Trace elements in road-deposited and water bed sediments in Kogarah Bay of Sydney, Australia: Enrichment, sources, and fractionation

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Abridged title: Trace elements in road-deposited sediments

Summary text

Road dusts deposited along busy roads commonly have toxic chemical elements which may be washed down by rain into rivers, lakes and bays causing water pollution that affects fish and other living creatures. Our study in a specific area of Australia showed that five such elements mainly originating from vehicle brakes, tyres and road surface abrasion were highly enriched in dust deposits. This did not, however, increase the elemental contents in the water sediments in a nearby bay. **Abstract.** Trace elements (TE) in road-deposited sediments (RDS) can be transported by stormwater to neighbouring water bodies to cause aquatic pollution. A study was conducted in Kogarah Bay of Sydney, Australia to assess the possible sources and potential mobility of TE in RDS and its contribution to the TE load to the adjacent water bed-sediments (WBS) in canals and bay. Of the 11 TE analysed, pseudo-total concentrations of Zn, Cu, V, Cr, and Sb were greatly enriched in RDS than in baseline soils (BS) (top 10 cm depth) collected in bushlands. All TE concentrations in WBS (top 10 cm depth) were similar to those in BS but lower than those in RDS. Correlation and principal component analyses revealed that Zn, Cu, Cr and Sb were related to each other in RDS, and they probably originated from tyres and brake linings. Vanadium occurred in another component, likely to have originated mainly from road asphalt. Pseudo-total and mobile fraction (0.1 M acetic acid, pH 2.85 extraction) TE concentrations in RDS were: Fe >Mn, Zn > Cu, Pb > Cr, Ni, V, Sb, Cd. The potential ecological TEs risk was low to medium in RDS but low in BS and WBS.

Additional keywords: heavy metals, road-deposited sediments, water sediments, trace elements fractionation, ecological risk.

Introduction

The escalating rate of vehicle usage as a result of the urban population's rapid growth is causing increasing amounts of road-deposited sediments (RDS) in many parts of the world. RDS contain high concentrations of inorganic and organic pollutants, and of these, trace elements (TE) particularly heavy metals, constitute a major component (Loganathan *et al.* 2013). Many of the TE at elevated concentrations have harmful effects on humans and aquatic environments (Hakanson 1980; Lynam and Pfeifer 1991; Scanlon 1991). The sources

of the TE in RDS are vehicle parts and exhaust emissions, road surfaces, and lithology of the area (Taylor and Owens 2009; Loganathan *et al.* 2013). The mobile fraction of TE in RDS can easily be dissolved in stormwater and can potentially be transported into adjacent water bodies. Other fractions can also be transported along with suspended solids in the water. Trace elements stored in water bed-sediments (WBS) are released slowly over time and become bioavailable to aquatic organismms.

The degree of TE enrichment in RDS depends on the relative contribution of the various sources of TE such as baseline soils (BS), vehicle activities and abrasion of road surfaces. The contribution of vehicle activities and roads can be assessed by comparing the TE concentrations in RDS with those in nearby BS away from the roads with minimum contamination using different types of indices, for example: a geo-accumulation index (I_{geo}) (Faiz *et al.* 2009; Lu *et al.* 2009), an enrichment factor (EF) (Lu *et al.* 2009), and a pollution index (PI) (Faiz *et al.* 2009; Lu *et al.* 2009).

Source identification of TE is important so that the correct measures to prevent pollution are implemented. Multivariate statistical approaches such as correlation coefficient, cluster and principal component analyses have been employed for source identification (Yongming *et al.* 2006; Mohammed *et al.* 2012). In these analyses, TE which are intercorrelated to form principal components are considered to have originated from common sources.

The mobility and bioavailability of TE in RDS, WBS, and NBS depends mostly on the chemical forms in which the metals exist, rather than on the total concentration. Sequential extraction methods are one method by which the operationally defined readily mobile, less mobile and immobile fractions can be determined. Two general sequential extraction procedures have been used previously. The method developed by Tessier *et al.* (1979) is a five-step sequential extraction method which uses reagents to extract the following fractions in successive order of decreasing mobility: exchangeable, carbonatebound, Fe/Mn oxide-bound, organic-bound and residual. The other sequential extraction method is a simpler three-step procedure developed by the Standards, Measurements and Testing programme of the European Union (Rauret *et al.* 1999; Kartal *et al.* 2006). This method extracts water soluble + exchangeable, reducible (Fe/Mn oxide-bound) and oxidisable (organic and sulphide-bound) fractions. In these methods the water soluble + exchangeable fraction is considered to be readily mobile and the residual fraction originating mainly from natural geological materials is the most immobile fraction.

Numerous studies have been conducted on TE accumulation in RDS (Taylor and Owens 2009; Loganathan *et al.* 2013). However, very few studies have been reported on comparing metal concentrations between RDS, WBS, and BS within a single catchment area (Birch 2011). Such a study is important in assessing the RDS and BS contribution of TE load contributions to local water bodies, so that control measures can be adopted, if necessary, to reduce RDS polluting the water bodies. The aims of the study were to determine TE: (1) distributions in RDS, WBS, and BS; (2) enrichments in RDS and WBS by comparing concentrations in RDS and WBS with those in BS reference; (3) potential mobility and bioavailability using a sequential chemical extraction method; and (4) possible sources using statistical methods.

Materials and methods

Sediments and soils sampling

Samples used in this study were subsamples of those collected for another study conducted to

determine the polycyclic aromatic hydrocarbons distribution in the same area (Nguyen *et al.* 2014). Eleven samples each of RDS and WBS, and 7 samples of BS were collected in and around Kogarah Bay, Sydney, the largest and most populated city in Australia. The sites where the samples were collected are shown in Fig. 1. No major industries were presented close to the catchment. RDS were collected during summer months on asphalted pavement within the shoulder of the roads excluding the gutters within an area of approximately 2 m². Two to three weeks of dry weather were allowed for sufficient RDS to accumulate at each site before samples were collected. The areas were carefully brushed using a clean plastic dustpan and then transferred into self-sealing polyethylene bags for transport to the laboratory for analysis.

Most parts of the catchment area had roads and buildings, leaving only very small areas under bushland which had minimum effect from the above sources with no visible signs of contamination. These sites were 200 to 2000 m away from the various RDS sampling sites. Due to the limited land area available under bushland a smaller number of BS samples were collected. Before collection of BS samples, the vegetation on the land was removed and approximately 1 kg soil collected to a depth of 0.1 m depth from each of three areas 10 m apart using a stainless steel spade. The samples were mixed thoroughly and subsamples of 1 kg soil were transported to the laboratory for analysis.

A total of 11 WBS samples were collected. Six samples were collected (top 0.1 m depth) during summer season using a stainless steel grab sampler in canals that carry stormwaters from the roads (Fig. 1). Each sample was a composite of 3-5 samples collected at 1 m apart. Similarly, five WBS samples were collected (top 0.1 m depth) within the bay at distances of 10 to 100 m from the places where the canals drains to the bay. The depth of water at the sampling sites was 0.2-0.5 m in the canals and 1-2 m in the bay. The bay is > 7 km from the coastline of the Pacific Ocean.

Figure 1

Chemical analysis

The soil and sediment samples were dried at 40°C inside an oven. The dried samples were ground to pass through a 2 mm stainless steel sieve and subsamples of approximately 50 g each were ground using a stainless steel electric grinder before the TE analysis. Pseudo-total concentrations of Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sb, V, and Zn were determined by digestion of 0.5-1.0 g soil or sediment sample using 3 mL 70% HNO₃ and 3 mL 32% HCl for 2 h at 100°C. Of these TE, Sb is a metalloid, V is a light metal, and the rest are heavy metals. The digestion was followed by analysis of the diluted digests using ICP-OES and ICP-MS at the National Measurement Institute, Sydney, Australia. The instruments used were Varian 730-ES ICP-OES and Perkin Elmer DRC 2 ICP-MS. An Australian 'national certified reference standard' (river sediment sample AGAL-10) analysed in parallel with the unknown samples indicated 92-108% recovery of TE (Table 1). The near-100% recovery suggests that the extraction method and chemical analysis used in the study are suitably accurate.

Asphalt samples were collected from three roads in the study area to determine the concentration of V. In each road, approximately 100 g composite samples from five points at distances of 1-5 m apart were collected. The samples were processed, acid digested, and analysed as described for soil and sediments.

Fractionation of the TE was conducted according to the Standards, Measurements and Testing Program of the European Union (Rauret *et al.* 1999; Kartal *et al.* 2006). In this procedure, exchangeable and water and weak-acid soluble TE were extracted first by acetic acid (0.1 M, pH 2.85), followed by extraction of the reducible TE fraction by hydroxylamine

hydrochloride (0.5 M, pH 2), and finally extraction of the oxidisable TE fraction by hydrogen peroxide (8.8M)/ammonium acetate (1M, pH 2). The concentration of TE in the residual fraction was determined by subtracting the sum of the above three fractions' concentrations from the pseudo-total TE concentration. TE concentrations in the fractions were measured using ICP-OES and ICP-MS.

TABLE 1

Pollution index

The Pollution Index (PI) is defined as the ratio of TE concentration in RDS to the corresponding TE background concentration in BS. The PI of each TE was calculated and classified as either low (PI \leq 1), medium (1 \leq PI \leq 3) or high (PI \geq 3) (Faiz *et al.* 2009; Lu *et al.* 2009; Sun *et al.* 2010).

Ecological risk

The method of determining ecological risks of TE originally introduced by Hakanson (1980) has recently been used in many studies (Qiu *et al.* 2010; Sun *et al.* 2010; Saeedi *et al.* 2012). This method was adopted to assess the ecological risks of TE pollution in RDS and WBS:

$$RI = \sum_{i=1}^{m} Er$$

 $Er = T_r \times C_f$ and $C_f = C_r / C_b$

where C_r is TE concentration in RDS or WBS and C_b is TE concentration in BS. Er is the ecological risk of each TE and RI is the ecological risk of multiple TE. Tr is defined as a

"toxic-response factor" for a given TE and Hakanson (1980) demonstrated this value for Cd, Cu, Pb, Cr and Zn to be 30, 5, 5, 2 and 1, respectively. These values are related to the degree of toxicity per unit concentration of the TE.

Statistical analysis

Correlation analysis was conducted between TE separately for RDS and BS to determine their inter-element relationships (Chang *et al.* 2009). Pearson correlation coefficients were employed to examine the degree of significance of the above relationships. A significant and positive correlation between TE often indicates that they are derived from the same sources (Yongming *et al.* 2006; Chang *et al.* 2009; Faiz *et al.* 2009). The correlation was performed using SPSS Statistics version 20.

Principal component analysis (PCA) is a multivariate statistical tool employed to reduce a set of original variables to extract a small number of latent factors called principal components for analysing relationships among the observed variables. Variables with similar characteristics are grouped into factors and factors with eigenvalues greater than one are selected as the principal components. The analysis provides information on the percentage of variance explained by each factor and the factor loadings for each variable. The results of PCA reveal the primary portion of the data variance through these factors; consequently, the TE representative of each factor can be selected as source tracer. Factors having high loading values (high variance percentage of variance) represent the possible TE sources.

The correlation and PCA were done using Predictive Analysis Software (PASW) Statistics version 20, previously called SPSS Statistics.

Results and discussion

Trace element concentrations and enrichments

Concentrations of TE in RDS, WBS and BS in this study were generally in the lower half of the concentration ranges reported in other countries (Table 2). The concentrations of most of the TE in BS did not vary much as evidenced by the low standard errors of means of concentrations (Cd 0.2 ± 0.03 , Sb 0.7 ± 0.09 , V 8.4 ± 1.8 , Cr 4.4 ± 0.9 , Cu 19.2 ± 3.9) which indicates that the mineral composition among the different sampling points in BS was similar. Mean concentrations of all TE were generally higher in RDS than in BS (Fig. 2), indicating that vehicle activity and road surface abrasion have contributed to the accumulation of TE in RDS. The pollution index (PI), the ratio of TE concentration in RDS to that in BS, was more than 3 for Zn, Cu, V, Cr, and Sb in all samples (Fig. 3). Furthermore the medians of the values were more than 5 indicating that these TE were highly enriched in RDS according to the classification of Faiz et al. (2009) and Lu et al. (2009). Vehicle brakes and tyre wear might have been the possible predominant sources for Zn, Cu, Cr, and Sb in RDS and mainly road surface for V (Ozaki et al. 2004; McKenzie et al. 2009; Loganathan et al. 2013). Vanadium can also be contributed by brake linings (Figi et al. 2010) and tyres (Ozaki et al. 2004). Many others also reported enrichment of Zn, Cu, and Sb in RDS (Sweden, Cu, Zn, and Sb, Hjortenkrans et al. 2006; NW China, Cu and Zn, Lu et al. 2009; Pakistan, Cu and Zn, Faiz et al. 2009; Australia, Cu and Zn, Mohammed et al. 2012). The TE concentrations in WBS were the same or marginally higher than in BS for all TE except Pb (median PI lower than ≤ 2 for all TE, except for Pb it was 5), probably because WBS originated from both RDS and BS with the latter being the larger contributor.

TABLE 2, 3;FIGURES 2, 3

Metal sources

Pearson's correlation coefficient analysis showed that in BS, the metals Sb and Cd had no significant correlation with any of the metals (Table 3). In RDS, however, Sb was significantly correlated to Pb, Cu, Zn, Cd, and Cd to Sb, Pb, and Zn. This suggests that Sb, Pb, Cu, Zn, and Cd in RDS might have derived from vehicle activities. These metals are components of tyre treads/or brake linings (Loganathan *et al.* 2013). Therefore these vehicle components would have contributed to these TE in RDS. However, of these TE, only Cu, Zn, and Sb were enriched in RDS when their concentrations were compared to BS producing high PI values (Fig. 3).

Vanadium was significantly correlated with Pb, Cu, Zn, Fe, Cr, and Ni in BS but only with Mn in RDS (Table 3). Therefore, V in RDS would have originated from a source other than BS, possibly from the erosion of asphalt road surface which contains V (Lindgren 1996; Ozaki *et al.* 2004; Béze *et al.* 2012) by abrasion during vehicle transport. The mean V concentration of the asphalt samples from three roads in the study area was $121 \pm 11 \text{ mg/kg}$. This value is nearly double the value of the V concentration in RDS (65 ± 5 mg/kg) and much higher than the value of $8 \pm 2 \text{ mg/kg}$ for BS.

The correlation data for WBS is more similar to BS than RDS (Table 3). For example, Sb had no significant positive correlation to any other metal in both WBS and BS, but was significantly correlated to Pb, Cu, Zn, Cd, and Fe in RDS. These results show that BS would have contributed to the TE load in WBS more than RDS did.

The principal component analysis of RDS, WBS, and BS data grouped the metals into two statistically significant components that together explained 72.5%, 79.6%, and 79.8% of

the total variance of metal concentrations in RDS, BS, and WBS, respectively (Table 4). In BS, the metals, Sb and Cd had high factor loadings in component 2, whereas the other metals had high loadings in component 1 (Table 4, Fig. 4). This is in agreement with the correlation data where these two metals had no significant correlation with the rest whereas the other metals were related to each other. In RDS the TE, Sb, Pb, Cu, Zn, Cd, and Fe which were significantly correlated in the correlation analysis fitted into one component with high factor loadings in PCA (Table 4, Fig. 4). Similarly, V which was significantly correlated with many metals in BS, formed a common component with these metals in PCA but separated from them in RDS into another component. Therefore PCA, as concluded in the correlation analysis, suggests that of the five TE enriched in RDS, Zn, Cu, Cr, and Sb originated from the latter four elements, the dominant source of V is considered to be asphalt road surface which contains significant amounts of this element.

TABLE 4; FIGURE 4

Metal fractionation

Fractionation data for RDS showed that the concentration of the most mobile of the four fractions, the exchangeable TE fraction (Fraction 1), decreased in the order Fe > Mn, Zn > Cu, Pb > Cr, Ni, V, Cd, Sb (Fig. 5). The mobile fraction as a percentage of total concentration was, however, lowest for Fe and highest for Cd, because of the high total concentration of Fe and low total concentration of Cd. Zinc was found to be the second largest percentage of

mobile fraction. Other studies also reported Cd or Zn to be the most mobile TE (Charlesworth *et al.* 2003; Mohammed *et al.* 2012). Considering the total concentration of mobile fraction, Fe, Mn, Zn, Cu, and Pb from RDS were expected to have been transported more than the other TE to the neighbouring water bodies by stormwaters. Transportation of other fractions may have also occurred through the movement of suspended particles in stormwater. However, no major difference in total concentration between WBS and BS was noticed for these or other TE probably due to: firstly, the low amounts of TE transported from RDS compared to the amounts already present in WBS; and secondly, the wider distribution of transported TE between the sediments and large volume of water in the bay.

FIGURE 5

Ecological risk

The Australian and New Zealand Interim Sediment Quality Guidelines (ISQG) concentrations for TE reported to be at low risk based on a battery of biological toxicity tests were 1.5, 2.0, 21, 50, 65, 80, and 200 mg/kg for Cd, Sb, Ni, Pb, Cu, Cr, and Zn, respectively (McCready *et al.* 2006). The corresponding concentrations for high risk were 10, 25, 52, 220, 270, 370, and 410 mg/kg. Based on these guidelines, all WBS and BS sites were at low risk for all TE. In RDS sites, however, Sb, Pb, Cu, and Zn concentrations were between the low and high risk values. It is interesting to note that of these four TE, Zn, Sb, and Cu were enriched in RDS as shown by the PI index (Fig. 3).

Ecological risk assessments of TE determined using Hakanson's (1980) method showed that the RDS sampling sites had low to moderate risk (Table 5). The range of risk index (RI) values for RDS (65-234) are similar to those (46-353) reported for different functional areas in seven districts of Shenyang in China (Sun *et al.* 2010). The RI values for RDS are, however, much lower than the values reported for RDS in Tehran, Iran (Saeedi *et al.* 2012). This is probably due to the smaller concentrations of the TE in this study compared to the study in Iran. The much lower risks in WBS sites are due to the smaller concentrations of Cr, Pb, Cu, and Zn at these sites than at RDS sites (Fig. 2). Of the TE in RDS, Cu had the highest potential ecological risk (Er value) and Zn the least.

TABLE 5

Conclusions

Road-deposited sediments contained elevated concentrations of TE. The ratio of TE concentration in RDS to that in BS was more than 5 for Zn, Cu, V, Cr, and Sb. Both the correlation and principal component analyses showed that Zn, Cu, Cr and Sb in RDS originated mainly from one source, probably vehicle brakes and tyre wear and V mainly from another source, likely to be road asphalt surface abrasion. The TE concentrations were similar in WBS and BS. TE fractionation data showed that TEs' potential mobility, an indication of their transportation by stormwater, decreased in the order Fe > Mn, Zn > Cu, Pb > Cr, Ni, V, Cd, Sb. However, the mobile fraction as a percentage of total TE concentration was highest for Cd and Zn, and lowest for Fe, Cr, Ni and V. Ecological risk as assessed by ISQG and the method developed by Hakanson (1980) was low to medium in RDS but low in BS and WBS.

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	Cert	ified mater	ial reference	(AGAL-10))
ТЕ	Measure (mg/	d values 'kg)	Certified (mg/	l values kg)	Recovery (%)
	Mean	S.D	Mean	S.D	
Cd	10.1	1.0	9.33	0.64	108
Cr	87.0	5.0	82.0	11	106
Cu	23.6	2.0	23.2	1.9	102
Fe	18880	1160	19950	1170	95
Mn	230	20	241	11	96
Ni	18.3	2.0	17.8	2.7	103
Pb	41.4	3.0	40.4	2.7	102
Sb	5.90	3.0	6.40	3.2	92
V	25.1	5.0	25.3	4.0	99
Zn	61.0	5.0	57.0	4.2	107

Table 1. Comparison of measured TE concentrations (mg/kg) obtained using HNO₃/HCl digestion and certified values on the Australian reference standard

Recovery = (measured value/certified value) x 100

Sample	City	Cd	Cr	Cu	Ni	Pb	Mn	Fe	Zn	Sb	V	Reference
RDS	Sydney, Australia	0.2	42	263	14	165	567	19645	544	6.5	65	This study
	Sydney, Australia	0.2-1.7	49-486	314-730	20- 208	36-379	489- 3966	22036- 103000	557-2117	-	-	Mohammed et al. (2012)
	Sydney, Australia	-	34	160	27	880	-	-	850	-	-	Birch and Scollen (2003)
	Tokyo, Japan	0.5	0.3	30	53	11	-	-	1450	1.1	35	Ozaki <i>et al.</i> (2004)
	Birmingham, UK	1.6	-	467	41	48	-	-	534	-	-	Charlesworth et al. (2003)
	Coventry, UK	0.9	-	226	130	47	-	-	680	-	-	Charlesworth et al. (2003)
	London, UK	3.5	-	155	-	1030	-	26000	653	-	-	Schwar <i>et al.</i> (1988)
	Manchester, UK	-	-	113	-	265	-	8767	653	-	-	Robertson et al. (2003)
	Oslo, Norway	1.4	-	123	41	180	833	-	412	-	-	De Miguel et al. (1997)
	Madrid, Spain	-	61	188	44	1927	362	-	476	-	17	De Miguel <i>et al.</i> (1997)
	Aviles, Spain	22.3	42	183	28	514	1661	42200	4892	-	28.1	Ordonez et al. (2003)
	Lulea, Sweden	0.1	13	24	8	34	-	-	60	-	-	Karlsson and Viklander (2008)
	Amman, Jordan	1.7	-	177	88	236	-	7132	358	-	-	Jiries (2003)
	Istanbul, Turkey	1.9	-	208	32	212	398	-	521	-	-	Sezgin et al. (2003)
	Tokat, Turkey	5.4	41	38	128	266	415	-	98	-	-	Tuzen (2003)
	Kayseri, Turkey	10.1	-	67	57	165.5	274	-	-	-	-	Al-Khashman (2007)
	Bahrain	72	144	-	-	697	-	-	152	-	-	Akhter and Madany (1993)
	Dhaka, Bangdalesh	-	136	104	35	53	-	-	169	-	-	Ahmed <i>et al.</i> (2007)
	Tehran, Iran	10.7	33	225	35	257	1215	47935	873	-	-	Saaedi et al. (2012)
	Luanda, Angola	1.1	26	42	10	351	258	-	317	-	20	Baptista and De Miguel (2005)
	Ottawa, Canada	0.4	43	66	15	39	431	-	113	-	34	Ramussen et al. (2001)
	Hawaii, USA	-	273	167	177	106	-	-	434	-	-	Sutherland et al. (2000)
	Urumqui, China	1.2	54	95	43	54	926.6	-	294	-	-	Wei et al. (2010)
	Xian, China	-	167	95	-	231	687	-	421	-	-	Han <i>et al.</i> (2006)
	Baoji, China	-	126	123	49	433	804	-	715	89	-	Lu et al. (2009)
	Beijing, China	0.6	69	72	26	202	-	-	219	-	-	Du et al. (2013)
	Hangzhou, China	3.6	60	212	28	194	-	-	709	-	-	Zhang and Wang (2009)
	HongKong	3.8	-	173	-	181	-	-	1450	-	-	Li et al. (2001)
	Taiwan	-	-	51	17	102	338	25467	227	-	-	Kuo et al. (2009)
	Singapore	2.4	1245	9069	186	338	-	-	1696	-	-	Joshi et al. (2009)

Table 2. Mean or range of concentration of TE (mg/kg) in road deposited sediments in Sydney area and other cities in the world

 Table 2. (Continued)

Sample	City	Cd	Cr	Cu	Ni	Pb	Mn	Fe	Zn	Sb	V	Reference
WBS	Sydney, Australia	0.2	13	48	7	45	72	11329	117	0.2	23	This study
	Sydney, Australia	-	20	110	20	200	-	-	260	-	-	Birch and Scollen (2003)
	Sydney, Australia	-	59	180	15	310	-	-	640	-	-	Birch and Mc Cready (2009)
	Alberoni, Italia	0.4	82	31	26	60	360	20	-	-	-	Sfriso <i>et al.</i> (1995)
	Venice, Italia	2.5	82	57	31	60	430	22	480	-	-	Sfriso et al. (1995)
	Manzala, Egypt	85	-	315	-	135	420	2430	22	-	-	Saeed and Shaker (2008)
	Tamar, UK	1.0	47	330	44	235	59	35124	452	-	-	Bryan and Langston (1992)
BS	Sydney, Australia	4.4	0.2	19	3	59	108	5972	74	0.7	8	This study
	Sydney, Australia	-	16	51	15	330	-	-	330	-	-	Birch and Scollen (2003)
	Sydney, Australia	-	20	62	12	410	-	-	340	-	-	Snowdon and Birch (2004)
	Sydney, Australia	0.0-0.5	10-79	6-225	27-242	24-198	16-2460	6175-47210	71-238	-	-	Mohammed et al. (2012)
	Brisbane, Australia	0.1	0.0	8	0.0	272	1	61	353	-	-	Gunawardana et al. (2012)
	Earth's crust	0.2	100	50	80	14	950	41000	75	-	-	Karbassi et al. (2005)
	Beijing, China	0.2	76	28	-	18	-	-	-	-	-	Liu et al. (2005)
	Chengdu, China	0.4	60	43	-	77	-	-	-	-	-	Liu et al. (2006)
	Hainan, China	0.3	23	30	16	48	-	-	52	-	-	Zhao et al. (2007)
	Kunshan, China	0.2	88	34	31	30	-	-	105	-	-	Chen and Pu (2007)
	Taiwan	-	-	12	132	20	298	27110	68	-	-	Kuo et al. (2009)

-	TE	Sb	Cr	Ni	V	Pb	Cu	Mn	Zn	Cd	Fe
RDS -	Sb	1.00									
KD5	Cr	0.24	1.00								
	Ni	0.19	0.62 (*)	1.00							
	V	0.20	0.67 ([*])	0.34	1.00						
	Pb	0.71 ([*])	0.01	0.22	0.18	1.00					
	Cu	0.72 ([*])	0.68 ([*])	0.73 ([*])	0.38	0.57	1.00				
	Mn	-0.12	0.54	0.05	0.77 ([*])	0.02	0.13	1.00			
	Zn	0.81 (^{**})	-0.01	-0.06	0.34	0.72 ([*])	0.42	0.14	1.00		
	Cd	0.67 ([*])	0.02	0.10	0.23	0.79 ([*])	0.48	-0.09	0.70 ([*])	1.00	
	Fe	0.65 (*)	0.72 ([*])	0.58	0.48	0.41	0.87 (^{**})	0.22	0.42	0.37	1.00

 Table 3. Inter-element correlation in RDS, WBS and BS samples at Kogarah, Sydney (values represent correlation coefficient)

 Table 3. (Continued)

-	ТЕ	Sb	Cr	Ni	V	Pb	Cu	Mn	Zn	Cd	Fe
-		1.00									
WBS	Cr	-0.75 ([*])	1.00								
	Ni	-0.30	0.74(*)	1.00							
	V	-0.82 (**)	0.94(**)	0.59	1.00						
	Pb	-0.54	0.67 ([*])	0.57	0.67(*)	1.00					
	Cu	-0.09	0.36	0.09	0.76(**)	-0.14	1.00				
	Mn	-0.18	0.39	0.04	0.32	-0.19	0.86(**)	1.00			
	Zn	-0.56	0.82(**)	0.87(**)	0.44	0.86(**)	-0.10	-0.08	1.00		
	Cd	0.02	0.27	0.33	0.19	0.72(*)	-0.09	-0.16	0.49	1.00	
	Fe	-0.82 (***)	0.94(**)	0.69 ([*])	0.97 (**)	0.75(*)	0.14	0.22	0.87(**)	0.21	1.00

 Table 3. (Continued)

Sb1.00Cr-0.271.00Ni-0.21 $0.87(^{**})$ 1.00V-0.15 $0.90(^{**})$ $0.72(^{*})$ 1.00Pb-0.09 $0.85(^{**})$ 0.65 $0.99(^{**})$ 1.00Cu-0.15 $0.77(^{*})$ 0.63 $0.95(^{***})$ $0.96(^{***})$ 1.00Mn 0.33 0.50 0.35 0.60 0.63 0.45 1.00 Zn-0.12 $0.94(^{**})$ $0.95(^{***})$ $0.80(^{**})$ $0.74(^{*})$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00	ТЕ	Sb	Cr	Ni	V	Pb	Cu	Mn	Zn	Cd	Fe
Cr -0.27 1.00 Ni -0.21 $0.87(^{**})$ 1.00 V -0.15 $0.90(^{**})$ $0.72(^{*})$ 1.00 Pb -0.09 $0.85(^{**})$ 0.65 $0.99(^{**})$ 1.00 Cu -0.15 $0.77(^{*})$ 0.63 $0.95(^{**})$ 1.00 Mn 0.33 0.50 0.35 0.60 0.63 0.45 1.00 Zn -0.12 $0.94(^{**})$ $0.95(^{***})$ $0.80(^{**})$ $0.74(^{*})$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00	Sb	1.00									
Ni-0.21 $0.87(*)^*$ 1.00 V-0.15 $0.90(*)^*$ $0.72(*)^*$ 1.00 Pb-0.09 $0.85(*)^*$ 0.65 $0.99(*)^*$ 1.00 Cu-0.15 $0.77(*)^*$ 0.63 $0.95(**)^*$ $0.96(***)^*$ 1.00 Mn 0.33 0.50 0.35 0.60 0.63 0.45 1.00 Zn-0.12 $0.94(**)^*$ $0.95(***)^*$ $0.80(**)^*$ $0.74(*)^*$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00 Fe 0.10 0.57 0.43 $0.86(**)$ $0.90(**)$ $0.89(**)$ 0.67 0.51 0.02	Cr	-0.27	1.00								
V -0.15 $0.90(^{**})$ $0.72(^{*})$ 1.00 Pb -0.09 $0.85(^{**})$ 0.65 $0.99(^{**})$ 1.00 Cu -0.15 $0.77(^{*})$ 0.63 $0.95(^{**})$ $0.96(^{***})$ 1.00 Mn 0.33 0.50 0.35 0.60 0.63 0.45 1.00 Zn -0.12 $0.94(^{**})$ $0.95(^{***})$ $0.74(^{*})$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00 Fe 0.10 0.57 0.43 $0.86(^{**})$ $0.90(^{**})$ $0.89(^{**})$ 0.67 0.51 0.02	Ni	-0.21	0.87 (^{**})	1.00							
Pb -0.09 $0.85(*)$ 0.65 $0.99(*)$ 1.00 Cu -0.15 $0.77(*)$ 0.63 $0.95(**)$ $0.96(**)$ 1.00 Mn 0.33 0.50 0.35 0.60 0.63 0.45 1.00 Zn -0.12 $0.94(**)$ $0.95(***)$ $0.80(**)$ $0.74(*)$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00 Fe 0.10 0.57 0.43 $0.86(**)$ $0.90(**)$ $0.89(**)$ 0.67 0.51 0.02	V	-0.15	0.90 (^{**})	0.72 ([*])	1.00						
Cu-0.15 $0.77(^*)$ 0.63 $0.95(^{**})$ $0.96(^{***})$ 1.00Mn0.330.500.350.600.630.451.00Zn-0.12 $0.94(^{**})$ $0.95(^{***})$ $0.80(^{**})$ $0.74(^*)$ 0.660.591.00Cd0.350.110.500.050.020.130.160.361.00Fe0.100.570.430.86(^{**})0.99(^{**})0.89(^{**})0.670.510.02	Pb	-0.09	0.85 (^{**})	0.65	0.99 (^{**})	1.00					
Mn 0.33 0.50 0.35 0.60 0.63 0.45 1.00 Zn -0.12 $0.94(^{**})$ $0.95(^{***})$ $0.80(^{**})$ $0.74(^{*})$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00 Fe 0.10 0.57 0.43 $0.86(^{**})$ $0.99(^{**})$ $0.89(^{**})$ 0.67 0.51 0.02	Cu	-0.15	0.77 ([*])	0.63	0.95 (**)	0.96 (***)	1.00				
Zn -0.12 $0.94(^{**})$ $0.95(^{***})$ $0.80(^{**})$ $0.74(^{*})$ 0.66 0.59 1.00 Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00 Fe 0.10 0.57 0.43 $0.86(^{**})$ $0.90(^{**})$ $0.89(^{**})$ 0.67 0.51 0.02	Mn	0.33	0.50	0.35	0.60	0.63	0.45	1.00			
Cd 0.35 0.11 0.50 0.05 0.02 0.13 0.16 0.36 1.00 Fe 0.10 0.57 0.43 $0.86(^{**})$ $0.99(^{**})$ 0.67 0.51 0.02	Zn	-0.12	0.94 (^{**})	0.95 (***)	0.80(**)	0.74 ([*])	0.66	0.59	1.00		
F o 0.10 0.57 0.43 $0.86(^{**})$ $0.00(^{**})$ $0.80(^{**})$ 0.67 0.51 0.02	Cd	0.35	0.11	0.50	0.05	0.02	0.13	0.16	0.36	1.00	
$\mathbf{Fe} = 0.10 = 0.57 = 0.45 = 0.00(2) = 0.00(2) = 0.07 = 0.01 = 0.02$	Fe	0.10	0.57	0.43	0.86 (**)	0.90 (^{**})	0.89 (^{**})	0.67	0.51	0.02	1.00

(*) correlation is significant at the 0.05 level (2 tailed) (**) correlation is significant at the 0.01 level (2 tailed)

		RDS			WBS			BS	
Element -	PC1	PC2	Comm.	PC1	PC2	Comm.	PC1	PC2	Comm.
Sb	0.81	-0.45	0.85	-0.76	-0.15	0.59	-0.10	0.83	0.73
V	0.60	0.51	0.62	0.94	0.25	0.95	0.98	-0.12	0.70
Cr	0.62	0.73	0.91	0.96	0.22	0.97	0.92	-0.12	0.98
Cu	0.91	0.12	0.84	0.19	0.86	0.77	0.91	-0.19	0.87
Ni	0.68	-0.52	0.50	0.77	-0.15	0.61	0.81	-0.01	0.87
Mn	0.28	0.61	0.45	0.22	0.91	0.88	0.67	0.55	0.66
Pb	0.72	-0.53	0.79	0.83	-0.45	0.88	0.96	-0.09	0.75
Fe	0.86	0.22	0.80	0.97	0.59	0.94	0.83	0.02	0.94
Cd	0.67	-0.56	0.76	0.41	-0.50	0.41	0.12	0.85	0.69
Zn	0.56	0.44	0.74	0.92	-0.32	0.95	0.89	0.11	0.80
Initial Eigen values	4.78	2.47	4.78	5.68	2.28		6.18	1.80	
Variance (%)	47.8	24.7	47.8	56.84	22.75		61.77	17.98	
Cumulative variance (%)	47.8	72.5	47.8	56.84	79.60		61.77	79.75	

Table 4.1 Interpar component analysis for The concentrations of the KDS, who and h	Table 4.	Principal	component	analysis for	TE concentration	ns of the	e RDS,	WBS and P
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Comm : Communalities

Table 5. The potential ecological risk indices	(RI) for RDS and W	BS from different sampling
sites in Kogarah, Sydney		

	Sampling		Me	ean value of	f Er		DI	Ecological risk
	site	Cr	Pb	Cu	Cd	Zn	- KI	
RDS	1	12.8	5.0	28.6	15.7	3.8	65.8	Low
	2	18.7	5.3	33.8	28.2	2.8	88.8	Low
	3	22.3	12.0	96.3	17.2	5.7	153.5	Moderate
	4	20.5	8.5	19.0	15.7	4.7	68.4	Low
	5	16.4	6.3	24.7	26.6	6.7	80.8	Low
	6	26.0	24.9	140.5	36.0	6.3	233.8	Moderate
	7	17.3	16.6	65.1	29.8	10.1	138.8	Low
	8	16.4	34.1	75.5	40.7	12.0	178.7	Moderate
	9	15.0	29.5	85.9	36.0	9.7	176.1	Moderate
	10	29.2	12.0	119.7	28.2	9.4	198.5	Moderate
	11	16.0	12.9	65.1	31.3	9.3	134.5	Low
	Ave.						138.0	Low-moderate risk
WBS	1	1.9	1.8	2.9	15.7	0.7	22.9	Low
	2	3.6	3.3	6.2	15.7	1.2	30.0	Low
	3	4.2	1.9	41.6	15.7	0.7	64.2	Low
	4	2.2	2.3	5.7	15.7	1.5	27.4	Low
	5	5.9	4.0	7.5	36.0	1.6	55.1	Low
	6	12.8	5.2	28.6	31.3	2.3	80.2	Low
	7	4.4	4.1	6.8	18.8	1.3	35.4	Low
	8	8.2	5.3	9.1	23.5	2.0	48.1	Low
	9	9.1	5.5	12.2	18.8	2.2	47.8	Low
	10	6.4	5.3	7.5	29.8	1.8	50.8	Low
	11	5.9	7.1	8.6	72.1	2.0	95.7	Low
	Ave.						46.6	Low risk

Figure captions

Fig. 1. Map of sampling sites

Fig. 2. Mean TE concentrations in RDS, WBS, and BS samples (vertical lines on top of bars are standard errors)

Fig. 3. Box-plot of pollution indices for RDS (A) and WBS (B) (The line separating the dark and light shaded area inside the box represents the median; the boxes mark the 25th and 75th percentiles; the horizontal line outside the box, the whisker, denotes the maximum and minimum values)

Fig. 4. PCA analysis of TE concentrations in RDS, WBS and BS

Fig. 5. Concentrations of TE fractions in RDS (F1, exchangeable + water and acid soluble; F2, reducible; F3, oxidisable; F4, residual)



Fig. 1



Fig. 2



Fig. 3







Fig. 5