

## Analysis of build-up of heavy metals and volatile organics on urban roads in gold coast, Australia

Parvez Mahbub, Ashantha Goonetilleke, Godwin A. Ayoko, Prasanna Egodawatta and Tan Yigitcanlar

### ABSTRACT

Urban water quality can be significantly impaired by the build-up of pollutants such as heavy metals and volatile organics on urban road surfaces due to vehicular traffic. Any control strategy for the mitigation of traffic related build-up of heavy metals and volatile organic pollutants should be based on the knowledge of their build-up processes. In the study discussed in this paper, the outcomes of a detailed experimental investigation into build-up processes of heavy metals and volatile organics are presented. It was found that traffic parameters such as average daily traffic, volume over capacity ratio and surface texture depth had similar strong correlations with the build-up of heavy metals and volatile organics. Multicriteria decision analyses revealed that the 1–74  $\mu\text{m}$  particulate fraction of total suspended solids (TSS) could be regarded as a surrogate indicator for particulate heavy metals in build-up and this same fraction of total organic carbon could be regarded as a surrogate indicator for particulate volatile organics build-up. In terms of pollutants affinity, TSS was found to be the predominant parameter for particulate heavy metals build-up and total dissolved solids was found to be the predominant parameter for the potential dissolved particulate fraction in heavy metals build-up. It was also found that land use did not play a significant role in the build-up of traffic generated heavy metals and volatile organics.

**Key words** | heavy metals, pollutant build-up, traffic pollutants, urban water quality, volatile organics

**Parvez Mahbub** (corresponding author)

**Ashantha Goonetilleke**

**Prasanna Egodawatta**

**Tan Yigitcanlar**

School of Urban Development, Faculty of Built Environment and Engineering, Queensland University of Technology, Brisbane, Australia  
E-mail: [s.mahbub@qut.edu.au](mailto:s.mahbub@qut.edu.au)

**Godwin A. Ayoko**

School of Physical and Chemical Sciences, Faculty of Science and Technology, Queensland University of Technology, Brisbane, Australia

### INTRODUCTION

Rapid urbanisation is a global phenomenon that is happening as a result of increased demand in urban activities throughout the world. One of the significant impacts of urbanisation is the increase in vehicle usage on urban roads. The scenario of changes in urban traffic due to increased urbanisation can readily affect the pollutant build-up on road surfaces. In this regard, the environmental impacts and more specifically the water quality impacts of road transport and their mitigation strategies have received limited attention (Tomerini & Brown 1998; Brown *et al.* 2004; BITRE 2008). Some proposed models such as Transport Planning Add-on Environmental Modelling System (Brown *et al.* 1998) and Vehicle Contaminant Load Model (Gardiner & Armstrong 2007) have assumed a simplified pollutant accumulation process to describe pollutant inputs into receiving water bodies from road traffic. Wu *et al.* (1998) used long term average pollutant loading rates

to characterise highway pollutant loading. Charlesworth & Lees (1999) studied particulate associated heavy metal pollutants and identified the dominant heavy metal species in the source-transport-deposition cascade. Pollutant accumulation in the urban environment is a complex process and an in-depth understanding of the process of build-up of significant pollutants on the road surface will strengthen the knowledge base leading to improved stormwater quality mitigation strategies.

The major pollutants to water bodies that are generated by transport activities include polycyclic aromatic hydrocarbons, total petroleum hydrocarbons, volatile organics and heavy metals (Hoffman *et al.* 1982, 1984; Sansalone & Buchberger 1997). These have significant human health impacts. This paper investigates the build-up processes of heavy metals and volatile organics generated by urban traffic on road surfaces. The outcome of this study will contribute

to the development of robust mitigation measures of improve urban water quality in terms of vehicle generated heavy metals and volatile organics in build-up on urban road surfaces.

## SITE SELECTION

The research study was undertaken in the Gold Coast region of south-east Queensland, Australia. The study adopted a suburb based approach by selecting ten road sites which represented a combination of residential, commercial and industrial land uses in two suburbs. These two suburbs reflected the transport infrastructures that were developed over the last decade in the Gold Coast region. The selected suburbs were Helensvale and Coomera. The selection of different land uses provided a cross-section of traffic activities on road surfaces in the Gold Coast region. The average daily traffic (ADT) and congestion are two important traffic parameters in terms of characterising urban roads. Congestion on urban roads mainly occurs during peak hours. The volume to capacity (V/C) ratio describes the traffic activities on the stretch of a road during peak hours (Ogden & Taylor 1999). Hence, ADT and V/C were selected as traffic parameters in this study. A number of pavement parameters such as surface texture depth (STD) and lane width were also used to characterise the sample collection sites. A sophisticated transport model called 'ZENITH' (GCCC 2006), which is currently being used by the Gold Coast City Council for their pavement infrastructure planning and design activities, was used to predict the current ADT and V/C ratios for the selected sites. The STD and the lane widths of the roads were measured at the study sites. Figure 1 shows the ground level photo of one of the build-up sites. This was a typical residential site having DG14 grade asphalt with 5.1% aggregate binder.



Figure 1 | Build-up sample collection site.

Table 1 | Traffic and pavement characteristics data of the selected sites

Site name (Labels)	Land Use	ADT <sup>a</sup>	Volume to capacity ratio (V/C) <sup>a</sup>	STD (mm)	Lane width (m)
Abraham Road (1C)	Commercial	13,028	1.11	0.6467	3.5
Reserve Road (2R)	Residential	6,339	0.45	0.7505	3.5
Peanba Park Road (3R)	Residential	581	0.15	0.6844	2.8
Beattie Road (4I)	Industrial	2,670	0.24	0.7074	3.5
Shipper Drive (5I)	Industrial	7,530	0.55	0.6788	3.5
Hope Island Road (6C)	Commercial	7,534	0.57	0.7254	3.4
Lindfield Road (7C)	Commercial	2,312	0.33	0.9417	3.3
Town Centre Drive (8C)	Commercial	24,506	0.62	0.6416	3.5
Dalley Park Drive (9R)	Residential	3,534	0.42	0.8342	2.9
Discovery Drive (10R)	Residential	9,116	0.25	0.6957	2.9

<sup>a</sup>GCCC (2006).

The STD of this site is 0.75 mm. Table 1 shows the selected sites and their corresponding traffic and pavement data.

## BUILD-UP SAMPLE COLLECTION

A sample collection method referred to as 'Wet and Dry Vacuum System' (Mahbub *et al.* 2009) was used in this study. Deionised water was sprayed using a high pressure sprayer on a 2 × 1.5 m plot followed by a thorough vacuuming using a domestic vacuum cleaner. In terms of collecting samples from an actual road surface subject to atmospheric wear and tear as well as daily traffic, this method achieved the same level of efficiency as described in earlier studies performed on synthetic surfaces (Deletic & Orr 2005; Egodawatta 2007). The build-up samples were collected in 25 L plastic containers containing deionised water. Homogeneous 500 mL subsamples were transferred to high density 1 L polyethylene bottles using a churn splitter. The particulate analytes were fractionated into four sizes namely 300, 150–299, 75–149 and 1–74 μm using wet sieving. The filtrate that passed through a 1 μm membrane filter was

considered to contain the potential dissolved analytes. Samples were collected from the road surfaces after a minimum antecedent dry period of 7 days (Egodawatta 2007).

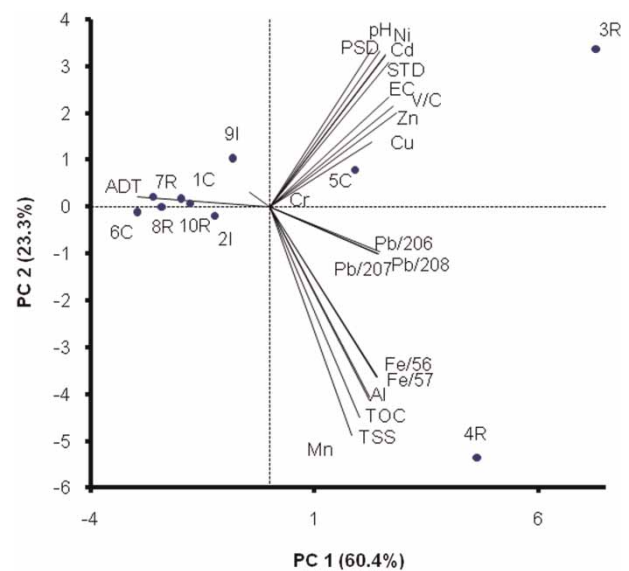
## TEST RESULTS AND DATA ANALYSES

The methods used for sample collection, digestion and determination are covered in USEPA 200.8 (EPA 1994). The methods followed for the determination of volatile range organics were USEPA 5035, 5030B, 8015, 8021 and 8260 (EPA 2008). The road STD was measured according to the recommendations of the US Federal Highway Administration (FHWA 2005).

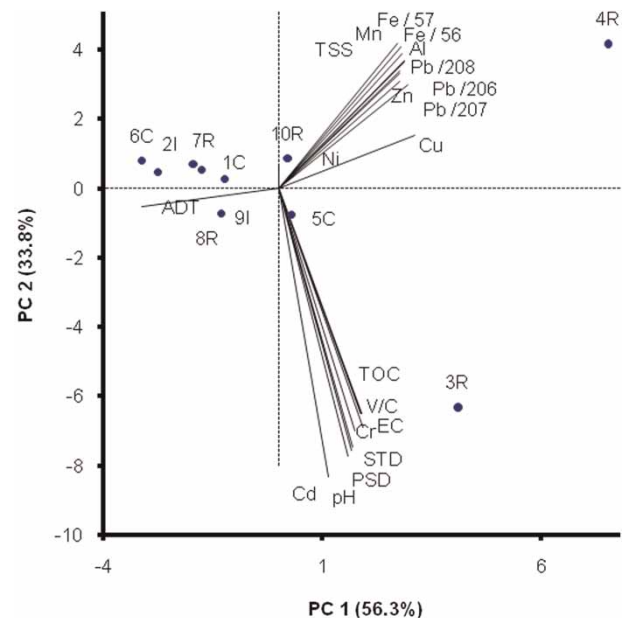
### Heavy metals

The heavy metals selected for this investigation were cadmium (Cd), chromium (Cr), nickel (Ni), lead (Pb), zinc (Zn), copper (Cu), manganese (Mn), aluminium (Al) and iron (Fe). Iron and lead respectively had further two and three species in terms of different isotopes. The selection of heavy metals for analysis was based on a detailed literature review on heavy metal pollution generated by road traffic (for example Drapper *et al.* 2000; Deletic & Orr 2005; Herngren *et al.* 2006). The particulate and potentially dissolved heavy metals were tested in the build-up samples. For quality control, calibration standards, internal standards, blanks and certified reference materials were used. The trace metal detection was performed using inductively coupled plasma/mass spectrometry (ICP/MS). The percentage recovery of the target heavy metals ranged within 85–115%. The relative standard deviations of the repetitive samples were found within 1.5–15% for different heavy metals.

Principal component analysis (PCA) is regarded as an effective tool for pattern recognition (Massart *et al.* 1997) and hence used in this study to identify inherent patterns in the data. All twelve heavy metals along with other parameters including total and dissolved organic carbon (TOC and DOC), particle size distribution (PSD), pH, electrical conductivity (EC), ADT, volume to capacity ratio (V/C), total and dissolved suspended solid (TSS and TDS) and STD were considered as variables. The ten study sites were considered as objects. The pollutant concentrations were expressed in mg/m<sup>2</sup> of the road surface. The PCA biplots in Figures 2 and 3 show the patterns observed in two of the four particulate size fractions investigated.



**Figure 2** | PCA biplot for heavy metals build-up on urban roads at 150–299 μm fractions (objects are represented with numbered labels with suffix C = commercial, I = industrial or R = residential).



**Figure 3** | PCA biplot for heavy metals build-up on urban roads at 1–74 μm fractions (objects are represented with numbered labels with suffix C = commercial, I = industrial or R = residential).

To differentiate the isotopes of iron and lead, corresponding molecular weights are shown alongside the biplots.

In Figure 2, the 150–299 μm particulate fraction shows two groups of variables that are negatively correlated with ADT. In this fraction, TSS has a strong positive correlation with the iron species, manganese, TOC and aluminium in

one group, whilst copper, zinc, nickel and cadmium has strong positive correlation with STD, pH, EC and V/C. This indicates that the iron species, manganese and aluminium could be from sources other than traffic, whilst copper, zinc, nickel and cadmium would be generated from traffic. It is also noticeable that these two groups are nearly perpendicular to each other which indicate that they were independent of each other. The lead species had moderate positive correlation with TOC. Nonetheless, the lead species were significant as they had large loadings in the biplot. In Figure 2, only three objects (3R, 4R and 5C) had positive scores along with positive loadings of all variables except ADT on PC1 whilst the rest of the objects had negative scores along with negative loading of ADT on PC1. None of the objects except 3R and 4R had noteworthy scores on PC2. Similar findings regarding other fractions (e.g. Figure 3) underlined the fact that traffic related heavy metals build-up was not directly influenced by the land use; rather the traffic and pavement characteristics were more directly correlated with the heavy metals build-up.

Initially, the fact that the decrease in ADT was related to the increase of heavy metals build-up on roads appeared to be contradictory. However, after a close examination of V/C and STD, it was found that these traffic parameters were also negatively correlated with ADT. This can be explained by the fact that as the capacity of a lane is fixed, the increase in V/C indicates congestion on the road with low movement of traffic. Hence, it is postulated that the decrease in ADT as noted indicates lane congestion which in turn caused the increase in trace element build-up on the road surface. Also, low traffic movement during congestion could affect the texture of the road surface in a different way than high traffic movement during little or no congestion. Table 2 gives the correlation matrix between target heavy metals

and traffic, pavement and other significant chemical parameters.

In Figure 3, the 1–74  $\mu\text{m}$  fraction also revealed two distinct groups of variables that were again pointing towards different sources. In this fraction, TSS has a strong correlation with copper, zinc, aluminium, manganese, iron and lead species. In the other group, cadmium and chromium has a very strong positive correlation with TOC, V/C, pH, EC, PSD and STD. Nickel has an insignificant loading score in this fraction. There was no impact of land use on traffic related heavy metal build-up detected in this fraction as well.

The PCA of the potential dissolved fraction of heavy metals in the build-up is shown in Figure 4. Unlike the particulate fractions discussed so far, the potential dissolved fraction did not show any particular groups of variables as evident in Figure 4. However, two important similarities with the particulate fractions were still found. These were as follows:

- ADT has negative correlation with all the variables including V/C and STD.
- No impact of land use could be found in the potential dissolved fraction.

In this fraction, zinc, copper, cadmium, chromium and lead species were very strongly correlated with V/C, STD and TDS.

### Volatile organics

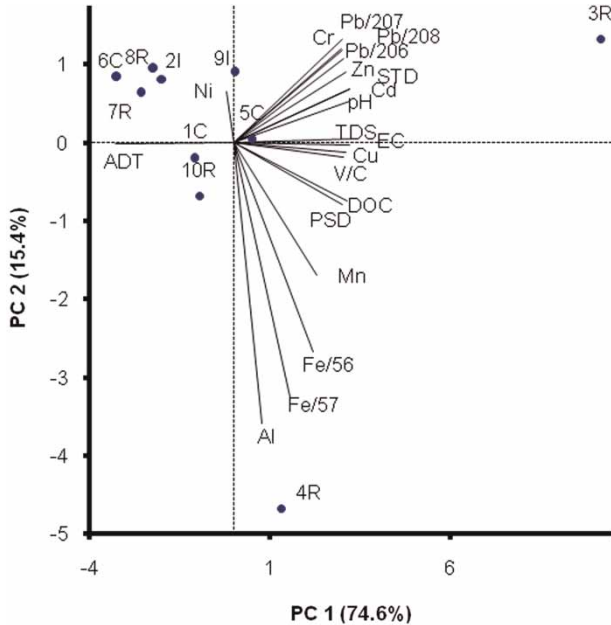
The volatile organics investigated were toluene (TLE), ethylbenzene (ETB), meta and para xylene (MPX) and ortho xylene (OX). A purge and trap system along with gas chromatograph mass spectrometry were used for sample extraction and determination. The lower reporting limit for

**Table 2** | Total correlation matrix between heavy metals and other parameters

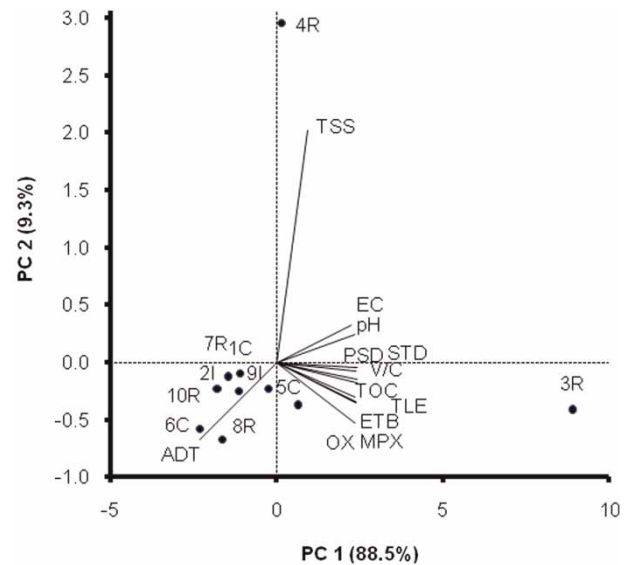
Heavy metals	ADT	V/C	STD	pH	EC	PSD	TSS	TOC
Al	-0.82	0.47	0.39	0.37	0.50	0.36	0.97	0.98
Cr	0.13	-0.04	-0.10	-0.11	-0.04	-0.15	-0.10	-0.08
Mn	-0.63	0.25	0.12	0.10	0.25	0.08	0.99	0.98
Fe/56	-0.82	0.47	0.39	0.37	0.50	0.36	0.97	0.98
Ni	-0.79	0.82	0.94	0.96	0.94	0.99	0.15	0.20
Cu	-0.53	0.76	0.71	0.64	0.53	0.55	0.20	0.31
Zn	-0.82	0.93	0.95	0.91	0.86	0.85	0.32	0.43
Cd	-0.83	0.91	0.99	0.99	0.96	0.99	0.19	0.27
Pb/206	-0.60	0.67	0.55	0.47	0.44	0.38	0.52	0.61

each analyte was 0.001 mg/L. The particulate and potential dissolved fractions were prepared same as for the heavy metals. For quality control, calibration standards, internal

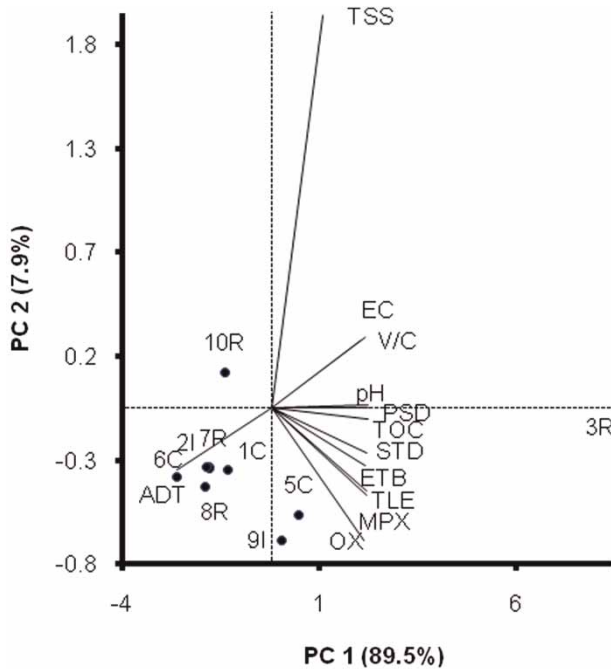
standards and surrogates were used as recommended. Analyses of all particulate and the dissolved fractions of volatile organics revealed that the target volatiles form a group of variables that has very strong positive correlations with TOC for all particulate fractions. The high percentage of carbon in the molecular structures of the volatile organics is attributed to be the reason. This also indicates that the



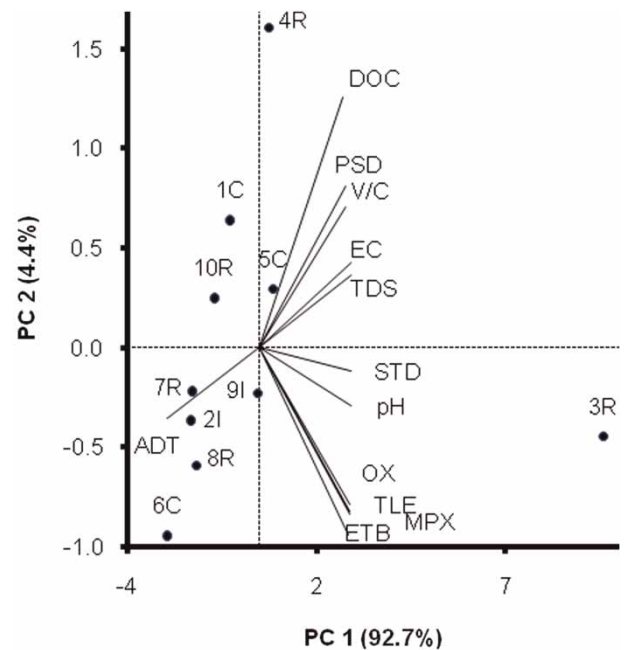
**Figure 4** | PCA biplot for heavy metals build-up on urban roads at  $<1\mu\text{m}$  fractions (objects are represented with numbered labels with suffix C = commercial, I = Industrial or R = residential).



**Figure 6** | PCA biplot for volatile organics build-up on urban roads at 75–149  $\mu\text{m}$  fractions (objects are represented with numbered labels with suffix C = commercial, I = Industrial or R = residential).



**Figure 5** | PCA biplot for volatile organics build-up on urban roads at  $>300\mu\text{m}$  fractions (objects are represented with numbered labels with suffix C = commercial, I = Industrial or R = residential).



**Figure 7** | PCA biplot for volatile organics build-up on urban roads at  $<1\mu\text{m}$  fractions (objects are represented with numbered labels with suffix C = commercial, I = Industrial or R = residential).

target volatiles which are hydrophobic in nature generally inclined towards the organic carbons primarily in particulate form. Figures 5 and 6 show two PCA biplots for the particulate fractions of volatile organics.

Unlike for some heavy metals, it was noted that TSS has no impact on the volatile organic build-up in any particulate fraction. The result for the potential dissolved fraction is shown in Figure 7. The potential dissolved fraction, showed opposite results to the particulate fractions. In Figure 7, the volatile organics have moderately positive correlations with TDS and weak positive correlations with DOC. No impact of land use on traffic related volatile organic build-up was found for any fraction. A summary of the outcomes of the PCA is given in Table 3 below.

Table 3 is a generalised view of a pollutant's affinity during build-up. A closer analysis of Table 3 point to two specific issues that merited further investigation in order to better understand the build-up processes of heavy metals and volatile organics on urban roads. These issues are:

- For heavy metals, which chemical parameter out of TOC, DOC, TSS and TDS is predominant in terms of pollutant's affinity towards them, both in particulate and dissolved form;
- For both heavy metals and volatile organics, which particle fraction is predominant in terms of the pollutant's affinity towards predominant chemical parameters in particulate form.

Table 3 | Affinity of individual pollutants towards different chemical parameters

Principal pollutant category	Individual pollutant	Affinity in Particulate fractions (1-300 µm and higher) in build-up	Affinity in potential dissolved fraction (<1 µm) in build-up
Heavy Metal	Lead (Pb)	-	TDS
	Nickel (Ni)	TOC	-
	Cadmium (Cd)	TOC	DOC
	Chromium (Cr)	TOC	DOC
	Zinc (Zn)	-	TDS
	Copper (Cu)	-	TDS
	Iron (Fe)	TSS, TOC	-
	Aluminium (Al)	TSS, TOC	-
	Manganese (Mn)	TSS, TOC	-
Volatile Organic Carbon	Toluene (TLE)	TOC	TDS
	Ethylbenzene (ETB)	TOC	TDS
	Meta and Para Xylene (MPX)	TOC	TDS
	Ortho Xylene (OX)	TOC	TDS

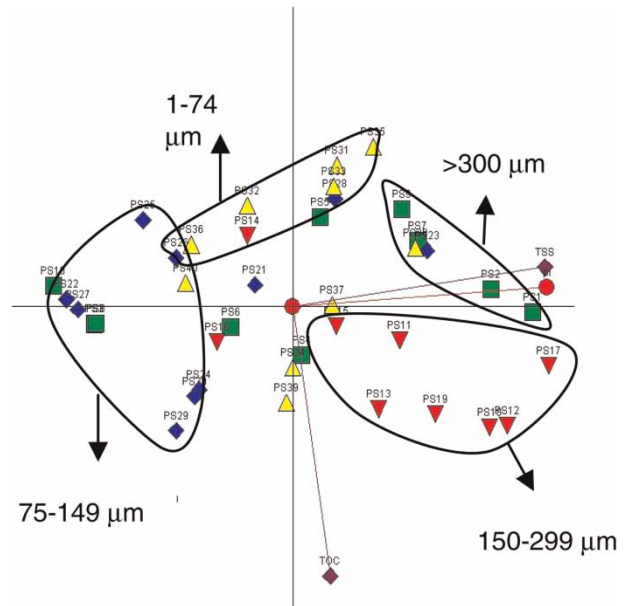


Figure 8 | GAIA biplot for predominant chemical parameter scenario (◆) for particulate heavy metals; (▼) pi-decision axis; (▲) >300 µm fractions; (▼) 150-299 µm fractions; (◆) 75-149 µm fractions; (▲) 1-74 µm fractions.

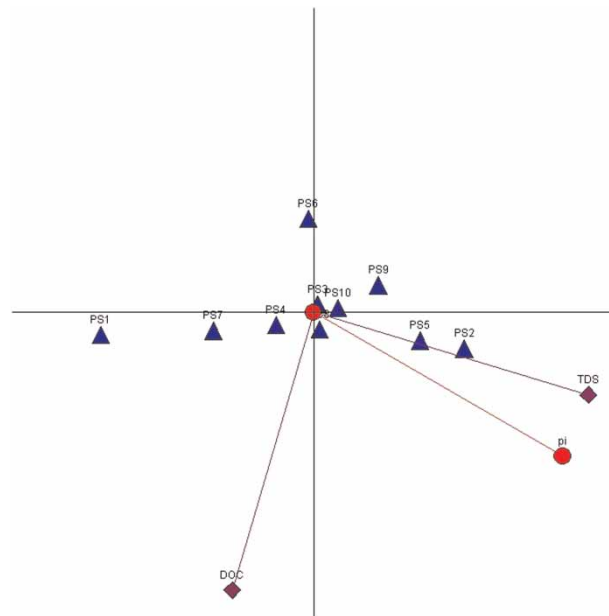


Figure 9 | GAIA biplot for predominant chemical parameter scenario (◆) for the dissolved heavy metals; (●) pi-decision axis; (▲) <1 µm fraction.

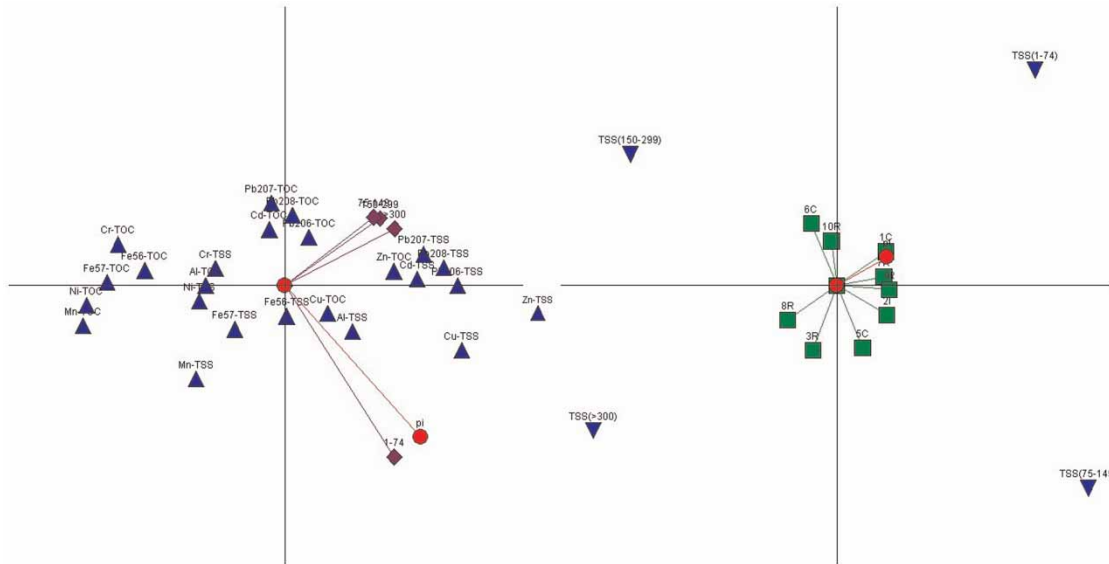
### Multicriteria decision analyses for heavy metals and volatile organics build-up

Multicriteria decision analyses (Keller et al. 1991) incorporating geometrical analysis for interactive aid (GAIA) was used to investigate the two issues highlighted in Table 3. In order

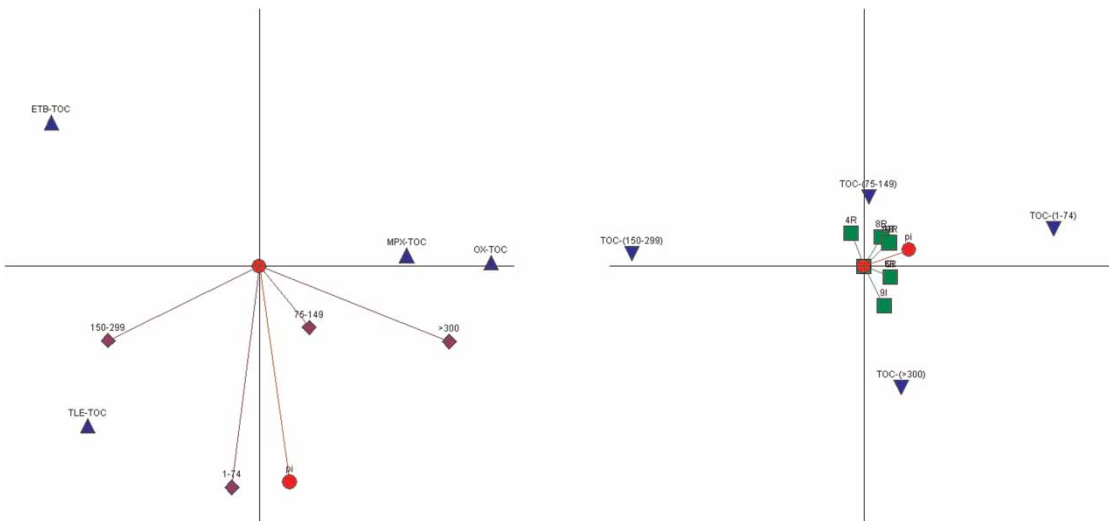
to determine the affinity of heavy metals and volatile organics towards different chemical parameters such as TSS, TDS, TOC and DOC, the concentrations were expressed as loadings (mg of target analytes/mg of predominant chemical parameter). According to Figure 8, for heavy metals the combined particulate fraction from 1 to 300  $\mu\text{m}$  and higher showed that TSS was the predominant parameter rather than TOC and in Figure 9, the combined potential dissolved fraction revealed that TDS as the predominant parameter rather than DOC in terms of their build-up

loadings. The inclination of the pi-decision axis towards a chemical parameter determined its predominance.

The predominant particulate fraction in build-up was also analysed for both heavy metals and volatile organics. Figure 10 shows that the particle size fraction 1 to 74  $\mu\text{m}$  was predominant for heavy metals build-up on the road surfaces. As TSS was primarily represented by the 1–74  $\mu\text{m}$  particulate fraction in Figure 10, TSS can be regarded as a surrogate indicator for particulate heavy metals in build-up. In the case of volatile organics, the particle size fraction



**Figure 10** | GAIA biplot for predominant heavy metals particulate fraction (◆); (●) pi-decision axes; (▲) metals' affinity towards predominant chemical parameters; (■) study sites; (▼) TSS' presence in particulate fractions.



**Figure 11** | GAIA biplot for predominant volatile organics particulate fraction (◆); (●) pi-decision axes; (▲) organics' affinity towards predominant chemical parameter; (■) study sites; (▼) TOC's presence in particulate fractions.

1–74  $\mu\text{m}$  was also the predominant fraction for the volatile organics in build-up as shown in Figure 11. As this study found that TOC was mainly present with this fraction, TOC can be regarded as a surrogate indicator for volatile organics in build-up on urban road surfaces.

Herngren *et al.* (2006) found that 0.45–75  $\mu\text{m}$  range contained the highest heavy metal concentration in road deposited sediments whilst Deletic & Orr (2005) found fractions less than 63  $\mu\text{m}$  had maximum concentration. In this regard, the finding of this study is significant as it has identified the predominant particulate fraction for heavy metals build-up and characterised the affinity of heavy metals in terms of predominant chemical parameters both in particulate and the potential dissolved fraction.

## CONCLUSIONS

This study has undertaken an in-depth investigation into the inherent processes in the build-up of heavy metals and volatile organics on urban roads due to vehicular traffic. It was found that the decrease in ADT was associated with the increase in V/C ratio and STD for both heavy metals and volatile organics in build-up. Hence, the congestion of vehicles in a traffic lane was found to be primarily responsible for the pollutants build-up rather than the vehicle counts during a specified time period. It was also found that land use did not affect the build-up of traffic related heavy metals and volatile organics on urban road surfaces.

Multicriteria decision analyses revealed that total suspended solids (TSS) in the 1–74  $\mu\text{m}$  fraction could be regarded as a surrogate indicator for particulate build-up of heavy metals whilst TOC in the 1–74  $\mu\text{m}$  fraction could be regarded as a surrogate indicator for particulate build-up of volatile organics. TSS was found to be the predominant parameter in particulate heavy metals build-up whilst TDS was found to be the predominant chemical parameter in dissolved heavy metals build-up in terms of pollutants affinity towards them.

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