Ecotoxicology and Environmental Safety 164 (2018) 696–703

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

journal homepage: www.elsevier.com/locate/ecoenv

Heavy metals transport pathways: The importance of atmospheric pollution contributing to stormwater pollution

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ARTICLE INFO

Keywords:
Heavy metals
Atmospheric pollution
Stormwater quality
Stormwater pollutant processes
Multivariate analysis

ABSTRACT

Pollution has become a serious issue in the urban water environment as stormwater runoff transports a range of pollutants to receiving water bodies, undermining water quality and posing human and ecosystem health risks. Commonly, the primary focus of stormwater quality research is on the role of pollutants directly accumulating at the ground phase. However, atmospheric phase can also exert a significant impact on stormwater quality through atmospheric deposition. Unfortunately, only limited research has focused on the linkage between atmospheric and ground phases in relation to urban stormwater quality. The study discussed in this paper characterised the four primary transport pathways, atmospheric build-up (AB), atmospheric deposition (AD) and road surface build-up (BU) and wash-off (WO) in relation to heavy metals, which is a key urban stormwater pollutant. The research outcomes confirmed the direct linkage between atmospheric phase and ground phase and in turn the significance of atmospheric heavy metals as a contributing source to stormwater runoff pollution. Zn was the most dominant heavy metal in all four pathways. For the AB pathway, atmospheric heavy metal pollution on weekdays is more serious than weekends. For the AD pathway, dry atmospheric deposition of heavy metals is positively correlated to dry days, whilst wet (bulk) deposition is related to rainfall depth. For the BU pathway, heavy-duty vehicle traffic volume was found to be the most important source. For the WO pathway, industrial and commercial areas tend to produce higher heavy metal concentrations in stormwater runoff than residential areas. The study results will contribute to the creation of effective urban stormwater pollution mitigation strategies and thereby enhancing the quality of the urban water environment.

1. Introduction

Urban stormwater pollution has received significant attention since it can undermine receiving water quality, leading to adverse impacts on the urban water environment (Drake et al., 2014; Wang et al., 2010). Past research on urban stormwater quality have generally focused on the role played by various influential factors including vehicular traffic and land use characteristics (Borris et al., 2013; Lee et al., 2009; Wijesiri et al., 2018) in relation to pollution deposition on ground surfaces such as road surfaces and entrainment in stormwater runoff. Essentially, these past research studies have been mainly undertaken based on the ground phase (Bian et al., 2015; Shorshani et al., 2014; Zhao et al., 2014) as the primary sources of urban stormwater pollutants. The common research methodology is to collect stormwater runoff samples from catchment surfaces and to correlate their concentrations/loads with the surrounding environment (such as Liu et al., 2018; Zannoni et al., 2016).

However, urban stormwater quality is dependent not only on pollutants contributed by the ground phase, but also pollutants contributed by the atmospheric phase (air) (Gunawardena et al., 2013; Weerasundara et al., 2018). Pollutants generated in the urban environment can firstly accumulate in the air and then transported to ground surfaces through atmospheric deposition (Wang et al., 2016). Atmospheric deposition can occur as dry and wet deposition. In this context, atmospheric pollution can also be a significant contributor to stormwater quality degradation. However, research studies which have

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https://doi.org/10.1016/j.ecoenv.2018.08.072
Received 17 May 2018; Received in revised form 13 August 2018; Accepted 20 August 2018
0147-6513/

focused on the linkage between atmospheric and ground phases in terms of urban stormwater quality are extremely limited. This is a serious constraint to the development of a comprehensive understanding of stormwater pollutant transport pathways for the creation of effective stormwater pollution mitigation strategies.

Heavy metals are a common pollutant present in the urban environment. The presence of heavy metal loads is strongly related to urban traffic and land use characteristics (Duong and Lee, 2011; Mejia et al., 2013; Zhao and Li, 2013). For example, traffic congestion is an important factor in heavy metals build-up on road surfaces and subsequent wash-off (Shorshani et al., 2014), while industrial land use tends to generate stormwater runoff with relatively high heavy metal concentrations (Shooshtarian et al., 2018; Tiefenthaler et al., 2008). In this regard, an in-depth understanding of heavy metals transport pathways and their relationships with influential factors can provide essential insights into stormwater treatment design, particularly for systems targeting heavy metal removal (Cristaldi et al., 2017).

The research study discussed in this paper investigated heavy metals which are widely present in the urban environment and harmful to human and ecosystem health (Adel et al., 2016; Mazzei et al., 2014; Sciacca and Conti, 2009). The study entailed the characterisation of the four primary transport pathways of heavy metals, namely, atmospheric build-up (accumulation in the air, AB), atmospheric deposition (AD, including dry deposition, DD and wet deposition, WD), and build-up (BU) and wash-off (WO) from road surfaces. The knowledge created is expected to improve effective stormwater treatment design strategies and thereby improving the water quality in the urban environment.

2. Materials and methods

2.1. Study sites

The study sites selected were located in Gold Coast City, Australia. This City has experienced high population growth, leading to increased traffic volume. Fifteen study sites were selected where AB and AD samples as well as BU samples were collected while data on WO were generated using a modelling approach (see Supplementary Information). Eleven of the study sites were road surfaces while four sites were located in areas with various anthropogenic activities and were selected to collect only atmospheric phase samples. Fig. S4 and Table S1 in the Supplementary Information show the selected study sites and relevant land use characteristics. The study sites encompassing typical land uses including residential, commercial and industrial areas are characterised by varied traffic activities such as vehicular traffic volume and degree of congestion. This permitted the investigation of heavy metal transport pathways influenced by a wide range of traffic and land use characteristics.

2.2. Sampling, testing and data collection

Samples of AB (representing pollutant accumulation in the atmosphere), AD (including dry deposition during dry weather and wet deposition during rainy days) and BU (representing pollutant accumulation on road surfaces) were collected while data related to WO (representing pollutants washed-off by stormwater runoff) were generated using a a modelling approach developed in a previous study undertaken by the authors (Ma et al., 2016). Table S1 gives the number of sampling episodes for each transport pathway investigation. AB samples were collected during weekdays and weekends as there are significant differences in traffic characteristics. The sample collection procedures are detailed in the Supplementary Information. Heavy metals investigated included Manganese (Mn), lead (Pb), copper (Cu), Zinc (Zn), Nickel (Ni), Cadmium (Cd) and Chromium (Cr) as these are commonly present in the urban environment and closely related to urban traffic (Duong and Lee, 2011; Johansson et al., 2009; Mejia et al., 2013). It is noteworthy that this study primarily focused on traffic related heavy metals and hence other metal species such as those related to manufacturing (for example, mercury) and geogenic sources (for example, ferrous) (Egodawatta et al., 2013) were not included. Testing was undertaken using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) specified in the EPA Method 200.8 (EPA, 1994) and standard QC/QA procedure was adopted during the testing. It was found that the recovery was within 85–115%, which was considered acceptable.

Additionally, physio-chemical parameters including total airborne particulate (total suspended particulate) matter (TSP) for AB samples and total organic carbon (TOC) and total solids (TS) in BU samples were also analysed as these parameters influence pollutants in both, atmospheric and ground phases (Borne et al., 2013; Kayhanian et al., 2008). In the case of road build-up, each BU sample was sieved to separate into five particle sizes (namely, < 1 μm, 1–75 μm, 75–150 μm, 150–300 μm and > 300 μm). This was undertaken due to the fact that pollutant adsorption to solids including heavy metals is influenced by the particle size (Li et al., 2015; Samara and Voutsa, 2005). Table S2 in the Supplementary Information provides details of the laboratory testing undertaken in relation to the four transport pathways and the test data are provided in Table S3-S7 in the Supplementary Information.

Data relating to wash-off (see Table S7 in the Supplementary Information) were generated using the modelling approach noted above (Ma et al., 2016). The modelling approach uses a number of influential factors (traffic and land use related to parameters) to determine the heavy metal concentrations in stormwater runoff. Eq. (1) gives the modelling equation used while Eq. (2) was used to calculate Cu wash-off concentration (mg/L). Equations for calculating other heavy metal concentrations as well as detailed information about the calculation process are given in the Supplementary Information. The heavy metal wash-off concentrations determined for each road site were the average concentrations. Eq. (1) (Ma et al., 2016) and Eq. (2) (Ma et al., 2016).

$$C = f(DTV, \quad C, \quad I, \quad R) \tag{1}$$

$$\mathrm{Cu} = \exp(-8.653 + 0.4779 \times \ln\mathrm{DTV} + 5.841 \times \mathrm{C} + 24.78 \times \mathrm{I} - 93.96 \times \mathrm{I}^2 + 0.557 \times \mathrm{R}) \tag{2}$$

where: C – heavy metal concentrations in stormwater;

DTV – total average daily traffic volume (DTV);
C, I, R – percentage of commercial, industrial and residential land use area within 1 km radius from sampling points.

Data relating to influential parameters including land use related parameters (C, I and R as shown in Eq. (1)), traffic related parameters (average daily traffic DTV, heavy-duty vehicle traffic HDTV and parameter representing traffic congestion condition V/C) and climatic parameters (wind speed for atmospheric build-up analysis, antecedent dry days for DD and BU analysis and rainfall characteristics for WD) were also collected and provided in Table S3–S7 in the Supplementary Information. Traffic data was provided by Gold Coast City Council, while climatic parameters were obtained from the Australian Bureau of Meteorology.

2.3. Study approach

This study included four types of data analysis representing the four transport pathways of heavy metals, namely AB, AD, BU and WO on road surfaces. Principal component analysis (PCA) was the main technique applied to identify the relationships between influential factors and each transport pathway. The use of PCA was due to its exceptional ability for identifying correlations between objects and variables. The identified correlations are considered as reliable if the total percentages of significant principal components are larger than 60% (Adams, 1995). In this study, when the percentage is less than 60%, a correlation matrix is provided to support the PCA analysis. Origin 9.5 software was used to