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Linking source characterisation and human health risk assessment of metals to rainfall characteristics☆

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A B S T R A C T

Metals deposited on urban road surfaces and incorporated in stormwater runoff are discharged into receiving waters, influencing their quality and can pose human health risks. Effective design of stormwater treatment measures is closely dependent on the in-depth understanding of stormwater pollutant sources and the associated health risks. The study discussed in this paper has linked the sources of metals in stormwater runoff and the accompanying human health risk to rainfall characteristics. The study outcomes confirmed that the metal contributions to stormwater runoff from the primary sources were in the order of sea salt > soil > traffic. Although traffic contributes a relatively lower percentage to wash-off, the human health risks posed by traffic sourced metals were relatively much higher. This implies that traffic sources should receive particular attention in treating stormwater. These outcomes have the potential to contribute to enhancing effective source control measures in order to safeguard natural waterways from polluted road wash-off.

1. Introduction

Urban road surfaces are receptors of pollutants generated by a range of natural and anthropogenic sources while also forming an effective platform for wash-off during rainfall events. The polluted wash-off from urban road surfaces drains into receiving waters, influencing their quality and the health of the aquatic environment (Wu et al., 2017b). Among pollutant types related to urban road surfaces, metals have been investigated to a significant extent. This is due to the fact that metals can be toxic and exert adverse impacts on human health (Liu et al., 2016b; Ma and Singhirunnusorn, 2012; Zeng et al., 2015). Therefore, implementation of measures to mitigate stormwater pollution is common. However, the design of effective stormwater treatment measures is closely dependent on the in-depth understanding of stormwater pollutant sources and contributions (Mummullage et al., 2016a).

A range of studies have been conducted to identify the sources and their contributions to pollutant build-up on road surfaces (such as Zannoni et al., 2016; Zheng et al., 2016). However, there are only a limited number of studies which have focused on source quantification of pollutants wash-off from road surfaces. Egodawatta et al. (2007) noted that only a fraction of pollutants available on road surfaces (pollutants build-up) are washed-off during rainfall events. This could lead to differences in the sources of pollutants and their contributions in build-up and wash-off. Hence, studies undertaken to investigate the sources and contributions of pollutants in build-up, do not necessarily directly relate to what is present in wash-off. This phenomenon has not been explicitly addressed in past research literature.

Furthermore, as metals on urban roads can be from different sources (Hussain et al., 2015; Liao et al., 2017; Mummullage et al., 2016b), their toxicity could be different. For example, soil primarily introduces metals such as Al and Mn with toxicities that are relatively lower than those of traffic sourced metals such as Cu and Cr (Hakanson, 1980; Wu et al., 2017a). This means that human

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health risks posed by metals when they are washed-off by stormwater runoff could vary with their sources. Furthermore, the risks could vary with rainfall and the resulting stormwater runoff characteristics. For instance, the first flush effect leads to higher concentrations of pollutants at the beginning of a rainfall event (Kang et al., 2008; Li et al., 2007; Liu et al., 2010). Therefore, this could lead to higher human health risks from the initial fraction of stormwater runoff when compared to the later fraction. Unfortunately, the investigation of the influence of rainfall characteristics on human health risks posed by metals in stormwater runoff, in terms of their sources, is extremely limited.

This paper presents the results of an extensive study undertaken to identify metal sources and quantify the contributions in wash-off from road surfaces. Based on the identified sources and their contributions, human health risks posed by metals from different sources were assessed in terms of their relationships with rainfall characteristics. This study also linked metal sources and their human health risk to rainfall characteristics. The outcomes from this study is expected to contribute to enhancing the formulation of effective source control measures in order to safeguard receiving waters from polluted urban road wash-off.

2. Materials and methods

2.1. Study sites

Pollutant wash-off samples were collected from two selected road sites, namely, Yarrimbah Drive (YD) and Via Roma Drive (VRD) from Gold Coast, Australia as shown in Fig. 1. The selected sites consisted of residential (YD) and commercial (VRD) land uses, respectively. The commercial and residential land uses were selected due to the fact that roads within these two areas have relatively good surface conditions. YD is the connector road to the main arterial while VRD is a major arterial road. Both roads carry relatively high traffic volumes, which are around 3000 vehicles per day.

2.2. Sample collection

Rainfall events were simulated using a rainfall simulator and pollutant wash-off samples were collected as described in Egodawatta et al. (2007). The use of simulated rainfall overcame the practical issues relating to uncertainties associated with the occurrence and consistency of natural rainfall and enabled effective control of a range variables influencing wash-off such as rainfall intensity and duration (Brodie and Rosewell, 2007; Egodawatta and Goonetilleke, 2008; Liu et al., 2012).

The simulator used in this study was calibrated for the selected rainfall intensities prior to sample collection. Details of the rainfall simulator and its calibration procedure can be found in Herngren et al. (2005). Rainfall intensities simulated were 83, 64, 38 and 25 mm/h. Simulated rainfall durations were varied from 6 min to 36 min. The selection of rainfall intensities and durations were based on statistical analysis of long term rainfall records (historical rainfall records over a period of 19 years, see Figure S1 in the Supplementary Information) for the region. These rainfall events simulated are generally less than 1-year annual recurrence interval (ARI) at the study sites (see Table S1 and Figure S2) since most stormwater treatment system designs are based on this ARI range (Liu et al., 2016a). The rainfall event selection process is provided in the Supplementary Information. It is important to note that rainfall characteristics in the study primarily refer to rainfall duration and intensity and seasonal variations were not considered. Accordingly, 17 samples were collected from each study site. Table 1 gives the rainfall intensities and durations for the 17 samples collected.

2.3. Laboratory analysis

Collected samples were tested for metals which included Li, Na, Mg, Al, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Mo, Rh, Cd, Sn, Sb, Ba, Pt and Pb. This selection of metals was to ensure that they act as tracers to facilitate clear differentiation between various sources (Gasperi et al., 2014; Legret and Pagotto, 1999; Mummullage et al., 2016b; Thorpe and Harrison, 2008). Testing was based on EPA 200.8 method (EPA, 1994) and duplicate samples were tested in order to ensure the reliability of the test results. An Inductively Coupled Plasma- Mass Spectrometry (ICP/MS, Agilent 8800 Triple Quadrupole) was used for the analysis. Quality control/assurance procedures found that the percentage recovery of metals was 90–115%, which is acceptable for road dust samples (Mahbub et al., 2010). Detailed descriptions of the analyses undertaken and relevant quality assurance/quality control measures adopted can be found in the Supplementary Information together with the metal concentrations obtained, which is given in Table S2.

2.4. Analytical tools for source identification and apportionment

The statistical analysis of the collected data was primarily

Fig. 1. Selected study roads.