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COMPOSITION AND SOURCE IDENTIFICATION OF ROAD DEPOSITED POLLUTANTS

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Abstract: Road deposited solids are a mix of pollutants originating from a range of anthropogenic sources common to urban land uses and soil inputs from surrounding areas. These particles accumulate potentially toxic pollutants thereby posing a threat to receiving waters. Reliable estimation of sources of particulate pollutants in build-up and quantification of particle composition is important for the development of best management practices for stormwater quality mitigation. The research study analysed build-up pollutants from sixteen different urban road surfaces and soil from four background locations. The road surfaces were selected from residential, industrial and commercial land uses from four suburbs in Gold Coast, Australia. Collected build-up samples were analysed for solids load, organic matter and mineralogy. The soil samples were analysed for mineralogy. Quantitative and qualitative analysis of mineralogical data, along with multivariate data analysis were employed to identify the relative source contributions to road deposited solids.

The build-up load on road surfaces in different suburbs showed significant differences due to the nature of anthropogenic activities, road texture depth and antecedent dry period. Analysis revealed that build-up pollutants consists primarily of soil derived minerals (60%) and the remainder is composed of traffic generated pollutants and organic matter. Major mineral components detected were quartz and potential clay forming minerals such as albite, microline, chlorite and muscovite. An average of 40-50% of build-up pollutants by weight was made up of quartz. Comparison of the mineral component of build-up pollutants with background soil samples indicated that the minerals primarily originate from surrounding soils. About 2.2% of build-up pollutants were organic matter which originates largely from plant matter. Traffic related pollutants which are potentially toxic to the receiving water environment represented about 30% of the build-up pollutants at the study sites.

Key words: Pollutants build-up, Pollutant source identification, Traffic pollutants, Urban water quality.

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1 INTRODUCTION

Road deposited particulates in the urban environment result from complex and diverse sources (Adachi & Tainosho, 2004). Accumulation of particulates on road surfaces is very rapid. As noted by Egodawatta and Goonetilleke (2006), the rate of pollutant build-up on urban residential road surfaces in Australia is in the range of 1 to $2g/m^2/day$. Furthermore, it is a well known fact that the particles accumulated on urban road surfaces carry potentially toxic pollutants. This underlines the importance of solids as a significant stormwater pollutant (Hoffman, Latimer, Hunt, Mills, & Quinn, 1985).

Solids deposited on a road surface are heterogeneous resulting from atmospheric deposition, inputs from surrounding soil and pollutants originating from traffic related activities (Fergusson & Kim, 1991). Traffic related pollutants originate from tyre and brake abrasion products, combustion exhaust and pavement wear (Adachi & Tainosho, 2004; Rogge, Hildemann, Mazurek, & Cass, 1993; Kreider, Panko, McAtee, Sweet, & Finley, 2010). As noted by Beckwith, Ellis and Revitt (1986) these particles are subject to complex mixing processes occurring during transport and on-road processes which continue to alter their chemical composition. Such changes in chemical composition are also common for natural soil inputs found on road surfaces. Furthermore, due to frequent traffic activities, traffic related particles can combine with soil mineral components and produce unique mixtures (Kreider et al., 2010). Therefore, it is important to characterise the composition of road deposited particulates so that the contributing sources can be identified.

The primary aim of the research study discussed in the paper was to characterise the solids composition on road surfaces based on their mineralogy and to determine the dominant pollutant sources. This would enable the identification of physical and chemical characteristics of solids. Particle composition and pollutant source characteristics can vary with a range of factors including geographical location, land use, traffic conditions and antecedent dry period. The particle composition of road deposited particulates is also closely related to the background soil of the adjacent land (Xie, Dearing, & Bloemendal, 2000).

2 MATERIALS AND METHODS

2.1 The Study Sites

The study sites were located in the Gold Coast, Southeast

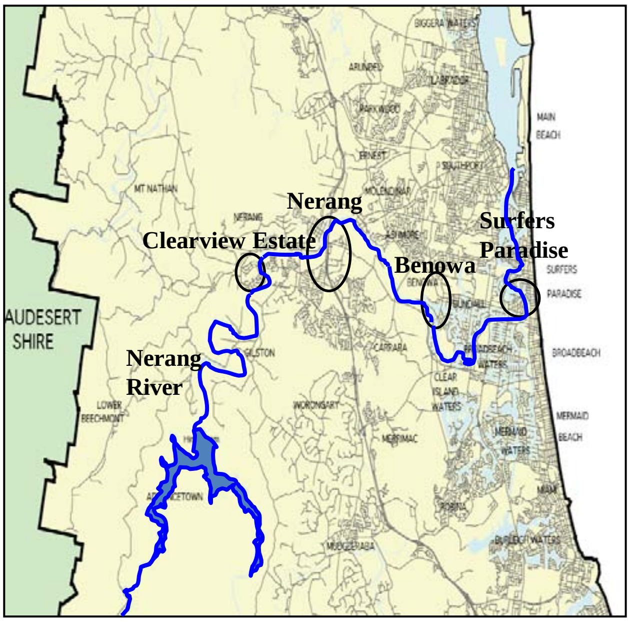

Figure 01: Study site locations

Queensland, which is one of Australia's rapidly developing urban regions. Four suburbs with different soil profiles were selected stretching from inland to the coastline along the Nerang River (Fig. 01). The selected suburbs lie along a 12.5km traverse with almost equal distance apart between each suburb. Selected suburbs were Clearview Estate, Nerang, Benowa and Surfers Paradise. A total of sixteen road surfaces with varying levels of traffic density were selected, representing four road sites from each suburb and different land uses; Clearview Estate - Residential, Nerang - Industrial, Benowa - Residential, industrial and commercial and Surfers Paradise - Commercial land uses.

2.2 Sample Collection

2.2.1 Build-Up Sample Collection

Pollutant build-up samples from each (road) study site were collected using a dry and wet vacuuming system. The area demarcated for sample collection was initially vacuumed in a dry state using a vacuum cleaner (Delonghi Aqualand make) with a water filtration system. To enhance the collection efficiency of fine particles, the road surface was then moistened by spraying deionised water (using Swift 60L compact sprayer with pressure control) under a control pressure for 3 min. and the same vacuum system was re-used. The complete collection methodology was tested under controlled field conditions and found to be 90% efficient in collecting and retaining pollutants (Mahbub, Goonetilleke, & Egodawatta, 2009).

Sample requirement for the laboratory analyses was about 45g. Build-up samples were collected from each sampling area from four to six plots with each plot having an area 2m x 1.5m. Plots were demarcated equally spaced between the median strip and the kerb or in the middle of parking spaces, assuming uniform pollutant build-up across the surface.

Two separate samples were collected from each road surface representing different antecedent dry periods to account for changes in sample composition. Furthermore, previous research studies have found that pollutant accumulation in the initial period is rapid and tend to reach equilibrium in around seven to nine dry days (Egodawatta & Goonetilleke, 2006; Ball, Jenks, & Aubourg, 1998). Consequently, sampling times were selected such that the antecedent dry period for one sampling episode was less than eight days and the other greater than eight days. The sampling was carried out in two different seasons to account for the seasonal variations in pollutant build-up. Additionally, the road surface texture depth was measured according to procedures specified in (Federal Highway Administration [FHWA], 2005).

2.2.2 Soil Sample Collection

Soil samples adjacent to study sites were collected to obtain background information about the surrounding soil properties. This data was used to interpret the influence of soil inputs to the road surface pollutant build-up. Soil sample collection and preservation was carried out according to the methods specified in the Australian Laboratory Handbook of Soil and Water Chemical Methods (Rayment & Higginson, 1992).

2.3 Laboratory Analysis

The collected build-up samples were analysed for total solids (TS) and total organic carbon (TOC). Additionally, the mineralogy was determined using X-ray diffraction (XRD) analysis on powdered build-up solids and soil samples using PANalytical X'Pert PRO Multi-purpose diffractometer. In the case of low proportions of clay minerals, orientated sample analysis was carried out by drying a dilute suspension of clay on a silicon wafer. Orientated samples improve the detection of small clay quantities present in the sample.

2.4 Data Analysis

The experimental data formed a large data set including mineralogical components of solids and soil samples. Analysis of a large data set with multiple variables require analytical approaches capable of clustering similar data together whilst identifying relationships between variables. In these circumstances, the application of multivariate analytical techniques has been found to be the most appropriate (for example Settle, Goonetilleke, & Ayoko, 2007; Herngren, Goonetilleke, & Ayoko, 2005). In this study, Principal Component Analysis (PCA), which is an analytical technique frequently applied in the analysis of environmental data was used.

The PCA technique is used to transform the original variables to a new orthogonal set of Principal Components (PCs) such that the first PC contains most of the data variance and the second PC contains the second largest variance and so on. Objects that exhibit similar variances for the analysed variables have similar PCA scores forming a cluster when plotted on a biplot. Additionally, strongly correlated variables have the same magnitude and orientation when plotted, whereas uncorrelated variables are orthogonal to each other. Detailed descriptions of PCA can be found elsewhere (Adams, 1995; Massart, Vandeginste, Deming, Michotte, & Kaufman, 1988). Data pre-treatment is carried out to reduce 'noise' which interferes with the data analysis (Kokot, Grigg, Panayiotou, & Phuong, 1998). Accordingly, the data matrix was column centred and standardised (auto scaled). Build-up pollutants were taken as load per unit area of road surface (mg/m^2).

3 RESULTS AND DISCUSSION

3.1 Characteristics of Build-up Pollutants

Initially, the overall variability of build-up pollutant loads based on site and land use was compared. The road surface texture depth, antecedent dry days, average build-up loads and total organic carbon content (TOC) for each suburb are given in Tab. 01. As evident in Tab. 01, the build-up load in the majority of the study sites is not proportional to the antecedent dry period.

TABLE 01: Site Description and Build-up Pollutant Load Characteristics

Suburb	**Land use**	**Road texture depth range (mm)**	**Antecedent dry days**	**Avg. TS load (g/m^2)**	**Avg. TOC (%)**
Clearview Estate	R-4	0.76-0.92	8	1.79	1.54
			17	0.81	4.22
Nerang	I-4	0.93-1.14	5	3.53	0.76
			9	7.03	0.77
Benowa	I,C-1, and R-2	0.80-0.91	8	1.39	1.81
			14	0.78	5.12
Surfers	C-4	0.63-1.11	4	2.22	2.73

Paradise			10	1.75	0.25

Notes:
R: Residential; I: Industrial; C: Commercial
Numbers in second column indicates the corresponding number of road sites representing each land use.

However, previous studies have shown that there is an increase in build-up load with the increase in dry days (Egodawatta & Goonetilleke, 2006). The seemingly contradictory results were attributed to pre-existing pollutant load on the road surfaces, which would have been influenced by the last rain event (Egodawatta, Thomas, & Goonetilleke, 2007).

The build-up load on road surfaces in different suburbs showed significant differences. It can be postulated that the differences are due to the nature of anthropogenic activities, traffic density, road texture depth and antecedent dry period (Egodawatta & Goonetilleke, 2006; Ball et al., 1998). This was quite evident in industrial road surfaces of Nerang, where considerably high pollutant load was recorded for shorter dry periods. These roads also had relatively high average surface texture depth. The results further showed that solids from residential land use contain a high percentage of organic material compared to commercial and industrial areas. This could be attributed to the surrounding vegetation at these road sites.

3.2 Mineralogy of Solids and Surrounding Soil

3.2.1 Analysis of Pollutant Characteristics

As hypothesised, solids composition can vary with a range of factors including geographical location, land use, traffic characteristics and antecedent dry period. Detailed understanding of the mineralogy and its variation with the underlying factors is important to derive knowledge on solids composition. Major mineral components found in solids were quartz and potential clay forming minerals such as albite, microline, chlorite and muscovite. Furthermore, minor proportions of orthoclase, kaolinite, riebeckite which are clay minerals were also detected in a few samples. Other than that, a significant proportion of amorphous content (un-identified fraction of about 40%) was detected in all the samples irrespective of the study site. The results showed that the road deposited solids consist primarily of mineral matter which account for a minimum of about 60% of the sample. Among these minerals, quartz was the dominant mineral which commonly ranged between 40-50%. The present results are consistent with previously published data. Roger, Montrejaud-Vignoles, Andral, Herremans and Fortune (1998) observed similar mineralogical components based on the analysis of stormwater runoff solids particles. Based on a study in Brisbane, Al-Chalabi and Hawker (1996) reported high sand content and very low clay content in the road deposited

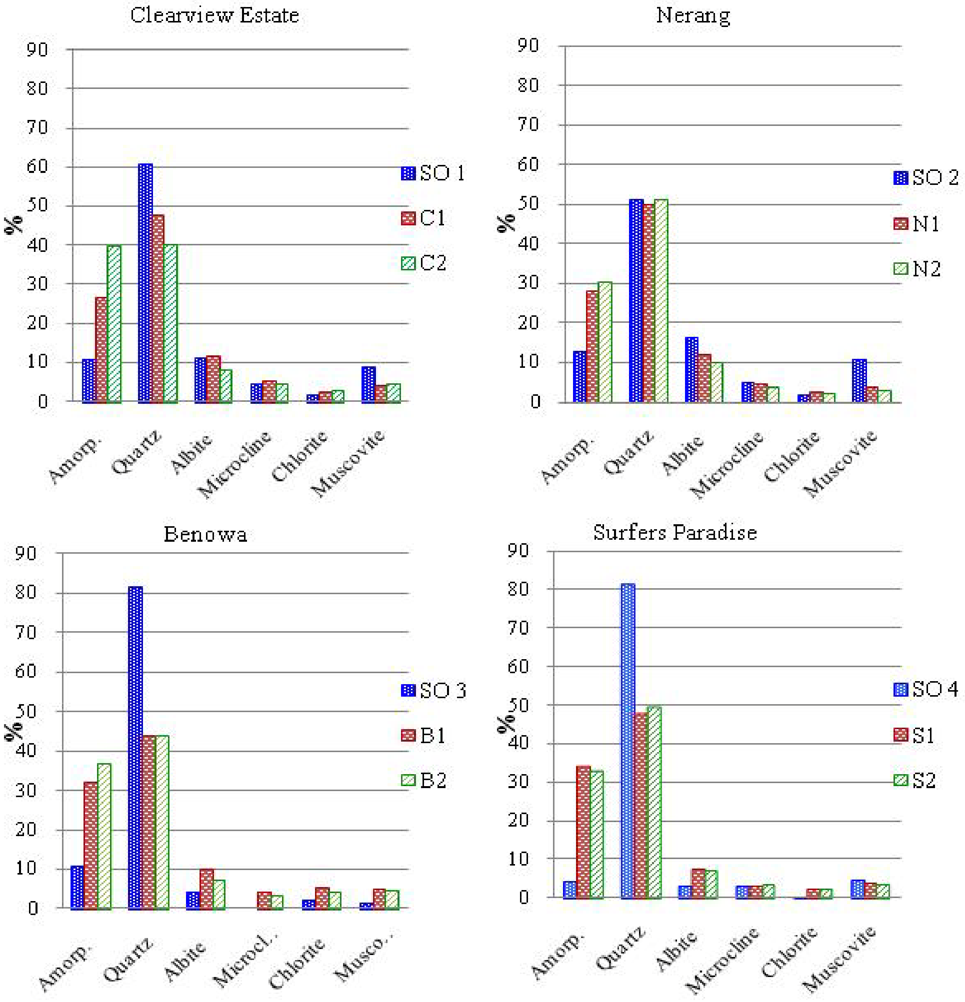

FIGURE 02: Mineralogical variations of solids and soil samples in four suburbs

Note: SO - soil, C – Clearview Estate, N- Nerang, B- Benowa and S- Surfers Paradise, 1 and 2 - first and second sampling event.

solids, similar to the present study.

Minerals detected in soil samples included, quartz, albite, microline, chlorite, muscovite and kaolinite. Minerals identified in soil samples were identical to those found in solids samples except kaolinite. This clearly confirmed that there are inputs from surrounding soil to the road surfaces. To further confirm this observation, the mineral composition of soil samples was compared to the road deposited solids samples from the two sampling episodes in each suburb. Fig. 02 shows solids and soil samples have identical mineral components with soil inputs dominating the road surface pollutants. This is in agreement with findings of Xie et al. (2000) who noted that surface soils form a significant component of street deposited solids.

The effect of geographical location on mineralogical composition in road deposited solids is evident in Fig. 02. For example, in the soil samples, the quartz content in the coastal suburbs of Surfers Paradise and Benowa is around 80% and in the inland suburbs such as Nerang and Clearview Estate, it is in the range of 50-60%. Though the quartz content in the solids samples in the four suburbs do not vary significantly, the effect of geographical location is evident in clay forming minerals. Clay forming minerals such as albite, microline, muscovite and chlorite in solids samples in inland suburbs are about 6% higher than the coastal suburbs. However in soil samples, this difference is about 20%. The substantial differences in mineral percentages in road deposited solids compared to soil are attributed to the higher amorphous content in solids.

Amorphous content was the second largest component in solids at all study sites and it was the most significant difference between road deposited solids and surrounding soil samples. According to Fig. 02, the amorphous content in soil ranged between 4.5-10%. The amorphous content in soil was in similar range as reported by Khalil (2005) and Dawes (2006). Amorphous content could be due to partially weathered minerals (non crystalline mineral), low content of certain clay minerals (that do not contribute to diffractograms due to the limit of detection), amorphous silicate (non crystalline quartz) and organic matter, as noted by Bish and Post (1989). However, the amorphous content in solids was 10-40% higher compared to soil samples. On average, the organic matter content in the solids samples were <2.2% (see Tab. 01). This suggested that the solids samples contained on average about 30% of material that can be classified as amorphous but different to the amorphous material commonly present in soil samples. A proportion of amorphous content could be attributed to low content of clay minerals (Khalil, 2005). Therefore, in order to identify potential components of clay minerals in solids particles, thin film clay analysis was carried out which can detect the presence of small fractions of clay minerals.

3.2.2 Clay Analysis

Samples of thin film X-ray diffraction patterns for a solids sample and soil sample from Nerang are shown in Fig. 03. Fig. 03 (a) thin film X-ray patterns show reflections of illite, semectite, kaolinite, chlorite and mixed layer illite-smectite. Illite, mica and chlorite minerals were present in all the samples and kaolinite and sepolite were present in a majority of the samples. Poorly defined reflections of smectite, amphibole, palygorsite and mixed layer illite-smectite were present in a few of the samples. The analysis indicated that the crystalline form of the above clay minerals were present in the finer solid particles which were not detected in the powder diffraction patterns and accounted as amorphous material. As seen in Fig. 03 (b) detected clay minerals in soil samples were smectite, illite-mica, kaolinite, chlorite and mixed layer of illite-smectite.

Qualitative measurements obtained from clay analysis of solids and soil samples indicated that a portion of the amorphous content present in solids could account for the minor portions of clay minerals. The clay minerals detected in soil and solid samples did not show significant differences. Yet high proportions of amorphous content were detected in the solids. This indicated that material other than clay minerals was present in the road deposited solids.

Therefore it can be postulated that the high percentage of amorphous content in solids samples are generated from traffic sources. As noted by Kreider et al. (2010) traffic related pollutants on road surfaces are subject to change with a variety of driving conditions, for example vehicle speed, load, acceleration, braking and steering. Influence of above noted traffic parameters was clearly evident in the present study as well, which indicated that approximately 5-10% high amorphous content in Surfers Paradise and Benowa suburbs than Clearview Estate which has a relatively low traffic density. In addition to the traffic parameters, surface texture is also an important parameter in relation to traffic related pollutant generation as it influences tyre wear or the frictional force between the tyre and pavement (Dahl et al., 2006). Therefore, the surface texture would have influenced the higher amorphous content in the solids in Nerang and Surfers Paradise suburbs compared to Clearview Estate (See Tab. 01 and Fig. 03). In addition, increase in the amorphous content with different antecedent dry days can be noted in the solids from the two different sampling events. This is attributed to the re-distribution, segregation and particle generation due to frequent vehicle movement. During the antecedent dry period, mineralogical composition could change which may then influence the physical and chemical characteristics of the road deposited solids. Thus, the

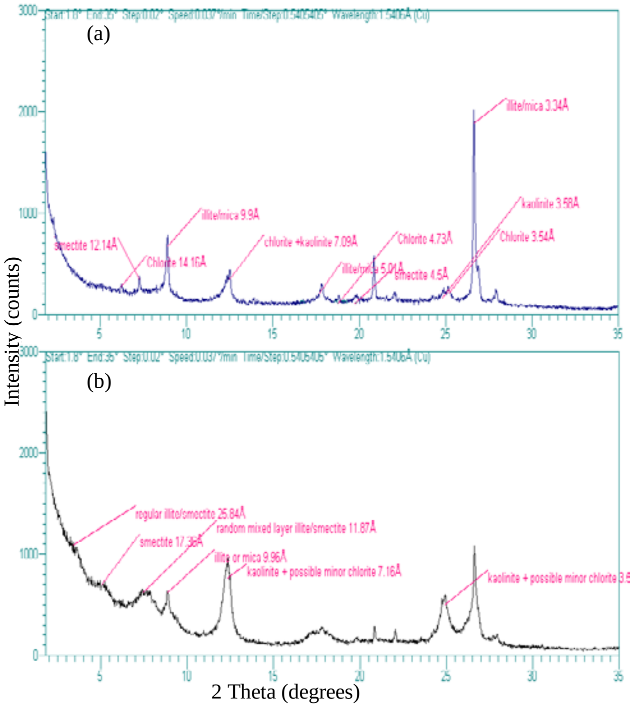

FIGURE 03: Thin film X-ray diffraction patterns of (a) Solids collected at Stevens Street at Nerang (b) Soil collected at Nerang

road deposited solids composition show a dynamic behaviour in particulate accumulation and it confirms that amorphous material is primarily originating from traffic related sources.

Similarly, Robertson, Taylor and Hoon (2003) employing geochemical and mineralogical analysis identified that urban road deposited solids are predominantly composed of non-soil derived material containing high levels of heavy metals. As noted by Rogge et al. (1993), 7.6% of the fine particulate road dust in street surfaces in Los Angeles area is vehicular exhaust particles. Besides, there could be particles derived from pavement surface wear which account for non-mineral particles. Thus, the mineralogical data provide valuable insights into the signature characteristics of road deposited solids and the role of surrounding soil deposition on road surfaces.

3.2.3 Characterisation of Build-up and Pollutant Source Identification

To further confirm whether the amorphous content found in samples originate from soil inputs or anthropogenic sources, the analysis was further refined using PCA.

A pre-treated data matrix of 36 x 6 which consisted of mineralogical data of road deposited solids and soil samples were subjected to PCA. The resulting biplot is shown in Fig. 04. As seen in Fig. 04, PC 1 explains the mineralogical variation of particles which are associated with high negative loading of quartz and positive loadings of other mineral components. PC 2 explains the pollutant source variability which is associated with strong negative loading of amorphous material while all of the other mineral components show positive loading on PC 2.

All of the soil samples show positive scores on PC 2 and are clustered orthogonal to the direction of amorphous and quartz vectors. This was primarily due to the low content of amorphous material generally present in these soils. In the biplot, soil samples develop a cluster in the perimeter of the road deposited solids cluster (dotted line in Fig. 04). However they are not entirely separated from the solids sample cluster. This indicates that road deposited solids have close correlation with soil, but there are differences due to the amorphous content. Furthermore, this reflects the pollutant source variability of the solids due to soil input and traffic related pollutants. Therefore, it can be argued that the amorphous content is the primary reason for the separation of soils from road deposited solids. Furthermore, the analysis indicated that soil originating from the adjoining area as a result of erosion or atmospheric deposition is the major source of the mineral components to the road deposited solids. Also, in Fig. 04 the amorphous content has a negative correlation with quartz and no correlation with albite, microline, and muscovite minerals. This indicates that the amorphous material contains relatively low mineral components. Therefore, it can be postulated that the major portion of the amorphous content in the solids could be material other than clay or non-crystalline mineral components. This further confirms the contribution of traffic related pollutants to road deposited solids.

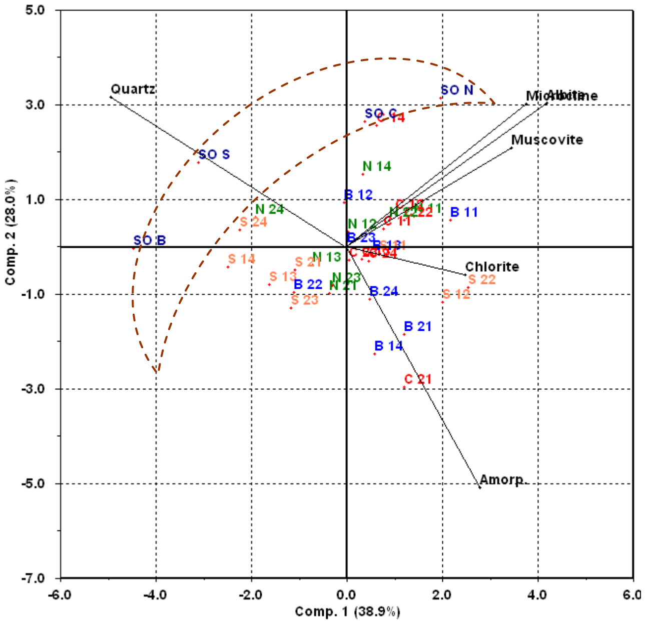

FIGURE 04: PC 1 vs PC 2 biplot obtained from PCA analysis for solids and soil samples

Note: SO - soil, C – Clearview Estate, N- Nerang, B- Benowa and S- Surfers Paradise, 1 and 2 - first and second sampling event.

The results obtained confirmed that mineralogical analysis can be applied to characterise the composition of solids on road surfaces. The mineralogical analysis of road deposited solids along with the background soil allowed the assessment of the fraction of soil in pollutants build-up and to identify the pollutant sources. Furthermore, the study outcomes provide quantification of anthropogenic pollutants on road surfaces in the urban environment.

4 CONCLUSIONS

The research study characterised road deposited solids particles in different urban land uses. The following important conclusions were derived from the study:

- The highest pollutant load was noted in the industrial land use sites.
- Residential land uses had the highest organic matter content. It is likely that the organic content is mainly derived from the surrounding vegetation.
- The solids particles on road surfaces were predominantly composed of soil derived mineral components. Quartz was the dominant mineral which ranged between 40-50% of the sample, irrespective of the geographical location.
- The influence of geographical location on mineralogical composition was evident. For example, in the soil samples, the quartz content in the coastal suburbs was around 80% and in the inland suburbs it was in the range of 50-60%. In contrast, the clay mineral content in solids samples in the inland suburbs were always higher compared to the coastal suburbs.
- Mineralogical analysis coupled with clay analysis of the road deposited solids and the surrounding soils indicated that vehicular sources contribute about 30% of the non-soil derived pollutants to the build-up pollutant load. This indicates that anthropogenic activities (ie. traffic related sources) are significant pollutant contributors to urban road surfaces.
- Road deposited solids composition show dynamic behaviour of accumulation, especially anthropogenic pollutants. Site specific parameters such as geographical location, anthropogenic activities, road texture depth and antecedent dry period have significant influence on the

composition of traffic related particles and clay forming minerals in pollutants build-up.

- The study results confirmed that mineralogical analysis can provide important information on signature characteristics of the pollutant sources on road deposited solids.

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