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# Time as the critical factor in the investigation of the relationship between pollutant wash-off and rainfall characteristics

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**Abstract:** The approach adopted for investigating the relationship between rainfall characteristics and pollutant wash-off process is commonly based on the use of parameters which represent the entire rainfall event. This does not permit the investigation of the influence of rainfall characteristics on different sectors of the wash-off process such as first flush where there is a high pollutant wash-off load at the initial stage of the runoff event. This research study analysed the influence of rainfall characteristics on the pollutant wash-off process using two sets of innovative parameters by partitioning wash-off and rainfall characteristics. It was found that the initial 10% of the wash-off process is closely linked to runoff volume related rainfall parameters including rainfall depth and rainfall duration while the remaining part of the wash-off process is primarily influenced by kinetic energy related rainfall parameters, namely, rainfall intensity. These outcomes prove that different sectors of the wash-off process are influenced by different segments of a rainfall event.

**Keywords:** Pollutant wash-off; Water Sensitive Urban Design; Stormwater quality; Stormwater pollutant processes; Multivariate analysis

# **1** Introduction

It is commonly known that pollutant wash-off is strongly influenced by rainfall characteristics (Kleinman et al., 2006; Greenstein et al., 2004; Egodawatta et al., 2007; Lindblom et al., 2011; Freni et al., 2008). Consequently, there has been significant research effort on investigating the relationships between the pollutant wash-off and rainfall characteristics in order to enhance the effectiveness of stormwater treatment system design such as Water Sensitive Urban Design (WSUD) (For example Miguntanna et al., 2013; Liu et al., 2012; Wium-Andersen et al., 2013). These research studies have used stormwater quality data such as event mean concentrations (EMC) and common rainfall parameters such as average rainfall intensity (Shigaki et al., 2007; Liu et al., 2012; Berndtsson et al., 2009). Unfortunately, this approach represents a significant limitation as parameters such as these

represent the entire rainfall event (lumped parameters) and does not permit the investigation of the influence of rainfall characteristics on different sectors of the wash-off process.

It is hypothesised that different sectors of the wash-off process are influenced by different segments of a rainfall event. This hypothesis needs to be viewed in the context of the occurrence of the first flush phenomenon, which refers to the wash-off of a relatively higher pollutant load at the initial part of a runoff event (Lee et al., 2002; Li et al., 2007). Accordingly, it can be argued that the common approach of using lumped rainfall parameters could overshadow the critical relationships between pollutant wash-off and rainfall characteristics and hence cannot provide an in-depth understanding of the wash-off process. For investigating the different sectors of the wash-off process, the research study discussed in this paper adopted an innovative approach by partitioning the wash-off process and rainfall characteristics in order to create new knowledge to enhance the effectiveness of stormwater treatment system design.

# 2 Methods and materials

# 2.1 Study sites

Data required for the study was generated by monitoring residential catchments located at Coomera Waters, Gold Coast, Australia. Coomera Waters is a residential estate developed around a 17 ha lake and natural wetlands based on Water Sensitive Urban Design (WSUD) principles in order to protect the natural waterways from being polluted by stormwater runoff. Three small catchments in Coomera Waters were selected as the study catchments and named as Catchment A, Catchment B and C. Details of the catchment characteristics are given in Figure 1.

## 2.2 Data collection and testing

The three catchments have been continuously monitored for stormwater quality and quantity and rainfall since 2007 using automatic monitoring stations established at the outlets to collect flow measurements and stormwater runoff samples and pluviograph stations for rainfall data collection. The data collection procedures are outlined in the Supplementary Information. The samples collected were tested for total nitrogen (TN), total phosphorus (TP) and total suspended solids (TSS) which are the primary stormwater pollutants (Goonetilleke et al., 2005; Qin et al., 2010; Collins et al., 2010; Francey et al., 2010; Dierberg et al., 2008). Sample testing was undertaken according to test methods specified in Standard Methods for the Examination of Water and Wastewater (APHA, 2005). Field blanks and laboratory blanks were used as part of the QA-QC procedure. Sample collection, transport and storage complied with Australia New Zealand Standards, AS/NZS 5667.1:1998 (AS/NZS, 1998).

### 2.3 Determination of innovative parameters

Two sets of innovative parameters were derived as detailed in the Supplementary Information to meet the needs of the envisaged investigation. These parameters which represented the pollutant load percentages washed-off by different sectors of runoff volume are described below.

### Interval parameters

The interval wash-off parameter (termed as LV) is the cumulative pollutant load percentage for every 10% of runoff volume interval until 90% of runoff volume. The LV parameter was selected in order to compare rainfall events, based on temporal variations. For example, LV20 is the cumulative pollutant load percentage washed-off from the beginning until 20% of the runoff volume. The same approach was used to calculate interval rainfall parameters for every 10% of the rainfall hyetograph up to 90%. As examples, RDep20 and RD20 are the

rainfall depth and rainfall duration from the commencement of rain until 20% of effective rainfall, respectively, while the AI20 gives the average intensity from the commencement of rainfall to 20% of effective rainfall. RDep is rainfall depth; RD is rainfall duration and AI is the average intensity. Accordingly, the interval wash-off and rainfall parameters determined are LV10-LV90, RDep10-RDep90, RD10-RD90 and AI10-AI90.

### Section parameters

The section wash-off parameter (termed as P) is the increment in pollutant load percentage washed-off at 10% of runoff volume interval while the section rainfall parameters represented the rainfall characteristics at 10% of effective rainfall interval. P parameter was selected in order to investigate the variations in the wash-off process within a rainfall event. For example, P2030 represents the percentage of pollutant load washed-off between 20% and 30% of runoff volume; RDep2030 and RD2030 give the rainfall depth and rainfall duration between 20% and 30% of effective rainfall, respectively; AI2030 represents the average rainfall intensity which occurred between 20% and 30% of the effective rainfall depth. Accordingly, the section wash-off and rainfall parameters are P0010-P8090, RDep0010-RDep8090, RD0010-RD8090 and AI0010-AI8090.

# 3. Results and discussion

### 3.1 Univariate data analysis

Table 1 gives the mean and relative standard deviation (RSD) values for LV and P for TSS, TP and TN for the 23 rainfall events that were investigated in the study. In terms of mean values of LV, most pollutant loads are washed-off by the first 40% of runoff volume regardless of the pollutant type (LV40 is 61.22%, 60.74% and 52.43% for TSS, TP and TN respectively). From the perspective of first flush, this observation means that the occurrence and characteristics of first flush could be determined by assessing the pollutant loads in the first 40% of the runoff volume.

For RSD, it can be noted that the corresponding values generally decrease from LV10 to LV90 for all three pollutants, where the RSD values are larger than 10% for up to LV60 (except for LV50 of TN). Since a data set with RSD greater than 10% is considered as having a high variability (Hamburg, 1994), this means that there is high variation in the wash-off process with rainfall characteristics, particularly the pollutant loads carried by the first 60% of runoff volume.

In terms of P parameters, the mean values reduce from P0010 to P8090 for TSS, TP and TN, which indicates that relatively higher percentage of pollutant loads are washed-off by the initial portion of the runoff volume (first flush phenomenon). It is also noteworthy that the RSD values are relatively higher for the initial P parameters (P0010) and the later ones (from P5060 to P8090), while the RSD values for the middle P parameters (from P1020 to P4050) are relatively lower.

These observations derived from the LV and P data suggest that different sectors of the washoff process behave differently and their variations with rainfall characteristics are also different. It is important to understand the reasons for differences in wash-off for rainfall events with different characteristics and for different segments in a rainfall event. This underlines the inadequacy of investigating pollutant wash-off using lumped rainfall parameters and the importance of further investigating the relationship between different sectors of the wash-off process and different segments of a rainfall event.

# 3.2 Multivariate data analysis

In order to undertake a comprehensive investigation of the relationship between rainfall characteristics and the wash-off process, the two innovative sets of rainfall parameters discussed above were derived. Additionally, lumped rainfall parameters (for the entire rainfall events) were also included in the analysis. These were, the total rainfall depth (TRDep), average rainfall intensity (AgI), total rainfall duration (TRD) and antecedent dry period (ADP), which are considered as important rainfall characteristics influencing the wash-off process (Liu et al., 2012, Kleinman et al., 2006).

Principal Component Analysis (PCA) was used to explore the influence of rainfall characteristics on pollutant wash-off due to its versatility for identifying correlations among variables and objects (Kokot et al., 1998). An introduction to PCA including the interpretation of PCA biplot is detailed in the Supplementary Information. StatistiXL software (StatistiXL, 2007) was used for PCA. Analysis of interval parameters and section parameters were undertaken separately using PCA in order to individually investigate their ability to derive an in-depth understanding of the wash-off process.

### **3.2.1** Analysis of interval parameters

A data matrix  $(23\times58)$  was submitted to PCA. The objects were the 23 monitored rainfall events while the variables were the nine interval wash-off parameters (LV) for TSS, TN and TP, the nine interval rainfall parameters for rainfall depth (RDep), rainfall duration (RD) and rainfall intensity (AI) and the four common rainfall parameters, TRDep, AgI, TRD and ADP. According to the Scree plot method, the first three PCs were selected. Figure 2 gives the resulting PCA biplots, PC1 vs. PC2 and PC1 vs. PC3.

It can be noted that all LV parameters are divided into two parts regardless of the pollutant type. As evident in the PC1 vs. PC2 biplot (Figure 2A), most of LV10-LV50 vectors are projected on the negative PC1 axis while most of LV60-LV90 vectors are on the positive PC1 axis. In the PC1 vs. PC3 biplot (Figure 2B), all initial LV parameters (LV10-LV50) are projected on the positive PC3 axis while most of LV60-LV90 vectors are on the negative PC3 axis. Additionally, it can be observed from Figure 2A that different LV parameters are correlated with different rainfall characteristics as the LV parameter vectors are in different directions and accordingly correlate with different rainfall characteristics vectors.

The initial LV parameter such as LV10, LV 20 and LV30 tend to relate to runoff volume related rainfall characteristics such as rainfall duration and rainfall depth while the later LV parameters such as LV80 and LV90 are correlated to kinetic energy related rainfall characteristics such as rainfall intensity. Antecedent dry period tends to correlate to the middle and later LV parameters including LV 60, LV70 and LV80. These outcomes confirm that the different sectors of the pollutant wash-off process are related to different rainfall characteristics.

Although the analysis of LV parameters indicated the variability of pollutant wash-off processes with rainfall characteristics, it still provides limited ability to interpret the relationship between different sectors of the wash-off process with different segments of a rainfall event due to the use of cumulative parameters. This highlighted the need for more detailed analysis of section wash-off and rainfall parameters.

### **3.2.2 Analysis of section parameters**

The data matrix  $(23 \times 58)$  was submitted to PCA. The objects were the 23 rainfall events monitored and the variables were the nine section wash-off parameters (P) for TSS, TN and

TP, the nine section rainfall parameters (RDep, RD and AI) and the four common rainfall parameters for the entire events (TRDep, AgI, TRD and ADP). Based on the Scree plot method, the first three PCs were selected. Figure 3C and 3D give the resulting PCA biplots, including PC1 vs. PC2 and PC1 vs. PC3. According to the PCA biplots, all P parameters and section rainfall parameters are separated into three clusters independent of the pollutant type.

<u>Cluster 1</u>: P0010 vectors for TSS, TP and TN are projected on the positive PC1 axis, on which most of the rainfall depth and duration section parameters (RDep and RD) and parameters for the total depth (TRDep) and duration (TRD) are also projected. The different behaviour of the different sectors of the wash-off process is also evident in the PC1 vs. PC3 biplot (Figure 3B). For example, P0010 vectors for TSS, TP and TN are in different directions compared to the middle sectors of the wash-off process such as P2030 and P3040. This means that the initial 10% of the wash-off process is significantly influenced by rainfall depth and duration. This is attributed to the weakly adhered solids available (free solids) on the catchment surface which will undergo wash-off easily (Miguntanna et al., 2013). Accordingly, high rainfall depth and long duration resulting in high runoff volume can wash-off a relatively larger pollutant load. This implies that high rainfall depth and long duration events tend to produce a high magnitude first flush.

<u>Cluster 2</u>: Most of P1020-P5060 vectors for TN, TP and TSS (except for TSSP5060) are projected on the positive PC2 axis, on which the antecedent dry period (ADP) and AI0010 vectors are also projected. These observations mean that rainfall intensity in the initial 10% of effective rainfall (AI0010) exerts a different influence on pollutant wash-off from the remaining section rainfall intensity parameters (AI1020-AI8090). AI0010 plays an important role in influencing up to almost the first 60% of the overall pollutant wash-off process while the rainfall intensity after 10% of effective rainfall primarily affects the later sectors of the wash-off process such as P6070-P8090. Additionally, the antecedent dry period primarily affects the initial sectors of the wash-off process such as P1020-P5060. This indicates that the pollutants built-up during the dry period has a more significant influence on the early part of the wash-off process.

<u>Cluster 3</u>: P6070-P8090 vectors for TSS, TP and TN and TSSP5060 are projected on the negative PC2 axis, on which AI1020-AI8090 and average rainfall intensity (AgI) are also projected. This implies that the rainfall intensity after 10% of effective rainfall has a significant influence on the later sectors of the wash-off process while intensity in the initial 10% of effective rainfall has no significant impact on the end sectors of the wash-off process. As the weakly adhered solids diminish after the early stages of the rainfall-runoff event, the strongly adhered solids which depend on the magnitude of the rainfall kinetic energy to detach, are exposed. Therefore, the pollutant load detached towards the end part of a runoff hydrograph is strongly dependent on the rainfall kinetic energy.

#### 3.2.3 Summary

The above analysis highlights the importance of investigating the relationship between the wash-off process and rainfall characteristics in segments. It was found that different sectors of the wash-off process are influenced by different segments in a rainfall event. This implies that the use of lumped rainfall parameters representing an entire rainfall event is inappropriate to investigate the influence of rainfall characteristics on wash-off. This conclusion applies particularly in the case of rainfall intensity, since the intensity during the initial 10% and after 10% of effective rainfall exerts completely different influences on the wash-off process.

# **4** Conclusions

This paper presents the outcomes of a research study into the relationship between the pollutant wash-off process and rainfall characteristics using two sets of innovative parameters which partitioned wash-off and rainfall characteristics, as interval and section parameters. It was found that the initial 10% sector of the wash-off process is closely related to the runoff volume related rainfall characteristics including rainfall depth and duration while the remaining sectors are primarily influenced by kinetic energy related characteristics, namely rainfall intensity. Additionally, it was noted that lumped characteristics representing the entire rainfall event is inappropriate to investigate the influence exerted by rainfall characteristics on pollutant wash-off. This is particularly the case for rainfall intensity, as the intensity in the initial 10% and after 10% of effective rainfall exerts completely different influences on the wash-off process. In this context, the use of innovative parameters proposed in this study such as interval and section parameters can provide an in-depth understanding of the pollutant wash-off process. This in turn will enhance the design of stormwater treatment systems.

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Interval		LV10	LV20	LV30	LV40	LV50	LV60	LV70	LV80	LV90
TSS	Mean%	20.78	36.00	49.30	61.22	71.35	78.96	85.74	91.87	96.61
	RSD%	49.01	31.70	23.20	19.44	16.48	13.80	9.11	5.29	2.20
ТР	Mean%	22.43	36.91	50.30	60.74	70.96	77.91	84.57	91.13	96.30
	RSD%	55.87	34.15	25.13	17.57	13.18	10.88	6.96	4.11	2.13
TN	Mean%	15.13	28.30	40.65	52.43	63.22	73.91	81.83	89.39	95.61
	RSD%	37.86	19.75	15.48	11.70	9.52	10.50	7.83	4.78	2.26
Section										
Se	ection	P0010	P1020	P2030	P3040	P4050	P5060	P6070	P7080	P8090
	ection Mean%	<b>P0010</b> 20.78	<b>P1020</b> 15.22	<b>P2030</b> 13.30	<b>P3040</b> 11.91	<b>P4050</b> 10.13	<b>P5060</b> 7.61	<b>P6070</b> 6.78	<b>P7080</b> 6.13	<b>P8090</b> 4.74
Se TSS										
TSS	Mean%	20.78	15.22	13.30	11.91	10.13	7.61	6.78	6.13	4.74
	Mean% RSD%	20.78 49.01	15.22 37.12	13.30 28.68	11.91 27.22	10.13 27.35	7.61 40.23	6.78 52.70	6.13 56.98	4.74 64.73
TSS TP	Mean% RSD% Mean%	20.78 49.01 22.26	15.22 37.12 14.48	13.30 28.68 13.39	11.91 27.22 10.43	10.13 27.35 10.22	7.61 40.23 6.96	6.78 52.70 6.65	6.13 56.98 6.57	4.74 64.73 5.17
TSS	Mean% RSD% Mean% RSD%	20.78 49.01 22.26 53.21	15.22 37.12 14.48 31.74	13.30 28.68 13.39 30.97	11.91 27.22 10.43 36.50	10.13 27.35 10.22 47.42	7.61 40.23 6.96 113.46	6.78 52.70 6.65 48.68	6.13 56.98 6.57 41.61	4.74 64.73 5.17 38.51

Table 1 Interval and section wash-off data

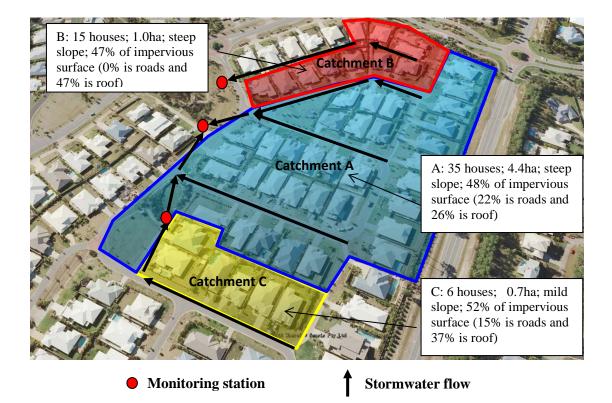


Figure 1 Study catchments and their characteristics

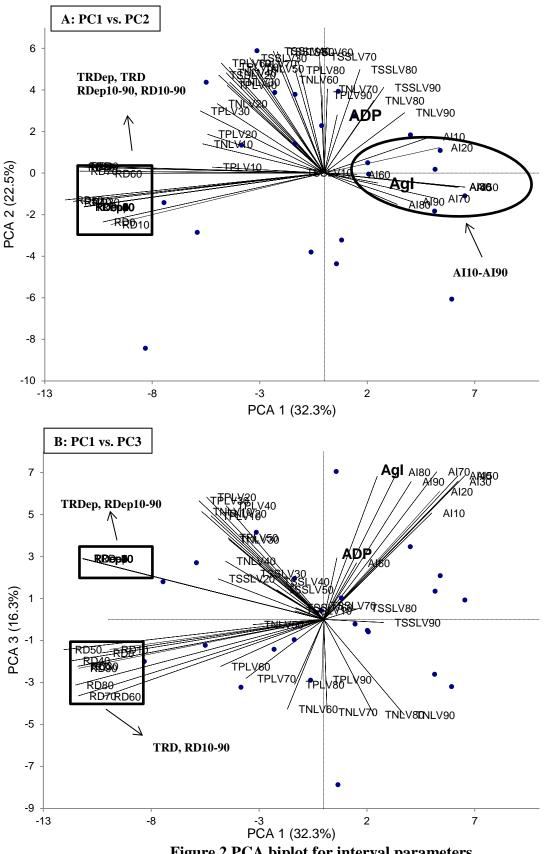
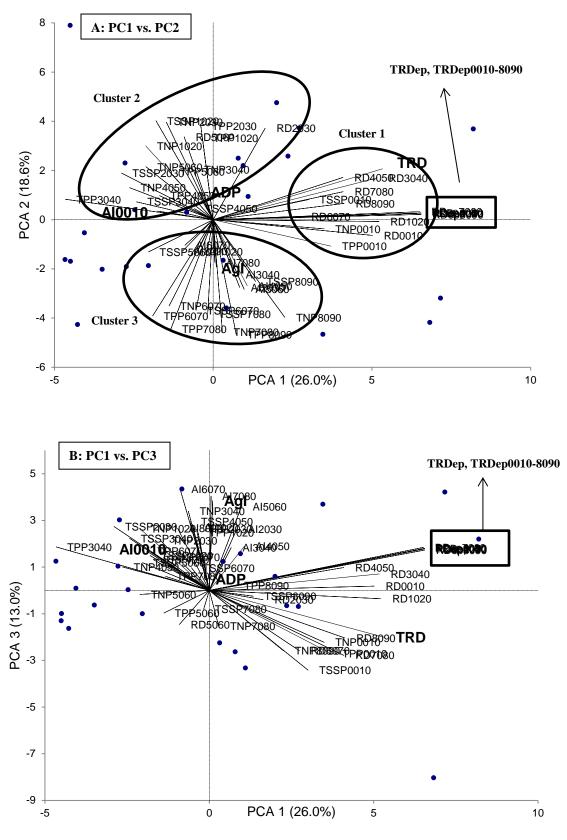
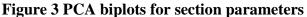


Figure 2 PCA biplot for interval parameters

(TRDep=total rainfall depth; AgI=average rainfall intensity; TRD=total rainfall duration; ADP=antecedent dry period; TRDep10-90=interval rainfall depth; TRD10-90=interval rainfall duration; AI10-90=interval average intensity)





(TRDep=total rainfall depth; AgI=average rainfall intensity; TRD=total rainfall duration; ADP=antecedent dry period; TRDep0010-8090=section rainfall depth; TRD0010-8090=section rainfall duration; AI0010-8090=section average intensity)

# Time as the critical factor in the investigation of the relationship between pollutant wash-off and rainfall characteristics

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# The selected rainfall events and their characteristics

A total of 13 rainfall events were selected for analysis after assessment of the available data. However, suitable rainfall-runoff records and water quality data for the 13 rainfall events was not available for all of the three catchments. Three events were common for all three catchments, while the rest of the events comprised of data suitable for one or two catchments. Accordingly, the total number of sampling episodes selected for analysis totalled 23 rainfall events. Rainfall characteristics are given in Table S1.

Event ID	Total rainfall depth	Average intensity	Antecedent dry period	Catchments
	( <b>mm</b> )	( <b>mm/h</b> )	( <b>h</b> )	
1	1.4	6	23.65	A, B, C
2	1.4	21	9.24	А
3	0.8	12	216	А
4	2.2	3	164	A, B, C
5	3.2	7.38	396	A, B
6	3.2	4	24	A, B, C
7	2.6	13	202	A, B
8	4.2	18	9	А
9	5.8	7.9	3	А
10	0.8	8	170.4	B, C
11	0.8	1.6	20	С
12	5.8	6.69	16	С
13	0.6	1.833	4	B, C

#### **Table S1 Rainfall characteristics and applicable catchments**

### Stormwater sampling and runoff volume data collection

### Stormwater sample collection

Stormwater samples were collected using automatic samplers, which were programmed to collect discrete samples. This was to investigate the variation in water quality during a runoff event. The samples were collected in 1L plastic bottles and up to 24 bottles were collected. In this research study, the automatic samplers have been set to collect samples based on predefined flow where the samplers are triggered once the flow reaches a pre-set depth at the weir. The sampling time was recorded and then transferred to the computer. The collected samples were transported back to the laboratory for analysis, normally within 24 hours.

#### **Runoff volume measurement**

A V-notch weir was used to measure the runoff volume at the catchment outlet. It was installed either in the stormwater culvert or at the inlet of the WSUD devices such as the bioretention basin at Catchment C. The weir apparatus consisted of a weir box, baffles, face plate and pressure transducer. A weir box with dimensions of 400mm length, 30mm wide and 30mm high made of UV resistant plastic was used to measure the runoff volume. The use of a plastic weir box enabled the installation and calibration process to be carried out efficiently. Baffles were installed towards the back of the weir box to reduce flow velocity and create a still water body over the water level sensor in the weir box.

## Principal Component Analysis (PCA)

Principal Component Analysis (PCA) is performed on transformed data by reducing a set of raw data to a number of principal components (PCs). PC1 describes the largest data variance and PC2 the next largest data variance and so on. There are as many PCs as the number of variables, but most of the variance is accounted for in the first few PCs (Adams, 1995). Each object is identified by a score, and each variable by a loading value or weighting. The data display may be obtained by plotting (i) PCi vs PCj scores (score plot, i, j = PC number), (ii) loadings for a given PC (loadings plot) and (iii) scores and loading vectors on the one plot (biplot). The various display plots indicate relationships between objects, the significance of variables on each PC, and correlations between objects and variables. This analytical method can provide useful guidance regarding the relationship of objects and variables in a data matrix.

In the PCA biplot, the variables are considered as correlated when the angles between the vectors are small. An obtuse angle indicates a weak correlation. An angle of 90° is considered as uncorrelated parameters and 180° as inversely correlated. Objects with similar characteristics make clusters and are strongly correlated to the variables when their vectors point in the same direction as the variables.

### Development of interval and section parameters

The cumulative pollutant load vs. cumulative runoff volume was plotted in order to develop the innovative parameters. Prior to plotting, the rainfall events were carefully selected. An important criterion in the selection of rainfall events was the availability of sufficient water quality samples during the occurrence of the runoff hydrograph. Since water quality is relatively more variable during the rising limb of the hydrograph and decreases relatively consistently along the falling limb, it was important that sufficient water quality samples were available for the rising limb. In addition to that, the relatively small study catchment area and its 'flashy characteristics' result in accelerated response time to pollutant peak concentration. Therefore, the accurate depiction of the pollutant peak required samples to be collected at short time intervals and the availability of a sufficient number of samples. Accordingly, it was specified that a minimum of five water quality samples should have been collected for any selected runoff event.

The laboratory test results of runoff samples were in the form of concentration. The pollutant loads were obtained by multiplying the pollutant concentrations with corresponding runoff volumes. The cumulative pollutant loads and runoff volumes were derived to form the cumulative pollutant load vs. cumulative runoff volume plot. Both interval and section parameters were determined from this plot (see Figure S1).

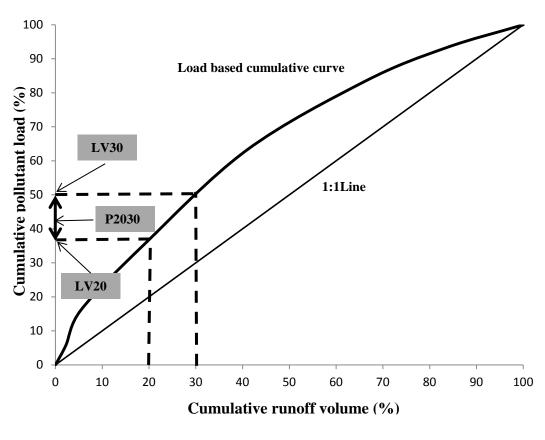


Figure S1

## References

Adams, M.J., 1995. Chemometrics in Aanlytical Chemistry. The Royal Society of Chemistry, Cambridge.