Environmental Pollution 216 (2016) 80–85

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Short communication

Sources of hydrocarbons in urban road dust: Identification, quantification and prediction☆

Sandya Mummullage, Prasanna Egodawatta*, Godwin A. Ayoko, Ashantha Goonetilleke

Science and Engineering Faculty, Queensland University of Technology (QUT), G.P.O. Box 2434, Brisbane, QLD 4001, Australia

ARTICLE INFO

Article history:
Received 29 February 2016
Received in revised form
28 April 2016
Accepted 16 May 2016
Available online 26 May 2016

Keywords:
Stormwater quality
Urban road dust
Hydrocarbons
Source identification
Source contributions

ABSTRACT

Among urban stormwater pollutants, hydrocarbons are a significant environmental concern due to their toxicity and relatively stable chemical structure. This study focused on the identification of hydrocarbon contributing sources to urban road dust and approaches for the quantification of pollutant loads to enhance the design of source control measures. The study confirmed the validity of the use of mathematical techniques of principal component analysis (PCA) and hierarchical cluster analysis (HCA) for source identification and principal component analysis/absolute principal component scores (PCA/APCS) receptor model for pollutant load quantification. Study outcomes identified non-combusted lubrication oils, non-combusted diesel fuels and tyre and asphalt wear as the three most critical urban hydrocarbon sources. The site specific variabilities of contributions from sources were replicated using three mathematical models. The models employed predictor variables of daily traffic volume (DTV), road surface texture depth (TD), slope of the road section (SLP), effective population (EPOP) and effective impervious fraction (EIF), which can be considered as the five governing parameters of pollutant generation, deposition and redistribution. Models were developed such that they can be applicable in determining hydrocarbon contributions from urban sites enabling effective design of source control measures.

1. Introduction

Among urban stormwater pollutants, hydrocarbons from anthropogenic sources are a significant environmental concern due to their toxicity and relatively stable chemical structure (Kucklick et al., 1997; US EPA, 1999). These can found in abundance in the urban environment due to high intensity of vehicular traffic and the presence of anthropogenic activities where fossil fuels are heavily used. Many hydrocarbons have strong associations with particulates (Ellis et al., 1997; Lopes and Dionne, 1996; Means et al., 1980), and hence can be found in high concentrations in urban road dust. Dust accumulated over antecedent dry days and consequent washoff during rain events is considered as the most significant pathway for hydrocarbon pollution.

Abatement of hydrocarbon pollutants at their sources is one of the fundamental principles of stormwater pollution control. Success of source control measures depends on the accuracy and reliability of identifying important urban hydrocarbon sources and estimating their relative contributions. Past research studies have used multivariate data analysis techniques to identify the sources of pollutants (Hussain et al., 2015; Valotto et al., 2015) and to determine their contributions to pollutant mixtures (Larsen and Baker, 2003; Liu et al., 2008; Ozaki et al., 2015; Zhang et al., 2008). While these studies are primarily focused on atmospheric pollutants, soils and sediments, none of them have been extended to develop models which can predict source contributions based on site specific characteristics. These limitations can result in inadequate design of stormwater treatment measures and ineffective implementation of pollution source control strategies.

This paper provides a comprehensive methodology for the identification of contributing hydrocarbon sources in the urban environment and assessment of hydrocarbon load generated by each identified source. The paper also presents predictive models that can be used to estimate variations in hydrocarbon loads due to changes in traffic and land use characteristics.

2. Material and methods

2.1. Study sites

Road dust samples were collected from 16 selected road sites

☆ This paper has been recommended for acceptance by Chen Da.

* Corresponding author.
E-mail address: p.egodawatta@qut.edu.au (P. Egodawatta).

http://dx.doi.org/10.1016/j.envpol.2016.05.042
0269-7491/

belonging to four suburbs in the Gold Coast region, South East Queensland, Australia (see Fig. 1). The four suburbs represented different land uses and four road sites were selected from each suburb to represent the traffic characteristics specific to that suburb. The selected 16 road sites also have significant differences in geometric and surface characteristics as evident in Fig. 1. The variations in land use and traffic and geometric and surfaces characteristics ensured variations in source contributions to the pollutant mixture in the road dust.

2.2. Road dust sample collection

Road dust samples from each site were collected using a dry and wet vacuuming system (see Mahbub et al., 2010, for details). The system used consisted of a vacuum cleaner with a water filtration system and a pressure controlled sprayer. The system was first used to collect dust accumulated on a 1.5 m × 2.0 m road surface plot by vacuuming in a dry state. Then the plot surface was moistened using deionized water and wet vacuumed. Dust collection and retention efficiency of the system was tested under controlled field conditions prior to use and was found to be 98%.

Samples were collected in two sampling episodes from each of the 16 sites representing two different antecedent dry periods to allow for possible variation in road dust mixtures due to differences in antecedent dry days. Altogether 32 samples were collected representing antecedent dry days less than 7 days and more than 7 days. Research literature has noted that road dust build-up is rapid within the first 7 days of dry weather after a rain event, while showing a reduced build-up rate afterwards (Egodawatta et al., 2013). As the samples were collected during an antecedent dry period of around 7 days, pollutant transformations such as volatilisation of hydrocarbons was not considered to be a significant issue.

2.3. Laboratory analysis

The road dust samples were analysed for 16 hydrocarbon compounds from C8 to C40. Hydrocarbon compounds were extracted from the road dust using liquid-liquid extraction based on US EPA method 3510C and cleaned using US EPA method 3630C (US EPA, 2008). Testing was undertaken using a HP 6890 gas chromatograph coupled with MS 5973 mass selective detector (Agilent technologies, Australia). The precision of the method (percentage recovery) was determined by spiking a known concentration of calibration standard solution prior to starting the extraction procedure. The recovery was found to be within 85%–115%. Surrogate standards and blanks were used for QA/QC purposes. Replicate samples were analysed to test the repeatability of the method. Detailed information relating to laboratory testing is included in the Supplementary Information.

2.4. Analytical methods for source identification and quantification

Principal component analysis (PCA) and hierarchical cluster analysis (HCA) were used for source identification. PCA is a pattern recognition technique that can be used for distinguishing variables and objects with similarities. HCA is typically used for clustering complex data sets and is considered as a complementary technique to PCA. The data set was subjected to pre-treatment (standardisation and outlier detection) prior to source identification using PCA and HCA. In this regard, Hotelling T^2 test was used for outlier detection. Further details regarding PCA, HCA and Hoteling T^2 test can be found in Brady et al. (2014), Gunawardena et al. (2014), and Mummullage et al. (2016).

Principal component analysis/absolute principal component scores (PCA/APCS) receptor model in conjunction with multi linear regression was used to quantify the contributions from identified sources. The PCA/APCS receptor model is capable of analysing a series of observations simultaneously in order to determine the number of sources, their chemical signature profiles (referred to as source profiles) and their contribution to each observation. The technique primarily estimates the mass of pollutants at a given site to its key sources by solving a mass balance equation. Analytical procedure adopted in relation to PCA/APCS is presented in the Supplementary Information. More details of the model can be found elsewhere (eg. Guo et al., 2004).

3. Results and discussion

3.1. Concentrations of hydrocarbons in road dust

Descriptive statistics of individual hydrocarbon compounds are presented in Table 1, including frequency of detection (FOD). The low molecular weight compounds with carbon numbers ranging from C8 to C16 were found to be less frequently detected across all the suburbs, which is attributed to losses due to volatilisation. As evident in Table 1, the distribution of hydrocarbons among suburbs is random, which highlight the fact that there is substantial variation in hydrocarbons concentrations even within a suburb and is not solely influenced by the predominant land use. A range of site specific factors such as traffic characteristics was hypothesised to play a significant role.

3.2. Identification of hydrocarbon sources

Potential sources of hydrocarbons in road dust were identified using PCA. Initially, four outliers identified using Hotelling T^2 test (Settle et al., 2007) was removed. The resulting dataset in the form of standardised measured concentrations of hydrocarbons (in μg/g) were introduced to PCA which extracted three components with eigenvalues >1 explaining 83% of the total data variance. The resulting Varimax rotated component matrix is shown in Table 2.

Potential sources were assigned based on the featured hydrocarbon elements of each identified component in Table 2. This was done with the support of published research studies outlining the characteristics of hydrocarbon emissions from different urban sources. Component 1 which explained 43% of the total data variance is strongly related to C20 to C32 hydrocarbons. Carlsson et al. (1999) noted that the hydrocarbon range C17 to C40 is primarily derived from non-combusted engine lubrication oil depending on the engine type and lubricant in use. Wang et al. (2003) have presented the hydrocarbons distribution for a range of commercially available products, suggesting that C20–C32 range predominantly belongs to heavy fuel oils. Accordingly, Component 1 was assigned as non-combusted lubrication oil.

Component 2 explained 21% of total data variance and is highly correlated with C12 to C16 and C34 hydrocarbons. Published research studies have reported that C12 to C16 hydrocarbons can be related to diesel fuel (Draper et al., 1996). Carlsson et al. (1999) have also reported that C12–C22 compounds primarily originate from non-combusted diesel fuel. Accordingly, Component 2 was assigned as non-combusted diesel fuel. High molecular weight C34 (tetratriacontane) can typically be a lubricant or asphalt related product. The reason for the association of C34 with Component 2 is not clear.

Component 3 explained 19% of total data variance and was characterised with high loadings of C36 to C40 compounds and moderate loadings of C10 and C12 compounds. Rogge et al. (1993) noted that C35 or greater hydrocarbon compounds are predominantly derived from tyre wear. However, studies by Kennedy and