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## The PM<sub>10</sub> fraction of road dust in the UK and India: Characterization, source profiles and oxidative potential

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### HIGHLIGHTS

- PM<sub>10</sub> fraction of road dust was analysed in Birmingham (UK) and New Delhi (India).
- Crustal matter/suspended dust was found to be the major source in both cities.
- Non-exhaust traffic particles influence composition differently in UK and India.
- Source profiles of the road dust PM<sub>10</sub> fraction were prepared.

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### ABSTRACT

Most studies of road dust composition have sampled a very wide range of particle sizes, but from the perspective of respiratory exposure to resuspended dusts, it is the PM<sub>10</sub> fraction which is of most importance. The PM<sub>10</sub> fraction of road dust samples was collected at two sites in Birmingham, UK (major highway and road tunnel) and one site in New Delhi, India. Dust loadings were found to be much higher for New Delhi compared to Birmingham, while concentrations of several species were much higher in the case of Birmingham. Detailed chemical source profiles were prepared for both cities and previously generated empirical factors for source attribution to brake wear, tyre wear, and crustal dust were successfully applied to the UK sites. However, 100% of the mass for the Indian site could not be accounted for using these factors. This study highlights the need for generation of local empirical estimation factors for non-exhaust vehicle emissions. A limited number of bulk road dust and brake pad samples were also characterized. Oxidative potential (OP) was also determined for a limited number of PM<sub>10</sub> and bulk road dust samples, and Cu was found to be a factor significantly associated with OP in PM<sub>10</sub> and bulk road dust.

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### 1. Introduction

Emissions from road vehicles are of importance for human health as they are emitted in the vicinity of human activity, and street canyon effects can enhance pollutant levels by limiting dispersion (Colville et al., 2001). Particulate matter (PM) emissions from road vehicles include both exhaust and non-exhaust (wear and tear of vehicle parts, resuspension of dust) constituents (Pant and Harrison, 2013). Non-exhaust

emissions are more abundant in the coarse size fraction (particles with aerodynamic diameter of 2.5–10 μm) and can deposit on the roadside and be resuspended subsequently due to vehicle activity. Road dust (RD) consists primarily of coarse-sized PM derived from different sources such as wear of vehicle components (brakes, tyres and clutches), the road surface, engine corrosion, tailpipe emissions, crustal dust and other emission sources. However, similarity in composition between direct emission sources such as brake wear and tyre wear and particles resuspended from the road surface renders it difficult to distinguish the contribution of the different sources (Bukowiecki et al., 2010). RD can be a significant source of trace metals, particle-bound polycyclic aromatic hydrocarbons (PAHs), and other chemical species in the atmosphere (Amato et al., 2009; Duong and Lee, 2011; Kwon and Castaldi, 2012). Several studies on RD have reported elemental and organic marker concentrations for RD in different size fractions (Han et al.,

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2007; Morillo et al., 2007; Agarwal, 2009; Amato et al., 2009; Faiz et al., 2009; Duong and Lee, 2011; Gunawardana et al., 2011; Martuzevicius et al., 2011). Crustal dust is characterized by elements such as Si, Al, Ti, Mn, and Fe while non-exhaust emissions are typically characterized by trace metals such as Cu, Ba, Sb, Sn (brake wear), and Zn (tyre wear) (Wahlin et al., 2006; Gietl et al., 2010; Hulskotte et al., 2014). However, several authors have also used organic markers (e.g. PAHs, n-alkanes, benzothiazoles) as source tracers (Lough et al., 2005; Kwon and Castaldi, 2012). Enrichment of both trace elements and organic species has been reported in RD compared to background soils worldwide (Han et al., 2007; Liu et al., 2007; Agarwal, 2009; Duong and Lee, 2011; Luo et al., 2011; Peltier et al., 2011). Elements such as Pt, Pd, and Rh have also been reported to be present in RD and are attributed to emissions from catalytic converters (Prichard and Fisher, 2012).

It is important to note that the chemical source profile (i.e. percentage of species with respect to total PM mass) for contaminated soil as well as non-exhaust emissions varies geographically dependent upon parameters such as traffic (volume and pattern, fleet characteristics), and road surface type, and the climate and geology of the region (Omstedt et al., 2005; Amato et al., 2011; Pant and Harrison, 2013; Kwak et al., 2013). Most of the research, as well as policy action, in the last few decades has focused largely on exhaust emissions, and with a decline in percentage contribution of vehicle exhaust emissions to ambient PM, the contribution of non-exhaust PM is of increasing importance (Thorpe and Harrison, 2008; Denier van der Gon et al., 2013). Detailed accounts of sources and properties of non-exhaust emissions and ambient measurements of non-exhaust components are provided elsewhere (Luhana et al., 2004; Thorpe and Harrison, 2008; Pant and Harrison, 2013; Amato et al., 2014). Typical methods for characterization of RD and non-exhaust emissions include use of a brake dynamometer (Garg et al., 2000), rolling resistance testing machine (Rogge et al., 1993), rotating drum method (Camatani et al., 2001), sweep/vacuum collection of particles followed by sieving or resuspension (Duong and Lee, 2011; Martuzevicius et al., 2011) and use of a specific RD sampler (Amato et al., 2009). Several researchers have analysed the chemical composition of brake components (Rogge et al., 1993; Garg et al., 2000; Kukutschova et al., 2010, 2011; Hulskotte et al., 2014) and tyres (Camatani et al., 2001; Adachi and Tainosho, 2004; Councell et al., 2004; Milani et al., 2004; Aatmeeyata and Sharma, 2010).

Much of the reported research has been conducted on bulk or very coarse fractions of RD, with a very few studies focused on the characterization and source apportionment of the PM<sub>10</sub> fraction of RD (Wahlin et al., 2006; Amato et al., 2009, 2011; Han et al., 2009). In order to quantify RD as a source of airborne PM, it is important to prepare detailed chemical source profiles. Both India and UK lack detailed chemical source profiles for the RD source. For New Delhi, source profiles for PM<sub>10</sub> RD (paved, unpaved and soil) were published as a part of a larger study but the molecular markers were not analysed in detail (Sethi and Patil, 2008).

The aims of this study were to characterize the chemical composition of the PM<sub>10</sub> fraction RD, to prepare a PM<sub>10</sub> RD source profile and to calculate the contribution of non-exhaust emission sources to samples of RD in Birmingham (UK) and New Delhi (India). In addition, oxidative potential of the RD samples from the UK was assessed. To the best of our knowledge, this is the first study to undertake detailed elemental as well as molecular marker and oxidative characterization of the PM<sub>10</sub> fraction of RD either in India or the UK.

## 2. Methods

### 2.1. Sampling sites

Samples were collected at two high traffic locations in Birmingham: Bristol Road – a heavily trafficked road (hereafter referred to as Site A) and Queensway A38 tunnel – a road traffic tunnel (hereafter referred to as Site B) and one heavily trafficked site in New Delhi: Mathura

Road – NH2 (hereafter referred to as Site C). Site A (UK roadside) is one of the major arterial roads in south-west Birmingham with a dual carriageway with 32,000 vehicles/day and includes both gasoline and diesel vehicles. RD samples were collected on both sides of the road close to the traffic junction. Site B (UK road tunnel) is one of the major road tunnels in Birmingham and runs in the North/South direction. The tunnel is naturally ventilated and typical driving speed through the tunnel is 30 miles/h with an estimated 89,000 vehicles travelling each day. Several studies have been conducted at both these sites in the past (Smith and Harrison, 1996; Birmili et al., 2006). Site C (India roadside) is one of the major roads in Delhi and falls on the National Highway 2. An estimated 174,200 vehicles per day run on this road including intra-state and interstate traffic. Public transport (including buses and auto-rickshaws) runs on compressed natural gas (CNG) while personal vehicles run on both gasoline and diesel. Fig. S1 (SI) provides the modal split at the various sites (Azzi, 2012; Department of Transport, 2014; Pant et al., 2015).

### 2.2. Sampling

A custom-built PM<sub>10</sub> dust sampler (as described in Amato et al., 2009) was used to collect the RD samples. Samples were collected onto 47 mm PTFE and quartz fibre filters to enable analysis of elements and organic species. Both PTFE (n = 10) and quartz filter (n = 10) samples were collected at Sites A and C (UK and India roadside) while only PTFE filter samples (n = 10) were collected at Site B (UK tunnel). All the samples were collected after five days of dry weather in September 2012 (Site B) and June–July 2013 (Sites A and C – UK and India roadside). Individual samples were collected over an area of 1 m<sup>2</sup> for a period of 15 min and include samples from the kerbside as well as middle of the road.

In spring 2014, soil, bulk RD, brake pad, and tyre samples were collected in Birmingham while soil and brake pad samples were collected in India in June 2014. The soil samples were collected from the lawns of University of Birmingham and the CRRI campus remote from traffic in Birmingham and New Delhi respectively while the brake pads were sourced from local garages. Only one brake pad per city was analysed. Care was taken to ensure that the soil samples were collected from an uncontaminated area. An additional set of PM<sub>10</sub> RD samples were also collected at Site A (UK roadside). The soil and RD samples were dried, ground using a pestle and mortar and sieved using a 2 mm sieve; 0.5 g of each sample was then extracted and analysed using the methods detailed in the next section. Brake pad (UK and India) and tyre (UK) samples were frozen using liquid nitrogen (N<sub>2</sub>) and subsequently ground. The samples were passed through a 2 mm sieve and extracted and analysed.

### 2.3. Chemical analysis

Details of the analytical procedures are provided in the SI.

## 3. Results and discussion

### 3.1. Mass loadings of PM<sub>10</sub> RD fraction

Average PM<sub>10</sub> mass loading on the road surface was found to be much higher in New Delhi compared to Birmingham:  $9.34 \pm 5.56$  mg/m<sup>2</sup>,  $12.1 \pm 9.3$  mg/m<sup>2</sup> and  $72.9 \pm 24.3$  mg/m<sup>2</sup> for sites A (UK roadside), B (UK road tunnel), and C (India roadside) respectively. Mass loadings varied between 3.78 and 21.8 mg/m<sup>2</sup>, 3.01 and 36.1 mg/m<sup>2</sup> and 44 and 106 mg/m<sup>2</sup> at Sites A, B, and C respectively. In comparison, an average mass loading of 9 mg/m<sup>2</sup> has been reported for the city centre in Barcelona (Spain) while mass loadings of 2.4–21.6 mg/m<sup>2</sup> have been reported for different site types across Spain (Amato et al., 2009, 2013). Higher mass loading in Delhi can be attributed to several factors including local meteorology (lower annual rainfall resulting in dry conditions),

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