

Identification of brake wear particles and derivation of a quantitative tracer for brake dust at a major road

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ABSTRACT

Traffic-generated air pollutant emissions can be classified into exhaust and non-exhaust emissions. Increased attention is focussing on non-exhaust emissions as exhaust emissions are progressively limited by regulations. To characterise metal-rich emission from abrasion processes, size-segregated analysis of atmospheric aerosol particles sampled with micro-orifice uniform deposit impactors (MOUDI) in March 2007 in London was performed. The samples were collected at a roadside and a background site and were analysed for Al, Ba, Cu, Fe, Sb, Ti, V, Zn, Ca^{2+} , K^+ , Mg^{2+} , Na^+ , and NH_4^+ . Most components showed a clear roadside increment, which was evident as a higher mass concentration and a change in the size distribution. In particular, Fe, Cu, Ba, and Sb correlated highly, indicative of a common traffic-related source. Using complementary information on the fleet composition, vehicle number and average speed, the brake wear emission was calculated using the EMEP/CORINAIR emission database. The total PM_{10} and barium emission of the traffic was determined by ratio to NO_x whose source strength was estimated from published emission factors. Barium was found to comprise 1.1% of brake wear (PM_{10}) particles from the traffic fleet as a whole, allowing its use as a quantitative tracer of brake wear emissions at other traffic-influenced sites.

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1. Introduction

With the introduction of European Union Limit Values for particulate matter (PM) (EU-Commission, 1999) source apportionment of PM has gained in importance as a tool for quantitative estimation of the contributions of different sources to airborne concentrations. In every larger agglomeration traffic contributes a large fraction of the aerosol mass. For traffic, not only the exhaust emissions, which are heavily regulated, but also the non-exhaust emissions need to be taken into account. Road dust resuspension, road surface abrasion, brake and tyre wear all contribute to non-exhaust emissions. Studies have shown that exhaust and non-exhaust emissions from traffic contribute almost equally to PM_{10} concentrations in roadside air (Harrison et al., 2001; Querol et al., 2004). The ratio of abrasion sources (brake, tyre and road wear) to resuspension was similarly calculated as approximately 1:1 for Marylebone Road (Thorpe et al., 2007).

Brakes and tyres vary considerably in their formulation and each manufacturer uses a different composition which makes it difficult to predict brake dust composition from the entire vehicle fleet. Brake lining materials comprise in general five main components (Stanford and Jain, 2001; Chan and Stachowiak, 2004): fibres for mechanical strength, abrasives for increasing friction, lubricants for stabilising the frictional properties, binders for maintaining structural integrity and fillers for adjusting the proportions of the different constituents. Thorpe and Harrison (2008) reviewed metals which are typically used in brake lining materials and road vehicle brake dust. Among the ones with highest abundances are barium, magnesium, antimony and zinc. At traffic-influenced sites it seems likely that brake wear is the main source of these elements and especially of barium, which is often used in the form of barite in the filler material of brake pads. Being contributed by only one major source is an important prerequisite for a substance to be used as a tracer for emissions, and other sources of barium are likely to be few.

In this study, size-segregated aerosol samples were collected at three sites in London, one roadside with heavy traffic (Marylebone Road) and two urban background sites (Regent's College and North Kensington) with a view to analysis of trace metals considered as candidates for tracers of non-exhaust vehicle emissions.

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2. Material and methods

2.1. Site description

Marylebone Road is a heavily-trafficked 6-lane road in central London which is frequently congested. The sampling site is located outside the University of Westminster within a street canyon. An urban background site was situated nearby at Regent's College, located in Regent's Park, 800 m north of Marylebone Road and free from local traffic influences.

Both the Marylebone Road and Regent's College sites housed eight stage micro-orifice uniform deposit impactors (MOUDI), Model 100, (MSP Corporation) and a Partisol Sampler (Dichotomous Partisol Plus Model 2025 Sequential Air Sampler; Rupprecht and Pataschnick Co. Inc). The MOUDI sampled through 2-metre vertical stainless steel tube with a rain cover, while the Partisol was free-standing out-of-doors. Size-segregated measurements with the MOUDI samplers were taken on 7 separate occasions during this campaign, with the air sampling intervals as shown in Table 1. The Partisol samplers installed at both sites collected samples for gravimetric determination of the PM_{2.5} and PM_{2.5–10} mass concentration. The sampling interval was 24 h daily between 6th to 30th March 2007.

NO_x and PM₁₀ were measured continuously at Marylebone Road, which is a site affiliated to the national Urban and Rural Network (www.airquality.co.uk). As these parameters were not measured at Regent's College, urban background data from the nearby North Kensington national network site were used. This site is located 4 km from Marylebone Road and is subject to the same central London background air quality influences.

2.2. Analysis techniques

The MOUDI sampler (Marple et al., 1991) used substrates of 47 mm diameter PTFE and a backup filter (37 mm diameter) of the same material, with 1 µm pore size (Whatman). The flow-corrected 50% cut off diameters (D₅₀) for the aerosol flow of 22 L min⁻¹ were in µm 21.0, 11.5, 7.2, 3.7, 2.2, 1.2, 0.7, 0.4, and 0.2. After sampling, one half of each filter was extracted by acid digestion for trace metal analysis by inductively coupled plasma mass spectrometry (ICP-MS) (Agilent 7500ce with an Octopole Reaction System) and the other half was extracted for the analysis of the main water-soluble cations by ion chromatography (Dionex DX 500, with Gradient Pump GP40 and Ion Pac[®] CS12A Cation-Exchange Column). The extraction of the trace metals aluminium (Al), barium (Ba), copper (Cu), iron (Fe), nickel (Ni), antimony (Sb), titanium (Ti), vanadium (V), and zinc (Zn) was performed in 2 mL of a mixture of 189 cm³ L⁻¹ Aristar Grade hydrochloric acid and 66 cm³ L⁻¹ nitric acid (Aristar Grade), according to the procedure of Harper et al. (1983). Validation of this method is described in Harper et al. (1983). The mixture was heated in an ultrasonic bath at 80 °C for 60 min. Ni was analysed by ICP-MS but due to insufficient data the trace metal was not included in further analysis. The extraction was

made without HF thus aluminosilicates were not fully digested, and hence elements bound in aluminosilicate matrices are not dissolved quantitatively. The filter segments for cation analysis of calcium (Ca²⁺), potassium (K⁺), magnesium (Mg²⁺), sodium (Na⁺), and ammonium (NH₄⁺), were prepared by adding 0.5 mL propan-2-ol on the filter, and after wetting of the filter, 10 mL distilled deionised water was added. The mixture was shaken for 20 min.

PM_{2.5} and PM_{2.5–10} particle samples were collected with a Partisol sampler onto 47 mm diameter, 1 µm pore size PTFE filters (Whatman). The Partisol samplers contain a virtual impactor and flow controllers located in the inlet line, separating the flow into fine and coarse fractions, at flow rates of 15.0 L min⁻¹ and 1.7 L min⁻¹, respectively. The mass was defined by weighing the filters before and after exposure using a Sartorius Model MC5 microbalance. The instrument collects a small proportion of fine fraction particles on the coarse filter, for which a standard correction is applied. The filters were conditioned 24 h at a relative humidity of 35–45% and a temperature of 20 °C. To reduce the effects of static electricity an ionizing blower and an α-particle source (²¹⁰Po) were used. The PM₁₀ mass was calculated by adding PM_{2.5} and PM_{2.5–10}.

Traffic data were registered for the six lanes in Marylebone Road. The data included vehicle count, speed and vehicle class using inductive loop monitoring. Two loops are installed in each lane which gives information concerning the speed (11 bins) and vehicle classification in 6 groups due to successive pulses (EUR6 scheme).

2.3. Emission rates

Emission rates for PM₁₀ were calculated based on the assumption that PM₁₀ and NO_x concentrations in the traffic increment (roadside minus background concentration) share traffic as a common source, and behave similarly in the atmosphere. In our earlier work the NO_x exhaust emissions from traffic (E_{NOx}) were calculated using the National Atmospheric Emissions Inventory (NAEI, 2002) database, which disaggregates emission factors according to vehicle type, traffic numbers, and mean speed using data taken at Marylebone Road. Correction factors were applied to account for the changes in the fuel composition and the improvements in emission-control technology with time (NAEI, 2000). Recently, the U.K. Department for Transport has re-evaluated road traffic emission factors, and hence the newer data (Department for Transport, 2009) were used. The traffic-related PM₁₀ emissions (E_{PM10}) can be determined as follows (Lohmeyer et al., 2002):

$$E_{PM10} = E_{NOx}(\Delta PM_{10}/\Delta NO_x) \quad (1)$$

in which ΔPM₁₀ and ΔNO_x refer to the respective traffic increments in concentration (roadside minus urban background).

Hourly data were used, and as suggested by Thorpe et al. (2007), data with a ΔPM₁₀:ΔNO_x ratio larger 0.1 was excluded from the analysis. This restriction reduced the dataset by one data pair.

The brake emission factor was determined according to the EMEP/CORINAIR methodology (EEA, 2005) using the traffic data, vehicle types, speeds and numbers from Marylebone Road. A more detailed account of the methods of estimation of the emission factors is given by Thorpe et al. (2007).

3. Results and discussion

3.1. Particulate matter

The PM₁₀ concentration was measured at both the roadside, Marylebone Road, and the urban background site, Regent's College,

Table 1
Dates of sampling with MOUDI impactor at Marylebone Road and Regent's College.

Sample Number	Date/Time of sampling
1	07.03. 14:00–08.03.2007 12:00
2	10.03. 14:00–11.03.2007 12:00
3	13.03. 14:00–15.03.2007 10:50
4	17.03. 18:00–18.03.2007 18:00
5	20.03. 15:00–21.03.2007 14:00
6	22.03. 13:00–26.03.2007 9:52
7	28.03. 13:00–30.03.2007 12:00

with a Partisol sampler. A time series of coarse and fine fraction mass measurements appears in Fig. 1 and the average concentrations and the differences for the Partisol measurements can be found in Table 2.

3.2. Traffic

In March 2007 the average number of vehicles passing the installed measurement loops per day was $76\,803 \pm 2899$ (median = 77 026). Passenger cars and light vans represented the highest traffic numbers, cars with trailers formed the class with the lowest numbers. The mean average vehicle composition covering the seven measurement periods is given in Table 3.

3.3. Particulate matter composition

Differences in the concentrations of PM components, trace metals and cations, between Marylebone Road and Regent's College were determined. The median for all elements showed a roadside increment (Table 4), as did the mean of all components except potassium.

When comparing data from the single measurement periods, Ca^{2+} , Na^+ , Sb, Ba, Cu, and Fe showed a positive roadside increment in all seven samples, clearly indicative of a common traffic source. Spearman rank correlations were calculated for the trace metals separately for each site and between the sites. Ba at the Regent's College site correlated highly ($R > 0.8$) with Fe, Cu, and Sb at the same site. The same correlations were found for the components at Marylebone Road. Ba at Regent's College correlated highly with the following trace metals at Marylebone Road: Fe ($R = 0.86$), Cu ($R = 0.86$) and Sb ($R = 0.83$), but Ba at the roadside only correlated highly with Sb at Regent's College. The Spearman rank coefficients for correlations between Ba at Marylebone Road and Fe and Cu respectively at Regent's College were 0.54 and 0.50. The strength of these correlations indicates that Ba, Fe, Cu and Sb have a common source. Birmili et al. (2006) also found that in the United Kingdom

Table 2

Partisol $\text{PM}_{2.5}$ and PM_{10} data ($\mu\text{g m}^{-3}$), daily measurements 6th to 30th March 2007.

		Marylebone Road	Regent's College	Roadside Increment
$\text{PM}_{2.5}$	Average	24.8	20.3	4.55
	SD	15.4	19.3	8.91
	Median	21.2	11.5	5.48
PM_{10}	Average	40.6	31.4	9.20
	SD	20.0	23.3	9.62
	Median	36.1	22.3	9.18

Cu, Ba, Fe and Mn (which was not analysed in this study) in the size range $1.5\text{--}3\text{ }\mu\text{m}$ were correlated in urban air due to vehicle emissions. At Marylebone Road a lower correlation ($R = 0.71$) of Ba was found with Al, and correlations below 0.5 with Ba for Ti and Zn. At Regent's College Ba was also correlated highly with Fe ($R = 0.86$), Cu ($R = 0.89$) and Sb ($R = 0.80$).

For a more detailed analysis the mean concentration values per stage were compared between the two sampling sites. Ba and Ti were elevated on all stages at the roadside in samples where both were detected simultaneously at both sites. V, Fe, Cu, and Al showed enhancements in one size class at the urban background site, but on other stages the roadside contribution was clearly predominant.

Not only were the mass concentrations different between the two sites but also the size distribution varied. K^+ showed a large increase in the largest size fraction, $>11.5\text{ }\mu\text{m}$ and a smaller one in the $7.5\text{ }\mu\text{m}\text{--}11.5\text{ }\mu\text{m}$ size class at the roadside. K^+ is a soil-associated element likely to be associated with (road) dust resuspension, mainly affecting the larger particles. This positive roadside effect is also visible when comparing the Ca^{2+} concentrations and less clearly with Mg^{2+} . Fe, Ba, Cu, and Sb were clearly increased at the roadside in the size range $3.7\text{--}7.2\text{ }\mu\text{m}$. For Sb this peak is broader, covering three Moudi impactor stages, from 1.2 to $7.2\text{ }\mu\text{m}$. For Zn a roadside increment was also detected in the coarse fraction with particle diameters from 2.2 to $3.7\text{ }\mu\text{m}$. The elements Fe, Ba, Sb and Cu have been widely associated in the literature with brake wear

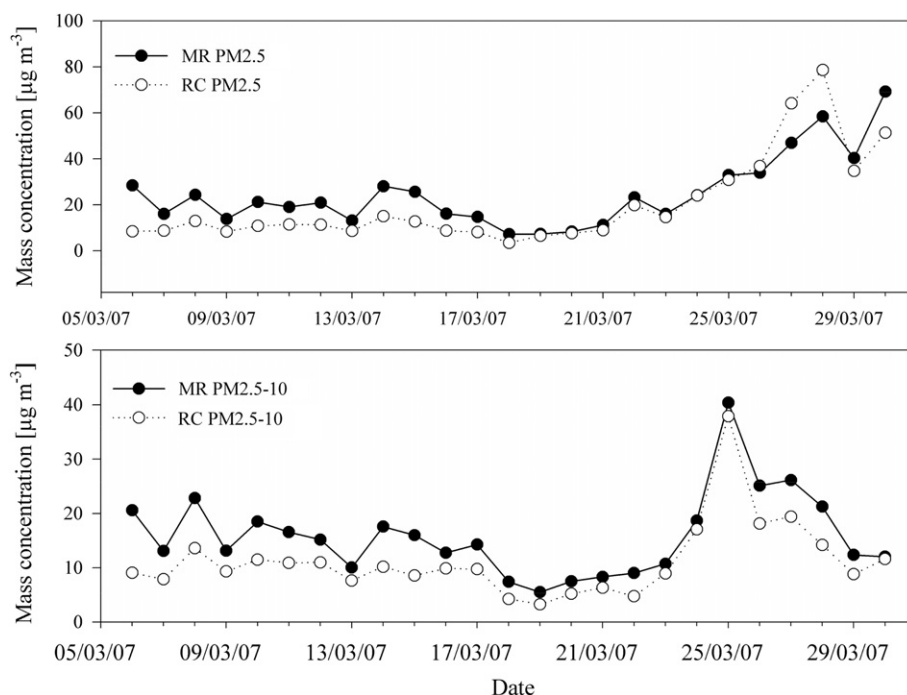


Fig. 1. Time series of mass concentrations of $\text{PM}_{2.5}$ and $\text{PM}_{2.5-10}$ measured with a Partisol at Marylebone Road (MR) and Regent's College (RC).

Table 3

Average vehicle fleet composition during seven sampling periods in March 2007 at the Marylebone Road site.

Vehicle type	Car/Light Van	Rigid Lorry, Heavy Van	Bus	2-Wheeler	Artic Lorry	Car with Trailer
Average	83%	10%	2.0%	1.9%	1.8%	0.87%

(Legret and Pagotto, 1999; Westerlund, 2001; von Uexküll et al., 2005; Iijima et al., 2007), and our data are also consistent with findings of Iijima et al. (2007) who reported that their laboratory generated brake dust particles peaked between 3 μm and 6 μm and consisted amongst other elements of Ba, Cu, K, Fe, Sb, and Zn. A smaller particle diameter, 2.8 μm , for brake wear was determined in a study by Wåhlin et al. (2006), with the trace metals Ba, Cu, Cr, Fe, Mo, Pb, Sb, Sn, Zn, and Zr as associated components. One large mode for Ba, Fe, and Sr was found by Allen et al. (2001) at three sites in the UK. A study by Brüggemann et al. (2009) in Dresden, Germany identified one main mode in the coarse size fraction between 1.2 and 3.5 μm for Cu and Fe which overlaps with the modes detected in this study. Cu was clearly increased at the roadside and showed high correlations with trace metals which occur in brake material. Denier van der Gon et al. (2007) estimated that 50–75% of the Cu emissions to the atmosphere in Western Europe are due to brake wear, which supports the assumption of brake wear being the main roadside source of Cu. Hjortenkrans et al. (2007) determined brake material as the main source for Cu as well as Sb with an emission ratio of Cu/Sb of 5.3 in Stockholm. The ratio calculated for this study was higher, being 9.1 on average. At the roadside Ba, Cu, Fe, and Sb showed great similarity in their size distributions. They all had their main peaks between 1.2 and 7.2 μm (Fig. 2). Furthermore Cu and Sb exhibited a second mode at 0.2 μm –0.4 μm .

The size distribution of Na^+ and V did not show any significant differences between the two sites. These elements are not normally associated with traffic emissions but with sea salt and biomass combustion (Na^+) and with soil material, oil combustion and oil refineries (V).

Considering the single measurement periods rather than the mean size distribution the behaviour of Ba became more complex. The main mode was again found in the larger size fractions, but the mode altered somewhat. The main mode could be either detected in the size class between 2.2 and 3.7 μm or split between two size fractions, 1.2–2.2 μm and 3.7–7.2 μm . Possible influences on the size-association of Ba are different driving patterns and the brake temperature. Both are influenced by the level of congestion and the meteorological conditions. PM originating from brake wear is expected to result from two processes. Mechanical abrasion

Table 4

Mean values for the concentrations at Regent's College, Marylebone Road and the roadside increment at Marylebone Road, concentrations in ng m^{-3} , SD = standard deviation.

	Regent's College			Marylebone Road			Roadside Increment	
	Average	SD	Median	Average	SD	Median	Average	Median
Aluminium	46.9	32.1	42.8	74.6	49.6	78.9	27.7	36.1
Ammonium	1123	1594	397	1197	1696	458	74.3	60.3
Antimony	1.31	0.807	1.49	6.73	3.48	6.24	5.41	4.75
Barium	3.90	2.31	3.65	17.9	12.8	15.6	14.0	11.9
Calcium	167	80.6	184	398	240	425	232	241
Copper	9.31	6.52	7.02	53.2	33.8	50.5	43.9	43.5
Iron	236	143	223	1214	722	1039	978	816
Magnesium	113	86.7	145	120	90.1	147	6.36	1.84
Potassium	743	331	720	684	468	773	−59.1	52.8
Sodium	995	610	1144	1108	617	1180	112	35.3
Titanium	1.42	1.10	1.13	3.05	1.87	2.71	1.63	1.58
Vanadium	1.87	1.70	1.08	2.13	1.72	1.62	0.26	0.54
Zinc	32.5	36.9	15.6	44.6	23.3	42.0	12.1	26.4

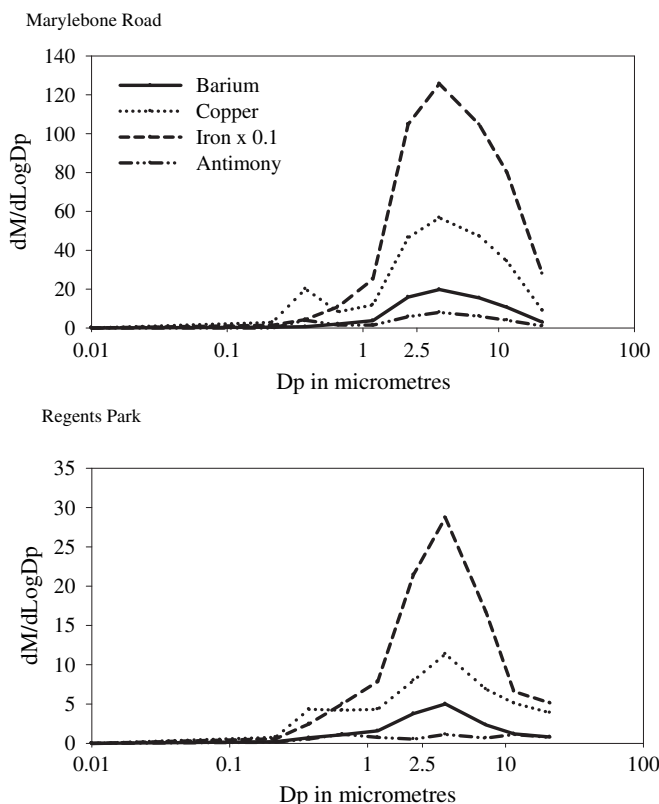


Fig. 2. Size distribution of Ba, Cu, Fe and Sb at Marylebone Road (above) and Regent's College (below).

generates particles of larger sizes, whereas brake pad materials contribute to the smaller particle size fractions by volatilisation and condensation. Volatilisation of some metals is possible as the brake temperatures can rise to several hundred degrees Celsius (Garg et al., 2000). Variations in modal diameter were also measured in some samples for Cu and Sb.

3.4. Emission rates

The NO_x and PM_{10} emission rates were calculated for Marylebone Road. Averaged over the seven sampling periods E_{NO_x} was $1.05 \text{ g vehicle}^{-1} \text{ km}^{-1}$. This NO_x emission factor was used to determine $E_{\text{PM}_{10}}$, which was calculated as $82.3 \text{ mg vehicle}^{-1} \text{ km}^{-1}$. A comparison with the PM_{10} emission rates for the years 2000–2003 for Marylebone Road showed that the emissions have reduced. This might be due to a change in the vehicle fleet

composition or improvements in abatement technology. For March 2007 the average brake wear emission based on CORINAIR emission factors was $10.7 \text{ mg vehicle}^{-1} \text{ km}^{-1}$ accounting for 13.0% of the total particulate matter emissions from traffic at Marylebone Road.

The ratio of the roadside increments of Ba in PM_{11} (13 ng m^{-3}) to the PM_{10} concentration increment averaged 0.14%. Combining this with the knowledge that brake wear emissions contributed 13% to the traffic-related PM_{10} concentration it is possible to calculate the contribution of Ba emissions to brake wear. Hence the mean proportion of Ba in estimated brake wear emissions (in PM_{10}) was 1.1%.

The ratio of the average roadside increment of Fe to Ba was 69.8. Assuming that the incremental Fe concentration was produced by brake wear as is suggested by the size distribution, the proportion of Fe in brake wear was 75%. Iron oxides are used as abrasives in brake lining material (Chan and Stachowiak, 2004), and brake discs, which are abraded in the braking process, are made of cast iron. In the review of Thorpe and Harrison (2008) metal concentrations in brake linings and brake dust were compiled. The percentage of Fe in brake linings was 1.2–63.7% and in brake dust was between 1.1 and 53.7%. These figures are hard to reconcile as the brake dust contains abrasion products from the iron disc or drum, and hence should have a higher Fe content than the lining. Iijima et al. (2008) reported that approximately 30% of brake dust originated from disc wear in their experiments. Sanders et al. (2003) report elemental analysis of brake wear debris, including the pad and disc component for low metallic, semi-metallic and non-asbestos organic brake linings. For both the former categories, the iron contribution to airborne dust is 47–48% whilst that of barium is 2.5–3.5%. They reported that wear rates are very material dependent. Given the very wide range of brake pad composition and wear rates relative to the disc, our data appear to be broadly consistent with earlier data derived from laboratory studies and instrumented vehicles. With 75% this study determined a higher value if brake dust is considered as the only source for the roadside incremental Fe concentration. However, Fe dust may also be emitted by tyre wear as studies of Hildemann et al. (1991) and Adachi and Tainosho (2004) have shown and this may be a contribution. It is thought unlikely that resuspension of road dust is an appreciable source of iron as the size distribution would be expected to be dominated by larger particles, as for aluminium, potassium and calcium. The very substantial roadside increment in iron concentrations observed at other U.K. sites (Harrison et al., 2003, 2004) is broadly consistent with a high abundance of iron in abrasion particles generated by road traffic, and wear of the disc or drum is clearly an important source of iron.

4. Conclusion

The elements Ba, Fe, Cu and Sb all show a large increment of roadside concentrations over the urban background (Table 4) and all show very similar particle size distributions in roadside air (Fig. 2). If barium is used as a marker element for brake dust particles, it is shown to comprise 1.1% of brake dust emissions estimated using CORINAIR emission factors. If Fe is assumed to arise solely from brake wear, it would represent 75% of the brake dust, which is considered feasible as brake discs and drums are subject to abrasion, but this may be an over-estimate, with there being possible contributions from tyre wear and resuspended particles.

The similarity between the size distributions of barium in roadside air and urban background air (Fig. 2) is strongly suggestive of brake dust being the predominant source of barium in background urban air as well as in the traffic increment at roadside. This being the case, it should be feasible to use barium as a quantitative tracer of brake dust in urban air, from which airborne concentrations of brake dust can be estimated.

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