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Accumulation of potentially toxic elements in road deposited sediments in residential and light industrial neighborhoods of Singapore

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ABSTRACT

Road deposited sediments (RDS) are a valuable environmental medium for characterizing contaminant levels in urban areas; and their associated potentially toxic elements (PTEs) can directly impact both human and aquatic health. In this study, RDS were collected from 15 co-located industrial and residential roads throughout Singapore to determine the effect of land use on contaminant levels. A second pilot study was designed to quantify the efficiency of road sweeping in removing different RDS grain size fractions from industrial and residential roads. The fine fraction (<63 μm) of all RDSs was analyzed for over 40 elements. Eleven elements that reflect geogenic and anthropogenic sources were examined in detail (Al, Co, Cr, Cu, Fe, Ni, Pb, Sb, Sc, Si, and Zn). Industrial RDS had statistically higher concentrations of Co, Cr, Fe, and Ni than residential RDS. Potentially toxic elements Cu, Pb, Sb, and Zn were enriched >10-fold at all locations compared to upper continental crust values. Concentrations of Cu, Pb and Zn exceeded aquatic sediment probable effect concentration levels, suggesting they could generate a toxic response in bottom-dwelling aquatic organisms. Traffic was equally heavy at both industrial and residential sites, but large trucks and machinery comprised a larger proportion of the traffic in the industrial areas. Traffic was not significantly correlated with the PTE (i.e., Cu, Pb, Sb and Zn) concentrations. Plausible anthropogenic contaminant sources include vehicles (e.g., brake and tire wear, vehicle emissions) and several industrial activities including metal works, oil processing, and waste incineration. Street sweeping was effective in removal of large organic debris and inorganic RDS, but it was ineffective in removing the geochemically important fraction, i.e., <125 μm.

1. Introduction

Roads are an essential component of the urban landscape that are often conspicuous point-sources of dissolved and sediment-associated contaminants (McKenzie et al., 2008; Tian et al., 2009; Zhao et al., 2009). Litter and oil spills are visible evidence of road contamination (Stenstrom et al., 1984; Allison et al., 1997; Walker and Wong, 1999; Kang et al., 2009). Less-visible contaminants include inorganic potentially toxic elements (PTEs), de-icing salts, and organic compounds such as polycyclic aromatic hydrocarbons (PAHs), perfluorinated surfactants (PFSs), polychlorinated biphenyls (PCBs), nutrients (e.g., phosphates and nitrates), and pesticides. Sources of inorganic PTEs on road surfaces include vehicular exhaust emissions, wear of *in situ* road surfaces and pavements, brake and tire wear, atmospheric deposition, and roadside soil erosion (Fergusson, 1984; Sutherland, 2003; Folkeson et al., 2008). Brake wear for example releases fine particles ranging from a few hundred nanometers to a few micrometers in diameter. The rate of wear and character of associated frictional debris is a function of vehicle speed, braking frequency, vehicle condition and the composition of brake lining materials (Thorpe and Harrison, 2008). Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb and Zn are some of the elements typically found in brake wear particles (Table 1; Davis et al., 2000; McKenzie et al., 2008; Thorpe and Harrison, 2008). Similarly, tire wear debris can be enriched with Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn (Table 1; Davis et al., 2000; McKenzie et al., 2008; Thorpe and Harrison, 2008; Duong and Lee, 2011). Rogge et al. (1993) postulated that friction between the road surface and tire treads results in an average tire wear rate of between 6 and 90 mg/km, depending on driving speed, tire age, and the road surface condition. Generally, higher speeds, older wheels and rougher surfaces result in more wear (Duong and Lee, 2011).

Elemental contaminants can also enter the environment by the combustion of fossil fuels including petrol and diesel fuels (Table 1).

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Table 1
Vehicular- and road-related sources of inorganic elements.

Metal/Source[a]	Bi	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Pd	Pt	Rh	Sb	V	Zn
Brakes		✓		✓	✓	✓	✓	✓	✓				✓		✓
Tires		✓	✓	✓	✓	✓	✓	✓	✓						✓
Fuels and Oils	✓	✓		✓	✓			✓						✓	✓
Vehicle Body									✓						
Catalytic Convertors										✓	✓	✓			
Engine Wear				✓	✓		✓	✓							
Road surface		✓			✓	✓		✓	✓					✓	✓
Road Markings									✓						
Road Equipment															✓

[a] Sources: Helmers, 1996; Lindgren, 1996; Davis et al., 2000; Deletic and Orr, 2005; Folkeson et al., 2008; McKenzie et al., 2008; Sutherland et al., 2008; Thorpe and Harrison, 2008; Duong and Lee, 2011.

Although leaded petrol is banned in many countries today, significant archives may still exist in the near-road environment as the low solubility of Pb allows it to have long residence times in the soil column (Turjoman and Fuller, 1987; Sutherland and Tolosa, 2001). Zinc is also present in petroleum as an additive to reduce engine wear (Table 1). Small amounts of Bi, Cd, Cr, Cu, Ni, and V are also used in petrol and diesel (Davis et al., 2000). Paint particles chipped from vehicle bodies and road lane markers are other sources of Pb (Deletic and Orr, 2005). Wear of 3-way catalytic converters can also release platinum group elements (Pd, Pt and Rh) to the road environment (Helmers, 1996; Sutherland et al., 2008). Engine wear releases Cr, Cu, Mn, and Ni (Table 1). Mineral and stone materials used in road building may contain Cd, Cu, Fe, Ni, Pb, V, and Zn, which are released to the environment as the road surface wears (Lindgren, 1996). Galvanized-steel road equipment such as crash barriers, road signs and lamp posts may release Zn as they corrode (Table 1) (Folkeson et al., 2008).

Soil, parent material and bedrock are natural sources of elements in the urban landscape, but concentrations are often elevated by anthropogenic activities (Carlosena et al., 1998). Elevated concentrations of metal(oid)s are commonly archived in roadside soils, and they potentially represent a long-term source of contaminants to the road surface via remobilization by wind, rain, vehicular turbulence and pedestrian movement (Sutherland and Tolosa, 2001; Jartun et al., 2008). Releases via industrial processes often provide an overriding anthropogenic signal on the spatial and temporal distribution of road contaminants (Lau and Stenstrom, 2005; Herngren et al., 2006). For example, Zhao et al. (2009) found that sediments collected from industry-influenced roads had higher concentrations of Cr, Cu, Ni, and Pb than commercial and residential roads in Beijing. The presence of many small factories producing wine, textiles and hardware were considered potential sources. Likewise, Zhang and Wang (2009) found that industrial sites in Hangzhou, China, home to food processing, engineering, electronics and textile industries, had the highest concentrations of nine PTEs (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Zn) tested. The presence of steel tool houses and machine shops in industrial districts of Seoul were potential contributors to the high levels of Cd, Cu, and Zn in RDS (Yun et al., 2000). Also, higher levels of contamination in industrial areas may be related to greater volumes of traffic, especially heavy vehicles (Wong et al., 2000; Charlesworth et al., 2003; Pandian et al., 2009).

Road deposited sediments (RDS, Sutherland and Tolosa, 2000) are a management concern because many contaminants are concentrated in the fine fractions that are readily re-suspended by traffic or strong winds, and they may also be conveyed by surface overland flow into nearby water bodies. For example, Zhao et al. (2010) found that particles <44 μm contributed more than 70% of total suspended solid (TSS) load in road runoff at a site in Beijing. Similarly, Nie et al. (2008) found the contribution of particles <45 μm to TSS was twice that of coarser particles (*cf.* Jartun et al., 2008). The threat of RDS contributing to the degradation of water bodies is substantial if they are not routinely removed by street sweeping (Walker and Wong, 1999; Schilling, 2005; Kang et al., 2009). This issue is of particular concern for industrial areas where metal contaminant concentrations may be greatly enriched over background levels.

The effectiveness of street sweeping as a contaminant control measure is to some extent uncertain, as fine sediments are not always removed efficiently (Walker and Wong, 1999). Sartor and Boyd (1972), for example, found that only 50% of all fractions of RDS could be removed by sweeping—and the removal rate of material <104 μm was only 15–20% (cf. USEPA, 1983; Sartor and Gaboury, 1984). Although technological advancements have improved sediment removal rates from roads, the value of street sweeping as a storm water best management practice is still debatable. For example, sweeping failed to reduce TSS loads, and hence sediment-associated contaminant levels, in a recent study in the USA (Yee, 2005). In contrast, German and Svensson (2001) showed that street sweeping removed up to 80% of RDS; and Sutherland and Jelen (1997) using a vacuum-assisted sweeper were able to remove 70% of the particles <63 μm. Variations between studies may be related to differences in sweeper type, for example mechanical broom versus regenerative air and vacuum-assisted sweepers. Other variables affecting sweeping efficiency include sweeping time, frequency of sweeping, sweeping speed, operator skill, and road surface type (Sartor and Gaboury, 1984; Allison et al., 1998; Curtis, 2002; Schilling, 2005).

Research on road contaminants in Southeast Asia is limited to a few locations: e.g., Kuala Lumpur, Malaysia (Ramlan and Badri, 1989), Bangkok, Thailand (Boonyatumanond et al., 2007), and Singapore (Joshi et al., 2009). In the Singapore study, the authors found that Cr, Cu, Fe, Ni, Pb and Zn levels were 3- to 42-fold higher in an industrial area than in a residential or a commercial area. Emissions from nearby industries and heavy vehicles were likely sources that resulted in these elements exceeding the maximum permissible limits in soils. Road contaminants in Singapore are potentially a significant source of water pollution because of the high density of roads, many of which drain into sewers, culverts and channels which lead directly to the reservoirs and sea. Metal contaminants, which are ubiquitous, non-biodegradable, and potentially toxic (Li et al., 2004; Herngren et al., 2005; Lee et al., 2006; Nie et al., 2008) are a concern in Singapore because of the high density of light industrial areas on the island. Herein we expand upon the study of Joshi et al. (2009) in Singapore to quantify the concentrations and enrichment ratios of 11 elements, some potentially toxic, in industrial and residential areas distributed across the entire island. Additionally, for the first time in Singapore, the effectiveness of street sweeping in removing different grain size fractions of RDS (and hence contaminants) is explored.

2. Materials and methods

2.1. Field sites

Singapore is located at the southern tip of the Malaysian Peninsula, between latitudes 1°09′ N and 1°29′ N and longitudes 103°36′ E and 104°25′ E (Hu and Balasubramaniam, 2003). The island is 42 km from east to west and 23 km from north to south. The local climate is characterized by a narrow range of temperatures (25–34 °C), high humidity and abundant rainfall throughout the year (annual mean = 2550 mm, mean rainfall for all months >150 mm (Ng et al., 2005)). Singapore is a fully industrialized nation with manufacturing and service sectors being key economic drivers. Specifically, the electronic and petrochemical industries dominate the manufacturing sector; tourism, the service sector (Atlas Singapore and the World, 2008). Singapore has a vehicle density of 276 vehicles per km^2 of road and a total vehicle count of 925,518. The road density is about 5 km km^{-2} (Singapore Land Transport Statistics in Brief, 2010).

Road deposited sediments were collected from 15 residential and 15 industrial sites. To ensure representative spatial coverage, the 30 sites were stratified across the island (Fig. 1). Additionally, all sampled roads were flat and made from asphalt (Fig. 2). Work was conducted between October 17 to November 6, 2010 (sediment collection), then again in January (sweeping experiments) and April (vehicle survey) of 2011.

2.2. Sampling

Non-metallic fiber bristle brooms and plastic dustpans were used to collect RDS in a manner similar to other studies (Charlesworth et al., 2003; Joshi et al., 2009; Zhang and Wang, 2009). This method is as efficient as vacuum cleaning (Tanner et al., 2008) and does not alter the chemical composition of sediments. Multiple sweeping passes were made along sampled road sections to remove all sediments as singular sweeps are generally inefficient in removing them. A new broom and dustpan were used at each field site to avoid cross contamination. Sediment samples were collected within 0.3 m of the curb for safety considerations and because most road sediments are generally stored in this area (Sartor and Boyd, 1972). Additional work involved sweeping a 50-m (length) × 0.30 m (wide) section at each of the 30 sites to quantify the amount and range of particles present on a given road. A fixed distance of 50 m was chosen because it was operationally manageable.

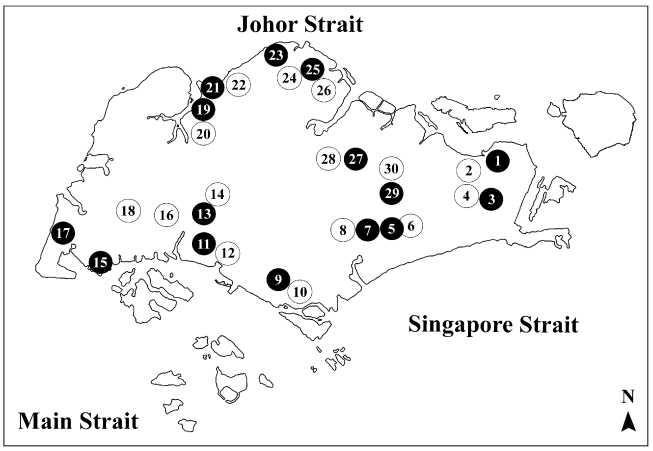

Fig. 1. Road deposited sediment sampling locations in Singapore. Industrial roads (black). Residential Roads (white).

The second phase of this study involved examining RDS removal effectiveness of mechanical broom street sweepers, which are deployed throughout Singapore. Two sites were selected, one in an industrial area (site #11), and one in a residential area (site #12). Three sets of two adjacent 50-m road sections were demarcated on 19 and 20 January 2011, respectively. For each set, material was collected on one section before the street was swept, then on the other within an hour after the street sweeper had passed.

2.3. Sample preparation

All RDS samples were transferred to Ziploc bags and transported to the laboratory where they were oven-dried at 105 °C for 24 h. Samples were passed through a suite of stainless steel sieves (16 mm, 8 mm, 4 mm, 2 mm, 1 mm, 500 μm, 250 μm, 125 μm and 63 μm) to determine the mass of each fraction. To prevent any cross contamination each sieve was thoroughly cleaned and oven-dried prior to reuse. For grain size partitioning, only the sediments less than 4 mm were considered because coarser fractions primarily consisted of organic debris and large stones.

2.4. Elemental analyses

The <63-μm fraction from phase one of this study was ground to homogenize samples prior to elemental analysis. The fusion-dissolution method used to totally digest samples involved combining 300 mg of RDS with 600 mg of lithium tetraborate ($Li_2B_4O_7$) flux. The powder mixture was homogenized and transferred to a graphite crucible that was heated in a muffle furnace (900 °C) for 20–30 min. After cooling, the fusion bead was transferred to a tared 500-mL HDPE bottle, and the following reagents were added: 2% HNO_3, 20 drops of H_2O_2, 10 drops of HF, bringing the final mass to 500 g with 2% HNO_3. The solution was shaken vigorously for 4–6 h until clear. The final solution was further diluted 2× online during analysis. Element concentrations were determined with a Thermo Scientific X Series2 Quadrupole ICP-MS with collision cell technology (CCT). Data was reduced with PlasmaLab software.

2.5. Quality control

Several approaches were used to assess the quality of analyses. Element concentrations from five independent digestions of US Geological Survey (USGS) reference material SDC-1 (mica schist) were compared to recommended values for 11 elements to determine precision and accuracy. Additionally, two Geosciences Laboratory (Boise State University) in-house standards (GMP and MON, both granites) were analysed five separate times and the precision for 11 elements was determined. Finally, precision for 11 elements was determined for three replicates of RDS from industrial site #1 and residential site #14. It is clear from the summary of quality control data that the 11 elements were measured precisely and accurately (Table 2).

2.6. Traffic survey

A one-time traffic survey was conducted simultaneously by 30 assistants at all field sites on April 11, 2011, between 8:30–10:30 AM (a Monday morning workday). Our intention was to quantify the average number of motorcycles and scooters, cars, buses, vans, lorries, and heavy vehicles (e.g., dump trucks, container trucks, cement mixers and oil tankers) using each road. This information was useful for examining relationships between traffic use and contaminant concentrations.

Fig. 2. Examples of two industrial and residential road sample locations in Singapore.

2.7. Level of contamination

Three approaches were used to characterize the degree of contamination of RDS. The first involved comparing concentrations of Cr, Cu, Ni, Pb and Zn in RDS to consensus-based sediment quality guidelines (SQGs) developed by MacDonald et al. (2000), who used matching sediment chemistry and toxicity data from USA field studies. For each contaminant, we compared concentrations with the threshold effect concentrations (TEC) and probable effect concentrations (PEC) as shown in Table 3. Concentrations below a TEC value for a given element rarely generate a toxicity response in bottom-dwelling aquatic organisms, whereas concentrations above a PEC value do.

Secondly, as not all elements have TEC and PEC values, an enrichment ratio (ER) approach was also used to characterize degree of contamination. An ER is the ratio between the concentration of a given element (n) in a RDS sample and its background concentration in the upper continental crust (UCC, Table 3). In the calculation, we normalized by grain size (i.e., <63-μm fraction) and used Al as the conservative element (Andrews and Sutherland, 2004):

$$\mathrm{ER_n} = \frac{C_n\mathrm{RDS}/C_{Al}\mathrm{RDS}}{C_n\mathrm{UCC}/C_{Al}\mathrm{UCC}} \quad (1)$$

where C_n is the concentration of element n in RDS or the UCC (values from Rudnick and Gao, 2003); and C_{Al} is the concentration of aluminum in RDS or the UCC. Although ER values greater than 1 can be considered anthropologically impacted, given the variability of element concentrations in the UCC and related weathering processes, we consider elements with ER values >10 as anthropogenically enriched.

Thirdly, as SQGs and ER-values are element-specific, the Tomlinson et al. (1980) multi-element pollution load index (PLI) was computed for each RDS site:

$$\mathrm{PLI_{Site}} = \sqrt[5]{\mathrm{CF_{Element\ 1}} \times \mathrm{CF_{Element\ 2}} \times \ldots \mathrm{CF_{Element\ 5}}} \quad (2)$$

where the concentration factors ($\mathrm{CF_{Element}}$) for the five elements with the highest concentrations are computed as follows:

Table 2
Quality control data for US Geological Survey (USGS) reference material SDC-1 (mica schist), two Boise State University in-house standards MON and GMP (granites), and replicate analyses of industrial (IND) and residential (RES) road-deposited samples.

Element	SDC-1[a] (mg/kg)	SDC-1[b] Precision (%)	SDC-1[c] Accuracy (%)	MON[d] Precision (%)	GMP[d] Precision (%)	IND[e] Precision (%)	RES[f] Precision (%)
Al	83,622	0.25	2.42	0.81	0.65	1.97	1.25
Co	18	0.77	−3.06	1.63	1.01	5.56	8.77
Cr	64	0.17	−6.49	2.12	0.80	3.49	9.04
Cu	30	0.50	−0.72	1.12	3.16	1.51	2.81
Fe	44,202	0.18	12.91	1.35	0.97	1.04	3.15
Ni	38	0.43	−8.67	1.80	1.97	2.64	7.64
Pb	25	0.29	4.13	0.60	0.34	1.05	3.96
Sb	0.54	2.24	1.67	6.40	9.56	2.20	2.78
Sc	17	0.64	2.13	1.97	4.64	2.79	8.63
Si	307,572	0.63	5.06	0.49	1.09	4.19	8.29
Zn	103	1.06	−4.39	1.59	0.92	3.35	1.18

[a] Recommended values from USGS Certificate of Analysis, March 1998 (Denver, Colorado).
[b] Calculated by the coefficient of variation (relative standard deviation); $n = 5$.
[c] Calculated as [(Measured value − USGS Recommended value)/(USGS Recommended value)] * 100.
[d] Calculated same as SDC-1 ($n = 5$).
[e] Calculated same as SDC-1 ($n = 3$ for industrial site #1).
[f] Calculated same as SDC-1 ($n = 5$ for residential site #14).

Table 3
Statistical summary of selected element concentrations (mg/kg) in road deposited sediment from 15 industrial and 15 residential sites in Singapore.

Element	TEC[a]	PEC[a]	#PEC[b]	UCC[c]	Industrial[d]	Residential[d]	*p*-value[e]
Al				81,505	51,110 ± 5090 (36,260; 59,730)	51,240 ± 2450 (44,920; 76,070)	0.724
Co				17.3	18.7 ± 7.1 (8.3; 144)	12.9 ± 3.4 (7.2; 28.2)	0.027
Cr	43.4	111	15, 15	92	402 ± 115 (232; 4647)	312 ± 103 (130; 1325)	0.038
Cu	31.6	149	15, 15	28	729 ± 265 (251; 4271)	498 ± 129 (203; 1094)	0.101
Fe				39,176	73,020 ± 20,890 (34,270; 213,700)	51,700 ± 15,300 (31,560; 94,470)	0.017
Ni	22.7	48.6	14, 14	47	204 ± 116 (40.7; 6660)	75.5 ± 16.8 (44.0; 446)	0.0023
Pb	35.8	128	15, 15	17	280 ± 91.3 (144; 744)	201 ± 28.7 (135; 395)	0.165
Sb				0.4	13.8 ± 3.2 (7.7; 45.4)	17.5 ± 3.8 (9.5; 36.2)	0.071
Sc				14	8.5 ± 0.50 (6.1; 11.6)	9.7 ± 0.61 (8.2; 17.2)	0.014
Si				311,405	230,500 ± 17,400 (113,900; 288,900)	243,500 ± 14,000 (208,100; 266,900)	0.468
Zn	121	459	15, 15	67	1649 ± 807 (826; 11,960)	1543 ± 425 (704; 2105)	0.141

[a] Consensus-based sediment quality guidelines: TEC = threshold effect concentration; and PEC = probable effect concentration (MacDonald et al., 2000); no guideline values are available for Al, Co, Fe, Sb, Sc, and Si.
[b] #PEC is the number of industrial and residential sites exceeding PEC (maximum number = 15).
[c] Values for the upper continental crust (Rudnick and Gao, 2003).
[d] Median ± median absolute deviation from the median; values in parentheses represent minimum and maximum concentrations.
[e] Tied *p*-values computed using the nonparametric (unpaired) Mann–Whitney *U*-test for industrial and residential site comparisons.

$$CF_{Element} = \frac{C_n RDS}{C_n UCC} \quad (3)$$

where, the concentration factor for a given element 'n' in RDS is normalized by the background concentration of the same element in the UCC (Table 3). Road deposited sediment with PLI values greater than one reflect increasing anthropogenic multi-element signals. A PLI of 1 or less indicates a site exhibits no significant anthropogenic signal. Finally, individual PLI_{Site} values were used to compute $PLI_{Landuse}$ values for industrial and residential land uses as follows:

$$PLI_{Landuse} = \sqrt[15]{PLI_{Site\ 1} \times PLI_{Site\ 2} \times \ldots PLI_{Site\ 15}} \quad (4)$$

Consideration of the results from all three approaches provides an investigator with suitable environmental screening tools to determine whether further toxicity testing is warranted, or whether additional geochemical investigations are necessary using operationally-defined sequential extraction analysis, or physiologically-based extraction tests.

2.8. Geometric mean particle diameter

The geometric mean particle diameter (GMPD) was computed for the sediments collected in the second round of fieldwork to determine the particle size distribution of RDS per 50 m stretch of road. Only the sediments less than 4 mm were considered because the coarser fraction consisted mainly of organic debris and large stones. The GMPD (μm) is calculated using the following equation (modified from Sutherland et al., 1996):

$$GMPD = \exp\left(\frac{\sum_{i=1}^{n} w_i \log \overline{D}_i}{\sum_{i=1}^{n} w_i}\right) \quad (5)$$

where, values of w_i are masses of sediments in the following size ranges: <63 μm; 63–125 μm; 125–250 μm; 250–500 μm; 500–1000 μm; 1000–2000 μm; and 2000–4000 μm. D_i refers to the mean diameter of seven size fraction ranges.

3. Results and discussion

3.1. Sediment on industrial and residential roads

The average RDS sample mass for 50 m × 0.3 m (15 m^2) swaths of industrial roads was 1895 ± 1492 g (± one standard deviation; range from 409 to 4410 g) versus 566 ± 403 g (81–1503 g) for residential sites (Table 4). The three-fold greater loading on industrial roads reflects improper waste disposal, as well as the presence of a greater number of parked vehicles and trailers along the roads that prevented street sweeper access. Additionally, sweeping frequency may be less in some industrial estates. Finally, some of the sites were enriched with sediment eroded from roadside soils where excavation had occurred or protective grass cover was not maintained.

The proportion of each of the grain size fractions in the RDS for both land uses was very similar. On average, the fines (<125 μm) accounted for 10.6 ± 5.9% of the industrial road sediment mass; 8.8 ± 3.0%, for residential roads. The dominant fraction for both land uses was the fraction between 250 and 500 μm, amounting to about 24% in the industrial sites and 25% in the residential sites. The three fractions between 125 and 1000 μm accounted for about 70% of the mass of RDS for the two land uses. The GMPD values for industrial RDSs ranged from 230 to 960 μm; and 300–760 μm for residential roads (Table 4). The nonparametric Mann–Whitney *U*-test indicated that there was no statistically significant difference in GMPD between land uses (i.e., tied *p*-value = 0.55).

3.2. RDS element concentrations and enrichment ratios

The median element abundance for RDSs at industrial locations followed the order: Si > Fe > Al > Zn > Cu > Cr > Pb > Ni > Co > Sb > Sc (Table 3). The sequence is the same for residential sites, except that Co and Sb are transposed. The Mann–Whitney *U*-test

Table 4
Bulk road sediment properties and median elemental concentrations for 15 industrial and 15 residential sites across Singapore.

Land use	Number	Land use activity	Mass g	GMPD[b] μm	<63 μm %	<63 μm Organic[a] %	Traffic density mean ± std dev	Al mg/kg	Co mg/kg	Cr mg/kg	Cu mg/kg	Fe mg/kg	Ni mg/kg	Pb mg/kg	Sb mg/kg	Sc mg/kg	Si mg/kg	Zn mg/kg
Industrial																		
Industrial	1	Aerospace industries	3266	380	1.9	21.8	457 ± 127	51 110	95	448	929	58 330	592	280	14	8	229 000	1224
Industrial	3	Aerospace industries	1917	550	1.6	5.6	267 ± 47	50 820	19	421	900	42 300	245	189	13	11	275 600	1540
Industrial	5	Auto repair, air con parts repair	672	480	1.6	9.8	1452 ± 168	51 410	9	263	464	41 280	89	338	19	9	230 400	1293
Industrial	7	Tire processing & warehouse	4410	960	0.6	15.3	69 ± 14	57 730	12	287	251	72 930	114	317	8	9	238 700	1814
Industrial	9	Metal works, woodwork & food production	442	570	1.0	17.2	314 ± 47	46 020	30	1997	1484	93 910	954	368	14	9	213 100	4141
Industrial	11	Food processing	409	490	0.9	10.8	219 ± 17	55 150	15	402	502	59 940	188	153	12	9	264 800	1493
Industrial	13	metal works, auto parts and printing	467	300	4.3	8.2	408 ± 8	40 100	144	4647	4271	213 700	6660	744	16	6	113 900	5451
Industrial	15	Shipyard	3907	380	2.2	10.3	300 ± 91	36 260	44	490	3585	204 600	313	387	45	8	182 100	11 960
Industrial	17	Incineration plant, petrochemical, transport	1006	320	3.3	8.5	473 ± 15	49 390	25	347	1253	87 590	204	221	27	11	237 200	2456
Industrial	19	Timber, chemicals and asphalt production	1233	230	5.9	3.8	415 ± 64	59 730	8	362	497	57 550	41	157	9	8	284 800	842
Industrial	21	Scrap metal processing, timber and furniture	3558	320	1.8	9.4	353 ± 4	44 340	17	567	697	104 200	69	170	18	8	239 700	1391
Industrial	23	Power station, oil depot, construction materials	4046	600	0.6	6.5	95 ± 16	53 300	16	269	374	90 100	163	283	10	9	223 400	2537
Industrial	25	Electronics, semiconductors	673	570	3.6	11.8	66 ± 23	49 200	19	369	729	73 020	425	532	15	8	187 000	1649
Industrial	27	Electronics, microelectronics, semiconductors	1234	520	1.7	13.5	279 ± 105	56 600	13	232	276	34 270	88	144	11	8	288 900	826
Industrial	29	Recycling, furniture manufacturing, metal works	1179	340	2.0	13.5	192 ± 30	57 360	30	1397	915	99 510	976	258	11	12	230 500	2883
Residential																		
Residential	2	Housing	380	550	1.4	14.5	443 ± 170	55 510	14	346	383	38 740	77	395	20	9	266 900	1608
Residential	4	Housing	350	560	1.7	16.9	136 ± 38	52 360	15	167	344	31 560	65	157	20	9	238 900	1118
Residential	6	Housing	1503	340	3.1	9.2	757 ± 120	76 070	27	868	498	56 290	446	173	13	17	258 700	1113
Residential	8	Housing	174	460	1.4	17.5	342 ± 18	46 170	7	162	263	36 400	44	182	14	8	215 300	1110
Residential	10	Housing	258	760	0.7	11.6	520 ± 2	48 790	8	130	203	33 180	51	173	10	10	243 500	704
Residential	12	Housing	435	380	1.4	9.8	267 ± 40	51 450	9	230	392	46 950	82	135	14	9	234 600	890
Residential	14	Housing	1198	570	1.4	10.2	208 ± 22	51 240	16	308	627	52 940	86	219	16	12	233 900	1200
Residential	16	Housing	81	470	2.1	11.8	176 ± 40	49 700	28	460	1094	80 970	141	201	21	10	250 100	2105
Residential	18	Housing	701	300	2.1	12.1	355 ± 66	50 510	15	315	868	68 150	101	163	36	9	262 700	1578
Residential	20	Housing	257	490	0.6	13.3	218 ± 70	49 380	18	382	968	71 670	112	230	22	11	249 500	2101
Residential	22	Housing	737	560	1.0	11.0	227 ± 20	58 910	9	362	623	51 700	75	232	18	10	257 500	1389
Residential	24	Housing	295	370	3.9	10.0	167 ± 42	52 470	9	209	585	94 470	59	224	17	9	222 200	2009
Residential	26	Housing	873	600	1.0	12.8	212 ± 10	58 960	10	199	268	37 740	65	260	14	10	217 200	1659
Residential	28	Housing	843	440	1.2	7.9	470 ± 27	47 720	13	1325	572	84 350	57	190	26	9	208 100	1543
Residential	30	Housing	398	490	1.3	32.0	726 ± 15	44 920	10	312	465	39 540	59	297	24	10	246 700	1585

[a] The organic fraction was established using loss-on-ignition, whereby >5-g, oven-dried subsamples were ignited in a muffle furnace for 4 h at 550 °C.
[b] GMPD represents the geometric mean particle diameter (Equation 5).

indicated that concentrations of Co, Cr, Fe, and Ni were significantly ($p \leq 0.05$) higher in industrial RDS. Scandium was significantly higher in residential RDS. Conversely, Al, Cu, Pb, Sb, Si, and Zn were not significantly different between land uses. Four of these elements, Cu, Pb, Sb, and Zn, had median enrichment ratios >10 (Fig. 3). Additionally, for the three elements with established sediment quality guidelines (i.e., Cu, Pb and Zn) all RDSs exceeded the PEC threshold (Table 3). These results require further exploration as they indicate substantial anthropogenic inputs to the road environment in Singapore; their off-site transport to receiving water bodies will likely negatively impact sediment-dwelling aquatic organisms. Elemental concentrations and ERs of the 11 elements we studied are discussed in detail below.

3.2.1. Cobalt

The median Co concentration of 19 ± 7 mg/kg (± one median absolute deviation from the median; MAD) at the industrial site was marginally greater than UCC value (17.3 mg/kg; Table 3). Computations of Al-normalized ERs indicate that Co had low median values of 1.8 and 1.2 for industrial and residential sites, respectively (Fig. 3). However, the industrial site #13 had an exceptionally high Co concentration (144 mg/kg) and ER_{Co} (17), indicating a strong anthropogenic signal. This site was potentially impacted by several nearby sources, such as scrap metal processing, stainless steel fabrication, printing, and automotive-related businesses.

3.2.2. Chromium

Concentrations of Cr were significantly greater in RDS on industrial roads (402 mg/kg) compared with residential roads (312 mg/kg). The concentrations for both land uses were 3–4 fold greater than concentrations in the UCC (i.e., 92 mg/kg; Table 3). Automotive sources of Cr include brake linings, tires, fuels, and engine wear particles (Table 1). Metal works, microelectronic production and petrochemicals are industrial sources of Cr (Joshi et al., 2009). The observed differences between industrial and residential Cr concentrations are likely caused by the abundance of Cr emitting sources in the former; however, our current methodology does not allow us to verify this conclusively. The Cr values at all 30 locations exceeded the probable effect concentration (PEC) for aquatic sediments (111 mg/kg), and would likely cause adverse effects to sediment-dwelling organisms (Table 3). Although median ER_{Cr} values were <10 for both industrial (6.6) and residential sites (5.4), five of 15 industrial sites and two of 15 residential sites had ER_{Cr} values >10 (not shown). As with Co, industrial site #13 had the highest Cr concentration (4647 mg/kg), which was 2-fold higher than the next highest RDS concentration at industrial site #9 at 1997 mg/kg. Compared with ER_{Cr} of sediments sampled by Wood et al. (1997) along the east–west axis of Johor Strait, between Malaysia and Singapore (range of 0.4–0.9; $n = 24$), our RDS ER_{Cr}-values reflect site specific anthropogenic signals, even though the median ER_{Cr} values were <10.

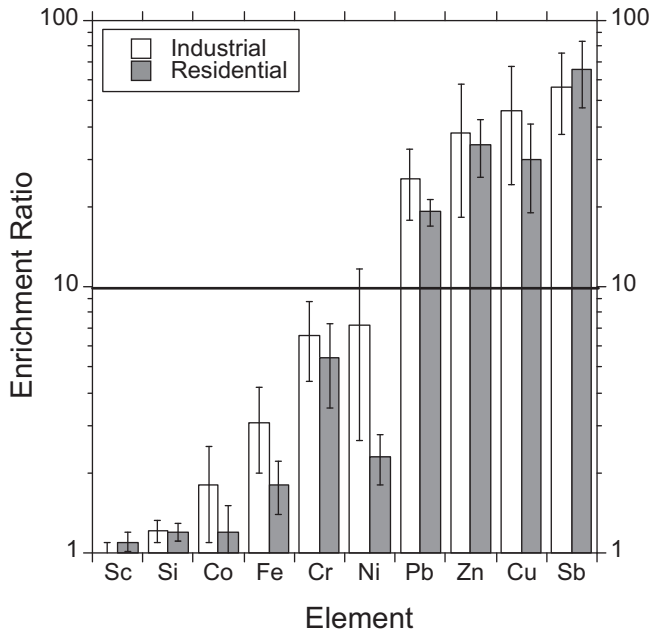

Fig. 3. Element enrichment ratios of road deposited sediments in industrial and residential areas in Singapore Equation (1). Values are medians ± one median absolute deviation from the median. An ER value >10 indicates anthropogenic enrichment. Aluminum was not included as it is the conservative element used to calculate enrichment ratios.

3.2.3. Iron

Concentrations of Fe were significantly higher in industrial RDS (73,020 mg/kg) compared with residential RDSs (51,700 mg/kg) (Table 3). Iron is a common major element in the UCC, with an average concentration of about 39,000 mg/kg (Rudnick and Gao, 2003). Given the high background levels of Fe, it is unusual to identify sites that clearly exhibit anthropogenic Fe signals from concentration data alone. Enrichment ratios on average were <10 for both industrial (3.1) and residential (1.8) sites (Fig. 3). Only two industrial sites had ER_{Fe} >10: site #15 (12) and site #13 (11). In comparison, Johor Strait sediments ($n = 19$) had a median ER_{Fe} of 0.7 (range = 0.5–1.8), much lower than both our residential and industrial samples, which exhibit some moderate degree of anthropogenic input (Wood et al., 1997).

3.2.4. Nickel

The final element exhibiting a statistically significant higher concentration in industrial sites was Ni, which had a median concentration 3-fold higher than residential RDS (204 vs. 76 mg/kg) (Table 3). The background Ni concentration in the UCC is 47 mg/kg. Fourteen sites in each land use had RDS that exceeded the PEC threshold value of 48.6 mg/kg. Thus, Ni may have potential adverse effects on sediment-dwelling aquatic organisms. Six of 15 industrial sites had ER_{Ni} values >10, ranging from 15 to 288 (the median of all 15 sites was 7.2). Residential sites had a median ER_{Ni} value of 2.3 (Fig. 3); and only site #6 had an ER_{Ni} >10. As with Co, Cr, and Fe, industrial site #13 had an anomalously high Ni concentration (6660 mg/kg). The concentration was almost 7-fold higher than the second ranked industrial RDS site (i.e., site #29 at 976 mg/kg). In general, high Ni concentrations in RDS from Singapore industrial sites were related to scrap metal processing, recycling and metal works (cf. Balasubramaniam and Qian, 2004).

3.2.5. Scandium

Scandium was the only element of the 11 examined in detail that had a significantly higher concentration in residential sites compare to industrial sites (Table 3). However, an absolute median Sc concentration difference of 1.2 mg/kg between the two areas is likely not practically significant (Table 3). In fact, median Sc concentrations for both land uses (8.5 and 9.7 mg/kg) were lower than the average (14 mg/kg) reported in the UCC (Table 3). Scandium is commonly used in the geochemical literature as a normalizing agent since anthropogenic sources are minimal. The geogenic nature of this element is reflected in ER_{Sc} values of 1.0 and 1.1 for industrial and residential sites, respectively (Fig. 3). Similarly, ER_{Sc} values calculated for sediments from the Johor Strait (Wood et al., 1997) ranged from 0.4 to 1.1.

3.2.6. Copper

Copper is an element commonly found to be anthropogenically enriched in road sediments (Yun et al., 2000; Andrews and

Sutherland, 2004; Zhang and Wang, 2009; Zhao et al., 2009). Copper in the UCC (28 mg/kg) is much lower than the median values found in our industrial (729 mg/kg), and residential (498 mg/kg) RDS samples (Table 3). The median ER_{Cu} values for industrial and residential sites were 46 and 30, respectively, indicating excessive enrichment (Fig. 3). Industrial sites #13, #15, and #9 had the highest ER_{Cu} values: 310 (4271 mg/kg), 288 (3585 mg/kg), and 94 (1484 mg/kg), respectively (Table 4; Fig. 4a). The average Cu concentration from industrial RDS samples collected across Singapore in this study were lower than the single site sampled by Joshi et al. (2009): i.e., 1142 vs. 9069 mg/kg (*cf.* Table 5). Anthropogenic enrichment of Cu at all residential sites likely reflected a combination of vehicle-related sources and local deposition of atmospheric particulate material (APM) from industrial and waste incineration sources. Residential sites #16, #20, and #18 had the highest ER_{Cu} values: 64 (1094 mg/kg), 57 (968 mg/kg), and 50 (868 mg/kg), respectively (Table 4; Fig. 4b). The residential RDS from this study had an average Cu value 2-fold higher than RDS from a single residential site sampled by Joshi et al. (2009) in prior work in Singapore (544 versus 247 mg/kg); and it was similar to the 570 mg/kg fly ash value determined by Wu and Ting (2006) (*cf.* Table 5). Vehicular sources for Cu include brake wear, tire wear, fuel and oil leakage, and wear of engine parts (Table 1). Potential industrial sources of Cu in this study include metals works, auto parts and printing, and various shipyard related activities (Hu and Balasubramaniam, 2003; Joshi et al., 2009).

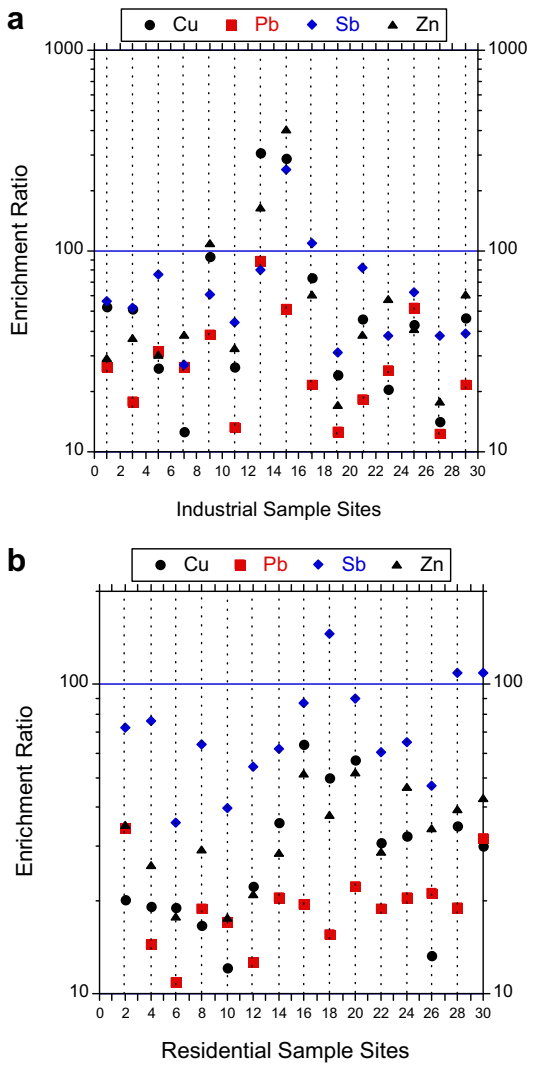

Fig. 4. Road deposited sediment enrichment ratios for Cu, Pb, Sb and Zn at each of the (a) 15 industrial sites and (b) 15 residential sites. An ER value >10 indicates anthropogenic enrichment.

3.2.7. Lead

Lead concentrations were less variable at residential sites, with a coefficient of variation (CV) of 30% compared with 53% for industrial RDS sites (CV = std dev/mean; Table 5). Lead at industrial sites ranged from 144 to 744 mg/kg (median = 280 mg/kg); residential RDS sites ranged between 135 and 395 mg/kg (median = 201 mg/kg) (Tables 3 and 4). In comparison, Pb in the UCC is 17 mg/kg. The median ER_{Pb} for industrial and residential sites was 25 (range: 12–89) and 19 (range: 11–34), respectively (Fig. 3). The three highest ER_{Pb} values were recorded in the following industrial sites (Fig. 4a): #13 (89), #25 (52) and #15 (51). Residential EF_{Pb} values were more constrained, with the two highest values recorded at the following sites (Fig. 4b): #2 (34) and #30 (32). Most residential sites had EF values of approximately 20.

Lead concentrations in RDS sampled at both land-use locations are within the range of data reported by Zhou et al. (1997) for Singapore roadside soils (27–1981 mg/kg; Table 5). Mean Pb concentrations in residential RDSs in our study (215 mg/kg) were 3-fold higher than determined by Joshi et al. (2009) in RDS at a single residential location (69 mg/kg; Table 5). The mean Pb concentration in RDS from our industrial sites was comparable to the average reported by Joshi et al. (2009) for their site (303 versus 338 mg/kg). Road deposited sediment Pb concentrations were also substantially lower than those reported for fly ash in Singapore: i.e., 2000–2120 mg/kg (Tan et al., 1997; Wu and Ting, 2006).

Lead contamination in urban environments has been widely studied because of its ubiquitous distribution in soils and RDSs, and its known toxicity (Naja and Volesky, 2009). In the past, the largest source of atmospheric Pb in Singapore was associated with leaded-gas combustion, as well as the manufacture of batteries, metal products, paints, and ceramics (Ng et al., 2005). Additional sources for Pb contamination include tire wear, long-range atmospheric transport from other Southeast Asian countries, and municipal solid waste incineration. Lead concentrations in petrol in Singapore have steadily decreased from 840 to 800 mg/L in 1980, to 600 mg/L in 1981, to 400 mg/L in 1983, to 150 mg/L in 1987, to 13 mg/L currently (Chin, 1996; Foo and Tan, 1998; Banerjee, 2011). Unleaded petrol was introduced to Singapore in January, 1991 (Chin, 1996). The reduction of Pb in petrol has been reflected in temporal decreases in Pb in APM collected in Singapore. For example, Neo et al. (2000) found that Pb in roadside APM decreased by an order of magnitude from 1.1 $\mu g/m^3$ in 1986 to 0.13 $\mu g/m^3$ in 1997. In 2005, Pb levels in $PM_{2.5}$ and PM_{10} averaged 15.6 and 12.6 ng/m^3, respectively (Karthikeyan et al., 2006). Reductions and the eventual ban of Pb in petrol sold in Singapore have also been reflected in blood-Pb levels in non-occupationally exposed Singaporeans. For example, geometric mean blood-Pb levels decreased from 151 $\mu g/L$ in 1986, to 76.6 $\mu g/L$ in 1990/1991, to 66 $\mu g/L$ in 1995/1997 (Neo et al., 2000). Despite Pb decreases, which are exhibited worldwide, there may still be a substantial store of Pb in the environment, particularly in roadside soils. Remobilisation of stored Pb from soils by erosion processes or human disturbance can contribute to contemporaneous Pb levels in RDS (Sutherland and Tolosa, 2001; Sutherland, 2003); and this may be the case in Singapore.

3.2.8. Antimony

Antimony was the most enriched of all elements in RDS, regardless of land use (Fig. 3). The median concentration of Sb in industrial (13.8 mg/kg) and residential (17.5 mg/kg) RDS were substantially higher than Sb in the UCC (0.4 mg/kg) (Table 3). They

Table 5
Summary of mean (± one standard deviation) Cu, Pb and Zn concentrations in solid media in Singapore (mg/kg).

Reference	Solid media	Description	*n*	Cu	Pb	Zn
This study	Road sediment	Residential (<63 μm)	15	544 ± 263	215 ± 65	1447 ± 427
		Industrial (<63 μm)	15	1142 ± 1190	303 ± 162	2767 ± 2841
Chen et al. (1996) [a]	Reservoir sediment	Kranji A, B, C	3	21.1–28.6	34.8–82.4	112–367
		MacRitchie A, B, C	3	12.6–20.6	19.1–64.3	75.0–133
Chen (1999) [b]	Soil	Residential (0–5 cm)	25	16.2 (2.9–90.3)	20.1 (6.0–69.6)	33.5 (8.6–92.5)
		Industrial (0–5 cm)	13	106 (1.1–485)	101 (50.8–235)	411 (16.0–3594)
Cuong et al. (2005) [b]	Mangrove sediment	S. Buloh	9	7.1 ± 6.03	12.3 ± 5.2	51.2 ± 40.0
		S. Khatib Bongsu	4	32.0 ± 14.3	31.0 ± 6.2	120 ± 13.9
Cuong and Obbard (2006) [b]	Marine sediment	Kranji – W. Johor Strait	triplicates	17.9 ± 3.2	26.1 ± 2.7	62.1 ± 2.9
		Pulau Tekong	triplicates	7.7 ± 1.3	29.8 ± 6.2	49.8 ± 10.9
Flammang et al. (1997) [a]	Coastal sediment	Coral reefs, south of Singapore	8	113–401	27–56	56–95
Goh and Chou (1997) [a]	Coastal sediment	Various offshore & mainland locations	20	1.4–1781	1.4–82.2	94.9–281
Joshi et al. (2009) [b]	Road sediment	Residential (<355 μm)	26–27	247 ± 53.5	68.6 ± 25.9	275 ± 55.1
		Industrial (<355 μm)	26–27	9069 ± 3742	338 ± 55.6	1696 ± 446
Joshi and Balasubramanian (2010) [b]	Road runoff sediment	Residential	120	33.6	52.8	389
		Industrial	120	217	87.1	892
Nayar et al. (2004) [a]	Estuary sediment	Ponggol estuary	72	34.7 (3.7–439)	17.3 (1.2–157)	n/a
Ng et al. (2005) [b]	Soil	Main island (0–5 cm), <2 mm	6	5.2–15.6	13.6–25.6	10.7–31.3
Orlic and Tang (1999) [b]	Marine sediment	Seven coastal regions around Singapore	38	13–242	24–88	81–451
Tan et al. (1997) [b]	Fly ash	Tuas municipal solid waste incineration	n/a	990	2120	8080
Wood et al. (1997) [b]	Marine sediment	Strait of Johor	13–23	28.4 ± 24.2	50.5 ± 30.2	135 ± 67.9
Wu and Ting (2006) [b]	Fly ash	Municipal solid waste incineration	n/a	570	2000	6288
Zhou et al. (1997) [b]	Soil	Roadside	6	30.2–67.7	27.3–1981	124–938
	Soil	Residential	7	12.5–57.4	15.7–46.4	70–151
	Soil	Industrial	8	11.8–194	28.7–167	111–837

n/a – not available.
[a] HNO_3 digestion.
[b] Total.

were also higher than those in 24 surface soils from Japan (mean = 0.37 mg/kg), unpolluted B-horizon soils in the USA (mean = 0.52 mg/kg), and various soil types in China (mean = 1.06 mg/kg) (Filella et al., 2002). Furthermore they were lower than those for sediments from 23 stations in the Johor Straits: median = 1.4 mg/kg; range = 1–3 mg/kg (Wood et al., 1997). The median ER_{Sb} value in industrial RDS was 57 (range: 27–255); 65 (range: 36–146) for residential sites. Although the median enrichment ratio was higher at residential sites, the difference was not significant ($p = 0.21$). The two highest ER_{Sb} values in RDS from industrial sites were sites #15 (255) and #17 (110) (Fig. 4a). The three highest residential ER_{Sb} values were recorded at #18 (146), #28 (109), and #30 (109) (Fig. 4b). Enrichment ratios computed from the Johor Straits data of Wood et al. (1997) ranged from 2.2 to 9.3.

Antimony has received much less attention than Pb in the environmental contaminant literature despite its toxicity and carcinogenic properties (van Velzen et al., 1998). Antimony and its compounds are considered pollutants of priority interest by the US Environmental Protection Agency and the European Union (Filella et al., 2002). The most common anthropogenic source for Sb in the urban environment is the wear of asbestos-free brake linings (Table 1; Helmers, 1996; Dietl et al., 1997). Antimony concentrations in car brake dust have been found to range from 4 to 16,900 mg/kg (Thorpe and Harrison, 2008).

The median Sb/Pb ratio for industrial RDS in Singapore (0.057 ± 0.021) was significantly lower ($p = 0.021$) than those in RDS in residential areas (0.079 ± 0.022). The difference in Sb/Pb ratios may reflect different break lining compositions between vehicles in industrial versus residential areas—i.e., a greater percentage of heavy vehicles are used in the former (see section 3.5). For comparison, the median Sb/Pb ratio for RDS in Honolulu, Hawaii was 0.030 (Sutherland and Tolosa, 2000). Ratios of Sb/Pb in road runoff sediments in both residential (0.029) and thoroughfare (0.053) locations were reported from Hamburg, FRG (Dannecker et al., 1990).

3.2.9. Zinc

Zinc is a common element found to be anthropogenically enriched in RDS worldwide (Yun et al., 2000; Andrews and Sutherland, 2004; Zhang and Wang, 2009). Zinc concentrations were substantially more variable in industrial versus residential RDSs (CV = 103% versus 30%; Table 5). The median concentration of Zn in industrial RDSs was 1649 mg/kg (range: 826 to 11,960 mg/kg), and 1543 mg/kg (range: 704 to 2105 mg/kg) in residential RDSs (Table 3). These values are substantially higher than Zn in the UCC (67 mg/kg; Table 3). Zinc concentrations from all 30 sites exceeded the PEC threshold value of 459 mg/kg (Table 3), and therefore, could potentially pose a significant toxic threat to aquatic-dwelling organisms.

Median ER_{Zn} values for industrial and residential RDSs were 38 (range: 17 to 401) and 34 (range: 18 to 52), respectively. The three highest ER_{Zn} values in RDS from industrial sites were (Fig. 4a): #15 (401), #13 (165), and #9 (109). The three highest residential ER_{Zn} values were recorded at the following sites (Fig. 4b): #20 (52), #16 (52), and #24 (47).

The arithmetic mean Zn concentration in residential RDSs from this study (1447 mg/kg) was comparable to the highest value sampled by Zhou et al. (1997) in Singapore roadside soil (938 mg/kg), suspended sediment sampled by Joshi and Balasubramanian (2010) from industrial road runoff (892 mg/kg), and the RDS sampled by Joshi et al. (2009) in an industrial site (1696 mg/kg). The average Zn value in RDS from industrial roads (2767 mg/kg) was exceeded only by fly ash values (6288 mg/kg and 8080 mg/kg) reported by Wu and Ting (2006) and Tan et al. (1997) (Table 5).

Overall, zinc shows a significant anthropogenic signal in RDSs from Singapore. The high concentration of 11,960 mg/kg from industrial road #15 was more than 2-fold higher than the next highest RDS site (#13; 5451 mg/kg). Zinc is a common element in vehicle brake linings, tires, fuels and oils (Table 1). Thorpe and Harrison (2008) report Zn concentrations in car tire treads up to

10,250 mg/kg; between 25 and 188,000 mg/kg in brake linings; and 120 to 27,300 mg/kg in car brake dust. Zinc in Singapore is also associated with oil-fired power plants, oil refineries, metal-related industries and waste incineration (Hu and Balasubramaniam, 2003; Balasubramaniam and Qian, 2004). All of these vehicular and industrial sources are potential contributors to RDS at the industrial and residential sites sampled in this study.

3.2.10. Silicon

Silicon was found to be a geogenically-controlled element, as ER values were 1.2 for RDS in both land uses (Fig. 3). The results for Si validate the ER approach used in this study: i.e., an ER value of approximately 1 confirms, as expected, that there are no significant anthropogenic sources of these elements in Singapore.

3.3. Correlations of significantly enriched elements

Spearman rank-order correlation was used to explore the strength of association between Cu, Pb, Sb, and Zn—the four elements that were identified as having ER values >10 (Fig. 3). Our assumption was that statistically-related elements may reflect similar sources of contamination. Six pairwise comparisons were made for the two separate land uses explored in this study. Copper in RDS was significantly correlated with Sb and Zn in industrial and residential sites (Table 6). This association suggests a common source, likely dust from brake linings, which are known sources of all three elements. Lead was not significantly correlated with Sb, suggesting that the Pb source to road surfaces is something other than brake linings. Based on prior work in Honolulu, Hawaii (Sutherland et al., 2003), we believe Pb in RDS could be mobilized from previously stored Pb in roadside soils (i.e., legacy Pb). Additionally, we do not exclude local deposition of atmospheric particulate matter emitted from waste incinerators, or possibly long-range sources in locations throughout Southeast Asia. Lead was also significantly correlated with Zn in industrial and residential sites. Potential sources that could account for such an association between Pb and Zn include tire wear, fuel and oil leakage from vehicles, and exhaust emissions as Pb is still in unleaded gasoline in Singapore (13 mg/kg) and Zn additives are used in engine oil for wear protection (Davis et al., 2000).

3.4. Pollutant load index (PLI)

The Tomlinson et al. (1980) PLI (Equation (2)) was used to characterize the multi-element contamination level for the 30 RDS samples. Values of PLI_{Site} greater than 1 reflect an increasing multi-element contaminant signal. It is evident that the anthropogenic signal is generally stronger for industrial sites (Fig. 5): average PLI_{Site} was 25 ± 19 (± one standard deviation) (range: 9.3 to 82.8) for industrial sites versus 15 ± 4 (range: 7.6 to 21.7) for residential sites. The industrial sites that exhibited the highest contamination from a multi-element perspective were site #13 (PLI = 83) followed by site #15 (52). For residential areas, the PLI_{Site} was highest for sites #28 (22) and #16 (21). For similarly ranked sites between the two land uses, PLI_{Site} for industrial locations exceeded those for residential sites ($p = 0.041$). Finally, a calculation of the overall $PLI_{Landuse}$ (Equation (4)) for the two contrasting areas indicated that the industrial site's value of 20.6 exceeded that of the residential site's value of 14.6.

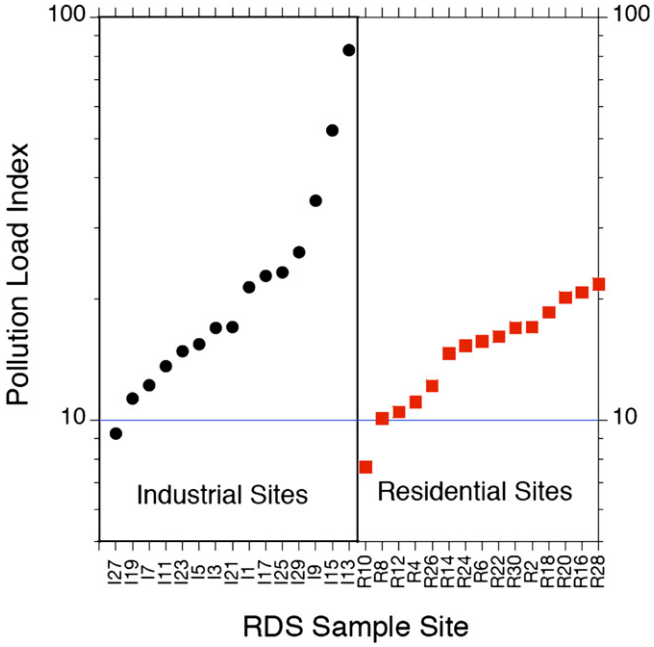

Fig. 5. Pollutant Load Index for road deposited sediments in industrial and residential sites in Singapore Equation (2).

3.5. Correlations with traffic intensity

Data from the intensive one-time traffic survey indicated that the mean number of vehicles on industrial and residential roads were similar (357 versus 348 vehicles/hour) (Table 7). Cars were the dominant vehicle on residential (68%) and industrial (43%) roads, 235 versus 153 vehicles/hour, respectively. Heavy vehicles and lorries were more common on the industrial roads (126 vehicles/hour; 35%) compared to residential roads (37 vehicles/hour; 11%). Buses and van traffic was similar in industrial and residential areas (approximately 50 vehicles/hour). Sites #5 and #6 were the busiest industrial and residential roads, having an average of 1452 and 757 vehicles per hour, respectively (Table 4). Although heavy metal concentrations are acknowledged to have a positive association with vehicular volumes or heavy vehicles (Wong et al., 2000; Charlesworth et al., 2003; Pandian et al., 2009), no such significant relationship ($\alpha = 0.05$) was observed in this study using the Spearman rank-order correlation statistic for the four elements with median enrichment ratios >10, i.e., Cu, Pb, Sb and Zn (not

Table 6
Pairwise comparisons between Cu, Pb, Sb, and Zn in road deposited sediment from Singapore for industrial ($n = 15$) and residential sites ($n = 15$) using Spearman rank-order correlation.

Pairwise Element Comparison	Industrial[a]	Residential[a]
Cu vs. Pb	0.46 (0.087)	0.11 (0.68)
Cu vs. Sb	0.64 (0.017)*	0.58 (0.031)*
Cu vs. Zn	0.61 (0.023)*	0.58 (0.031)*
Pb vs. Sb	0.40 (0.13)	0.21 (0.43)
Pb vs. Zn	0.67 (0.012)*	0.63 (0.018)*
Sb vs. Zn	0.28 (0.30)	0.56 (0.037)*

[a] Spearman correlation coefficients (r_s) are reported with tied p-values in parentheses. Values identified with an asterisk are considered statistically significant at an α-level of 0.05.

Table 7
Mean numbers of vehicles on both industrial (IND) and residential (RES) roads per hour (April 11, 2011)

	Total[a]	Motorcycle & Scooter	Car	Bus & Van	Lorry	Heavy vehicles
IND	357 ± 329	26 ± 32	153 ± 219	52 ± 51	104 ± 61	22 ± 37
RES	348 ± 200	27 ± 18	235 ± 135	48 ± 36	36 ± 28	1 ± 2

[a] Values are means ± one standard deviation ($n = 15$ for each road class).

Table 8
Variation in sweeping efficiency for eight grain size fractions of road deposited sediment from one industrial and one residential site, Singapore.

	Unit	All	>4000 μm	2000 μm[b]	1000 μm[c]	500 μm[d]	250 μm[e]	125 μm[f]	63 μm[g]	<63 μm
Industrial Site										
Mass Before Sweeping	g	261	42	19	27	45	55	51	20	3
Inorganic Fraction	%	78	34	78	86	92	90	85	85	81
Organic Fraction[a]	%	22	66	22	14	8	10	15	15	19
Removal Efficiency (Total)	%	41	85	66	59	55	41	21	−35	−317
Removal Efficiency (Inorganic)	%	33	73	69	59	55	39	15	−42	−360
Removal Efficiency (Organic)	%	68	91	55	55	54	53	54	6	−134
Residential Site										
Mass Before Sweeping	g	848	31	38	95	260	226	150	39	10
Inorganic Fraction	%	92	64	86	90	94	94	94	92	89
Organic Fraction[a]	%	8	36	14	10	6	6	6	8	11
Removal Efficiency (Total)	%	57	74	69	66	74	53	34	18	28
Removal Efficiency (Inorganic)	%	58	79	70	66	74	54	35	19	29
Removal Efficiency (Organic)	%	52	65	62	66	68	46	14	8	19

[a] The organic fraction was determined using loss-on-ignition, whereby >5-g, oven-dried subsamples were ignited in a muffle furnace for 4 h at 550 °C.
[b] 2000 μm represent sediments between 4000 and 2000 μm.
[c] 1000 μm represent sediments between 2000 and 1000 μm.
[d] 500 μm represent sediments between 1000 and 500 μm.
[e] 250 μm represent sediments between 500 and 250 μm.
[f] 125 μm represent sediments between 250 and 125 μm.
[g] 63 μm represent sediments between 125 and 63 μm.

shown). Thus, the interaction between traffic and sediment-associated contaminant loads is not straight forward. On one hand these traffic count data reflect only a single snapshot of the total daily traffic volumes. However, lack of correlation may also indicate that at least some of the contaminants may come from a mixture of point and non-point sources including vehicles, industry, and long-range transport.

3.6. Effectiveness of street sweeping

The bulk of the material found on the road surfaces was inorganic (78–92%; Table 8). The street sweeper removed 41–57% of all material on the road surface (Table 8). Sweeping was significantly more effective at removing all material >500 μm at all sites combined (55–74% removal of both organic and inorganic material). In general, sweeping was of limited effectiveness in removing material <125 μm, as no more than 30% was removed at either site. In fact, the industrial site showed an anomalous increase in material <125 μm following sweeping.

Several studies report limitations of road sweeping (Sartor and Boyd, 1972; USEPA, 1983; Sartor and Gaboury, 1984; Walker and Wong, 1999; Yee, 2005). The results of our pilot sweeping study are particularly alarming because of the enrichment of the <63-μm fraction – the very fraction to which road contaminants are preferentially sorbed. In partial explanation, the scrubbing action of the rotating sweeper brushes may have detached fine sediments that were attached to the road surface (*cf.* Vaze and Chiew, 2002). In general, most street sweepers are designed to remove large debris (Chang et al., 2005). Additionally, the rotating action of the brush may move sediments from the center of the road to the curbside where RDS samples were collected.

4. Summary and recommendations

The greater amount of sediment stored on the surface of industrial versus residential roads may reflect failure to control erosion on near-road sources, inability of sweepers to access the full length of road because of parked vehicles, and possibly less frequent sweeping in industrial areas. Concentrations of Co, Cr, Fe, and Ni were higher in industrial RDS than those in residential RDS, but Sc was higher in residential RDS. However, none of these five contaminants were greatly enriched over upper continental crust background values. In contrast, significant enrichment signals were observed for Cu, Pb, Sb, and Zn in both industrial and residential RDS, i.e., ER-ratios >10. Furthermore, Cu, Pb and Zn concentrations exceeded sediment quality guideline PEC threshold values, indicating that their off-site transport to receiving water bodies would likely negatively impact sediment-dwelling aquatic organisms in Singapore. Anthropogenic inputs of these contaminants to Singapore roads likely include both vehicular (e.g., brakes, tires, emissions, etc.) and industrial sources (e.g., oil-fired power plants, oil refineries, metal related industries, ship building/repair, and waste incineration). The sampling methodology employed in this study demonstrated that hotspots of high concentrations were spread throughout the island, but not all industrial roads could be considered equally contaminated. Two industrial sites (#13 and #15), a shipyard and an area supporting metal works shops, hardware stores, auto parts, stainless steel works, were notable for having exceedingly high concentrations of several potentially toxic elements. In addition, while the focus of our study was on differences between industrial and residential sites, sample collection multiple times throughout the year could be undertaken in future to ascertain seasonal variability in contamination levels, as found earlier by Joshi et al. (2009).

Street sweeping was found to be of limited effectiveness for removing fine fractions (<125 μm) from road surfaces. These preliminary data have important management implications for future reductions in sediment-associated transport of potentially toxic elements to receiving water bodies in Singapore. Failure to remove fine material allows for the re-suspension by wind or vehicles or overland flow transport into water bodies by storm runoff, thereby potentially adversely affecting human and aquatic health. Sweeping was effective at removing gross pollutants, such as litter, vegetation and coarse sediments. Multiple passes of the road sweeper may not prove to be an effective strategy, as subsequent passes might dislodge additional fine material that will not be removed by simple brush and vacuum sweeping technology. Investment in new technologies may be needed to improve the overall efficiency of removing small grain size fractions, which contain preferentially high concentrations of PTEs, from the roads in Singapore.

It is also important to consider alternative means of reducing the input of inorganic PTEs from vehicles to the road environment. The use of alternative fuels such as liquefied petroleum gas, compressed

natural gas, and dimethyl ether are examples. One drawback though, is that emissions of certain elements such as Hg might increase from their use. The use of battery powered electric and hybrid vehicles is another alternative. Another solution would be to reduce the concentrations of PTEs (Cu, Pb, Zn) in brake linings and tires. Legislation, such as that of the European Parliament and Council mandating the non-usage of Cd, Cr, Hg and Pb in vehicular components, can reduce contaminant input to roads.

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