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## **Understanding nutrient build-up on urban road surfaces**

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#### **Abstract**

This article discusses the outcomes of a research project on nutrients build-up on urban road surfaces. Nutrient build-up was investigated on road sites belonging to residential, industrial and commercial land use. Collected build-up samples were separated into five particle size ranges and were tested for total nitrogen (TN), total phosphorus (TP) and sub species of nutrients, namely, NO<sub>2</sub>-, NO<sub>3</sub><sup>-</sup>, TKN and PO<sub>4</sub><sup>3-</sup>. Multivariate analytical techniques were used to analyse the data and to develop detailed understanding on buildup. Data analysis revealed that the solids loads on urban road surfaces are highly influenced by factors such as land use, antecedent dry period and traffic volume. However, the nutrient build-up process was found to be independent of the type of land use. It was solely dependent on the particle size of solids build-up. Most of the nutrients were associated with the particle size range < 150 µm. Therefore, the removal of particles below 150 µm from road surfaces is of importance for the removal of nitrogen and phosphorus from road surface solids build-up. It is also important to consider the difference in the composition of nitrogen and phosphorus build-up in the context of designing effective stormwater quality mitigation strategies.

2005).

Key words: nutrient pollution; pollutant build-up; stormwater pollution

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#### Introduction

Urban stormwater runoff has been recognised as a major source of pollutants into receiving water bodies (Goonetilleke et al., 2005). With the growing awareness of the adverse impacts of stormwater pollution, regulatory authorities strive to mitigate these impacts by planning and implementing a range of stormwater quality mitigation strategies. In this context, the detailed understanding of pollutants and pollutant processes is imperative for the provision of efficient and cost effective mitigation strategies. Even though the types of pollutants which significantly influence water quality is known, the effectiveness of mitigation strategies is hampered by the lack of understanding relating to pollutant processes.

Road surfaces have been recognised as a major source of pollutants to receiving urban water bodies (Ball et al., 1998; Bannerman et al., 1993). This is because of the efficient conversion of rainfall to runoff by the impervious surfaces and its conveyance to receiving water bodies by the drainage system, with minimal delay. This is further compounded by the high concentration of directly contributing anthropogenic activities such as vehicular traffic. The other pollutant contributors to urban road surfaces are atmospheric deposition and soil inputs. The composition of pollutant build-up and the pollutant loadings on road surfaces can vary with factors such as traffic volume and traffic characteristics, road surface condition, land use and antecedent dry days (Ball et al., 1998; Herngren et al.,

Nutrients have been identified as a primary stormwater

pollutant and in particular nitrogen (N) and phosphorus (P)

concentrations are sufficient to allow excessive growth of aquatic plants such as algae in receiving water bodies.

Past research studies which investigated nutrient pollution in stormwater runoff have primarily focussed on the quantification of nutrient loads from different land uses. However, an appreciable knowledge gap exists in relation to the nutrient build-up process on urban road surfaces. This article presents the outcomes of an investigation into nutrient build-up process on urban road surfaces for three different land uses; residential, industrial and commercial.

#### 1 Materials and methods

## 1.1 Study area

The research sites were selected from the Gold Coast, Queensland State, Australia. The Gold Coast is the sixth

<sup>(</sup>Bannerman et al., 1993; Carpenter et al., 1998). N and P are most commonly derived from lawn fertilizer, vehicular traffic, atmospheric deposition, organic matter, detergents and animal waste (Carpenter et al. 1998; Puckett, 1995). Excess nutrients are considered as pollutants when their

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largest city in Australia and encompasses a network of waterways, which is made up of five main rivers and numerous creeks, many of which connect to lakes and canals. Rapid urban development close to water bodies has influenced a change in the water quality due to a variety of pollutants generated from anthropogenic activities common to urban areas (GCCC, 2008).

Three road surfaces belonging to residential, industrial and commercial land uses were selected for the pollutant build-up investigations. The residential road surface investigated was located in a typical suburban area with detached family houses with small gardens. The road is used by the residents for access and reflected a relatively satisfactory surface condition. The gardens were well maintained which could have been the result of frequent use of fertiliser.

The industrial road surface investigated was an access road to a light industrial area. The activities in the area include paint, metal and cement based industries. The road surface was not in a good condition compared to the residential road surface. It was considerably degraded and had been subjected to oil spills due to the regular movement of heavy vehicles. The surface was coarse textured suggesting that a large number of fine particles would be embedded within the voids.

The surrounding land use of the commercial road surface investigated included a vehicle service station, car park area and motorcycle dealer shop. The road accommodated a relatively higher traffic volume compared to both the residential and industrial roads.

## 1.2 Sample collection

According to research literature, a significant fraction of stormwater pollutants is attached to the finer fraction of solids (defined as particles <150  $\mu m$  in this research study) (Deletic and Orr, 2005; Herngren et al., 2006). Therefore, the build-up sampling technique was selected to enhance the collection and retention of fine solids particles. Prior to deciding on the preferred sample collection technique, a review was undertaken to understand the performance of vacuuming, sweeping and brushing techniques which have been used in previous research studies (Bris et al., 1999; Robertson et al., 2003).

Consequently, a combination of brushing and vacuuming techniques was employed to collect build-up samples. A domestic vacuum cleaner which was verified to have a 95% efficiency in collecting fine particles was used for this purpose. The vacuum cleaner used was a Delonghi Aqualand model, which incorporates an efficient filtration system. The fine brush attached to the vacuum nozzle replicated the brushing action which enhanced the collection efficiency by dislodging the finer particles from the surface.

The vacuum cleaner compartment was filled with 3 L of deionised water as the filtration agent and the solids build-up was collected into the filtration compartment. The build-up samples from each road surface were collected from a 2 m  $\times$  1.5 m plot area located in the middle of the kerb and the median strip of the road surface (Fig. 1). The

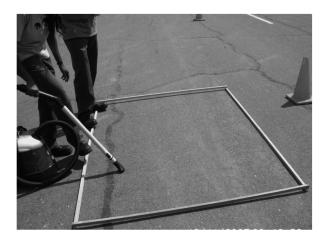


Fig. 1 Collection of build-up.

selection of a small plot area was to avoid the influence of physical factors which arise due to the heterogeneity of the surfaces and thereby limit the transferability of research outcomes between catchments (Goonetilleke et al., 2009). The collected solids were transferred to a prewashed polyethylene container. The samples were then transported to the laboratory for the analysis of the selected physico-chemical parameters.

#### 1.3 Laboratory analysis

Firstly, each build-up sample was analysed for the particle size distribution using a Malvern Mastersizer S particle size analyser. Particle size of solids is an important parameter to characterise their mobility during wash-off and their association with other pollutants (Bian and Zhu, 2008). Secondly, based on the results of the particle size distribution analysis, the build-up samples were separated into five particle size ranges by wet sieving and the potential soluble fraction was determined by filtering through a 1  $\mu m$  glass fibre filter paper. As such, each build-up sample was separated into five sub samples; > 300  $\mu m$ , 150–300  $\mu m$ , 75–150  $\mu m$ , 1–75  $\mu m$  and <1  $\mu m$ . The original build-up sample and wet sieved build-up samples were analysed for the physico-chemical parameters listed in Table 1.

#### 1.4 Data analysis

Multivariate data analysis techniques were used to identify the linkages between nutrient parameters, TS, TOC, particle size ranges and land use. In this regard, multicriteria decision making methods (MCDM) namely PROMETHEE and GAIA (Visual Decision Inc., 2000) were selected for the data analysis. PROMETHEE and GAIA have been increasingly employed to handle multivariate data in environmental data analysis (Herngren et al., 2006; Ayoko et al., 2007). These methods have the capability for ranking and pattern recognition even with a small number of data sets. As only five wet sieved build-up samples were analysed from each road surface, the application of PROMETHEE and GAIA methods were considered appropriate.

Prior to the application of PROMETHEE and GAIA a number of modelling options including a ranking order,

Table 1	Toot	parameters	a = 4	ma atha da	mod
Table I	Test	parameters	ana	mernoas	usea

Parameter	Test method
Particle size distribution	Malvern Mastersizer S particle size analyser
Total solids (TS)	Method 2540C and 2540D (APHA, 2005)
Total organic carbon (TOC)	Shimadzu TOC-5000A Total Organic Carbon Analyzer according to Method 5310C (APHA, 2005)
Nitrite nitrogen (NO <sub>2</sub> <sup>-</sup> )	SmartChem 140 discrete analyser using Method 4500-NO <sub>2</sub> -B (APHA, 2005)
Nitrate nitrogen (NO <sub>3</sub> <sup>-</sup> )	SmartChem 140 discrete analyser using Method 4500-NO <sub>3</sub> -F (APHA, 2005)
Total kjeldahl nitrogen (TKN)	SEAL discrete analyser using Method 351.2 (US EPA, 1993)
Total nitrogen (TN)	Summation of NO <sub>2</sub> , NO <sub>3</sub> and TKN
Phosphate (PO <sub>4</sub> <sup>3-</sup> )	SEAL discrete analyser using Method 4500-P-F (APHA, 2005)
Total phosphorus (TP)	SEAL discrete analyser using Method 365.4 (US EPA, 1983)

weighting condition, a specific preference function and a threshold value must be defined for each variable. The variables were maximised such that the most polluted particle size range was ranked first in the PROMETHEE analysis. The parameters were given the same weighting and hence no variable was favoured over the other. In the MCDM software, six preference functions with six specific shapes are available. The V-shaped preference function was selected for all the variables and the threshold value was set to the maximum concentration of each variable (Herngren et al., 2005). To summarise the results of all the comparisons done using the preference function, the net out ranking flow  $(\Phi)$  is computed. The larger the net flow of an object, the higher the ranking order relative to the other samples. More details on these modelling options can be found in Visual Decision Inc. (2000). PROMETHEE displays the results based on two options; partial ranking (PROMETHEE I) and complete ranking (PROMETHEE II) which can be selected according to the preference of the user.

GAIA biplot shows visually how the objects relate to one another and to the variables, as well as how variables relate to each other. In addition, it displays the decision axis,  $\pi$ , which represents the weights of the criteria. The orientation of the decision axis emphasises which criteria are predominant (Visual Decision Inc., 2000).

#### 2 Results and discussion

The data analysis was conducted in three stages. As most of the primary stormwater pollutants such as nutrients and organic carbon are attached to solids, an understanding of the primary characteristics of solids build-up was considered important prior to developing knowledge on the nutrient build-up process. Secondly, as organic matter has been identified as a major source of nutrients, test results obtained for total organic carbon were analysed (Bian and Zhu, 2008). Finally, knowledge on the nutrient build-up process was developed based on the understanding generated on solids and organic carbon build-up. In turn, the

knowledge on nutrient build-up process was extended to understand the physico-chemical parameters that influence this process.

# 2.1 Investigation of primary characteristics of total solids build-up

The amount of nutrients attached to solids varies with the particle size (Vaze and Chiew, 2004). As such, the particle size distribution of solids and the total solids load collected from each road surface were analysed to understand the primary characteristics of solids build-up on road surfaces.

According to Table 2, the TS load is significantly different between the particle size ranges. Several researchers have considered 150  $\mu m$  as a cut-off range to differentiate the finer and coarser fractions of solids build-up (Goonetilleke et al., 2009; Herngren et al., 2006). For all the road surfaces, the particle size range 75–150  $\mu m$  contains the highest percentage of total solids load. Overall, particle size ranges less than 150  $\mu m$  contains more than 70% of the total solids load for all road surfaces. This suggests that particles below 150  $\mu m$  are of specific concern in urban areas.

As evident in Table 2, the particle size distribution of solids for each road surface is different. This is attributed to factors such as the nature of anthropogenic activities and traffic characteristics (Bian and Zhu, 2008; Herngren et al., 2006). For example, the industrial road surface shows a relatively higher solids load in the particles below 150  $\mu m$  compared to the other two road surfaces. It is hypothesised that this is due to the presence of industrial enterprises such as cement based industries in the surrounding area and redistribution of fine particles due to the regular movement of heavy vehicles in the area.

Additionally, as shown in Table 2, the variation of TS load for each road surface is related to the antecedent dry days. The differences in the nature of the anthropogenic activities at each site could also be a factor that influences the build-up loads (Bian and Zhu, 2008; Zafra et al., 2008). For example, the highest solids load collected from

 Table 2
 Amount of TS load in each size range as a percentage of total solids load

Road surface		Solids load (%)				TS load (g/m <sup>2</sup> )	Antec	cedent dry days
	<1 μm	1–75 μm	75–150 μm	150–300 μm	>300 μm			
Residential	29.33	14.18	42.50	11.82	2.17	2.25	8	
Industrial	33.01	8.27	48.62	6.60	3.50	3.44	9	
Commercial	26.41	7.36	38.10	19.33	8.81	4.06	11	$C_{\Lambda}^{\diamond}$

the commercial road surface could well be attributed to longer antecedent dry days as well as high traffic volumes compared to the other two sites.

## 2.2 Investigation of total organic carbon in solids buildup

Several researchers have identified organic matter as a major source of nutrients to solids build-up (Flanagan and Forster, 1989; Makepeace et al., 1995). In addition to the contribution of nutrients from the decomposition of organic matter such as plant debris, Flanagan and Foster (1989) have suggested that the disproportionately larger surface area of organic matter can hold nutrients, thus causing an increase in the nutrients load. Consequently, the understanding of TOC build-up was important to obtain a detailed understanding of the nutrient build-up process. The organic carbon load for each particle size range per unit weight of total solids is given in Table 3.

Table 3 TOC in the solids build-up for each road surface

Road surface	TOC (mg/g)				
	<1	1-75	75-150	150-300	>300
	μm	μm	μm	μm	μm
Residential	16.32	11.43	23.50	5.63	4.25
Industrial	3.08	2.55	5.03	0.78	1.14
Commercial	4.93	2.31	7.39	2.47	3.47

As seen in Table 3, the particle size range 75–150 µm shows the highest amount of TOC and the overall size range below 150 µm contains the predominant amount of TOC for all road surfaces. These results confirm the findings of past researchers who noted significantly higher amounts of TOC in finer particles than in coarser particles (Bian and Zhu, 2008; Sartor and Boyd, 1972). Sartor and Boyd (1972) attributed this to the low structural strength of organic matter which results in being easily ground into fine particles. Additionally, the TOC loads on the surfaces vary with the type of land use with the highest load in the residential road surface for all the particle size ranges compared to the other two road surfaces. This is attributed to the greater presence of vegetation in the surrounding area.

### 2.3 Investigation of nutrients build-up process

Build-up of different species of nutrients can be different due to differences in the primary sources of nutrient species. Also, nutrient build-up can be influenced by physico-chemical parameters such as organic carbon. To understand the build-up process for different nutrient species, a detailed analysis using multivariate techniques was conducted. The linkage between different nutrient species and influential physico-chemical parameters was investigated using PROMETHEE and GAIA.

The analysis was based on the laboratory test results obtained for nitrite-nitrogen (NO<sub>2</sub><sup>-</sup>), nitrate-nitrogen (NO<sub>3</sub><sup>-</sup>), total kjeldahl nitrogen (TKN), total nitrogen (TN), phosphate (PO<sub>4</sub><sup>3-</sup>) and total phosphorus (TP) for different particle size ranges. Furthermore, as TOC showed considerable variability between the different particle size

ranges, this parameter was also included in the analysis. Additionally, TS was also included for the completeness of the analysis.

PROMETHEE and GAIA analysis was undertaken considering all the wet sieved build-up samples for all the road surfaces. This was based on the hypothesis that even though the pollutant loads are influenced by the type of land use, the underlying physical and chemical processes of pollutant build-up would not be influenced by the type of land use. However, in order to eliminate any bias due to different loadings of pollutants in the three road surfaces, the data were pre-treated prior to analysis (Ayoko et al., 2007). Table 4 shows outcomes of the PROMETHEE analysis. Figure 2 shows the principal component biplot obtained from GAIA analysis. The total data variance of 72.69% explained by the GAIA biplot indicates that the majority of the information is included in the analysis.

As evident in Table 4, the finer fraction of solids build-up is the most polluted. According to Table 4, all the particle size ranges ranked first are below 150  $\mu$ m for all the road surfaces. These are the most polluted objects. On the other hand six out of eight least polluted objects are greater than 150  $\mu$ m. Furthermore, Fig. 2 shows that all the particle size ranges with positive scores on PC1 are below 150  $\mu$ m whilst the variables NO<sub>2</sub><sup>-</sup>, TKN, TN, PO<sub>4</sub><sup>3-</sup>

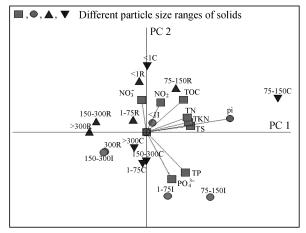


Fig. 2 GAIA analysis for all the road surfaces. R: residential, I: industrial, C: commercial.

 Table 4
 PROMETHEE II ranking

Sample	Net Φ	Ranking order
75–150C	0.42	1
75-150I	0.19	2
<1C	0.10	3
1-75I	0.08	4
75-150R	0.06	5
<1I	0.04	6
<1R	0.05	7
150-300C	-0.01	8
1-75C	-0.04	9
>300C	-0.05	10
1-75R	-0.10	11
>300I	-0.17	12
150-300I	-0.18	13
150-300R	-0.19	14
>300R	-0.21	15

R: residential; I: industrial; C: commercial.

and TP show positive loadings on PC1. This confirmed the higher affinity of nutrients to the particle size range below 150  $\mu$ m irrespective of the land use. Nutrients in these size ranges can be easily washed-off with the stormwater runoff and impose a significant threat to receiving waters. This is of serious concern as the investigation of solids build-up showed that particle size range smaller than 150  $\mu$ m contains the majority of the TS load. Therefore, the stormwater quality mitigation strategies designed to remove nutrients from road surfaces should be capable of trapping the particle size range below 150  $\mu$ m.

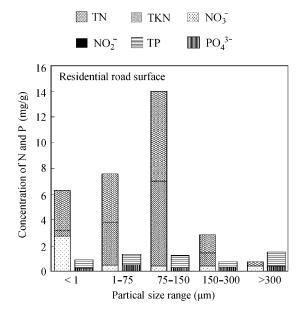
Table 4 further indicates that the particle size range 75–150  $\mu$ m for all road surfaces ranks highest, which highlights the highly polluted nature of this particle size range. On the other hand, the GAIA biplot (Fig. 2) shows particle size range 75–150  $\mu$ m for all road surfaces have high PC1 scores. Furthermore, TN and TP vectors show high positive loadings on PC1. This suggests that particle size range 75–150  $\mu$ m is the most significant for the nutrient build-up process. Notably, the decision axis  $\pi$  vector points towards the particle size range 75–150  $\mu$ m confirming the significance of this particle size range in the nutrient build-up process.

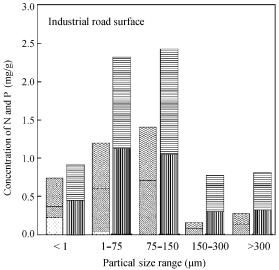
The findings from the PROMETHEE and GAIA analysis were further evaluated using the base data. The amount of nutrients available per unit weight of solids in each particles size range was determined for each road surface. Figure 3 provides the graphical representation of nutrients loads available in each particle size range for each road surface.

According to Fig. 3, the highest TN load is in the particle size range  $75{\text -}150~\mu m$  for all the road surfaces. The particle size range  $75{\text -}150~\mu m$  shows the highest TP load for both industrial and commercial road surfaces. The residential road surface shows the second highest amount of TP for the same particle size range. Therefore, it can be surmised that the particle size range  $75{\text -}150~\mu m$  exerts the strongest influence on the nutrient build-up process irrespective of the type of land use.

The nitrogen build-up on road surfaces is primarily in organic form where TOC exerts a strong influence. According to Fig. 2, TN is strongly correlated to TOC and TKN which is the organic form of nitrogen. As evident in Fig. 3, more than half of the TN amount is attributed to TKN for most of the particle size ranges. This indicates that TKN is the most dominant nitrogen species in the solids build-up on road surfaces. The importance of TOC to nitrogen build-up process is further confirmed by the highest amount of both TOC and TN in the particle size range 75–150  $\mu m$  as noted in the investigation of TOC in solids build-up (Table 3, Fig. 2). Additionally, the GAIA biplot (Figs. 2 and 3) indicate that NO2 $^-$  and NO3 $^-$  are associated mostly with the particle size range <1  $\mu m$  which confirms their high degree of solubility.

Unlike nitrogen build-up, phosphorus build-up is mostly in inorganic form. As shown in Fig. 2, TP is strongly correlated to  $PO_4^{3-}$ . As evident in Fig. 3, more than half of the TP content is attributed to  $PO_4^{3-}$  in a majority of the different particle size ranges for all the road surfaces.





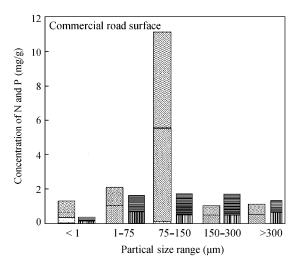


Fig. 3 Comparison of nutrients build-up.

This suggests that PO<sub>4</sub><sup>3-</sup> which is an inorganic form of phosphorus is the most dominant form of phosphorus.

Additionally, Fig. 3 shows that the particle size range 1–75  $\mu$ m has the highest amount of PO<sub>4</sub><sup>3–</sup>. This is further confirmed by the findings of Sartor and Boyd (1972) who noted that PO<sub>4</sub><sup>3–</sup> is mainly associated with the particle size of less than 43  $\mu$ m. This is a significant concern as high amounts of PO<sub>4</sub><sup>3–</sup> can be easily washed-off with the runoff and cause nutrient enrichment of receiving water bodies due to its high degree of bioavailability compared to other phosphorus species.

However, according to Fig. 3, the  $PO_4^{3-}$  fraction in TP in the particle size ranges <1  $\mu$ m, 75–150  $\mu$ m and >300  $\mu$ m at the residential road surface and particle size range 75–150  $\mu$ m at the commercial road surface is limited. This suggests that some other phosphorus forms such as organic phosphorus could also contribute to the TP load in the solids build-up. However, this relationship is not very clear as TP shows no correlation to TOC in the GAIA biplot (Fig. 2).

It is important to note that the nutrients build-up process is solely dependent on the particle size of solids rather than the type of land use. All objects in the GAIA biplot (Fig. 2) are discriminated based on the particle size range of the solids and not on the type of land use. This strengthens the importance of the design of stormwater quality mitigation strategies based on the understanding of underlying processes of nutrient build-up rather than the nutrient loads which could vary with the type of land use. Consequently, it could lead to a more robust approach for the design of stromwater quality mitigation strategies targeting the removal of nutrients from stormwater runoff.

## **3 Conclusions**

Targeting of the finer fraction of solids should be the fundamental approach to stormwater quality mitigation strategies for the removal of nutrients from road surfaces irrespective of the type of land use. Nutrient build-up process is dependent on the particle size of the solids build-up and not on the type of land use. The finer fraction (particles below 150  $\mu m$ ) of solids is the most important for the nutrient build-up process and as a general rule, the particle size range 75–150  $\mu m$  exerts the strongest influence. However, in the case of phosphate, the particle size range 1–75  $\mu m$  was also found to be significant. Therefore, a stormwater quality mitigation strategy should be capable of trapping particles < 150  $\mu m$  for the effective removal of nutrients.

Furthermore, in the context of designing stormwater quality mitigation strategies, it is also important to note that the difference in the nitrogen and phosphorus build-up processes in terms of their composition. The nitrogen build-up on road surfaces is primarily in organic form, whereas phosphorus build-up on road surfaces is primarily in inorganic form. The influence of the organic and inorganic forms of nutrients to eutrophication of receiving water bodies could vary depending on the differences in the degree of bioavailability. Therefore, the design of stormwater quality mitigation strategies should also take into consideration the differences between the composition

of nitrogen and phosphorus build-up to be more effective, instead of the design of a common set of approaches.

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