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Build-up and wash-off dynamics of atmospherically derived Cu, Pb, Zn and TSS in stormwater runoff as a function of meteorological characteristics

Louise U. Murphy [a], Thomas A. Cochrane [a,*], Aisling O'Sullivan [a,b]

[a] University of Canterbury, Department of Civil and Natural Resources Engineering, Private Bag 4800, Christchurch, New Zealand
[b] Pattle Delamore Partners Ltd., 295 Blenheim Rd., Christchurch 8041, New Zealand

HIGHLIGHTS

- We have monitored atmospherically derived Cu, Pb, Zn and TSS loads in stormwater.
- Mixed-effect models predicted pollutant loads from rain characteristics.
- Pollutant build-up was related to antecedent dry days.
- Pollutant wash-off of Cu and Zn were related to rain depth.
- Pollutant wash-off of Pb and TSS were related to rain intensity and duration.

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ABSTRACT

Atmospheric pollutants deposited on impermeable surfaces can be an important source of pollutants to stormwater runoff; however, modelling atmospheric pollutant loads in runoff has rarely been done, because of the challenges and uncertainties in monitoring their contribution. To overcome this, impermeable concrete boards ($\approx 1\ m^2$) were deployed for 11 months in different locations within an urban area (industrial, residential and airside) throughout Christchurch, New Zealand, to capture spatially distributed atmospheric deposition loads in runoff over varying meteorological conditions. Runoff was analysed for total and dissolved Cu, Zn, Pb, and total suspended solids (TSS). Mixed-effect regression models were developed to simulate atmospheric pollutant loads in stormwater runoff. In addition, the models were used to explain the influence of different meteorological characteristics (e.g. antecedent dry days and rain depth) on pollutant build-up and wash-off dynamics. The models predicted approximately 53% to 69% of the variation in pollutant loads and were successful in predicting pollutant–load trends over time which can be useful for general stormwater planning processes. Results from the models illustrated the importance of antecedent dry days on pollutant build-up. Furthermore, results indicated that peak rainfall intensity and rain duration had a significant relationship with TSS and total Pb, whereas, rain depth had a significant relationship with total Cu and total Zn. This suggested that the pollutant speciation phase plays an important role in surface wash-off. Rain intensity and duration had a greater influence when the pollutants were predominantly in their particulate phase. Conversely, rain depth exerted a greater influence when a high fraction of the pollutants were predominantly in their dissolved phase. For all pollutants, the models were represented by a log–arctan relationship for pollutant build-up and a log–log relationship for pollutant wash-off. The modelling approach enables the site-specific relationships between individual pollutants and rainfall characteristics to be investigated.

1. Introduction

Knowledge of stormwater pollution loads (or concentrations) throughout rain events is required to evaluate the impact of stormwater on the health and the ecology of an urban waterway. This knowledge is also required to help select appropriate stormwater attenuation and treatment infrastructure and to determine the effectiveness of existing stormwater management strategies (Vaze and Chiew, 2003a). Thus, stormwater models are increasingly being relied on as an aid to inform solutions to stormwater quality problems in the urban environment (Obropta and Kardos, 2007). They are an indispensable prediction tool when optimising mitigation and management measures for waterways

* Corresponding author.
E-mail address: tom.cochrane@canterbury.ac.nz (T.A. Cochrane).

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protection (Egodawatta and Goonetilleke, 2008; Vaze and Chiew, 2003a). Additionally, stormwater models can be integrated into a stormwater quality monitoring campaign which subsequently can continue providing information on the analysed system after the monitoring campaign is concluded (Birch et al., 2013). Typically, either "process-based" stormwater models (Egodawatta et al., 2009; Huber and Dickinson, 1988; Wang et al., 2011; Wicke et al., 2010) or regression models (Driver and Tasker, 1990; Irish et al., 1998; Jewell and Adrian, 1982) are used to estimate pollutant loads in stormwater runoff (Vaze and Chiew, 2003a). Process-based stormwater models simulate pollutant build-up and the subsequent pollutant wash-off as two distinct processes (Vaze and Chiew, 2003a). Whereby pollutant build-up describes the accumulation of pollutants on an impermeable surface during antecedent dry days, which can expressed as a linear, exponential, power, or another function of time (Kim et al., 2006). Pollutant wash-off describes the removal of pollutants by the shear stress generated by surface runoff flow and the energy imparted by the falling precipitation (Vaze and Chiew, 2002).

Regression models fit a line to the average relationship between a dependent variable and one or more independent variables, assuming that the variation in the dependent variable can be explained scientifically by quantitative changes in the independent variables that govern the process (Irish et al., 1998). In stormwater modelling, the regression equation combines the variables controlling pollutant build-up and pollutant wash-off processes into a single unified equation. Regression models are a quick and relatively friendly tool for predicting stormwater quality outputs and they can be customised to different situations. For example, independent models for individual pollutants and for different land-use conditions can be developed based on monitoring data for those conditions. A particular advantage of regression modelling is that the uncertainty associated with the inputted variables is also integrated in the model (Zoppou, 2001). In addition, regression models are a useful tool in identifying pollutant specific casual variables (Irish et al., 1998), i.e. determining which independent variable(s) influence specific pollutants loads. However, regression models, like all stormwater quality models, have difficulties in accurately predicting stormwater quality because modelling water quality involves many highly uncertain variables. This explains why stormwater quality models are inferior to stormwater quantity models (Obropta and Kardos, 2007). For example, any advancement of stormwater quality models is hindered by the complexity and dynamic nature of stormwater pollution (Beck and Birch, 2013). Other deficiencies in stormwater quality modelling are: incomplete water quality data on complete storms (Deletic and Maksimovic, 1998); lack of fully reliable water quality data due to low accuracy in measurement; different monitoring quality assurance methods; and deficient frequency of data collection, i.e. the time between collecting samples is inadequate to correctly record extreme storms (Deletic and Maksimovic, 1998).

Atmospheric deposition, which is defined as the returning of atmospheric gases and particles to the earth's surface, has been acknowledged as an important source of heavy metals to urban runoff (Gunawardena et al., 2013; Huston et al., 2009; Murphy et al., 2014; Sabin et al., 2005, 2006; Wicke et al., 2012). Atmospheric deposition occurs in two processes: wet or dry deposition. Wet deposition occurs when particles leach from the atmosphere with water droplets in the form of rain, snow, fog, mist, dew, and frost (Göbel et al., 2007). Dry deposition is the direct settling of particles and gases onto land or water surfaces (Azimi et al., 2003). Airborne particulates deposit onto impervious surfaces during the dry period and are subsequently transported to waterways as urban runoff following precipitation events (Burian et al., 2001). Davis and Birch (2011) found that atmospheric deposition accounted for 12%, 33%, and 5% of Cu, Zn, and Pb in urban stormwater in Sydney, Australia. However, few studies have modelled the effects of atmospheric deposition as a source of heavy metals in stormwater pollution; therefore, many uncertainties and challenges remain with managing these airborne pollutants in runoff. Wicke et al. (2010) modelled airborne pollutants in stormwater runoff using process-based models. Antecedent dry period was used as the defining variable for pollutant build-up while runoff rate explained pollutant wash-off. The model was found to replicate the experimental values well and was very informative for determining the controls that affect airborne pollutant build-up and wash-off. However, pollutants were washed-off by simulated rainfall which does not take into account the variability throughout a rain event (e.g. rain intensity), nor does it take wet deposition into account. As wet deposition is considered an important component of bulk deposition (Morselli et al., 2003), measuring both wet and dry deposition is crucial in order to model catchment-wide atmospheric deposition loads in runoff (Davis and Birch, 2011).

The objective of this research was to develop an event-process driven stormwater quality model that can estimate bulk atmospheric deposition loads for total Cu, total Zn, total Pb, and TSS from three different locations within an urban area: residential, industrial, and airside (within an airport grounds). As stormwater runoff was collected from modular concrete boards, the factors that confound typical stormwater models, such as sewer sediment transport and microbial degradation, were not present. Therefore, a simplified model on bulk deposition loads in stormwater was generated. In addition, this research aimed to identify the casual variables influencing airborne pollutant build-up and wash-off in stormwater runoff. Knowledge of the mechanisms of pollutant build-up and wash-off is an essential component of stormwater modelling (Nazahiyah et al., 2007). Improving the accuracy of stormwater modelling will lead to better understanding of local stormwater quality, and thus, increase the appropriateness of selected treatment systems (Liu et al., 2013).

2. Materials and methods

2.1. Study approach and study sites

Atmospheric total and dissolved Cu, Zn, Pb, and TSS loads in runoff were quantified from three different locations within Christchurch, New Zealand (Fig. 1) from February 2013 to December 2013. The areas studied were airside (Air), industrial (Ind), and residential (Res). The main characteristics of each sampling site are represented in Table 1. Air was within the grounds of Christchurch International Airport; Res was within a low housing density residential area and had a university nearby; Ind was within a light industrial area. Twelve impermeable concrete boards (four replicates for each sampling sites) were used to capture atmospheric pollutant build-up and wash-off from a conventional urban pavement section. Curbs and other features typically associated with the perimeter of a pavement were not represented as it went beyond the scope of this research. There were neighbouring buildings near the concrete boards at the Ind and Res research sites (detailed in Murphy et al. (2014)). The boards were near buildings because they better represented typical residential and industrial areas were open, uninterrupted space is uncommon. The concrete boards were elevated 450 mm above the ground and inclined to a 4° slope. A collection area, where runoff was solely collected from, was incorporated into the board design (Fig. 2). The collection area was designed to prevent pollutant loss from splash and spray. This assumed that any pollutants lost from the collection area via splash/spray equated to the pollutant gained from splash. Residual pollutants remaining (i.e. the fixed load) on the concrete boards were removed via brushing (Vaze and Chiew, 2002) and flushing with deionised water after each rain event. A polycarbonate stormwater collection funnel was used to collect runoff from the defined area and quickly convey it to a 20 L High Density Polyethylene (HDPE) collection chamber. Runoff samples were collected from the collection chamber within 24 h of the rain event finishing. All runoff samples were homogenised in-situ before sub-sampling into head-space free HDPE sampling bottles. To remove