The use of mineral magnetic measurements as a particulate matter (PM) proxy for road deposited sediments (RDS): Marylebone Road, London

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Abstract

Road deposited sediments (RDS) are a recognised pollution problem and a worrying public health concern of many urban environments. Linkages between the magneto characteristics of RDS and their particle size properties have been explored to determine the extent to which magnetic technologies can be utilised as a proxy for proffering insights to address pollution challenges. Samples (n = 60) were collected (May, 2008) along both sides of a busy urban road (Marylebone Road) in central London, UK. Magnetic concentration parameters (χLF, χARM and SIRM) reveal high levels of magnetic material, when compared to previous urban RDS studies. Correlation analysis between the magnetic parameters and textural parameters (χLF, χARM, SIRM and PM1.0, PM2.5, PM10) show significantly strong relationships but, unlike earlier studies, the trends display negative correlations. Despite this kinship not adhering to previously identified trends, this does not mean that mineral magnetic measurements cannot be used as a proxy. Moreover, it simply implies that the nature of any trends needs to be established for specific places before it can be reliably applied as a proxy.

Keywords: environmental magnetism, particle size, street dust, built environment, epidemiology, public health.

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1 Introduction

Road deposited sediments (RDS) (sometimes referred to as street dust) can be toxic [1–3] and contribute to the *particulate matter* (e.g. $PM_{1.0}$, $PM_{2.5}$, PM_{10}) loadings of urban sediments. Given the microscopic characteristics of these grains they are easily re-suspended and can become a significant human exposure source. When particles are absorbed through inhalation it can lead to serious health problems, such as cardiovascular disease and respiratory illness [4–6]. In the UK, for instance, it is estimated that 24,000 deaths occur annually due to poor air quality [7].

From an environmental perspective, RDS also cause urban drainage system issues, where urban runoff transfers the mix of sediments and toxic substances to receiving drainage systems and/or watercourses, causing detrimental effects on water quality and the health of the natural environment [8–10]. Other studies have highlighted linkages between road surface runoff and the deleterious influence of inorganic metal toxicants on benthic community structure and function in receiving water bodies [11, 12]. This is because the composition of RDS comprises a variety of both natural materials (quartz, clay and carbonates) and anthropogenic particles [13, 14]. Typically, anthropogenic sediments are derived from industrial and vehicle-generated sources, causing them to contain both metals (metallic fragments and iron oxides) and organic matter [13, 15].

Previous studies [16–18] have demonstrated the distribution of heavy metals within the built environment, in particular their proximity to roadsides and urban catchments. Heavy metals have been found to be associated with road traffic in urban areas and are known to contain particles associated with vehicle wear (such as tyres, body, brake linings), road surface wear, road paint degradation, vehicle fluids, and particulate emissions [19–22].

There is growing awareness of the issues associated with PM pollution [23], particularly within the built environment arena. As such, programmes of PM monitoring are now commonplace in major towns and cities of many countries. For instance, the UK currently has a network of 64 automatic monitoring sites (using gravimetric analysers of PM concentrations). The suite of sites, forming the 'Automatic Urban and Rural Network' (AURN), operated by the Department for Environment, Food and Rural Affairs (DEFRA), are positioned at strategic urban locations where data is continually measured and monitored (www.airquality.co.uk), recording hourly and daily measurements of PM_{10}, $PM_{2.5}$, Nitrogen oxides, Sulphur dioxide and Ozone, amongst others.

The European Air Quality Framework Directive (96/62/EC) and the First Air Quality Daughter Directive (1999/30/EC) legislation require PM_{10} levels not to exceed 50 μgm^{-3} for more than 35-days per year and set the maximum annual mean limit at 40 μgm^{-3}. That said, Marylebone Road is one of the most widely publicised air pollution sites in the UK, where PM concentrations are known to regularly exceed regulatory standards [24–27]. Therefore, this venue was selected so as to validate the potential of adopting an alternative technology for monitoring the PM sizes of RDS. Previous RDS studies, elsewhere, have already shown kinships exist between particle sizes and heavy metal content [28–31] but the magnetic signature of RDS has only recently been identified to exhibit significant correlations with these characteristics [32, 33]. However, magneto-techniques have not been widely applied to sites where there are known PM issues and, thus, allow new findings, herein, to be compared with other studies.

This work aims to demonstrate the extent to which particular magnetic concentration parameters can be used as a particle size proxy for urban RDS and attempt to highlight whether any data associations follow the predictable trends of similar studies.

2 Case study

Marylebone Road (Fig. 1) is a major arterial route (A501) for traffic (up to seven lanes, including bus lanes) and pedestrians within the city of Westminster (central London) that forms part of the inner London ring-road and marks the northern limit of the London congestion charging zone.

Roadside buildings create an asymmetric street canyon with a height to width ratio of ~0.8 [34]. The road has consistently high daily mean PM_{10} levels that regularly exceed legislative requirements (e.g. 185 incidents 2002–2004 [34]; 47

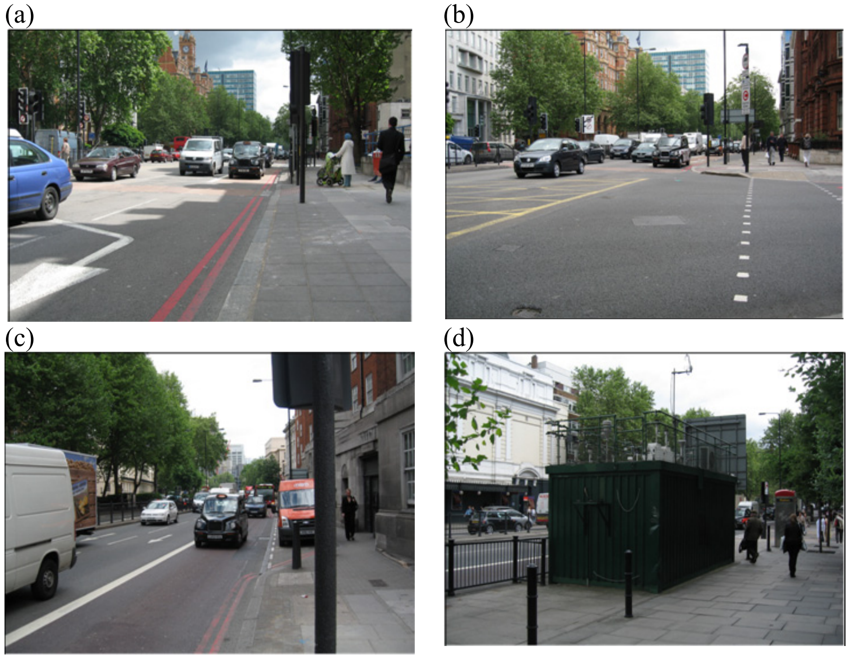

Figure 1: Views of Marylebone Road (May, 2008): (a) Site 9, East facing (Grid reference: 527833 181940); (b) Site 14, East Facing (Grid reference: 527963 181995); (c) Site 29, West facing (Grid reference: 528783 182200); and (d) Site 61, East facing (Grid reference: 526243 180535).

incidents in 2007; 29 incidents in 2008; and 36 incidents in 2009). These exceedences are thought to be due to congestion and high traffic flows [35], with over 80,000 vehicles per day using the road [34]. This has led to a number of independent studies on PM [25, 34, 36–39], which reveal increases in iron rich dusts [39] and a greater frequency of PM_{10} exceedences during weekdays when traffic conditions peak [34].

3 Materials and methods

3.1 Sample collection and preparation

Street dust was collected from the pavements (sidewalks) at regular spacings along both sides of the road. Typically, 10–50 g dust samples were collected (from ~1 m^2) by brushing with a small hand-held fine-bristle brush. Dust was then transferred to clean, pre-labelled, self-seal, airtight plastic bags. In the laboratory, samples were visibly screened to remove macroscopic traces of hair, animal and plant matter [21].

3.2 Mineral magnetic measurements

All samples were subjected to the same preparation and analysis procedure. Samples were dried at room temperature (<40 °C), weighed, packed into 10 ml plastic pots and immobilized with clean sponge foam and tape prior to analysis. Initial, low–field, mass–specific, magnetic susceptibility (χ) was measured using a Bartington (Oxford, England) MS2 susceptibility meter. By using a MS2B sensor, low frequency susceptibility was measured (χ_{LF}). Anhysteretic Remanence Magnetisation (ARM) was induced with a peak alternating field of 100 mT and small steady biasing field of 0.04 mT using a Molspin (Newcastle–upon–Tyne, England) A.F. demagnetiser. The resultant remanence created within the samples was measured using a Molspin 1A magnetometer and the values converted to give the mass specific susceptibility of ARM (χ_{ARM}). The samples were then demagnetized to remove the induced ARM and exposed to a series of successively larger field sizes up to a maximum ‘saturation’ field of 1000 mT, followed by a series of successively larger fields in the opposite direction (backfields), generated by two Molspin pulse magnetisers (0-100 and 0-1000 mT). After each ‘forward’ and ‘reverse’ field, sample isothermal remanent magnetisation (IRM) was measured using the magnetometer [40].

3.3 Laser diffraction measurements

All samples were subjected to the same textural preparation and analysis procedure, using sieving (2000 μm aperture) followed by laser diffraction analysis. Low Angle Laser Light Scattering (LALLS), using a Malvern (Malvern, England) Mastersizer Long-bed X with a MSX17 sample presentation unit, enabled rapid measurement of particle sizes within the 0.1-2000 μm range. Macroscopic traces of organic matter were removed from representative subsamples before being dampened by the dropwise addition of a standard chemical solution (40 g/l solution of sodium hexametaphosphate ($(NaPO_3)_6$) in distilled water) to help disperse aggregates. To ensure complete disaggregation, each slurry was then subjected to ultrasonic dispersion in a Malvern MSX17 sample presentation unit. For greater precision, the mean of five replicate analyses was measured with a mixed refractive indices presentation setting. A standard range of textural parameters was calculated, including the percentage of sand, silt and clay class sizes and their sub-intervals. The Malvern instrumentation was regularly calibrated using latex beads of known size [40].

4 Results

Particle size data (Table 1) indicates samples are dominated by sand (~79%), silt (~19%) and clay (~2%), in respective orders. From a respiratory-health perspective, PM_{10} grains represent ~7%, $PM_{2.5}$ ~3% and $PM_{1.0}$ ~1% of the sediments at pavement level. This is noteworthy because, once suspended, particles <10 μm in diameter are able to remain airborne for hours or days and, in some cases, even weeks [41]. Therefore, the presence of PM of these sizes on pavement surfaces indicates either the sediments have not been disturbed recently or they have only just settled-out. Since the pavements normally receive frequent and heavy foot-traffic, it is assumed the time of sampling (0500 – 0900) and weather conditions (warm, dry and still) have permitted sizeable PM accumulations.

Mineral magnetic characteristics have been summarised (Table 2). χ_{LF} is roughly proportional to the concentration of ferrimagnetic minerals within the sample, although in materials with little or no ferrimagnetic component and a relatively large antiferromagnetic component, the latter may dominate the signal. χ_{ARM} is particularly sensitive to the concentration of magnetic grains of stable

Table 1: Summary particle size properties of the RDS: (a) traditional sediment size fractions and (b) respiratory health-related size fractions (n = 60 samples).

(a)	Mean (%)	Minimum (%)	Maximum (%)	Standard Deviation
Sand (63-2000 μm)	78.88	45.92	91.45	7.98
Silt (2-63 μm)	19.08	7.27	53.01	7.85
Clay (<2 μm)	2.04	0.90	8.50	1.31

(b)	Mean (%)	Minimum (%)	Maximum (%)	Standard Deviation
$<PM_{10}$	6.57	2.84	23.99	3.88
$<PM_{2.5}$	2.64	1.25	9.82	1.69
$<PM_{1.0}$	1.37	0.66	5.39	1.02

single domain size, e.g. ~0.03–0.06 μm. SIRM is related to concentrations of all remanence-carrying minerals in the sample, but is also dependent upon the assemblage of mineral types and their magnetic grain size. These data indicate the samples contain moderate to high magnetic concentrations. Compared to previous urban magneto-dust studies, the mean values are sizeably greater than those of Liverpool (23.7 x10-7 m^3 kg^{-1}) [42] and Shanghai (29.9 x10-7 m^3 kg^{-1}) [43].

Spearman's rank correlation coefficient values (r_s) between the mineral magnetic concentration parameters and particle size parameters have been grouped according to traditional sediment size fractions (Table 3) and respiratory health-related size fractions (Table 4). Significant relationships ($p < 0.001$; n = 50) exist between clay content and all of the magnetic concentration parameters,

Table 2: Summary mineral magnetic properties of the RDS (n = 60 samples).

	Units	**Mean**	**Minimum**	**Maximum**	**Standard Deviation**
χ_{LF}	$10^{-7}m^3kg^{-1}$	47.88	11.13	87.97	16.12
χ_{ARM}	$10^{-7}m^3kg^{-1}$	2.18	0.04	11.37	1.53
SIRM	$10^{-5}Am^2kg^{-1}$	3779.00	956.00	6161.00	119.80

Table 3: Spearman's rank correlation coefficients (rs) between mineral magnetic concentration and particle size parameters for the RDS based on traditional sediment size fractions.

(a)	**Clay <2 μm**	**Silt 2-63 μm**	**Sand 63-2000 μm**
χ_{LF}	-0.453***	0.010	0.076
χ_{ARM}	-0.384**	0.109	-0.017
SIRM	-0.386**	-0.049	0.135

Note: Significance levels: p <0.05 = *; p <0.01 = **; p <0.001 = ***.

Table 4: Spearman's rank correlation coefficients (rs) between mineral magnetic concentration and particle size parameters for the RDS based on respiratory health-related size fractions (n = 60 samples).

(b)	**$<PM_{1.0}$**	**$<PM_{2.5}$**	**$<PM_{10}$**
χ_{LF}	-0.589***	-0.575**	-0.526**
χ_{ARM}	-0.511**	-0.471**	-0.402**
SIRM	-0.554**	-0.553**	-0.519**

Note: Significance levels: p <0.05 = *; p <0.01 = **; p <0.001 = ***.

which is similar for each of the PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ sizes. Therefore, this indicates all the magnetic concentration parameters could potentially be used as a particle size proxy, particularly if the kinship is required with particles < PM_{10}.

5 Discussion

Earlier sedimentological works have noted significant correlations exist between magnetic and particle size properties. Oldfield *et al.* [44] identified anhysteretic remanent magnetisation (ARM) measurements reflect the concentration of fine-grained magnetite (<0.1 μm) in clay fractions and χ_{LF} measurements can be used to infer the presence of coarser multi-domain magnetite (>1.0 μm) in sands and coarse silts. Clifton *et al.* [45] found χ_{LF} was strongly associated with sands and medium silts, χ_{ARM} was strongly associated with clay and fine silts, and SIRM was strongly associated with very fine to medium silts. Zhang *et al.* [46] suggested that both percentage frequency-dependent magnetic susceptibility ($\chi_{FD\%}$) and χ_{ARM} can be used as a proxy for clay content.

These studies have illustrated sand to correlate negatively with χ_{LF} ($r = -0.94$), χ_{ARM} ($r = -0.96$) and SIRM ($r = -0.91$); silt to correlate positively with χ_{LF} ($r = 0.96$), χ_{ARM} ($r = 0.96$) and SIRM ($r = 0.96$); and clay to correlate positively with χ_{LF} ($r = 0.82$), χ_{ARM} ($r = 0.94$) and SIRM ($r = 0.81$). When data presented here are compared to earlier investigations, it is apparent that each magnetic parameter also correlates with particle size but the significance is only notable with clay fraction and not the sand and silt fractions. Nevertheless, on first observation, this highlights the potential use of mineral magnetic data as a means of normalizing compositional analytical data (i.e. geochemical) for particle size. However, an important disparity is that the trends observed in this work show negative relationships with the clay fraction, while previous works have always revealed positive trends with the clay fraction. This highlights a sizeable issue for the universal application of mineral magnetic measurements as a particle size proxy.

Previous RDS works have also noted significant correlations exist between χ_{LF}, χ_{ARM}, SIRM and respiratory health-related particle size fractions. Booth *et al.* [32] revealed PM_{10} to correlate with χ_{LF} ($r = 0.69$; $p < 0.001$; $n = 50$); $PM_{2.5}$ to correlate with χ_{LF} ($r = 0.71$; $p < 0.001$; $n = 50$), χ_{ARM} ($r = 0.30$; $p < 0.05$; $n = 50$) and SIRM ($r = 0.33$; $p < 0.05$; $n = 50$); and $PM_{1.0}$ to correlate with χ_{LF} ($r = 0.66$; $p < 0.001$; $n = 50$), χ_{ARM} ($r = 0.41$; $p < 0.01$; $n = 50$) and SIRM ($r = 0.32$; $p < 0.05$; $n = 50$). Similarly, Crosby *et al.* [33] revealed PM_{10} to correlate with χ_{ARM} ($r = 0.44$; $p < 0.01$; $n = 35$) and SIRM ($r = 0.43$; $p < 0.01$; $n = 35$); $PM_{2.5}$ to correlate with χ_{ARM} ($r = 0.45$; $p < 0.01$; $n = 35$) and SIRM ($r = 0.43$; $p < 0.01$; $n = 35$); and $PM_{1.0}$ to correlate with χ_{ARM} ($r = 0.42$; $p < 0.01$; $n = 35$) and SIRM ($r = 0.40$; $p < 0.05$; $n = 35$).

Marylebone Road displays similar correlation significance levels to those of the towns of both Southport [32] and Scunthorpe [33]. However, a notable discrepancy in the correlations is that Marylebone Road again displays significant negative trends; whereas, both of the other places have significant positive trends. As such, this represents a potential flaw in the use of mineral magnetic measurements as a universal proxy. However, despite this apparent setback, it does not mean that mineral magnetic measurements cannot be used as a proxy. Moreover, it simply implies that the nature of any trends needs to be established for specific places before it can be reliably applied as a proxy.

These differences offer an opportunity to provide speculative reasoning to explain the outcomes; whereby, it is postulated that the differences are due to the RDS being derived from different sources that have varying characteristics and/or are derived from several mixed sources. To support this argument, attention is drawn to the similarities and disparity of the venues already mentioned. For instance, given the size and restrictive (street canyon) nature of Marylebone Road it is proposed that the RDS are chiefly derived from a singular source (i.e. vehicular); whereas, Southport is a seaside resort with no noteworthy industry so it is proposed that the RDS are probably derived from more than one main source (i.e. a mix of wind-blown coastal sediments and vehicular derived dusts); likewise, Scunthorpe is celebrated as an iron and steel town so it is proposed that the RDS are also probably derived from more than one main source (i.e. a mix of industrial emissions and vehicular derived dusts). Verification of this reasoning would require detailed investigation, such as SEM analyses and/or complex sediment source modelling. However, as with most investigations, these findings promote the need for further research on the reliability of using magnetic technologies as a pollution proxy but, likewise, it also offers a provoking avenue to expand the work to provenance studies.

6 Future work

This work forms part of a wider investigation that includes other places in the UK (Dumfries, Norwich, Oswestry, Runcorn, Salford, Scunthorpe and Wolverhampton), which is attempting to address the same aims as those posed in this particular work. It is anticipated that it will offer better insights into the reliability of using mineral magnetic measurements as a particulate matter proxy.

7 Conclusions

As with previous studies, this work indicates each of the magnetic concentration parameters could be reliably employed as a particle size proxy for urban RDS, where the finest fraction (<10μm) is the focus. However, the trends displayed in this work are negative correlations and, since this is unlike other studies, it indicates that the perceived universal relationship does not always exist like previously proposed. Despite this potential discrepancy in its suitability, it does not mean that mineral magnetic measurements cannot be used as a proxy. Moreover, it simply implies that the nature of any trends needs to be established for specific places before it can be reliably applied as a proxy.

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