

Queensland University of Technology Brisbane Australia

This is the author's version of a work that was submitted/accepted for publication in the following source:

Gunawardana, Chandima, Goonetilleke, Ashantha, Egodawatta, Prasanna, Dawes, Les A., & Kokot, Serge (2012) Source characterisation of road dust based on chemical and mineralogical composition. *Chemosphere*, *87*(2), pp. 163-170.

This file was downloaded from: http://eprints.qut.edu.au/49070/

© Crown Copyright © 2011 Published by Elsevier Ltd.

This is the author's version of a work that was accepted for publication in <Chemosphere>. Changes resulting from the publishing process, such as peer review, editing, corrections, structural formatting, and other quality control mechanisms may not be reflected in this document. Changes may have been made to this work since it was submitted for publication. A definitive version was subsequently published in Chemosphere, [VOL 87, ISSUE 2, (April 2012)] DOI: 10.1016/j.chemosphere.2011.12.012

Notice: Changes introduced as a result of publishing processes such as copy-editing and formatting may not be reflected in this document. For a definitive version of this work, please refer to the published source:

http://dx.doi.org/10.1016/j.chemosphere.2011.12.012

Source characterisation of road dust based on chemical and mineralogical composition

Chandima Gunawardana¹, Ashantha Goonetilleke¹*, Prasanna Egodawatta¹, Les Dawes¹, Serge Kokot²

¹School of Urban Development, Queensland University of Technology, G.P.O. Box 2434, Brisbane, QLD 4001, Australia

² Faculty of Science and Technology, Queensland University of Technology, G.P.O. Box 2434 Brisbane, QLD 4001, Australia

Email: chandima.gunawardana@qut.edu.au a.goonetilleke@qut.edu.au p.egodawatta@qut.edu.au l.dawes@qut.edu.au s.kokot@qut.edu.au

*Corresponding author: Tel.:+61 7 3138 1539; fax: +61 7 3138 1170

Email: a.goonetilleke@qut.edu.au

Source characterisation of road dust based on chemical and mineralogical composition

Abstract: Road dust contain potentially toxic pollutants originating from a range of anthropogenic sources common to urban land uses and soil inputs from surrounding areas. The research study analysed the mineralogy and morphology of dust samples from road surfaces from different land uses and background soil samples to characterise the relative source contributions to road dust. The road dust consist primarily of soil derived minerals (60%) with quartz averaging 40-50% and remainder being clay forming minerals of albite, microcline, chlorite and muscovite originating from surrounding soils. About 2% was organic matter primarily originating from plant matter. Potentially toxic pollutants represented about 30% of the build-up. These pollutants consist of brake and tire wear, combustion emissions and fly ash from asphalt. Heavy metals such as Zn, Cu, Pb, Ni, Cr and Cd primarily originate from vehicular traffic while Fe, Al and Mn primarily originate from surrounding soils. The research study confirmed the significant contribution of vehicular traffic to dust deposited on urban road surfaces.

Key words: pollutants build-up; pollutant source identification; road dust; road surface pollutants; traffic generated particles

1 Introduction

Among particulate pollutants on urban road surfaces, fine solids form the dominant fraction and exert a significant influence on stormwater due to ease of mobility (Deletic et al., 2000; Herngren et al., 2006; Goonetilleke et al., 2009). Furthermore, fine solids particles on road surfaces are also a significant source of air pollution in urban areas (Thorpe and Harrison, 2008; Amato et al., 2009, 2011). It is commonly known that particles accumulated on urban road surfaces carry toxic pollutants such as heavy metals (Rogge et al., 1993; Gunawardana et al., 2011). Dust particles function as a carrier of other pollutants which is largely dependent on the particulate composition in terms of the nature of different minerals and organic compounds and their proportions (McBride, 1994). Therefore, characterising the sources and composition of dust particles on urban road surfaces is important for the development of appropriate pollution mitigation strategies.

Road deposited particulates in the urban environment are a heterogeneous mix originating from diverse sources, and source characterisation can be complex (Adachi and Tainosho, 2004). Potential sources include the erosion of surrounding soil, atmospheric deposition and anthropogenic activities including traffic related activities (Fergusson and Kim, 1991; Amato et al., 2009, 2011; Kreider et al., 2010). Traffic related pollutants include tire and brake abrasion products, combustion exhaust and pavement wear (Amato et al., 2011; Rogge et al., 1993; Kreider et al., 2010). As noted by Beckwith et al. (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 clearly identified. The

primary aim of the research study discussed in the paper was to source characterise the solids deposited on road surfaces based on their mineralogy, heavy metal content and morphology.

2 Materials and methods

2.1 Study sites

The study sites consisting of road surfaces were selected from the Gold Coast, Southeast Queensland, which is one of Australia's rapidly growing urban regions. The primary criterion considered in study site selection was the anticipated variation in particle composition of road dust. Consequently, four suburbs with different soil profiles were selected extending from the inland to the coast along the Nerang River (Figure 1). It was hypothesised that soil inputs from the surrounding area would be a primary source of particulates to road surfaces. The most common soils in the Gold Coast region are Dermosols (known as Red and Yellow Podzolic) and Kurosols which are mainly derived from the Neranleigh–Fernvale meta sediments and clayey sub-soils (Noble, 1996). The four selected suburbs; Clearview Estate, Nerang, Benowa and Surfers Paradise lie along a 12.5 km traverse in an approximate line extending from east to west and are equal distance apart from each other. 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 – mostly commercial.

Insert Fig. 1

2.2 Sample collection

2.2.1 Build-up sample collection

Past research has indicated that the wash-off process resulting from a rainfall event removes only a portion of the road dust available on the surface (Sartor et al., 1974; Egodawatta et al., 2007). Therefore, investigating wash-off for pollutant source characterisation could be misleading. Accordingly, the research study discussed in the paper investigated the dust build-up on urban road surfaces which would represent all available pollutants.

Road dust samples from each 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 brand) with a water filtration system. To enhance the collection efficiency of fine particles, the road surface was then moistened by spraying deionised water (using a Swift 60L compact sprayer with pressure control) under a control pressure for 3 min. and the same vacuum system was re-used to collect the wet sample (Mahbub et al., 2009). The complete collection methodology was tested under controlled field conditions and found to be 90% efficient in collecting and retaining dust samples. Other studies have reported similar collection efficiencies on artificial surfaces under laboratory conditions, using only vacuum systems or in combination with sprayers (for example Deletic and Orr, 2005; Herngren et al. 2006). The method adopted for this research study achieved the same efficiency in collecting from an actual road surface which is subject to wear and tear from daily traffic. Build-up samples were collected from each sampling area from four to six plots with each plot having an area of 3 m^2 . The plot size selected ensured that the road dust build-up was homogeneous (Gunawardana et al., 2011). The plots were equally spaced between the median strip and the kerb or in the middle of parking spaces, to obtain representative samples particularly in relation to chemistry and mineralogy and to

maintain consistency in the sampling.

Two separate samples were collected from each road surface representing different antecedent dry periods to account for changes in sample composition. Past research studies have found that total dust build-up in the initial period is rapid and tends to reach equilibrium in around seven to nine dry days (Egodawatta and Goonetilleke, 2008). 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. Additionally, the road surface texture depth was measured according to the recommendations provided by FHWA (2005).

2.2.2 Soil sample collection

At each suburb, surface soil samples adjacent to road surfaces were collected to gather background information about the surrounding soil properties. Samples to a depth of 10-15 cm were collected using a stainless steel shovel from transects parallel to the road at three to four meters away from the kerb. Two samples were collected from each site at approximately 20 m apart and mixed together. This data was used to interpret the influence of soil inputs to the road surface dust build-up. Soil sample collection and preservation were carried out according to the methods specified in Rayment and Higginson (1992).

2.3 Laboratory analysis

Road dust samples collected in the water filtration compartment of the vacuum system was wet sieved and the <425 μ m size fraction was separated and the large size fraction discarded. The soil samples were collected in dry state and were also sieved to separate out the particles larger than <425 μ m. The collected build-up solids and soil samples were analysed for a range of physical and chemical parameters including total solids (TS), total organic carbon (TOC), pH and electrical conductivity (EC). Sample testing was undertaken according to test methods specified in Standard Methods for the Examination of Water and Wastewater (APHA, 2005).

Mineralogy was determined by X-ray diffraction (XRD) analysis of powdered build-up solids (<425 μ m) and soil (<2 mm) samples using PANalytical X'Pert PRO Multi-purpose diffractometer. A 10% weight of corundum (Al₂O₃) was used as the internal standard in the quantitative analysis of mineralogical components. Percentage recoveries of corundum were within acceptable limits for this analysis (Khalil, 2005). Sample preparation and pretreatment was carried out according to the techniques described by Brindley and Brown (1984). In the case of low proportions of clay minerals, orientated sample analysis to improve the detection of small clay quantities was carried out by drying a dilute suspension of clay on a silicon wafer.

Build-up solids and soil samples were also examined for morphology and elemental composition using a Quanta 3D FIB (Focused Ion Beam) scanning electronic microscope (SEM). The samples were gold coated to improve conductivity and better image resolution after mounting on a carbon tape attached to aluminium studs (Kreider et al., 2010). Elemental compositions of the particles were determined with an energy dispersive X-ray spectroscope (EDX) to characterise the heavy metal elements attached to the particles. The elemental analyses of particles were carried out from the centre of the particle with an acceleration voltage of 30 kV, a working distance of 15 mm, and an EDX collection time of 20 s. Backscattered electron images (BEIs) were also taken over 0.01 mm² area at a 1000 magnification.

Both, build-up solids and soil samples were tested for Iron (Fe), Aluminium (Al), Lead (Pb), Zinc (Zn), Cadmium (Cd), Chromium (Cr), Manganese (Mn), Nickel (Ni) and Copper (Cu) (Method 200.8, US EPA, 1994). According to research literature, these heavy metals are among the most common in dust build-up on road surfaces (Herngren et al., 2006). Nitric acid (HNO₃) digestion for heavy metals extraction was carried according to the Method 200.8 (US EPA, 1994). Precision and accuracy of analysis and the digestion procedure was monitored using internal standards, certified reference material and quality control blanks. Multi-element standards supplied by Accustandard were used as calibration standards. Multielement standards from Sigma-Aldrich were used as the certified reference material. The percentage recovery was in the range of 85% to 115%, which is within the specified limits.

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 (Settle et al., 2007; Herngren et al., 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 to 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 (Massart et al., 1988). Data pretreatment was carried out to reduce 'noise' which interferes with the data analysis (Kokot et al., 1998). Accordingly, the data matrix was column centred and standardised (auto scaled). This was done by subtracting the column mean from each cell value and dividing by the column standard deviation. This ensured equal significance of variables with a standard deviation of 1 (Kokot et al., 1998).

3 Results and discussion

3.1 Characteristics of road dust

Initially, the overall variability of road dust loads based on site and land use was compared. The road surface texture depth, average build-up solids loads and total organic carbon content (TOC) for each suburb are given in Table 1.

Insert Table 1

The road dust load characteristics for different suburbs indicated significant differences (Table 1). It was postulated that the differences are due to the nature of anthropogenic activities, traffic density, road texture depth, traffic lane characteristics and antecedent dry period (Egodawatta et al., 2007). This was quite evident in the industrial road surfaces in Nerang, where considerably high road dust loads were recorded for relatively shorter dry periods in comparison to residential and commercial land use sites. These roads also had relatively high average surface texture depth. The results confirmed that road dust from

residential land use areas 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 soils 3.2.1 Analysis of road dust characteristics

Road dust composition can vary with a range of factors including geographical location, land use, traffic characteristics and antecedent dry period (Amato et al., 2011; Roger et al., 1998). Therefore, for obtaining a detailed understanding of the mineralogy of the solids particles and its variation with the underlying factors, the mineral percentages of the samples from the two sampling episodes in each suburb were compared (Figure 2).

Insert Fig. 2

The major mineral components were quartz which ranged between 40-50% and potential clay forming minerals such as albite, microcline, chlorite and muscovite. Additionally, minor proportions of orthoclase, kaolinite, riebeckite, which are also clay minerals were detected in some samples. Furthermore, a significant proportion of amorphous content (an unidentified fraction of about 40%) was detected in all the samples irrespective of the study site. The results showed that road dust primarily consist of mineral matter which account for a minimum of about 60% of the sample (Figure 2). The results obtained were consistent with previously published research data. For example, Roger et al. (1998) observed similar mineralogical components based on the analysis of solids particles from stormwater runoff. Based on a study in Brisbane, Australia, Al-Chalabi and Hawker (1996) reported high sand content and very low clay content in the road dust, similar to the present study.

Minerals identified in soil samples were similar to those found in road dust samples except kaolinite. Minerals detected in soil samples included, quartz, albite, microcline, chlorite, muscovite and kaolinite. This clearly confirmed that there are inputs from surrounding soil to the road surfaces. This is in agreement with findings of Xie et al. (2000) who noted that surface soils form a significant component of road dust.

The influence of the geographical location on the mineralogical composition of both, road dust and soils are evident in Figure 2. Clay forming minerals in soils such as albite, microcline, muscovite and chlorite content is higher in the inland suburbs compared to the coastal suburbs. The same characteristics can be observed for the soil samples.

The amorphous content was the second largest component in road dust at all the study sites. According to Figure 2, the amorphous content in soils ranged between 4.5-10%, which is in a similar range as reported by Dawes et al. (2006). Amorphous content could be due to partially weathered minerals (non-crystalline mineral), certain clay minerals present in low content (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 road dust was 10-40% higher compared to the soil samples. On average, the organic matter content in the road dust samples were <2.2% (see Table 1). This suggested that the road dust 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. The consistent appearance of high amorphous content in road dust leads to the conclusion that the amorphous content could be of anthropogenic

origin, being incorporated in the road dust as a result of traffic activities.

3.2.2 Clay analysis

It was hypothesised that a fraction of the amorphous content could consist of clay minerals present in low proportions. Accordingly, thin film clay analysis was carried out to detect the presence of small fractions of clay minerals (Khalil, 2005). Thin film X-ray patterns of road dust samples showed reflections of illite, smectite, kaolinite, chlorite and mixed layer illite-smectite. Illite, mica and chlorite minerals were present in all the road dust samples. Kaolinite and sepiolite were present in a majority of the samples whilst poorly defined reflections of smectite, amphibole, palygorskite and mixed layer illite-smectite were present in a few of the samples. Thus, it confirmed that the crystalline form of the above clay minerals were present in the road dust particles which were not detected in the powder diffraction patterns and accounted as amorphous material. The clay minerals detected in the soil samples were smectite, illite-mica, kaolinite, chlorite and mixed layer of illite-smectite. Qualitative measurement showed no significant difference in clay minerals in soil and road dust samples, yet higher amount of amorphous content was detected in road dust. This indicated that material other than clay minerals was present in the road dust which confirmed possible anthropogenic contribution to the amorphous content.

The presence of traffic related amorphous content can be further confirmed by the relationships with traffic related factors. As noted by Kreider et al. (2010), traffic related pollutants on road surfaces are subject to change with a variety of factors such as vehicle speed, load, acceleration, braking and steering. Approximately 5-10% high amorphous content in Surfers Paradise and Benowa suburbs compared to Clearview Estate residential suburb clearly confirm the contribution of above noted traffic factors on pollutants generation. In addition to the traffic parameters, the surface texture can also be an important parameter in relation to traffic related pollutant generation as it influences tire wear or the frictional force between the tire and pavement and consequent increase in the solids load on the road surface (Dahl et al., 2006; Amato et al., 2009). Therefore, the possibility exists that surface texture would have influenced the higher amorphous content in the road dust at Nerang and Surfers Paradise suburbs (refer Table 1). However, relatively high amorphous content was detected at Benowa suburb which had a lower surface texture depth. It is hypothesised that this is due to the presence of industrial and commercial activities in this suburb.

3.2.3 Characterisation of road dust and pollutant source identification

The data analysis was further refined using PCA. A pre-treated (auto scaled) data matrix of 36 x 6 was subjected to PCA. The 36 objects in the matrix represented the 32 road dust samples collected from each of the four suburbs consisting of four road sites each with two sampling episodes per site and four soil samples from each of the four suburbs. The six variables consisted of the mineralogical components. The resulting biplot is shown in Figure 3. As evident in Figure 3, PC 1 primarily explains the variation of mineralogical composition of particles which is associated with quartz on the negative PC 1 axis and other mineral components associated with amorphous material along the negative PC 2 axis and all of the other mineral components along the positive PC 2 axis.

Insert Fig. 3

All of the soil samples show positive scores on PC 2 and are clustered orthogonal to the direction of amorphous and quartz vectors. This is primarily attributed to the low content of amorphous material generally present in these soils. In the biplot, soil samples form a cluster (dotted line in Figure 3) at the perimeter of the road dust cluster. However, the soil cluster is not entirely separated from the road dust sample cluster. This indicates that road dust is closely correlated with soil, but there are differences due to the amorphous content. Furthermore, this reflects the source variability of the road dust due to soil inputs and traffic related pollutants. Consequently, this further confirms that the amorphous content is the primary reason for the separation of soils from road dust. The analysis outcomes also indicate that soil originating from adjoining areas as a result of erosion or atmospheric processes is the major source of the mineral constituents in road dust. Also, in Figure 3, the amorphous content is negatively correlated with quartz and has no correlation with albite, microcline 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 road dust would be material other than clay or non-crystalline mineral components. This further confirms the contribution of traffic related pollutants to road dust.

3.3 Chemical characterisation of road dust and soil

Analysis of the chemical composition (heavy metals, TOC, pH and EC) of road dust is another approach for source identification of road surface build-up solids. Heavy metals content in road dust primarily indicates the chemical composition of the particles generated via traffic related activities. The heavy metal concentrations were investigated to identify the significance of each of the metal elements in road dust and soils. The average and standard deviation of heavy metal concentration (per unit weight of solids mg g⁻¹) in the soils and road dust samples are given in Table 2.

Insert Table 2

Fe and Al were the most abundant metal elements in solids and soil samples. It is possible that Fe and Al could originate from vehicle component rust, brake lining material, Fe products in brake-pads and vehicle exhaust (Amato et al., 2011; Adachi and Tainosho, 2004; Manno et al., 2006). However, a significant amount of Fe and Al could also originate from soil sources (Pierson and Brachaczek, 1983). Zn was the most abundant metal element after Fe and Al in all of the particle sizes regardless of the study site. As noted in past literature, sources of Zn in road dust are primarily from tire wear (Adachi and Tainosho, 2004). Among the nine metal elements investigated, Cd was the least detected in all of the study sites followed by Cr and Ni. Cd is typically used as plating in brake pads to prevent corrosion (McKenzie et al., 2009). Cr and Ni are generated from tire wear and brake pad wear (McKenzie et al., 2009).

Soil samples indicated significantly high Fe, Al and Mn content compared to road dust and Fe content in soils being ten times higher. Similarly, elevated Pb content was detected in soil samples at all study sites. However, Zn, Cu, Ni, Cr and Cd were significantly low in soil samples compared to the road dust. For example, a negligible content Ni and Cr was present in the soil samples. Similarly, Cd and Cu content in soils were significantly low (10 times)

and were about $<12 \ \mu g \ g^{-1}$ and $0.25 \ \mu g \ g^{-1}$ respectively. These observations confirm that Zn, Cu, Ni, Cr and Cd would have originated from traffic related activities while soil inputs would be the source of Fe, Al and Mn in road dust.

Since it was difficult to obtain a conclusive understanding of the chemical characteristics of road dust using conventional statistical data analysis methods, PCA was performed to identify the potential sources of road dust. A pre-treated (auto scaled) data matrix of 36 x 18 was used for the analysis. The 36 objects represented the thirty road dust samples collected from 16 road surfaces and four soil samples collected from each suburb. The eighteen variables consisted of the nine heavy metal elements, six mineralogical components, together with TOC, pH and EC of the road dust and soil. The resulting PCA biplot is shown in Figure 4.

Insert Fig. 4

As evident in Figure 4, road dust is separated from soil objects indicating two different clusters in the PCA biplot. The road dust forms a cluster characterised by the high Zn, Cu, Ni, Cr and Cd heavy metal elements. Strong correlation of amorphous content with these heavy metals implied that traffic related material would be the main source of amorphous content, as Zn, Cu, Ni, Cr and Cd originate from sources such as tire wear, brake pad wear and asphalt. This confirms that road dust contain a high amount of heavy metals of anthropogenic origin.

These heavy metals would have originated from traffic related particles present in the amorphous content of the build-up solids. Additionally, the soil objects cluster is associated with Fe, Al, Mn, TOC and quartz content. This confirms that Fe, Al and Mn in road dust primarily originate from soil sources, which is typically present in the form of metal oxide associated with minerals. The analysis confirmed that Fe, Al and Mn in road dust are primarily heavy metal elements of geochemical origin. Natural organic matter in surface soil could be the main source of TOC in road dust. This conclusion was derived based on the association of TOC with the soil cluster rather than with road dust objects. Chemical composition of road dust further confirmed that the sources of amorphous content are traffic related.

3.4 Morphology

It is possible to detect the presence of particles derived from traffic related activities or other anthropogenic sources by their detailed morphology and elemental composition (Adachi and Tainosho, 2004). Accordingly, SEM images of build-up solids and soil samples were investigated to distinguish anthropogenic particles. Figure 5a and 5d shows the morphology and elemental composition of road dust and soils, respectively. Traffic related particles are distinguished from mineral components by the presence of C, Al, Si, Ti, S, Ca, Fe and Zn (Camatini et al., 2001). As further confirmation, the morphology and elemental analysis of tire wear particles generated under laboratory conditions were compared with the road dust samples.

Insert Fig. 5

SEM images of road dust indicated distinctly angular well separated particles (Figure 5b). Road dust contain a high amount of elongated particles with rough surfaces resulting from abrasion and were identified as tire wear particles (See Figure 5b). Elemental analysis indicated that these particles contain heavy metal elements of Fe, Cu, Zn, Ni and Pb. Similar morphology was found in tire wear particles generated under laboratory conditions. These characteristics were similar to the morphology reported in past research literature (Adachi and Tainosho, 2004; Kreider et al., 2010). Furthermore, a large amount of spherical particles with smooth surfaces were noted which could be fly ash originating from asphalt and coal combustion (Goodarzi, 2006) (See Figure 5c). As shown in Figure 5c, these particles contain Si, Al, O, Fe, Na, K and Ti. In addition, the samples were predominantly composed of angular particles comprising of Si, Al, Ca and Na as shown by the elemental analysis. These mineral components would primarily originate from surrounding soil.

In contrast, a majority of particles in soil samples had highly weathered and aggregated solids (Figure 5d and e). A small fraction of the soil consisted of organic matter derived from plant debris (See Figure 5f). Al, Fe and Mn were the most common elements identified in soil samples, in addition to Si, O, Ca, K and Mg. The SEM images and elemental analyses results indicated that the road dust contain a high amount of particles embedded with heavy metals, whilst soil particles contain mineral and organic particles originating from plant debris.

4 Conclusions

Reliable identification of the sources of particulate pollutants in build-up on road surfaces and characterisation of particle composition is important for the development of best management practices for stormwater quality mitigation. The research study analysed dust samples from 16 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. The following important conclusions were derived from the study:

- The highest dust load on road surfaces 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 primarily derived from the surrounding vegetation.
- The particulates on road surfaces are predominantly composed of soil derived mineral components. Quartz was the dominant mineral which ranged between 40 and 50% of the sample, irrespective of the geographical location.
- Mineralogical analysis coupled with clay analysis of the road dust and the surrounding soils indicated that vehicular sources contribute about 30% of the non-soil derived pollutants to road dust. Chemical composition of road dust and soils indicated that traffic related material is the main source of amorphous content which consists of a significant amount of heavy metals. Furthermore, it was found that the surrounding soil is the primary source of Fe, Al and Mn in road dust whilst the sources of Zn, Cu, Pb, Ni, Cr and Cd are traffic related.
- SEM images and elemental analysis further indicated that the main constituents present in the amorphous content are derived from traffic related activities including tire abrasion and fly ash from asphalt and combustion by-products.
- The research study confirmed the significant role of traffic related activities as a source of particles of anthropogenic origin to urban road surfaces. These particles

contain heavy metals which may become available for transport with stormwater runoff and eventually impact on receiving water quality.

References

- Adachi, K., Tainosho, Y., 2004. Characterization of heavy metal particles embedded in tire dust. Environ. Internat. 30, 1009–1017.
- Al-Chalabi, A.S., Hawker, D., 1996. Retention and exchange behaviour of vehicular lead in street dusts from major roads. Sci. Total Environ. 187, 105-119.
- Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N., Hopke, P.K., 2009. Quantifying road dust resuspension in urban environment by Multilinear Engine: A comparison with PMF2. Atmos. Environ. 43, 2770-2780.
- Amato F., Pandolfi M., Moreno, T., Furger, M., Pey J., Alastuey, A., Bukowiecki, N., Prevot, A.S.H., Baltensperger, U., Querol X., 2011. Sources and variability of inhalable road dust particles in three European cities. Atmos. Environ. 45, 6777-6787.
- APHA, 2005. Standard Methods for the Examination of Water and Wastewater. Twentieth ed. American Public Health Association, American Water Works Association, Water Environment Federation, Washington.

Beckwith, P.R., Ellis, J.B., Revitt, D.M., 1986. Heavy metal and magnetic relationships for urban source sediments. Phys. Earth Planet. Inter. 42, 67–75.

- Bish, D.L., Post, J.E., 1989. Modern Powder diffraction, Reviews in Mineralogy Volume 20. Chelsea, MI: Geological Society of America.
- Brindley, G.W., Brown, G., 1984. X-ray Diffraction procedures for clay minerals identification: Brindley, G.W. and Brown, G. (Eds.) Crystal Structures of Clay Minerals and Their X-Ray Identification, Mineralogical Society, London.
- Camatini, M., Crosta, G.F., Dolukhanyan, T., Sung, C., Giuliani, G., Corbetta, G.M., Cencetti, S., Regazzoni, C., 2001. Microcharacterization and identification of tire debris in heterogeneous laboratory and environmental specimens. Mater. Charact. 46, 271– 283.
- Dahl, A., Gharibi, A., Swietlicki, E., Gudmundsson, A., Bohgard, M., Ljungman, A., Blomqvist, G., Gustafsson, M., 2006. Traffic generated emissions of ultrafine particles from pavement-tire interface. Atmos. Environ. 40, 1314–23.
- Dawes, L., Goonetilleke, A., Cox, M., 2006. Assessment of physical and chemical attributes of Sub-Tropical soil to predict long term effluent treatment potential. Soil Sediment Contam. 14, 211-229.
- Deletic, A., Ashley, R., Rest, D., 2000. Modelling input of fine granular sediment into drainage systems via gully-pots. Water Res. 34, 3836-3844.
- Deletic, A., Orr, D.W., 2005. Pollution build-up on road surfaces. Journal J. Environ. Eng. 131, 49-59.
- Egodawatta, P., Goonetilleke, A., 2008. Understanding road surface pollutant wash-off and underling physical processes using simulated rainfall. Water Sci. Technol. 57, 1241-1246.
- Egodawatta, P., Thomas, E., Goonetilleke, A., 2007. Mathematical interpretation of pollutant

wash-off from urban road surfaces using simulated rainfall. Water Res. 41, 3025-3031.

- Federal Highway Administration (FHWA), 2005. Surface Texture for Asphalt and Concrete Pavements: Technical Advisory Report (T5040.36). Washington, DC: US Department of Transportation.
- Fergusson, J. E., Kim, N., 1991. Trace elements in street and house dusts: source and speciation. Sci. Total Environ. 100, 125–150.
- Goodarzi, F., 2006. Characteristics and composition of fly ash from Canadian coal-fired power plants. Fuel. 85, 1418–1427.
- Goonetilleke, A., Egodawatta, P., Kitchen, B., 2009. Evaluation of pollutant build-up and wash-off from selected land uses at the Port of Brisbane, Australia. Mar. Poll. Bull. 58, 213-221.
- Gunawardana, C., Goonetilleke, A., Egodawatta, P., Dawes, L., Kokot, S., 2011. Role of solids in heavy metal pollution of receiving waters. J. Environ. Eng., do:10.1061/(ASCE)EE.1943-7870.0000487.
- Herngren, L., Goonetilleke, A., Ayoko, G.A., 2005. Understanding heavy metal and suspended solids relationships in urban stormwater using simulated rainfall. J. Environ. Manage. 76, 149-158.
- Herngren, L., Goonetilleke, A., Ayoko, G.A., 2006. Analysis of heavy metals in roaddeposited sediments. Anal. Chim. Acta. 571, 270-278.
- Khalil, W., 2005. Integrated Land Capability for Ecological Sustainability of On-site Sewage Treatment Systems. PhD Thesis, Queensland University of Technology.
- Kokot, S., Grigg, M., Panayiotou, H., Phuong, T.D., 1998. Data interpretation by some common chemometrics methods. Electroanal. 10, 1-8.
- Kreider, M.L., Panko, J.M., McAtee, B.L., Sweet, L.I., Finley, B.L., 2010. Physical and chemical characterization of tire-related particles: Comparison of particles generated using different methodologies. Sci. Total Environ. 408, 652-659.
- Mahbub, S.M.P., Goonetilleke, A., Egodawatta, P., 2009. Experimental methodology for determining heavy metals and total petroleum hydrocarbons in pollutant build-up and wash-off due to urban traffic and climate change. In Proceedings of the Second Infrastructure Theme Postgraduate Conference (pp.355-363). Brisbane, Australia: Queensland University of Technology.
- Manno, E., Varrica, D., Dongarra, G., 2006. Metal distribution in road dust samples collected in an urban area close to a petrochemical plant at Gela, Sicily. Atmos. Environ. 40, 5929–5941.
- Massart, D.L., Vandeginste, B.G.M., Deming, S.M., Michotte, Y., Kaufman, L., 1988. Chemometrics-A Text Book. Amsterdam: Elsevier.
- McBride, M.B., 1994. Environmental Chemistry of Soil, Oxford University Press, New York.
- McKenzie, E.R., Money, J.E., Green, P.G., Young, T.M., 2009. Metals associated with stormwater-relevant brake and tire samples. Sci. Total Environ. 407, 5855-5860.
- Noble, K.E., 1996. Resource Information. Noble K.E. (Ed.) Understanding and Managing Soils in the Moreton region. Department of Primary Industries Training Series QE96003, Brisbane.

- Pierson, W.R., Brachaczek, W.W., 1983. Particulate matter associated with vehicles on the road, II. Aerosol Sci. Tech. 4, 1-40.
- Rayment, G.E., Higginson, F.R., 1992. Australian Laboratory Handbook of Soil and Water Chemical Methods. Melbourne, Australia: Inkata Press.
- Roger, S., Montrejaud-Vignoles, M., Andral, M.C., Herremans, L., Fortune, J.P., 1998. Mineral, physical and chemical analysis of the solid matter carried by motorway runoff water. Water Res. 32, 1119-1125.
- Rogge, W.F., Hildemann, L.M., Mazurek, M.A., Cass, G.R., 1993. Sources of fine organic aerosol. 3. Road dust, tire debris, and organometallic brake lining dust: Roads as sources and sinks. Environ. Sci. Technol. 27, 1892-1904.
- Sartor, J.D., Boyd, B.G., Agardy, F.J., 1974. Water pollution aspects of street surface contaminants. U.S. Environmental Protection Agency, Washington, DC, USA, Report No.EPA- R2-72/081.
- Settle, S., Goonetilleke, A., Ayoko, G.A., 2007. Determination of surrogate indicators for phosphorus and solids in urban stormwater: Application of multivariate data analysis techniques. Water, Air, Soil Poll. 182, 149-161.
- Thorpe, A., Harrison, R.M., 2008. Sources and properties of non-exhaust particulate matter from road traffic: A review. Sci. Total Environ. 400, 270-282.
- US EPA., 1994. Method 200.8: Trace Elements in Waters and Wastes by Inductively Coupled Plasma- mass Spectrometry. U.S. Environmental Protection Agency, Ohio.
- Xie, S., Dearing, J.A., Bloemendal, J., 2000. The organic matter content of street dust in Liverpool, UK, and its association with dust magnetic properties. Atmos. Environ. 34, 269-275.

$1 \mathbf{\alpha}$	Table	1:	Site	descri	ption	and	road	dust	characteristi	C۶
---------------------	-------	----	------	--------	-------	-----	------	------	---------------	----

Suburb	Land use	Road texture depth range (mm)	Avg. TS load (g/m ²)	Avg. TOC (%)
Clearview Estate	D /	0.76.0.92	1.79	1.54
Clear view Estate	N-4	0.70-0.92	0.81	4.22
Norong	ТЛ	0.02.1.14	3.53	0.76
Ineralig	1-4	0.93-1.14	7.03	0.77
D		0.80.0.01	1.39	1.81
Benowa	I-1, C-1, and K-2	0.80-0.91	0.78	5.12
	0.4	0.72.1.11	2.22	2.73
Surfers Paradise	C-4	0.03-1.11	1.75	0.25

Note: R: Residential; I: Industrial; C: Commercial

The number of study sites is shown along with the land use label

Sample		Al (mg/g)	Cr (µg/g)	Mn (mg/g)	Fe (mg/g)	Ni (µg/g)	Cu (µg/g)	Zn (µg/g)	Cd (µg/g)	Pb (µg/g)
	Clearview Estate	5.88±2.30	14.8±8.70	0.2±0.10	7.22±4.00	7.92±5.00	131.4±26.0	296.6±78.0	0.51±0.10	32.5±12.2
Road	Nerang	2.03±1.60	3.96±0.40	0.09±0.01	4.23±1.20	6.11±1.80	65.5±20.0	176.4±100	0.19±0.10	25.7±11.0
dust	Benowa	3.96±1.50	9.37±2.30	0.15±0.20	5.73±1.90	7.01±1.60	98.4±24.0	236.5±42.0	0.35±0.10	29.1±4.70
	Surfers Paradise	1.40±0.90	3.16±0.40	0.06±0.00	2.98±0.10	4.53±0.70	70.8±20.0	90.4±23.0	0.54±0.10	38.4±9.50
Soils		23.2±1.60	0.00	1.05 ± 0.60	61.2±20.0	0.00	8.09±2.10	352.9±101	0.11±0.08	272.1±98

Table 2: Average heavy metal concentration in road dust and soils in different suburbs (average ± standard deviation)

Figure captions

Fig. 1: Locations of study sites

Fig. 2: Comparison of minerals in road dust and soil samples at the study sites Note: SO - soil, C – Clearview Estate, N- Nerang, B- Benowa and S- Surfers Paradise, 1 and 2 - first and second sampling events, respectively.

Fig. 3: PCA biplot for mineralogical composition of road dust and soil samples Note: SO - soil, Solids - notation indicating the sampling location and sampling event, C -Clearview Estate, N - Nerang, B - Benowa and S - Surfers Paradise, 1 and 2 - first and second sampling events, respectively.

Fig. 4: PCA biplot obtained for heavy metal content in road dust and soil samples Note: SO - soil, Solids - notation indicating the sampling location and sampling event, C -Clearview Estate, N - Nerang, B - Benowa and S - Surfers Paradise, 1 and 2 - first and second sampling events, respectively.

Fig. 5: SEM images and EDX spectrums (at the indicated location) (a) road dust, (b) tire wear particle, (c) fly ash, (d) soil, (e) organic matter and (f) aggregated mineral



Fig. 1



Fig. 2









