THE SPATIAL AND TEMPORAL VARIABILITY OF AIRBORNE POLLUTANTS IN STORMWATER RUNOFF

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ABSTRACT

Atmospheric deposition is increasingly being recognised as a significant source of total suspended solids (TSS) and heavy metals in urban runoff. However, many uncertainties and challenges remain with measuring and managing these pollutants in runoff. Impermeable concrete boards were deployed in a residential, industrial, and airside land-use area in Christchurch for almost one year in 2013 to determine the spatial and temporal variability of airborne pollutant loads (principally TSS, Cu, Pb, and Zn) in runoff. Results showed that each land-use area displayed similar trends of increasing/decreasing pollutant loads throughout the monitoring period, suggesting that the pollutants originated from a similar source. Consistently higher pollutant loads were found for the industrial area, which was attributed to local topographic conditions rather than land-use activity. All pollutants had a statistically significant relationship with antecedent dry days, illustrating its importance on pollutant build-up. Pollutants dominated by their particulate-phase were influenced by peak rainfall intensity, which was explained by the energy from an intense rainfall event dislodging more particulate pollutants; however, this relationship was weak. Dissolved-phased pollutants displayed a greater relationship to rain depth showing that the quantity of rain influences the dissolution of pollutants from a surface.

KEYWORDS

Atmospheric Deposition, Stormwater, Metals, TSS, Spatial and Temporal Variability

PRESENTER PROFILE

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1 INTRODUCTION

Urban development leads to increased impervious landscapes, which interrupts the hydrological cycle by creating an impermeable barrier to natural infiltration of precipitation (Lindh, 1972). Precipitate, unable to infiltrate, flows over impermeable surfaces as sheet runoff, carrying the pollutants from the land with it; thus, compromising the quality of the stormwater. This highly polluted runoff is redirected (frequently untreated) to nearby waterways altering their water quality and quantity, and thereby, adversely affecting receiving aquatic ecosystems (Göbel *et al.*, 2007).

Stormwater signatures typically comprise of suspended sediments from building and pavement weathering; heavy metals from weathered building materials, wear and tear from vehicle components; hydrocarbons from industrial and vehicle emissions; and nutrients from excessive fertiliser usage on vegetation (Davis *et al.*, 2010a). In New Zealand, total suspended sediments (TSS) and heavy metals (in particular Cu, Pb, and Zn) are of greatest concern due to their abundance in stormwater signatures and their pernicious effects on aquatic ecosystems (Auckland Regional Council, 2003, Christchurch City Council, 2003).

Building (i.e. roofs and sidings) and road runoff are deemed the major *direct* sources of heavy metals in urban runoff. Roofing materials, such as rolled Cu and Zn, are frequently used worldwide as they are considered to be relatively "maintenance-free", durable and adaptable to many different design styles (He *et al.*, 2001). However, these roofing materials are subject to natural atmospheric corrosion processes and can be a significant source of heavy metal pollution (Rocher *et al.*, 2004, Pennington and Webster-Brown, 2008, Karlén *et al.*, 2001). Additionally, building sidings can contribute substantial quantities of heavy metals to stormwater, for example, contributing 22%, 59%, and 79% of Cu, Zn, and Pb to urban residential runoff as monitored in Maryland, USA (Davis *et al.*, 2001). Road infrastructure can contribute approximately 35-75% of the heavy metals in stormwater runoff, although they only encompass approximately 10-20% of an urban catchment (Pandey *et al.*, 2005). Metal pollutants in road runoff originate from tyre wear, brake lining, exhaust fumes, road construction, and resuspension of road dust (Sternbeck *et al.*, 2002, Beasley and Kneale, 2002). However, the majority of traffic-derived pollutants (80-95%) are not deposited directly onto the road surface, but are carried away by atmospheric and mechanical motion to elsewhere in the local environs (Göbel *et al.*, 2007). Therefore, atmospheric deposition (i.e. the returning of elements in the atmosphere to the earth's surface) can be a significant indirect source of heavy metals and TSS in urban runoff (Hu and Balasubramanian, 2003). For example, for the semi-arid catchment of Los Angeles, USA, atmospheric deposition is reported to contribute 57% to 100% of the metal loadings in stormwater (Sabin *et al.*, 2005).

The quantities of atmospherically-derived pollutants in stormwater runoff are influenced by (1) land-use area, (2) metrological conditions, and (3) pavement type (Kim *et al.*, 2002, Gunawardena *et al.*, 2011, Wicke *et al.*, 2012b). Land-use activity can impose a strong influence on airborne pollutant concentrations in stormwater runoff as different activities release varying quantities of heavy metals into the atmosphere (Kim *et al.*, 2002). For instance, metal fluxes were three to four times greater in urban and industrial areas in comparison to rural sites over 10 km away (Rossini *et al.*, 2005). Additionally, the topography of an area is an important factor affecting pollutant deposition. Deposition is increased on a windward slope (on a hillside) in comparison to the baseline (flat terrain) conditions (Goossens, 1988, Goossens, 1996). Conversely, deposition is significantly reduced from the baseline condition all over the leeward face of a slope (Goossens, 1996).

The relationship between meteorological conditions and stormwater runoff are commonly expressed as a two-stage process: pollutant build-up and pollutant wash-off (Vaze and Chiew, 2002). Pollutant build-up describes how pollutants accumulate on a surface during antecedent dry days (ADD); pollutant wash-off describes pollutant removal by a precipitate event (Vaze and Chiew, 2002). However, the relationship between different metrological conditions on pollutant build-up and pollutant wash-off is widely debated in the literature, as illustrated in Table 1.

Table 1: Relationships found between meteorological characteristics (ADD – antecedent dry days, RD – rain depth, RI – rain intensity, Vol – rainfall volume, Dur – rainfall duration) and runoff quality.

		Pollutant studied	**ADD**	**RD**	**RI**	**Vol**	**Dur**
Highway pollutants	Berhanu Desta *et al.* (2007)	TSS			x		
	Crabtree *et al.* (2006)	Metals			x		
	Gan *et al.* (2008)	Metals, COD, OP, & O&G	x	x			
	Hewitt and Rashed (1992)	Pb	x				
	Vaze and Chiew (2002)	Dry deposition	x				
Airborne pollutants	Gunawardena *et al.* (2011)	PM^1	x				
	Gunawardena *et al.* (2011)	PM^2		x			
	Hu and Balasubramanian (2003)	Metals				x	
	Rocher *et al.* (2004)	Metals	x	x			
	Wicke *et al.* (2012a)	Metals & TSS	x				

^{1}PM in dry deposition, ^{2}PM in total (dry and wet) deposition. Where, COD = chemical oxygen demand, OP = ortho-phosphorus, O&G = oils & greases, PM = particulate matter

Pollutant loadings in urban runoff can also be affected by pavement type. For instance, pollutants on a coarse asphalt surface are less likely to be mobilised due to the deeper cavities and greater adhesion of the material compared to a smooth concrete surface that enables pollutants to be more easily dislodged during a rain event (Wicke *et al.*, 2012b). On the other hand, a concrete surface will show some potential for Cu retention (Bahar *et al.*, 2008). Even over a distance of 0.61 m on a concrete surface, dissolved Cu was reduced by 7%, 8%, and 13% at a flow rate of 0.75 L/min, 1.5 L/min, and 0.325 L/min, respectively (Perkins *et al.*, 2005). This is due to the pH and alkalinity (bicarbonate) of stormwater runoff increasing as it contacts a concrete surface (Davis *et al.*, 2010b), which subsequently reduces the bioavailable form of Cu (Bahar *et al.*, 2008).

With a myriad of factors influencing airborne pollutant loadings in stormwater runoff, adequate knowledge on the dynamics of airborne pollutant build-up and wash-off is lacking. In particular, there is a dearth of knowledge regarding how spatial and temporal variations influence atmospheric pollutant loads in stormwater runoff. Therefore, research was conducted to improve the understanding of spatial and temporal variability of airborne pollutants in stormwater runoff from different land-use areas, specifically in Christchurch.

2 METHODOLOGY

To simulate atmospheric pollutant wash-off from a typical urban surface, stormwater runoff was collected from impervious concrete boards (surface area = 1 m^2). The concrete boards were deployed in different land-use areas (industrial, residential and airside of an airport) throughout Christchurch, New Zealand (Figure 1), from February to December 2013. Each area had four replicate boards elevated 450 mm above ground at a 4° slope. A collection area (718 x 400 mm), i.e. an area was runoff was solely collected from, was incorporated into the board design to minimise pollutant loss via splash and spray, as exemplified in Figure 2. This assumed that the splash and spray leaving the collection area equaled the splash and spray entering into it from the remainder of the board, which was adequately confirmed from preliminary experiments.

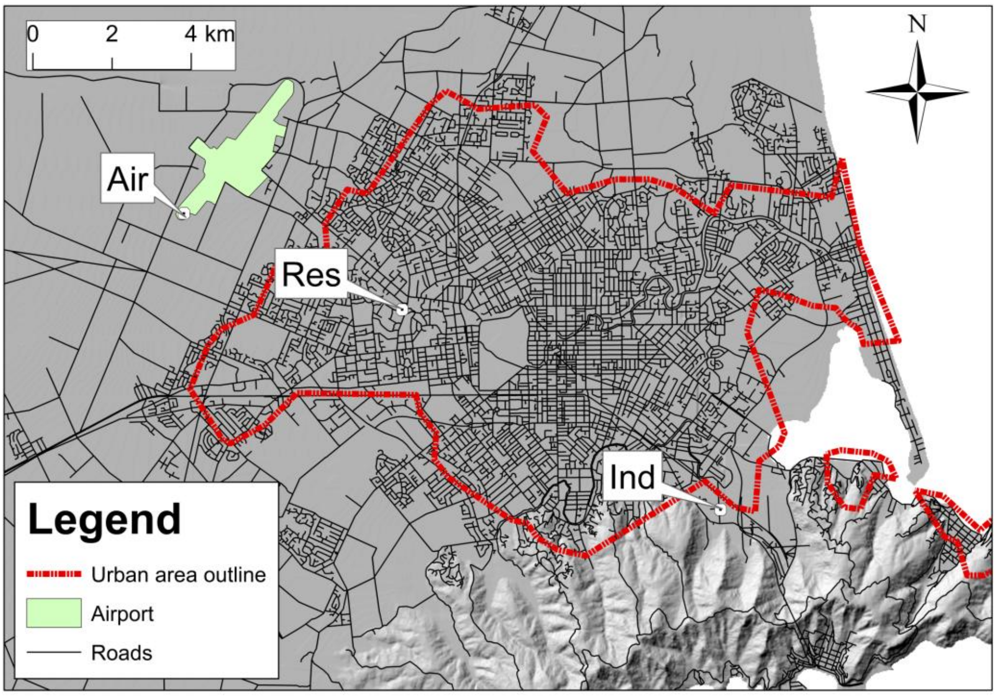

Figure 1: Air - airside (S43.501055 E172.520372), Res - residential (S43.523230 E172.588347), and Ind - industrial (S43.569078 E172.687519) monitoring sites in Christchurch.

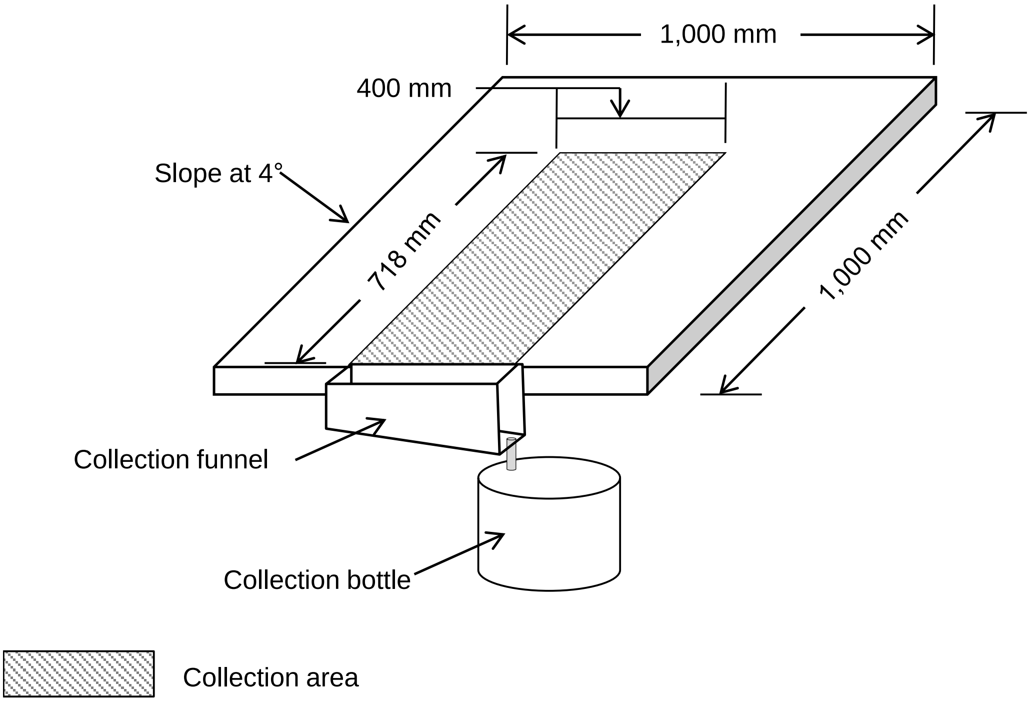

Figure 2: Experimental setup of the concrete boards and the stormwater collection system.

Runoff from each board was analysed for TSS, total Cu, total Pb, and total Zn in accordance with the SM2540D and SM3125-B guidelines (APHA, 2005). Total recoverable metals were digested using the methodology described in Wicke *et al.* (2012a) and Good *et al.* (2012). When runoff volumes were insufficient for TSS measurements, turbidity was used as a surrogate via a TSS-turbidity curve relationship established for each sampling area. Additionally, different metrological conditions (rainfall parameters, antecedent dry days, and wind patterns) were monitored for each site (Table 2).

All statistical analyses (i.e. bivariate Pearson's correlation, partial correlation, and analysis of variance) were conducted using the IBM SPSS Statistics 20 package. When required, variables were natural-log transformed to conform with the assumptions of normality and linearity (Field, 2013).

Table 2: Information regarding the collection of weather data.

	Parameter	**Weather station**	**Instrument type**	**Accuracy**	**Distance from sampling area**
Airside	Rainfall	Local rain gauge	Davis tipping bucket	± 4% (when intensity <50 mm/h) ± 5% (when intensity >50 mm/h)	On-site
	Wind	Local wind sensor	RainWise Wind-Log	Speed: ± 2% Direction: ± 22.5°	On-site
Industrial	Rainfall	Local rain gauge	Davis tipping bucket	± 4% (when intensity <50 mm/h) ± 5% (when intensity >50 mm/h)	On-site
	Wind	Environment Canterbury air monitoring station	Vector Instruments W200P & A100M	Speed: ± 1% Direction: ± 3°	1.44 km
Residential	Rainfall	University of Canterbury weather station	Rain-O-Matic Professional	± 2%	0.45 km
	Wind	NIWA weather station	Vector Instruments W200P & A100M	Speed: ± 1% Direction: ± 3°	1.77 km

3 RESULTS AND DISCUSSION

3.1 SPATIAL VARIABILITY OF AIRBORNE POLLUTANTS IN STORMWATER RUNOFF

The variability of Cu, Pb, Zn, and TSS measured in all land-use areas displayed similar trends of increasing and decreasing pollutant loads over time (Figure 3). Additionally, the airside and residential areas displayed significantly similar pollutant loadings throughout the sampling period. Therefore, the similar temporal trends suggest there is a homogeneous distribution of atmospherically-deposited pollutants within the greater Christchurch airshed. Studies on PM_{10} (PM<10 μm diameter) and carbon monoxide distributions within the Christchurch airshed found contradictory results, i.e., no uniform distributions at an intra-urban scale (Wilson *et al.*, 2006, Kossmann and Sturman, 2004). However, these studies quantified fine particle pollutants in the troposphere rather than pollutants deposited on impermeable surfaces during antecedent dry days, like in this research. The homogenous distribution of TSS and metal loadings within the Christchurch airshed was likely due to these pollutants being associated with larger particle sizes (>10 µm).

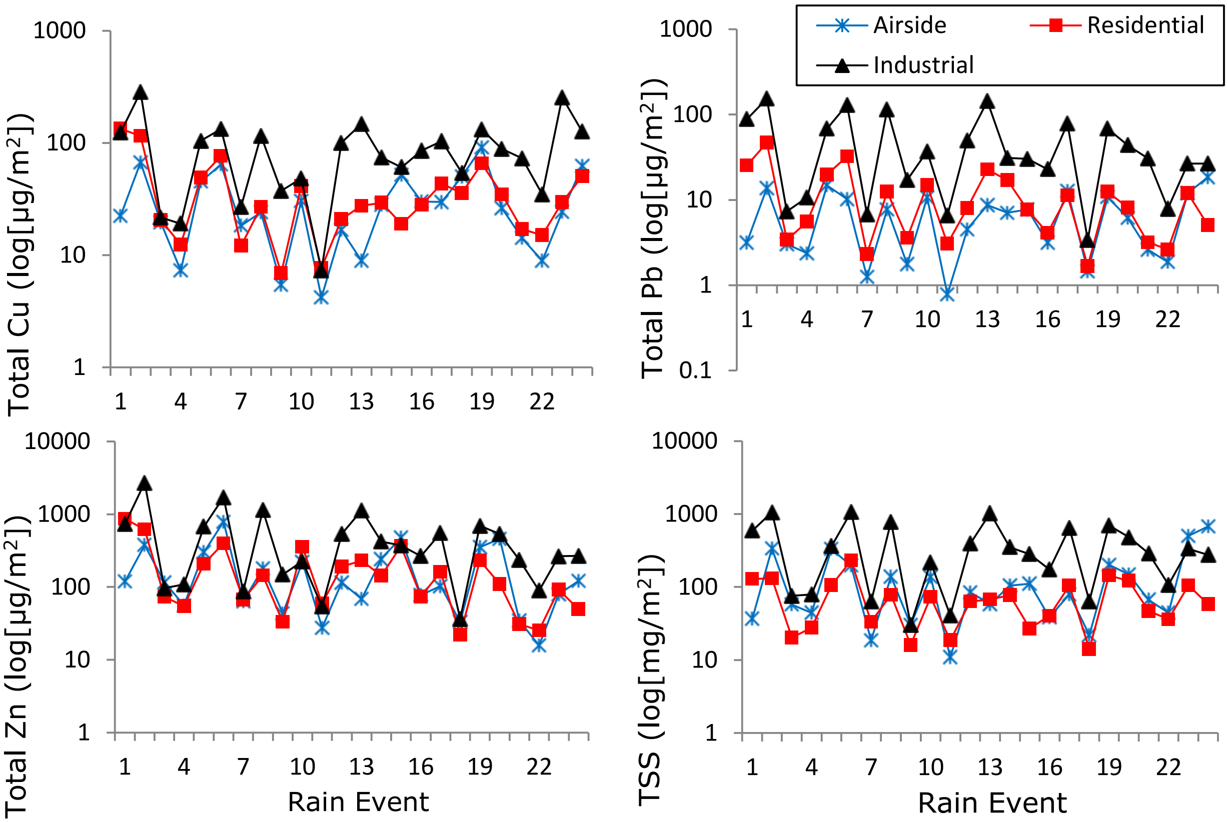

Figure 3: TSS, total Cu, Pb, and Zn loadings in stormwater runoff from 24 precipitation events that simultaneously occurred in all three land-use areas.

To ascertain whether the homogenous distribution of airborne TSS and metals originated from a similar atmospheric source, atmospheric pollutant ratios were generated for all land-use sites throughout the monitoring period (the ratios of Zn and Pb to Cu are exemplified in Table 3). The ratios were comparable between all land-use areas, suggesting that the pollutants originated from a single source. Moreover, land-use area was not an important factor for regulating pollutant loadings in runoff because the atmospheric pollutant ratios and the pollutant trends were analogous between the three research sites. Although the industrial area had consistently higher pollutant loadings compared to the other areas, this was not attributed to local land-use activities, but rather due to local topographic conditions influencing the rate of deposition. As the industrial study area was located next to a hill range (Port Hills), greater deposition rates were expected to occur during a northerly and easterly wind (the predominant wind) when the study area was located on the windward side of a slope. The residential and airside areas were both located on flat terrain; therefore, explaining the similar pollutant loadings between those areas.

Table 3: The ratio of pollutant build-up (µg/m^2/d) for Zn and Pb to Cu (mean ± SE). Within each group, those sharing a common letter are significantly similar ($p > 0.05$).

Ratio to Cu	**Industrial**	**Residential**	**Airside**
Cu	1	1	1
Pb	0.54 ± 0.036^b	0.34 ± 0.029^a	0.25 ± 0.014^a
Zn	5.39 ± 0.37^a	4.98 ± 0.43^a	5.58 + 0.51^a

This study found that, irrespective of land-use activity, atmospheric TSS and metal pollutants originated from a similar source. It is typically considered that vehicular activity is the dominant source of Cu, Pb, and Zn in the atmosphere (Hjortenkrans *et al.*, 2007, Chu-Fang *et al.*, 2005, Sternbeck *et al.*, 2002). However, associating the source of pollutants with vehicular activity, in this instance, was questionable. As traffic behaviours were different for each site, i.e. different traffic volumes, different braking conditions, different speed limits, it was reasonable to assume that there would be a greater variance between the pollutant trends and pollutant ratios to Cu. However, this was not observed in this study. Additionally, as different road usage patterns occurred in each research area, varying trends of metals would likely have occurred between the areas since Cu and Pb are related to traffic congestion, and Zn is related to traffic volume (Gunawardena *et al.*, 2013). Instead it is more likely that another source of metals was responsible for metal loads found in this study, e.g. the long-range transport of metal pollution from Australia to New Zealand (Marx *et al.*, 2008, Marx *et al.*, 2014).

3.2 TEMPORAL VARIABILITY OF AIRBORNE POLLUTANTS IN STORMWATER RUNOFF

Varying metrological conditions exert an important control over pollutant build-up and pollutant wash-off. Antecedent dry days were found to significantly influence pollutant build-up in all land-use areas. Figure 4 illustrates increasing pollutant loadings when antecedent dry days increases for the industrial area only (the industrial area was chosen as a representative for the other land-use areas studied). This result is similar to the findings by Gunawardena *et al.* (2011) and Wicke *et al.* (2012a). They found that pollutant build-up increases asymptotically with antecedent dry days and ultimately plateauing after 6 days. This asymptotic trend was not discerned in this study due to confounding factors (i.e., varying RD).

The relationship between rainfall characteristics and pollutant wash-off appears to be dependent on the pollutant of concern (Figures 4 and 5). For the airside area, Cu and Zn displayed a significant relationship with rain depth; total Pb and TSS showed a significant relationship with peak hourly rainfall intensity. This suggests that rain intensity had a significant relationship with pollutants that were associated with the particulate phase. This is explained by a greater quantity of particulates mobilising from an impermeable surface during high energy storm events. Rain depth had a greater influence on pollutants that had a high fraction in the dissolved phase (approximately 50% of total Cu and Zn were in the dissolved phase) as more pollutants dissociate from a surface with increasing volume of precipitate. However, no statistically significant relationships between rainfall characteristics and pollutant loadings were found for the industrial and residential area (perhaps due to other factors, i.e. antecedent dry days, confounding the data), except for TSS, which exhibited a significant relationship with rain intensity in the residential area. Although not statistically significant, rain intensity appeared to exert a greater influence on total Pb and TSS wash-off rather than rain depth, while rain depth illustrated a greater influence on total Cu and Zn. Figure 5 illustrates increasing Cu and

Zn with increasing rain depth, and increasing Pb and TSS with increasing rain intensity for the industrial research area only (a representative for the other research areas).

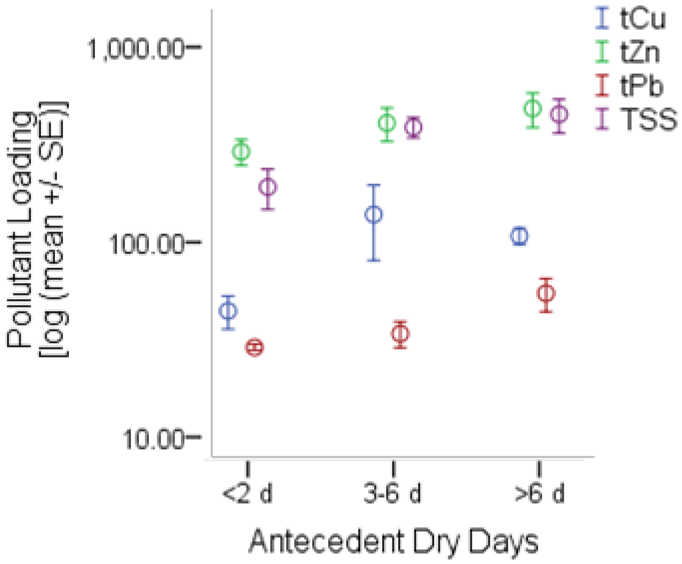

Figure 4: Varying pollutant loadings (Cu, Pb, and Zn in μg/m^2 and TSS in mg/m^2) with increasing antecedent dry days during medium rain intensity events (1-3 mm/h) for the industrial research area.

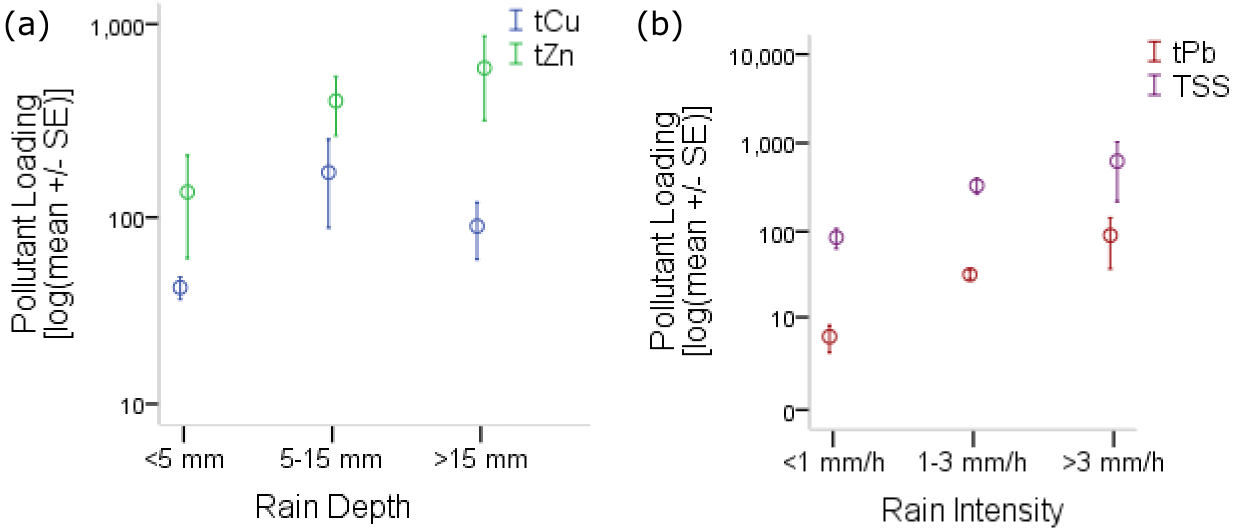

Figure 5: (a) Total Cu and total Zn loadings (μg/m^2) versus rain depth; (b) total Pb (μg/m^2) and TSS loadings (mg/m^2) versus rain intensity when antecedent dry days are constant (3-6 d) for the industrial research area.

4 ON-GOING WORK

On-going research is addressing the influence of pavement type (concrete versus asphalt) on atmospheric pollutant loadings in stormwater. Sets of impermeable asphalt, permeable asphalt, impermeable concrete, and permeable concrete were deployed to the residential sampling area for this purpose. Runoff is monitored for TSS, total and dissolved Cu, Pb, and Zn. The influence of the pavement composition and surface roughness is being accessed to determine how it affects airborne pollutant wash-off throughout 2014.

5 CONCLUSIONS

Accurate knowledge of the dynamics of airborne pollutant wash-off is an essential component of stormwater modelling. Improving the precision of stormwater models will lead to a better comprehension of local stormwater quality, and thus, increasing the effectiveness of stormwater abatement systems (Liu *et al.*, 2013). This research demonstrated that land-use activities do not necessarily exhibit a strong influence on total Cu, Zn, and Pb deposition in the Christchurch airshed, suggesting a similar pollution source was likely to be dominating metal deposition. However, the topographic features of an area were found to greatly influence the rate of deposition. It is recommended that a source appointment study on atmospheric metal pollution is conducted for the Christchurch airshed to help address the poorly understood distribution of airborne metals within the city-scale.

Antecedent dry days were found to exert the most influence on pollutant loads in stormwater runoff. Rainfall intensity and rainfall depth may also have an influence, although this is dependent on the speciation phase of the pollutant. Particulate pollutants are likely to be controlled by rainfall intensity as more particulates have the potential to be mobilised from a surface. Rain depth seems to influence pollutants that have a high proportion in their dissolved phase because more pollutants have the ability to dissociate from a surface.

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