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The impacts of traffic and rainfall characteristics on heavy metals build-up and wash-off from urban roads

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ABSTRACT

An investigation into the effects of changes in urban traffic characteristics due to rapid urbanisation and the predicted changes in rainfall characteristics due to climate change on the build-up and wash-off of heavy metals was carried out in Gold Coast, Australia. The study sites encompassed three different urban land uses. Nine heavy metals commonly associated with traffic emissions were selected. The results were interpreted using multivariate data analysis and decision making tools, such as principal component analysis (PCA), fuzzy clustering (FC), PROMETHEE and GAIA. Initial analyses established high, low and moderate traffic scenarios as well as low, low to moderate, moderate, high and extreme rainfall scenarios for build-up and wash-off investigations. GAIA analyses established that moderate to high traffic scenarios could affect the build-up while moderate to high rainfall scenarios could affect the wash-off of heavy metals under changed conditions. However, in wash-off, metal concentrations in 1-75µm fraction were found to be independent of the changes to rainfall characteristics. In build-up, high traffic activities in commercial and industrial areas influenced the accumulation of heavy metal concentrations in particulate size range from 75 - >300 µm, whereas metal concentrations in finer size range of <1-75 µm were not affected. As practical implications, solids <1 µm and organic matter from 1 - >300 µm can be targeted for removal of Ni, Cu, Pb, Cd, Cr and Zn from build-up whilst organic matter from <1 - >300 µm can be targeted for removal of Cd, Cr, Pb and Ni from wash-off. Cu and Zn need to be removed as free ions from most fractions in wash-off.

Keywords: Climate change, Heavy metals, Build-up, Wash-off, Water pollution

INTRODUCTION

Rapid urbanisation and climate change are two global phenomena that are attracting increased debate amongst the scientific community throughout the world in terms of their impacts on the environment. Rapid urbanisation and the consequent changes to urban traffic characteristics such as increased volume and congestion will in turn affect pollutant build-up on road surfaces (*1, 2*). Additionally, predicted changes in rainfall characteristics (*3*) due to climate change can readily affect pollutant wash-off from urban road surfaces. An in-depth knowledge of the build-up and wash-off processes of pollutants under such phenomena is the key to developing adaptive measures to mitigate the impacts of these changes on the water environment and to create a sustainable urban environment. For example, Delpla et al. (*4*) suggested that monitoring and analysis of the occurrence and fate of micro-pollutants including heavy metals must be considered in adaptive measures in the treatment of drinking water with regards to the climate change impacts.

In this regard, the environmental impacts and more specifically the water quality impacts of road transport in combination with climate change have received limited attention in research literature (*2, 5*). Pollutant accumulation and its subsequent wash-off in the environment are complex processes. Pollutant build-up in urban areas is influenced by anthropogenic activities related to population density, commerce and industry, land use, average daily traffic (ADT) and traffic volume to capacity ratio (V/C) (*2, 5, 6*). Pollutant wash-off is mainly influenced by the rainfall characteristics and the subsequent surface runoff from both pervious and impervious surfaces (*7*).

Heavy metals are among the most important pollutants in stormwater runoff that are generated by transport activities (*8, 9*). These pollutants have significant human and ecosystem health impacts (*10-12*). Several research studies have been undertaken in the past to characterise the build-up and wash-off of heavy metals under static environmental conditions (*13, 14*). However, in order to develop improved management practices for urban water quality, an in-depth understanding of the impacts of dynamic scenarios under changing urban traffic and rainfall conditions on the build-up and wash-off processes of heavy metals is critical. The research study discussed in this paper investigated the impacts of such changes in urban traffic and rainfall characteristics on the build-up and wash-off of heavy metals. A fuzzy clustering classification system is presented to characterise these dynamic scenarios. The outcome of this study is expected to contribute to the development of adaptive mitigation measures for the improvement of urban water quality.

EXPERIMENTAL SECTION

Site Selection

The research study was undertaken in the Gold Coast region of Southeast Queensland, Australia. The urban areas in this region have undergone rapid development. A suburb based approach was followed by selecting 11 road-sites in two suburbs, namely Helensvale and Coomera, which reflected the transport infrastructure that were developed during the last decade in the region. The sites selected for sample collection encompassed typical urban land uses, namely, residential, commercial and industrial, in order to obtain a cross-sectional view of traffic activities on road surfaces in the region. The residential sites comprised of single detached houses as well as multi-storied apartment blocks, the commercial sites were located near shopping mall car parks and the industrial sites were located in light industrial areas.

Build-up and Wash-off Sample Collection

A build-up sample collection method referred to as 'Wet and Dry Vacuum System' (*15*) was adopted for this study. A domestic vacuum cleaner with a water filtration system was used to collect the road dust from a 2 x 1.5 m plot area in the middle of the traffic lane in 25L plastic containers. Immediately afterwards de-ionised water was sprayed at 2 bar pressure on the collection plots and vacuuming was undertaken again to collect any remaining dust into the plastic containers.

Based on the climate change studies (*3*, *16*), the predicted rainfall characteristics for year 2030 were identified for the Gold Coast region. This study used a specially designed rainfall simulator (*13*) to replicate the design rainfall events common to the study region. It consisted of an A-frame structure with three Veejet 80100 nozzles, mounted on a stainless steel boom at a height of 2.4 m. In this paper, a total of 22 wash-off events were simulated. The runoff water was vacuumed continuously into 25L plastic containers using a domestic vacuum cleaner. For both build-up and wash-off, the total particulate analytes were fractioned into four size ranges, namely, 300 µm, 150-299 µm, 75-149 µm, 1-74 µm using wet sieving. The filtrate that passed through a 1 µm membrane filter was considered as the potential dissolved fraction. Detailed sample collection techniques are described in the supporting information.

Sample Testing

The heavy metals selected for analysis of build-up and wash-off samples were cadmium (Cd), chromium (Cr), nickel (Ni), lead (Pb), zinc (Zn), copper (Cu), antimony (Sb), manganese (Mn), aluminium (Al) and iron (Fe). These are mainly vehicle generated heavy metals (*13, 17*, *18*). Samples were preserved in 1 L polyethylene bottles with 3 mL (1+1) reagent grade nitric acid in 4˚C for at least 16 hours and then verified for pH< 2 just prior to withdrawing 50 mL aliquot for nitric acid digestion. The trace metal determination was performed by Inductively Coupled Plasma/Mass Spectrometry (ICP/MS).The test methods used are described in USEPA 200.8 (*19*).

As a measure of quality control, the percentage recovery, relative standard deviation and limit of detection for the target heavy metals were established. Details of these results are discussed in the Supporting Information.

The total and dissolved organic carbon (TOC and DOC), total and dissolved suspended solid (TSS and TDS) concentration, pH and electrical conductivity (EC) were also determined in all samples. The surface texture depth (STD) of pavements was measured according to the recommendations of the US Federal Highway Administration (*20*).

Data Analyses

In the build-up data matrices, the 11 sites were designated with object identifiers CH, IS, RB, RP, RD, CL, CT, RA, RDS, IBT and RR where the initials C, I and R were used to represent commercial, industrial and residential sites respectively. The 22 wash-off events were designated as rainfall objects assigned with numerical object identifiers starting from 1. The attributes of the objects in build-up and wash-off are described in the supporting information. After initial observation of the probability distribution of the build-up and wash-off objects, normalisation of each object was undertaken as a pre-treatment measure.

Considering the number of variables involved and the large amount of data generated, a range of multivariate analytical methods including principal component analysis (PCA), fuzzy clustering (FC) and the multicriteria decision making methods (MCDM) PROMETHEE and GAIA were employed for in-depth analyses of the data. A brief description of these data analyses techniques is outlined in the Supporting Information. Detailed discussion of these techniques can be found in the referenced literature (*21-24*).

RESULTS AND DISCUSSION

Exploratory PCA of Heavy Metals Build-up

PCA was performed on the pre-treated build-up data matrices. The resulting PCA biplot given in Figure 1 shows the patterns of variation for all particle size fractions taken together with the heavy metal elements. This initial PCA investigation revealed that Mn, Fe and Al form a group (A) of variables that are strongly correlated to each other while Ni, Cr, Zn, Pb, Cu, Sb and Cd form a separate group (B). These two groups are relatively orthogonal to each other which suggest that the two groups are uncorrelated and originating from different sources. Sb is found to have the strongest correlation with Cu amongst other group (B) variables. Vehicle brakes, bushings and thrust-bearings are described as the possible sources of both Cu and Sb with a very high Cu ratio. For example, 90% Cu in brake wear dust has been reported by Johansson et al. (*17*) and a Cu:Sb ratio of 4.6±2.3:1 by Sternbeck et al. (*18*).

Figure 1 also includes all the traffic related attributes of the objects. It is evident that STD and V/C are strongly correlated to group (B). Hence, it is postulated that the elements in group (B) are more likely to be generated from traffic activities. The elements in group (A) could be originating from the surrounding area as Mn, Fe and Al are among the most common elements in soil. Two objects with positive scores on PC1 (IBT and RDS) are strongly correlated with the groups (A) and (B) variables while objects IBT, RP and CL with positive scores on PC2 are strongly correlated to most of the group (B) variables and ADT, V/C and STD. These objects are a mix of residential, commercial and industrial land uses with varying traffic related attributes.

Insert Figure 1

In order to better understand the relationship between the object scores and variable loadings, individual PCA biplots of each fraction were analysed similarly as described above. Considering the individual biplots, it was observed that for particle fractions from 75 µm to >300 µm, the traffic related heavy metals are primarily generated in the commercial and industrial sites (objects CL, CT and IBT). High traffic densities in these areas are attributed to this fact. For the finer fractions such as 1 to 75 µm and the potential dissolved fraction of <1 µm, the traffic related heavy metals are present in all three types of land uses (objects CL, RDS, CT, IS, IBT, RR).

In a study by Patra et al. (*25*), the coarser particles were found to resuspend faster than finer particles due to vehicle induced turbulence on urban roads. They also found that fine particles remained on the ground for a longer time with very slow decay whilst the grinding of coarser materials under the wheels of traffic replenished the fine material reservoir on the road surface which further slowed the fine particle decay pattern. Manoli et al. (*26*) also observed strong contribution of resuspended road dust to the coarse particles on urban roads. Thus, in the current study, the predominance of coarser particles from 75 µm to >300 µm in commercial and industrial areas would mean their relatively rapid re-suspension and redistribution by traffic activities in these areas. However, the finer particles in the three land uses would not be affected by traffic activity.

Another important observation was the significant presence of TOC in the particulate fractions >300 µm, 150-300 µm, 75-150 µm and 1-75 µm with group (B) variables containing Ni, Cr, Zn, Pb, Cu and Cd whilst TSS was weakly present in these particulate fractions. Charlesworth and Lees (*27*) studied the association of heavy metals in group (B) with organic matter at source, transport and deposition phases. They found that irrespective of particle size between 63 µm and 2 mm, organic matter acted as the most dominant binding material with heavy metals at source. In the current research study, the particulate fraction was investigated further down to 1 µm. Thus, the presence of TOC with the group (B) heavy metals pointed to the fact that organic matter was acting as the binding agent for traffic related heavy metals at fractions higher than 1 µm. In the case of the potential dissolved fraction, the traffic related group (B) heavy metals were positively correlated with total dissolved solids (TDS) whilst dissolved organic carbon (DOC) was negatively correlated on PC1. This suggested that solids act as the binding agent for traffic related heavy metals for the potential dissolved fraction <1 µm.

FC Analysis and PROMETHEE Ranking of Heavy Metals Build-up

In order to classify the study objects consisting of traffic parameters such as daily traffic and congestion and hence better understand the correlation between the objects and the traffic generated variables discussed above, fuzzy clustering (FC) and PROMETHEE II net rankings were undertaken. The FC model consisted of three clusters (in keeping with the three land uses) and the cluster exponent p was set to 1.9 (moderately soft clustering). Thus, the class membership threshold value was set at 0.33 for an object to qualify as a member of a class. Table 1 shows the membership values obtained by fuzzy clustering of each object and the clusters are defined accordingly.

Insert Table 1

Cluster 1 is associated with high traffic volume ranging from 9000 to 24000 ADT with relatively high congestion. Cluster 2 is associated with moderate ADT values ranging from 2300 to 5900 with moderate congestion whilst cluster 3 is mainly associated with low traffic volume ranging from 500 to 3500 ADT with low congestion. In Table 1 there is one object (CH) left unclassified (fuzzy) as this object did not belong to a particular cluster according to the set membership threshold. The net ranking information produced by PROMETHEE II was used to generate the ϕ ranking values of all objects. The ϕ value determines how one object outranks the others in terms of its preference. The PROMETHEE II net ranking flow values of both the classified and fuzzy objects are given in Table 2.

Insert Table 2

It was found that the PROMETHEE II net ranking values could be used to classify the fuzzy object in Table 2. In this case the ‘Hope Island’ object could be classified into cluster 2 even though it had a slightly higher ADT value than this cluster. As the ‘Hope Island’ object had a relatively low congestion and similar texture depth as the other objects in cluster 2, the PROMETHEE II net ranking allowed this object to fall into this cluster. This type of classification simplified the analysis by decomposing the objects into high, low and moderate traffic objects.

Exploratory PCA of Heavy Metals Wash-off

The simulated rainfall events causing the wash-off of heavy metals were characterised by three attributes, namely, duration, intensity and average recurrence interval (ARI). PCA was performed on both particulate and dissolved wash-off data matrices as shown in Figures 2a and 2b.

Insert Figure 2

The PCA biplot of the total particulate wash-off given in Figure 2a revealed that pH and TOC have very strong correlation with all of the group (B) heavy metals except Cu and Zn. TSS was moderately correlated to the group (B) heavy metals except for Cu which has a very strong negative correlation with TSS. Charlesworth and Lees (*27*) found that organic matter and carbonates dominate the binding of particulate associated Cd, Cu, Ni, Zn and Pb during their transport from source to deposit. However, during the transport phase Cu and Zn were found to be bound more abundantly with carbonates than organic matter. Hence, the strong correlation between TOC and the group (B) metals except Cu and Zn reflected the fact that the organic matter could be acting as the binding agent for the particulate associated Cd, Cr, Pb and Ni during their wash-off.

In a sorption test using road dust leachates from roads with heavy traffic, Murakami et al. (*28*) found high concentrations of Cu and Zn in the soakbeds, suggesting traffic activities were contributing to the accumulation of these pollutants and that Zn might be released into the groundwater as free ions. The electrical conductivity (EC) of Cu and Zn is higher than that of Pb, Cd, Cr and Ni. In the case of the current study, the strong correlation of Cu and Zn with EC and their weak correlation with TOC in the particulate fraction meant that Cu and Zn could remain as free ions and were less likely to be associated with organic matter than the other group (B) heavy metals in particulate wash-off.

The PCA biplot for the dissolved fraction in Figure 2b shows that TDS has a relatively small loading vector and hence does not play a significant role in the dissolved fraction. Furthermore, in the dissolved fraction, Zn was found to have positive correlation with EC. Figure 2b suggests that organic matter is acting as the predominant binding agent for Ni, Cr, Cu, Pb and Cd whilst Zn still remained as free ions in the dissolved fraction.

FC Analysis and PROMETHEE Ranking of Heavy Metals Wash-off

From Figure 2, the impacts of rainfall attributes such as the intensity, frequency and duration on the wash-off of heavy metals were not easily discernible. Hence, fuzzy clustering (FC) coupled with PROMETHEE II net ranking was undertaken in order to further clarify the heavy metals wash-off. The cluster exponent was set to 1.9 and the number of clusters to four. This approach was chosen in order to incorporate low, moderate, high and extreme rainfall events. The events with intensity <40 mm/hr with relatively low average recurrence interval (ARI) were classified as low events; those having intensity between 50 to 100 mm/hr but with relatively higher ARIs of up to 50 years were classified as moderate events; events with intensities >100 mm/hr with very high frequency were classified as high events whilst events with similar intensities to moderate and high with extremely rare occurrence ($ARI \geq 100$ years) were classified as extreme events. The membership threshold was set to 0.25. Table 3 shows the membership values obtained by fuzzy clustering of each object and the clusters were defined accordingly.

Insert Table 3

As some of the rain events remained unclassified (fuzzy) after clustering, the PROMETHEE II routine was used to obtain their ϕ net ranking flow. The range of this value was used to determine the extent to which one cluster outranks another cluster. Hence, a fuzzy rain event from Table 3 could still be classified if its ϕ value falls within the range of a cluster. Table 4 presents the ϕ ranking values for both the classified and fuzzy events.

Insert Table 4

It is evident from Table 4 that the low and moderate clusters do not decisively outrank each other as the ranges of their ϕ values overlap. Hence, some fuzzy events (events 4, 11, 14, 18, 20 and 22) were inclusive to both low and moderate clusters after PROMETHEE II ranking. These events were further classified into a new sub-cluster called "low to moderate". Consequently, event 10 could now be classified as moderate while event 21 as extreme as their ϕ values fall exclusively within the ranges of the corresponding clusters. Due to changes in rainfall characteristics, this approach decomposed the heavy metal wash-off scenarios into five easily identifiable clusters, namely, low, moderate, low to moderate, high and extreme. Further heavy metal wash-off analyses were based on these clusters.

GAIA Analysis Incorporating Impacts of Traffic and Climate Change

As the final step, the study incorporated the impact of traffic and climate change into the heavy metal build-up and wash-off analyses by considering together all the clusters obtained. The five size fractions (>300μm, 150-300μm, 75-150μm, 1-75μm and <1μm) were considered as separate scenarios and were combined together for both build-up and wash-off. Under the existing conditions of heavy metals build-up and wash-off, the dominant build-up and wash-off clusters (which include the objects or actions) were investigated. The magnitude and inclination of the π decision vector indicated the dominance of a particular action or a group of actions under the combined scenarios. GAIA multicriteria routine was used to generate the biplots of the build-up and wash-off scenarios shown in Figure 3a and 3b respectively.

Insert Figure 3

In Figure 3a the decision vector π was located between moderate and high traffic (between the objects CL and IS). The particle size >150μm was strongly correlated with moderate to high traffic (CL and IS) whilst smaller particle sizes from 1 to 150μm were strongly correlated with moderate to low traffic (RB and RP). The rapid re-suspension of the larger particles (>150μm) in high traffic areas was attributed to this fact. The potential dissolved fraction (<1μm) and the finer fraction of 1-75μm were also strongly correlated with both moderate and high traffic (IS and IBT for <1μm; CL for 1-75μm). This pointed to the fact that the attributes of the moderate to high traffic sites will dominate heavy metals build-up in a mixture of urban traffic clusters for both particulate and dissolved fractions.

The GAIA biplot for heavy metals wash-off (Figure 3b) shows that the decision axis is situated between moderate (events 10 and 19) and high (events 7 and 8) rain events. The high rain events and moderate rain events have significant correlations with both, particulate and dissolved fractions. The low to moderate rain events (events 1 and 20) have limited impacts on these fractions as evident from their low correlations with both particulate and dissolved fractions. The extreme and low rain events did not have any noteworthy correlations with the wash-off of any heavy metal species. These findings suggest that the attributes of the moderate to high rainfall events will dominate heavy metal wash-off arising from predicted changes to rainfall characteristics due to climate change. Another important point to note in the GAIA biplot for wash-off is the magnitude of the loading vector for the 1-75μm fraction which is greater than all other fractions with little or no correlations with most of the rain events. Hence, it is postulated that the wash-off of heavy metals in this fraction is independent of any rain event and will not be affected by the predicted changes to rainfall characteristics due to climate change.

The research study provides a framework to investigate and classify urban traffic or rain events for given attributes and to incorporate this information to study the impact of urban traffic and climate change on heavy metals build-up and wash-off. The key findings from the study can be summarised as follows:

- Urban traffic characteristics in a moderate to high traffic environment will affect heavy metals build-up on urban roads for all particulate and dissolved fractions.
- The characteristics of moderate to high rainfall events will affect heavy metals wash-off from urban roads for most particulate and dissolved fractions. However, in wash-off, the 1-75μm fraction is independent of the predicted changes to rainfall characteristics due to climate change. Therefore, any adaptive measure incorporating the impacts of climate change on heavy metals wash-off must include the 1-75 μm fraction irrespective of the classified rain events described in this paper.
- In the build-up of heavy metals, coarser particles from 75 μm to >300 μm were predominant in the commercial and industrial areas. This indicates that high traffic activities in these areas affect the particulate bound heavy metals. However, finer particles of 1-75 μm and the potential dissolved fraction of <1 μm in all three land uses indicate that their presence is not affected by traffic related activities.
- In the build-up of heavy metals on urban road surfaces, Ni, Cu, Pb, Cd, Cr and Zn tend to be associated with organic matter for all particulate fractions. However, these heavy metals tend to be associated with solids in the potential dissolved fraction of <1 μm. Therefore, as a practical implication of this finding, in build-up, solids can be targeted up to <1 μm and organic matter can be targeted from 1 μm to >300 μm for the removal of traffic generated heavy metals on urban roads.
- In the wash-off of heavy metals, organic matter acts as a binding agent for both particulate bound and dissolved Cd, Cr, Pb and Ni. However, Cu and Zn are strongly associated with EC in particulate fractions whilst Zn is also strongly correlated with EC in the <1 μm fraction. Therefore, organic matter can be targeted from 1 μm to >300 μm size fraction for the removal of Cd, Cr, Pb and Ni whilst Cu and Zn will need to be removed as free ions. In the dissolved fraction of <1 μm, organic matter can be targeted for the removal of Cd, Cr, Pb, Cu and Ni whilst Zn still needs to be removed as free ions.

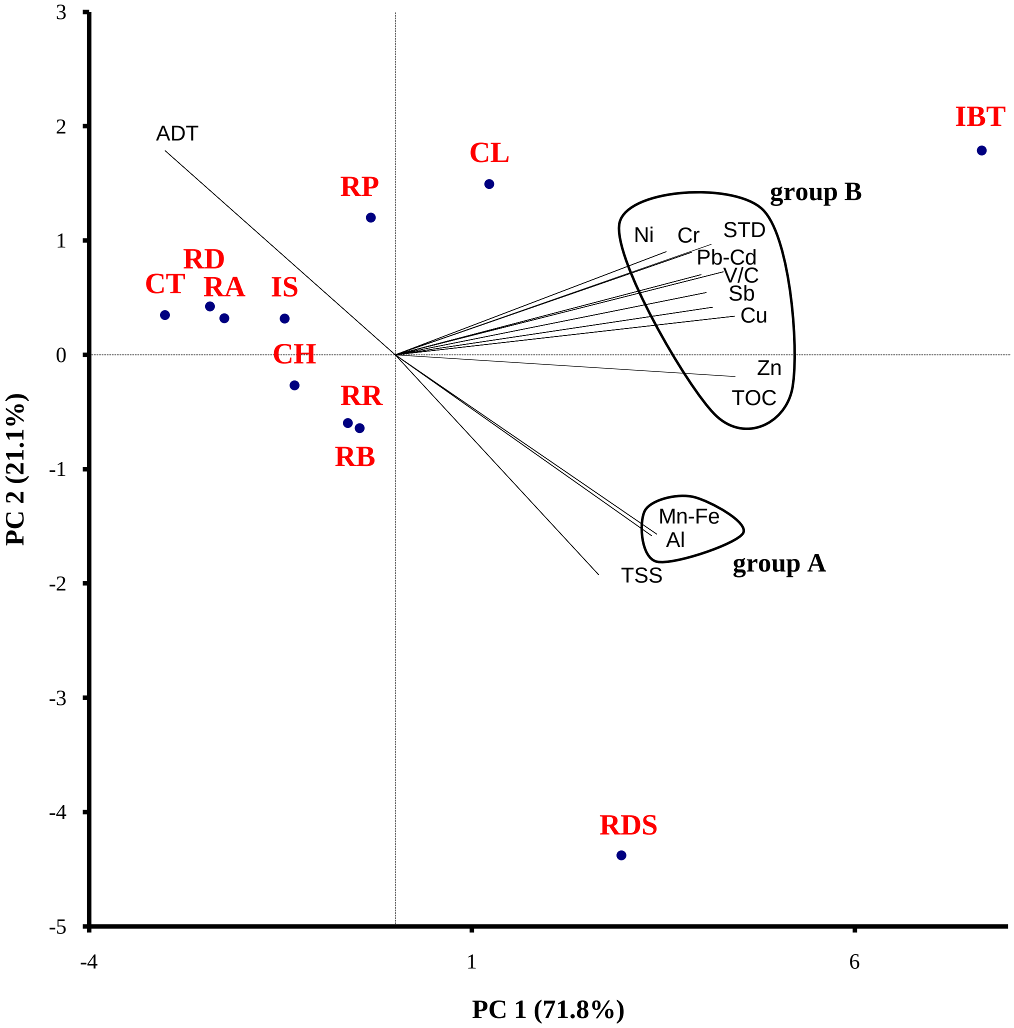
This study provides guidance for the development of stormwater quality mitigation strategies for the removal of heavy metals taking into consideration the dynamic scenarios of urban traffic and climate change. The key findings can contribute to the development of adaptive measures in stormwater quality mitigation for urban areas undergoing such changes.

LIST OF FIGURES

Figure 1 PCA biplot of build-up of all heavy metal size fractions; object identifiers are described in Table 1

Figure 2 PCA biplot for (a) particulate and (b) dissolved fractions for wash-off of heavy metals; objects are indicated by numbers starting from 1; numerical object identifiers are described in Table 3

Figure 3 GAIA biplots for (a) build-up :(◇) low traffic, (▽)moderate traffic and (○) high traffic: object identifiers are described in Table 1;and (b) wash-off : (○) low event, (◇) low to moderate event, (●) moderate event, (◆) high event and (□) extreme event: numerical object identifiers are described in Table 3



3
2
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PC 1 (71.8%)
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IBT
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RP
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CT
RA
IS
CH
RR
RB
RDS
group B
Ni
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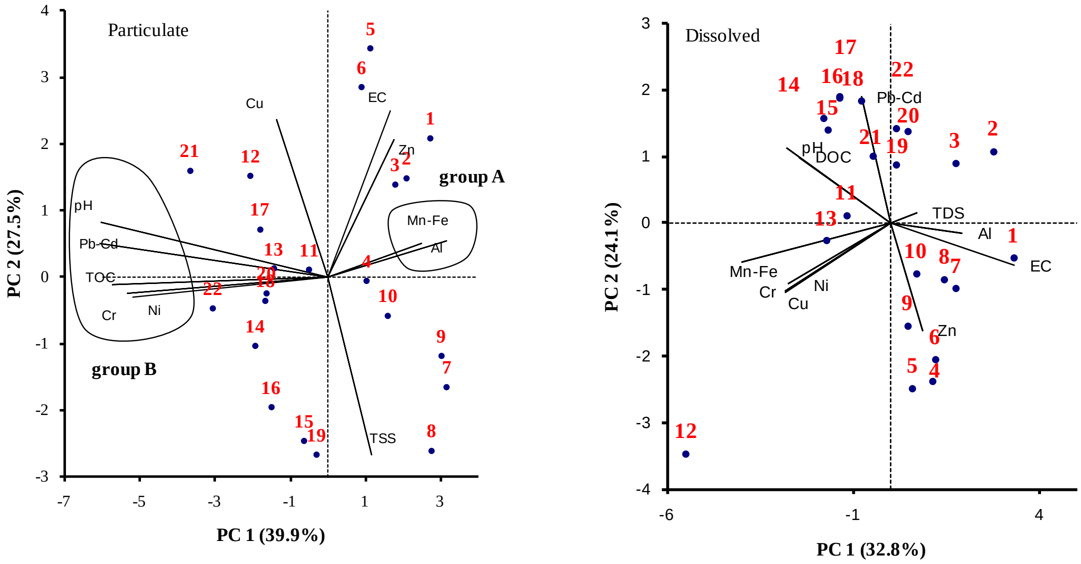
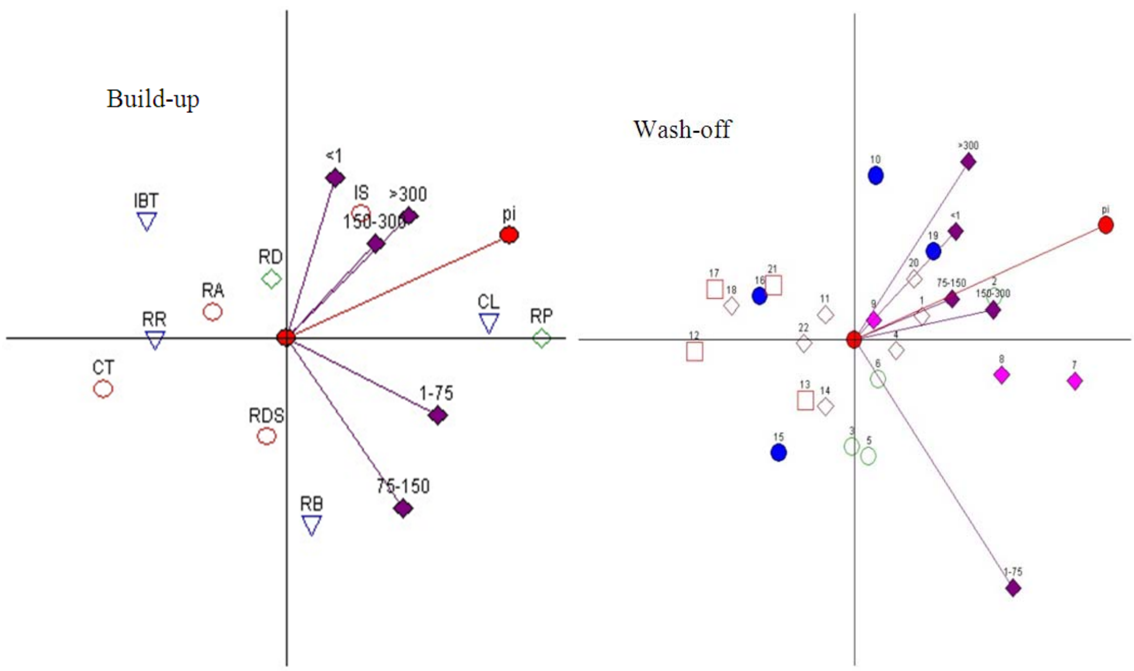
STD
Pb-Cd
V/C
Sb
Cu
Zn
TOC
Mn-Fe
Al
group A
TSS

Figure 1



(a) **(b)**

Figure 2



(a) **(b)**

Figure 3

Table 1 Membership values of different objects in heavy metals build-up after fuzzy clustering

Object names(labels or identifiers)*	**Cluster 1**	**Cluster2**	**Cluster 3**	**Cluster names**
Hope Island Road(CH)	0.651	0.343	0.006	fuzzy
Shipper Drive(IS)	0.723	0.272	0.005	High traffic
Billinghurst Crescent(RB)	0.012	0.988	0.001	Moderate traffic
Peanba Park Road(RP)	0.013	0.021	0.966	Low traffic
Dalley Park Drive(RD)	0.045	0.102	0.853	Low traffic
Lindfield Road(CL)	0.052	0.937	0.011	Moderate traffic
Town Centre Drive(CT)	0.956	0.041	0.003	High traffic
Abraham Road(RA)	0.998	0.002	0.000	High traffic
Discovery Drive(RDS)	0.998	0.002	0.000	High traffic
Beattie Road(IBT)	0.008	0.992	0.000	Moderate traffic
Reserve Road(RR)	0.000	1.000	0.000	Moderate traffic

* The initials C, I and R were used in each object identifier to represent commercial, industrial and residential sites respectively

Table 2 PROMETHEE II net ranking (ϕ) values showing the fuzzy object in heavy metals build-up could still be classified as a member of moderate traffic cluster as its ϕ value lies within the range of that cluster

Object names (clusters)	**Net ranking values of classified objects**	**Fuzzy Object**	**Net ranking value of fuzzy object**
Town Centre Drive (high)	0.94	Hope Island Road	-0.25
Shipper Drive (high)	0.67		
Discovery Drive (high)	0.58		
Abraham Road (high)	0.56		
Billinghurst Crescent (moderate)	-0.03		
Beattie Road (moderate)	-0.06		
Reserve Road (moderate)	-0.22		
Lindfield Road (moderate)	-0.25		
Dalley Park Drive (low)	-0.67		
Peanba Park Road (low)	-0.86		

Table 3 Membership values of the rainfall events in heavy metals wash-off after fuzzy clustering

Rainfall Events*	Duration, minutes	Intensity, mm/hr	ARI	Cluster 1	Cluster 2	Cluster 3	Cluster 4	Cluster names
1	60	39.3	1	0.035	0.840	0.082	0.043	low
2	65	37.39	1	0.018	0.929	0.032	0.020	low
3	90	39.3	2	0.049	0.836	0.065	0.050	low
4	120	24.6	1	0.258	0.386	0.201	0.155	fuzzy
5	133	39.3	5	0.047	0.812	0.054	0.086	low
6	160	39.3	10	0.030	0.886	0.031	0.053	low
7	5	125	1	0.048	0.053	0.874	0.026	high
8	5.75	119	1	0.069	0.042	0.859	0.030	high
9	10.5	120	2	0.055	0.066	0.838	0.041	high
10	16	125	5	0.105	0.100	0.531	0.264	fuzzy
11	21	122	10	0.256	0.125	0.122	0.497	fuzzy
12	45	125	100	0.105	0.090	0.050	0.755	extreme
13	49	115	100	0.050	0.014	0.010	0.926	extreme
14	52.5	77	10	0.494	0.032	0.034	0.440	fuzzy
15	67.5	77	20	0.915	0.015	0.032	0.039	moderate
16	86.8	77	50	0.955	0.007	0.009	0.029	moderate
17	101.25	77	100	0.011	0.005	0.003	0.981	extreme
18	105	75	100	0.409	0.051	0.036	0.504	fuzzy
19	25	63	1	0.831	0.032	0.073	0.064	moderate
20	42.5	61.2	2	0.362	0.053	0.036	0.550	fuzzy
21	69	59.2	5	0.250	0.092	0.049	0.609	fuzzy
22	85	58.3	10	0.279	0.048	0.038	0.636	fuzzy

*Numerical object identifiers used elsewhere are same as the rainfall events numbers

Table 4 PROMETHEE II net ranking (ϕ) values showing two fuzzy events, 10 and 21 could still be classified as members of moderate and extreme clusters, respectively as their ϕ values fall exclusively within the ranges of corresponding clusters

Rain events (clusters)	Net ranking values of classified rain events	Fuzzy Rain Events	Net ranking values of fuzzy rain events
2(low)	0.18	4	0.18
1(low)	0.09	20	0.14
3(low)	0.06	11	0.02
6(low)	0.02	10	0.20
5(low)	-0.22	14	0.00
19(moderate)	0.20	22	-0.07
15(moderate)	0.03	18	-0.14
16(moderate)	-0.17	21	-0.43
13(extreme)	-0.24	-	-
17(extreme)	-0.44	-	-
12(extreme)	-0.50	-	-
7(high)	0.55	-	-
8(high)	0.50	-	-
9(high)	0.12	-	-

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SUPPORTING INFORMATION AVAILABLE
Detailed description of sample collection, quality control and data analyses techniques are presented in the supporting information section, along with additional tables and figures for interested readers. This information is available free of charge via the internet at http://pubs.acs.org.

BRIEF
The build-up and wash-off of heavy metals on urban roads have been investigated from a dynamic point of view incorporating the changes in urban traffic and rainfall characteristics.

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SUPPORTING INFORMATION FOR JOURNAL PAPER

The impacts of traffic and rainfall characteristics on heavy metals build-up and wash-off from urban roads

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Supporting Information

Number of Pages: 19

Number of Figures: 3

Number of Tables: 6

BUILD-UP SAMPLE COLLECTION

This study used 'Wet and Dry Vacuum System' (*1*) for the collection of build-up samples from the urban road surfaces. A domestic vacuum cleaner with a water filtration system was used to collect the road dust from a 2 x 1.5 m plot area in the middle of the traffic lane in 25L plastic containers. Immediately afterwards de-ionised water was sprayed at 2 bar pressure on the collection plots and vacuuming was undertaken again to collect any remaining dust into the plastic containers. In terms of collecting samples from a road surface subject to natural and traffic related degradation, calibration studies confirmed that this method achieved a collection efficiency of over 90%, same as described in earlier studies performed on synthetic surfaces (*2, 3*). Samples were collected from the road surfaces after an antecedent dry period of 7 days. This was in conformity with the findings of Egodawatta (*4*), who noted that pollutant build-up on road surfaces asymptote to an almost constant value after a seven day antecedent dry period. Homogeneous 500 mL subsamples were transferred to high density 1 L polyethylene bottles using a churn splitter. The particle size distributions of the suspended solids in the subsamples were obtained using a Malvern Mastersizer S Particle Size Analyser capable of analysing particles between 0.05 and 900 µm. Based on the particle size distributions, the total particulate analytes were fractioned into four size ranges, namely, 300 µm, 150-299 µm, 75-149 µm, 1-74 µm using wet sieving. The filtrate that passed through a 1 µm membrane filter was considered as the potential total dissolved fraction.

WASH-OFF SAMPLE COLLECTION

The study used a specially designed rainfall simulator to replicate the design rainfall events common to the study region. The rainfall simulator was based on the design of simulators used in agricultural research as described by Floyd (*5*) and Silburn et al. (*6*). It consisted of an A-frame structure made of aluminium tubing of 40-mm diameter. Three Veejet 80100 nozzles, spaced 1 m apart, were mounted on a stainless steel boom at a height of 2.4 m to obtain rainfall drop size and terminal velocity similar to natural rainfall. Further details on the design of the rainfall simulator can be found in Herngren et al. (*7*).

Based on the detailed study by CSIRO (*8*) and the regional climate change study (*9*), the predicted rainfall characteristics for year 2030 can be identified for the Gold Coast region. The wash-off simulation methodology is described in detail by Mahbub et al. (*1*). In this paper, a total of 22 wash-off events were simulated. The runoff water was vacuumed continuously into 25L plastic containers using the same vacuum cleaner as for the build-up sample collection. Subsequently, 500mL event mean concentration (EMC) samples were extracted using a churn splitter. The EMC represented a flow weighted average concentration of the pollutants computed based on the total pollutant mass divided by the total runoff volume for a given rainfall event. As pollutant concentrations may vary by orders of magnitude during a runoff event, the EMC samples (representing single indices) were found to be appropriate for evaluating the impacts of stormwater runoff on receiving waters (*10*). The particulate and dissolved fractions in wash-off were separated in similar manner as described for build-up.

QUALITY CONTROL MEASURES IN SAMPLE TESTING

For quality control, calibration standards, internal standards, blanks and certified reference materials were used. The calibration standard supplied by Accustandard® contained each of the target heavy metal analytes at a concentration of 100 mg/L. Six different calibration standards at concentrations of 20, 10, 5, 1, 0.1 and 0.01 mg/L were prepared. The internal standard containing Indium (In), Bismuth (Bi), Terbium (Tb), Scandium (Sc) and Yttrium (Y) was prepared at a concentration of 1 mg/L.

The certified reference material (TraceCERT, Sigma-Aldrich®) contained 10 mg/L of each target analyte except iron (100 mg/L). The volumes of samples, standards and blanks were all kept at 50 mL after digestion while the concentration of internal standards was kept at 0.02 mg/L for analysis. The laboratory fortified blanks were prepared by adding the certified reference materials to the deionised water to obtain a concentration of 0.1 mg/L for each target analyte except iron (1 mg/L) and were treated exactly as samples for analysis. The analytical technique used for the analysis of the metal analytes was Inductively Coupled Plasma/Mass Spectrometry (ICP/MS). The percentage recoveries of the spiked blanks with known concentration of analytes were estimated using the following equation:

$$R = (LFB - LRB)/C \times 100 \text{ --(1)}$$

where R= percent recovery, LFB= laboratory fortified blank, LRB= blank and C= stated concentrations of analytes in the LFB. For different heavy metals, percent recoveries were found to be 89% to 115%. The percentage recovery data was used to verify the accuracy of the analytical methodology.

To determine the repeatability of the process, seven replicates of a randomly chosen sample from each batch were analysed. The relative standard deviations (RSD) of the above samples were measured using the equation:

$$RSD = (S_{rsd} / \overline{X}_{rsd}) \times 100 \text{ ---(2)}$$

where S_{rsd} = Standard deviation of the replicate samples and $\overline{X}_{rsd}$ = mean of the replicate samples. The relative standard deviations were from 2.1% to 14.7%.

The reporting limits of the method were established by estimating the limits of detection (LOD) using seven separate blank samples. The LOD is the lowest concentration of an analyte measured by a method that could be reliably distinguished from zero. The LOD was calculated using the equation:

$$LOD = \overline{X}_{LOD} + 3S_{LOD} \text{ -- (3)}$$

where $\overline{X}_{LOD}$ = mean of the seven blanks and S_{LOD} = standard deviation of the seven blanks. The LOD for different heavy metals were established from 0.008 mg/L to 0.053 mg/L. All of the test results were found within the specified limits of the test methods described in USEPA 200.8 (*11*)

DATA ANALYSIS TECHNIQUES

PCA

PCA is a pattern recognition technique employed to investigate the correlations among different variables and clusters among objects. The PCA technique is used to transform the original variables to a new orthogonal set of Principal Components (PCs) such that the first PC contains most of the data variance and the second PC contains the second largest variance and so on. The application of PCA to a data matrix generates a loading for each variable and a score for each object on the principal components. Consequently, the data can be presented diagrammatically by plotting the loading of each variable in the form of a vector and the score of each object in the form of a data point. This type of plot is referred to as a 'Biplot'. Detailed descriptions of PCA can be found elsewhere (*12*).

Fuzzy Clustering

Fuzzy clustering is an object classification method that assigns a degree of class membership for a given object over several classes (*13, 14*). The classification is performed with a user specified membership function which, in the case of 'SIRIUS' software (*15*) used for the analysis is similar to that described by Bezdek (*13*). An example of a membership function is $m(x) = 1 - c\left|x - a\right|^{P}$, where a and c are constants and p is called cluster exponent with a suggested value between 1 to 3 (*15*).

Values closer to 1 result in hard clustering where the objects are placed into their most preferred classes while values closer to 3 result in soft clustering where the objects are allowed to spread over as many classes as possible. The sum of the membership values of each object is 1. The main advantage of fuzzy clustering is that it facilitates the distinction between the objects that clearly belong to one cluster and those that are members of several clusters. A class membership threshold is defined as 1/ n (n = number of clusters).

PROMETHEE and GAIA

PROMETHEE (preference ranking organisation method for enrichment evaluation) is a method that is designed to rank a number of objects in terms of the data criteria. The ranking for each variable or criterion is performed by a user specified preference function. The positive and negative partial outranking flows, ϕ^{+} and ϕ^{-} are calculated from the preference functions for each object or action. The ϕ^{+} values indicate how each action outranks all the others, while the ϕ^{-} values indicate how each action is outranked by all the others. This procedure is known as PROMETHEE I. In some instances, objects may perform equally well for a different set of variables. To eliminate such outcomes, the net outranking flow ϕ, which is the difference between ϕ^{+} and ϕ^{-}, for each action is calculated. This process is termed as PROMETHEE II. Further details can be found in Keller et al. (*16*).

The GAIA (Geometrical Analysis for Interactive Aid) is essentially a principal component analysis (PCA) biplot (*16*). It is generated from the matrix derived from the decomposition of ϕ net outranking flow values from PROMETHEE II. GAIA provides a graphical display of the relationships between objects, variables and each other. An additional feature of GAIA is the inclusion of the decision axis, π, which gives an indication of the degree of decision power (length of the π vector) as well as the quality of the preferred object or action.

ADDITIONAL FIGURES AND TABLES

The additional Figure S1 shows the individual PCA biplots for five size fractions as mentioned in the original paper in order to better understand the the relationship between the object scores and variable loadings. Figures S2 and S3 show the results of the particle size distributions in the build-up and wash-off samples respectively from the Mastersizer S Particle Size Analyser. The additional Table S1 shows the traffic related attributes of the study sites, Table S2 provides the rainfall simulation plan based on the climate change studies (*8, 9*), Table S3 provides individual results of the quality control parameters for each target heavy metals, Table S4 illustrates the possible sources of elemental emissions from vehicles based on literatures (*17-26*), and finally Tables S5 and S6 show the chemical compositions of build-up and wash-off respectively.

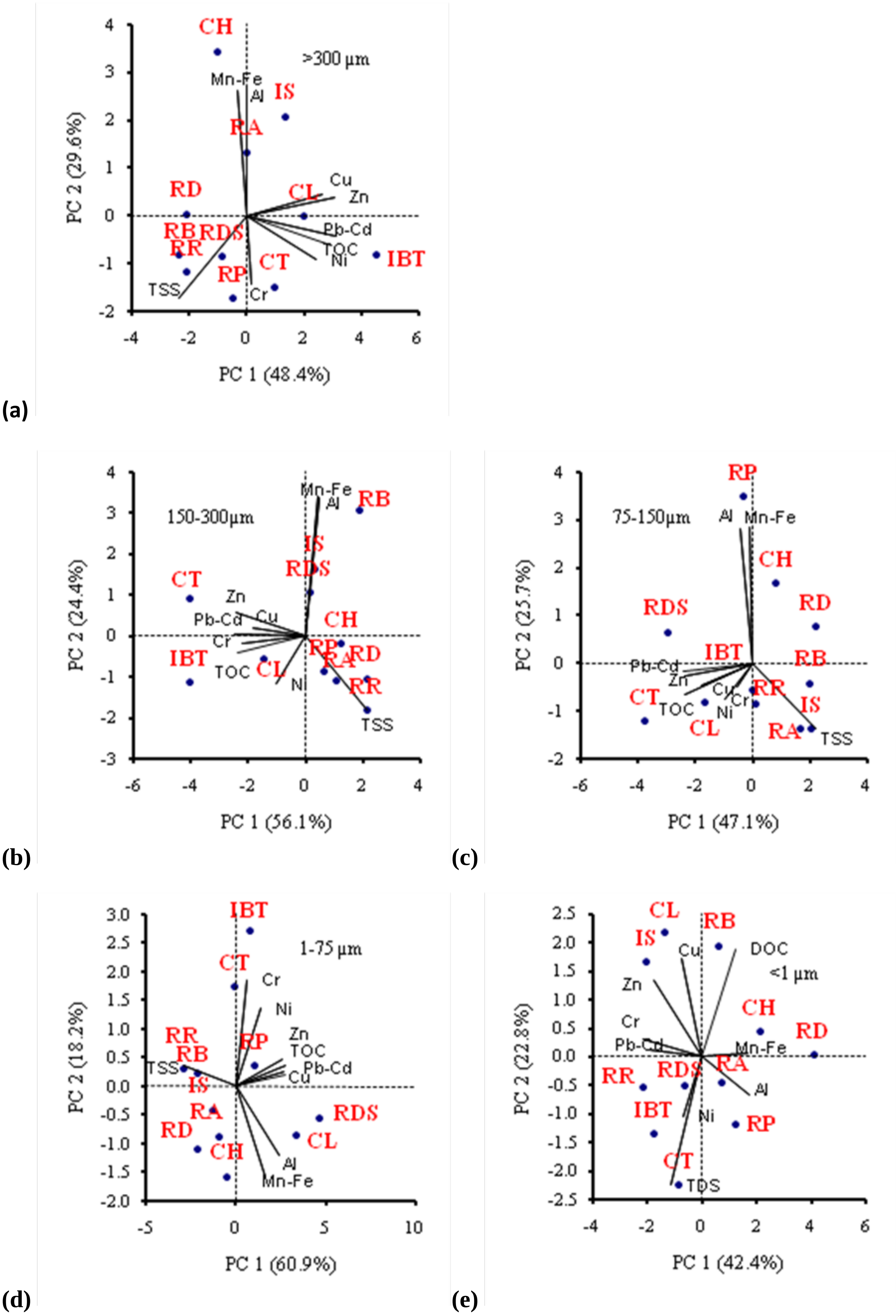

Figure S1 PCA biplots of particulate and potential dissolved fraction for heavy metals build-up for (a) >300µm, (b)150-300 µm, (c)75-150 µm, (d)1-75 µm and (e) <1 µm; objects are indicated by labels with the prefix I, C or R starting for industrial, commercial and residential sites, respectively

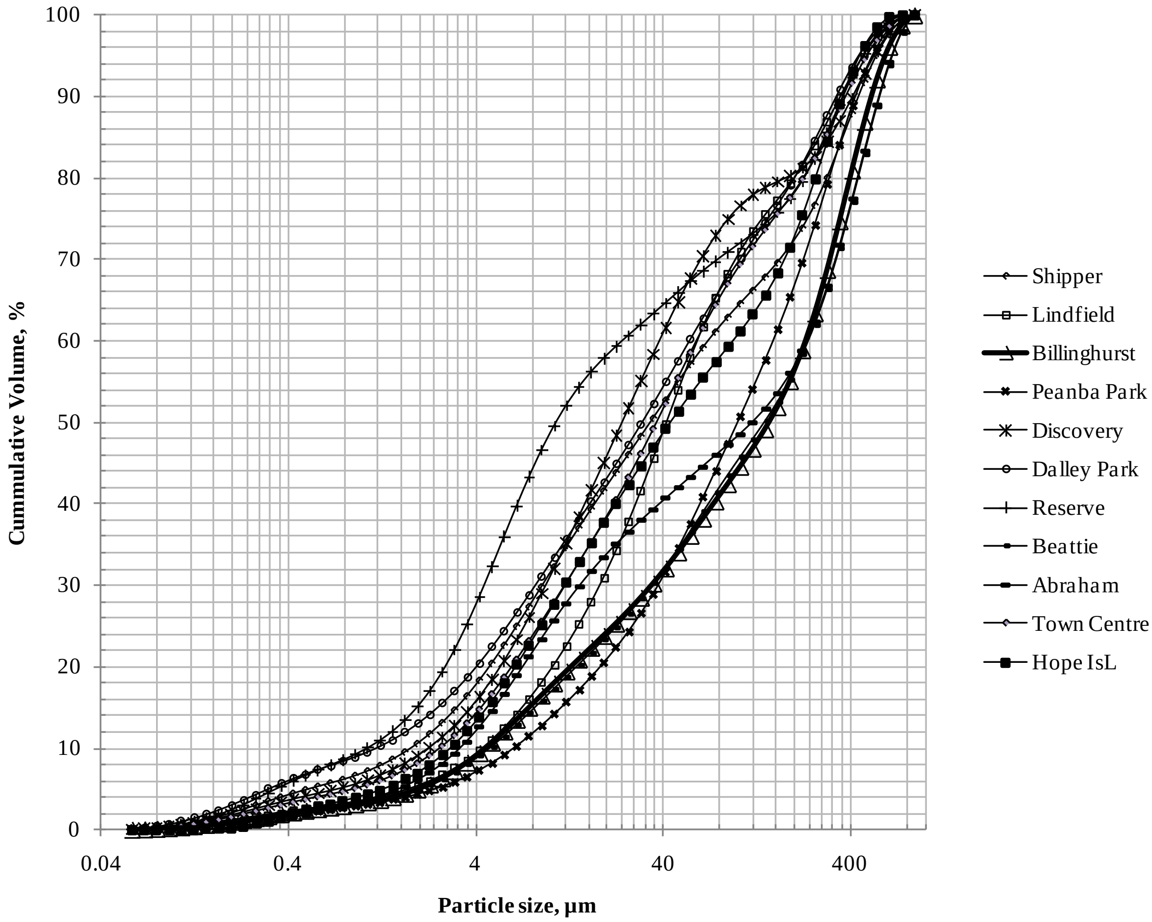

Figure S2 Results from Mastersizer S particle size distribution analysis showing that fractions <1µm constituted 2%-10% whilst fractions up to 300µm constituted almost 68% to 90% volume of the total build-up particles in samples collected from the 11 sites

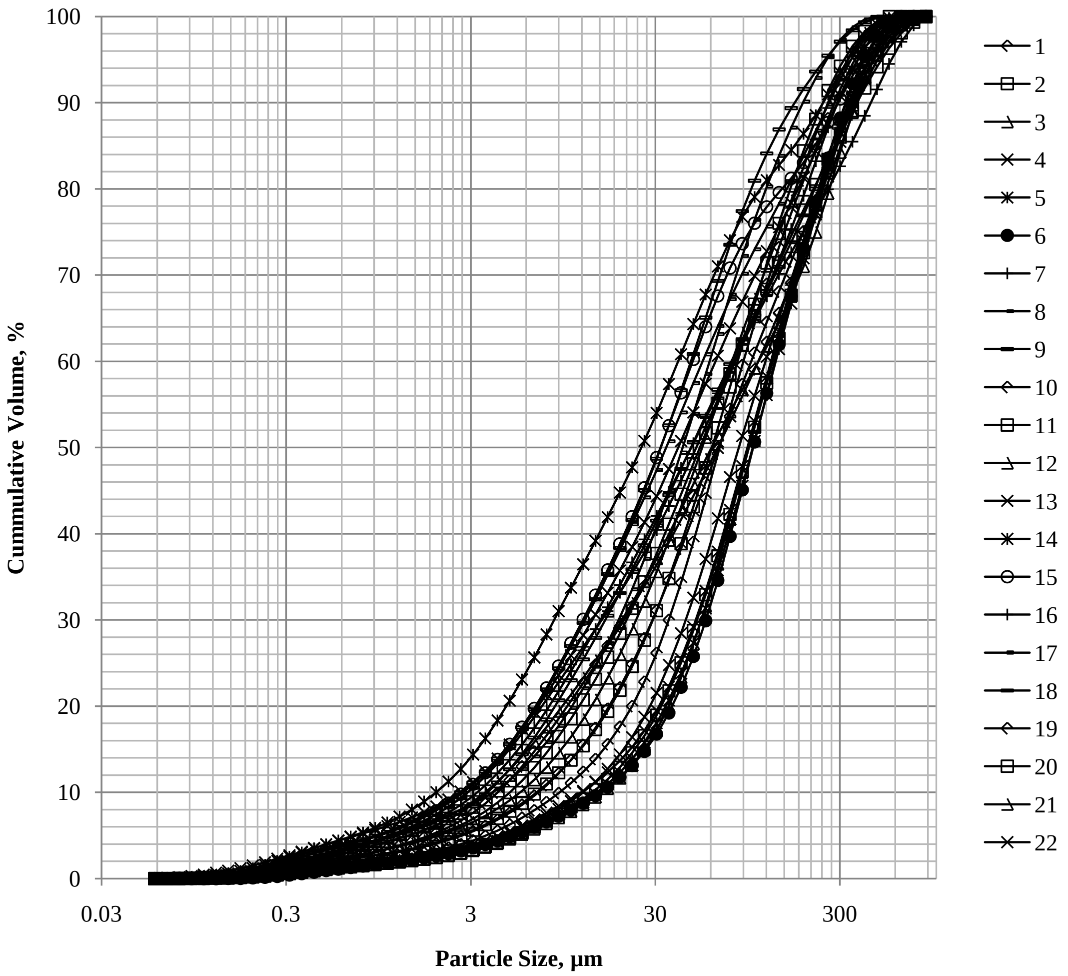

Figure S3 Results from Mastersizer S particle size distribution analysis showing that fractions <1µm constituted around 2% whilst fractions up to 300µm constituted almost 82% to 96% volume of the total wash-off particles in samples collected from the 22 simulated rain events

Table S1 Traffic data for the study sites

Site Name Site Label	Land Use	Coordinate Location	Average Daily Traffic (ADT), vehicles/day	Volume to Capacity Ratio (V/C)	Surface Texture Depth (STD), mm	Lane Width, m	Pavement type % of Aggregate Binder
Abraham Road RA	Residential	27.865°S 153.307°E	13028	1.11	0.6467	3.5	DG14[a] 5.1
Reserve Road RR	Residential	27.870°S 153.301°E	6339	0.45	0.7505	3.5	DG14[a] 5.1
Peanba Park road RP	Residential	27.851°S 153.281°E	581	0.15	0.6844	2.8	DG10[b] 5.3
Billinghurst Cres RB	Residential	27.856°S 153.298°E	5936	0.74	0.7015	2.9	DG10[b] 5.3
Beattie Road IBT	Industrial	27.868°S 153.324°E	2670	0.24	0.7074	3.5	DG14[a] 5.1
Shipper Drive IS	Industrial	27.861°S 155.332°E	7530	0.55	0.6788	3.5	DG14[a] 5.1
Hope Island Road CH	Commercial	27.882°S 153.328°E	7534	0.57	0.7254	3.4	DG14[a] 5.1
Lindfield Road CL	Commercial	27.922°S 153.334°E	2312	0.33	0.9417	3.3	DG10[b] 5.3
Town Centre Drive CT	Commercial	27.929°S 153.337°E	24506	0.62	0.6416	3.5	DG14[a] 5.1
Dalley Park Drive RD	Residential	27.887°S 153.346°E	3534	0.42	0.8342	2.9	DG10[b] 5.3
Discovery Drive RDS	Residential	27.899°S 153.327°E	9116	0.25	0.6957	2.9	DG14[a] 5.1

[a]Dense Grade Bitumen Asphalt with 5.1% aggregate binder
[b]Dense Grade Bitumen Asphalt with 5.3% aggregate binder

Table S2 Simulation rain events for the Gold Coast region at present and as predicted for year 2030

Current events				Predicted events for 2030*			
ARI (year)	**Duration (min)**	**Intensity (mm/hr)**	**Object Identifiers**	**ARI (year)**	**Duration (min)**	**Intensity (mm/hr)**	**Object Identifiers**
1	120	24.6	4	1	65	37.39	2
10	300	24	-	1	120	24.6	4
100	45	125	12	1	5	125	7
10	160	39.3	6	10	85	58.3	22
1	60	39.3	1	1	25	63	19
2	90	39.3	3	2	42.5	61.2	20
5	133	39.3	5	5	69	59.2	21
10	52.5	77	14	5	16	125	10
20	67.5	77	15	10	21	122	11
50	86.7	77	16	2	10.5	120	9
100	101.25	77	17	1	5.75	119	8
100	105	75	18	100	49	115	13

* Based on the climate change studies (*8, 9*)

Table S3 Limits of detection, percent recovery and relative standard deviation percentage found in the heavy metal analysis with corresponding molecular weights shown alongside each element

Heavy metal elements	LOD (mg/L)	Recovery, %	Relative Standard Deviation, %
Al / 27	0.010	105.939	11.297
Cr / 53	0.020	100.767	9.449
Mn / 55	0.026	89.914	12.567
Fe / 56	0.007	106.537	2.098
Fe / 57	0.005	97.355	2.433
Ni / 60	0.053	99.805	4.946
Cu / 63	0.019	110.402	7.460
Zn / 66	0.012	115.707	14.657
Cd / 111	0.078	114.481	-
Pb / 206	0.026	111.988	4.573
Pb / 207	0.045	110.841	4.991
Pb / 208	0.008	102.777	3.874

Table S4 Possible sources of elements frequently found in exhaust and non-exhaust emissions of motor vehicle

Elements	**Possible Sources**
Cu, Sb	Bushing, thurstbearing, brake (*17-21*)
Zn, Cu	Lubricants, engine oil (*22, 23*)
Zn, Cd	Tyre (*19, 24, 25*)
Cr	Alloy wheel plate, crankshaft, metal plating, yellow paint of pavement (*19, 24*)
Pb, Ni	Exhaust emission (*18*)
Cu, Sb, Ba	Rush hour stop-start (*26*)

Table S5 Chemical compositions (mean±standard deviations) of the Build-up of Heavy metals in the selected sites

Site Name **Site Label**	**Al, mg/m^2**	**Cr, mg/m^2**	**Mn, mg/m^2**	**Fe, mg/m^2**	**Ni, mg/m^2**	**Cu, mg/m^2**	**Zn, mg/m^2**	**Cd, mg/m^2**	**Pb, mg/m^2**	**TSS, mg/m^2**	**TOC, mg/m^2**
Hope Island Rd CH	13.18±4.8	0.06±0.05	0.48±0.11	39.58±4.3	0.06±0.03	1.20±0.09	2.53±0.21	0.52±0.48	0.34±0.02	260.27±19.2	39.49±8.1
Shipper Dr IS	7.26±3.2	0.04±0.02	0.40±0.31	15.67±4.5	0.02±0.01	2.46±1.21	3.22±1.4	0.52±0.48	0.93±0.09	186.16±31.8	14.85±6.3
Billinghurst Cres RB	13.17±7.2	0.04±0.11	0.40±0.08	33.75±5.9	0.02±0.01	1.44±0.04	3.57±0.9	0.52±0.48	0.52±0.05	469.07±33.8	39.78±17.2
Peanba Park RP	3.33±0.69	0.11±0.05	0.11±0.09	9.51±2.8	0.14±0.11	0.76±0.05	1.12±0.7	0.52±0.48	0.22±0.1	62.43±5.7	12.18±5.9
Dalley Park RD	36.74±10.1	0.07±0.01	2.63±0.98	110.58±3.6	0.10±0.03	2.86±0.94	4.01±0.29	0.42±0.29	1.21±0.61	1097.87±140.8	52.63±6.7
Lindfield Rd CL	1.96±0.38	0.04±0.02	0.05±0.01	4.91±3.1	0.03±0.01	2.15±0.25	2.12±1.1	0.52±0.48	0.59±0.07	55.01±12.1	15.03±5.4
Town Centre Dr CT	1.74±0.92	0.11±0.09	0.03±0.01	2.89±2.1	0.06±0.01	0.76±0.08	1.43±0.52	0.52±0.48	0.29±0.07	71.20±20.2	11.05±3.8
Abraham Rd RA	6.91±1.2	0.09±0.03	0.26±0.11	14.61±2.6	0.08±0.01	1.01±0.04	2.36±0.22	0.52±0.48	0.29±0.06	212.00±33.9	19.70±3.7
Discovery Dr RDS	2.66±0.94	0.03±0.01	0.06±0.01	4.96±1.4	0.20±0.08	0.83±0.06	2.25±0.49	0.52±0.48	0.48±0.11	67.20±12.5	17.69±4.9
Beattie Rd IBT	1.35±0.32	0.11±0.09	0.06±0.04	3.71±1.1	0.27±0.01	0.89±0.71	1.61±1.1	0.52±0.48	0.38±0.08	63.20±15.7	11.06±5.1
Reserve Rd RR	13.11±3.2	0.24±0.12	0.42±0.15	23.98±2.8	0.48±0.09	1.56±0.91	2.59±0.71	0.52±0.48	0.34±0.09	644.27±51.1	28.62±1.8

Table S6 Chemical compositions (mean±standard deviations) of the wash-off of Heavy metals for the simulated rain events

Rain Events*	Al, mg/L	Cr, mg/L	Mn, mg/L	Fe, mg/L	Pb, mg/L	Ni, mg/L	Cu, mg/L	Zn, mg/L	Cd, mg/L	TSS, mg/L	TOC, mg/L	pH	EC, micro-siemens/cm
1	4.13±1.6	0.02±0.01	0.02±0.01	1.01±0.21	0.09±0.02	0.02±0.01	0.34±0.1	1.65±0.82	0.20±0.06	30.60±8.1	9.56±3.6	7.08±0.03	56.80±0.01
2	0.78±0.11	0.02±0.01	0.01±0.005	0.75±0.1	0.10±0.05	0.03±0.01	0.14±0.05	1.11±0.32	0.20±0.06	31.10±3.8	9.69±1.5	7.08±0.02	53.70±0.05
3	0.70±0.13	0.03±0.01	0.02±0.005	0.75±0.09	0.09±0.01	0.04±0.01	0.21±0.08	2.03±0.12	0.20±0.02	35.00±10.2	8.78±1.8	7.01±0.01	43.20±0.04
4	0.63±0.12	0.02±0.01	0.02±0.005	0.80±0.06	0.05±0.01	0.09±0.02	0.38±0.11	10.46±2.1	0.16±0.05	36.10±11.3	7.64±4.2	7.06±0.02	42.00±0.06
5	1.48±0.05	0.02±0.01	0.04±0.01	0.83±0.07	0.05±0.01	0.04±0.01	0.40±0.1	11.17±1.32	0.16±0.09	20.40±9.2	7.69±0.98	7.06±0.01	39.30±0.02
6	0.33±0.09	0.01±0.005	0.02±0.01	0.74±0.12	0.08±0.02	0.01±0.005	0.30±0.05	9.81±2.2	0.16±0.04	16.70±7.1	7.27±2.4	7.07±0.01	38.30±0.02
7	1.16±0.25	0.02±0.01	0.02±0.01	1.44±0.12	0.05±0.01	0.01±0.005	0.26±0.09	10.21±2.4	0.16±0.04	60.70±18.2	28.60±5.4	7.01±0.01	74.80±0.02
8	0.81±0.21	0.02±0.01	0.02±0.01	1.58±0.1	0.04±0.01	0.01±0.005	0.29±0.06	7.80±2.1	0.16±0.04	69.80±18.1	26.22±5.9	7.01±0.01	63.40±0.02
9	4.33±1.2	0.02±0.01	0.02±0.01	1.33±0.09	0.06±0.02	0.01±0.005	0.25±0.12	8.62±3.2	0.16±0.01	24.86±3.6	21.18±6.4	7.10±0.01	44.80±0.01
10	0.80±0.38	0.03±0.01	0.02±0.01	1.53±0.1	0.04±0.01	0.04±0.01	0.17±0.04	7.82±1.05	0.16±0.04	30.40±12.1	18.17±8.1	7.09±0.02	38.70±0.01
11	0.53±0.07	0.03±0.01	0.05±0.01	1.02±0.3	0.08±0.04	0.03±0.01	0.24±0.06	2.50±1.3	0.20±0.03	22.10±10.7	16.25±8.6	7.09±0.03	33.80±0.01
12	0.48±0.1	0.09±0.03	0.05±0.01	0.96±0.04	0.13±0.01	0.06±0.01	0.25±0.08	2.22±0.95	0.16±0.01	15.80±6.5	12.92±6.4	7.07±0.01	28.00±0.01
13	0.43±0.2	0.05±0.02	0.03±0.01	0.66±0.4	0.10±0.06	0.06±0.02	0.24±0.09	1.93±0.98	0.20±0.01	20.00±7.2	8.71±1.1	7.06±0.01	28.10±0.01
14	0.18±0.05	0.03±0.01	0.04±0.01	0.54±0.23	0.13±0.06	0.03±0.01	0.16±0.06	1.52±0.31	0.20±0.06	20.20±7.8	17.57±4.3	7.04±0.01	23.20±0.02
15	0.56±0.1	0.03±0.01	0.03±0.01	0.62±0.21	0.11±0.02	0.06±0.02	0.17±0.1	1.42±0.12	0.20±0.05	28.20±3.4	15.98±3.5	7.00±0.01	23.80±0.02
16	0.40±0.08	0.03±0.01	0.01±0.005	0.52±0.2	0.10±0.05	0.09±0.01	0.16±0.03	1.49±0.15	0.20±0.05	26.30±2.5	16.44±4.2	7.02±0.01	23.70±0.02
17	0.45±0.05	0.03±0.01	0.01±0.005	0.64±0.12	0.07±0.02	0.03±0.01	0.17±0.06	1.11±0.14	0.20±0.05	16.10±9.3	14.42±2.6	7.01±0.01	22.80±0.01
18	0.26±0.06	0.02±0.01	0.01±0.005	0.55±0.41	0.14±0.05	0.06±0.02	0.19±0.01	1.29±0.24	0.20±0.02	21.80±1.4	13.70±2.8	6.99±0.01	23.50±0.02
19	0.27±0.06	0.04±0.02	0.02±0.01	0.60±0.31	0.07±0.01	0.08±0.02	0.17±0.09	1.68±0.31	0.20±0.04	35.10±1.8	15.82±3.2	7.16±0.01	33.40±0.03
20	0.31±0.01	0.03±0.01	0.01±0.005	0.51±0.12	0.13±0.04	0.08±0.02	0.17±0.06	1.30±0.34	0.20±0.04	20.10±6.4	12.20±6.7	7.20±0.02	29.40±0.06
21	0.26±0.05	0.05±0.02	0.02±0.01	0.46±0.13	0.09±0.01	0.11±0.03	0.19±0.08	1.31±0.25	0.20±0.07	12.50±6.7	9.36±2.8	7.19±0.02	25.90±0.02
22	0.55±0.1	0.03±0.01	0.03±0.005	0.42±0.11	0.13±0.05	0.11±0.05	0.18±0.06	1.16±0.24	0.20±0.07	18.60±3.1	10.39±4.1	7.17±0.03	25.40±0.02

*Numerical object identifiers used elsewhere are same as rain events numbers

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