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Understanding nutrient dynamics for effective stormwater treatment design

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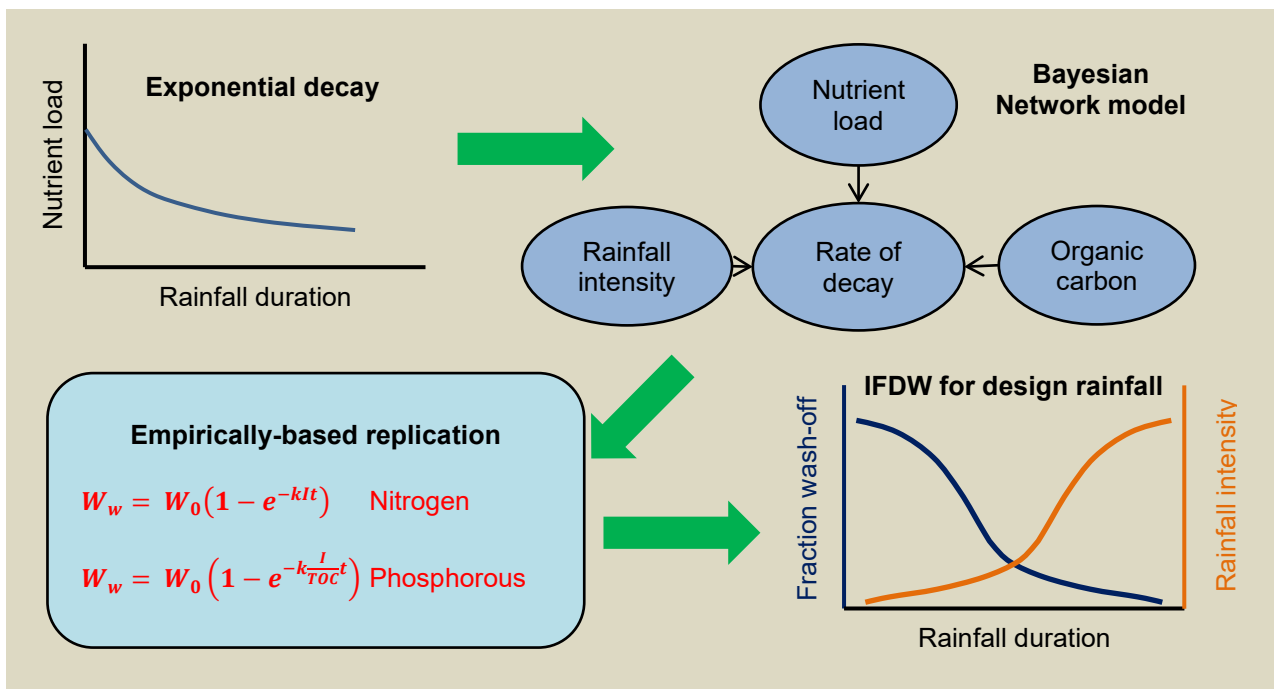
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highlights

- Empirically-based, statistical modelling integrated to replicate nutrient dynamics
- Nitrogen species wash-off at similar rates
- Phosphorous species can wash-off at different rates
- Created Intensity-Frequency-Duration-Wash-off (IFDW) distributions
- IFDW can be applied as a reference tool for designing stormwater treatment measures

Graphical Abstract



Abstract:

Current stormwater quality modelling tools lack robust mathematical replication of nutrient entrainment in runoff. This makes it challenging to design effective stormwater treatment systems such as nature based solutions with adequate resilience to future changes in nutrient inputs in urban environments. Consequently, poorly treated stormwater can be discharged into receiving waters, leading to nutrient enrichment and in turn, environmental and human health impacts. This study integrated empirically based with statistical modelling techniques to incorporate nutrient dynamics into commonly used Intensity-Frequency-Duration (IFD) distributions of design rainfall. Field based nutrient wash-off experiments were conducted to understand nutrient behaviour during a runoff event. New mathematical formulations were derived to describe the decay (wash-off) of nutrients. Rainfall intensity, duration and initially accumulated pollutant load exert positive influence on the decay of nitrogen and phosphorous, while organic carbon has a negative impact on phosphorus decay. It was also evident that nitrogen species would decay at a similar rate, while phosphorus species may decay at different rates. Compared to nitrogen species, phosphorous species were found more likely to be washed-off during a rainfall event. Using the mathematical formulations developed, wash-off of nitrogen and phosphorous was simulated for 435 very frequent and frequent/infrequent design rainfall events leading to the creation of Intensity-Frequency-Duration-Wash-off (IFDW) curves. Analysis of uncertainty associated with IFDW indicated that total phosphorous could be completely washed-off during most of the design rainfall events, while total nitrogen would only be completely washed-off by very few events that are rarer than 10% AEP (annual exceedance probability). IFDW can act as a tool for supporting effective stormwater treatment design in order to promote sustainable stormwater management and reuse.

Keywords: Design rainfall; Stormwater pollutants; Stormwater management; Stormwater quality; Stormwater pollutant processes

1. Introduction

Current stormwater quality modelling tools lack robust mathematical replications of the generation and dispersion of nutrients during dry and wet weather periods. Even though some of the advanced and emerging techniques such as machine learning have been found to be effective in replicating nutrient processes in runoff, these tools present computational limitations and have been tested for their preference using only limited field-based experimental data (Gorgoglione et al., 2021). Further, commercially available modelling tools such as Mike Urban and SWMM have limited capability for replicating variability in pollutant processes and often suffer from a lack of reliable data (MikeUrban, 2017; Rosa et al., 2015). While similar knowledge gaps in relation to particulate solids and toxicants such as heavy metals have been addressed in the past (Duncan, 1995; Egodawatta and Goonetilleke, 2006b; Li et al., 2018; Liu et al., 2015), research in relation to nutrient processes has shown poor progress. Moreover, research on fundamental pollutant processes such as build-up and wash-off typically focuses on particulate solids, while assuming similar behaviour for other pollutants attached to solids (Liu et al., 2010). However, outcomes of recent studies show particle-bound pollutants could exhibit behaviour different from those of solids during runoff events as a result of adsorption and desorption processes (Gunawardana et al., 2014). Consequently, lack of understanding of how load and composition of nutrients vary while undergoing build-up and wash-off creates significant challenges in designing pollution mitigation measures (Goonetilleke et al., 2005; Hvitved-Jacobsen et al., 2010).

Nutrients are pollutants that are discharged from a range of urban land uses via sources such as domestic waste, excess fertiliser usage and vehicle exhaust. Once entrained in runoff, nutrients can be transported to and enrich receiving waters, resulting in eutrophication. This not only poses risks to aquatic ecosystem health, but also reduces the potential of stormwater as an alternative water resource compared to relatively more expensive sources that also have a high carbon footprint such as desalinated sea water and recycled wastewater (Adams et al., 2020; Lapointe et al., 2020; Segurado et al., 2018; Wijesiri et al., 2020). Therefore, the control of generation, transport and dispersion of nutrients in urban environments is critical for safeguarding the quality of water resources (Nixdorff et al., 2021; Miguntanna et al. 2013), for enhancing urban liveability and for enhancing the reuse potential of urban stormwater (Delkash et al., 2018; Van Puijenbroek et al., 2019). In order to mitigate risks to urban water bodies, nutrients in runoff should be treated before being discharged into creeks and rivers (Walaszek et al., 2018). In this context, stormwater modelling plays a key role as the primary tool used for simulating stormwater quality for treatment system design (Obropta and Kardos, 2007; Tsihrintzis and Hamid, 1997; Zoppou, 2001).

There is a wealth of studies that have investigated a range of treatment systems such as nature-based solutions, relying on historical patterns of nutrient inputs (Barron et al., 2019; Wang et al., 2021; Zhang et al., 2021). While novel measures such as nature based solutions have proven to be effective in treating nutrients, there is an opportunity to improve their long-term resilience if potential changes to nutrient loads in runoff can be predicted into the future. The nutrients being discharged into the urban environment is likely to change due to the increase in the urban population and changing land uses, which are inevitable in the coming decades (Delkash et al., 2018). Further, the predicted effects of climate change such as longer dry periods and intense rainfall events (IPCC, 2018) will result in higher nutrient loads entrained in runoff. Therefore, the consequences of designing and implementing mitigation measures with limited resilience can lead to ineffective performance of treatment systems in the long term. Such failures will result in the discharge of significant nutrient loads into urban receiving waters (Booth et al., 2002).

The study discussed in this paper replicated the wash-off process of nutrients (nitrogen and phosphorous compounds) by combining empirically based and statistical modelling. As such, a statistical modelling technique, namely, Bayesian Networks (BNs), was used to characterise interdependencies between nutrients and influential factors, which were then translated into empirically based functions. This in turn led to the development of Intensity-Frequency-Duration-Wash-off (IFDW) distributions. The IFDW can be applied as a reference tool for designing stormwater treatment measures. As such, the knowledge and approaches created in this study will support effective stormwater quality treatment design in order to promote sustainable stormwater management and reuse.

2. Materials and Methods

2.1 Sampling sites

Field based experiments of nutrient wash-off were undertaken in the Gold Coast region, Australia. Gold Coast is situated to the south of Brisbane, the capital of Queensland State. The region hosts five major rivers and several creeks, while high density urban areas are spread along the east coast. The occurrence of rapid urban development in the area has put natural water bodies at risk of degradation.

Three sampling sites were selected for rainfall-runoff simulations, each representing residential, commercial and industrial land uses (see Figure 1). The selection criteria included fair to good road surface conditions, adequate space and slope of road surface for gravity flow of runoff during rainfall simulation, convenient access to the site and minimum disturbance to local residents and traffic. The residential area (Armstrong Way – 61% imperviousness; 1.32% slope) consisted of detached family houses with small gardens. The gardens were well maintained which could have been the result of the frequent use of fertiliser, contributing nutrients to the road surface. The road is used by residents for access and surface condition was considered good. The commercial area (Lawrence Drive – 68% imperviousness; 1.59% slope) has a vehicle service station, a parking area, a motorcycle sales centre and food stores. The road is located very close to a motorway and one end is connected to a busy highway. Therefore, the road carries a relatively high traffic volume. The industrial area (Stevens Street – 89% imperviousness; 5.91% slope) is surrounded by many industries, including paint, furniture, metal-working and cement-based industries. The road width is approximately 8 m and the road slope is relatively steep. The road surface is in a poor condition compared to the roads in the residential and commercial areas. The road is degraded and has been subjected to oil leakages and spills due to the regular movement of heavy vehicles.



Armstrong Way 0.61
(Residential)



Stevens street 0.68
(Industrial)



Lawrence Drive 0.89
(Commercial)

Figure 1. Aerial view of the study area and street views of the sampling sites (images extracted from Google Maps/Google Earth via *nearmap*). Note: The values are the impervious surface ratios for the three sampling sites

2.2 Sample collection

The amount of pollutants that can wash-off during a rainfall event depends on the pollutant load accumulated on the road surface over the preceding dry period. Therefore, it was necessary to collect two types of samples of road deposited solids: build-up samples and wash-off samples. It is important to note that build-up and wash-off samples were collected from separate plot areas, but ensured that both plot areas would be representative of the road site.

A. Build-up sampling

The build-up samples were collected using a wet and dry vacuum system (Figure S1 in Supplementary Information) from a 3 m² plot area from each road surface. The number of antecedent dry days prior to the build-up sample collection was over seven days. This was due to the fact that the pollutants build-up load asymptotes to a constant value after seven dry days (Egodawatta and Goonetilleke, 2006a). Prior to sample collection, a field blank was prepared using de-ionised water. Then, a plot area of 3 m² (1.5m × 2m) was demarcated on the road surface using a frame (Figure S2). In order to collect representative samples, the frame was placed between the road kerb and centreline since more dust generally accumulate near a kerb rather than at the centreline. The build-up samples were collected according to following the procedure adopted in performance verification of the vacuum system (no materials were dispersed, instead actual road dust accumulated on the surface were collected). It is important to note that the duration of water spraying (3 min) had to be adjusted to avoid generation of runoff and material loss. Sample handling was conducted according to Australian/New Zealand Standards (AS/NZS, 1998). Further details on sample collection can be found in Miguntanna (2009).

B. Wash-off sampling

Wash-off samples were collected using a mechanical rainfall simulator (Figure S3 in Supplementary Information), which could simulate events with different rainfall intensities and durations. Simulated rainfall was used in order to avoid practical constraints associated with natural rainfall events such as the lack of control over intensity and duration, and uncertainty in event occurrence. The use of rainfall simulation enables the creation of a large database in a relatively short period of time. Accordingly, rainfall events at intensities of 20, 40, 65, 86, 115 and 135 mm/h were simulated. The events simulated were consistent with long-term records of typical rainfall in the Gold Coast region (BoM, 2022). The simulated rainfall events are given in Table S1 in the Supplementary Information.

Six different test plots (six 3 m² road surface areas for the six different rainfall intensities) were used at each road site. First, a field blank was prepared using de-ionised water. Then, a frame of 1.5m × 2m was placed on the road surface (Figure S4). The rainfall simulator was aligned over the plot area and the collection trough was fixed to the open end of the frame and the plot area was water-sealed using gutter tape and silicon sealant. The rainfall events were simulated for 30 min. The washed-off materials were collected in 5 min intervals from the collection trough employing the vacuum cleaner used for build-up sampling. Sample handling was conducted according to Australian/New Zealand Standards (AS/NZS, 1998). Miguntanna (2009) provides detailed information on the sample collection procedure.

Both build-up and wash-off sample collection equipment were calibrated prior to use. Further, these have been widely applied in past studies (Liu et al., 2019; Mahbub et al., 2011; Ziyath et al., 2016). Detailed information on build-up and wash-off sample collection methods can be found in the Supplementary Information.

2.3 Laboratory analyses

For each sample (build-up and wash-off), the concentrations of nitrites (NO₂⁻), nitrates (NO₃⁻), total Kjeldahl nitrogen (TKN), total nitrogen (TN), phosphates (PO₄³⁻), total phosphorus (TP), and

total organic carbon (TOC) were tested. These were determined according to the standard methods specified in APHA (2020). Standard QA and QC procedures were employed, including triplicates, field blanks and laboratory blanks, during the testing processes. The test results are given in Tables S2 – S5 in the Supplementary Information.

2.4 Data analysis

A primary objective of the data analyses was to identify the most significant factors that influence nutrient wash-off and to characterise the nature of their interdependencies. This was done by employing BNs modelling, of which the outcomes (see Section 3.1) formed the basis for developing empirically based replication of nutrient wash-off (see Section 3.2).

BNs enable visualisation of the problem being addressed, formulation of complex non-linear relationships between variables as a set of simple linear relationships, and easy interpretation of model outcomes to understand the causality. BNs firstly create a graphical structure of the system being modelled. As such, a Directed Acyclic Graph (DAG) is created by connecting a set of random variables selected based on the state-of-the-art knowledge and expert opinion. Then, BNs learn the model structure using *Structure Learning Algorithms*, considering a particular system that is defined by a set of random variables $V = \{X_1, X_2, \dots, X_6\}$ as shown in Figure 2. Accordingly, the model structure uses the *Markov Property* of BNs (Equations 1 and 2) to define a factorisation of the joint probability distribution of V (i.e. global probability distribution) into local probability distributions of individual variables. The *Markov Property* states that a given variable is dependant only on its immediate parent variables (Korb and Nicholson, 2010).

For discrete variables:

$$P(X_1, X_2, \dots, X_6) = \prod_{i=1}^6 P(X_i | \prod X_i) \quad (\text{Eq. 1})$$

For continuous variables:

$$f(X_1, X_2, \dots, X_6) = \prod_{i=1}^6 f(X_i | \prod X_i) \quad (\text{Eq. 2})$$

The model parameters are estimated typically using *Maximum Likelihood Estimates* approach. For discrete and continuous variables, parameters are estimated in terms of conditional probabilities and conditional regression coefficients, respectively (Scutari, 2009). Further, the significance of each relationship identified was validated using leave-one-out cross validation (LOOCV).

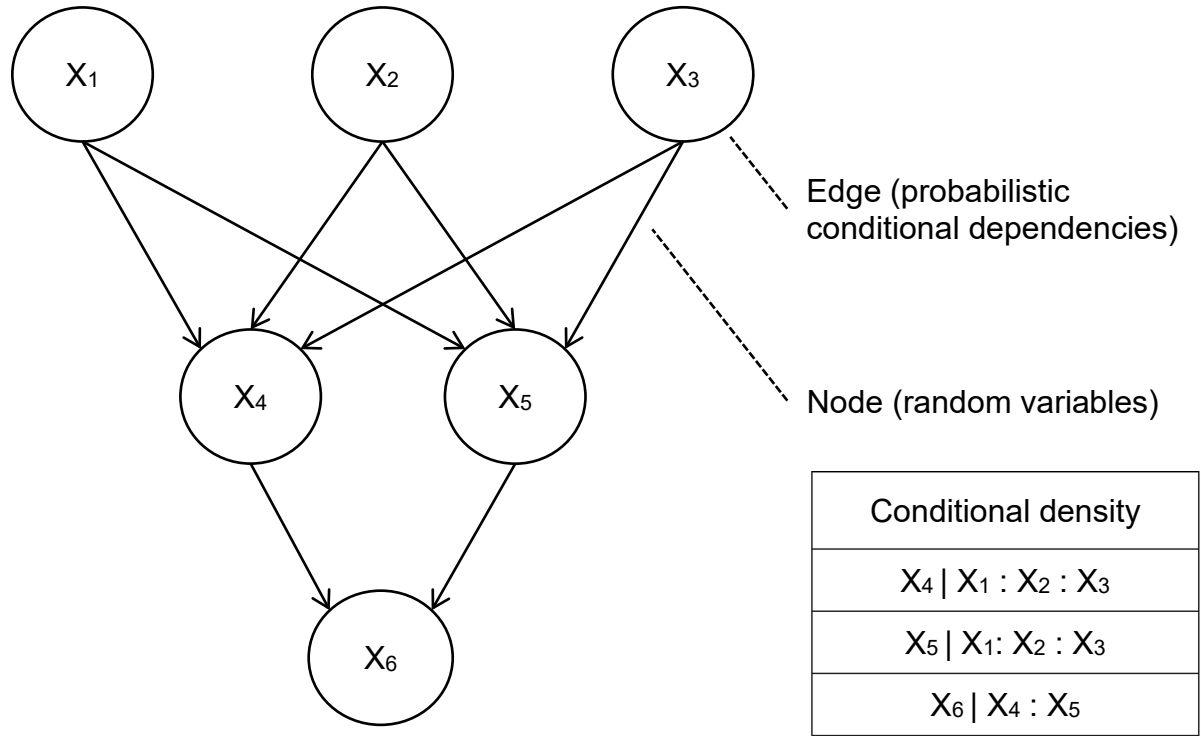


Figure 2. Directed Acyclic Graph (DAG) of a Bayesian Networks (BNs) model. *Note:* Conditional density refers to the probability density functions of a variable given its immediate parent variables.

3. Results and Discussion

3.1 Understanding nutrient wash-off

During the course of a rainfall event, a fraction of pollutant load accumulated during the preceding dry period gets washed-off. In the following sections, the wash-off process is described as ‘decaying of initially available pollutant load’ (i.e. decay) for the convenience of mathematical interpretation. As such, the rate of decay of pollutants depends on the pollutant load available (W) at a given time (T) and intensity of the rainfall event (I). In the case of particulate solids, higher the intensity, more particulates can be detached from impervious surfaces and entrained in runoff. It has been well established that wash-off of particulate solids exhibits exponential decay behaviour as shown in Figure 3a (Egodawatta et al., 2007; Sartor et al., 1974; Tsihrintzis and Hamid, 1998). Similarly, pollutants attached to particulate solids are likely to undergo exponential decay behaviour. However, it is necessary to characterise interdependencies between rate of decay and influential factors, including pollutant affinity to solids, which was accounted for in this study using organic carbon. Organic carbon represents the organic matter present in road deposited solids, which is a key source of chemically reactive functional groups that bind stormwater pollutants together (Sposito, 2008). As such, a Bayesian Networks (BNs) model was proposed (Figure 3b), which describes nutrient decay during a rainfall event.

The proposed BNs model structure replicates non-linear behaviour (likely) of nutrient decay by integrating linear interdependencies (likely) between the rate of decay (R) and available nutrient

load (W), rainfall intensity (I) and total organic carbon (TOC). Accordingly, R could be represented in the form of a conditional probability density function of W , I and TOC.

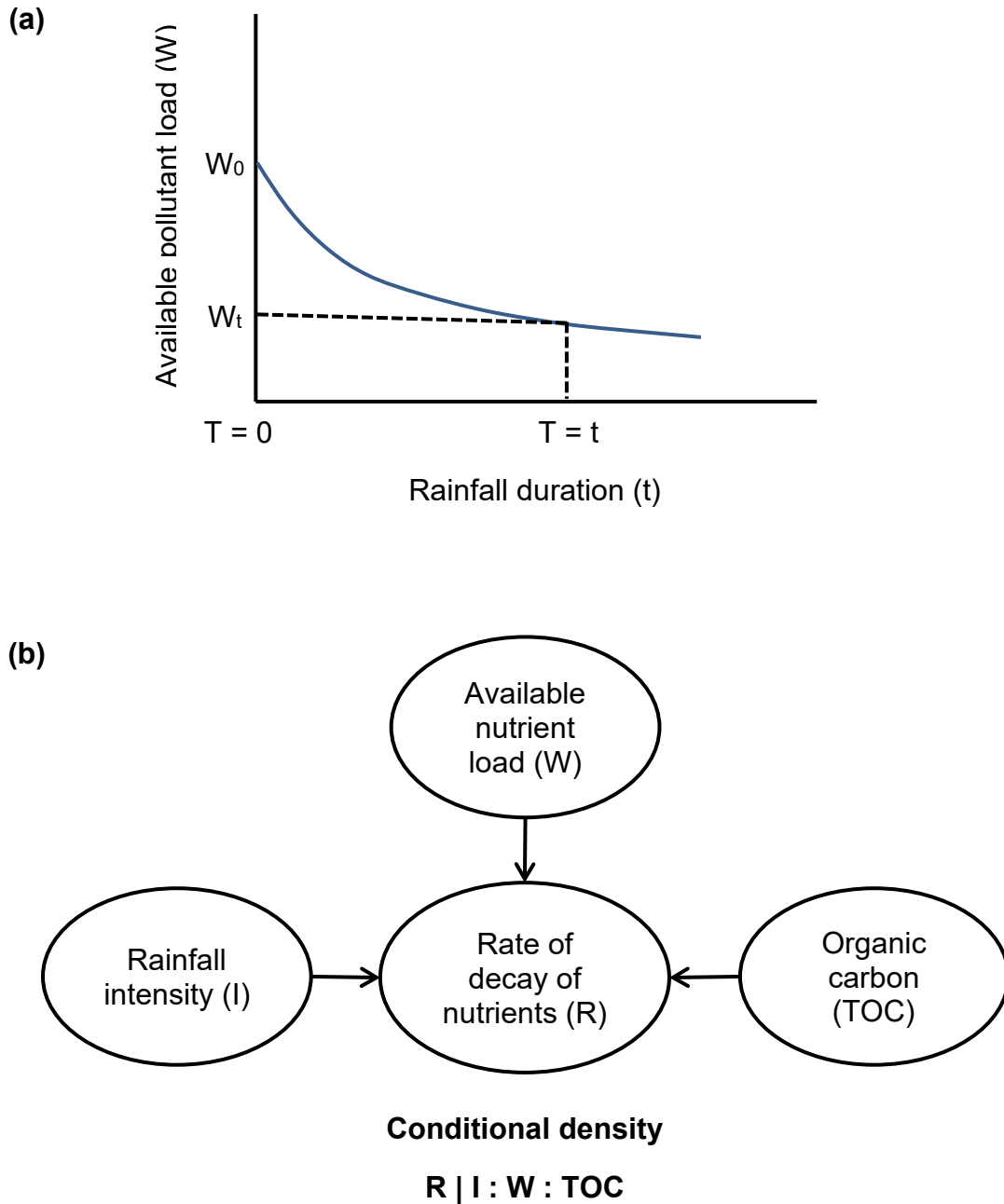


Figure 3. (a) Exponential decay behaviour of pollutant wash-off; (b) Bayesian Networks (BNs) model showing interdependencies between rate of nutrient decay and influential factors.

Table 1 shows the outcomes of the BNs model, where the overall performance is satisfactory for all species of nutrients, except NO_2^- . More importantly, rainfall intensity and available nutrient load exert statistically significant positive influence on all nutrients, while organic carbon only show significant, but inverse relationship with the rate of decay of total PO_4^{3-} and TP. To better

understand the interdependencies between nutrient decay and influential factors, the BN model outcomes are visualised in Figure 4.

Table 1. Summary of the estimates of interdependencies between rate of nutrient decay and influential factors.

Conditional probability density function: <i>Rate of nutrient decay I : W : TOC</i>						
Nutrient Type	Estimated conditional regression coefficient			RSE	Adjusted R ²	F-statistic
	I	W	TOC			
TN	0.676***	0.829***	0.145	0.166	0.740	91.27***
NO ₃ ⁻	0.897***	1.610***	0.177	0.200	0.776	110.4***
NO ₂ ⁻	0.890***	1.727***	-5.477***	0.403	0.574	43.8***
TKN	0.643***	0.782***	0.207	0.174	0.711	78.9***
TP	1.604***	1.811***	-1.861***	0.183	0.936	465.8***
PO ₄ ³⁻	1.134***	0.819***	-1.254**	0.199	0.925	391.7***
<p>Bayesian Networks (BNs) model parameter estimations made with Gaussian distribution and using log-transformed data. Model validated via leave-one-out cross validation (LOOCV) with Residual Standard Errors (RSE) reported and significance reported via t-test for individual coefficients and F-test for overall model: *p<0.05, **p<0.01, ***p<0.001.</p> <p>I – rainfall intensity; W – available pollutant load at a given point time; TOC – total organic carbon; TN – total nitrogen; TKN – total Kjeldahl nitrogen; and TP – total phosphorous</p>						

As shown in Figure 4, 50% increase in rainfall intensity would increase the rate of decay of nitrogen compounds up to 44% (86% at 100% increase in rainfall intensity), and phosphorous compounds up to a staggering 92% (204% at 100% increase in rainfall intensity). This is attributed to the higher kinetic energy associated with intense rainfall, which can rapidly detach and disintegrate nutrient-carrying particulates deposited on impervious surfaces (Liu et al., 2012; Liu et al., 2016). Increased frequency of high-intensity short-duration rainfall is predicted for regions such as Australia, due to changes to rainfall patterns as a result of climate change (IPCC, 2018), which can substantially alter nutrient wash-off and in turn enrichment of receiving waters. Similarly, a 50% increase in initially available nutrient load would increase the rate of decay of nitrogen compounds up to 92% (205% at 100% increase in initial load) and phosphorus compounds up to 108% (250% at 100% increase in initial load). This would result in concerning

effects in the absence of appropriate source control measures for mitigating nutrient discharge associated with increasing urban population and the consequent spread of urbanisation.

Unlike rainfall intensity and initially available load, TOC show conflicting, but interesting associations with the decay of nutrients. TOC does not exert significant influence on the decay of nitrogen compounds. This is hypothesised to be due to the competitive sorption of other pollutants present in runoff. For example, heavy metals, one of the ubiquitous stormwater pollutants, show competitive and preferential adsorption behaviour with functional groups in organic matter, while they tend to form soluble complexes with nitrogen (metal-nitrogen complexation) (Ghasemi et al., 2013; Jayarathne et al., 2019). In the case of phosphorous compounds, a 50% increase in TOC could substantially decrease the decay of phosphorous compounds up to 112% (263% at 100% increase in TOC). This is attributed to enhanced adsorption to particulate solids and the low solubility of phosphorous compounds, reducing the dissolved amount of phosphorous in runoff (Asomaning, 2020; Yang et al., 2019). Further, it also implies that phosphorous wash-off relies on the wash-off of particulate solids.

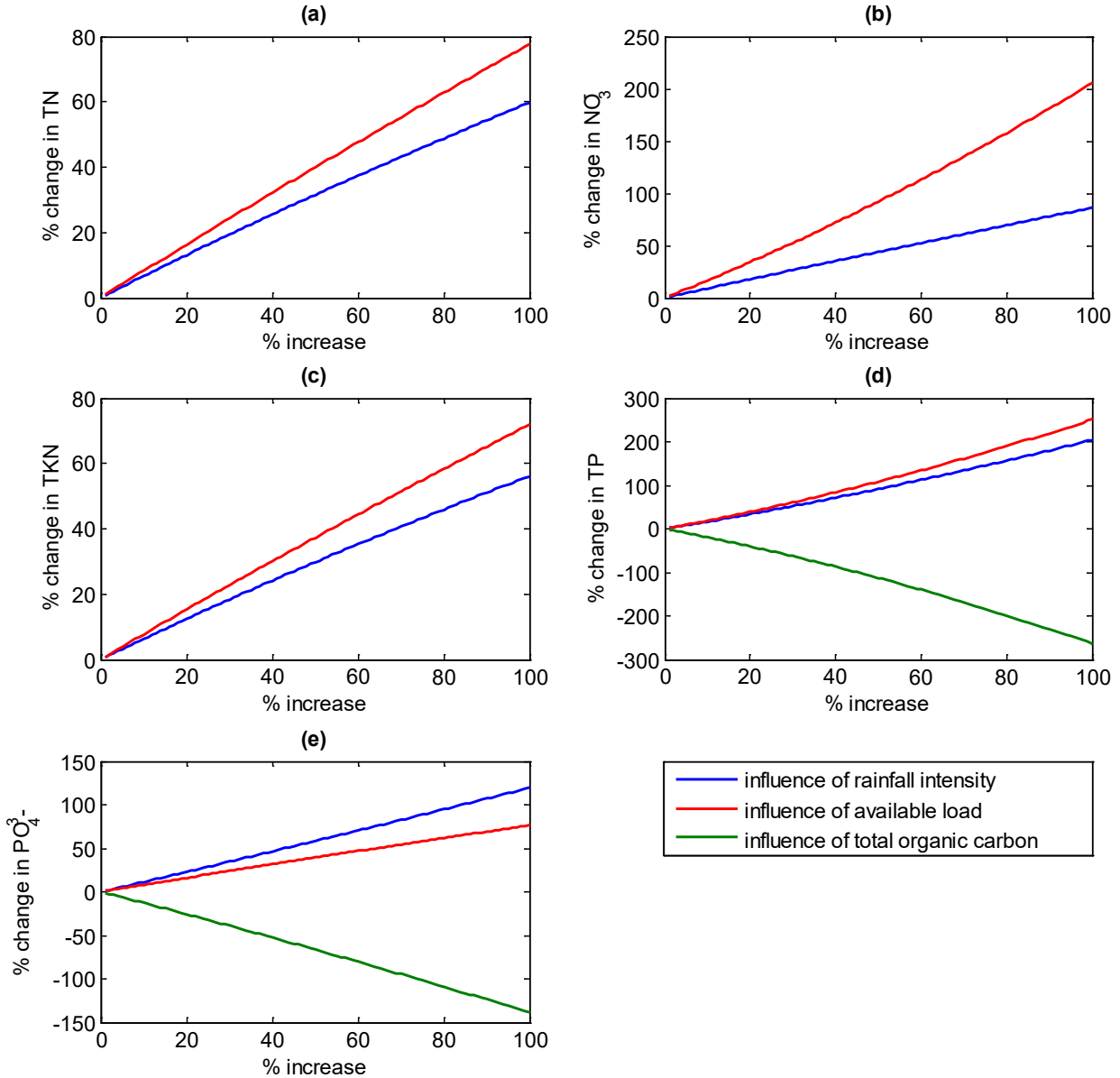


Figure 4. Influence of rainfall intensity, initially available load, and total organic carbon on the rate of nutrient decay (% change in decay is shown in response to % increase in each influential factor): (a) Total Nitrogen (TN); (b) Nitrates (NO_3^-); (c) Total Kjeldahl Nitrogen (TKN); (d) Total Phosphorous (TP); and (e) Phosphates (PO_4^{3-}). *Note: only significant influential factors and nutrient species for which the BNs model performed satisfactorily are included in the figure.*

3.2 Replication of nutrient wash-off

According to BNs analysis, it was found that the rate of decay of nitrogen compounds is directly proportional to the available load at a given point in time and rainfall intensity. Further, the rate of decay of phosphorous compounds is directly proportional to available load and rainfall intensity, and inversely proportional to the organic carbon load. Accordingly, respective exponential decay functions which describe the wash-off of nutrients were derived as given below.

Decay of nitrogen compounds

$$-\frac{W}{t} \propto W ; -\frac{W}{t} \propto I$$

$$\frac{dw}{dt} = -kWI$$

$$\int \frac{dw}{W} = -kI \int dt$$

$$\ln W = -kIt + c$$

At $t = 0$, $W = W_0$ and at $t = t$, $W = W_t$

$$\ln \frac{W_t}{W_0} = -kIt$$

$$W_t = W_0 e^{-kIt}$$

Hence wash-off:

$$W_w = W_0(1 - e^{-kIt}) \quad (\text{Eq. 3})$$

Decay of phosphorous compounds

$$-\frac{W}{t} \propto W ; -\frac{W}{t} \propto I ; -\frac{W}{t} \propto \frac{1}{TOC}$$

$$\frac{dw}{dt} = -k \frac{WI}{TOC}$$

$$\int \frac{dw}{W} = -k \frac{I}{TOC} \int dt$$

$$\ln W = -k \frac{I}{TOC} t + c$$

At $t = 0$, $W = W_0$ and at $t = t$, $W = W_t$

$$\ln \frac{W_t}{W_0} = -k \frac{I}{TOC} t$$

$$W_t = W_0 e^{-k \frac{I}{TOC} t}$$

Hence wash-off:

$$W_w = W_0 \left(1 - e^{-k \frac{I}{TOC_0} t}\right) \quad (\text{Eq. 4})$$

Where, t – time (hr)

I – rainfall intensity (mm/hr)

k – wash-off coefficient (/mm or mg/mm)

W – available nutrient load at any given point in time (mg/m²)

W_0 – available nutrient load at the beginning of rainfall event (mg/m²)

W_t – available nutrient load after time t during rainfall event (mg/m²)

W_w – wash-off nutrient load after time t during rainfall event (mg/m²)

TOC – available total organic carbon load at any given point in time (mg/m²)

TOC_0 – available total organic carbon load at the beginning of rainfall event (mg/m²)

The empirical models derived for nutrient wash-off were fitted with the experimental data and wash-off coefficients were estimated. The coefficient of determination for nitrogen and phosphorous species were 0.42 and 0.34, respectively, and root mean squared error for nitrogen and phosphorous species were 0.40 and 0.54, respectively. Even though the model fit with the experimental data was not satisfactory, it is important to note that these models have been derived based on the validated outcomes of a Bayesian Networks analysis (see Section 3.1). While noting that there is an opportunity to further refine these empirical models, this study quantified uncertainty associated with each model to enhance the realistic interpretation of the outcomes. The uncertainty indicates the variability associated with the model outcomes. Hence, is critical for decision making in the context of designing pollution mitigation measures. Non-linear regression was employed using MATLAB *nlinfit* function together with the proportional error model defined

in the form of $y = f + \theta f \varepsilon$, where the error term is specified as $\varepsilon \sim N(0,1)$, ‘f’ is function value, and ‘ θ ’ is error parameter (MATLAB, 2019b). This enabled accounting for parameter estimation errors and residual errors in uncertainty quantification. As such, 95% uncertainty intervals were determined by simulating a large number of (i.e. 10,000) the predicted values of nutrient wash-off.

Table 2 shows the estimated wash-off coefficients. It is evident that nitrogen species would decay at a similar rate, which is attributed to the solubility of nitrogen compounds. However, TP can decay almost three times as rapidly as phosphates. Further, fraction wash-off predicted from the above models showed that compared to nitrogen, phosphorous is more likely to be washed-off during any rainfall event. These observations on phosphorous compounds indicate that their wash-off is highly dependent on particulate solids and some forms of phosphorous is likely to be attached to solids than other species such as phosphates. While pollutant source control can always reduce the burden on stormwater treatment systems (Müller et al., 2020), the high likelihood of phosphorous wash-off implies that source control of phosphorous is more crucial than nitrogen for stormwater management.

Table 2. Estimated wash-off coefficient (k) for different nutrient species.

Nutrient Type	k (/mm)*	Nutrient Type	k (mg/mm.m ²)*
1TN	0.0020	2TP	0.3128
4NO ₃ ⁻	0.0019	3PO ₄ ³⁻	0.1257
TKN	0.0021		
*the difference in units was because rainfall intensity and initially available pollutant load influenced decay of both nitrogen and phosphorous species, but TOC only influenced decay of phosphorous species.			

3.3 Development of Intensity-Frequency-Duration-Wash-off (IFDW) curves

The design of stormwater management measures is typically based on design rainfall events. Common approaches such as Water Sensitive Urban Design (WSUD) target very frequent to frequent rainfall events (TMR, 2019). For stormwater management measures that are intended to control the volume/flow, the design process requires information on intensity, frequency and duration of rainfall events and their likelihood of occurrence over a particular period of time. In the case of Australia, this information is available in the form of intensity-frequency-duration (IFD) for a range of design rainfall events (435 design rainfall events were used in this study) via the Bureau of Meteorology (BoM, 2016).

The design rainfall data for the Gold Coast region were generated using the IFD information (Figures S5 and S6 in Supplementary Information), which was then used to calculate respective fraction wash-off $\left(\frac{W_w}{W_0}\right)$ for TN and TP. In the case of TP wash-off, the typical TOC content in the road deposited solids in the region was considered to be 27.6 mg/m² (average TOC build-up load obtained from experimental data given in Tables S2 – S5). Figure 5 and Figure S7 in Supplementary Information show the resulting Intensity-Frequency-Duration-Wash-off (IFDW)

curves for very frequent events (Figure 5) and frequent/infrequent events (Figure S7), respectively. These curves can be used to determine the fraction wash-off of a particular type of nutrient for the design rainfall event.

For example, 1 EY (exceedance per year) and 10% AEP (annual exceedance probability) design rainfall events that last for 30 min would have intensities of 57.9 mm/h and 104 mm/h, respectively (see Figures S5 and S6). Here, ‘exceedance per year’ is defined as the number of times a particular event can occur or exceed in a given year, and ‘annual exceedance probability’ is the probability of an event occurring or being exceeded in a given year. According to Figure 5, the 1 EY event, which is a very frequent event, could wash-off 6% of TN and 28% of TP accumulated on impervious surfaces. Similarly, the 10% AEP event, which is a frequent/infrequent event, could remove 10% of TN and 45% of TP from impervious surfaces (see Figure S7). Even though nitrogen compounds are water soluble, higher wash-off fractions of phosphorous compounds can be expected as phosphorous wash-off is dependent more on particulate solids (see Section 3.2).

Given typical nutrient build-up load in a catchment of interest, the information generated from IFDW curves will inform input nutrient load that needs to be managed. This approach can avoid the need for field-based data collection for stormwater modelling. Consequently, this would reduce the cost and time typically spent on designing stormwater treatment systems.

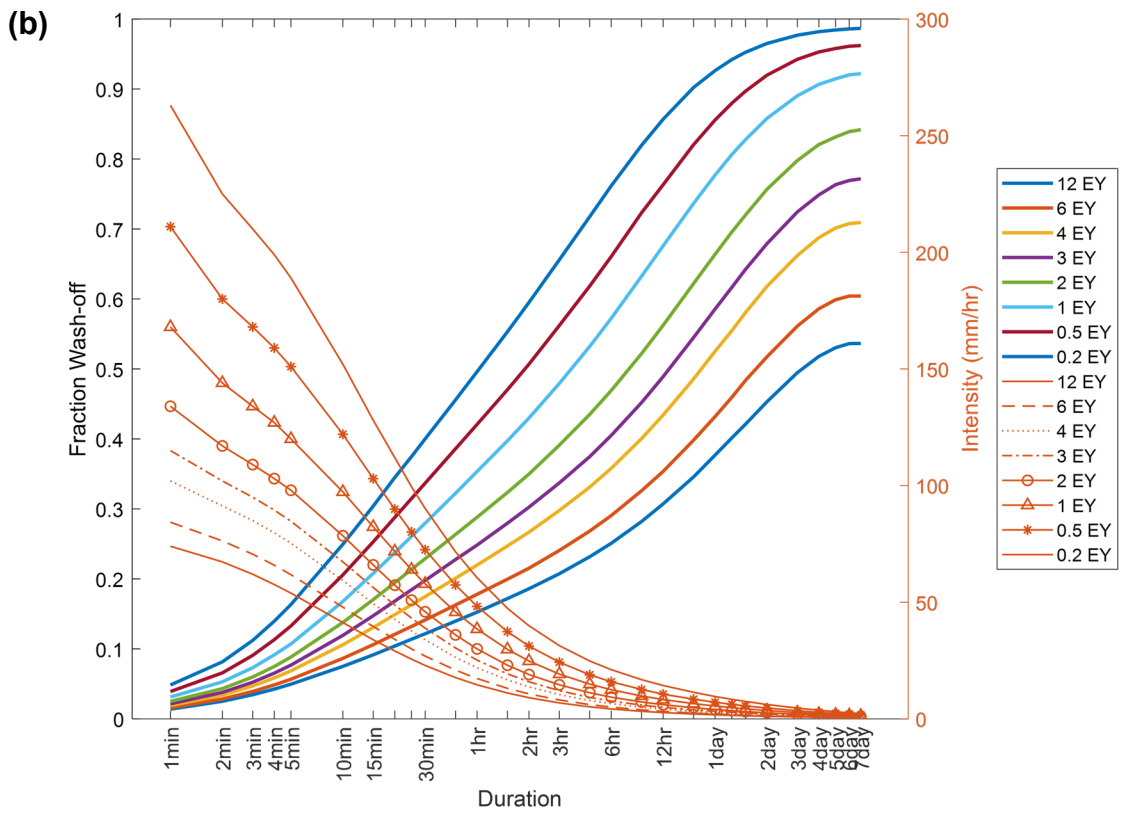
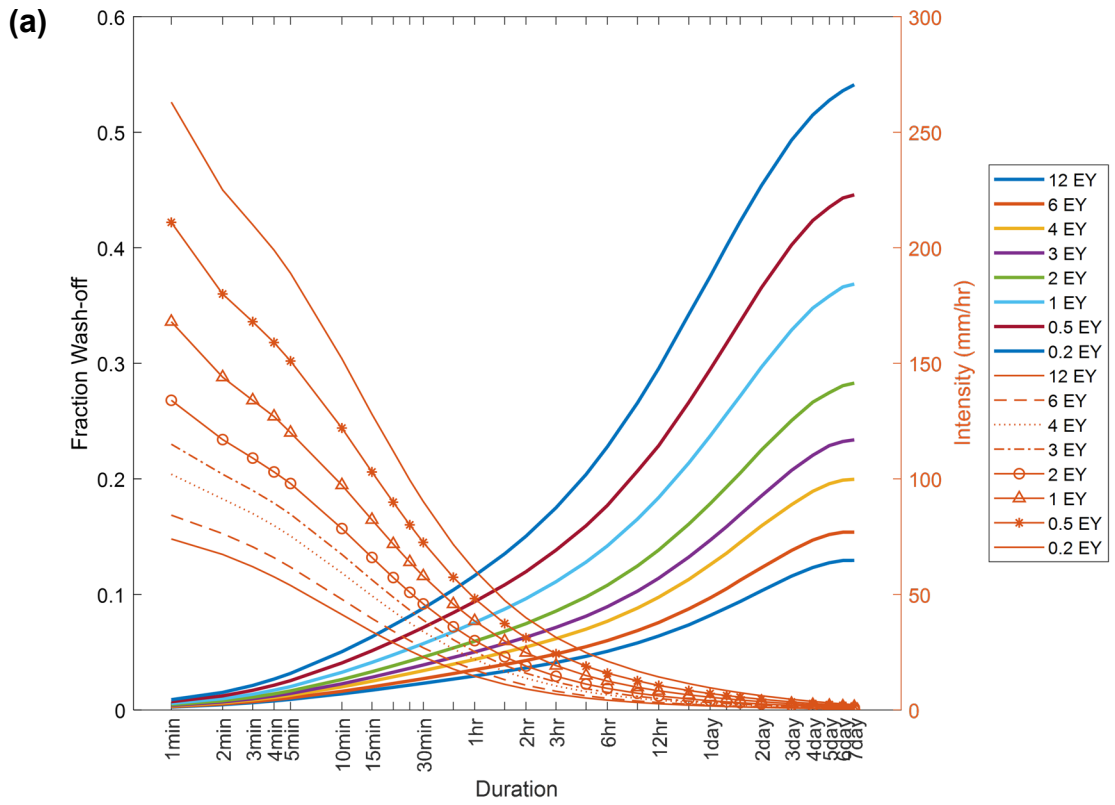


Figure 5. Intensity-Frequency-Duration-Washoff (IFDW) distributions for very frequent design rainfall events for Gold Coast region (28.0167° S, 153.4000° E): (a) Total Nitrogen (TN); (b) Total Phosphorous (TP). *Note 1: horizontal axis is in log scale; Note 2: EY – number of exceedances of a particular rainfall event per year.*

Moreover, it is important that the uncertainty associated with stormwater quality estimations is accounted for in the design of treatment measures. This uncertainty can arise from a range of sources such as data, model structure and model parameters, and inherent variability in the processes. As stated in Section 3.2, the uncertainty arising from the modelling process has been quantified in terms of parameter estimation errors and residual errors. The natural variability in nutrient processes is deemed accounted for given that the interdependencies between nutrient decay and influential factors are accurately characterised (see Section 3.1). Accordingly, the overall uncertainty quantified indicates possible range of variation of a given estimation of the decay of nutrients.

The uncertainty limits associated with estimations of fraction wash-off of TN and TP are shown in Figures 6 and 7 (very frequent design rainfall) and Figures S8 and S9 in the Supplementary Information (frequent/infrequent design rainfall). Accordingly, the point estimates of 6% TN and 28% TP wash-off for the 1 EY event could be as high as 10% and 66%, respectively (see Figures 6 and 7). Similarly, the 10% AEP event could result in 18% TN wash-off (exceeding the point estimate of 10% TN) and 103% TP wash-off (exceeding the point estimate of 45% TP) (see Figures S8 and S9).

Even though the above 103% TP wash-off is not observable in the field, it indicates that the 10% AEP event could potentially completely wash-off TP accumulated on the road surface. Interestingly, this behaviour of TP can be expected during the majority of both, very frequent and frequent/infrequent design rainfall events (see Figures 7 and S9). However, complete wash-off of TN would only occur during very few events that are rarer than 10% AEP (see Figures 6 and S8). These findings highlight the critical need for accounting for the variability in nutrient wash-off when designing a stormwater treatment system with adequate capacity to reduce nutrient loads in runoff.

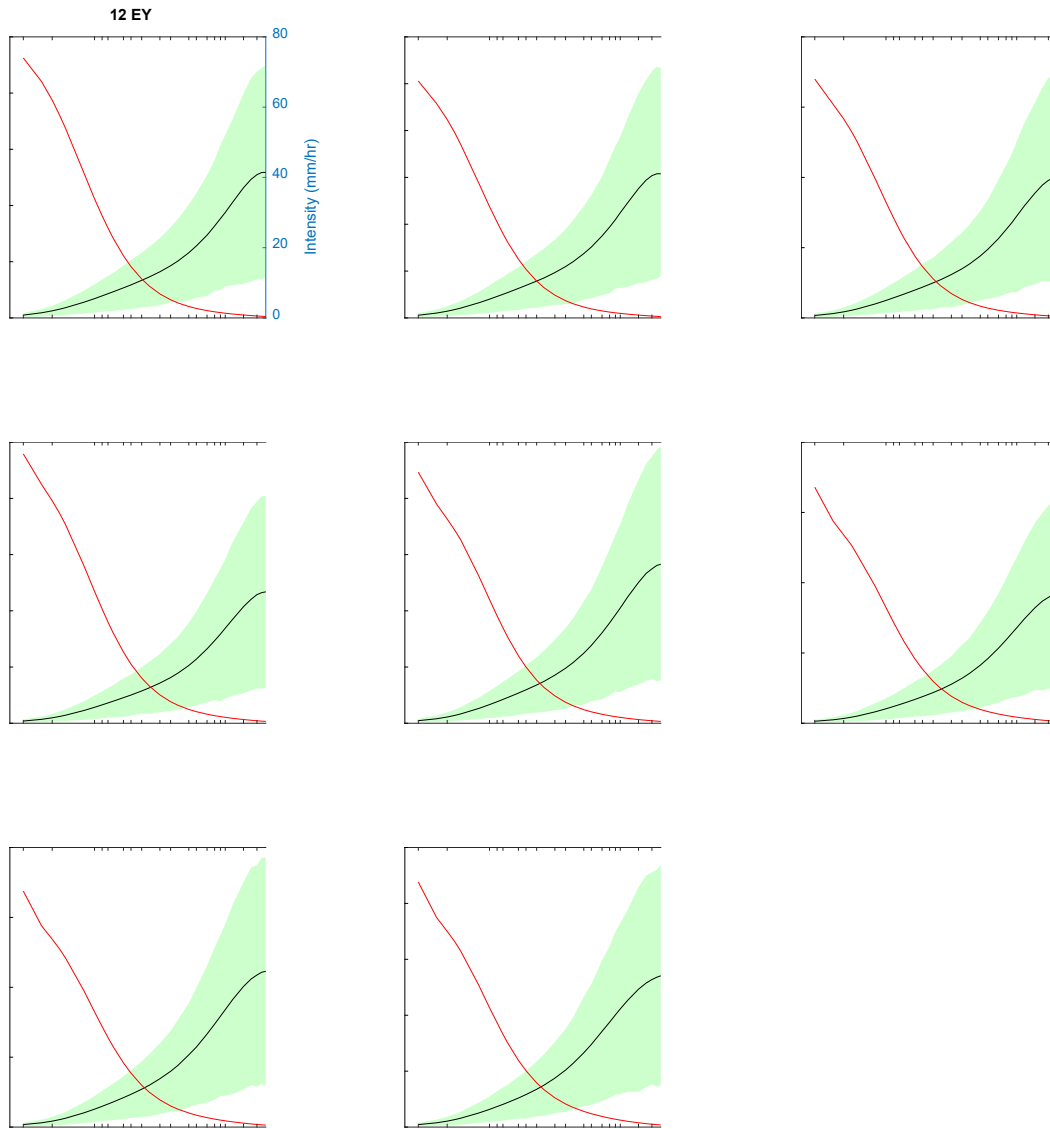


Figure 6. Intensity-Frequency-Duration-Washoff (IFDW) distributions of Total Nitrogen (TN) and associated uncertainty for very frequent design rainfall events for the Gold Coast region (28.0167° S, 153.4000° E). *Note 1: horizontal axis is in log scale; Note 2: EY – number of exceedances of a particular rainfall event per year.*

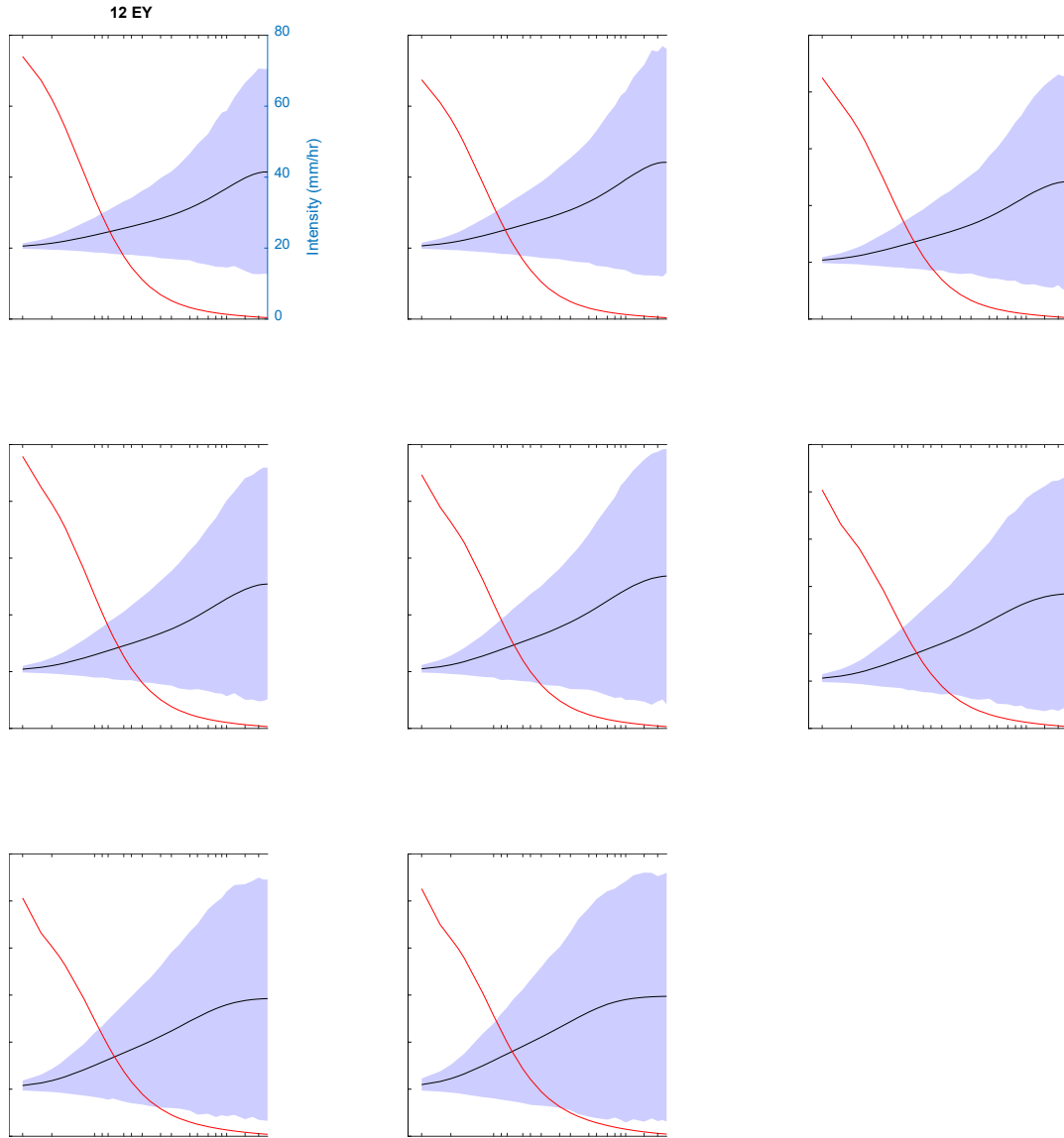


Figure 7. Intensity-Frequency-Duration-Washoff (IFDW) distributions of Total Phosphorous (TP) and associated uncertainty for very frequent design rainfall events for Gold Coast region (28.0167° S, 153.4000° E). *Note 1: horizontal axis is in log scale; Note 2: EY – number of exceedances of a particular rainfall event per year.*

4. Conclusions

This research study combined empirically based and statistical modelling techniques to develop two new mathematical formulations which can describe the decay (wash-off) of nutrients during storm events. It was found that rainfall intensity, rainfall duration and initially accumulated load exert positive influence on the decay of both, nitrogen and phosphorous species, while organic carbon only exerts influence (negative) on phosphorus decay. The results show that nitrogen

species would decay at a similar rate, while phosphorus species may decay at different rates. This knowledge was used to develop Intensity-Frequency-Duration-Washoff (IFDW) curves to understand how nutrients are washed-off during rainfall events with different probability of occurrence (i.e. very frequent events and frequent/infrequent events). The study outcomes provide a robust tool to support decision making by providing reliable knowledge on stormwater quality and can make a significant contribution to the design of effective stormwater treatment measures in order to promote sustainable stormwater management and reuse. Additionally, it is also noteworthy that the IFDW developed in this study took the entire rainfall into consideration while only a portion of the rainfall is responsible for pollutants wash-off in the real environment. This means the worst case scenarios (the highest pollutant loads washed-off) were estimated. This would ensure the effectiveness of designed stormwater treatment systems.

Supplementary Information

Supplementary Information provides details of sample collection, rainfall events simulated, original build-up and wash-off data, IFD data of very frequent and frequent/infrequent design rainfall events for Gold Coast region, and IFDW curves and associated uncertainty of TN and TP for frequent and infrequent design rainfall events in Gold Coast region.

Acknowledgement

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Supplementary Information

Understanding nutrient dynamics for effective stormwater treatment design

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1. Build-up sampling

Several sampling methods have been used in the past, such as brushing and sweeping, dry vacuuming, and wet vacuuming (Deletic and Orr 2005, Sartor and Boyd 1972, Vaze and Chiew 2002). However, a dry and wet vacuum system has been proven to effectively collect fine particles with efficiency > 90%, and generate minimum abrasion material (Gunawardana, 2011, Mahbub, 2011, Mummullage, 2015).

1.1 Wet and dry vacuum system

The sample collection system (Figure S1) included a 1500 Watt Delonghi Aqualand portable vacuum cleaner integrated with a highly efficient water filtration unit, a 12 Volt Swift Compact

water sprayer, and a generator. The water sprayer has a control unit to adjust the pressure to the desirable level. The system was verified for its performance under controlled conditions similar to road sites in the field, and the sample collection efficiency was found to be 95%. The details of the procedure followed can be found in Miguntanna (2009).



Fig

Vacuum cleaner

Water sprayer

1.2 Field sampling process



Figure S2. Build-up sampling process

2. Wash-off sampling

There are significant constraints to collect samples under natural storm events due to uncertainty in rainfall occurrence and lack of control over rainfall characteristics such as intensity and duration. Therefore, a mechanical rainfall simulator designed by Hergren (2005) was used in this study.

2.1 Rainfall Simulator

Hergren (2005) designed the rainfall simulator (Figure S3a) based on the characteristics of natural storm events occurring in Southeast Queensland, Australia. The simulator is capable of creating near-natural rainfall characteristics such as intensity, drop velocity and size, and drop-size

distribution. The intensity and duration can be varied by oscillating the nozzle boom controlling its cycle time.

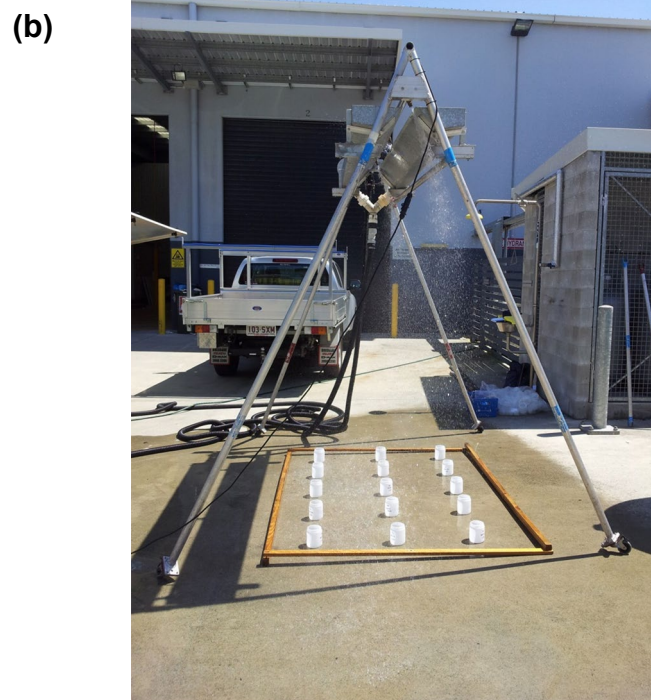
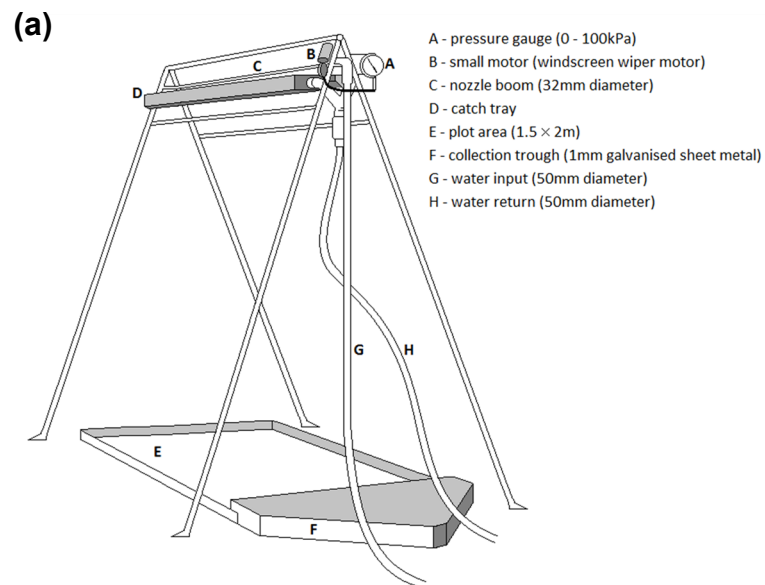


Figure S3. (a) Design of rainfall simulator; (b) Performance verification of rainfall simulator.

2.2 Performance verification

The performance of the simulator was verified under laboratory conditions. Accordingly, 15 polyethylene containers were placed under the rainfall simulator as shown in Figure S3b. Then a

series of rainfall events were simulated over a duration of 5 min. The volume of water in each container was measured, and the intensity of each rainfall event was calculated given the container surface area. The Uniformity Coefficient (C_u) defined by Equation S1 was used to determine the spatial variability of rainfall intensity. The C_u was found to be around 70%, which was similar to the values reported elsewhere (Herngren 2005, Egodawatta 2007).

$$C_u = \left(1 - \frac{\sum |x|}{m \times n}\right) \times 100 \quad \text{Equation S1}$$

Where: m – mean intensity
 n – number of observations
 x – deviation of individual observation from mean

The rainfall events were simulated for different control settings. For field simulation, it was necessary to select control settings that can create typical regional rainfall intensities with low spatial variability. After analysing historical records of rainfall, it was decided to select control settings that corresponded to intensities within the range of 20 – 133mm/hr. More details on performance verification of the rainfall simulator can be found in Miguntanna (2009).

2.3 Field Sampling process



Figure S4. Wash-off sampling process.

Table S1. Rainfall events simulated in this study.

Rainfall intensity (mm/h)	Rainfall duration (min)			
	Event 1	Event 2	Event 3	Event 4
20	10	20	30	40
40	10	15	25	35
65	10	15	20	30
86	10	15	20	25
115	5	10	15	20
135	5	10	15	20

Table S2. Original build-up data used in the study.

Land use	Sample volume (L)	Antecedent dry days	TOC	NO ₂ ⁻	NO ₃ ⁻	TKN	TN	PO ₄ ³⁻	TP
			(mg/L)						
Residential (Armstrong Way)	8.42	8	28.19	0.101	1.841	5.268	7.210	0.795	1.670
Industrial (Stevens Street)	7.04	9	19.22	0.018	0.639	3.576	4.233	5.756	7.005
Commercial (Lawrence Drive)	9.76	11	23.30	0.063	0.757	12.43	13.245	3.779	6.855

Note: samples were collected using a 3 m² plot area.

Table S3. Wash-off data for residential site (Armstrong Way).

Intensity (mm/hr)	time (min)	TOC (mg)	NO2- (mg)	NO3- (mg)	TKN (mg)	TN (mg)	PO43- (mg)	TP (mg)
20	5	0.867	0.000	0.008	0.179	0.187	0.007	0.008
20	10	1.557	0.000	0.014	0.338	0.352	0.011	0.015
20	15	2.176	0.001	0.019	0.467	0.487	0.014	0.022
20	20	2.745	0.001	0.024	0.576	0.601	0.016	0.029
20	25	3.275	0.001	0.028	0.671	0.700	0.018	0.034
20	30	3.774	0.001	0.032	0.756	0.789	0.020	0.040
20	35	4.248	0.001	0.036	0.833	0.870	0.021	0.045
20	40	4.699	0.001	0.039	0.902	0.943	0.023	0.049
40	5	1.874	0.001	0.019	0.349	0.368	0.015	0.057
40	10	3.545	0.001	0.036	0.653	0.690	0.025	0.097
40	15	5.115	0.002	0.053	0.910	0.965	0.034	0.133
40	20	6.615	0.002	0.068	1.142	1.212	0.040	0.162
40	25	8.018	0.002	0.081	1.356	1.440	0.045	0.186
40	30	9.366	0.003	0.093	1.558	1.654	0.050	0.208
40	35	10.667	0.003	0.103	1.749	1.855	0.054	0.229
65	5	2.381	0.001	0.047	0.332	0.380	0.020	0.110
65	10	4.461	0.002	0.089	0.587	0.677	0.033	0.219
65	15	6.381	0.002	0.128	0.800	0.930	0.043	0.326
65	20	8.217	0.002	0.166	0.989	1.158	0.052	0.431
65	25	9.982	0.003	0.204	1.161	1.367	0.058	0.532
65	30	11.689	0.003	0.240	1.320	1.563	0.065	0.629
86	5	5.126	0.002	0.021	0.619	0.642	0.028	0.185
86	10	9.750	0.003	0.040	1.229	1.272	0.050	0.348
86	15	14.029	0.005	0.060	1.782	1.847	0.069	0.503
86	20	18.048	0.006	0.076	2.278	2.360	0.085	0.652
115	5	7.041	0.002	0.015	0.818	0.835	0.027	0.262
115	10	12.181	0.003	0.027	1.443	1.473	0.053	0.521
115	15	16.382	0.005	0.037	1.926	1.967	0.071	0.700
135	5	4.236	0.002	0.025	0.746	0.773	0.014	0.313
135	10	7.595	0.004	0.043	1.280	1.327	0.025	0.626
135	15	10.564	0.006	0.057	1.706	1.769	0.034	0.892
135	20	13.303	0.007	0.070	2.076	2.153	0.048	1.116

Table S4. Wash-off data for industrial site (Stevens Street).

Intensity (mm/hr)	time (min)	TOC (mg)	N2 (mg)	N3 (mg)	TKN (mg)	TN (mg)	TP4 (mg)	TP (mg)
20	5	0.588	0.000	0.014	0.131	0.145	0.069	0.243
20	10	1.129	0.000	0.025	0.231	0.256	0.127	0.474
20	15	1.638	0.001	0.034	0.316	0.351	0.178	0.699
20	20	2.095	0.001	0.042	0.390	0.433	0.227	0.885
20	25	2.514	0.001	0.049	0.457	0.506	0.272	1.048
20	30	2.905	0.001	0.055	0.517	0.572	0.315	1.194
20	35	3.275	0.001	0.060	0.573	0.634	0.356	1.329
40	5	2.587	0.001	0.027	0.259	0.287	0.154	0.599
40	10	4.498	0.002	0.049	0.468	0.519	0.317	1.118
40	20	5.675	0.002	0.062	0.612	0.677	0.433	1.483
40	25	6.765	0.002	0.075	0.747	0.825	0.551	1.860
40	30	7.793	0.002	0.088	0.874	0.964	0.670	2.211
65	5	4.680	0.000	0.044	0.325	0.370	0.263	1.253
65	10	7.616	0.001	0.087	0.552	0.640	0.494	2.403
65	15	9.891	0.001	0.128	0.744	0.873	0.696	3.402
65	20	11.793	0.001	0.166	0.917	1.085	0.877	4.281
65	25	13.448	0.002	0.203	1.088	1.293	1.027	5.011
65	30	14.929	0.002	0.238	1.247	1.487	1.153	5.640
86	5	4.369	0.001	0.082	0.509	0.592	0.354	1.914
86	10	7.497	0.002	0.153	0.918	1.073	0.676	3.405
86	15	10.058	0.003	0.218	1.286	1.507	0.976	4.729
86	20	12.528	0.003	0.278	1.690	1.971	1.247	5.951
86	25	14.870	0.003	0.334	2.075	2.413	1.500	6.980
115	5	5.641	0.003	0.113	0.453	0.569	0.594	2.721
115	10	9.327	0.004	0.193	0.821	1.018	1.074	4.819
115	15	12.224	0.006	0.256	1.137	1.399	1.499	6.615
115	20	14.590	0.006	0.309	1.415	1.730	1.865	8.095
135	5	5.272	0.004	0.090	0.482	0.576	0.913	3.331
135	10	8.911	0.007	0.153	0.833	0.993	1.545	5.767
135	15	12.081	0.008	0.201	1.127	1.337	2.064	7.861
135	20	14.816	0.010	0.241	1.376	1.627	2.500	9.780

Table S5. Wash-off data for commercial site (Lawrence Drive).

Intensity (mm/hr)	time (min)	TOC (mg)	N2 (mg)	N3 (mg)	TKN (mg)	TN (mg)	TP4 (mg)	TP (mg)
20	5	3.392	0.007	0.045	0.777	0.829	0.064	0.223
20	10	5.830	0.011	0.079	1.333	1.423	0.126	0.445
20	15	7.853	0.015	0.108	1.789	1.912	0.185	0.668
20	20	9.624	0.018	0.134	2.193	2.345	0.243	0.891
20	25	11.223	0.020	0.158	2.550	2.729	0.297	1.103
20	30	12.705	0.023	0.181	2.876	3.079	0.348	1.307
20	35	14.100	0.025	0.202	3.179	3.405	0.397	1.500
20	40	15.402	0.027	0.221	3.459	3.707	0.439	1.673
40	5	5.702	0.008	0.090	1.250	1.348	0.090	0.446
40	10	10.246	0.013	0.155	2.199	2.367	0.179	0.832
40	15	14.137	0.017	0.210	2.999	3.226	0.259	1.198
40	20	17.544	0.021	0.258	3.694	3.972	0.335	1.536
40	25	20.608	0.024	0.301	4.307	4.632	0.406	1.842
40	30	23.409	0.027	0.341	4.859	5.227	0.474	2.126
40	35	26.008	0.030	0.378	5.361	5.768	0.540	2.394
65	5	4.404	0.007	0.106	0.956	1.070	0.153	0.739
65	10	7.937	0.013	0.193	1.668	1.874	0.299	1.446
65	15	11.004	0.018	0.273	2.242	2.533	0.439	2.178
65	20	13.721	0.023	0.349	2.743	3.115	0.573	2.908
65	25	16.202	0.027	0.422	3.195	3.644	0.698	3.592
65	30	18.499	0.031	0.489	3.595	4.115	0.803	4.173
86	5	6.725	0.008	0.159	1.273	1.440	0.210	1.143
86	10	11.719	0.014	0.309	2.154	2.476	0.408	2.179
86	15	16.131	0.019	0.455	2.832	3.307	0.586	3.176
86	20	20.022	0.024	0.580	3.403	4.008	0.748	4.148
86	25	23.598	0.029	0.691	3.901	4.621	0.878	4.992
115	5	11.558	0.011	0.081	0.914	1.006	0.405	1.670
115	10	19.983	0.020	0.155	1.817	1.992	0.801	3.202
115	15	26.956	0.029	0.226	2.619	2.874	1.185	4.564
115	5	32.904	0.036	0.295	3.314	3.645	1.473	5.775
135	10	4.948	0.007	0.096	0.697	0.801	0.885	3.508
135	15	8.870	0.013	0.185	1.232	1.430	1.538	6.802
135	20	12.326	0.019	0.270	1.647	1.935	2.085	9.574
135	25	15.442	0.023	0.352	1.999	2.374	2.568	12.002

Duration	Exceedance per Year (EY)							
	12EY	6EY	4EY	3EY	2EY	1EY	0.5EY#	0.2EY*
1 min	74.0	84.3	102	115	134	168	211	263
2 min	67.3	76.3	91.3	102	117	144	180	225
3 min	62.0	70.6	85.0	95.0	109	134	168	210
4 min	57.6	65.9	79.8	89.5	103	127	159	199
5 min	53.9	61.8	75.3	84.7	98.0	120	151	189
10 min	41.3	47.9	59.3	67.3	78.5	97.3	122	152
15 min	33.9	39.6	49.4	56.3	66.0	82.3	103	128
20 min	29.1	34.1	42.7	48.8	57.3	71.8	89.9	112
25 min	25.6	30.0	37.7	43.2	50.9	64.0	80.1	99.5
30 min	22.9	26.9	33.9	38.9	45.9	57.9	72.5	90.2
45 min	17.7	20.9	26.4	30.4	36.0	45.8	57.4	71.6
1 hour	14.6	17.3	21.9	25.2	30.0	38.5	48.2	60.5
1.5 hour	11.1	13.1	16.7	19.3	23.0	29.8	37.4	47.4
2 hour	9.08	10.7	13.7	15.9	19.0	24.8	31.2	39.9
3 hour	6.84	8.10	10.4	12.1	14.6	19.2	24.3	31.4
4.5 hour	5.17	6.13	7.89	9.21	11.2	14.9	18.9	24.8
6 hour	4.25	5.05	6.52	7.64	9.32	12.5	15.9	21.1
9 hour	3.25	3.87	5.02	5.90	7.24	9.82	12.6	16.8
12 hour	2.70	3.21	4.19	4.94	6.08	8.29	10.6	14.3
18 hour	2.08	2.49	3.26	3.86	4.77	6.54	8.42	11.4
24 hour	1.74	2.08	2.74	3.24	4.01	5.52	7.12	9.59
30 hour	1.51	1.81	2.38	2.82	3.50	4.83	6.23	8.38
36 hour	1.34	1.62	2.13	2.52	3.12	4.31	5.57	7.47
48 hour	1.11	1.34	1.77	2.09	2.60	3.59	4.64	6.17
72 hour	0.837	1.01	1.33	1.58	1.96	2.71	3.50	4.62
96 hour	0.671	0.811	1.07	1.27	1.58	2.18	2.81	3.69
120 hour	0.556	0.672	0.889	1.06	1.31	1.81	2.33	3.06
144 hour	0.471	0.568	0.754	0.899	1.12	1.55	1.99	2.61
168 hour	0.404	0.487	0.649	0.776	0.969	1.34	1.72	2.27

Note:

The 0.5 EY design rainfall corresponds to the 2 year Average Recurrence Interval (ARI) IFD **not** the 50% AEP IFD.

* The 0.2 EY design rainfall corresponds to the 5 year Average Recurrence Interval (ARI) IFD **not** the 20% AEP IFD.

Figure S5. Very frequent design rainfall intensity (mm/hr) for Gold Coast (28.0167° S, 153.4000° E) (BoM 2016).

Duration	Annual Exceedance Probability (AEP)						
	63.2%	50%#	20%*	10%	5%	2%	1%
1 min	168	190	257	303	348	407	452
2 min	144	162	221	262	302	364	415
3 min	134	151	206	244	282	338	383
4 min	127	143	195	231	266	316	356
5 min	120	136	185	219	252	297	333
10 min	97.3	110	149	175	200	233	257
15 min	82.3	92.8	126	147	169	195	216
20 min	71.8	81.0	109	128	147	171	188
25 min	64.0	72.1	97.5	115	131	153	169
30 min	57.9	65.3	88.4	104	119	139	155
45 min	45.8	51.7	70.2	83.0	95.5	113	126
1 hour	38.5	43.4	59.3	70.3	81.4	96.8	109
1.5 hour	29.8	33.7	46.5	55.6	64.8	78.1	88.7
2 hour	24.8	28.2	39.1	47.1	55.3	67.1	76.7
3 hour	19.2	21.9	30.8	37.4	44.3	54.2	62.4
4.5 hour	14.9	17.0	24.3	29.8	35.6	43.9	50.7
6 hour	12.5	14.3	20.6	25.4	30.5	37.7	43.7
9 hour	9.82	11.3	16.5	20.4	24.5	30.3	35.0
12 hour	8.29	9.58	14.0	17.4	20.9	25.8	29.8
18 hour	6.54	7.59	11.1	13.8	16.5	20.3	23.3
24 hour	5.52	6.42	9.41	11.6	13.9	16.9	19.4
30 hour	4.83	5.62	8.21	10.1	12.0	14.6	16.6
36 hour	4.31	5.02	7.32	8.95	10.6	12.9	14.6
48 hour	3.59	4.18	6.05	7.35	8.64	10.4	11.8
72 hour	2.71	3.15	4.52	5.45	6.34	7.63	8.62
96 hour	2.18	2.53	3.61	4.33	5.03	6.05	6.84
120 hour	1.81	2.10	3.00	3.60	4.18	5.03	5.70
144 hour	1.55	1.79	2.56	3.08	3.59	4.34	4.92
168 hour	1.34	1.55	2.23	2.69	3.16	3.83	4.35

Note:

The 50% AEP IFD **does not** correspond to the 2 year Average Recurrence Interval (ARI) IFD. Rather it corresponds to the 1.44 ARI.

* The 20% AEP IFD **does not** correspond to the 5 year Average Recurrence Interval (ARI) IFD. Rather it corresponds to the 4.48 ARI.

Figure S6. Frequent and infrequent design rainfall intensity (mm/hr) for Gold Coast (28.0167° S, 153.4000° E) (BoM 2016).

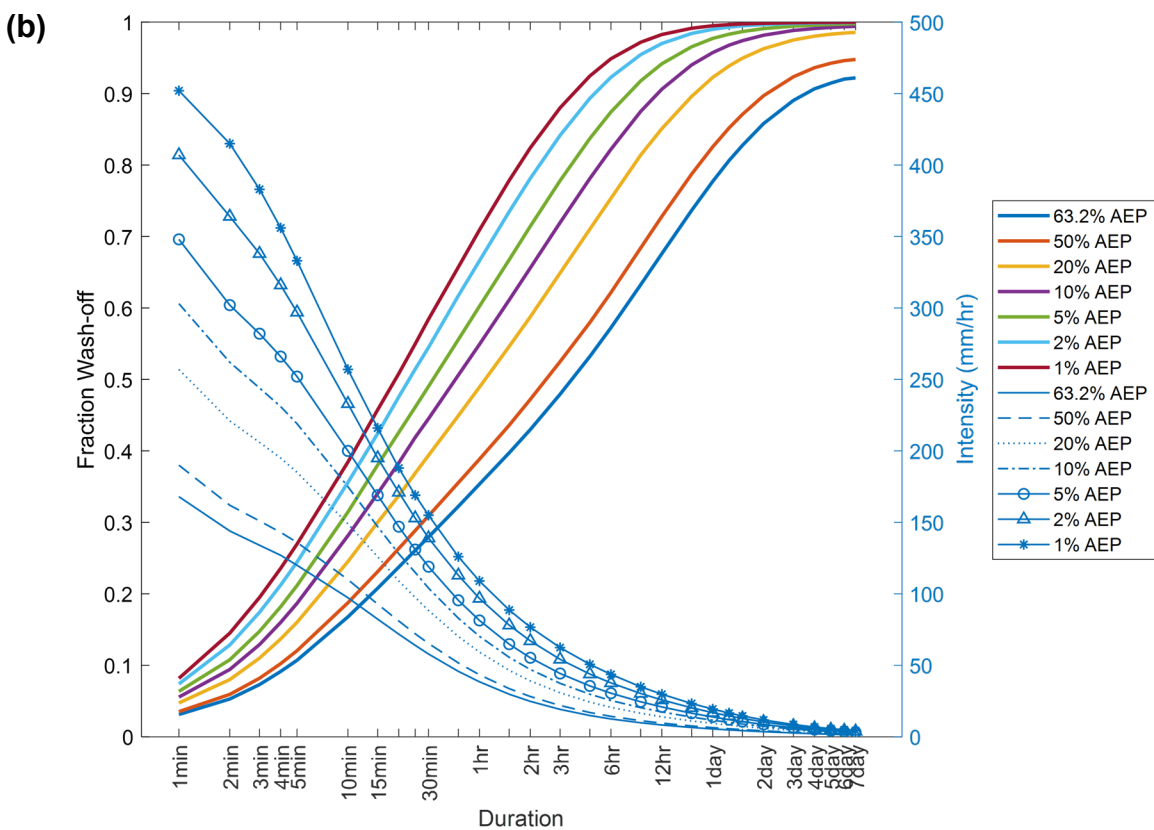
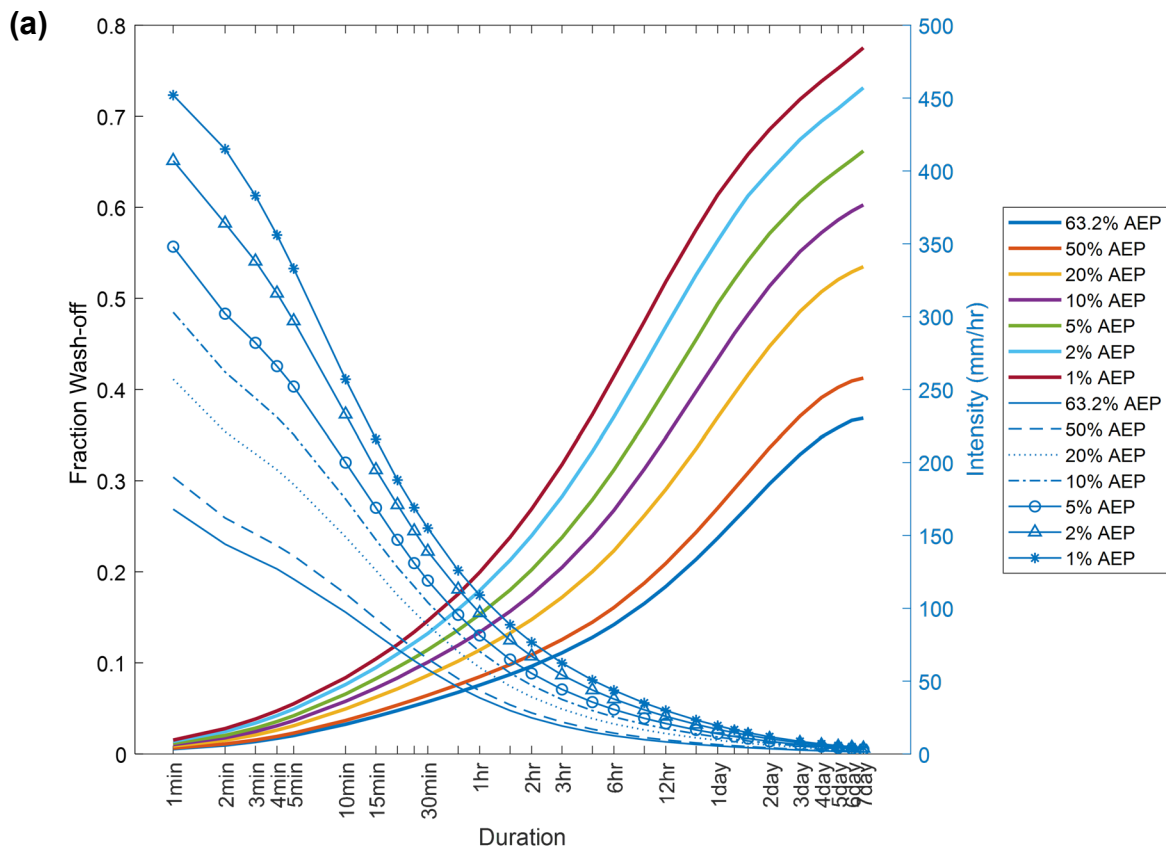


Figure S7. Intensity-Frequency-Duration-Washoff (IFDW) distributions for frequent and infrequent design rainfall events for Gold Coast region (28.0167° S, 153.4000° E): (a) Total Nitrogen (TN); (b) Total Phosphorous (TP). *Note 1: horizontal axis is in log scale; Note 2: AEP – annual exceedance probability of a particular rainfall event.*

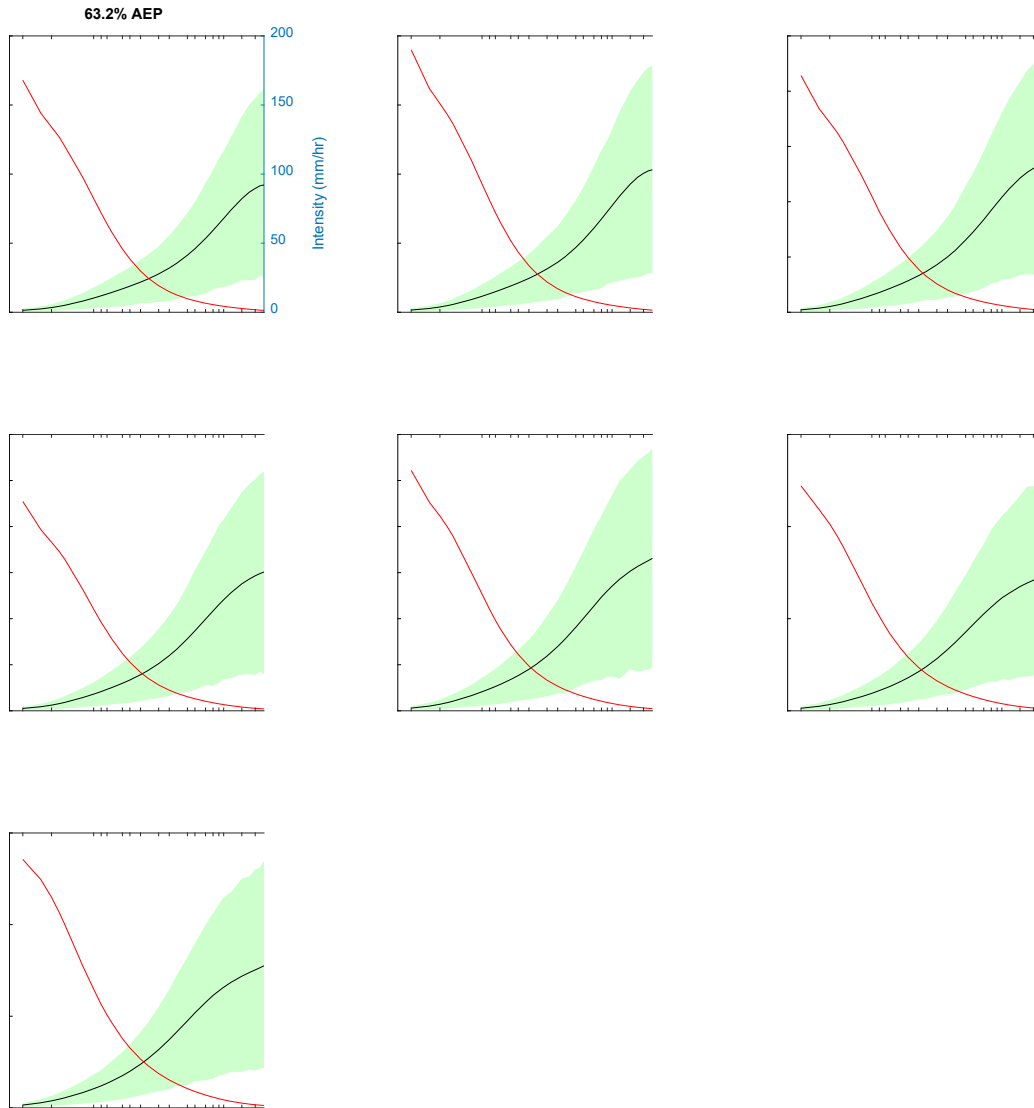


Figure S8. Intensity-Frequency-Duration-Washoff (IFDW) distributions of Total Nitrogen (TN) and associated uncertainty for frequent and infrequent design rainfall events for Gold Coast region (28.0167° S, 153.4000° E). *Note 1: horizontal axis is in log scale; Note 2: AEP – annual exceedance probability of a particular rainfall event.*

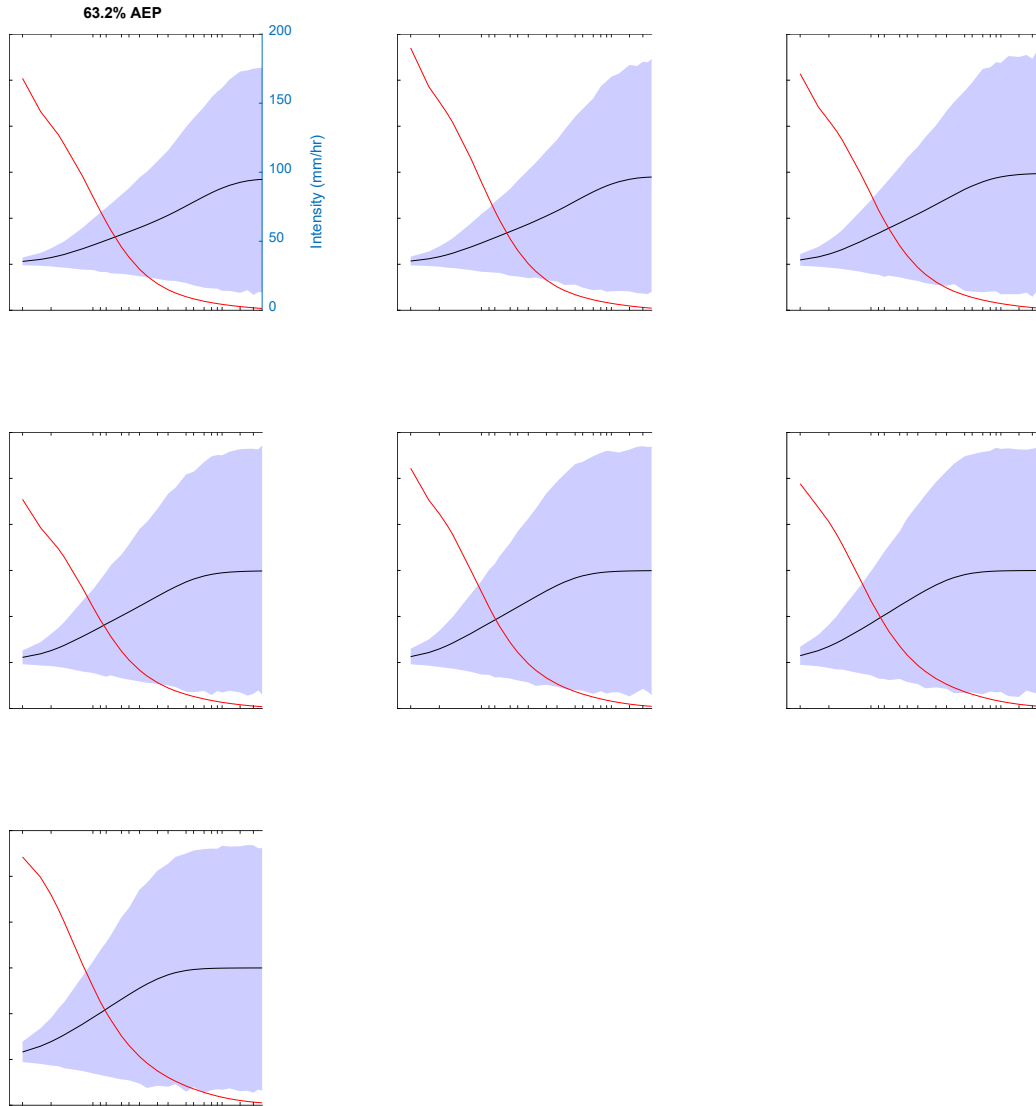


Figure S9. Intensity-Frequency-Duration-Washoff (IFDW) distributions of Total Phosphorous (TP) and associated uncertainty for frequent and infrequent design rainfall events for Gold Coast region (28.0167° S, 153.4000° E). *Note 1: horizontal axis is in log scale; Note 2: AEP – annual exceedance probability of a particular rainfall event.*

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