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Carpark pollutant yields from first flush stormwater runoff

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ABSTRACT

Stormwater runoff from carparks should be treated to remove pollutants before they enter urban waterways; however, differences in traffic characteristics and surrounding land use activities can result in varying first flush pollutant types and concentrations requiring specific treatment approaches. An understanding of potential variations in first flush pollutant characteristics from carparks is necessary to design adequate treatment systems. Stormwater runoff from over 20 runoff events in three different carparks (university, hospital and industrial) in Christchurch, New Zealand were thus analyzed for Total Suspended Solids (TSS) and heavy metals (Zn, Cu and Pb) over a year. Pollutant concentrations were found to vary across the carparks, which were largely driven by land use activities such as traffic count, size of the vehicles, and surrounding topography. Mean concentrations of heavy metals (both dissolved and particulate) and TSS were significantly higher in the industrial carpark than in the other two urban carparks, which had statistically similar mean pollutant concentrations. Specific ratios of metal species (Zn:Cu, and Zn:Pb) were relatively high for the industrial carpark, indicating a greater contribution from the wear and tear of large commercial vehicles. TSS and total Zn wash-off concentrations from the hospital carpark when it was non-operational (passive) were found to be significantly lower than the hospital carpark when it was operational (active), confirming that pollutant concentrations increase with vehicular activity. Rainfall characteristics such as antecedent dry days and rain intensity and duration were found to only have a low positive correlation to pollutant concentrations for all carparks. The findings from this study highlight the importance of considering carpark characteristics in the implementation of on-site stormwater treatment systems.

1. Introduction

Source-based urban stormwater quality evaluation has become a fundamental step in the management of water quality of receiving water bodies (Collins et al., 2010). Urban carparks represent a major source of stormwater pollutants such as Total Suspended Solids (TSS), heavy metals (*e.g.* Zn, Cu and Pb), anthropogenic organic compounds, nutrients and pathogens (Göbel et al., 2007; Xu et al., 2020). Runoff from carparks is therefore considered to be a significant source of pollutants to local receiving waterways affecting aquatic life (Tiefenthaler and Schiff 2001; Cochrane et al., 2011; Brown and Peake, 2006; Reddy et al., 2014 Wang et al., 2013). The quality of carpark runoff can be improved through the installation of on-site stormwater treatment systems, which require a smaller footprint and a lower maintenance cost as compared to conventional best management practices such as rain gardens or wetlands. Hence, it is important to better understand the initial higher concentration of pollutants in order to design or select the most appropriate on-site treatment system.

Vehicles are the primary source of metal pollutants in carparks and the majority of suspended solids picked up by stormwater runoff are related to traffic, surrounding land use, and atmospheric contamination, as influenced by rainfall and environmental conditions (Valtanen et al., 2014; Huber et al., 2016). Traffic patterns such as speed and vehicular movement, vehicle count and type (car, van or truck), maintenance activities (washing/cleaning of vehicles) and institutional regulations (such as posted speed sign, parking time limits, etc.) influence the accumulation of TSS and heavy metals on impervious surfaces (Davis et al., 2001; Councell et al., 2004). Heavy metals from vehicles originate from brake pad wear products (Cu), combustion exhaust, galvanized parts and railings, fuel and oil, wear of plating, bearings and bushings and other moving parts (Cu), wire corrosion, brake lining (Pb, Cu and Zn), and radiator fluid (Cu) (Amrhein and Strong, 1990; Amrhein et al., 1992; Glenn, 2001). Pollutants from vehicular traffic, atmospheric deposition and from pavement wear accumulate on carparks over time and subsequently are washed-off during storm events. The accumulation of these pollutants is influenced by atmospheric deposition, surrounding topography and the size of the drainage area (Lau et al., 2009). In addition, intentional and accidental spills have also been identified as major sources of pollutants (Christensen and Guinn, 1979; Davis et al., 2001).

Pollutant concentrations are usually higher during the initial period of a storm as compared to later periods (Lee et al., 2002). First flush has been identified as one of the primary causes of deterioration of the

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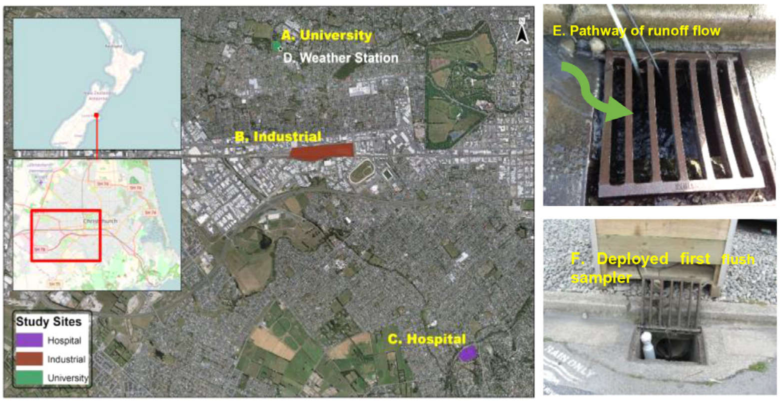

Fig. 1. Sampling sites: A. University carpark, B. Industrial carpark, C. Hospital carpark. D. weather station (all weather data were retrieved from Campbell weather station situated at University of Canterbury), E. pathway of runoff flow and F. deployed first flush sampler in the sump.

quality of urban waterways associated with impervious areas such as highways and carparks (Barco et al., 2008). Quantifying first flush pollutant concentrations is important since most stormwater treatment systems need to be designed to treat this initial portion of runoff events (Deng et al., 2005). Build-up and wash-off mechanisms influence first flush stormwater quality in urban carparks. Previous studies showed that carpark stormwater samples collected during the first 10 min of a rain event contained the highest pollutant concentrations, indicating the presence of a first flush phenomenon (Schiff et al., 2016).

Most stormwater quality studies have only considered large catchment areas (*i.e.* over 100 ha), long precipitation durations and moderate to high-intensity rainfalls (> 5 mm/h) for the analysis of first flush pollutant concentrations (Bach et al., 2010; Sansalone et al., 1998). Stormwater quality behavior from urban carparks (almost 100% imperviousness) with a low-intensity rainfall climate in a similar geographical area is not well understood. Furthermore, most studies have focused on urban roads and highway runoff with a wider range of geographic coverage and rainfall conditions (Lee et al., 2002; Wang et al., 2013). The contribution of low-intensity rainfall on runoff quality from different carparks within the same geographic area and under similar rainfall characteristics is not well understood. There is a dearth of information regarding how carpark characteristics (especially traffic pattern, vehicle type, and size of carpark) influence runoff pollutant concentrations and metal partitioning (dissolved and particulate) under low-intensity rainfall climate (> 85% of storms event monitored had rainfall intensity lower than 5 mm/h).

The main objective of this research was thus to quantify potential differences in first flush TSS and heavy metals pollutant concentrations from three different carparks (hospital, university, and industrial) within the same geographical location and under similar low-intensity rainfall conditions. A secondary objective was to assess the influence of antecedent dry days, initial 10 min rainfall intensity and initial rain depth on the pollutant concentrations in first flush stormwater. Findings will help inform the selection of the most appropriate on-site stormwater treatment systems for individual carparks.

2. Materials and methods

2.1. Sampling sites

Three sampling sites were selected to assess the influence of vehicular traffic on the quality of urban carpark runoff. The sampling sites were carparks located at a hospital, a university and an industrial area in Christchurch, New Zealand (Fig. 1). The sites are representative of major carpark types (commercial, industrial and residential) in the city. There is minimal variability of rainfall across the three carparks as they are relatively close to each other. The major differences observed in these carparks are traffic and carpark characteristics. Geographically, the carparks were within a radius of 6 km, but the hospital carpark was located at the foothills of the Banks Peninsula whereas the other two carparks were located in the flat part of the city. Average daily traffic differs among the carparks, ranging from 600 to over 1000 vehicles a day. Carpark surfaces were mainly asphalt and their drainage area ranged between 1752 and 5036 m^2 (Table 1).

2.2. Sample collection and analysis procedure

Twenty storm events were sampled from September 2015 to October 2016. Due to logistic limitations, the first flush stormwater runoff from the industrial site was not sampled for events 1 and 2 while the hospital carpark was not sampled for event 11. At the hospital, 11 storm events were sampled from September 2015 to June 2016 when the carpark was operational (active carpark) and 9 storm events were sampled from July 2016 to October 2016 after when the carpark was non-operational (passive carpark).

Although the term first flush has been broadly used in the literature, for this study we define first flush as the raw runoff required to fill a standard 1-L sample bottle placed in a carpark sump. Nalgene™ stormwater first flush samplers (1-L HDPE bottles), which meet the US-AEPA grab sampling requirements, were used for runoff sampling. The samplers were placed into a sump or manhole that received runoff of the corresponding urban carpark. The first flush samplers were suspended from the sump grate with a plastic cable tie in the corner of the sump where the initial runoff would flow into the sampler Fig. 1E. First flush samplers were deployed prior to a storm event and left in place until after the storm Fig. 1F. When the sampling bottle was filled, a floating ball valve sealed off the sample collection port. This mechanism prevented the collected first flush sample from carpark surfaces from being mixed with and diluted by later runoff. It is important to note that the samplers capture a 1 L sample from the first flush runoff, but this is not limited to the first liter of runoff from the carpark. The 1 L samplers fill gradually as the stormwater runoff flows over them, thus capturing a representative sample of that "first flush". Previous studies in this same geographic region (an elsewhere around the world) have used these commercial first flush samplers successfully to demonstrate the first flush phenomenon from various impermeable urban surfaces (Charters et al., 2021). All first flush samples were transported immediately to the laboratory where they were stored at 4 °C until chemical analysis within 24 h of sampling. For TSS analysis, the samples were vacuum filtered with 1.2 µm filter paper disks which were then oven-dried at 105 °C for 1 h following Murphy et al. (2015). Total heavy metals (Zn, Cu and Pb) samples were preserved with concentrated HNO_3 (70% Fisher, trace analysis grade) to a pH < 2.0 (APHA, 2005). Total and dissolved metals samples were then prepared on the same day of each storm event. Dissolved metals were prepared by filtering the sample through a 0.45 µm syringe filter and then preserved with trace grade acid to pH ≤ 2 (APHA method 3125 B). Total metal digestions were prepared by mixing 5 mL of trace grade nitric acid with 25 mL of preserved sample into a

Table 1
Land use characteristics and estimated daily traffic from urban carparks.

Carpark	[1]Estimated daily traffic	Characteristics of vehicles and land use category	[2]Drainage area (m^2)
University	900 vehicles/day	Residential/institutional: private cars, occasionally trucks for loading/unloading	5036
Hospital	600 vehicles/day	Commercial/institutional: private cars and occasionally buses and trucks	1752
Industrial	> 1000 vehicles/day	Industrial: trucks (mainly 16-wheeler), vans and private cars	3042

[1] Total vehicle counts were estimated based on field observations at the hospital carpark and a traffic monitoring device at the university and industrial carparks.
[2] Drainage areas were estimated using ArcGIS 10.3.

50 ml polypropylene centrifuge. The mixture was boiled for one hour in a heating block at 105 °C. Then the cooled samples were filtered through a 0.45 µm syringe filter before ICP-MS analysis following Wicke et al. (2012) and Murphy et al. (2015). The detection limit for Zn, Cu and Pb was 0.1 μg/L. Quality assurance protocols including blanks, duplicates (at least 10% of samples), analytical standards, and instrument calibration were conducted on all occasions. Also, all first flush samplers were soaked in 10% HCl for 3 days, rinsed with deionized water and air dried before use to avoid any potential contamination (Charters et al., 2016). Following each sampling event, the first flush samplers were replaced with fresh acid-washed first flush samplers.

Weather data from a Campbell weather station situated at the University of Canterbury's Engineering building were used for the analysis. The weather station data were compared against meteorological records from the nearby National Institute of Water and Atmosphere's (NIWA) weather station to verify its accuracy. These two sites were located within a radius of 8 km. Rainfall data from both stations were found to be similar and therefore University's weather data, which was the closest to the carparks, were used for this research. Event average rainfall intensity, antecedent dry days (ADD), rain duration and total rain depth were monitored for all 20 storm events. The average (minimum-maximum) values were found to be: initial 10 min rainfall intensity = 3.5 (1.2–10.8) mm/h, ADD = 6 (0.25–20.18) days, rain duration = 3 (0.9–11.8) h and total rain depth = 5.4 (1–12.6) mm (Supplementary information: Table A).

2.3. Data analysis

Statistical analysis was conducted using IBM® SPSS® Statistics (version 23) software. Scatter and box plots were used for initial graphical inspection of the distribution patterns of runoff data. The distribution patterns of first flush runoff data were further confirmed with the Shapiro-Wilk test (Liang et al., 2009). Carpark pollutant concentrations were from independent observations (independent storm events), and thus non-parametric tests were selected. The Kruskal-Wallis test was performed to ascertain whether any difference in TSS and total metal concentrations between the three carparks was statistically significant or not. Since the data were not normally distributed and had an unequal sample size (*i.e.* 20 storm events from university, 18 from the industrial carpark, and 8 from active hospital carpark), mean ranks were compared instead of median values (Charters et al., 2016). The Kruskal-Wallis method ranks each data point for the dependent variable (*i.e.* the water quality parameter) irrespective of which carpark surface it is associated with Kruskal William and Wallis (1952) and Charters et al. (2016). To further identify which particular carpark differed significantly from the others, pairwise comparisons of the differences in mean rank were then performed using the Mann-Whitney U test with a Bonferonni adjustment (Grech and Calleja, 2018). Data were screened for outliers prior to the analysis and only one data point for total Pb at the hospital (passive) carpark (storm event 19) was found to be an extreme outlier and was removed from the dataset.

Relationships between total metals were assessed for each carpark using Pearson's correlation. Scatter plots were used for initial visual inspection to confirm the occurrence of a linear relationship. Metal to metal species ratios were calculated for each metal to infer wear and tear from smaller and heavy commercial vehicles for each carpark.

Total and dissolved metals concentrations were also compared for each carpark using Pearson's correlation. Percentage partitioning was calculated to assess the dominant phase of each metal (dissolved vs particulate). Pair wise comparisons of TSS and metals were used to identify if particular carparks differed significantly between the wet and dry seasons.

3. Results

3.1. Influence of rainfall characteristics on first-flush pollutant concentrations

The concentration of TSS and heavy metals from the three different urban carparks varied over the twenty storm events monitored (Supplementary Information: Fig. A). However, the industrial carpark consistently showed the highest pollutant concentrations over all storm events. Accordingly, TSS and Total Zn, Total Cu and Total Pb concentrations in the industrial carpark were significantly different from those in the university and hospital carparks (Table 2) which had statistically similar mean pollutant concentrations between them.

Pollutants in first flush runoff from the university and industrial carparks were influenced by rainfall characteristics. Low positive correlations were found between TSS and first 10 min initial rainfall intensity ($r = 0.49$, $p < 0.005$) and between TSS and initial 10 min rain depth ($r= 0.471$, $p < 0.005$) at the university carpark. At the industrial carpark, total Cu (TCu) was positively correlated with ADD ($r = 0.470$, $p < 0.005$) and initial 10 min rain depth ($r = 0.509$, $p < 0.005$). Total Pb (TPb) was also positively correlated with ADD ($r = 0.503$, $p < 0.005$). The Hospital carpark did not show any correlation with rainfall characteristics.

TSS mean concentrations were the highest during the wet season (June-Sep) at the university carpark whereas mean concentrations of heavy metals were the highest during the dry season (Oct-May) at the industrial carpark. There were no consistent trends observed for particular seasons at each carpark. The observed inconsistent pollutant distribution trends suggest that the season had no influence on carpark runoff quality. The hospital carpark was not included in this analysis since the hospital was not operational (passive) after the dry season.

3.2. Carpark characteristics and first-flush pollutant concentrations

3.2.1. Total suspended solids

TSS concentrations between carparks were significantly different from each other (X^2 (2) = 34, $p < 0.001$, (Kruskal-Wallis Test). In addition, a post hoc analysis for pair wise comparisons identified that TSS concentrations for all the carparks were statistically different except for the university and hospital active carpark (Table 2). TSS concentrations in first flush from the industrial carpark were higher than in the first flush from the two other carparks (Fig. 2). Mean and median TSS values for the hospital active carpark were found to be four times higher than for the passive hospital carpark (Table 3). The hospital carpark's vehicle activity, being non-operational after a dry period, had significant influence on reduction of TSS and heavy metals concentration.

Table 2
Pairwise comparison of TSS and total metals from carparks. A post hoc significant test was performed using Mann-Whitney *U* test.

Pairwise combination of car park types	Bonferonni adjustment			
	TSS	TZn	T Cu	TPb
University-hospital (active carpark)	0.215	0.646	0.13	0.23
University-hospital (passive carpark)	0.010*	0.02*	0.02*	0.18
University-industrial	< 0.001*	< 0.001*	< 0.001*	< 0.001*
Hospital (active carpark)-industrial	< 0.001*	< 0.001*	< 0.001*	< 0.001*
Hospital (active carpark)-hospital (passive carpark)	0.004*	= 0.05*	0.214	0.021*
Hospital (passive carpark)-industrial	< 0.001*	< 0.001*	< 0.001*	< 0.001*

*denotes a statistically significant result. The significance level is 0.05.

Table 3
Mean, median, range, and mean rank of TSS and total metals (Zn, Cu, Pb) concentrations from different carparks, and mean ranks from Kruskal-Wallis test.

Carpark type	TSS (mg/L) mean, median(range) mean rank	TZn (µg/L)	TCu (µg/L)	TPb (µg/L)
University	174, 154 (17 – 651) 22	401, 272 (55 – 2128) 22	50, 41 (12 – 157) 24	58, 14 (4 – 532) 21
Hospital (active)	237, 196 (64 – 476) 28	332, 236 (108 – 997) 20	34, 32 (21 – 51) 17	27, 25 (11 – 50) 24
Hospital (passive)	63, 62 (6 – 120) 9	151, 126 (41 – 300) 11	28, 26 (14 – 58) 13	16, 11 (2 – 42) 12
Industrial	781, 657 (185 – 3002) 46	2716, 2035 (761 – 8277) 46	178, 153 (56 – 390) 45	171, 137 (36 – 359) 44

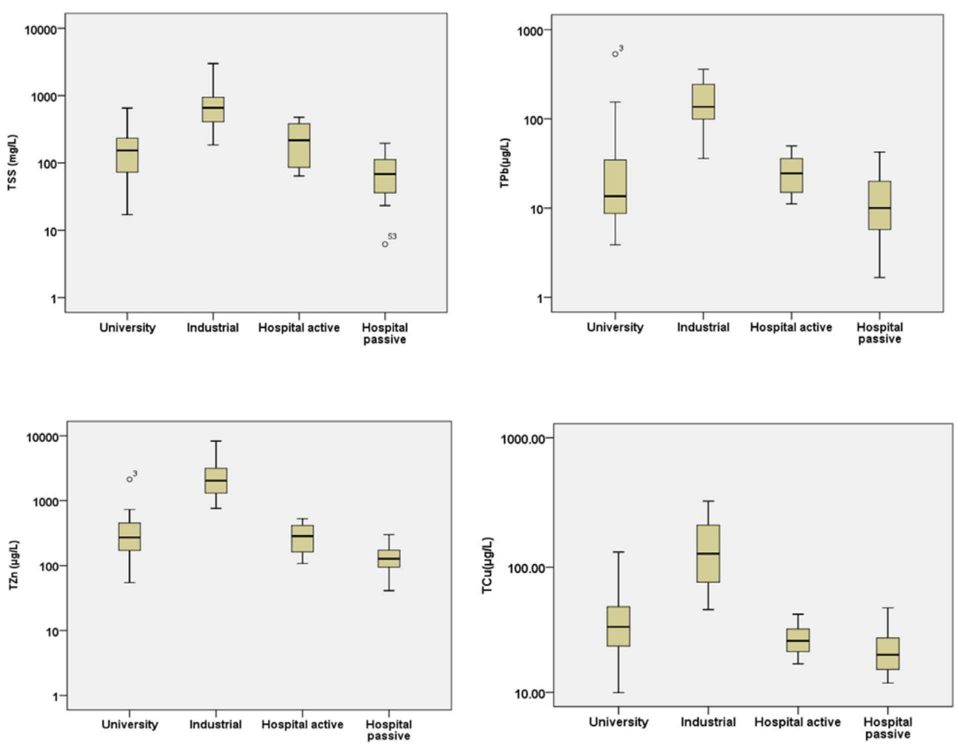

Fig. 2. First flush pollutant concentration at each carpark (denotes outliers' ± 1.5x Inter Quartile Range).

3.2.2. Total heavy metals (TZn, TCu and TPb)

The carparks were found to be significantly different from each other according to the Kruskal-Wallis analysis of their metals concentrations (TZn (X^2 (3) = 36, $p < 0.001$), TCu (X^2 (3) = 35, $p < 0.001$) and TPb (X^2 (3) = 29, $p < 0.001$)). A post hoc analysis for pair wise comparison identified that TZn, TCu and TPb concentrations for each carpark were statistically different except for the university and hospital active carpark (Table 2). TZn and TPb concentrations at the hospital active and hospital passive carparks were statistically different, but the TCu concentrations were statistically similar.

The highest TZn concentrations were observed at the industrial carpark (mean and maximum of 2716 µg/L and 8277 µg/L). TZn concentrations from the industrial carpark runoff was generally at least one order of magnitude higher than those from the other carparks studied (Fig. 2). The hospital and the university carparks had similar mean concentrations despite differences in nature and numbers of vehicles. TZn concentrations were reduced by half following the hospital carpark shut down. This clearly shows that metal concentrations decreased with lower vehicular activity; however, it took a significant amount of time to wash off all metals from carpark surfaces (Table 3). Similarly, the industrial carpark produced the highest concentrations of TCu and TPb

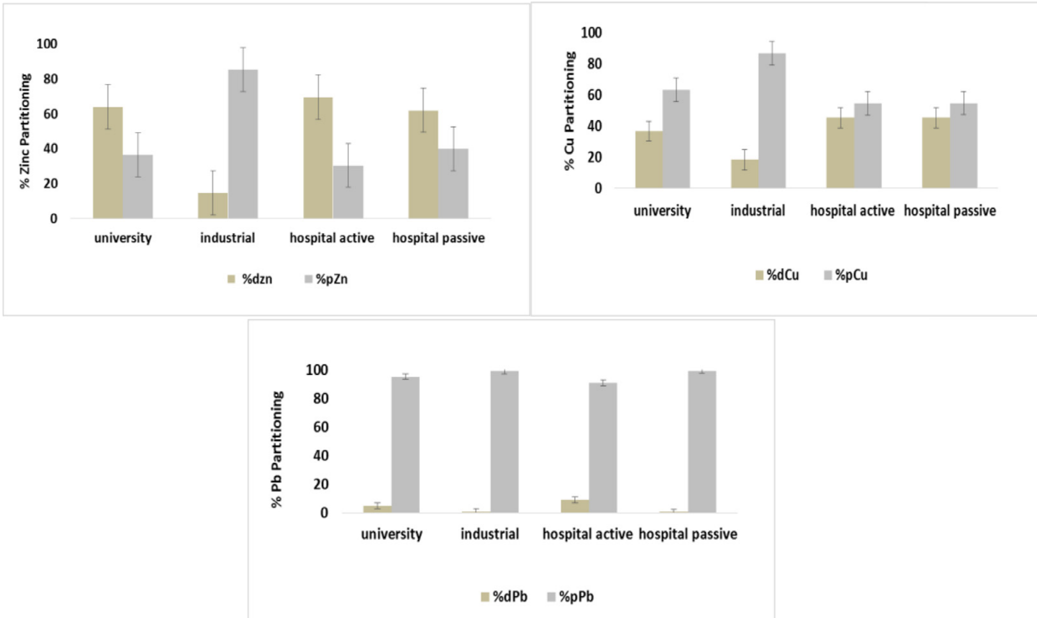

Fig. 3. Dissolved (dZn, dCu, and dPb) and particulate (pZn, pCu, and pPb) metal partitioning for each carpark studied, + / - 1 standard deviation.

(mean and maximum: 178 μg/L, 390 μg/L and 171 μg/L, 351 μg/L, respectively). TCu in that carpark was one order of magnitude higher than in the university and hospital carparks. Surprisingly, the hospital active and passive carpark had similar mean and max TCu values.

3.2.3. Relationship between metal concentrations

In each carpark, linear relationships were observed between each metal (TZn and TCu, TCu and TPb, and TPb and TZn) with the exception of the hospital (both active and passive) carpark (Supplementary information: Fig. B). Pearson's correlation analysis was performed to confirm the relationship between each metal for all of the carparks (Supplementary information: Table B). The industrial carpark had the highest TZn to TCu ratio (15:1) suggesting higher wear and tear from larger commercial vehicles. The university and the hospital active carparks had a similar and lower TZn to TCu ratio (8:1) implying consistent wear and tear from small vehicles (Supplementary information: Table C).

3.2.4. Fractionation of heavy metals

A linear relationship was observed between TZn and dissolved Zn (dZn) for all carparks except the industrial carpark (Supplementary information: Fig. C). Strong positive correlations were found between TZn and dZn concentrations for the university carpark ($r = 0.97$, $p < 0.001$) and at the hospital operational carpark ($r = 0.9$, $p < 0.001$) (Supplementary information: Table D). A moderate positive correlation was found for TCu and dissolved Cu (dCu) for the university carpark ($r = 0.595$, $p < 0.005$). In contrast, strong positive total metals to particulate metals relationship were found for the industrial carpark (Zn: $r = 0.99$, $p < 0.001$, Cu: $r = 0.98$, $p < 0.001$, Pb: $r = 1$, $p < 0.001$). The highest percentage of dZn was 70% at the hospital operational carpark, followed by the university (65%) and the hospital non-operational carpark (62%) (Fig. 3). The industrial carpark exhibited a lower percentage of dZn (15%) as compared to the other two carparks studied, and dCu ranged from 18 to 45% (Supplementary information: Table E). A smaller percentage (below 9%) of dPb was measured for all sites. A large variation on % metal partitioning between the events was seen for all of the carparks. Even though the hospital operational carpark had the highest percentage of dissolved metals as compared to the two other carparks, mean, and maximum dissolved metal concentrations were found to be higher at the industrial carpark (Table 4).

4. Discussion

4.1. Discharge of total suspended solids

The three urban carparks differ noticeably in their vehicular activities such as traffic pattern and size of vehicles. As such, differences in TSS concentrations between the three carparks are most likely primarily due to carpark traffic characteristics. Results suggest that TSS build-up at a higher rate in the industrial carpark resulting in higher TSS concentrations in the first flush stormwater. These higher TSS concentrations seem to be related to the industrial carpark having higher traffic counts as compared to the other two carparks studied with an average daily traffic of > 1000 of which 20% were 16-wheeler commercial trucks. Furthermore, TSS concentrations in this carpark seem also to be influenced by higher wear and tear common in larger commercial vehicles as has been reported by Garg et al. (2000) and Smolders and Degryse (2002). The higher mean TSS concentrations at the industrial carpark suggest that depending upon frequency and nature of the vehicles, TSS concentrations can be many times higher than road runoff (Göbel et al., 2007). Similarly, median TSS concentrations at the university carpark were found to be higher than in road runoff as reported by Charters et al. (2016) for a similar catchment area. Although, vehicle count at the hospital carpark was lower than at the university, mean and median TSS concentrations were higher for the hospital operational carpark. The location of the hospital carpark, at the foot of Christchurch Port Hills, might have influenced TSS deposition on it. In addition, several other factors such as visiting and parking hours, the extent of maneuvering and the size of the vehicles were consistent throughout the year at the hospital carpark whereas at the university traffic was more irregular as the university is closed during term breaks as well as at the end of year. It is likely that despite its larger drainage area and higher traffic count, these other factors contributed to the lower TSS concentrations in the university carpark.

The size of the carpark area did not appear to have an effect on first flush concentrations, when it was considered independently or when the ratio of the carpark areas to traffic count was considered (Lau et al., 2009). In this study, the carpark areas ranged from 1752 to 5036 m^2, but given that the rainfall is the same at each carpark, it is likely that the stormwater collected in the 1 L samplers comes from a similar area within each carpark, and thus first flush stormwater collection area is

Table 4
Mean, median and ranges of particulates and dissolved metals (Zn, Cu and Pb) concentration.

Carpark type	pZn (µg/L) mean, median (range)	dZn (µg/L)	pCu (µg/L)	dCu (µg/L)	pPb (µg/L)	dPb (µg/L)
University	144, 104 (0 – 528)	256, 118 (31 – 2175)	31, 22 (0 – 115)	19, 16 (6 – 43)	56, 12 (0.1 – 32)	3, 1 (0.14 – 32)
Hospital (active)	92, 84 (0 – 240)	302, 231 (54 – 1001)	19, 15 (6 – 32)	15, 15 (9 – 22)	24, 23 (10 – 49)	2.3, 2 (0.40 – 11)
Hospital (passive)	57, 32 (0 – 212)	94, 62 (45 – 325)	15, 12 (2 – 50)	13, 12 (7.8 – 23)	14, 10 (1.19 – 35)	0.5, 0.5 (0.10 – 0.9)
Industrial	2315, 1481 (93 – 8123)	400, 283 (88 – 944)	145, 105 (34 – 326)	33, 27 (8 – 72)	170, 135 (35 – 359)	1.3, 1.4 (0.15 – 2.7)

consistent between carparks. On the other hand, the ratio of the carpark area (m^2) to daily traffic count was higher at the university carpark (5.6:1) than at the industrial and the hospital carparks (3:1 in both). Even though the industrial and the hospital carparks had similar areas to daily traffic ratios, mean TSS concentrations were three times higher at the industrial carpark. This finding reinforces the notion that apart from vehicular traffic and drainage area, mode of driving (speed turning and wheel movement) and size of vehicles are also likely to influence substantially on TSS loadings.

4.2. Discharge of total metals

The high Zn concentrations observed at the industrial carpark are likely to be contributed from large vehicle wear and tear. Tire wear is a major contributor of Zn (Kennedy and Sutherland, 2008). Trucks tires have higher weighted average ZnO in the thread (2.1%) than those of cars (1.2%) (Smolders and Degryse, 2002; Murphy et al., 2015). Similarly, Cu emitted from brakes pads range from 3.17 to 8.70 mg/km for small cars to large trucks respectively (Garg et al., 2000). The higher Zn and Cu at the industrial carpark is also likely due to the frequent braking and acceleration due to loading and maneuvering to deliver goods. The total Zn to total Cu ratios at the university and the hospital active carparks are lower and consistent with small vehicle wear and tear. The similar mean Cu concentrations for both hospital active and passive carparks is likely due to the lower desorption property of Cu which remains longer on the carpark surfaces (Gunawardana et al., 2015).

Recent studies on atmospheric deposition of heavy metals in urban runoff in the same geographic location found average atmospheric deposition contributions of 26.3 µg/L TZn, 7.9 µg/L TCu, and 2.2 µg/L TPb (Murphy et al., 2015, Charters et al., 2016). These values indicate that the contribution of metals from atmospheric deposition is lower as compared to vehicular activities in the study area.

4.3. Relationship to rainfall conditions and variabilities within carpark types

Pollutants in first flush runoff from the university and industrial carparks were found to have a low positive correlation. The Hospital carpark did not show any correlation with rainfall characteristics. In additional, no consistent seasonal pollutant concentration trends were observed. The relatively low rainfall intensities in this study region seem to dampen the inherent variability due to rain. On the other hand, differences in pollutant loadings from defined carpark types could result from variability in other factors within the carpark and their surroundings. For example, “residential” defined carparks could have sufficient differences in traffic, topography, or other characteristics that could result in differences in observed runoff quality.

4.4. Active vs passive carparks

A significant decrease in TSS and metal concentrations at the hospital passive carpark further suggests that vehicular traffic is the main contributor of TSS and heavy metals build-up on carpark surfaces. TCu concentration at the active and passive carparks were similar suggesting that metals, especially Cu will remain in the environment for a longer period as not all storm events completely clean carpark surfaces and the effect is prolonged even after many storm events. Even when the carpark was passive, it took a significant amount of time to wash off metals from the hospital carpark, especially in a low-intensity rainfall climate.

4.5. Implications for treatment of first flush TSS and heavy metals concentrations

Results from TSS and heavy metals concentrations (both dissolved and particulate) indicate that the industrial carpark had the highest concentrations of pollutants. Removal of TSS from the industrial carpark first flush would significantly reduce the concentration of heavy metals since the majority of metals were in particulate form. However, the industrial carpark also had a significant amount of dissolved metals, which need to be addressed while designing stormwater treatment systems. Many of the stormwater treatment systems are designed to remove particulate matter such as TSS including particulate heavy metals through filtration and sedimentation, but most stormwater treatment systems currently used for carparks do not target the removal of dissolved metals. Significant differences in the pollutant concentrations between the industrial and other two carparks suggest land use based treatment systems are essential.

The findings also indicate that stormwater treatment systems that capture or treat the initial portion of stormwater discharge from these carparks are likely to provide long term environmental benefit. Different treatment criteria such as treating the first 13 mm of runoff per impervious area (Schueler, 1987), or volume of runoff by a 19 mm rainfall in the State of California (California Regional Water Quality Control, 2001) or removal of at least half (50%) of the constituent mass in the first 25% of the runoff volume (Wanielista and Yousef, 1993) are all overly conservative for the carparks analyzed in this study. The average rain depth monitored during the sampling was found to be 5.4 mm with a maximum of 12.6 mm. For these carparks, treating the first flush runoff from a small portion of the event runoff volume could be considered a more economical approach for reducing a large fraction of pollutants (Barco et al., 2008). Although the treatment of first flush runoff can lead to overall improvements to discharge water quality (Li et al., 2008), further studies are needed to quantify the contribution from steady state runoff.

5. Conclusions

Urban carpark first flush pollutants concentrations varied across the three carparks studied. TSS and heavy metals concentrations were particularly influenced by traffic count and the size of the vehicles. Metal partitioning and metal species ratios also differed between the carparks. Differences in the ratios of heavy metals at the industrial carpark, for example, indicated relatively higher wear and tear from larger commercial vehicles. A significant decrease in pollutants concentrations at the hospital passive carpark further reinforces that vehicular traffic is the main contributor of TSS and heavy metals build-up on carpark surfaces. Vehicular activities and types were thus determined to be the dominant source of deposited TSS and metal pollutants in the carparks studied.

Surrounding topography and traffic patterns also influenced pollutant concentrations, but to a smaller extent. ADD, initial 10 min rainfall intensity, and rain depth only had a low positive correlation with first flush pollutant concentrations for all carparks.

The data collected in this study suggests that consideration of pollutant concentrations and loads is necessary for the selection of suitable stormwater treatment systems for individual carparks. The university and hospital carparks, for example, had high concentration of dissolved metals compared to the industrial carpark, and thus treatment priorities for those should be to remove dissolved metals.

Further data collection from other carparks for understanding variability from broadly defined carpark types is also warranted, particularly with respect to traffic and specific carpark characteristics. In addition to first flush, understanding of pollutant loads during steady-state is necessary to ensure enhanced treatment. Sizing, cost-effectiveness, trapping, and treatment efficiency of concerned pollutants are also factors to be considered when designing/implementing suitable land use based treatment systems.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envc.2021.100301.

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