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Taxonomy of factors which influence heavy metal build-up on urban road surfaces.
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Taxonomy of factors which influence heavy metal build-up on urban road surfaces

An Liu[1]*, Chandima Gunawardana[2], Janaka Gunawardena[3], Prasanna Egodawatta[4]
Godwin A. Ayoko[4], Ashantha Goonetilleke[4]

[1] College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen 518060, People's Republic of China

[2]Department of Civil Engineering, Faculty of Engineering, University of Peradeniya, 20400 Sri Lanka

[3]Logan City Council, PO Box 3226, Logan City DC., Qld 4114, Australia

[4]Science and Engineering Faculty, Queensland University of Technology (QUT), P.O. Box 2434, Brisbane, Qld 4001, Australia

*Corresponding author:

E-mail: liuan@szu.edu.cn; Tel: 86-755-26557315; Fax: 86-755-26536141

Abstract: Heavy metals build-up on urban road surfaces is a complex process and influenced by a diverse range of factors. Although numerous research studies have been conducted in the area of heavy metals build-up, limited research has been undertaken to rank these factors in terms of their influence on the build-up process. This results in limitations in the identification of the most critical factor/s for accurately estimating heavy metal loads and for designing effective stormwater treatment measures. The research study undertook an in-depth analysis of the factors which influence heavy metals build-up based on data generated from a number of different geographical locations around the world. Traffic volume was found to be the highest ranked factor in terms of influencing heavy metals build-up while land use was ranked the second. Proximity to arterial roads, antecedent dry days and road surface roughness has a relatively lower ranking. Furthermore, the study outcomes advances the conceptual understanding of heavy metals build-up based on the finding that with increasing traffic volume, total heavy metal build-up load increases while the variability decreases. The outcomes from this research study are expected to contribute to more accurate estimation of heavy metals build-up loads leading to more effective stormwater treatment design.

Keywords: Heavy metals; Traffic volume; Stormwater quality; Stormwater pollutant processes; Multivariate analysis

1. Introduction

Heavy metals deposited (build-up) on urban roads, which are primarily attached to road dust, is of particular concern in the urban water environment since stormwater runoff transport these pollutants to receiving waters, degrading water quality [1, 2]. Due to their high toxicity [3, 4], accurate estimation of heavy metal loads is essential for the design of effective stormwater treatment strategies.

Heavy metal build-up on urban road surfaces is complex and multifaceted, influenced by a range of factors. These can be categorised as; external factors (such as traffic volume [5], land use [6], distance to arterial roads [7, 8] and road surface roughness [9]), inherent factors (such as heavy metal species [10] and particle size distribution [2, 11]) and climate related factors (antecedent dry days [12]). Past studies have reported on the individual role of these influential factors and their relationship with heavy metals build-up, as evident from the references cited above. However, few research studies have undertaken a comprehensive analysis of these factors and their role in heavy metals build-up. This can be attributed to two primary reasons. Firstly, it is difficult to investigate a wide range of influential factors during an individual research study due to the specific study focus. For example, Gunawardena et.al. [13] investigated the role of traffic volume and land use characteristics on heavy metals build-up while Gunawardena et.al. [14] focused on the adsorption of heavy metals to road deposited solids for different particle sizes. Secondly, it is essential to select appropriate data analysis techniques which have the capability to undertake the requisite investigations as these factors differ in terms of their characteristics, order of magnitude and the degree of influence on heavy metals build-up.

Although previous researchers have reported that the factors which were categorised above as external, inherent and climate related factors, play specific roles in relation to heavy metals build-up on urban road surfaces, they do not exert an equal influence. Consequently, there are obvious benefits in ranking them in order to identify the most critical factors for more accurate estimation of heavy metal build-up loads, for improved stormwater quality modelling, for enhanced interpretation of modelling outcomes and for the effective design of stormwater treatment measures.

It is equally important to understand how the heavy metals build-up vary with the highly ranked influential factor/s (critical factor/s) [15], because the accurate accounting of variability is closely related to the accuracy of interpretation of modelling outcomes. This is primarily related to stormwater quality modelling uncertainty. Among sources of model uncertainty, the variability of input parameters can undermine model performance because lumped parameters are commonly used to represent the entire catchment characteristics without adequately considering their specific characteristics [16]. This is particularly important when specific characteristics are highly ranked in terms of their influence on pollutants processes such as pollutant build-up. Therefore, understanding the variability in heavy metals build-up can help in the formulation of a robust modelling strategy to enable the nature of the variability in heavy metal build-up loads associated with highly ranked factors to be taken into consideration. Additionally, it can also assist in the more accurate interpretation of the modelling results by taking into account the variability of input parameters.

In this context, the research study undertook a comprehensive analysis of the range of factors identified in past research, which influence heavy metals build-up (attached to dust on the road surfaces). These factors included traffic volume, land use, distance to arterial roads, road surface roughness, antecedent dry days and particle size distribution. Data in relation to these factors were obtained from a comprehensive study undertaken by the authors as well as three previous research studies. The data was analysed employing a range of data analysis techniques as appropriate, including stepwise linear regression (SLR), principal component analysis (PCA) and Preference Ranking Organization Method for Enrichment of Evaluations (PROMETHEE). The objectives of the study were: (1) to rank the relevant factors in terms of their influence on heavy metals build-up; and (2) to analyse the variability of heavy metal build-up with the top-ranked factor/s. The study outcomes were validated using data obtained from seven previous research studies. The outcomes from this study will contribute to the accurate estimation heavy metals build-up loads and more effective heavy metal targeted treatment design.

2. Methods and materials

2.1 Study sites

The study sites were located at Gold Coast, Australia, where an arterial highway called Pacific Highway traverses the whole region from the north to the south. The Pacific

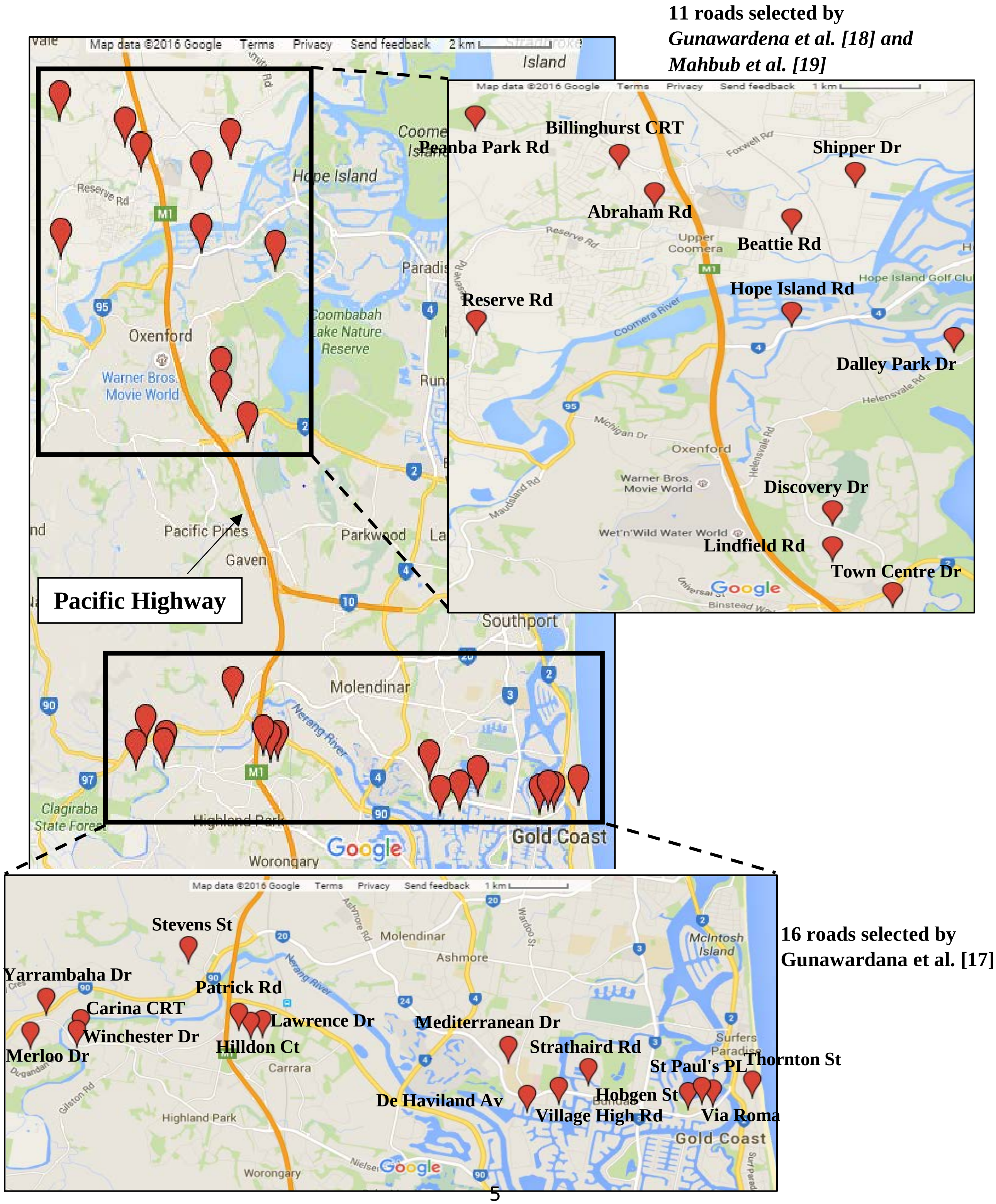
Highway which is 960 km long and links Sydney to Brisbane is a major transport route along the central east coast of Australia. A total of 27 urban road sites were selected close to the highway with different distances to the highway and having differing traffic characteristics, land use and road surface roughness. The data used in the research study were primarily obtained from three previous studies. This included 16 urban road sites selected by Gunawardana et al. [17] while 11 road sites were selected by Gunawardena et al. [18] and Mahbub et al. [19]. The sampling was conducted in two episodes for 16 road sites by [17], where one episode was for shorter antecedent dry days (ADD) and another one was for longer ADD. For the remaining 11 road sites, the ADD was 7 [18, 19]. Accordingly, a total of 43 build-up samples (16 road sites ×2 sampling episodes +11 road sites) were collected. Data related to ADD for each sample are provided in Table S3 in the Supporting Information. The study sites are shown in Figure 1.

2.2 Study approach

The total heavy metal loads and road dust loads per unit area were initially investigated. This was to derive a general understanding of heavy metal build-up loads on the road surfaces. Then, the study was divided into two primary stages. The first stage was to comprehensively analyse key factors and to rank them in terms of their influence on heavy metals build-up. The second stage was to investigate the variability in heavy metals build-up with the top-ranked factor identified in the first stage. As identified in past research literature, the influential factors investigated were average daily traffic volume (DTV), distance to highway (DHW, representing the distance to the closest arterial road), commercial area fraction (C), industrial area fraction (I), residential area fraction (R), antecedent dry days (ADD) and road surface texture depth (STD, representing road surface roughness). The heavy metal species investigated were Cr, Mn, Ni, Cu, Zn, Cd and Pb since these are metal pollutants commonly present in stormwater runoff from traffic areas [8, 11, 13, 20, 21]. Figure 2 illustrates the study approach adopted, including data analysis techniques used and the type of data used in each stage.

The dataset used in this stage was obtained from three recent publications [17-19] and data generated from an independent research study undertaken by the authors. It is noteworthy that although the data were obtained from the three previous publications, the studies were all undertaken at Gold Coast, Australia and sample collection, transport and laboratory testing were carried out using identical methods. This ensured the compatibility of the data sets used.

The dust samples from each road site were collected using a vacuum system for subsequent testing for the heavy metals attached to the road dust. The detailed information regarding road dust build-up sample collection, transport and laboratory testing is provided in the Supporting Information. A total of 43 data points (mean values of duplicate samples) including total heavy metal loads attached to road dust, loads associated with four particle size ranges (<75 µm, 75-150 µm, 150-300 µm and >300 µm) and seven influential factors were obtained. Table S1 in the Supporting Information gives the data availability which formed the basis for the study for this stage and explains how the data were derived (from previous publications or measured by the authors). Table S2 in the Supporting Information gives the collection methods used for obtaining the influential factors. The values for the influential factors (Table S3) and heavy metal loads (Table S4-S9) are provided in the Supporting Information.



11 roads selected by *Gunawardena et al. [18] and Mahbub et al. [19]*

16 roads selected by Gunawardana et al. [17]

Figure 1 Study sites

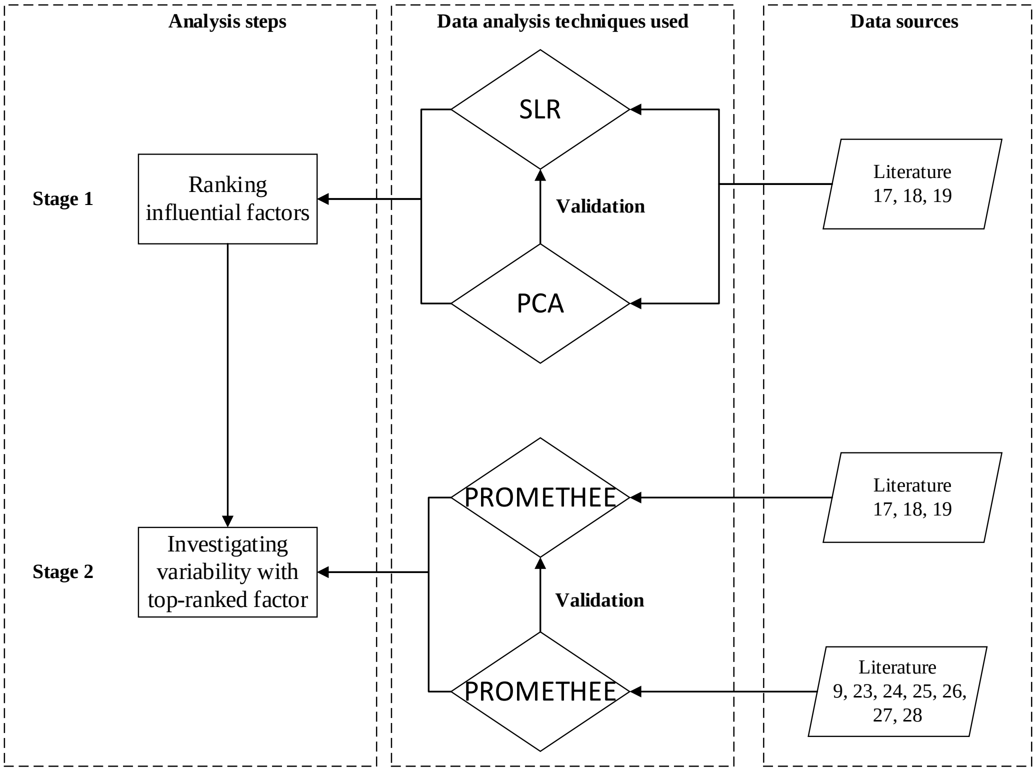


Figure 2 Study approach

(Note: SLR - stepwise linear regression; PCA - principal component analysis; PROMETHEE - Preference Ranking Organization Method for Enrichment of Evaluations)

2.2.1 Stage 1

Stepwise linear regression (SLR) was initially undertaken to determine the important factors according to their influence on heavy metals build-up and subsequently principal component analysis (PCA) was undertaken to validate the results obtained from SLR. SLR is a systematic method for adding and removing independent variables (such as DTV, DHW, C, I, R, ADD and STD in the case of this research study) from a linear or generalised linear model based on their statistical significance in explaining the dependent variable (such as heavy metal build-up loads in the case of this research study). The method commences with an initial model and then compares the explanatory power of incrementally larger and smaller models. MATLAB R2015a was used to undertake SLR. For the SLR analysis, the sum of the seven heavy metal loads for each sample was considered as the dependent variables while DHW, DTV, R, I, C, ADD and STD were the independent variables. This was to initially identify the ranking of influential factors in relation to heavy metals build-up. The MATLAB code used is provided in the Supporting Information.

Principal component analysis (PCA) was used to validate the results of SLR. For PCA, the objects were, the seven heavy metals and the seven influential factors, namely, DHW, DTV, R, I, C, ADD and STD while the variables were the 43 build-up samples.

2.2.2 Stage 2

This stage was conducted using multi criteria decision making method, PROMETHEE, to investigate heavy metals build-up variability with the top-ranked factor identified in the first stage. Detailed information regarding the PROMETHEE method can be found in the literature (e.g. [22]). The corresponding values of the heavy metals associated with the sub-samples representing different particle size ranges were obtained from the previously mentioned publications [17-19] and were included in the analysis. This was to derive a detailed understanding of the variability of heavy metals in road surface build-up. The study outcomes related to heavy metals build-up variability were validated using data from other cities reported in seven previous publications, including two cities in China [9, 23], one city in US [24], Korea [25], Jordan [26], Canada [27] and Malaysia [28], respectively.

3. Results and discussion

3.1 Preliminary investigation of heavy metal loads

Table 1 summarises the data in relation to the total heavy metal load per unit area (mg/m^2) for the 43 samples. Additionally, since the heavy metals are attached to road dust, the total dust loads per unit area (g/m^2) is also given. Values for the total heavy metal loads and road dust loads for each sample are provided in Table S4 in the Supporting Information. It can be noted in Table 1 that the mean value of road dust load per unit area was 3.70 g/m^2 while the standard deviation was 4.92 g/m^2.

Table 1 Total heavy metals and road dust loads

	Mean	SD*	Data range
Cr (mg/m^2)	0.0920	0.172	0.00-0.978
Mn (mg/m^2)	1.40	1.60	0.0350-7.95
Ni (mg/m^2)	0.115	0.158	0.00-0.697
Cu (mg/m^2)	1.40	1.33	0.170-7.67
Zn (mg/m^2)	3.33	4.14	0.282-21.0
Cd (mg/m^2)	0.0170	0.0240	0.00-0.115
Pb (mg/m^2)	0.661	0.802	0.0470-4.30
Road dust (g/m^2)	3.70	4.92	0.0510-28.8

*standard deviation

Total Zn load (mean value was 3.33 mg/m^2 and standard deviation was 4.14 mg/m^2) was the highest among the seven heavy metals while the total Cd load was the lowest (mean value was 0.0170 mg/m^2 and standard deviation was 0.0240 mg/m^2). This means that Zn is the most dominant heavy metal on the road surface while Cd is relatively low in terms of total loads.

As evident in Table 1, the road dust loads collected had a wide range (0.0510-28.8 g/m^2). This could result in heavy metal loads among road sites not being comparable. For example, a high heavy metal load value could just be due to the high load of total road dust load collected. Therefore, in order to compare heavy metal loads attached to road dust among different road sites, total heavy metal load (mg/m^2) for each sample was converted to the load per unit road dust mass (mg/g). Accordingly, the unit of heavy metal load data in the following analysis is mg/g.

3.2 Ranking influential factors

Table 2 shows the SLR outcomes. It was found that DTV was the first factor to be added to the stepwise regression procedure, followed by factors related to land use, namely, I, R and C. The last factor to be added was DHW. However, ADD and STD were not added in the regression procedure. These results imply that average daily traffic volume has the closest relationship with heavy metal build-up on urban road surfaces while land use (I, R, C) is the second most important influential factor, followed by distance to highway. However, antecedent dry days and road surface texture depth play a relatively less important role in influencing heavy metals build-up.

The R^2 of the final linear regression model is 0.748 while p-value is less than 0.0001, which means that the sum of heavy metal loads on each road surface is reasonably predicted by DTV, R, I, C and DHW. This further confirms the important role of traffic volume, land use and distance to highway in heavy metals build-up, compared to antecedent dry days and road surface roughness.

The relationship between influential factors and heavy metal build-up was validated using a PCA biplot (Figure 3). DTV vector forms acute angles with all heavy metal vectors and is projected on the positive PC1 axis same as all the heavy metal vectors. This means that a high traffic volume produces high heavy metal build-up loads. Additionally, all land use vectors are also projected on the positive PC1 axis and form acute angles with some of the heavy metal vectors such as ‘C’ and ‘R –Cd’ and ‘Cr’ and ‘I –Mn’. These observations further confirm that traffic volume is the most important influential factor in heavy metal build-up, followed by land use, while other parameters are less influential since DHW, ADD and STD vectors are positioned at the negative PC1 axis, opposite to the heavy metal vectors. Considering the fact that DHW was added as one of the factors in SLR although it was the last one to be added, the ranking of influential factors in relation to heavy metals build-up is DTV (rank 1), land use (rank 2), DHW (rank 3), ADD and STD (rank 4).

Table 2 Stepwise linear regression (SLR) outcomes

Regression step	Factors added	Regression Coefficients	SE*	p-value**
1	DTV	0.0009	0.0016	0.5817
2	I	19.326	36.986	0.6051
3	R	121.45	19.504	<0.0001
4	C	182.13	62.437	0.0066
5	DHW	0.0169	0.0036	<0.0001
6	DHW×R	-0.0329	0.0066	<0.0001
7	DHW×C	-0.0341	0.0131	0.0140
8	DHW×DTV	-2.1462×10^{-7}	5.5408×10^{-7}	0.7012
9	I×DTV	0.0140	0.0079	0.0853
10	DHW×I	0.0193	0.0146	0.1963
11	C×I	-310.06	226.33	0.1809
12	C×DTV	-0.0057	0.0043	0.1930
Intercept		-62.547	12.232	<0.0001
R^2 of linear regression model		0.7480		<0.0001

*standard error
**significant level

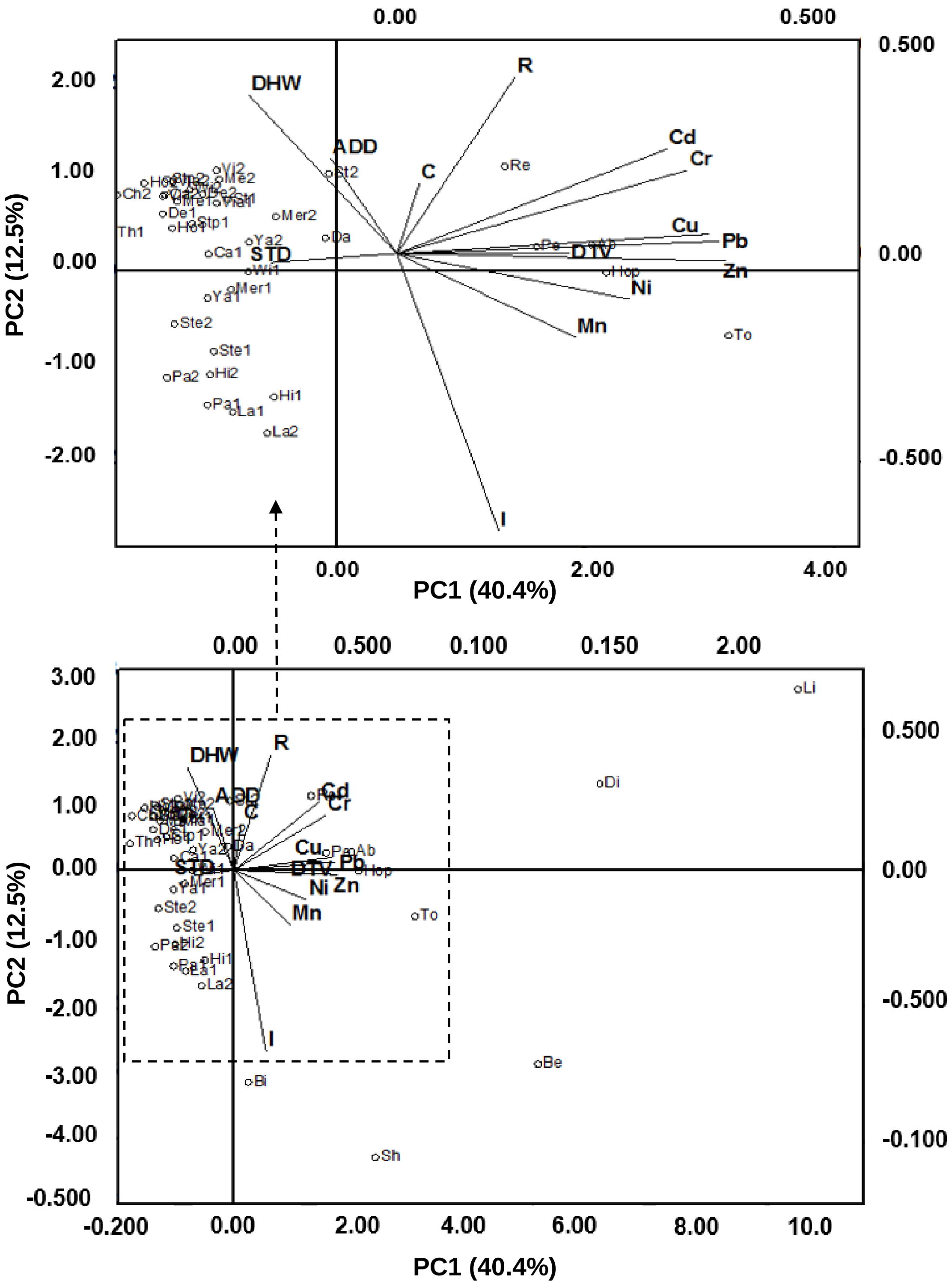


Figure 3 PCA biplot for validating relationship between influential factors and heavy metal build-up

(DHW=distance to highway; C, I and R=commercial, industrial and residential area fractions; DTV=average daily traffic volume; ADD=antecedent dry days and STD=surface texture depth)

According to SLR and PCA results, it was concluded that although traffic, land use, distance to highway, antecedent dry days and surface texture depth significantly influence heavy metal build-up as pointed out by past researchers, traffic volume exerts the strongest influence, compared to other factors while land use is ranked the second most important influential factor. This can be attributed to the fact that urban traffic activities are the primary source of heavy metals. This implies that when locating heavy metal targeted stormwater treatment measures, traffic volume should be the first factor to be considered. This is in agreement with previous research outcomes such as by Huber et al. [21]. Generally, treatment measures should be placed in high traffic volume areas. Secondly, the treatment measures should be located according to the local land use characteristics (the second ranked factor). For example, a treatment system should be located in high-traffic industrial areas rather than a high-traffic residential or commercial area since the factor related to industrial land uses was added in the SLR procedure prior to commercial and residential land use related factors (See Table 2).

3.3 Variability of heavy metals build-up with the top-ranked factor

As average daily traffic volume is the first ranked influential factor in relation to heavy metals build-up, due to reasons discussed above, it was essential to investigate the variability of heavy metals build-up with traffic volume. It is evident from the PCA biplot (Figure 3) that road objects positioned far along the positive PC1 axis are those with high daily traffic volume (To, Li and Di) of greater than 5000 vehicles (average number of vehicle on a daily basis, see Table S3) while roads such as Bi and Sh which have relatively lower loading values on the positive PC1 axis have moderate daily traffic volume, between 1000 and 5000 vehicles (see Table S3). Most of the roads located on the negative PC1 axis have lower daily traffic volume of less than 1000 vehicles (see Table S3). Accordingly, the following data analysis was based on the three traffic volume groups, namely, low traffic (20 samples, DTV<1000), moderate traffic (15 samples, 1000<DTV<5000) and high traffic (8 samples, DTV>5000).

For investigating the variability of heavy metals build-up with traffic volume, the coefficient of variance (CV) rather than the data range was used. This was because the orders of magnitude of heavy metal build-up data for the three traffic groups are quite different. For example, the build-up loads in the high traffic group can be a 1-2 order of magnitude higher than the corresponding values within the low traffic group. For example, Cr build-up in De1 (DTV is 500) was 0.00400 mg/g while the corresponding value was 0.534 mg/g in Li (DTV is 8600) (see Table S3 and Table S5). Similar outcomes can also be found in past research literature. For instance, Al-Khashman [26] found that the Zn build-up load in a high-traffic area was 231 mg/kg, while the corresponding value was only 48.0 mg/kg in a low-traffic area. Therefore, it was important to standardise their data distribution. The standardisation procedure was to divide the standard deviation value by the corresponding mean value, resulting in CV values.

CV values were generated for each heavy metal for the four particle size fractions within each traffic group. Accordingly, 84 CV values (7 heavy metals ×4 particle size fractions × 3 traffic volume groups) were obtained. The original matrix of CV values is provided in Table S10 in the Supporting Information. There were 12 objects (3 traffic volume groups × 4 particle size fractions) and 7 variables (CV values of 7 heavy metals). PROMETHEE method was used for the analysis. For undertaking PROMETHEE, it was required: (1) to assign the ranking sense (maximise/minimise); (2) to choose a preference function from linear, V-shape and usual functions and (3) to weigh each variable according to their importance. In this case, each variable was maximised so that the high CV values could be ranked first. The V-shape preference function was selected since it is commonly used in the stormwater quality research field [22, 29, 30]. Additionally, each variable was equally weighted since the variability of the seven heavy metals was considered to be equally important.

For better interpretation, a GAIA biplot was derived as well as PROMETHEE analysis outcomes. GAIA (Graphical Analysis for Interactive Assistance) is a PC1 vs. PC2 principal component biplot that provides a visual complement to the PROMETHEE ranking. Figure 4 shows the resulting GAIA biplot, while Table 3 gives the PROMETHEE ranking results.

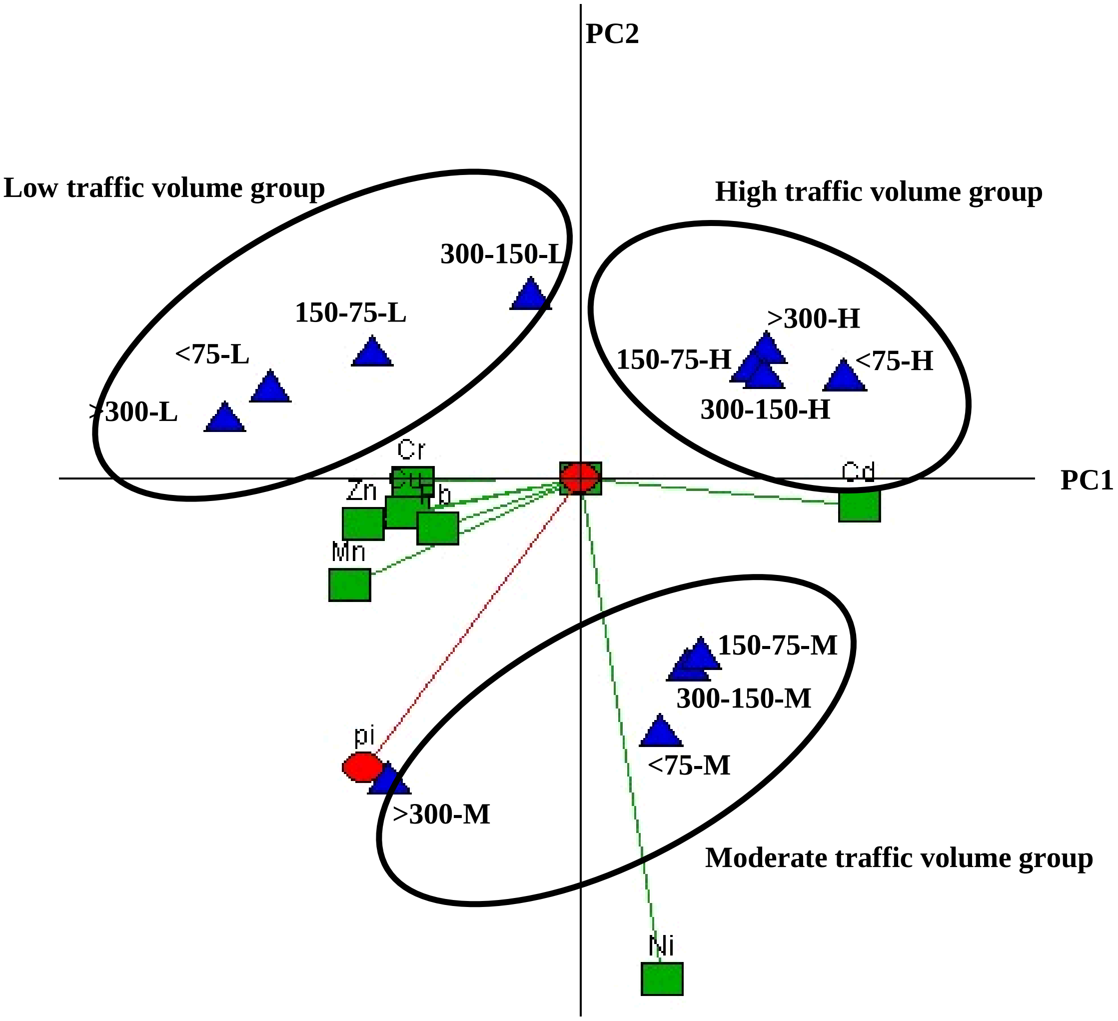


Figure 4 GAIA biplot for CV values for investigating the variability of heavy metals build-up (Δ=68.9%)

(H=high traffic volume group; M=moderate traffic volume group; L=low traffic volume group; each object represents the CV value on the particular particle size within one traffic volume group. For example, <75-M object represents the CV value for heavy metal build-up loads on <75 µm collected from the road sites which are within the moderate traffic volume area)

Table 3 PROMETHEE ranking results

Objects	Φ value	Ranking
>300-M	0.1868	1
>300-L	0.1237	2
<75-M	0.1210	3
<75-L	0.0583	4
300-150-M	0.0339	5
150-75-L	-0.0061	6
>300-H	-0.0345	7
150-75-M	-0.0401	8
300-150-H	-0.0644	9
150-75-H	-0.0659	10
300-150-L	-0.1417	11
<75-H	-0.1709	12

H=High traffic volume; M=Moderate traffic volume; L=Low traffic volume

As shown in Figure 4, the CV values are well clustered based on the three traffic groups rather than particle size fractions. This means that the variability of heavy metal build-up is primarily characterised by traffic volume, overtaking the influence of particle size. Additionally, it can be observed that the Cd vector is closely related to the CV values within the high traffic volume group while Ni is correlated with the moderate traffic volume group. Other heavy metals are related to the low traffic volume group. This implies that different traffic characteristics can lead to different types of variability in relation to different heavy metal species. Cd build-up tends to be highly variable in the road sites in the high traffic volume areas. This means that Cd concentrations in stormwater runoff from high traffic volume areas could be highly variable. This can be also supported by the fact that Cd is a very mobile heavy metal and could be easily transported by runoff, airborne or even splash water [31]. Ni build-up shows higher variation in the road sites in the moderate traffic volume areas. However, other heavy metals are more variable in the low traffic volume areas.

It can be noted in the PROMETHEE ranking results (Table 3) that high traffic volume objects are relatively bottom ranked while moderate and low traffic volume objects are generally ranked at the top. This observation suggests that heavy metals build-up has relatively lower variability with road sites in busy traffic areas, but highly variable in relatively lower traffic areas. In other words, heavy metal build-up loads are relatively similar from site to site within a high traffic volume area, but would be appreciably different in areas with relatively low traffic volume. The possible reason for the PROMETHEE ranking results is that high traffic volume produces high heavy metal loads and hence its influence could overtake other influential factors such as land use and antecedent dry days. However, low traffic volume cannot exert a strong influence on heavy metal build-up as other influential factors will also play a role in producing heavy metal build-up which results in high variability since different road sites have different influential site specific characteristics other than traffic, such as land use, road surface roughness and distance to highway.

These results imply that due to the high variability in heavy metal build-up on road sites, a decentralised stormwater treatment strategy should be adopted in a low traffic area, particularly for heavy metal targeted treatment systems. This is because, the relatively high variation in heavy metals build-up will also be reflected in the resulting stormwater runoff quality and specific treatment systems would be required to be strategically located to treat the stormwater runoff from different areas. However, for a high traffic area such as a busy highway, a centralised treatment system would be adequate due to the relatively low variation in heavy metal concentrations in stormwater runoff. This could also be related to possible savings in land area and cost.

Furthermore, in terms of particle sizes, >300 µm objects are ranked first within each traffic volume group. This means that the heavy metal load on large particles tend to be more variable. This is attributed to the fact that large particles are relatively more influenced by external factors such as road surface roughness, antecedent dry days and land use, as discussed below.

Large particles are less likely to be strongly adhered to road surfaces and hence are easily re-distributed by wind and traffic turbulence [5, 9] Additionally, Egodowatta et al. [32] have noted that the large particle size fraction is highly variable with antecedent dry days, compared to small particles. Longer antecedent dry days produce a relatively higher large particle size fraction. Furthermore, road sweeping primarily removes large particles [33] and different land use areas have different road sweeping frequency. For example, commercial areas are subject to more regular road sweeping compared to industrial areas. Consequently, these facts combine to contribute to the higher variability in heavy metal build-up in large particles.

3.4 Validation of the study outcomes on heavy metal build-up variability

In order to validate the study outcomes relating to the variability of heavy metals build-up with traffic volume, a dataset was compiled comprising of heavy metals build-up and traffic data, which have been reported in seven previous publications listed in Table S11 in the Supporting Information. However, heavy metal build-up load values did not have similar orders of magnitudes and even units (two studies [24, 27] had measured heavy metals build-up as ppm while other studies reported the data as µg/g, mg/g or mg/kg). Furthermore, it was noted that all of the seven publications concluded that higher traffic volume leads to higher heavy metals build-up loads. Unfortunately, none of the studies actually investigated the variability of heavy metals build-up with traffic volume.

Similar to the PROMETHEE analysis discussed above, the CV values were determined for high, moderate and low traffic conditions using the data extracted from the previous publications. Consequently, a CV matrix (38×7) was generated as shown in Table S12 in Supporting Information and the matrix was submitted to PROMETHEE analysis. Figure 5 gives the resulting GAIA biplot.

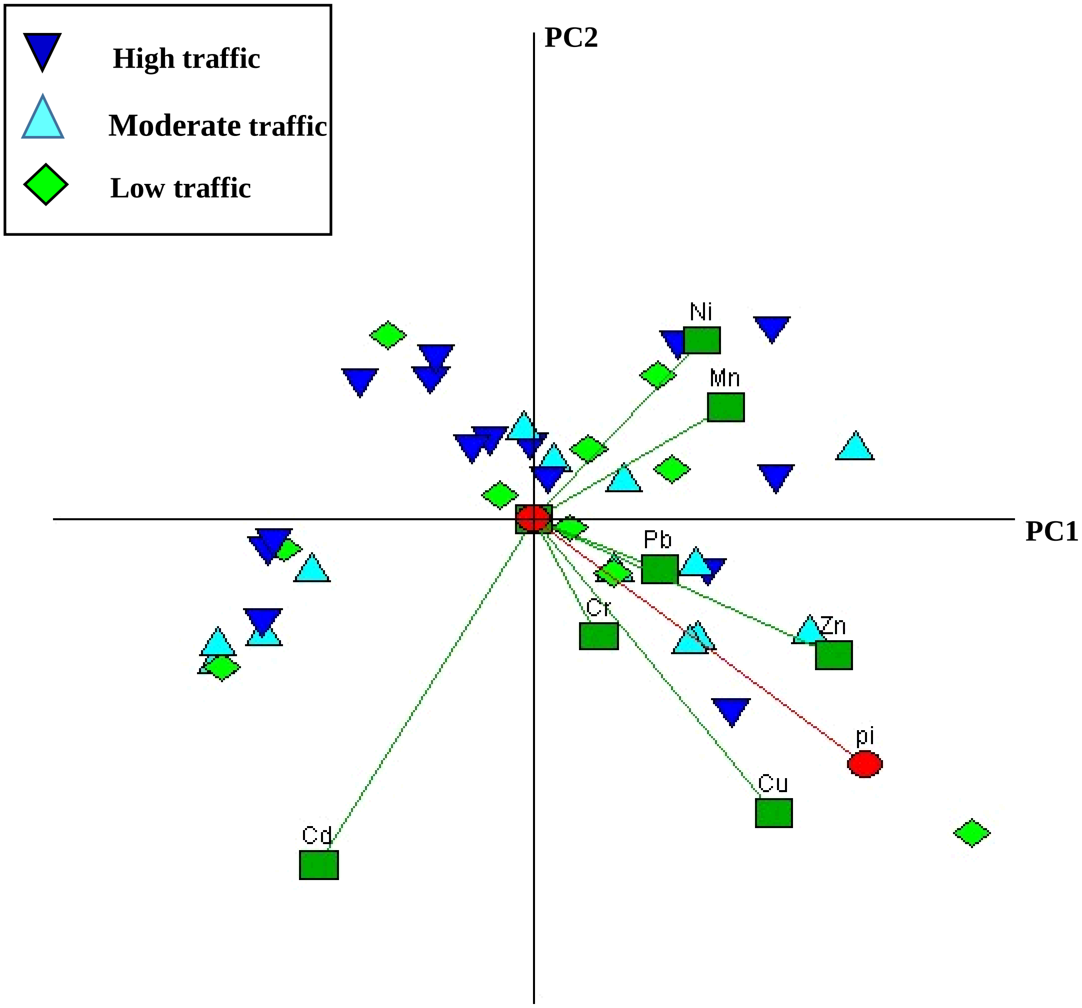


Figure 5 GAIA biplot for validation of heavy metal build-up variability (Δ=71.9%)

As evident in Figure 5, most of the high traffic objects are located opposite to the decision axis, pi, while most of moderate and low traffic objects tend to be in the same direction as the pi axis, which indicates large CV values. It is noteworthy that the pi axis points to low traffic objects. These observations confirm that heavy metals build-up have relatively lower variability in road sites in busy traffic areas while they are highly variable in relatively lower traffic areas. In addition, these results also confirm that heavy metal build-up variability with traffic volume is independent of geographical location and hence is a universal phenomenon.

3.5 Development of conceptual theory on variability of heavy metals build-up

It can be noted from the discussions above that the variability of heavy metals build-up is generally inversely proportional to traffic volume as high traffic areas tend to produce relatively low variability in heavy metals build-up while the low traffic generates high variability. This concept is illustrated by Figure 6. The intercept on heavy metal load axis represents the influence exerted by other parameters, excluding traffic. With increasing traffic volume, total heavy metal build-up loads increase whilst the variability decreases. The high variability in the low traffic volume scenarios represents the influence exerted by other factors other than traffic, while the influence contributed by other factors reduces with increasing traffic volume. The inherent uncertainty which changes with traffic volume affects the accuracy of heavy metal build-up load estimation. This means that the changing uncertainty with traffic volume should be considered when estimating heavy metal build-up loads.

Therefore, a wider uncertainty range should be taken into account when estimating heavy metal build-up loads for a low traffic volume area such as a low-density residential area or natural areas. Furthermore, when undertaking stormwater quality modelling, an appropriate modelling strategy should be adopted to enable the nature of the variability in heavy metal build-up loads associated with traffic characteristics to be taken into consideration. For example, it is recommended that when undertaking stormwater quality modelling, a low-traffic catchment should be disaggregated into a greater number of sub-catchments compared to a high-traffic catchment in order to adequately account for the associated variability. This is to minimise the impact of high variability and hence accurately estimate heavy metal loads in stormwater. For a high traffic area, a reduced disaggregation of sub-catchments or using a lumped heavy metals build-up load value might be acceptable. Additionally, the changing variability with traffic volume also implies that estimating heavy metal build-up loads for a given catchment should be based on the traffic characteristics rather than land use only, which is the conventional practice used in current stormwater modelling approaches [16, 34].

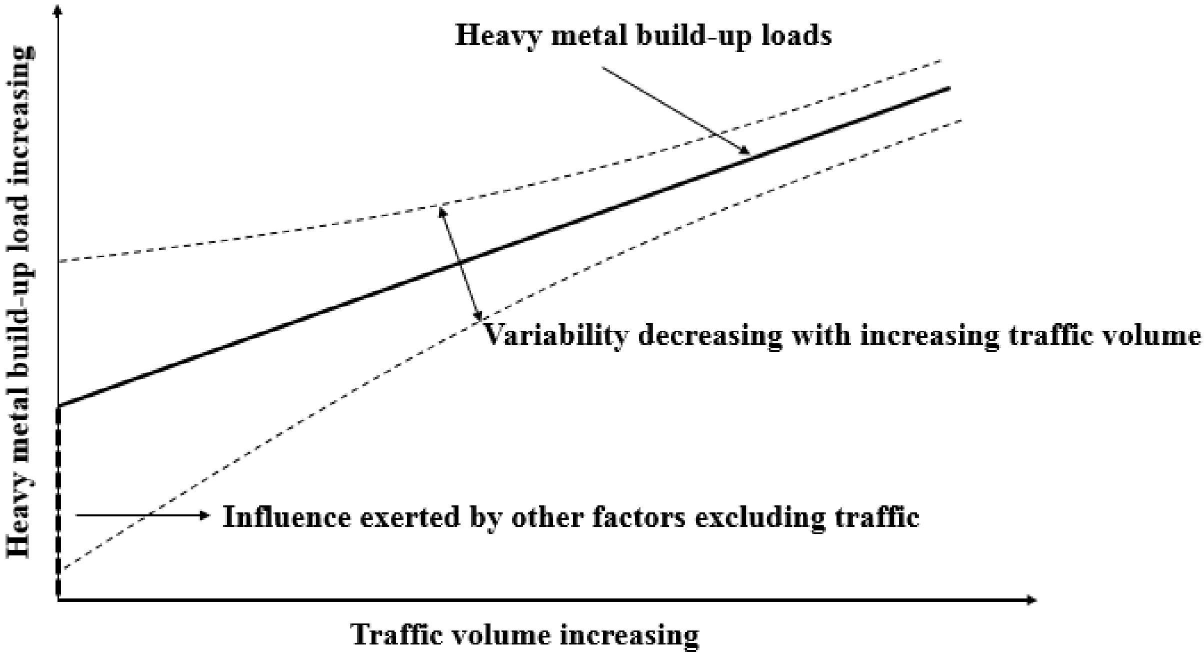


Figure 6 Conceptual illustration of the variability of heavy metals build-up with traffic volume

4. Conclusions

This research study ranked factors in terms of their influence on heavy metals build-up by using data generated from a number of different geographical locations around the world. The results showed that traffic volume was the highest ranked factor while land use was ranked second. Proximity to arterial roads, antecedent dry days and road surface roughness has a relatively lower ranking. Additionally, the study outcomes indicated that heavy metals build-up loads increase with increasing traffic volume while the variability decreases. These results can contribute to enhancing stormwater quality modelling strategies and thereby contribute to effective stormwater treatment design.

Supporting Information

The supporting information provides sample collection, transport and laboratory testing methods adopted, the data collection methods used and the raw data used in the analysis. This includes the data derived in relation to average daily traffic volume, commercial, industrial and residential land use area fractions, antecedent dry days and road surface texture depth, the original heavy metal loads and road dust loads per unit area, heavy metal build-up loads per unit dust mass for the total sample and for the four particle size ranges and the original CV value matrix for the seven heavy metal species for the four particle size fractions investigated for each traffic volume category. Additionally, the Matlab code and results from the stepwise linear regression analysis undertaken is provided. Furthermore, the data extracted from previous publications and used in the current research study is also provided.

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Supporting Information

Taxonomy of factors which influence heavy metal build-up on urban road surfaces

An Liu[1]*, Chandima Gunawardana[2], Janaka Gunawardana[3], Prasanna Egodawatta[4]
Godwin A. Ayoko[4], Ashantha Goonetilleke[4]

[1] College of Chemistry and Environmental Engineering, Shenzhen University, Shenzhen 518060, People's Republic of China

[2]Department of Civil Engineering, Faculty of Engineering, University of Peradeniya, 20400 Sri Lanka

[3]Logan City Council, PO Box 3226, Logan City DC., Qld 4114, Australia

[4]Science and Engineering Faculty, Queensland University of Technology (QUT), P.O. Box 2434, Brisbane, Qld 4001, Australia

*Corresponding author:

E-mail: liuan@szu.edu.cn; Tel: 86-755-26557315; Fax: 86-755-26536141

Sample collection, transport and laboratory testing

Collection of build-up samples and derivation of influential factor values

Road dust build-up sample on each road surface was collected using a dry and wet vacuuming system (with 90% collection efficiency) from 3 m^2 plots from each road surface as described by [1-3]. Before using the vacuum system, all component parts including the water compartment, hoses and foot were cleaned with deionised water. 3L of deionised water was poured into the water compartment as the filtration medium. A deionised water sample was taken as a field blank. The process of dry sample collection was to vacuum the surface three times in perpendicular directions (see Figure S1a). The dry sample collection was to collect the particulate pollutants. Before wet sample collection, the surface was dampened with the sprayer without creating any wash-off (see Figure S1b). The wet sample collection was undertaken to collect any remaining pollutants on the road surfaces. The same vacuuming procedure as for the dry sample collection was applied. As the last step, the water compartment, hoses and brush were washed with deionised water thoroughly in order to minimise the loss of particulate pollutants. The collected water sample was transferred into a polyethylene container.

a: Dry sample collection

b: Sprayer without creating any wash-off

Figure S1 Build-up sampling procedure

The sampling was conducted in two episodes for 16 road sites in [1], where one episode was for shorter antecedent dry days (ADD) and another one was for longer ADD. For the remaining 11 road sites, the ADD was 7 [2, 3]. Accordingly, a total of 43 build-up samples (16 road sites ×2 sampling episodes +11 road sites) were collected. The other influential factors were derived for each road site from available information or were directly measured by the authors, including average daily traffic volume (DTV), residential area fraction (R), commercial area fraction (C), industrial area fraction (I), distance to highway (DTW, representing the distance to the closest arterial road) and road surface texture depth (STD, representing road surface roughness).

Handling and treatment of samples

Each sample was labelled with the street name and collection date. Additionally, deionised water blanks and field water blanks were included to maintain standard quality control procedures as stipulated in Australia/ New Zealand Standards, Water Quality-Sampling [4]. All collected samples were transported to the laboratory on the same day. According to Standard Methods for Water and Waste Water [5], samples were preserved by adding preservatives (sulfuric acid) and placing them in refrigeration at a temperature of 4 ^{0}C.

Laboratory testing

The collected build-up samples were separated into four particle size ranges, namely <75 µm, 75-150 µm, 150-300 µm and >300 µm, by wet sieving. Total samples and sub-samples were tested for Cr, Mn, Ni, Cu, Zn, Cd and Pb since these heavy metals are common stormwater pollutants such as traffic area stormwater runoff pollutants as identified in research literature [6-10]. Samples were digested using nitric acid (HNO_3) according to Standard Method 3030E [5]. Inductively Couple Plasma-Mass spectrometer (ICP-MS) was used to test heavy metals. Single element metal solutions of Scandium, Bismuth, Indium, Terbium and Yrrrium of 100 g/mL (2% HNO_3) prepared by Accustandard® were used to prepare the internal standards. The traceable certified reference material was prepared from multi-element standard solution from TraceCERT (Sigma-Aldrich®). After each batch of 10 samples, a blank was analysed to ensure that no residue was carried over from the previous samples. Precision and accuracy of the heavy metals analyses and the digestion procedure were monitored using the laboratory fortified blanks prepared by adding an aliquot of certified reference material. The percentage recovery ranged from 85% to 115%, which is within the specified limits.

Table S1 Details of the datasets used in the research study

Dataset sources	Reference*	Sample no.	Heavy metals		Influential factors						
			Total (mg/g)	Sub-samples (4 particle sizes, mg/g)	DHW (m)	C (%)	I (%)	R (%)	DTV (/day)	ADD (day)	STD (mm)
Study 1	[19]	11 samples									√
Study 2	[18]		√	√	×	√	√	√	√	√	
Study 3	[17]	32 samples	√	√	×	×	×	×	×	√	√

√=data directly from the publications or calculated using data in the publications; ×= no data provided in the publications and values were measured by the authors; DHW=distance to highway, the 43 samples in the three studies were in the vicinity of a highway called "Pacific Highway", which is 960 km long, links Sydney to Brisbane and is a major transport route along the central east coast of Australia; C, I and R=commercial, industrial and residential area fractions; DTV=average daily traffic volume; ADD=antecedent dry days; STD=surface texture depth

*The reference numbers correspond to the reference list in the main manuscript

Table S2 Collection methods of influential factors

Influential factors	**Collection methods**
Average daily traffic volume (DTV)	Obtained from a traffic survey. The traffic survey was carried out using automatic traffic counters and was conducted at the selected road sites covering two weekdays and one weekend day.
Residential area fraction (R)	C, I and R were derived by dividing the commercial, industrial and residential area fractions with the total area within 1km of each road site. This was undertaken by initially demarcating the area in an aerial photograph from Google Earth and then calculating each land use area fraction using ArcMap.
Commercial area fraction (C)	
Industrial area fraction (I)	
Road surface texture depth (STD, mm)	Collected using Sand Patch Method, according to the method specified by the US Federal Highway Administration [11]
Distance to highway (m)	Perpendicular distance to the highway, obtained by measuring from Google Map

Table S3 Data on influential factors

Road	ID	DHW (m)	C	I	R	DTV	ADD (days)	STD (mm)
Peanba Park Rd	Pe	2.54×10^{3}	0.0700	0.00	0.930	0.0300×10^{3}	7	0.680
Carina CRT	Ca1*	2.47×10^{3}	0.00	0.00	0.620	0.500×10^{3}	8	0.920
Carina CRT	Ca2	2.47×10^{3}	0.00	0.00	0.620	0.500×10^{3}	17	0.920
De Haviland Av	De1	4.29×10^{3}	0.190	0.00	0.520	0.500×10^{3}	8	0.900
De Haviland Av	De2	4.29×10^{3}	0.190	0.00	0.520	0.500×10^{3}	14	0.900
Merloo Dr	Mer1	3.30×10^{3}	0.00	0.00	0.320	0.750×10^{3}	8	0.760
Winchester Dr	Wi1	2.60×10^{3}	0.00	0.00	0.630	0.750×10^{3}	8	0.870
Merloo Dr	Mer2	3.30×10^{3}	0.00	0.00	0.320	0.750×10^{3}	17	0.760
Winchester Dr	Wi2	2.60×10^{3}	0.00	0.00	0.630	0.750×10^{3}	17	0.870
Patrick Rd	Pa1	0.193×10^{3}	0.140	0.190	0.310	0.750×10^{3}	5	1.14
Patrick Rd	Pa2	0.193×10^{3}	0.140	0.190	0.310	0.750×10^{3}	9	1.14
Mediterranean Dr	Me1	4.36×10^{3}	0.180	0.00	0.580	0.750×10^{3}	8	0.820
Village High Rd	Vi1	4.89×10^{3}	0.290	0.00	0.510	0.750×10^{3}	8	0.910
Mediterranean Dr	Me2	4.36×10^{3}	0.180	0.00	0.580	0.750×10^{3}	14	0.820
Village High Rd	Vi2	4.89×10^{3}	0.290	0.00	0.510	0.750×10^{3}	14	0.910
Hobgen St	Ho1	6.93×10^{3}	0.110	0.00	0.450	0.750×10^{3}	4	0.900
St Paul's PL	Stp1	7.19×10^{3}	0.180	0.00	0.400	0.750×10^{3}	4	0.630
Hobgen St	Ho2	6.93×10^{3}	0.110	0.00	0.450	0.750×10^{3}	10	0.900
St Paul's PL	Stp2	7.19×10^{3}	0.180	0.00	0.400	0.750×10^{3}	10	0.630
Dalley Park Dr	Da	2.90×10^{3}	0.0100	0.00	0.990	0.990×10^{3}	7	0.830
Billinghurst CRT	Bi	0.608×10^{3}	0.0700	0.640	0.290	$1.\ 96\times10^{3}$	7	0.700
Shipper Dr	Sh	2.06×10^{3}	0.0900	0.830	0.0800	2.24×10^{3}	7	0.680
Yarrambaha Dr	Ya1	3.50×10^{3}	0.00	0.00	0.340	3.00×10^{3}	8	0.840
Yarrambaha Dr	Ya2	3.50×10^{3}	0.00	0.00	0.340	3.00×10^{3}	17	0.840
Strathaird Rd	St1	5.43×10^{3}	0.500	0.00	0.340	3.00×10^{3}	8	0.800
Strathaird Rd	St2	5.43×10^{3}	0.500	0.00	0.340	3.00×10^{3}	14	0.800
Via Roma	Via1	7.33×10^{3}	0.160	0.00	0.420	3.00×10^{3}	4	0.850
Thornton St	Th1	8.05×10^{3}	0.290	0.00	0.210	3.00×10^{3}	4	1.11
Via Roma	Via2	7.33×10^{3}	0.160	0.00	0.420	3.00×10^{3}	10	0.850
Thornton St	Th2	8.05×10^{3}	0.290	0.00	0.210	3.00×10^{3}	10	1.11
Stevens St	Ste1	0.763×10^{3}	0.190	0.0200	0.240	3.50×10^{3}	5	1.10
Hilldon Ct	Hi1	0.405×10^{3}	0.140	0.200	0.310	3.50×10^{3}	5	0.930
Stevens St	Ste2	0.763×10^{3}	0.190	0.0200	0.240	3.50×10^{3}	9	1.10
Hilldon Ct	Hi2	0.405×10^{3}	0.140	0.200	0.310	3.50×10^{3}	9	0.930
Beattie Rd	Be	1.11×10^{3}	0.0700	0.640	0.290	4.63×10^{3}	7	0.710
Town Centre Dr	To	0.597×10^{3}	0.450	0.240	0.310	5.93×10^{3}	7	0.640
Lawrence Dr	La1	0.615×10^{3}	0.0600	0.170	0.220	7.00×10^{3}	5	1.06
Lawrence Dr	La2	0.615×10^{3}	0.0600	0.170	0.220	7.00×10^{3}	9	1.06
Lindfield Rd	Li	0.535×10^{3}	0.260	0.0100	0.730	8.60×10^{3}	7	0.940
Abraham Rd	Ab	0.379×10^{3}	0.300	0.0400	0.65	8.74×10^{3}	7	0.650
Reserve Rd	Re	3.03×10^{3}	0.0400	0.0200	0.94	1.00×10^{4}	7	0.750
Discovery Dr	Di	0.907×10^{3}	0.0300	0.00	0.97	1.07×10^{4}	7	0.700
Hope Island Rd	Hop	2.21×10^{3}	0.293	0.0300	0.67	2.56×10^{4}	7	0.730

*numerals represent the two sampling episodes for 16 roads; 1 indicates the sampling episode of short antecedent dry days while 2 indicates the episode of long antecedent dry days; DHW=distance to highway; C, I and R=commercial, industrial and residential area fractions; DTV=average daily traffic volume; ADD=antecedent dry days; STD=road surface texture depth

Table S4 Total heavy metal (mg/m^2) and road dust (g/m^2) loads per unit area

Sample ID	Cr	Mn	Ni	Cu	Zn	Cd	Pb	Dust
Pe	0.00	0.106	0.00	0.750	1.07	0.00	0.125	0.0570
Ca1	0.0580	1.22	0.0450	0.504	1.25	0.0110	0.186	2.65
Ca2	0.0270	0.540	0.0570	0.810	1.44	0.0120	0.210	3.00
De1	0.00900	0.213	0.00900	0.170	0.490	0.00900	0.149	2.13
De2	0.0720	1.59	0.0870	1.17	4.58	0.0100	0.872	2.49
Mer1	0.00100	0.223	0.00900	0.227	0.500	0.00300	0.0470	0.360
Wi1	0.0610	4.28	0.143	0.938	4.37	0.0160	0.449	4.08
Mer2	0.271	0.865	0.0590	1.40	2.32	0.00500	0.144	1.31
Wi2	0.0530	0.435	0.0200	0.390	0.600	0.00600	0.210	1.50
Pa1	0.204	2.82	0.697	3.60	3.99	0.0310	1.18	7.83
Pa2	0.115	4.32	0.202	1.15	7.49	0.115	1.44	28.8
Me1	0.0260	0.529	0.0260	0.397	0.930	0.0260	0.331	6.61
Vi1	0.0120	0.334	0.0210	0.365	1.70	0.0120	0.365	3.04
Me2	0.0220	0.392	0.0460	0.700	1.31	0.00300	0.210	0.700
Vi2	0.0410	0.865	0.0580	1.21	1.95	0.00800	0.786	1.31
Ho1	0.0560	0.917	0.0330	0.682	1.60	0.00900	0.306	2.35
Stp1	0.0110	0.990	0.0360	0.990	1.40	0.0110	0.248	2.75
Ho2	0.0460	0.179	0.0560	0.282	0.280	0.00500	0.0510	1.28
Stp2	0.0500	0.371	0.0420	0.398	0.500	0.0110	0.159	2.65
Da	0.0290	1.90	0.0550	1.18	1.77	0.00	0.693	1.10
Bi	0.00800	0.400	0.0120	1.43	3.51	0.00	0.421	0.469
Sh	0.0140	0.398	0.0160	2.45	3.17	0.00	0.829	0.186
Ya1	0.0150	1.96	0.0540	0.572	2.62	0.0120	0.361	3.01
Ya2	0.0610	0.640	0.0450	0.607	1.16	0.00300	0.145	0.660
St1	0.00700	0.894	0.0740	0.963	3.49	0.0120	0.516	1.72
St2	0.978	7.95	0.689	4.24	21.0	0.0240	4.30	5.89
Via1	0.109	3.44	0.227	2.45	4.08	0.109	1.18	9.06
Th1	0.0390	1.47	0.0540	2.17	2.94	0.0310	1.08	7.74
Via2	0.0160	1.20	0.0720	3.20	3.00	0.0240	0.920	4.00
Th2	0.152	1.14	0.130	2.76	3.04	0.0220	2.55	5.42
Ste1	0.213	3.66	0.153	1.46	6.19	0.0270	0.599	6.65
Hi1	0.609	4.10	0.273	7.67	19.9	0.0420	2.84	10.5
Ste2	0.0450	3.16	0.0900	0.904	8.14	0.0450	0.791	11.3
Hi2	0.0320	1.05	0.0410	0.973	3.00	0.0320	0.568	8.11
Be	0.0110	0.0700	0.279	0.900	1.58	0.00100	0.320	0.0620
To	0.0110	0.0350	0.0710	0.773	1.41	0.00100	0.334	0.0660
La1	0.156	1.40	0.0680	2.08	2.81	0.0210	0.520	5.20
La2	0.00800	2.68	0.0130	0.665	0.910	0.00800	0.171	1.900
Li	0.027	0.0550	0.0430	2.16	2.09	0.00300	0.637	0.051
Ab	0.024	0.265	0.0910	1.02	2.33	0.00300	0.330	0.212
Re	0.195	0.432	0.492	1.57	2.56	0.00200	0.162	0.644
Di	0.022	0.0720	0.212	0.834	2.22	0.00200	0.417	0.067
Hop	0.027	0.482	0.0490	1.19	2.47	0.00	0.303	0.252

Table S5 Total heavy metal build-up loads (mg/g)

Sample ID	Cr	Mn	Ni	Cu	Zn	Cd	Pb	Sum
Pe	0.00	1.85	0.00	13.1	18.7	0.00	2.19	35.8
Ca1	0.0220	0.460	0.0170	0.190	0.470	0.00400	0.0700	1.23
Ca2	0.00900	0.180	0.0190	0.270	0.480	0.00400	0.0700	1.03
De1	0.00400	0.100	0.00400	0.0800	0.230	0.00400	0.0700	0.492
De2	0.0290	0.640	0.0350	0.470	1.84	0.00400	0.350	3.37
Mer1	0.00400	0.620	0.0250	0.630	1.38	0.00900	0.130	2.80
Wi1	0.0150	1.05	0.0350	0.230	1.07	0.00400	0.110	2.51
Mer2	0.207	0.660	0.0450	1.07	1.77	0.00400	0.110	3.87
Wi2	0.0350	0.290	0.0130	0.260	0.400	0.00400	0.140	1.14
Pa1	0.0260	0.360	0.0890	0.460	0.510	0.00400	0.150	1.60
Pa2	0.00400	0.150	0.00700	0.0400	0.260	0.00400	0.0500	0.515
Me1	0.00400	0.0800	0.00400	0.0600	0.140	0.00400	0.0500	0.342
Vi1	0.00400	0.110	0.00700	0.120	0.560	0.00400	0.120	0.925
Me2	0.0310	0.560	0.0650	1.00	1.87	0.00400	0.300	3.83
Vi2	0.0310	0.660	0.0440	0.920	1.49	0.00600	0.600	3.75
Ho1	0.0240	0.390	0.0140	0.290	0.680	0.00400	0.130	1.53
Stp1	0.00400	0.360	0.0130	0.360	0.510	0.00400	0.0900	1.34
Ho2	0.0360	0.140	0.0440	0.220	0.220	0.00400	0.0400	0.704
Stp2	0.0190	0.140	0.0160	0.150	0.190	0.00400	0.0600	0.579
Da	0.0260	1.73	0.0500	1.08	1.61	0.00	0.631	5.12
Bi	0.0180	0.852	0.0260	3.05	7.49	0.00	0.897	12.3
Sh	0.0740	2.14	0.0840	13.1	17.0	0.00	4.45	36.9
Ya1	0.00500	0.650	0.0180	0.190	0.870	0.00400	0.120	1.86
Ya2	0.0930	0.970	0.0680	0.920	1.76	0.00400	0.220	4.04
St1	0.00400	0.520	0.0430	0.560	2.03	0.00700	0.300	3.46
St2	0.166	1.35	0.117	0.720	3.56	0.00400	0.730	6.65
Via1	0.0120	0.380	0.0250	0.270	0.450	0.0120	0.130	1.28
Th1	0.00500	0.190	0.00700	0.280	0.380	0.00400	0.140	1.01
Via2	0.00400	0.300	0.0180	0.800	0.750	0.00600	0.230	2.11
Th2	0.0280	0.210	0.0240	0.510	0.560	0.00400	0.470	1.81
Ste1	0.0320	0.550	0.0230	0.220	0.930	0.00400	0.0900	1.85
Hi1	0.0580	0.390	0.0260	0.730	1.89	0.00400	0.270	3.37
Ste2	0.00400	0.280	0.00800	0.0800	0.720	0.00400	0.0700	1.17
Hi2	0.00400	0.130	0.00500	0.120	0.370	0.00400	0.0700	0.703
Be	0.180	1.13	4.50	14.5	25.6	0.0180	5.17	51.1
To	0.174	0.535	1.07	11.7	21.3	0.00800	5.07	39.9
La1	0.030	0.270	0.0130	0.400	0.540	0.00400	0.100	1.36
La2	0.004	1.41	0.00700	0.350	0.480	0.00400	0.0900	2.35
Li	0.534	1.07	0.829	42.2	40.7	0.0570	12.4	97.8
Ab	0.113	1.25	0.430	4.81	11.0	0.0150	1.56	19.2
Re	0.302	0.670	0.763	2.43	3.98	0.00300	0.251	8.40
Di	0.323	1.07	3.15	12.4	33.1	0.0280	6.21	56.3
Hop	0.109	1.91	0.194	4.72	9.79	0.00	1.20	17.9

Table S6 Heavy metal build-up loads in >300 µm solids range (mg/g)

Sample ID	Cr	Mn	Ni	Cu	Zn	Cd	Pb
Pe	0.00	0.0470	0.00	0.741	1.68	0.00	1.21
Ca1	0.00100	0.0800	0.00200	0.0300	0.0500	0.00100	0.0100
Ca2	0.00600	0.0500	0.00900	0.0600	0.300	0.00100	0.0200
De1	0.00100	0.0300	0.00100	0.0200	0.0100	0.00100	0.0100
De2	0.00100	0.0300	0.00200	0.0400	0.180	0.00100	0.0200
Mer1	0.00100	0.0600	0.00600	0.100	0.120	0.00200	0.0200
Wi1	0.00100	0.120	0.00900	0.0200	0.0900	0.00100	0.0100
Mer2	0.0510	0.0800	0.0120	0.140	0.260	0.00100	0.0300
Wi2	0.0160	0.0300	0.00100	0.0900	0.110	0.00100	0.0800
Pa1	0.00300	0.0300	0.00200	0.0400	0.0200	0.00100	0.0100
Pa2	0.00100	0.0300	0.00100	0.00	0.0300	0.00100	0.0100
Me1	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0100
Vi1	0.00100	0.0200	0.00100	0.0100	0.0300	0.00100	0.0100
Me2	0.00100	0.0800	0.00400	0.130	0.360	0.00100	0.0800
Vi2	0.00100	0.160	0.00700	0.160	0.130	0.00300	0.0800
Ho1	0.00200	0.0500	0.00200	0.0300	0.0400	0.00100	0.0200
Stp1	0.00100	0.0800	0.00200	0.0300	0.0800	0.00100	0.0100
Ho2	0.00100	0.0200	0.00200	0.0400	0.00	0.00100	0.0100
Stp2	0.00100	0.0100	0.00100	0.0200	0.00	0.00100	0.0100
Da	0.00500	1.08	0.0150	0.138	0.326	0.00	0.437
Bi	0.00200	0.106	0.00100	0.219	0.777	0.00	0.521
Sh	0.0130	1.00	0.0370	3.19	4.81	0.00	3.22
Ya1	0.00100	0.0300	0.00100	0.0400	0.0500	0.00100	0.0100
Ya2	0.0360	0.0800	0.0180	0.190	0.220	0.00100	0.130
St1	0.00100	0.0600	0.00400	0.0500	0.120	0.00100	0.0300
St2	0.00400	0.100	0.00700	0.100	0.520	0.00100	0.0700
Via1	0.00100	0.0500	0.00200	0.0100	0.0400	0.00100	0.0100
Th1	0.00200	0.0300	0.00100	0.0200	0.0500	0.00100	0.0100
Via2	0.00100	0.0300	0.00300	0.0700	0.0800	0.00100	0.0700
Th2	0.0230	0.0600	0.0160	0.0300	0.0300	0.00100	0.340
Ste1	0.00600	0.0500	0.00300	0.0200	0.0500	0.00100	0.0100
Hi1	0.00900	0.0700	0.00300	0.0400	0.230	0.00100	0.0100
Ste2	0.00100	0.0300	0.00100	0.0100	0.170	0.00100	0.0100
Hi2	0.00100	0.0100	0.00100	0.0200	0.0200	0.00100	0.0100
Be	0.0540	0.0890	0.906	1.66	3.87	0.00400	3.76
To	0.0480	0.102	0.530	1.68	2.57	0.00100	3.02
La1	0.00500	0.0300	0.00100	0.0500	0.0500	0.00100	0.0100
La2	0.00100	1.00	0.00100	0.0400	0.00	0.00100	0.0100
Li	0.146	0.136	0.282	5.66	5.36	0.0120	6.47
Ab	0.0260	0.143	0.155	0.31	0.904	0.00100	0.292
Re	0.00100	0.0200	0.00400	0.0540	0.154	0.00	0.0800
Di	0.0830	0.149	0.256	1.39	3.95	0.00300	4.87
Hop	0.0120	0.643	0.114	0.425	1.23	0.00	0.477

Table S7 Heavy metal build-up loads in 150-300 µm solids range (mg/g)

Sample ID	Cr	Mn	Ni	Cu	Zn	Cd	Pb
Pe	0.0	0.181	0.00	1.55	2.04	0.00	0.338
Ca1	0.00100	0.0400	0.00100	0.0300	0.0500	0.00100	0.0100
Ca2	0.00100	0.0700	0.00700	0.130	0.0900	0.00100	0.0200
De1	0.00100	0.0300	0.00100	0.0200	0.120	0.00100	0.0200
De2	0.00200	0.120	0.00600	0.110	0.470	0.00100	0.0800
Mer1	0.00100	0.180	0.00600	0.290	0.390	0.00200	0.0400
Wi1	0.00100	0.130	0.00500	0.0600	0.190	0.00100	0.0200
Mer2	0.0280	0.220	0.00700	0.200	0.540	0.00100	0.0100
Wi2	0.0100	0.100	0.00600	0.0500	0.0700	0.00100	0.0100
Pa1	0.00500	0.0800	0.0330	0.0900	0.120	0.00100	0.0300
Pa2	0.00100	0.0400	0.00200	0.0100	0.0800	0.00100	0.0100
Me1	0.00100	0.0300	0.00100	0.0200	0.0600	0.00100	0.0100
Vi1	0.00100	0.0400	0.00200	0.0500	0.260	0.00100	0.0300
Me2	0.00100	0.170	0.0130	0.250	0.700	0.00100	0.100
Vi2	0.00	0.130	0.00700	0.270	0.330	0.00100	0.130
Ho1	0.0130	0.120	0.00700	0.0900	0.150	0.00100	0.0700
Stp1	0.00100	0.160	0.00500	0.220	0.210	0.00100	0.0400
Ho2	0.00100	0.0300	0.0240	0.0700	0.0800	0.00100	0.0100
Stp2	0.00100	0.0600	0.0100	0.0600	0.0600	0.00100	0.0100
Da	0.00700	0.321	0.0140	0.264	0.292	0.00	0.121
Bi	0.00200	0.250	0.00400	0.344	0.771	0.00	0.144
Sh	0.00700	0.388	0.0110	3.29	1.71	0.00	0.657
Ya1	0.00100	0.0800	0.00200	0.0600	0.160	0.00100	0.0200
Ya2	0.0270	0.300	0.0150	0.270	0.520	0.00100	0.0200
St1	0.00100	0.140	0.00900	0.230	0.660	0.00100	0.0900
St2	0.00100	0.0400	0.00300	0.0500	0.190	0.00100	0.0300
Via1	0.00900	0.150	0.0120	0.0900	0.130	0.00600	0.0400
Th1	0.00100	0.0700	0.00300	0.0800	0.130	0.00100	0.0400
Via2	0.00100	0.130	0.00700	0.460	0.320	0.00100	0.0600
Th2	0.00100	0.0500	0.00200	0.170	0.140	0.00100	0.0400
Ste1	0.00300	0.0700	0.00200	0.0300	0.0900	0.00100	0.0100
Hi1	0.0120	0.100	0.00600	0.210	0.600	0.00100	0.0700
Ste2	0.00100	0.0500	0.00100	0.0100	0.160	0.00100	0.0100
Hi2	0.00100	0.0100	0.00100	0.0100	0.0500	0.00100	0.0100
Be	0.0260	0.109	0.466	3.05	3.16	0.00300	0.587
To	0.0490	0.0980	0.154	2.55	2.37	0.00	1.05
La1	0.00900	0.130	0.00600	0.190	0.240	0.00100	0.0300
La2	0.00100	0.130	0.00100	0.100	0.0800	0.00100	0.0200
Li	0.179	0.126	0.115	8.81	6.04	0.00700	2.24
Ab	0.0120	0.0460	0.0410	0.455	0.886	0.00100	0.0980
Re	0.00300	0.0370	0.105	0.145	0.274	0.00	0.0290
Di	0.0680	0.300	0.498	2.41	4.48	0.00300	0.518
Hop	0.00	0.123	0.00	0.281	0.655	0.00	0.209

Table S8 Heavy metal build-up loads in 75-150 µm solids range (mg/g)

Sample ID	Cr	Mn	Ni	Cu	Zn	Cd	Pb
Pe	0.00	1.21	0.00	6.65	3.90	0.00	0.289
Ca1	0.00200	0.190	0.00900	0.0900	0.240	0.00100	0.0300
Ca2	0.00100	0.0500	0.00200	0.0800	0.0800	0.00100	0.0200
De1	0.00100	0.0300	0.00100	0.0300	0.0900	0.00100	0.0200
De2	0.00100	0.140	0.00800	0.110	0.420	0.00100	0.0800
Mer1	0.00100	0.180	0.00600	0.170	0.460	0.00300	0.0300
Wi1	0.00900	0.380	0.0140	0.100	0.540	0.00100	0.0500
Mer2	0.0320	0.190	0.0220	0.230	0.690	0.00100	0.0400
Wi2	0.00800	0.120	0.00500	0.110	0.210	0.00100	0.0300
Pa1	0.00600	0.0800	0.0250	0.160	0.130	0.00100	0.0500
Pa2	0.00100	0.0300	0.00100	0.0100	0.0600	0.00100	0.0100
Me1	0.00100	0.0200	0.00100	0.0200	0.0600	0.00100	0.0100
Vi1	0.00100	0.0400	0.00300	0.0500	0.260	0.00100	0.0400
Me2	0.00100	0.140	0.0280	0.390	0.400	0.00100	0.0700
Vi2	0.00100	0.0900	0.00700	0.140	0.190	0.00100	0.100
Ho1	0.00700	0.110	0.00300	0.0600	0.190	0.00100	0.0100
Stp1	0.00100	0.0700	0.00500	0.0500	0.150	0.00100	0.0200
Ho2	0.0330	0.0300	0.0110	0.0800	0.130	0.00100	0.0100
Stp2	0.0160	0.0600	0.00400	0.0600	0.120	0.00100	0.0300
Da	0.0150	0.322	0.0150	0.440	0.539	0.00	0.0640
Bi	0.00100	0.131	0.00300	0.624	0.970	0.00	0.0390
Sh	0.0190	0.132	0.00500	2.66	1.34	0.00	0.0730
Ya1	0.00100	0.0900	0.00400	0.0600	0.230	0.00100	0.0200
Ya2	0.0290	0.580	0.0340	0.450	1.01	0.00100	0.0600
St1	0.00100	0.310	0.0290	0.270	1.24	0.00400	0.170
St2	0.0540	0.390	0.0370	0.200	1.08	0.00100	0.210
Via1	0.00100	0.100	0.00600	0.100	0.170	0.00300	0.0400
Th1	0.00100	0.0600	0.00200	0.120	0.130	0.00100	0.0500
Via2	0.00100	0.130	0.00700	0.260	0.340	0.00100	0.0700
Th2	0.00300	0.0900	0.00500	0.300	0.380	0.00100	0.0800
Ste1	0.00500	0.0800	0.00300	0.0500	0.170	0.00100	0.0100
Hi1	0.0130	0.110	0.00800	0.310	0.550	0.00100	0.100
Ste2	0.00100	0.0700	0.00200	0.0200	0.160	0.00100	0.0200
Hi2	0.00100	0.0200	0.00100	0.0200	0.0700	0.00100	0.0100
Be	0.0210	0.405	1.02	4.43	3.20	0.00300	0.240
To	0.0220	0.0640	0.0730	2.91	2.50	0.001	0.156
La1	0.00800	0.110	0.00500	0.160	0.240	0.001	0.0500
La2	0.00100	0.0600	0.00100	0.110	0.0900	0.001	0.0200
Li	0.0870	0.230	0.128	16.8	6.69	0.009	0.961
Ab	0.0150	0.0840	0.0290	0.721	1.21	0.002	0.150
Re	0.270	0.0810	0.470	0.247	0.267	0.000	0.0100
Di	0.0580	0.230	0.728	3.52	4.46	0.004	0.269
Hop	0.0620	0.388	0.0390	0.946	1.70	0.000	0.166

Table S9 Heavy metal build-up loads in <75 µm solids range (mg/g)

Sample ID	Cr	Mn	Ni	Cu	Zn	Cd	Pb
Pe	0.00	0.411	0.00	4.20	11.0	0.00	0.355
Ca1	0.0180	0.150	0.00500	0.0400	0.130	0.00100	0.0200
Ca2	0.00100	0.0100	0.00100	0.00	0.0100	0.00100	0.0100
De1	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0200
De2	0.0250	0.350	0.0190	0.210	0.770	0.00100	0.170
Mer1	0.00100	0.200	0.00700	0.0700	0.410	0.00200	0.0400
Wi1	0.00400	0.420	0.00700	0.0500	0.250	0.00100	0.0300
Mer2	0.0960	0.170	0.00400	0.500	0.280	0.00100	0.0300
Wi2	0.00100	0.0400	0.00100	0.0100	0.0100	0.00100	0.0200
Pa1	0.0120	0.170	0.0290	0.170	0.240	0.00100	0.0600
Pa2	0.00100	0.0500	0.00300	0.0200	0.0900	0.00100	0.0200
Me1	0.00100	0.0200	0.00100	0.0100	0.0100	0.00100	0.0200
Vi1	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0400
Me2	0.0280	0.170	0.0200	0.230	0.410	0.00100	0.0500
Vi2	0.0290	0.280	0.0230	0.350	0.840	0.00100	0.290
Ho1	0.00200	0.110	0.00200	0.110	0.300	0.00100	0.0300
Stp1	0.00100	0.0500	0.00100	0.0600	0.0700	0.00100	0.0200
Ho2	0.00100	0.0600	0.00700	0.0300	0.0100	0.00100	0.0100
Stp2	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0100
Da	0.00	0.00600	0.00600	0.234	0.452	0.00	0.00900
Bi	0.0140	0.365	0.0180	1.86	4.97	0.00	0.193
Sh	0.0350	0.613	0.0310	4.02	9.17	0.00	0.498
Ya1	0.00200	0.450	0.0110	0.0300	0.430	0.00100	0.0700
Ya2	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0100
St1	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0100
St2	0.107	0.820	0.0700	0.370	1.77	0.00100	0.420
Via1	0.00100	0.0800	0.00500	0.0700	0.110	0.00200	0.0400
Th1	0.00100	0.0300	0.00100	0.0600	0.0700	0.00100	0.0400
Via2	0.00100	0.0100	0.00100	0.0100	0.0100	0.00300	0.0300
Th2	0.00100	0.0100	0.00100	0.0100	0.0100	0.00100	0.0100
Ste1	0.0180	0.350	0.0150	0.120	0.620	0.00100	0.0600
Hi1	0.0240	0.110	0.00900	0.170	0.510	0.00100	0.0900
Ste2	0.00100	0.130	0.00400	0.0400	0.230	0.00100	0.0300
Hi2	0.00100	0.0900	0.00200	0.0700	0.230	0.00100	0.0400
Be	0.0790	0.526	2.12	5.40	15.3	0.00900	0.585
To	0.0550	0.270	0.317	4.59	13.9	0.00700	0.843
La1	0.00800	0.00	0.00100	0.00	0.0100	0.00100	0.0100
La2	0.00100	0.220	0.00400	0.100	0.310	0.00100	0.0400
Li	0.121	0.575	0.304	10.9	22.7	0.0290	2.75
Ab	0.0600	0.979	0.206	3.32	8.03	0.0110	1.02
Re	0.0280	0.532	0.184	1.99	3.28	0.00200	0.131
Di	0.114	0.391	1.67	5.09	20.2	0.0190	0.556
Hop	0.0350	0.756	0.0420	3.07	6.20	0.00	0.348

Table S10 Matrix of CV values (×10^2)

	Mn	Zn	Cr	Ni	Cu	Pb	Cd
>300-H	1.27	1.13	1.28	1.10	1.59	1.35	1.69
300-150-H	0.652	1.20	1.52	1.44	1.60	1.48	1.37
150-75-H	0.750	1.10	1.35	1.46	1.78	1.40	1.41
<75-H	0.672	0.936	0.854	1.62	0.958	1.27	1.20
>300-M	2.05	2.02	1.52	3.47	2.33	2.21	0.771
300-150-M	0.819	1.42	1.42	3.28	1.92	1.70	1.08
150-75-M	0.902	1.11	1.50	3.35	1.87	0.907	0.83
<75-M	1.09	1.98	1.69	3.56	2.04	1.37	1.34
>300-L	2.22	1.93	2.39	1.07	1.74	2.64	0.576
300-150-L	0.684	1.41	1.74	1.04	1.74	1.39	0.415
150-75-L	1.51	1.88	1.44	0.977	3.24	1.23	0.562
<75-L	1.01	3.17	1.99	1.24	2.92	1.53	0.415

H=high traffic volume; M=moderate traffic volume; L=low traffic volume

Table S11 Heavy metals build-up data extracted from seven previous publications

Sites	Reference*		ID[8]	Cd	Cu	Pb	Zn	Ni	Mn	Cr
Ulsan, Korea[1] (ug/g)	[25]	Mean	U-L	1.90	139	110	203	20.8	-	-
			U-M	1.86	99.4	92.8	160	18.9	-	-
			U-H	1.13	121	93.0	123	14.0	-	-
		SD	U-L	1.46	38.8	43.2	101	11.1	-	-
			U-M	0.802	42.3	10.3	23.3	7.86	-	-
			U-H	0.504	56.0	35.7	13.9	5.09	-	-
Massachusetts, USA (ppm)[2]	[24]	Mean	M-H	-	149	79.0	381	-	424	142
			M-M	-	121	214	309	-	492	101
			M-L	-	50.0	73.0	203	-	446	153
		SD	M-H	-	163	29.0	218	-	110	63.0
			M-M	-	80.0	501	180	-	254	54.0
			M-L	-	61.0	22.0	265	-	183	116
Shenzhen, China (mg/g)[3]	[9]	Mean	SZ-L	0.00100	0.0880	0.0150	0.580	0.0600	-	0.00300
			SZ-M	0.00	0.0480	0.0100	0.361	0.0410	-	0.00400
			SZ-H	0.00100	0.0500	0.0220	0.379	0.0330	-	0.00600
		SD	SZ-L	0.00	0.00700	0.00100	0.0350	0.0130	-	0.00
			SZ-M	0.00	0.0180	0.00200	0.213	0.0170	-	0.00200
			SZ-H	0.00	0.0240	0.0220	0.252	0.0120	-	0.00500

Zhenjiang, China (mg/kg)^4	[23]	Mean	ZJ-M1	-	56.1	148	468	-	-	-
			ZJ-L	-	59.4	199	513	-	-	-
			ZJ-M2	-	51.6	193	483	-	-	-
			ZJ-H	-	158	589	687	-	-	-
		SD	ZJ-M1	-	38.2	89.1	356	-	-	-
			ZJ-L	-	21.2	30.2	310	-	-	-
			ZJ-M2	-	25.3	100	285	-	-	-
			ZJ-H	-	6.41	8.92	10.1	-	-	-
Ma'an, Jordan (mg/kg)^5	[26]	Mean	MA-H1	2.60	19.7	105	231	134	105	-
			MA-H2	2.10	20.1	80.0	178	105	85.0	-
			MA-L1	1.50	18.5	64.0	140	85.0	73.0	-
			MA-M1	1.30	20.7	33.0	105	76.0	66.0	-
			MA-M2	2.40	20.5	25.1	113	88.0	48.0	-
			MA-M3	1.10	18.9	20.7	136	44.0	56.0	-
			MA-H3	2.20	26.4	98.0	260	110	110	-
			MA-M4	1.90	13.0	18.1	184	55.0	70.0	-
			MA-L2	1.90	13.4	15.4	48.0	75.0	80.0	-
		SD	MA-H1	4.10	3.30	4.10	6.20	3.10	4.20	-
			MA-H2	4.50	5.10	6.10	7.80	3.10	3.20	-
			MA-L1	4.10	4.20	4.30	3.40	5.20	3.40	-

			MA-M1	3.50	3.10	2.10	3.50	4.30	5.60	-
			MA-M2	4.10	5.20	7.20	1.50	6.50	4.60	-
			MA-M3	3.20	3.10	2.00	4.80	4.30	5.10	-
			MA-H3	3.70	4.60	2.10	3.20	3.10	5.80	-
			MA-M4	4.30	3.40	3.30	3.10	3.20	3.60	-
			MA-L2	3.10	2.10	2.40	2.30	2.30	4.20	-
Toronto, Canada (ppm)[6]	[27]	Mean	T-M1	0.0490	179	170	456	0.106	1.33×10^{3}	149
			T-H	0.0530	186	205	222	0.160	1.51×10^{3}	230
			T-L	0.0500	135	152	156	0.0780	1.39×10^{3}	188
			T-M2	0.0500	154	196	184	0.117	1.37×10^{3}	203
		SD	T-M1	0.00100	77.0	35.9	468	0.0350	991	73.4
			T-H	0.00400	83.4	113	82.1	0.156	762	137
			T-L	0.00200	47.3	26.3	81.5	0.0320	616	52.6
			T-M2	0.00100	40.8	60.0	92.0	0.0500	779	80.0
Kuala Lumpur, Malaysia (ug/g)[7]	[28]	Mean	KL-H1-i[9]	1.25	225	239	505	49.5	371	86.9
			KL-H1-ii	0.560	130	117	243	20.5	216	40.9
			KL-H1-iii	0.280	57.1	77.0	130	15.8	144	27.6
			KL-H2-i	1.08	445	110	140	23.5	267	53.8
			KL-H2-ii	0.590	244	67.6	83.0	15.2	188	32.9
			KL-H2-iii	0.410	184	57.5	68.3	12.5	169	28.0

			KL-M-i	0.710	84.3	88.4	394	22.3	311	56.7
			KL-M-ii	0.600	50.1	40.3	243	14.1	203	28.7
			KL-M-iii	0.310	22.0	31.9	153	10.1	147	18.6
			KL-L-i	4.50	45.4	87.4	218	16.8	309	40.3
			KL-L-ii	5.03	39.6	67.4	173	19.2	212	31.7
			KL-L-iii	4.36	42.7	46.2	110	8.38	178	23.0
		SD	KL-H1-i	0.230	28.1	84.1	84.7	8.56	61.3	16.3
			KL-H1-ii	0.110	24.2	47.00	16.2	3.56	31.7	13.4
			KL-H1-iii	0.160	14.8	43.5	28.2	4.27	26.0	10.4
			KL-H2-i	0.620	146	22.6	37.0	4.60	45.4	9.32
			KL-H2-ii	0.310	104	24.0	32.2	3.95	34.7	7.81
			KL-H2-iii	0.300	113	34.3	34.2	8.36	107	15.4
			KL-M-i	0.290	57.4	42.5	237	9.42	48.1	29.2
			KL-M-ii	0.430	44.4	19.4	131	5.39	25.5	12.6
			KL-M-iii	0.170	7.28	18.1	73.0	4.07	10.0	3.10
			KL-L-i	2.71	13.5	49.7	83.4	3.72	10.3	14.8
			KL-L-ii	3.33	23.4	41.7	62.4	13.2	64.6	11.1
			KL-L-iii	2.10	34.7	34.5	15.9	0.500	70.5	8.60

[1] This study measured heavy metals build-up loads and traffic data for 11 roads in Ulsan, Korea. Therefore, the high, moderate and low traffic groups were categorised according to their traffic measurements. Then, the mean and standard deviation values were calculated for each traffic group.

[2] This study categorised the study sites into high, moderate and low traffic groups. Additionally, the mean and standard deviation values of heavy metal build-up loads for each traffic group were also provided.

[3] This study measured heavy metals build-up loads and traffic data for 10 roads in Shenzhen, China. The high, median and low traffic groups were categorised according to their traffic measurements. Then, the mean and standard deviation values were calculated for each traffic group.

[4] This study gave the mean and standard deviation values of heavy metals build-up loads for a commercial area, residential area, riverside park and intense traffic area. The authors suggested that riverside park was a low traffic area while commercial and residential areas were moderate traffic areas. The intense traffic area was the high traffic area.

[5] This study categorised the sites into high, moderate and low traffic groups. Additionally, the mean and standard deviation values of heavy metal build-up loads for each traffic group were also given.

[6] This study measured heavy metal build-up loads and traffic data for 4 highways in Toronto, Canada. Accordingly, the high, moderate and low traffic groups were categorised according to the traffic measurements given. Then, the mean and standard deviation values were calculated for each traffic group.

[7] This study measured heavy metals build-up loads for three particle sizes for 4 roads in Kuala Lumpur, Malaysia. Additionally, the authors categorised the study roads into two high traffic, one moderate traffic and one low traffic areas.

[8] In column 4, the first letter or the first two letters represent the city while the second letter after the hyphen represents the traffic group (H=high traffic; M=median traffic; L=low traffic). The numeral represents the ID within one traffic group. For example, T-M2 represents mean and standard deviation values of the second median traffic group in Toronto, Canada.

[9] In column 4, the roman numerals given as i, ii and iii represent <63µm, 63-125µm and 125-250 µm particle sizes.

*The reference numbers correspond to the reference list in the main manuscript

Table S12 CV values calculated based on data from seven previous publications

ID	Cd	Cu	Pb	Zn	Ni	Mn	Cr
U-L	77.0	28.0	39.2	49.7	53.2	-	-
U-M	43.0	42.6	11.1	14.6	41.6	-	-
U-H	44.7	46.4	38.4	11.3	36.5	-	-
M-H	-	109	36.8	57.2	-	25.9	44.4
M-M	-	66.1	234	58.3	-	51.6	53.5
M-L	-	122	30.1	131	-	41.0	75.8
SZ-L	41.9	7.71	6.95	6.10	20.8	-	14.8
SZ-M	51.7	37.5	25.4	59.2	41.6	-	46.9
SZ-H	61.7	48.5	99.8	66.4	37.2	-	75.4
ZJ-L	-	35.8	15.2	60.4	-	-	-
ZJ-M1	-	68.1	60.2	76.0	-	-	-
ZJ-M2	-	49.1	51.9	59.0	-	-	-
ZJ-H	-	4.05	1.51	1.50	-	-	-
MA-H1	158	16.8	3.91	2.70	2.30	4.00	-
MA-H2	214	25.4	7.63	4.40	3.00	3.80	-
MA-L1	273	22.7	6.72	2.40	6.10	4.70	-
MA-M1	269	15.0	6.36	3.30	5.70	8.50	-
MA-M2	171	25.4	28.7	1.30	7.40	9.60	-
MA-M3	291	16.4	9.66	3.50	9.80	9.10	-
MA-H3	168	17.4	2.14	1.20	2.80	5.30	-
MA-M4	226	26.2	18.2	1.70	5.80	5.10	-
MA-L2	163	15.7	15.6	4.80	3.10	5.30	-
T-M1	2.04	43.1	21.1	103	33.0	74.5	49.3
T-H	7.55	44.8	55.0	37.0	97.5	50.6	59.7
T-L	4.00	35.1	17.3	52.2	41.0	44.3	28.0
T-M2	2.00	26.5	30.6	50.1	42.7	57.0	39.4
KL-H1-i	18.4	12.5	35.2	16.8	17.3	16.5	18.7
KL-H1-ii	19.6	18.6	40.3	6.70	17.4	14.7	32.7
KL-H1-iii	57.1	25.9	56.5	21.7	27.0	18.1	37.9
KL-H2-i	57.4	32.7	20.5	26.5	19.6	17.0	17.3
KL-H2-ii	52.5	42.7	35.5	38.8	26.0	18.4	23.8
KL-H2-iii	73.2	61.3	59.6	50.1	67.1	63.0	55.0
KL-M-i	40.8	68.1	48.0	60.0	42.3	15.5	51.5
KL-M-ii	71.7	88.7	48.0	54.1	38.3	12.5	43.8
KL-M-iii	54.8	33.1	56.7	47.6	40.4	6.80	16.7
KL-L-i	60.2	29.8	56.8	38.4	22.2	3.30	36.8
KL-L-ii	66.2	59.1	61.9	36.1	68.7	30.5	34.9
KL-L-iii	48.2	81.3	74.7	14.5	6.00	39.7	37.4

Matlab code and results for stepwise linear regression

Matlab code

```
mdl = stepwiselm (x, y,'Criterion','adjrsquared')
```

Stepwiselm returns a linear model of the responses `y` to the predictor variables in the data matrix `X`, using stepwise regression to add or remove predictors. The adding or removal of predictor variables was based on the criterion of increase in the value of adjusted R^2

Results calculated by Matlab

```
1. Adding x5, AdjRsquared = 0.12295
2. Adding x3, AdjRsquared = 0.19491
3. Adding x4, AdjRsquared = 0.33164
4. Adding x2, AdjRsquared = 0.3592
5. Adding x1, AdjRsquared = 0.36011
6. Adding x1:x4, AdjRsquared = 0.51797
7. Adding x1:x2, AdjRsquared = 0.5921
8. Adding x1:x5, AdjRsquared = 0.62133
9. Adding x3:x5, AdjRsquared = 0.62547
10. Adding x1:x3, AdjRsquared = 0.63325
11. Adding x2:x3, AdjRsquared = 0.63831
12. Adding x2:x5, AdjRsquared = 0.64711
```

```
mdl =
Linear regression model:
   y ~ 1 + x1*x2 + x1*x3 + x1*x4 + x1*x5 + x2*x3 + x2*x5 + x3*x5
Estimated Coefficients:
                   Estimate          SE          tStat        pValue
                 ___________    __________    _________    __________

   (Intercept)      -62.547       12.232      -5.1134    1.6914e-05
   x1              0.016938    0.0036301       4.666    5.9731e-05
   x2                182.13       62.437       2.917     0.0066334
   x3                19.326       36.986     0.52254       0.60513
   x4                121.45       19.504       6.227    7.4043e-07
   x5            0.00089229    0.0016019       0.557       0.58166
   x1:x2          -0.034105     0.013075     -2.6083      0.014048
   x1:x3           0.019265     0.014577      1.3216        0.1963
   x1:x4           -0.03285    0.0066046     -4.9738    2.5088e-05
   x1:x5        -2.1462e-07   5.5408e-07    -0.38735       0.70123
   x2:x3            -310.06       226.33       -1.37       0.18087
   x2:x5         -0.0056864    0.0042703     -1.3316       0.19302
   x3:x5           0.014012    0.0078733      1.7797      0.085255
Number of observations: 43, Error degrees of freedom: 30
Root Mean Squared Error: 11.6
R-squared: 0.748,  Adjusted R-Squared 0.647
F-statistic vs. constant model: 7.42, p-value = 4.26e-06
```

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