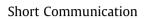
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Mathematical relationships for metal build-up on urban road surfaces based on traffic and land use characteristics



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HIGHLIGHTS

• Sources of metal build-up: Mn-geogenic; Pb, Cu, Zn, Cr, Ni and Cd-traffic related.

• Traffic sources: vehicle wear-Pb, Cu, Zn; exhaust-Cr, Ni; exhaust and wear-Cd.

• Land use does not exhibit clear pattern in influencing metal build-up process.

• Reliable prediction equation developed for cumulative fuel related metal load.

• Prediction equation for wear-related metal load is suitable for preliminary studies.

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The study investigated the influence of traffic and land use parameters on metal build-up on urban road surfaces. Mathematical relationships were developed to predict metals originating from fuel combustion and vehicle wear. The analysis undertaken found that nickel and chromium originate from exhaust emissions, lead, copper and zinc from vehicle wear, cadmium from both exhaust and wear and manganese from geogenic sources. Land use does not demonstrate a clear pattern in relation to the metal build-up process, though its inherent characteristics such as traffic activities exert influence. The equation derived for fuel related metal load has high cross-validated coefficient of determination (Q^2) and low Standard Error of Cross-Validation (SECV) values which indicates that the model is reliable, while the equation derived for wear-related metal load has low Q^2 and high SECV values suggesting its use only in preliminary investigations. Relative Prediction Error values for both equations are considered to be well within the error limits for a complex system such as an urban road surface. These equations will be beneficial for developing reliable stormwater treatment strategies in urban areas which specifically focus on mitigation of metal pollution.

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1. Introduction

Stormwater transports a range of potentially toxic metal ions deposited on urban impervious surfaces, particularly from road surfaces, to receiving waters causing adverse aquatic ecosystem health impacts (Herngren et al., 2006). Effective management of stormwater related metal pollution requires accurate estimation of metal loads present on road surfaces based on an in-depth understanding of the metal build-up process. Though solids build-up process has been widely understood, the transferability of the knowledge to specific pollutants such as metals is very limited (Liu et al., 2012).

Egodawatta et al. (2013) developed a mathematical model to replicate the metal build-up process based on antecedent dry days. However, the model did not consider the specific influence exerted by widely acknowledged major anthropogenic sources of metals on road surfaces, namely traffic and land use activities (Goonetilleke et al., 2009; Mahbub et al., 2010). This constrains its use in the development of effective traffic and land use related pollution mitigation strategies. Accordingly, the aims of this study were to: (1) investigate the influence of traffic and land use characteristics in the metal build-up process on urban road surfaces; and (2) develop quantitative mathematical relationships to predict metal loads in the build-up on road surfaces based on traffic and land use parameters. The outcomes from this study will contribute



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to the evaluation of metal pollution in the urban environment from future changes in traffic and land use characteristics and for the development of reliable stormwater treatment strategies in urban areas.

2. Materials and methods

Eleven road sites were selected in the Gold Coast region, Queensland, Australia with the study sites encompassing variations in traffic and land use characteristics (Fig. S1 in Supplementary information). The build-up samples were collected from 2.0 m \times 1.5 m plot areas in the middle of the traffic lane using the wet and dry vacuuming system described by Mahbub et al. (2011), which consisted of a domestic vacuum cleaner fitted with a water filtration system. The selected plots were firstly dry vacuumed to collect most of the dust samples, and then wet vacuumed after spraying deionized water on the plots at 2 bar pressure for 3 min in order to collect the remaining fine particles. Prior to the sample collection, the procedure was tested under laboratory conditions and was found to be 97.4% efficient (Refer to Supplementary information).

The samples collected were transported and stored in the laboratory under prescribed conditions until the analysis of the following metals commonly present on urban road surfaces was undertaken: cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), manganese (Mn), nickel (Ni) and zinc (Zn), using Method 200.8 for Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS) (US-EPA, 1994) with TraceSELECT (Product No. 54704) as the certified reference material. The detection limits for ICP-MS for the selected metals were in the range of 0.001–0.005 mg L⁻¹. Additionally, the total solids (TS) was determined using Methods 2540C and 2540D (APHA, 2004).

Pollutant build-up on road surfaces is influenced by traffic factors such as traffic volume, congestion and vehicle mix (EPASGV, 1999), and land use characteristics within the vicinity of the site (Goonetilleke et al., 2009). In this context, the following surrogate indicators were selected to represent these influential factors in the analysis: annual average daily traffic volume (ADT_to) as a surrogate for traffic volume; volume to capacity ratio (V/C) as a surrogate for congestion and total heavy duty traffic volume (ADT_hv) as a surrogate for vehicle mix. Relevant data was obtained by undertaking classified traffic counts at the respective study sites. Surrogates that represented land use factors were the percentage of, industrial (1%), commercial (C%) and residential (R%) land use within 1 km radius from the sampling points. The data analysis was conducted using multivariate methods including principal component analysis (PCA), factor analysis (FA) and multiple linear regression analysis (MLR).

PCA transforms a large set of variables into an orthogonal set of principal components. PCA transforms original variables to

Data matrix used in the study.

orthogonal principal components (PCs) so that highest variance is associate to first few PCs. This results in the reduction in the number of variables, thereby facilitating effective interpretation of the data set. PCA outcomes are often presented as biplots, which enable the identification of the underlying relationships between objects and variables (Mostert et al., 2010). A detailed description of PCA can be found elsewhere (Adams, 1995). Similarly, FA uses few factors to explain the correlation between the variables. By observing the characteristics of the variables correlated to a factor, it is possible to explain what a particular factor represents (Abdi, 2003). MLR is a regression technique that is often used to develop mathematical relationships for a dependent variable based on a number of independent variables (Ni et al., 2001). In this study, FA was performed using StatistiXL software (v. 1.8, 2008, StatistiXL, Broadway-Nedlands, Australia) while MATLAB R2009b (Mathworks Inc., Natick, MA, USA) was used for MLR and PCA analysis.

3. Results and discussions

3.1. Factor analysis (FA)

FA was performed on the raw data matrix (Table 1) using principal component extraction method with orthogonal VARIMAX rotation technique, which results in factors being strongly correlated to a specific set of variables, while weakly correlated with other variables as a result of rotating the original factors (Egodawatta et al., 2013). This simplifies the interpretation of a complex data set as each variable is primarily associated with a specific factor (Abdi, 2003). The factors were extracted based on the initial eigenvalue criteria ≥ 1 and the results are presented in Table 2. It was hypothesised that metals with the same source of origin and build-up process are grouped under the same factor.

As evident in Table 2, Cr and Ni are associated with Factor 1 because they have relatively higher loadings in Factor 1 compared to the other factors. Similarly, Pb, Zn and Cu have higher loadings in Factor 2, while Mn has higher loading on Factor 3. This suggests that the source and build-up process for Pb, Cu and Zn are different to those for Cr and Ni, while the source and build-up process for Mn is different to these two metal groups. Furthermore, Cd has a loading of 0.29 on Factor 1, 0.11 on Factor 2 and a relatively high negative loading on Factor 3. This suggests that the source and build-up process for Cd would be different from those of the other metals.

3.2. Exploratory principal component analysis (PCA)

To facilitate visual display and interpretation of the results, PCA were separately performed on standardised data matrices consisting of: (1) metal loads and traffic variables, and (2) metal loads and

| Site ID | Total solids (mg/100 m ²) | Metal loads (mg/100 m ²) | | | | | | | Traffic variables | | Land use variables | | | |
|---------|---------------------------------------|--------------------------------------|------|------|------|-----|------|------|-------------------|--------|--------------------|------|------|------------|
| | | Cd | Cr | Ni | Pb | Zn | Cu | Mn | ADT_to | ADT_hv | V/C | С% | I% | <i>R</i> % |
| Ab_c | 21.2 | 0.31 | 2.38 | 9.11 | 33.0 | 233 | 102 | 26.5 | 8739 | 101 | 0.60 | 0.28 | 0.04 | 0.68 |
| Re_r | 64.4 | 0.17 | 19.5 | 49.1 | 16.2 | 256 | 157 | 43.2 | 9973 | 193 | 0.72 | 0.04 | 0.02 | 0.94 |
| Pe_r | 5.71 | 0.00 | 0.00 | 0.00 | 12.5 | 107 | 75.0 | 10.6 | 30 | 0 | 0.00 | 0.03 | 0.00 | 0.97 |
| Bi_r | 46.9 | 0.00 | 0.86 | 1.24 | 42.0 | 351 | 143 | 40 | 1963 | 10 | 0.45 | 0.24 | 0.03 | 0.73 |
| Be_i | 6.19 | 0.11 | 1.11 | 27.9 | 32.0 | 158 | 90 | 6.99 | 4630 | 86 | 0.46 | 0.07 | 0.59 | 0.34 |
| Sh_i | 18.6 | 0.00 | 1.38 | 1.56 | 82.9 | 317 | 245 | 39.8 | 2234 | 31 | 0.22 | 0.05 | 0.48 | 0.47 |
| Ho_c | 25.2 | 0.00 | 2.75 | 4.90 | 30.3 | 247 | 119 | 48.2 | 25571 | 270 | 0.59 | 0.14 | 0.01 | 0.85 |
| Li_c | 5.13 | 0.29 | 2.74 | 4.25 | 63.7 | 209 | 216 | 5.47 | 8594 | 28 | 0.73 | 0.26 | 0.03 | 0.71 |
| To_c | 6.59 | 0.05 | 1.15 | 7.07 | 33.4 | 141 | 77.3 | 3.52 | 5922 | 61 | 0.18 | 0.30 | 0.17 | 0.53 |
| Da_r | 110 | 0.00 | 2.90 | 5.46 | 69.3 | 177 | 118 | 190 | 993 | 6 | 0.09 | 0.01 | 0.00 | 0.99 |
| Di_r | 6.72 | 0.19 | 2.17 | 21.2 | 41.7 | 222 | 83.4 | 7.20 | 10682 | 41 | 0.69 | 0.02 | 0.00 | 0.98 |

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