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Rates of particulate pollution deposition onto leaf surfaces: Temporal and inter-species magnetic analyses

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This research uses biomagnetic techniques to enable quantitative mapping of particulate pollution distribution at uniquely high spatial resolution.

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A B S T R A C T

Evaluation of health impacts arising from inhalation of pollutant particles <10 μm (PM_{10}) is an active research area. However, lack of exposure data at high spatial resolution impedes identification of causal associations between exposure and illness. Biomagnetic monitoring of PM_{10} deposited on tree leaves may provide a means of obtaining exposure data at high spatial resolution. To calculate ambient PM_{10} concentrations from leaf magnetic values, the relationship between the magnetic signal and total PM_{10} mass must be quantified, and the exposure time (via magnetic deposition velocity (MV_d) calculations) known. Birches display higher MV_d (~ 5 cm^{-1}) than lime trees (~ 2 cm^{-1}). Leaf saturation remanence values reached 'equilibrium' with ambient PM_{10} concentrations after ~6 'dry' days (<3 mm/day rainfall). Other co-located species displayed within-species consistency in MV_d; robust inter-calibration can thus be achieved, enabling magnetic PM_{10} biomonitoring at unprecedented spatial resolution.

1. Introduction

A growing body of literature (e.g. Curtis et al., 2006; Lipmann, 2007; Schwarze et al., 2006; Zeger et al., 2008) documents the adverse health effects of exposure to fine-grained pollutant particles, i.e. those with aerodynamic diameters below 10 μm in diameter (PM_{10}), and particularly those below 2.5 μm ($PM_{2.5}$). However, the pollution exposure data relied upon by many epidemiological studies are often sourced from low spatial-resolution networks of monitoring stations, which are unlikely to capture possibly fine scale variations in PM concentration and/or particle size across the diverse urban environment. For example, in large population studies (e.g. Dominici et al., 2006; Karr et al., 2006; Woodruff et al., 2006; Zeger et al., 2008), PM_{10} exposure has frequently been characterised as the average PM_{10} value at the 'nearest' monitoring station (i.e. within 5 miles of residence). Such coarse-scale data reduce the potential for identifying and quantifying specific causal links between the degree of exposure to PM_{10} and the likelihood of adverse health impacts within a population.

An additional problem with data from conventional monitoring stations is the height of their air inlets, often situated in excess of 3 m. Traffic-derived PM_{10} values decrease not only with increased distance from roads, but also with increased height (e.g. Maher et al., 2008; Mitchell and Maher, 2009). Thus, PM_{10} data from the conventional pollution monitoring networks may be a weak indicator of individual human exposure.

A number of studies have used the magnetic properties of deposited particles as a proxy for particulate pollution levels (e.g.Hanesch et al., 2007; Maher et al., 2008; Matzka and Maher, 1999; Szönyi et al., 2008). Magnetic techniques, using natural surfaces as passive collectors of particulate pollution, are sensitive, rapid, and relatively cheap; and passive collectors require no power source or protection from vandalism. Strong correlation has been demonstrated between leaf saturation remanent magnetisation (SIRM) and/or magnetic susceptibility values and the presence of pollution particles, produced by combustion and/or abrasion processes (e.g. Gautam et al., 2004; Halsall et al., 2008; Maher et al., 2008), and toxic metals, such as lead and iron (e.g. Maher et al., 2008). Strong correlation between roadside leaf SIRMs and ambient PM_{10} concentrations (from co-located pumped-air samples) suggests that magnetic biomonitoring, using tree leaves as passive pollution collectors, can be a robust technique for quantifying

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ambient concentrations of PM_{10} levels at unprecedentedly high spatial resolution and at pedestrian-relevant heights (Mitchell and Maher, 2009). So far, however, the rate and temporal variability of magnetic particle deposition on tree leaves, and on different species of trees, have yet to be examined.

Deposition to vegetation surfaces is also significant in its own right. It is a sink for atmospheric particles, and a route by which pollutants and particulate nutrients (such as ammonium nitrate) can enter the biosphere. Dry deposition of pollution particles (Pryor et al., 2007) is considered more important than wet deposition, particularly near to pollution sources (e.g. Businger, 1986; Pryor et al., 2007). Some data exist for particulate dry deposition rates to foliage both under controlled, laboratory and wind-tunnel conditions (e.g. Caffrey et al., 1998; Dai et al., 2001; Parker and Kinnersley, 2004), and field conditions (e.g. Petroff et al., 2008; Pryor et al., 2008). However, the range of published deposition values is large, reflecting both the differing methods of measurement and the complex dependence of deposition velocities (V_d) on variables such as particle size and density, terrain, vegetation, meteorological conditions, and chemical species (e.g. Zhang et al., 2001). Measurements under natural conditions are particularly difficult to obtain (Kinnersley et al., 1994). Artificial sample collectors currently available are unable to reproduce accurately the effects of real receptor surface morphology, canopy structure and texture (Wesely and Hicks, 2000), and therefore may provide unrealistic deposition estimates.

Here, we report a magnetic biomonitoring study which has measured deposition velocities to natural surfaces under field conditions. We examined glasshouse-grown 'clean' leaves and co-located pumped-air samples at a rural 'background' site, in order to assess the rate of PM_{10} deposition on tree leaves, as measured via leaf magnetic properties (saturation remanence). By sequential sampling, we identified the time period required for particle deposition to reach dynamic equilibrium, and thus the time over which the leaf magnetic record of PM_{10} deposition is effectively integrated. This time period has significance for application of magnetic biomonitoring as a proxy for ambient PM_{10} concentrations and sampling protocols, as it identifies the minimum required leaf exposure time, and/or the minimum time period required for re-equilibration after significant rainfall, for the leaves to provide a robust proxy of ambient PM_{10}. We repeated the experiment at a roadside site, with increased traffic-derived PM_{10} levels. Finally, we used the biomagnetic data to evaluate differences in the capacity of various deciduous tree species for PM_{10} collection. We show that biomagnetic monitoring, using a range of different tree species, can be used to identify ambient PM_{10} concentrations from magnetic measurements of particles accumulated on the leaf surface over $\sim$ 6 days, enabling collection of data at high spatial resolution with direct relevance to human exposure and health impacts.

2. Methods

To obtain magnetically-'clean' trees, lime (Tilia platyphyllos) and birch (Betula pendula) trees were grown from seed under glasshouse conditions. Leaf SIRMs were measured for the trees prior to exposure. Only trees taller than 30 cm and with a canopy comprising $>\sim$40 leaves were used for analysis of PM_{10} deposition over the experimental periods of 12 and 24 days. A pilot study was carried out between 27/04/2008–08/05/2008 at Site 1, Hazelrigg weather station (Meteorological Office Climatological Station Number 7236), a site with low PM_{10} concentrations ($<$12 $\mu g/m^3$), located on top of a small hill $\sim$ 1 km northeast of Lancaster University, UK. 'Clean' samples consisting of 6 leaves were collected (two leaves from three each of the lime and birch trees, to avoid over-sampling from any one young tree). The trees were then planted at the 'background' field site (site 1), proximal to an Andersen Hi-Vol air sampler (inlet height $\sim$ 1 m), and leaf samples collected daily from $\sim$0.3 m height, the height found to be associated with maximum PM_{10} deposition from traffic sources (Maher et al., 2008; Mitchell and Maher, 2009). Pumped-air samples (1636 m^3) were collected daily at a rate of 1133 l/min from the co-located Hi-Vol air sampler, fitted with magnetically clean (SIRM = 1×10^{-7} Amps (A)) PTFE filters (1 μm pore size). The conventional method for calculating deposition velocities (V_d) is to divide the particle flux to the sampling surface over a known period of time (mass/m^2/s) by the ambient concentration (mass/m^3). Magnetic deposition velocities were calculated following the conventional approach, but substituting SIRM for mass;

$$MV_d = F/C \tag{1}$$

where F = $SIRM_{leaf}/m^2$ s and C= $SIRM_{filter}/m^3$,

A second sampling campaign was carried out between 10/05/08–03/06/08 at the main access road to Lancaster University (site 2), a site with enhanced traffic-derived particulate pollution levels. Again, prior to exposure, 'clean' leaf samples were taken from six trees of each species; these trees were then placed at the roadside. Half of the trees were planted close to the road, on the uphill side of the roundabout approach to maximise exposure to vehicle-sourced pollution (Matzka and Maher, 1999; Mitchell and Maher, 2009). Samples were collected 48-hourly from $\sim$0.3 m height. The remaining trees were not planted but placed at the roadside only during the morning and evening peak traffic flow periods (08:15–10:15, 15:30–17:30). At all other times, they were stored in the glasshouse. Leaf samples were collected concurrently with the planted trees (i.e. after 4 $\times$ 2 h exposures) from $\sim$0.3 m height. Pumped-air samples (240 L) were collected, at a rate of 2 l/min, from co-located SKC Leland Legacy personal monitors with magnetically clean (SIRM = 1×10^{-7} A) PTFE filters (1 μm pore size) in IoM-type PM_{10}-selective sampler heads, during each peak traffic flow period. After exposure, all filters were immediately weighed then placed in magnetically clean (SIRM = 0.05×10^{-6} A/m) 10 cc plastic pots and taken to the laboratory for magnetic analysis.

Correlations were calculated between leaf magnetic values and the independently measured ambient PM_{10} concentrations. In order to identify the time period required for particulate accumulations on leaves to attain dynamic equilibrium with ambient concentrations, stepwise exclusion correlation calculations were used. Initially, all samples were included in the correlation calculation. Samples were then removed one at a time, in time-series order, and the correlation (R^2) and significance re-calculated. The optimum solution is the one in which the correlation and the sample number included in the calculation are maximised.

To examine the variability of leaf SIRMS between different tree species, leaves were sampled from 36 sites around an elevated stack point source, Oxfordshire, at monthly sampling intervals from May to September, 2008. Samples were collected from 1.5 to 2 m height at any location where two or more of the following tree species were co-located within a 2 m radius: birch (Betula pendula), beech (Fagus sylvatica), lime (Tilia platyphyllos), field maple (Acer campestre), ash (Fraxinus excelsior), sycamore (Acer pseudoplatanus), elder (Sambucus nigra), elm (Ulmus procera), willow (Salix alba) and oak (Quercus robur).

All leaf samples were refrigerated at 5 °C before being returned to the Centre for Environmental Magnetism and Palaeomagnetism (CEMP) at Lancaster University for magnetic analysis using the protocol given in the Appendix and outlined in Mitchell and Maher (2009).

3. Results

3.1. Leaf SIRM and air filter time series, sites 1 and 2

Fig. 1 shows the SIRMs of the pumped-air filters and ambient PM_{10} concentrations from sites 1 ('background') and 2 (roadside), together with roadside filter data from previously sampled sites around the city of Lancaster (Mitchell and Maher, 2009). As the site 1 and 2 filters, and the published Lancaster roadside data, all display similar correlation with ambient PM_{10} concentrations, the three datasets may be treated as one statistical population; strong correlation ($R^2 = 0.90$, $n = 54$, $p = 0.01$) is observed between the filter SIRMs and ambient PM_{10} concentrations in the combined dataset. The slight deviation from the straight line around the lowest concentration points (from Mitchell and Maher, 2009) may reflect the relatively short pumping time (2 h) resulting in very low mass values compared to the 24 h pumping time at site 1, relative to the sensitivity of the mass balance (0.1 mg).

Site 1 ('background') consistently exhibits low PM_{10} concentrations, with a mean ambient PM_{10} concentration of 5.6 ± 2.9 $\mu g/m^3$. Prior to exposure, the measured leaf SIRM values at site 1 are minimal ($<10 \times 10^{-7}$ A). Over the subsequent week's exposure, the SIRMs show an overall upward trend, although rainfall events ($>$3 mm/day) produce reductions both in the leaf and air filter SIRMs (Fig. 2).

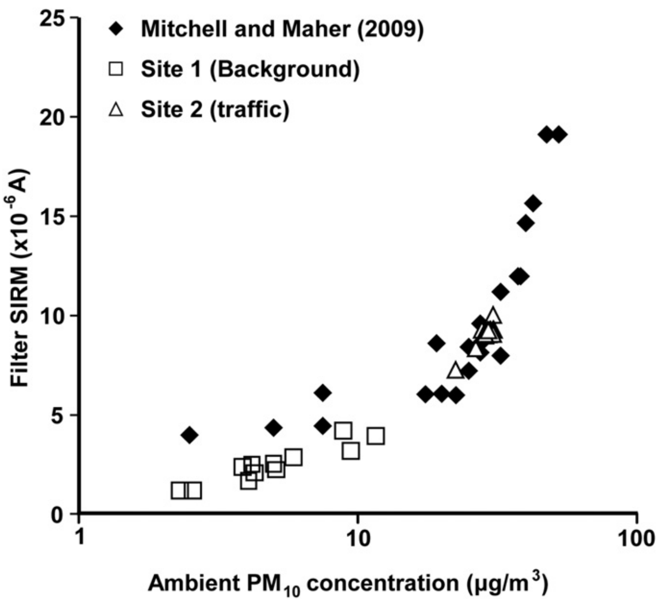


Fig. 1. Correlation ($R^2 = 0.90$, $n = 54$, $\sigma = 0.01$) between filter SIRM and ambient PM_{10} concentration, sites 1 and 2 (this study) and Lancaster roadside data (Mitchell and Maher, 2009).

Leaf sample SIRM values from the roadside, site 2, are presented in Fig. 3; values from both permanently and temporarily exposed lime and birch tree samples are shown. Again, leaf SIRM values – initially minimal – rapidly increase upon exposure. They exceed the site 1 'background' values after 48 h' exposure, and reach maximum values, after $\sim$ 14 days, which are 2 $\times$ higher than 'background'. SIRMs also again decrease after rainfall events

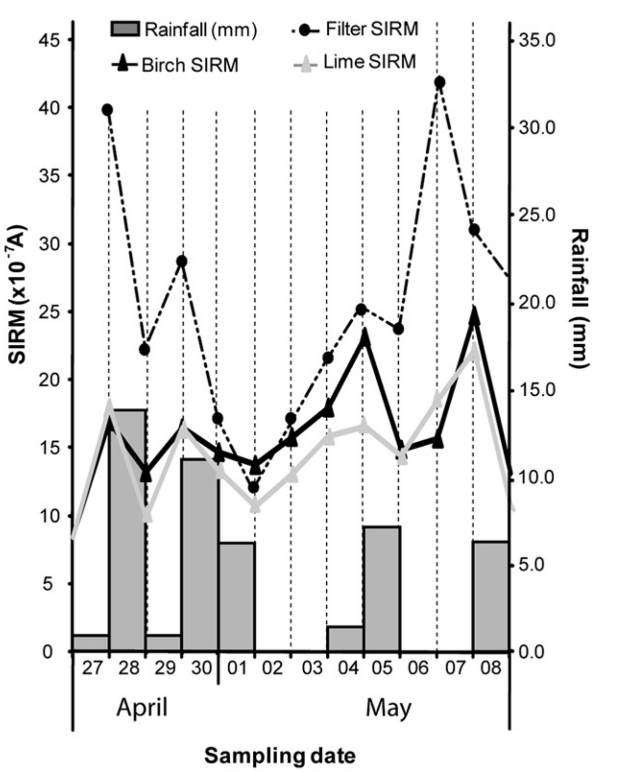


Fig. 2. Daily rainfall data and leaf area-normalized SIRM values of pumped-air filters and birch and lime tree leaves sampled at site 1 from 27/04/08–08/05/08.

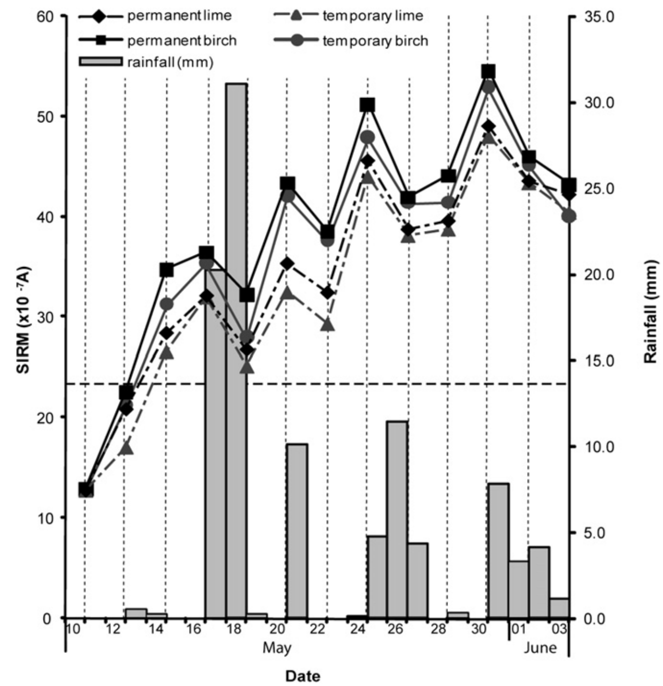


Fig. 3. Daily rainfall data and leaf area-normalized SIRM values of pumped-air filters and birch and lime tree leaves sampled at site 2 from 10/05/08–03/06/08. 'Temporary' trees were exposed only for the two peak traffic flow periods each day (08:15–10:15, 15:30–17:30).

exceeding 2 mm in a 24 h period. Notably, for both species, the trees with temporary, peak traffic exposure exhibit similar SIRM values to those which were exposed permanently throughout the measurement period.

Initially, both lime and birch leaf SIRMs increase independently of ambient PM_{10} concentrations. After $\sim$ 6 days, the magnetic values fluctuate in tandem with ambient PM_{10} concentrations (Fig. 4).

Stepwise–exclusion correlation calculations between leaf magnetic values and ambient PM_{10} concentrations over time indicate a substantial increase in R^2 values when samples 1 and 2 (i.e. representing the first two days of traffic exposure, 12–14th May) are excluded ($R^2 = 0.8–0.9$, $n = 10$, $p = 0.05$). The strongest correlation between the leaf SIRMs and the ambient PM_{10}

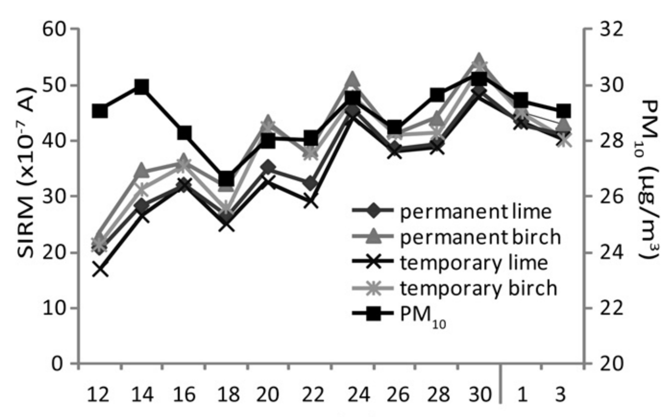


Fig. 4. Ambient PM_{10} concentrations (from co-located pumped-air samples) and SIRM values of birch and lime tree leaves sampled at site 2 from 10/05/08–03/06/08.

concentrations occurs after 6 days. As further samples are removed from the calculation, both the correlation and the significance decrease (until only two samples remain, giving a statistically meaningless correlation of R^2 equal to 1) (Fig. 5).

3.2. Between-species magnetic biomonitoring

Monthly samples taken from different, co-located tree species at 36 sites around a stack point source of pollution (Oxfordshire, UK) indicate systematic, between-species variation in magnetic particle deposition velocity to the leaf surface. Strong correlation is observed between leaf SIRMs from sycamore (the most ubiquitous tree species occurring within the sample area) and from the other species sampled (Fig. 6; Table 1), enabling classification of the majority of the tree species into three classes according to their relative magnetic particle-capturing ability. Lime and beech remove considerably more particles from the air than sycamore, while sweet chestnut, willow, elder and elm capture substantially fewer. The only exception to this marked between-species consistency is the oak, which shows very little correlation between its leaf SIRMs and those of any of the other species measured here.

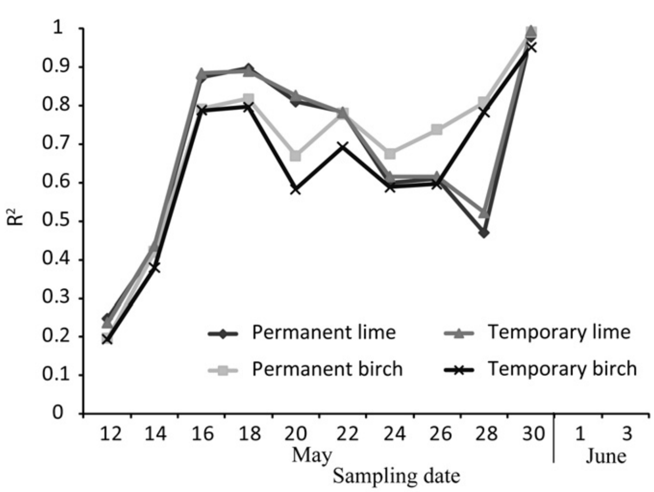
4. Discussion

At Site 1, the rural background site, the magnetic particle deposition velocity (MV_d) for both lime and birch tree leaves was $\sim$2 cm^{-1}, i.e. within an order of magnitude of published particle V_d values for deciduous trees (e.g. Freer-Smith et al., 2005; Pryor et al., 2007). After $\sim$ 6 days, the change in-leaf SIRMs tracked changes in measured ambient PM_{10} levels until heavy rainfall ($\sim$7 mm) occurred on 05/05/2008. Subsequent episodes of rainfall were consistently seen to give rise to similar reductions at both sites 1 and 2. The observed reduction of leaf SIRMs under this amount of rainfall is consistent with the 'wash-off' of particles exceeding their rate of wet deposition, as previously reported (e.g. Kinnersley and Scott, 2001). In contrast, for an evergreen species (holly oak), Szönyi et al. (2008) report no clear pattern of rainfall-induced reduction in-leaf magnetic (susceptibility) values. However this may be attributable to intra-species differences in-leaf structure, as discussed below. Site 2 (the roadside site) exhibited similar deposition velocities ($\sim$2 cm^{-1}) for the lime trees, but a slightly higher rate of deposition for the birch trees ($\sim$5 cm^{-1}), possibly reflecting leaf ageing (e.g. increasingly ridged) through the early part of the growing season. The between-species difference in MVd may reflect differences in the leaf surface morphology; the presence of hairs on the upper birch leaf surface may aid particle retention. After the initial period of exposure and PM_{10} accumulation ($\sim$6 days), leaf SIRMs closely track the trends in measured PM_{10} concentrations. The strong correspondence between leaf SIRM values for the trees exposed only to temporary, peak traffic conditions and those exposed permanently (Fig. 3) demonstrates that most leaf particle deposition occurs during the peak traffic periods, with minimal deposition during periods of low traffic flow. The implication of this is that the integrated MV_d signal is reflecting the peak concentration that the leaves regularly experienced.

Examination of the relationship between leaf SIRMs and measured ambient PM_{10} concentrations shows that strong correlation exists between them ($R^2 = 0.8$–0.9, $n = 10$, $p = 0.01$) once the initial period of leaf SIRM 'build-up' has been completed (i.e. after $\sim$ 6 days). It seems likely that after this period of net accumulation of particles on the initially clean leaf surface, particle deposition on the leaf surface reaches a dynamic equilibrium between the rates of particle deposition and particle loss. To achieve dynamic equilibrium, the mass of particles being deposited to the leaf surface must equal the mass being lost via re-suspension mechanisms (Chamberlain and Chadwick, 1972), supplemented by rainfall and biological shedding e.g. of wax cuticle (e.g. Lehndorff et al., 2006). The main processes involved in attainment of dynamic equilibrium are dry deposition and particle re-suspension, both driven by air turbulence. The number of particles lost via re-suspension is a constant proportion of the total present on the leaf surface. Therefore, the leaf particle loading can equilibrate upwards or downwards (independently of supplementary loss mechanisms such as rainfall), dependent upon atmospheric concentrations of particulate pollution. The length of time for such an equilibrium to be reached is the minimum exposure time required in order for leaf SIRMs to reflect ambient PM_{10} concentrations.

Pertubations which may affect the attainment of dynamic equilibrium, and must therefore be accounted for in sampling protocols, include rainfall and transient inputs of relatively 'non'-magnetic PM_{10} from distal sources (e.g. tropical/sub-tropical dust storms). From available fixed monitoring station data, the sample locations used in this study do not experience distal particulate input, but rainfall is accounted for as described.

One possible additional contribution to the SIRM which might demonstrate progressive increase with time is the 'take-up' of particles within the leaf structure (e.g. Matzka and Maher, 1999; Lehndorff et al., 2006; Szönyi et al., 2008). However, consecutive monthly measurements of deciduous trees over the entire in-leaf season (around a UK point source) exhibit no progressive increase in magnetic values (Mitchell et al. in prep). This indicates that the surface particles dominate the leaf magnetic signature. It seems likely that waxy evergreen leaves capture ambient particles at a slower rate than the deciduous tree species investigated here, and hence evergreens may incorporate a larger proportion of particles within the leaf structure, rather than upon the leaf surface. Deciduous trees therefore appear more suitable for monitoring of ambient particulate pollution than evergreen species. Robust inter-calibration of leaf SIRMs between tree species offers the opportunity for greatly enhanced spatial coverage of PM_{10} monitoring, both across the urban roadside environment and around pollution point



n	12	11	10	9	8	7	6	5	4	3	2	1
p	>0.1	0.05	0.01	0.01	0.05	0.05	0.1	>0.1	>0.1	0.1	n/a	n/a

Fig. 5. Stepwise-excluded R^2 values for the correlation between leaf SIRM and ambient PM_{10} concentrations. Initially, all samples were included, then stepwise removed to identify the time period after which the leaf SIRM is most reliably representative of ambient PM_{10} concentrations.

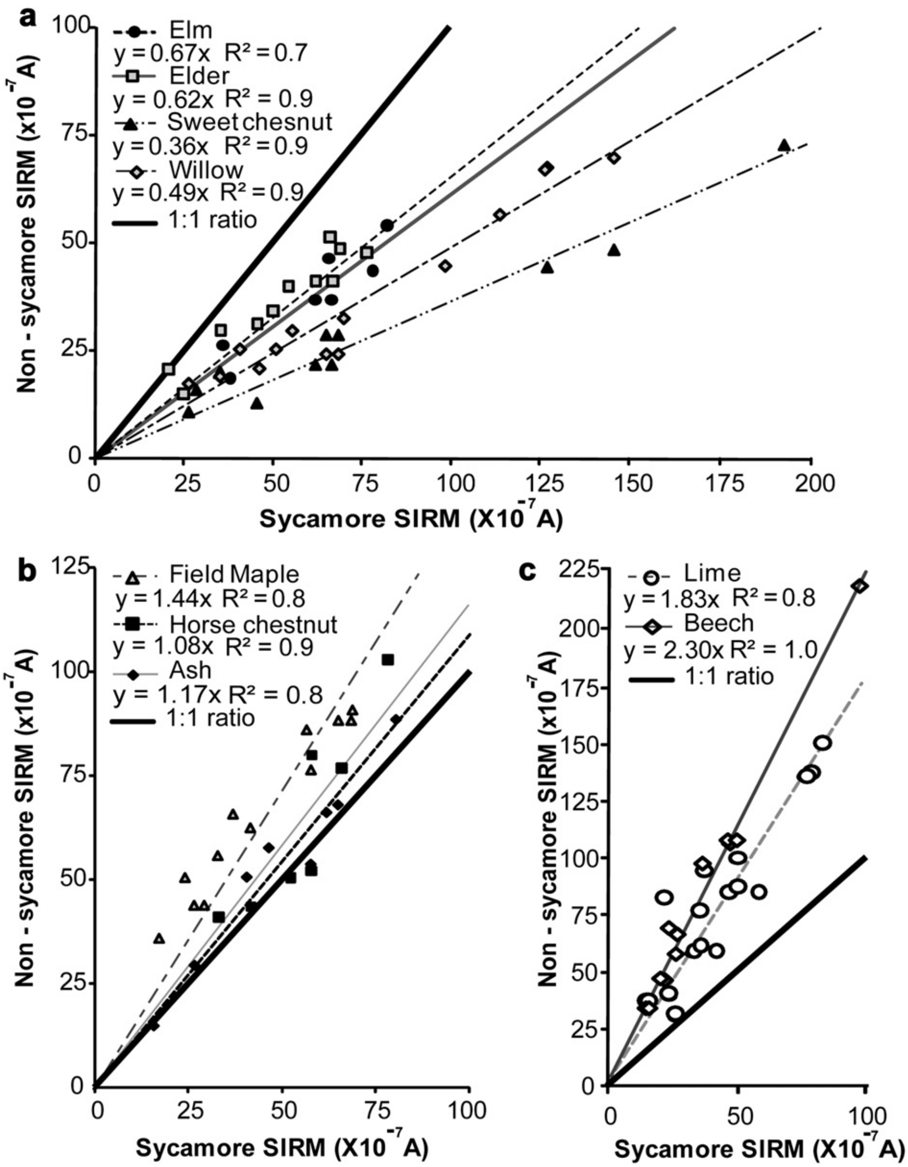


Fig. 6. Correlation between SIRM values of samples from sycamore and other co-located species. The different species can be subdivided into 3 classes by pollution-capturing efficiency with respect to sycamore: (a) less efficient; (b) ~ equal; and (c) more efficient.

sources. Measured magnetic properties of leaf samples collected monthly from 36 field sites around an elevated stack point source showed within-species consistency in magnetic deposition velocity to the leaf surface. The strong correlation between the measured SIRMs for lime and sycamore ($R^2 = 0.81$, $n = 17$, $p = 0.01$), and for sycamore and other non-sycamore species (Fig. 6; Table 1) enables use of the MV_d calculated for lime (2 ms^{-1}) to calculate MV_d for other species (Table 2).

Table 1
Correlation between SIRM values of samples from sycamore and other co-located species. R^2, n and p are given for each species.

Species	R^2	n	p
Sycamore	1.0	104	n/a
Elm	0.85	7	0.01
Elder	0.71	12	0.01
Sweet chestnut	0.87	10	0.01
Willow	0.93	10	0.01
Field maple	0.80	10	0.01
Horse chestnut	0.79	7	0.01
Ash	0.92	7	0.01
Lime	0.81	17	0.01
Beech	0.99	12	0.01
Oak	−0.33	12	0.00

The highest MV_ds are observed for leaves with a ridged and hairy morphology; the lowest rate of deposition occurs for leaves with smooth, waxy surfaces. Particles appear to accumulate around ridges in the leaf surface (Mitchell and Maher, 2009). Additionally, trees such as lime and birch attract aphids which secrete a 'honeydew' waste product making the surface of the leaf sticky, possibly enhancing particle retention. Other species such as field maple directly secrete honeydew to the same effect.

It should be noted that all species in this dataset are deciduous, therefore magnetic monitoring may only be undertaken during the 'in-leaf' season. However, deposition of magnetic particulate pollution onto pine tree needles has a reported 'equilibrium' time of ~26 months (Lehndorff et al., 2006), therefore pines appear unsuitable for monitoring of contemporary ambient air quality and associated human exposure.

Table 2
Calculated PM_{10} magnetic deposition velocities (MV_d) [±1 S.E.] and leaf arrangement and morphologies of 11 deciduous tree species (Allaby, 2006).

Species	V_d (cms^{-1})	Leaf arrangement	No. leaflets	Shape	Margin	Surface	
						Upper	Lower
Sweet chestnut	0.5 [0.13]	Simple alternate	–	Lanceolate	Serrate	Ridged waxy	Hairy (young)
Willow	0.6 [0.16]	Simple opposite	–	Linear	Serrate	Smooth waxy	Smooth
Elder	0.8 [0.20]	Compound pinnate	5–9	Obtuse or falcate	Serrate	Smooth	Hairy
Elm	0.9 [0.20]	Simple alternate	–	Aristate	Doubly-serrate	Smooth	Smooth
Sycamore	1.3 [0.16]	Simple opposite	–	Palmate	Serrate	Ridged	Hairy
Horse chestnut	1.4 [0.89]	Compound palmate	5–7	Cuneate	Doubly-serrate	Ridged	Ridged
Ash	1.5 [1.02]	Compound opposite	9–13	Obtuse	Serrate	Ridged	Hairy (young)
Field Maple	1.9 [0.46]	Simple opposite	–	Palmate	Entire	Smooth 'honeydew'	Ridged
Lime	2.4	Simple	–	Oblique cordate	Serrate	Ridged	Ridged hairy
Beech	3.0 [1.52]	Simple	–	Obtuse	Entire or ciliate	Ridged hairy	Hairy
Birch	4.6	Simple alternate	–	Deltoid	Doubly-serrate	Ridged, hairy	Ridged

5. Conclusions

- Biomagnetic monitoring of PM_{10} deposition on initially 'clean' (glasshouse-grown) tree leaves shows that particles gradually accumulate on the surfaces of deciduous tree leaves until a dynamic equilibrium between particle deposition and particle loss is reached. For birch and lime trees, the time required for equilibrium to be reached is of the order of 6 days.
- When dynamic equilibrium has been reached, leaf SIRMs reflect, and can act as a quantitative surrogate for, ambient PM_{10} concentrations. Rainfall events lower both ambient PM_{10} concentrations and leaf SIRMs.
- Surface morphology appears to be a dominating factor in particle deposition to the leaf surface, with ridged, hairy leaves exhibiting greatest deposition velocities.
- Particles on the leaf surface, rather than particles incorporated into the leaf structure, dominate the magnetic signature for the entire in-leaf season for deciduous species. Deciduous trees, demonstrating relatively rapid equilibration with ambient PM10 concentrations, appear preferable for monitoring purposes compared with more slowly accumulating evergreen species.
- SIRMs measured for a range of different deciduous tree species can be reliably inter-calibrated, thus enabling PM_{10} monitoring at unprecedentedly high spatial resolution.

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Appendix 1. Magnetic methods

All magnetic measurements were carried out at the Centre for Environmental Magnetism and Palaeomagnetism at Lancaster University.

The ARM was imparted at 80 milliTesla (mT) in a 0.08 mT dc biasing field (using a Molspin demagnetiser with ARM attachment), and subsequently AF-demagnetised at fields of 20, 50 and 100 mT. The susceptibility of anhysteretic remanent magnetisation (χ_{ARM}) was calculated by normalizing the ARM by the dc biasing field.

Room-temperature remanent magnetisation (IRM) was then incrementally acquired (in dc fields of 20, 50, 100 and 300 mT) using a Molspin pulse magnetizer. The 'saturation' remanence (SIRM), acquired by subjecting samples to an applied magnetic field of 1 T generated using a Newport electromagnet, was used to indicate the total concentration of magnetic particles (Muxworthy et al., 2003).

All leaf remanence values were measured using a Molspin Minispin magnetometer (sensitivity level $\sim 0.1 \times 10^{-8}$ Am^2) and normalized for leaf surface area (Matzka and Maher, 1999). The magnetometer was calibrated routinely (i.e. after ~ ten sample measurements) against a laboratory rock specimen. Magnetic measurements of the pumped-air filters were also carried out using a Molspin magnetometer and normalized for surface area (calculated from scanned leaf images using a pixel-counting programme).

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