draft Guidelines (2000) in all samples, except one (K3-Nov 2003), over the three sampling intervals. Lead concentrations exceeded ANZECC Draft Guidelines (2000) at four of the six sample locations on more than one occasion.

Figure 3 – Grain size distribution for 200 g of road sediments collected from the Kuranda Range Road, sieved for 30 minutes on mechanical shaker



Most (90%) of the road material collected from the Kuranda Range Road in October 2002 was evenly distributed between the 0.75µm to 4mm sieve range. Grains and debris with diameter greater than 2mm constituted over 30% of the weight of the entire material collected from the gutters. Visual inspection of the material captured in the >2mm sieve interval indicates that this coarse material is not just composed of light organic debris and litter; a large proportion of it comprises weathered clasts of rock, presumably eroded from the outcropping Hodgkinson Formation alongside the road.

Figure 4 – Heavy metal concentrations (mg/kg) in various grain size fractions of a composite sample of sediments collected from the Kuranda Range Road in October 2002



Heavy metal concentrations in the various size fractions are shown in Figure 4. Here it can be seen that Ni concentrations are elevated in the fractions coarser than 1mm as well as those grain fractions less than 106µm, but are lower in the grain size fractions between 106µm and 1mm. Elevated Cd and Zn concentrations are confined to the medium size grain fractions (75µm – 2mm), whereas Cu and Pb are concentrated in the finer and medium size fractions.

Heavy metals in runoff water samples

Table 3 – Heavy metal concentrations (µg/L) in filtered runoff samples from sample locations shown on Figure 2

		K1	***K2***	**K2**	**K3**	**K4**	**K6**
	Date	*5-Feb-03*	*21-Jan-03*	*5-Feb-03*	*5-Feb-03*	*5-Feb-03*	*5-Feb-03*
	ANZECC Guidelines**						
Cd	0.06	≤0.05	*≤0.05*	≤0.05	≤0.05	≤0.05	≤0.05
Cu	1	***6.25***	***14.3***	***14.2***	***6.65***	***3.66***	***12.6***
Pb	1	0.146	*0.618*	0.405	≤0.05	≤0.05	0.592
Ni	8	2.24	*5.24*	1.49	1.61	0.22	1.53
Zn	2.4	***104***	***562***	***141***	***101***	***155***	***266***

Notes: *ANZECC Guidelines*** = ANZECC Draft Guidelines for the Protection of Freshwater Ecosystems – 99% Protection Level (2000)
Shaded values exceed ANZECC Guidelines

Copper and Zn concentrations were above the ANZECC Draft Guidelines (2000) in all filtered runoff samples collected in January 2003 and February 2003. Cadmium was below the detection limit in all runoff samples. Nickel was detected but in concentrations below the ANZECC Draft Guidelines (2000) in all field filtered runoff samples, while Pb was detected in five of the seven filtered rainwater runoff samples in concentrations below the ANZECC Draft Guidelines (2000).

Background heavy metals in stream sediments and stream waters

Table 4 – Mean background heavy metal concentrations (mg/kg) in stream sediments from Streets Creek, Kuranda Range

Mean background heavy metal concentrations in stream sediments from Streets Creek, Kuranda Range: N = 8		
	Date	*Feb-04*
	*ANZECC Guidelines**	
Cd	1.5	0.028
Cu	65	22.96
Pb	50	21.24
Ni	21	13.73
Zn	200	27.38

Notes: *ANZECC Guidelines** = (*ANZECC Draft Guidelines for Sediments* 2000)

Table 5 – Background heavy metal concentrations (µg/L) in unfiltered stream waters from Streets Creek, Kuranda Range

Background heavy metal concentrations in stream waters from Streets Creek, Kuranda Range		
	Date	*Feb-04*
	*ANZECC Guidelines ***	
Cd	0.06	≤0.05
Cu	1	0.891
Pb	1	0.646
Ni	8	0.161
Zn	2.4	≤5

Notes: *ANZECC Guidelines*** = (*ANZECC Draft Guidelines for Freshwater* 2000)

Remediation experiments – results

Table 6 – pH values of various water samples

	Water sample	**pH**
Field sample	*Rainwater runoff from gutters on Kuranda Range Road (mean)*	*6.40*
Laboratory experiment samples	*Distilled water*	*5.64*
	Leachate from road sediment	*6.67*
	Leachate from road sediment mixed with zeolites	*6.59*

Results in Table 6 suggest that road sediments increase the pH of distilled water by approximately 1 unit. Similar pH values were recorded for the leachate from the road sediment and the rainwater runoff samples collected from the Kuranda Range Road.

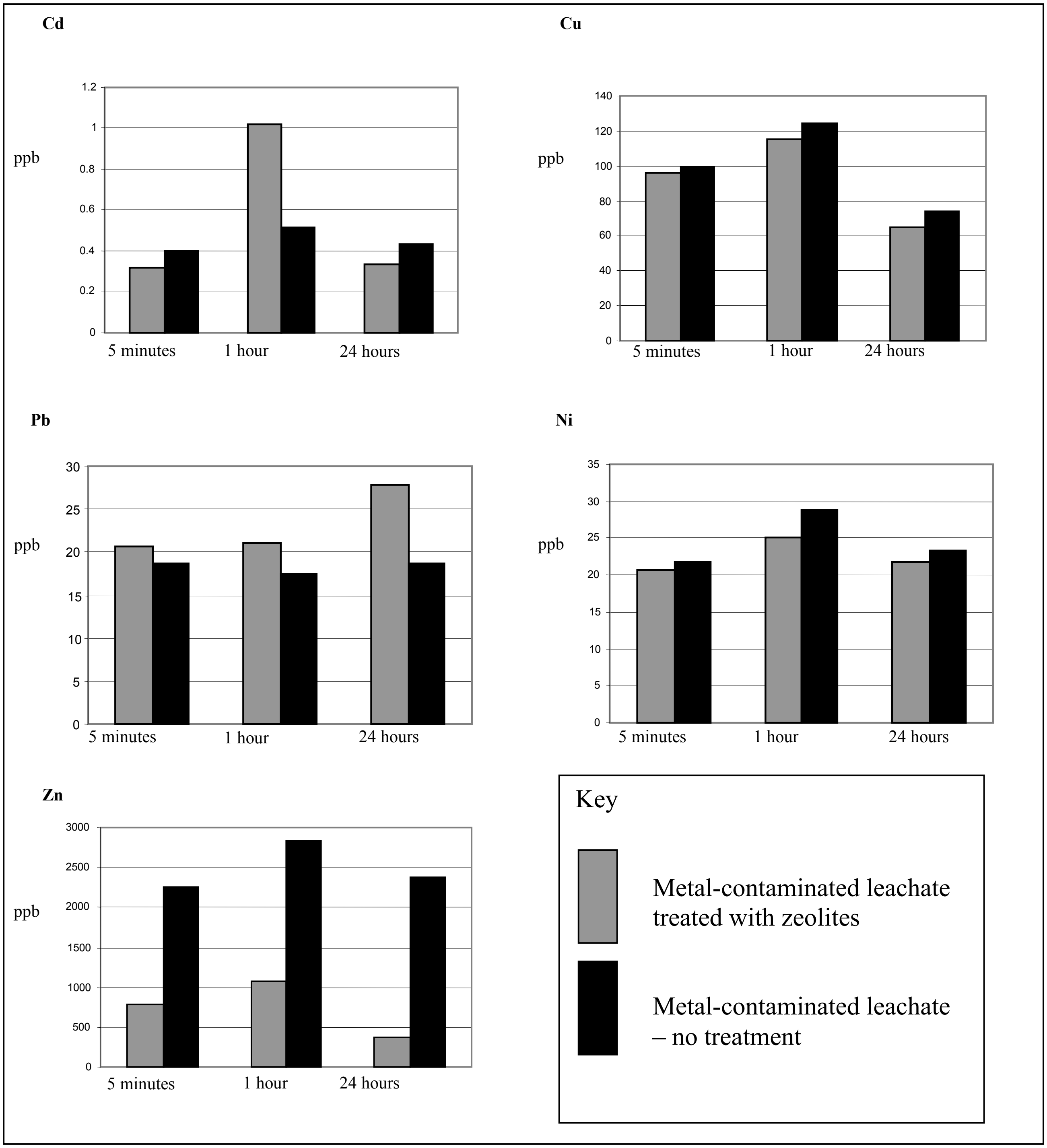
The pH of sediment-contaminated leachate is not altered by mixing with zeolites.

Results from Figure 5 show that Cd, Cu, Ni and Zn dissolved concentrations were lower in the zeolite-treated leachate than the untreated leachate (contaminated with road sediments) after 24 hours.

Lead concentrations were consistently greater in the zeolite-treated leachate than the untreated leachate over the three sampling intervals.

Dissolved Zn showed the greatest reduction between untreated and zeolite-treated leachate over all three sampling periods.

Figure 5 – Dissolved heavy metal concentrations (µg/L) in untreated leachate and leachate treated with zeolites after a) 5 minutes, b) 1 hour and c) 24 hours respectively



Discussion

Heavy metal levels in road sediments

Copper, Pb, Ni and Zn were detected in elevated concentrations in road sediments collected from a number of locations in concreted gutters along the Kuranda Range Road in October 2002, August 2003 and November 2003. Most samples collected over the three sampling intervals recorded Ni and Zn values above ANZECC Draft Guidelines (2000).

The concentrations of these metals, particularly Zn, in sediments from the Kuranda Range Road were above their concentrations recorded in the background stream sediment samples collected from Streets Creek (Table 4). Therefore, a source other than native rock material is likely account for these elevated metal concentrations in the road sediments.

Typically, abrasion of motor vehicle parts and vehicle emissions have been demonstrated to be the principal source of elevated levels of these types of metals in road sediments throughout the world (Harrison et al. 1981). Based on the lack of any metals-processing or mining land use within an approximate 100 km radius of the site it is reasonable to suggest that vehicles are the principal source of heavy metal contamination along the Kuranda Range Road. A potential additional source of metal contamination along this road includes galvanized material such as road signs and crash barriers. Galvanised material has been demonstrated to be a source of Zn to road corridors (Smolders and Degryse 2002).

Heavy metal concentrations detected in road sediments in this study are compared with values documented in other investigations in Table 7. It can be seen that Ni and Zn concentrations from the Kuranda Range Road typically exceeded values reported in sediments from more heavily trafficked roads. The high Ni concentrations detected in the road sediments in this investigation may be attributable to the considerable use of brakes along the steep and winding Kuranda Range Road. Zinc contamination in road sediments is more likely related to road design than traffic volume. Zinc is largely discharged from vehicles as Zn-oxides from abrasion of rubber tyres (Smolders and Degryse 2002) so it is likely to be present in elevated concentrations where corners on roads are sharp and abrasion rates are high. This is demonstrated by the consistently high Zn concentrations recorded at locations K1 and K2 on the Kuranda Range Road, which incorporate tighter bends than the other sampling locations (K3-K6).

Copper concentrations recorded in road sediments along the Kuranda Range Road appear to be high compared with Cu values reported in the literature, considering that this metal does tend to show an increase in concentration in road sediments with increasing traffic volume (Table 7). This trend is explained by the source of Cu in motor vehicles, as it is mainly derived from engine and brake pad wear (Viklander 1998).

Lead concentrations in road sediments along the Kuranda Range typically exceeded ANZECC Draft Guidelines (2000). Despite this, Pb levels recorded along the Kuranda Range Road are low compared with Pb levels reported in the overseas studies shown in Table 7.

This is perhaps a legacy of the phasing-out of leaded petrol in Australia in 2002, while countries abroad, such as Venezuela, Ecuador and even Great Britain, continue to use leaded gasoline (Hewitt and Candy 1990; Fernandez and Galarraga 2001; Pb-petrol UK, web 2004). Even so, Pb levels in sediments on the Kuranda Range Road are low compared to Pb values documented in road sediments in Sydney (Birch and Scollen 2003). This may reflect a footprint of longer Pb accumulation from leaded-petrol cars and industry in Sydney than in north Queensland. Gulson et al. (1981) reported Pb contamination extending to a depth of at least 10cm in Australian soils as a result of Pb fallout from vehicle emissions. Results from a study conducted by Chiaradia et al. (1997) indicated that despite the phasing out of leaded petrol in Sydney in the 1990s, petrol still accounted for more than 90% of the total Pb in the atmosphere of Sydney at that time. This highlights the lingering presence of Pb in the environment (Chiaradia et al. 1997).

Table 7 – Comparison of heavy metal concentrations in sediments collected from roads with varying traffic densities (all results in mg/kg)

	Mean concentration in road sediments	***Location***	***Author***
Cd	2.780	Rural major road, Lancaster University	(Harrison et al. 1981)
	2.630	Highway, Merter, Istanbul	(Sezgin et al. 2003)
	0.180	**Kuranda Range Road, 6126 vehicles/day**	**This study**
Cu	199.0	Rural major road, Lancaster University	(Harrison et al. 1981)
	73.0	Honolulu urban road, 45 200 vehicles per day	(Sutherland and Tolosa 2001)
	260.0	Parramatta Road, Sydney, 73 177 vehicles/day	(Birch and Scollen 2003)
	189.0	Marion Street, Sydney, 22 618 vehicles/day	(Birch and Scollen 2003)
	92.0	National Street, Sydney, <2000 vehicles/day	(Birch and Scollen 2003)
	40.9	**Kuranda Range Road, 6126 vehicles/day**	**This study**
Pb	5779.00	Tunnel sediments in Caracas	(Fernandez and Galarraga 2001)
	2540.00	Rural major road, Lancaster University	(Harrison et al. 1981)
	285.00	Honolulu urban road, 45 200 vehicles per day	(Sutherland and Tolosa 2001)
	1538.00	Parramatta Road, Sydney, 73 177 vehicles/day	(Birch and Scollen 2003)
	737.00	Marion Street, Sydney, 22 618 vehicles/day	(Birch and Scollen 2003)
	470.00	National Street, Sydney, <2000 vehicles/day	(Birch and Scollen 2003)
	50.13	**Kuranda Range Road, 6126 vehicles/day**	**This study**
Ni	24.00	Honolulu urban road, 45 200 vehicles per day	(Sutherland and Tolosa 2001)
	34.00	Parramatta Road, Sydney, 73 177 vehicles/day	(Birch and Scollen 2003)
	31.00	Marion Street, Sydney, 22 618 vehicles/day	(Birch and Scollen 2003)
	22.00	National Street, Sydney, <2000 vehicles/day	(Birch and Scollen 2003)
	52.20	**Kuranda Range Road, 6126 vehicles/day**	**This study**
Zn	458	Rural major road, Lancaster University	(Harrison et al. 1981)
	314	Honolulu urban road, 45 200 vehicles per day	(Sutherland and Tolosa 2001)
	706	Parramatta Road, Sydney, 73 177 vehicles/day	(Birch and Scollen 2003)
	578	Marion Street, Sydney, 22 618 vehicles/day	(Birch and Scollen 2003)
	1417	National Street, Sydney, <2000 vehicles/day	(Birch and Scollen 2003)
	1052	**Kuranda Range Road, 6126 vehicles/day**	**This study**

Heavy metal concentrations with respect to grain-size distribution

The grain size distribution of the heavy metal contaminants in the road sediments collected from the Kuranda Range Road suggests different settling patterns for the different metals. Cadmium and Zn appear to be concentrated most greatly in the medium size particles (500µm-70µm), Cu and Pb appear to be concentrated in the medium to fine material (<500µm) while Ni appears to be accumulated in two discrete grain size fractions - in the coarse (1mm-4mm) and finer-grained (<106µm) material.

In previous research into contamination of roadside sediments, heavy metals have been documented to be most concentrated in fine-grained particles in roadside sediments (<75µm), regardless of traffic volume (Viklander 1998; Birch and Scollen 2003; Varrica et al. 2003). Concentrations of anthropogenic heavy metals in road sediments tend to decrease with increasing particle size (Viklander 1998).

Varrica et al. (2003) conducted a detailed investigation into metal contamination in the fine-grained material (500µm to <10µm) of road sediments in Palermo, Sicily. In their study they found Cu and Zn were typically associated with other metals, including Ni, in clusters that were attracted to the surface of mineral phases in the 500µm grain size fraction. They also found Pb concentrations to be closely linked with Cl and Br levels (a common association of gasoline combustion) in the finer material (<40µm), while the presence of trace metals including Cu, Pb and Zn were detected in the finest particles (<10µm) (Varrica et al. 2003).

This brings into question the nature of the elevated Ni levels recorded in the coarse (>1mm) component of road sediments collected from the Kuranda Range Road. Are these elevated Ni concentrations suggestive of clusters of smaller Ni-rich particles attached to the surfaces of the coarser sediments? There is no clear answer to this question. It certainly seems that Ni contributions from the coarse and fine-grained background sediments do not account for most of the Ni detected in the road sediments collected from the Kuranda Range Road (see Table 4).

One line of reasoning suggesting that these elevated Ni concentrations in the coarse material are not produced by clusters of smaller particles is provided by the lower Ni concentrations in the medium-grained sediments from the Kuranda Range Road. It has been suggested that these medium sized particles (425µm to 850µm) contribute the greatest surface area in road sediments (Sansalone et al. 1998), so if Ni were present as smaller clusters it would be reasonable to suggest these clusters would be detected in the medium grain size fractions. However, in this investigation Ni levels were lowest in these medium size particles.

The elevated Ni concentrations in the coarse-grained sediments are therefore difficult to account for. Ni-rich basalt gravel used in asphalt is a possible explanation to account for these elevated concentrations.

Heavy metal mobility in road runoff waters

Knowledge of heavy metal distribution with respect to grain size fractions in road sediments provides some information regarding the potential mobility of these metals. The principal mobilising agent acting on sediments on the Kuranda Range Road is rainwater runoff, which is greatest in the Cairns region between the months of January and March (BOM Cairns climate, web page 2004).

The fine fraction of road sediments, which carries elevated loads of heavy metals, is readily transported by runoff waters (Birch and Scollen 2003). Furthermore, results from Table 3 indicate that some proportion of heavy metals in road sediments is soluble in runoff waters. These rainwater runoff samples were filtered through 0.45 µm pore-size paper, which produces filtrate thought to reasonably represent the 'dissolved' load of contaminants (Rose et al. 2001). However, there is some discrepancy in the literature regarding this assumption. Lee et al. (1997) claim that filtrate that has passed through 0.45 µm sized pore spaces in filter paper actually constitutes a mixture of dissolved solutes and colloids. Still, it is generally regarded that the contaminant loads present in this filtrate effectively represent the most labile portion of contaminants in runoff waters (Sansalone and Buchberger 1997; Dierkes and Geiger 1999; Rose et al. 2001). From here in, the term 'dissolved' will apply to concentrations detected in filtrate that has passed through 0.45 µm pore-size filter paper.

Table 3 shows elevated Cu and Zn concentrations in filtered rainwater runoff samples collected from the Kuranda Range Road in January and February 2003. Elevated dissolved Zn and Cu levels have been measured in runoff waters in roadside environments throughout the world (Sansalone and Buchberger 1997; Rose et al. 2001). Dissolved Pb and Ni were detected in runoff waters collected from the Kuranda Range Road in this study, although in concentrations well below the ANZECC Draft Guidelines (2000).

There is a dilution effect evident in the contaminant concentrations recorded in sample K2 in late January 2003 and the levels of those same contaminants detected in early February 2003. Copper, Pb, Ni and Zn concentrations collected at this location in late January 2003 (the onset of the first heavy rain for the season) were much greater than their respective concentrations in the sample collected at this location in early February (following the 'first flush'). However, the fact elevated metal concentrations persisted in runoff samples collected several weeks after the first significant rainfall event (see Table 3) indicates high loads of heavy metals are present in road sediments along the Kuranda Range Road.

Adsorption of aqueous heavy metals

The practical aspect of this research was to assess the potential of materials to adsorb metals from contaminated waters. In an environment such as the Kuranda Range, the installation of a large-scale remediation system such as a wetland or sediment retention pond is seen as unfeasible because of the sensitive nature of the location. Instead, small-scale filtration traps incorporating adsorptive substances are viewed to be the best-suited method to limit metal dispersal from road runoff into streams and over soils adjacent to the Kuranda Range Road.

The natural capacity of roads and gutters to immobilize metal contaminants is an important consideration in designing remediation strategies for these environments. Rubber particles in road sediments may also play a crucial role in keeping runoff waters at near-neutral levels. Smolders and Degryse (2002) state that tyre particles raise the pH of soil solution by approximately one unit. Rose et al. (2001) suggest that buffering runoff waters in this near-neutral region is critical to limit the solubility, and hence, mobility of trace metals.

Apart from the ability of tyre rubber to buffer pH levels of slightly acidic rainwater runoff, organic matter associated with rubber tyres shows a strong capacity to prevent metals entering solution. Organic matter has been demonstrated to remove metals from the aqueous phase in a number of investigations (Lee et al. 1997; Yin et al. 2002).

Despite the natural attenuation potential of roads to decrease metal solubility, a significant portion of these vehicle-derived metals have the potential to be mobilised by runoff waters, as evidenced by this study and a number of other investigations (Lee et al. 1997; Sansalone et al. 1998). Adsorption of soluble metals from runoff is one approach to limit the dispersal of these contaminants over roadside soils and into streams. It is critical to treat contaminated runoff water as close to its source (roads) as possible to avoid the build-up of metal-stores in sediments and soils, which contain compounds and chemical conditions that can actually remobilize the metals in more labile forms.

Results of the adsorption experiments involving zeolites indicated that zeolites are extremely effective in reducing dissolved Zn loads in contaminated waters even over a period of five minutes. However, the zeolites proved much less effective at adsorbing Cd, Cu and Ni. Furthermore, the zeolites appeared to be ineffective in adsorbing Pb.

The capacity of zeolites to adsorb Cd, Cu, Pb and Ni requires further research to validate their incorporation into filtration traps in gutters to reduce the levels of these metals in runoff waters. This research will most likely involve methods such as active stirring or some kind of agitation of solution treated with zeolites.

Conclusions

Elevated Cu, Pb, Ni and Zn concentrations were detected in road sediments from the Kuranda Range Road over a sampling period of one year between October 2002 and November 2003. These metals were detected in road sediments in concentrations well above background values recorded in the region and in many samples, Pb, Ni and Zn levels exceeded the ANZECC Draft Guidelines (2000). The grain size distribution analysis of these sediments indicated that a large portion of these metals may be readily mobilised by runoff waters into roadside streams and over soils. Elevated Cu and Zn concentrations were detected in rainwater runoff samples.

A series of adsorption experiments involving zeolites indicated that a large proportion of aqueous Zn was adsorbed by zeolites over a period between five minutes and one day.

The effectiveness of zeolites to adsorb Cd, Cu, Pb and Ni requires further research to validate the incorporation of zeolites into filtration traps in gutters to reduce the levels of these metals in runoff waters. This research will most likely involve methods such as active stirring or some kind of agitation of solution treated with zeolites.

Still, small-scale filtration traps that incorporate adsorptive substances are viewed to be the best-suited method to limit metal dispersal from road runoff waters into streams and over soils along the Kuranda Range Road.

References

Al-Chalabi, A. S. and Hawker, D. (2000). "Distribution of Vehicular Lead in Roadside Soils of Major Roads of Brisbane, Australia." *Water, Air and Soil Pollution* **118**: 299-310.

Alvarez-Ayuso, E. and Garcia-Sanchez, A. (2003). "Sepiolite as a Feasible Soil Additive for the Immobilisation of Cadmium and Zinc." *The Science of the Total Environment* **305**(1-3): 1-12.

Australian and New Zealand Environment and Conservation Council Draft Guidelines - Low Interim Sediment Quality Guidelines (2000).ANZECC Draft Guidelines for Sediments. Australian Department of Environment and Heritage.

Australian and New Zealand Environment and Conservation Council Draft Guidelines for the Protection of Freshwater and Marine Aquatic Ecosystems (2000).ANZECC Draft Guidelines for Freshwater. Australian Department of Environment and Heritage.

Birch, G. E. and Scollen, A. (2003). "Heavy Metals in Road Dust, Gully Pots an Parkland Soils in a Highly Urbanised Sub-catchment of Port Jackson, Australia." *Australian Journal of Soil Research* **41**: 1329-1342.

Cairns, Kuranda to Double Island Map, http://www.queensland-australia.com/100136.php (2003). Queensland Maps.

Chiaradia, M., Gulson, B. L., James, M., Jameson, C. W. and Johnson, D. (1997). "Identification of Secondary Lead Sources in the Air of an Urban Environment." *Atmospheric Environment* **31**(21): 3511-3521.

Climate of Cairns http://www.bom.gov.au/weather/qld/cairns/climate.shtml (2004). Commonwealth Bureau of Meterology.

Dierkes, C. and Geiger, W. F. (1999). "Pollution Retention Capabilities of Roadside Soils." *Water Science Technology* **39**(2): 201-208.

Fernandez, R. and Galarraga, F. (2001). "Lead Concentration and Composition of Organic Compounds in Settled Particles in Road Tunnels from the Caracas Valley - Venezuela." *Environmental Geochemistry and Health* **23**: 17-25.

Forbes, S., Grose-Hodge, S., Hewitt, G., O'Connor, S., Shrubb, S., Beaumont, C., Chiefetz, A., Double, C., Etherington, K., Imwold, D. and Jackson, H., Eds. (2002). *Geographica - the complete illustrated reference to Australia and the World*, James Mills-Hicks.

Gulson, B. L., Tiller, K. G., Mizon, K. J. and Merry, R. H. (1981). "Use of Lead Isotopes in Soils to Identify the Source of Lead Contamination Near Adelaide, South Australia." *American Chemical Society* **15**(6): 691-696.

Harrison, R. M., Laxen, D. P. H. and Wilson, S. J. (1981). "Chemical Associations of Lead, Cadmium, Copper and Zinc in Street Dusts and Roadside Soils." *Environmental Science and Technology* **15**(11): 1378-1383.

Hewitt, C. N. and Candy, G. B. B. (1990). "Soil and Street Dust Heavy Metal Concentrations in and around Cuenca, Ecuador." *Environmental Pollution* **63**: 129-136.

Leaded.petrol.co.uk http://www.leadedpetrol.co.uk/ (2004). Bayford and Co Ltd.

Lee, P. K., Baillif, P. B. and Touray, J.-C. (1997). "Geochemical Behaviour and Relative Mobility of Metals (Mn, Cd, Zn and Pb) in Recent Sediments of a Retention Pond Along the A-71 Motorway in Sologne, France." *Environmental Geology* **32**(2): 142-152.

Road Sections 32A and 20A - Kennedy Highway (49.2 km) from Peninsula District (MR) and Captain Cook Highway (75.1 km) from Peninsula District (MR), respectively: DMR Private

Database, ARMIS program - Daily traffic density data. (2002). Queensland Government - Department of Main Roads.

Rose, S., Crean, M. S., Sheheen, D. K. and Ghazi, A. M. (2001). "Comparative Zinc Dynamics in Atlanta Metropolitan Region Stream and Street Runoff." *Environmental Geology* **40**: 983-992.

Sansalone, J. J. and Buchberger, S. G. (1997). "Partitioning and First Flush of Metals in Urban Roadway Stormwater." *Journal of Environmental Engineering* **123**(2): 134-143.

Sansalone, J. J., Koran, J. M., Smithson, J. A. and Buchberger, S. G. (1998). "Physical Characteristics of Urban Roadway Solids Transported During Rain Events." *Journal of Environmental Engineering* **124**(5): 427-440.

Sezgin, N., Ozcan, H. K., Demir, G., Nemlioglu, S. and Bayat, C. (2003). "Determination of Heavy Metal Concentrations in Street Dusts in Istanbul E-5 Highway." *Environment International* **29**: 979-985.

Smolders, E. and Degryse, F. (2002). "Fate and Effect of Zinc from Tire Debris in Soil." *Environmental Science and Technology* **36**: 3706-3710.

Sutherland, R. A., Day, J. P. and Bussen, J. O. (2003). "Lead Concentrations, Isotope Ratios and Source Apportionment in Road Deposited Sediments, Honolulu, Oahu, Hawaii." *Water, Air and Soil Pollution* **142**: 165-186.

Sutherland, R. A. and Tolosa, C. A. (2001). "Variation in Total and Extractable Elements with Distance from Roads in an Urban Watershed, Honolulu, Hawaii." *Water, Air and Soil Pollution* **127**: 315-338.

Thomson, N. R., McBean, E. A., Snodgrass, W. and Mostrenko, I. (1997). "Sample Size Needs for Characterising Pollutant Concentrations in Highway Runoff." *Journal of Environmental Engineering* **123**(10): 1061-1065.

Turner, D., Maynard, J. B. and Sansalone, J. (2001). "Heavy Metal Contamination in Soils of Urban Highways: Comparison Between Runoff and Soil Concentrations at Cincinnati, Ohio." *Water, Air and Soil Pollution* **132**: 293-314.

Varrica, D., Dongarra, G., Sabatino, G. and Monna, F. (2003). "Inorganic Geochemistry of Roadway Dust from the Metropolitan Area of Palermo, Italy." *Environmental Geology* **44**: 222-230.

Viklander, M. (1998). "Particle Size Distribution and Metal Content in Street Sediments." *Journal of Environmental Engineering* **124**: 761-766.

Yin, Y., Impelliteri, C. A., You, S.-J. and Allen, H. E. (2002). "The Importance of Organic Matter Distribution and Extact Soil:Solution Ratio on the Desorption of Heavy Metals from Soils." *The Science of the Total Environment* **287**: 107-119.

Zhou, W., Beck, B. F. and Green, T. S. (2003). "Evaluation of a Peat Filtration System for Treating Highway Runoff in a Karst Setting." *Environmental Geology* **44**: 187-202.