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ATMOSPHERIC ENVIRONMENT

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Atmospheric Environment 33 (1999) 4565–4569

Magnetic biomonitoring of roadside tree leaves: identification of spatial and temporal variations in vehicle-derived particulates

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Received 12 January 1999; accepted 22 April 1999

Abstract

We report here the novel use of rapid and non-destructive magnetic measurements to investigate the spatial and temporal pattern of urban dust loadings on leaves of roadside trees. More than 600 leaves were collected from birch trees and their remanent magnetization ($IRM_{300\ mT}$) determined and normalized for the leaf area. The results show that this normalised 2-D magnetization is dominantly controlled by the tree's distance to the road. The magnetic analyses enabled detailed mapping of the spatial and temporal variations of vehicle-derived particulates. Higher 2D-magnetizations, indicating higher magnetic dust loadings, were measured for leaves collected adjacent to uphill road sections than for those next to downhill sections. This suggests that vehicle emissions, rather than friction wear or resuspended road dust, are the major source of the roadside magnetic particles. Additional magnetic analyses suggest that the particle size of the magnetic grains dominantly falls in the range classified for airborne particulate matter as $PM_{2.5}$ (<2.5 μm), a particle size hazardous to health due to its capacity to be respired deeply into the lungs. Thus, the leaf magnetizations relate directly to release into the atmosphere of harmful vehicle combustion products. For leaves from individual trees, magnetization values fall significantly from high values proximal to the roadside to lower values at the distal side, confirming the ability of trees to reduce aerosol concentrations in the atmosphere. Magnetic analysis of leaves over days and weeks shows that rainfall produces a net decrease in the leaf magnetic loadings. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Magnetic measurements; Vehicle pollution; Biomonitoring

1. Introduction

Strong links have been demonstrated between fine-grained (<10 μm) particulate matter and the risk of respiratory and circulatory morbidity and mortality (e.g. DoH, 1995; Pope et al., 1995). The greatest health impacts may be due to the *finest* particulate material (<2.5 μm, $PM_{2.5}$), which can be inhaled deeply into the unciliated and alveolar sections of the lungs. However, sources and loadings of $PM_{2.5}$, and its degree of association with larger particles, are presently poorly known.

Magnetic particles are found almost invariably amongst atmospheric particulate pollutants (e.g. Flanders, 1994; Hunt et al., 1984). Iron often occurs as an impurity in fossil fuels – during industrial, domestic or vehicle combustion, carbon and organic material are lost by oxidation whilst the iron forms a non-volatile residue, often comprising glassy spherules (due to melting). These spherules are *magnetic*, with magnetizations easily measurable using cryogenic magnetometers. Depending on the fuel type and temperature of combustion, the spherules contain variable amounts and grain sizes of

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1352-2310/99/$ - see front matter
PII: S1352-2310(99)00229-0

magnetite (Fe_3O_4) and/or haematite (αFe_2O_3). It has been shown that combustion processes simultaneously release hazardous substances and magnetic particles into the atmosphere. Morris et al. (1995), for example, found strong correlation between sample mutagenicity and magnetic susceptibility for urban dust samples. In addition to these combustion-related particles, non-spherical magnetite particles can be generated by vehicles, via exhaust emissions and abrasion/corrosion of engine and/or vehicle body material (Olson and Skogerboe, 1975). Hoffmann et al. (1999) mapped the magnetic enhancement of the soil surface due to vehicle pollution by measuring profiles of magnetic susceptibility along a German motorway. Maximum values were found to be localized within 2–5 m of the road and reflected the prevailing wind direction.

A magnetic study of urban dusts in the centre of Munich (Matzka, 1997) identified high correlation between total PM_{10} dust mass and its magnetic concentration (as indicated by its saturation remanent magnetization (SIRM) – the magnetization retained by a sample after exposure to a large magnetic field, e.g. 300 mT or 1 T). The correlation coefficient of SIRM and PM_{10} dust mass was $r = 0.879$, with a standard error of 0.15 (sample number of 12), indicating coherent co-existence of magnetic and other urban dust particles.

Due to a combination of high total surface area and 'favourable' surface properties, tree leaves can remove particulate matter from the atmosphere (e.g. Freer-Smith et al., 1997). As raindrops contain particles collected from the atmosphere, they could either contribute to dust accumulation on leaf surfaces or, by detaching previously collected particles, its reduction.

Magnetic monitoring of pollution by measurements of leaves is potentially efficient: samples are abundant and hundreds of samples can be collected and analysed in days. Schädlich et al. (1995), for example, measured the magnetic susceptibility of pine needles from 31 sites in the industrial region of Leipzig-Halle in Germany. They found higher magnetic values for needles affected by fly-ash deposition from power plants.

2. Sampling methods

The city of Norwich (England), and its surrounding area, is characterized by little industry but, due to 100,000 inhabitants, a substantial volume of traffic. Almost all the leaf samples were collected in the city centre, some in the suburbs, and also a few from the rural north Norfolk coast, at Weybourne, with only one minor road in the area.

Leaves were collected from one species of tree to avoid the possibility of species-dependent differences in dust absorbency. Birches (*Betula pendula*) proved most abundant in the urban environment (e.g. gardens, parks, roadsides). Each sample consisted of six leaves, taken from the outer canopy at a height of 1.5–2 m, packed into 10 cc plastic sample pots. To ensure leaves of similar age, only the oldest leaves of the newest twig growth were collected.

To determine the reliability of the method, two samples were taken for every sampling position and their mean value and deviation calculated. Consequently, the calculated magnetizations for each sample point are based on the measurement of 12 leaves.

3. Measurements

Measurements were carried out at the Centre for Environmental Magnetism and Paleomagnetism of the University of East Anglia. All samples were magnetized with a pulsed magnetic field of 300 mT; the isothermal remanent magnetization ($IRM_{300\ mT}$) was then measured with a CCL cryogenic magnetometer with a sensitivity of 10^{-10} A m^2 (the weakest leaf samples had magnetizations of $\sim 10^{-8}$ A m^2). The area of each leaf was calculated by digitising their computer-scanned images. The 2D-magnetization was calculated as the magnetic moment per leaf area, in units of Amperes (A = Am^2/m^2). After measurement, a small number of leaves, representative of different sampling locations, was cleaned with water, detergent and ultrasonics, to determine their 'background' magnetization.

To characterize the magnetic mineralogy and grain size, additional magnetic measurements were made, including room temperature IRM and AF demagnetization, described in more detail elsewhere (Matzka and Maher, in preparation).

4. Results

The magnetizations of leaves from trees at different distances from roads, and their background values, are shown in Table 1. Magnetizations are minimal for coastal and park trees, much higher for roadside trees and highest for trees most proximal to major roads. This magnetic pattern matches that reported by Impens and Delcarte (1979) for total dust interception by roadside trees. Maximum 2D-magnetizations were encountered for a sample taken between traffic lanes of an ascending major road. For the distal, least magnetic samples, $\sim 65\%$ of the measured signal is derived from adhering dust particles; for the proximal, most magnetic samples, $\sim 80\%$ is removed upon cleaning of the leaf (Table 1). The residual magnetization may indicate incorporation of some dust particles, imperfect cleaning of the leaf, or a non-airborne, biogenic magnetic contribution.

Leaf samples were collected at a dual carriageway at Grapes Hill, a major city road with a gradient of 12%.

Table 1
2D-magnetization ($IRM_{300\ mT}$) of sampled birch leaves for different locations versus 'background' $IRM_{300\ mT}$ of the subsequently cleaned leaves. Leaf samples from birch trees proximal to roads show much higher 2D-magnetizations than those from the north Norfolk coast and Norwich parks. Cleaning with detergent and subsequent ultrasonic cleaning removes between ~ 65 and 80% of the magnetization. Each data point represents a measurement integrating over 6 leaves

Sample location	2-D magnetization (10^{-6} A)	2-D magnetization after cleaning of leaf (10^{-6} A)	% magnetization removed by clearing
N. Norfolk coast (Weybourne)	5.12	1.9	63
Norwich park (Chapelfield Gardens)	8.46	2.94	65
Norwich park (Eaton Park)	9.34	2.01	78.5
Norwich park (Eaton Park)	10.51	2.33	78
Distal to street (Queens Road)	31.39	5.29	83
Proximal to street (Queens Road)	54.92	11.17	80
Central reservation of a dual carriage-way (Grapes Hill)	67.42	14.58	78

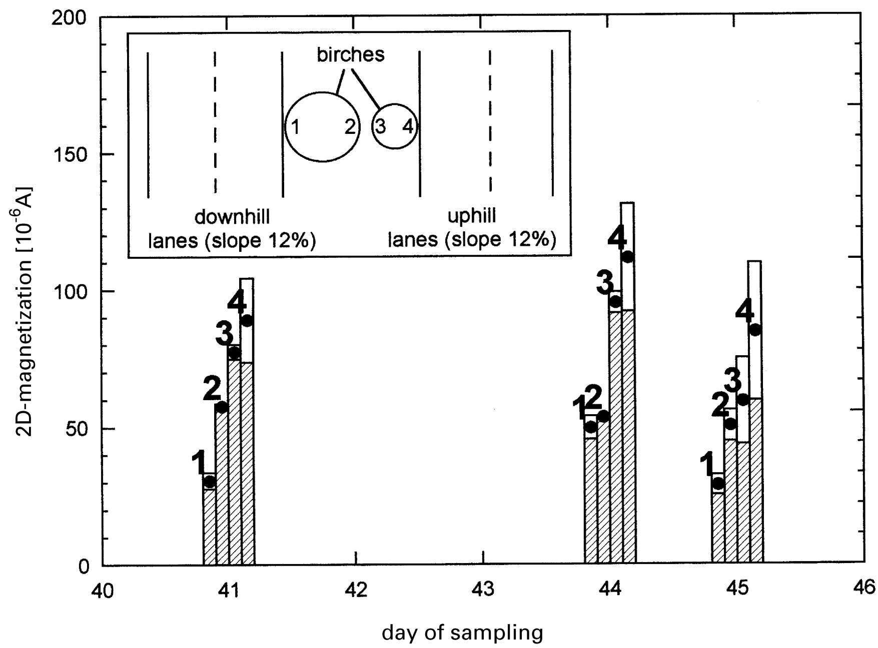

Fig. 1. $IRM_{300\ mT}$ of leaves sampled from between the downhill and uphill carriageways of a major road (Grapes Hill, Norwich) over several days. Every circle is the arithmetic mean of measurements of two samples (hashed and blank bars) each consisting of 6 leaves. Sampling positions are as indicated by the corresponding number in the inset sketch. Between day 1 (10 August 1997) and day 4 (13 August 1997), weather conditions were dry and sunny whereas rain fell between sampling on days 4 and 5 (14 August 1997).

Leaves were collected from two individual birch trees within a central reservation between the uphill and downhill lanes (Fig. 1). Sampling was carried out on day 1 (10 August 1997) and days 4 and 5. All three days show the same magnetization pattern, with higher values displayed by leaves from the tree adjacent to the uphill lanes. Maximum magnetization values range from 90 to 110×10^{-6} A, significantly higher than at the level section of this road. As the vehicles normally travel faster on the downhill lanes, they would probably raise more road dust, and due to greater braking, more friction-related particles. Conversely, driving uphill requires more energy, raising fuel consumption and exhaust emissions. The magnetization maximum next to the uphill lanes suggests that many of the magnetic particles are derived from combustion and exhaust-related sources.

The different sampling days not only show the reproducibility of this magnetization pattern, but also changes

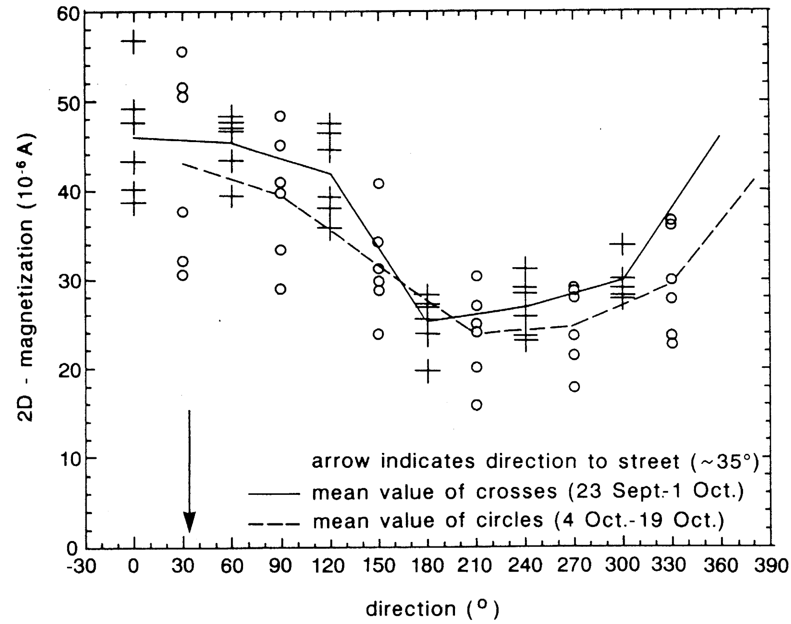

Fig. 2. $IRM_{300\ mT}$ of leaves sampled around a birch tree (QU12) within 5 m of a major road (Queens Road) in Norwich. Sampling positions are indicated by their direction clockwise from north as seen from the centre of the tree, with Queens Road sited at 35°N (arrows). Crosses at 0°, 60°,… are samples taken from 23 September 1997 to 1 October 1997 and circles are samples taken from 4 October 1997 to 19 October 1997 at 30, 90°,… . Each data point represents measurements of two samples, each consisting of 6 leaves; hence, the arithmetic mean (marked by the solid and dashed lines) for each sample position represents 72 leaves. The mean difference between the two samples measured at each sampling position is 13%, significantly lower than the magnetization differences between sampling positions.

with time and weather. In the three sunny days from day 1 to day 4, the magnetization increased by 20% of the mean value. After sampling on day 4, there were heavy rainfalls in Norwich. Resampling on day 5 showed a decrease in the mean magnetization of 28%, identifying the net removal of particles from leaf surfaces by rainwater.

Fig. 2 shows magnetizations of leaves sampled around an individual birch tree next to another major road (Queens Rd.) in Norwich city centre. From the centre of the tree, the road is located at 35° (clockwise from north) at a distance of 5 m. Traffic using the lane proximal to the birch tree runs from ESE to WNW. On the first 6 sampling days (23 September 1997 to 1 October 1997), samples were taken from a range of positions around the tree canopy: i.e. 0, 60,…, 300°; their $IRM_{300\ mT}$ values are plotted as crosses in Fig. 2. On the next 6 sampling days (4 October 1997 to 19 October 1997, indicated by circles), sample positions were changed to the intermediate positions 30, 90,…, 330°. In temporal terms, the decrease in mean magnetization seen over the second 6-day sampling interval can again be attributed to rainfall, on 7 October 1997. In terms of spatial pattern, both sample sets display maximum magnetization values proximal to the direction of the road. The side of the tree facing the approaching traffic (40–120°) shows significantly higher values; at the distal side, magnetization values are lower by 45%. This pattern confirms the effectiveness of trees as particulate filters when planted proximal to a source (Madders and Lawrence, 1981; Beckett et al., 1998). Sinusoidal fitting of the data for each sampling day identifies maximum magnetizations between 42 and 71°. The first sample set has its maximum at 48°, the second set at 60°. These different directions may indicate the additional influence of different local prevailing wind directions.

For 9 roadside samples, displaying high magnetizations, and 9 distal samples with low magnetizations, IRM acquisition and AF demagnetization were analyzed in more detail. Both sample sets display little IRM acquisition (~ 5%) in applied magnetic fields beyond 300 mT, indicating that the magnetic mineralogy is dominated by ferrimagnetic minerals like magnetite. AF demagnetization experiments show that the roadside samples display very similar magnetic stability. Their coercivity of remanence (the magnetic field required to demagnetize the IRM) is > 40 mT, indicating magnetic particles with a grain size from 0.03 μm to 3 μm (Heider et al., 1996), if magnetite is assumed as the remanence carrier. The distal, weakly magnetic leaf samples display more variable magnetic stabilities, indicating a range of possible magnetic grain sizes.

5. Conclusions

1. The magnetization of birch leaf samples collected from urban and rural areas of Norfolk, UK is controlled by their proximity to major roads and may be an easily measurable proxy for vehicle pollution loadings.
2. Highest leaf magnetizations were found adjacent to the uphill (12°) lanes of a dual carriageway, indicating a combustion- and/or exhaust-related source of the magnetic particles.
3. Magnetic analyses indicate that the grain size of the particles is of the order of 0.3–3 μm, a size of potential hazard to health due to its capacity to be respired deeply into the lungs.
4. For leaves from individual trees, magnetization values fall from higher values on the road-proximal side to low values on the distal side, indicating the ability of trees to reduce particulate concentrations at respirable height within the atmosphere.
5. Rainfall produces a net decrease in the concentrations of magnetic particles on leaf surfaces. Leaf drip and stem flow may thus act to remove fine-grained particulates from the roadside zone.

Acknowledgements

We appreciate the constructive comments of W. Morris and another anonymous reviewer. We thank M. Hounslow, S. Dorling, A. Watkinson and N. Petersen for helpful discussions and Norwich City Council and Norfolk Police for kind assistance. Thanks also to V. Karloukovski for computational help and M. Lemanski and T. Mutch for helping plugging leaves. JM was partially funded by the German Academic Exchange Service (DAAD).

References

Beckett, K.P., Freer-Smith, P.H., Taylor, G., 1998. Urban woodlands: their role in reducing the effects of particulate pollution. Environmental Pollution 99, 347–360.

Committee on the Medical Effects of Air Pollutants, 1995. Health Effects of Non-Biological Particles, Department of Health, HMSO, London.

Freer-Smith, P.H., Holloway, S., Goodman, A., 1997. The uptake of particulates by an urban woodland: site description and particulate composition. Environmental Pollution 95, 27–35.

Flanders, P.J., 1994. Collection, measurement, and analysis of airborne magnetic particulates from pollution in the environment. Journal of Applied Physics 75, 5931–5936.

Heider, F., Zitzelsberger, A., Fabian, K., 1996. Magnetic susceptibility and remanent coercive force in grown magnetic crystals from 0.1 μm to 6 mm. Physics and Earth Planet International 93, 239–256.

Hoffmann V., Knab M., Appel E., 1999. Magnetic susceptibility mapping of roadside pollution. Journal of Geochemical Exploration, in press.

Hunt, A., Jones, J., Oldfield, F., 1984. Magnetic measurements and heavy metals in atmospheric particulates of anthropogenic origin. Science of the Total Environment 33, 129–139.

Impens, R.A., Delcarte, E., 1979. Survey of urban trees in Brussels Belgium. Journal of Arboriculture 5, 169–176.

Madders, M., Lawrence, M.I.G., 1981. The role of woodland in air pollution control. Quaterly Journal of Forestry 76, 256–261.

Matzka J., 1997. Magnetische, elektronenmikroskopische und lichtmikroskopische Untersuchungen an Stäuben und Aschen sowie an einzelen Aschepartikeln. Diploma Thesis. Inst. Allg. Angew. Geophysik, University of München, München, Germany.

Morris, W.A., Versteeg, J.K., Bryant, D.W., Legzdins, A.E., McCarry, B.E., Marvin, C.H., 1995. Preliminary comparison between mutagenicity and magnetic susceptibility of respirable airborne particulates. Atmospheric Environment 29, 3441–3450.

Olson, K.W., Skogerboe, R.K., 1975. Identification of soil lead compounds from automotive sources. Environmental Science and Technology 9, 227–230.

Pope, C.A., Thun, M.J., Namboodiri, M.M., Dockery, W.D., Evans, J.S., Speizer, F.E., Heath, C.W., 1995. Particulate Air Pollution as a Predictor of Mortality in a Prospective Study of US Adults. American Journal of Respiration and Critical Care Medicine 151, 669–674.

Schädlich, G., Weissflog, L., Schüürmann, G., 1995. Magnetic susceptibility in conifer needles as indicator of fly ash deposition. Fresenius Environmental Bulletin 4, 7–12.