





Atmospheric Environment 35 (2001) 4651-4662

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Factors influencing measurements of PM₁₀ during 1995-1997 in London

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Received 30 June 2000; received in revised form 17 January 2001; accepted 24 January 2001

Abstract

A 3 year set of PM₁₀ monitoring data using the tapered element oscillating micro-balance from 3 sites in London was analysed, in conjunction with air stream back trajectories and meteorological data for the same 24 h periods. Geometric mean PM₁₀ for any site against groupings of trajectory wind direction (TWD) showed a south-easterly peak in PM₁₀. Analysis of variance was carried out using London meteorological parameters and also wind speed and direction information calculated from positions on air mass back trajectories at 4 different periods prior to the approach to London. This model explained 60-65% of the observed variation, and because the analysis enables the meteorological factors to be de-coupled from the TWD, it can be concluded that local weather is not the prime cause of the peak. It was found that for 6-7% of the time when the TWD was from $120^{\circ}-160^{\circ}$, at least $15-20 \ \mu g \ m^{-3}$ of extra PM_{10} was advected in. Evidence is presented that the local meteorology is responsible for re-suspension. By removing individually each parameter retained in the model, its relative contribution to explaining variance was assessed. TWD, then trajectory wind speed (TWS) were found to be the most important. There was a calculated reduction of 7.0 μ g m⁻³ (95% confidence 6.0-8.0) of PM₁₀ over the 1047 day monitoring period. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Particulate matter; Meteorology; Wind trajectory; Statistical analysis; Secondary particulates

1. Introduction

The near universal need for large cities to monitor their citizens' exposure to particulates, especially to PM_{10} which are believed injurious to health, (COMEAP, 1995), has resulted in some uniquely large data sets. Monitoring authorities seldom have the resources for a detailed statistical analysis, though a good example of a report produced by Local Authorities in London is that by Rickard et al. (1999), but this addresses local pollution interests rather than fundamental research issues.

The UK National Air Quality Strategy, DETR (2000), sets an E.C. manual gravimetric standard for particles of less than 10 μ m (PM₁₀), of 50 μ g m⁻³ measured as a 24 h mean, with the objective to be reached by 2005 of no more than 35 exceedances per year, and 40 μ g m⁻³ maximum as annual mean. However the tapered element oscillating micro-balance (TEOM) has been used in London for 6 years, and whilst excellent for producing results every 15 min, it has some well documented disadvantages, in particular that the 50°C operating temperature dissociates and under records ammonium nitrate, present as a secondary particulate (Smith et al., 1997).

Others have analysed data sets measured in large cities, e.g. Olcese and Toselli (1998), who carried out a

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stepwise regression on PM_{10} in Cordoba, Argentina, using as variables CO, NO_x and meteorological parameters. However CO and NO_x are widely known to be highly correlated with PM_{10} because all three species are produced by vehicles. Our study extends such work by putting together 3 years (1995–97) of PM_{10} monitoring in London with wind trajectory speed and direction data for each day, and also with extensive daily meteorological information. We believe such data sources have not been studied statistically by the technique of analysis of variance (ANOVA) which provides a complement to the modelling approach based on a theoretical knowledge of the relevant processes.

This work tests three specific hypotheses.

- (1) That the trajectory wind direction plays a major role in accounting for variation in TEOM PM_{10} measured in London and that the observed south-easterly peak is statistically significant.
- (2) That the peak apparent in the raw PM_{10} measurements when plotted against TWD is not caused by weather effects in London.
- (3) That despite (2) above, the meteorology in London has an influence on PM_{10} measurements, and this can be elucidated.

The weather experienced by the air masses during their trajectory to the capital has not been studied, despite a probable influence on PM_{10} . To include this effect would be complex, and it is assumed that these effects will remain part of the unexplained variation in our model.

Two further questions have been addressed.

- Do daily trajectories provide information on the sources and the magnitude of London's imported PM₁₀?
- Which are the most important London meteorological parameters for explaining the variation in PM₁₀, and what is the relationship between particulates and each parameter?

2. Data bases

2.1. PM₁₀ from London sites

The South East Institute for Public Health has collected PM_{10} monitoring data from sites in London since April 1994. The data analysed are 2720 TEOM PM_{10} measurements from 1995–97, largely when simultaneous measurements were available from all three sites of Haringey (909 observations), Kensington and Chelsea (K&C, 905 readings), and Eltham (906); see Fig. 1. The Haringey site is at a roadside, but the others are urban background sites. The distances between these locations are Eltham to K&C 21 km, Haringey to K&C 11 km



Fig. 1. Map of London showing the 3 monitoring sites.

and Eltham to Haringey 18 km. The means of the PM_{10} at the three sites are Haringey 27.5 µg m⁻³, K&C 25.1 and Eltham 23.0. All measured TEOM daily values are from midnight to midnight, and the mean of 15 min interval TEOM measurements. All three sites were affiliated in 1996 to the Automatic Urban and Rural Network and operated to the national quality control standards required by the relevant UK government department (DETR). Prior to 1996 the sites were operated to similar standards. Data is required for a minimum of 75% of the time available for a day's results to be valid.

The PM₁₀ at any one site can be well predicted from another, with R^2 (square of regression coefficient) between 81% and 89% (no improvement with a quadratic). This implies that despite local baseline differences, the main factors affecting PM₁₀ are common across London. Harrison et al. (1997) also showed highly significant inter-site correlations in Birmingham of both PM_{10} and NO_x . Similarly Buzorius et al. (1999) found that the aerosol number concentration correlates well when measurements are made at different sites in Helsinki. Local variations may be due to traffic and industrial emission differences, and the common factors relate to meteorological conditions and the particulate content of air masses coming in to the Greater London area. The distribution of mean annual particulate sulphate ion in the UK and adjacent coast of Europe shows spatial variation of about 0.4 μ g m⁻³ per 100 km (APEG, 1999).

2.2. Air trajectory data

Computer calculated trajectories of air masses arriving over London in 1995–97 were obtained from the British Atmospheric Data Centre (BADC). This provides a database of routine back trajectories, all located at 900 mbars, (ca. 1300 m above sea level).

The latitude and longitude (lat/long) of central London are very close to $51.5^{\circ}N$, 0° . The trajectory information is available for lats of 51° and 52°N, both at long 0°. The calculated trajectories for 1995 (plotted over 60 h) relate to air masses arriving at the specified UK position each day at 12 noon. To obtain back trajectories for the London latitude, the 51° and 52°N trajectories were averaged by taking a mean of the lat and long of each position along the trajectory. For 1996 and 1997, the trajectories are available for midnight, 06:00, 12:00 and 18:00 each day and plotted back over 5 days, so to calculate a composite trajectory representative of the air mass arriving in London for one whole day, the means of lat and longs of 8 sets (4 for 51°N and 4 for 52°N) of individual trajectories were calculated. This gave a back trajectory for arrival each day for a 3 year period to match the 24 h particulate measurements.

The lat and long of the estimated position of the air mass 12, 24, 48 h and for 1996–97 only, 5 days prior to arrival in London was used to calculate a trajectory wind direction (TWD) and a trajectory wind speed (TWS) assuming a straight line of approach. Caution is needed in their interpretation, since Fast and Berkowitz (1997) warn that the use of back trajectories to identify surface source regions for long-range transport is suitable for only a coarse approximation of the point of origin.

2.3. Meteorological data

To test the significance of the observed trend of PM_{10} with trajectory wind direction, and to assist interpretation of other variations in PM₁₀, meteorological parameters were modelled taking data mainly from the London Weather Centre. To fill gaps, London Heathrow and Wisley in Surrey were used. The same meteorological parameters were used for all three monitoring sites on any one day. Some are inexact fits for the midnight to midnight continuous measurement of PM_{10} by the TEOM (see list below). The meteorological data were passed through the UK ADMS preprocessor module, to estimate additional parameters including friction velocity (u^*) , Pasquill stability class and boundary layer height (h), based on the theory of Brost and Wyngaard (1978) and the boundary layer depth formula of Nieuwstadt (1981). The ground level wind speed and direction at 09:00 h from meteorological data were not used because u^* is derived from the former, and because TWD and TWS are better descriptors the parameters found significant were:

- Hours of sunshine (00:00–24:00 GMT).
- Daylight hours (calculated, sunrise to sunset).
- Today's rain (rain in 24 h from 09:00 GMT).

- Yesterday's rain (rain in 24 h from 09:00 on day before monitoring period).
- *T*-range (temperature difference between daily maximum and minimum).
- *T*-mean (mean of daily maximum and minimum temperatures).
- TWS (wind speed from trajectories m s⁻¹). This is calculated from the 12, 24, 48 h or 5 day trajectory as appropriate.
- Visibility (m at 09 : 00 h GMT).
- Vapour pressure (of water mbars at 09 : 00 h).
- Friction velocity u^* (m s⁻¹, 24 h mean).
- PSC-D (h in 24 of atmosphere in Pasquill stability class neutral).

Also tested were barometric pressure at 09:00, boundary layer height (24 h mean), daily maximum and minimum temperatures, rainfall earlier than the previous day, relative humidity and the time spent by the atmosphere in Pasquill stability classes other than neutral. However, these proved non-significant, or less significant in some variants.

3. Results and discussion

3.1. Initial analysis

TEOM PM₁₀ data plotted as a function of TWD for any one of the sites demonstrates great scatter, and exceedances of 50 μ g m⁻³ are possible at nearly all wind directions. A clearer picture results from taking the geometric mean of PM₁₀ for groups of generally 10 observations when the 905–909 readings for one site have been sorted by increasing bearing (0–360°). Fig. 2 for K&C shows a peak in PM₁₀ from about 90° to 200°, irrespective of whether wind direction is calculated from



Fig. 2. Kensington and Chelsea 1995–97 TEOM PM_{10} Data. Geometric mean of 90 groups of 10 observations sorted and plotted by 3 durations of TWD.

12, 24, or 48 h trajectories. This is not site specific, since almost identical plots are obtainable from all three, though generally the magnitude of PM_{10} is Haringey> K&C> Eltham for each wind direction category.

Statistical analysis to test the initial hypotheses was undertaken, based on analysis of variance (ANOVA) using the General Linear Model (GLM; Minitab V12.2). This analytical but non-predictive model enables variables that are categorical in nature (called factors), as well as smoothly varying numerical parameters, such as temperature or rainfall (called covariates) to be tested. Interactions between covariates, between factors, or between either can be checked for significance. The factors are site (discrimination between the 3 monitoring sites), weekday/weekend and also TWD, which was treated as categorical because 0° and 360° are the same. The model is able to give R^2 as the indicator of the amount of variation explained.

The analysis seeks to explain as much as possible of the variation, and to confirm significant p values (p < 0.05) for all of the factors, covariates and interactions in the model. Because variables are related, the effect of a given factor or covariate is assessed as the last item added to the model. F and p values are calculated on this basis, but there is no simple threshold for Fbecause of varying degrees of freedom, but most effects are highly significant.

Several groupings of TWD category were tried, firstly in 5° bands of TWD, and then 10° bands. The model showed a higher R^2 for the more numerous 5° bands, but a disadvantage was that there were far more days with winds in some of the bands (W–SW quarter) than in others (SE). This resulted in large differences of standard deviation between bands, so they were made to have equal numbers of observations in them. The 2720 PM₁₀ observations were represented as

- (a) 91 bands called TWD91, 90 with 30 measurements in each, and a final band with 20.
- (b) 45 bands called TWD45, $(44 \times 60, \text{ and } 1 \times 80)$.

TWD can be ascribed in several ways, for example by straight line from the co-ordinates of London to the co-ordinates of a trajectory at 12, 24, 48 h and 5 days prior to air mass arrival. Thus for each day and each composite averaged trajectory, 4 different TWD91 and 4 alternate TWD45 directions are possible (called e.g. 12hTWD91, etc., see Tables 1a and 1b) to indicate trajectory duration and wind rose grouping. Seven data points were deliberately removed from the original 2727 PM_{10} entries where 1 of the 3 sites appeared greatly different from the other two, and had a large standardised residual. Where the standardised residual was high for all 3 sites on a given day, the data were retained. This residual was large for some very high and very low PM_{10} values.

3.2. Linear models

Initially a simple linear model was used, with no covariates squared, cubed, square rooted, etc. This produced an R^2 of 85.0%, but two serious problems were encountered. Firstly, the high R^2 was achieved with 6 interactions between TWD91 and covariates, all p < 0.0005. The model first estimates a slope for the relationship between Log_e PM₁₀ and the covariate, and where an interaction between TWD91 and that covariate exists, a second coefficient is calculated which is an adjustment to the gradient, and different for each TWD91 category. This is acceptable if a gradually evolving slope between Log_e PM₁₀ and the covariate is observable as the wind changes between 0° and 360°, but inspection suggested a random character with little physical meaning.

The second problem comes from the assumption that relationships between covariates and $\text{Log}_{e} PM_{10}$ are linear. But a quadratic least squares regression between $\text{Log}_{e} PM_{10}$ and individual covariates makes apparent that some relationships are appreciably curved, albeit with low regression coefficients. So quadratic models are preferred, and are those analysed further.

3.3. Quadratic models

The aim in the choice of model is to explain as much as possible of the variation and achieve high significance in the terms included, and address any subjectivity in this process by running variants. Models with many interactions were found difficult to interpret physically, so the quadratic models chosen are relatively simple with no interactions between covariates.

All the meteorological parameters mentioned in Section 2.3 were offered to the model, including the same parameters squared. Tables 1a and 1b list the factors and covariates in the eight modelling runs. The six model variants using the 12, 24 and 48 h trajectories had all parameters significant at the 99% level (p < 0.01) and mainly very highly significant (p < 0.0005). The 5day TWD91 analysis had today's rain as not significant. The six models with 12–48 h trajectories have R^2 of 61–65%.

The remaining 35–39% of unexplained variance may partly come from the TEOM measurements. Ammonium nitrate is not retained quantitatively on the TEOM filter (e.g. Allen et al., 1997). We have ourselves determined low concentrations of nitrate on a TEOM filter, and it appears possible that it is initially collected, particularly under episode conditions, and then slowly volatilised. High humidity may also affect the TEOM. AT 0400 on 12 February 1996 all three instruments recorded negative readings of around $-2/-3 \ \mu g \ m^{-3}$. Further possible sources of unexplained variance are the absence of parameters that relate to traffic flows, which

Table 1

Results from analysis of variance (general linear model in minitab)

(a) Base case

No of wind rose groups					
Trajectory duration	Degrees of freedom	F	р	Coefficient	SD
48hTWD45	44	2.88	< 0.0005	Factor	
Site	2	111.15	< 0.0005	Factor	
Weekday or weekend	1	44.23	< 0.0005	Factor	
Trajectory wind speed (TWS)	1	22.90	< 0.0005	-0.027907	0.005831
48hTWD45 interaction with TWS	44	5.24	< 0.0005	Interaction	
Visibility	1	88.16	< 0.0005	-0.000124	0.000013
Today's rain	1	26.04	< 0.0005	-0.010525	0.002062
Yesterday's rain	1	61.95	< 0.0005	-0.016332	0.002075
Long term trend	1	180.99	< 0.0005	-0.000299	0.000022
T-range	1	139.41	< 0.0005	0.031070	0.002631
T-mean	1	34.64	< 0.0005	-0.027641	0.004696
Daylight hours	1	24.89	< 0.0005	0.114570	0.022970
Friction velocity u^*	1	62.58	< 0.0005	-1.153600	0.145800
Pasquill stability class-D	1	38.48	< 0.0005	0.010868	0.001752
Daylight hours squared	1	33.13	< 0.0005	-0.005259	0.000914
U^* squared	1	19.27	< 0.0005	0.580900	0.132300
T-mean squared	1	74.51	< 0.0005	0.001540	0.000178
Vapour press squared	1	69.50	< 0.0005	-0.001188	0.000142
Visibility sq./1000	1	21.90	< 0.0005	0.000011	0.000002
Wind speed squared	1	14.43	< 0.0005	0.001294	0.000341

R squared = 62.04%

(b) Variant cases^a

	Parameters			
Solutions All 7 below	All	P<0.005	R squared (%)	
Except:				
12hTWD45	u* squared	0.001	60.73	
24hTWD45	Wind speed sq.	0.003	61.40	
5dayTWD45	Today's rain	0.006	60.35	
12hTWD91	(No Exceptions)		65.10	
24hTWD91	Wind speed sq.	0.006	65.22	
48hTWD91	u [*] squared	0.001	65.21	
5dayTWD91	Today's rain	0.167	66.49	

^aFactors, covariates and interaction are the same as in base case.

apart from the weekday/weekend distinction, are assumed constant. Also not included are meteorological effects along the trajectories, and non-linearity in the air movements. The quadratic model assumed will be an approximation, and there are also other unconsidered influences in a complex problem.

To assess the influence of one factor or covariate on $\text{Log}_{e} \text{PM}_{10}$, each has been removed in turn from the base case, and then reinserted before removing the next, so that a ΔR^2 was measured between the base case and the model with one term removed. Where linear and

quadratic terms of the same covariate were significant, both were removed to estimate the combined contribution (Table 2). The greatest ΔR^2 comes from the effect of 48hTWD45, together with the interaction between direction and TWS. Removing these two together lowers the R^2 by 12–13%.

3.4. Trajectory duration and wind rose grouping

Fig. 3 shows 4 ANOVA variants, with 4 trajectory periods all using 45 wind rose groups. The geometric

mean PM_{10} data plotted are *estimated* by the ANOVA (assuming other factors and covariates are at means) and show calculations of the isolated influence on PM_{10} of the 4 different descriptors of TWD depending on duration of trajectory. The 48 h TWD data gives a slightly higher south-easterly peak than the 24 or 12 h analogues, and the 5 day data a substantially poorer peak.

The different trajectory periods give TWD bearings that are cross correlated. A simple regression is not

Table 2

Removal of factors and covariates in turn: effect on *R* squared of base case

	R squared (%)	Delta from base case
No 48hTWD45 or its interaction with wind speed	49.70	12.34
No wind speed or WS squared or interaction with 48hTWD45	58.07	3.97
No friction velocity u^* or u^* squared	58.14	3.90
No Interaction of wind direction with wind speed	58.69	3.35
No Site discriminator	58.81	3.23
No long term trend	59.41	2.63
No visibility or visibility squared	59.69	2.35
No T-range	60.02	2.03
No T-mean or T-mean squared	60.83	1.22
No vapour pressure squared	61.03	1.01
No daylight hours or daylight hours squared	61.19	0.85
No yesterday's rain	61.14	0.90
No weekday/weekend discriminator	61.40	0.64
No today's rain	61.66	0.38
No Pasquill stability class-D (neutral atmosphere)	61.48	0.56
Base case	62.04	0.00

possible, since bearings cannot differ by > 180°. So for example, 12hTWD and 24hTWD bearings were tabulated, and a third column constructed of the differences. Where the difference was > 180°, 360° was added to the numerically lower bearing. Linear regression was carried out between these corrected values, with the result that 12hTWD can be predicted from 24hTWD with $R^2 =$ 96.1% and n = 907, from 48hTWD ($R^2 = 78.8\%$, n =907), and from 5dayTWD ($R^2 = 48.0\%$, n = 654). All three regressions had p < 0.0005.

The choice between dividing the wind rose into 91 or 45 groups rests on two considerations. Firstly TWD91 solutions give 3.2-6.1% higher R^2 than the TWD45 ones, because for each of the 91 categories the model calculates a separate mean (and standard deviation SD) for the estimated PM₁₀. Secondly although TWD91 demonstrates a clear high south-easterly peak, many smaller non-significant peaks (95% limits estimated from ± 1.96 SD) are also apparent, and contribute to absorbing variance. So the smoother trend line of the TWD45 solutions with 45 wind rose groups, more PM_{10} readings per group, smaller SD but slightly lower R^2 are preferred. But this choice does not affect the qualitative conclusions drawn, and the quantitative conclusions only modestly, since all the six solutions with trajectory periods up to 48 h in Table 1 have the same factors and covariates as highly significant. The solution 48hTWD45, nominated as base case, shows the highest estimated PM₁₀ after allowance for other meteorological influences, and will be analysed further.

3.5. Evidence for south-easterly peak

Fig. 4 is plot of 48hTWD45, together with error bars indicating 95% significance limits for the estimates of



Fig. 3. Results from ANOVA. Calculated effect on PM_{10} of TWD, showing effect of 4 alternative periods in advance to establish wind direction.



Fig. 4. Results from ANOVA on base case. Calculated effect on $Log_e PM_{10}$ of TWD (other influences at mean values) plotted as function of 48hTWD45, showing 95% significance limits.

PM₁₀ produced by the model. The three highest data points (60 original PM₁₀ observations and 20 TWD estimates in each) which span the range from ca 120° to 160° (about 7% of the monitoring period) are significantly greater than all other 48hTWD45 data points. Linear extrapolation into Europe leads into NE France, but taking into account the curved clockwise circulation round high pressure cells, pollution sources could encompass many urban centres of Eastern Europe. If the TWD descriptor is the 12 or 24 h trajectory, then the 3 data points of the peak are also significantly greater than all others except the one on each side of the peak itself. So whenever the geostrophic wind is from the southeast, as described by 12-48 h trajectories, there is a probability that London PM₁₀ will be at least 15-20 μ g m⁻³ higher. For the 5 day TWD data on the smaller 1996-97 data set, the SE peak is lower and the 95% significance limits much wider, suggesting serious trajectory curvature if straight line approach is assumed from this long in advance, or that particulate sources are nearer than 5 days away at mean wind speed.

The error bars on the points at the top of the SE peak in Fig. 4 are longer than those on the base line points, suggesting a wider spread in PM_{10} measurements contributing to the peak points. But if only occasional pollution episodes were responsible for the peak, even longer error bars might be anticipated. To investigate this, the well known episode of March 1996 was removed by excluding that month's data, using for analysis the 24 h TWD data. The highest 3 points in the 24hTWD44 plot are lower by $1-5 \,\mu g \, m^{-3}$ than when March 1996 is included, but the reduced peak is still statistically significant.

Since the local London meteorology is included in the model, it can be concluded that the SE peak made manifest by TWD is not caused by the former. But meteorological effects on the European continent during a typical passage of the air mass could be a contributing cause, since this is not modelled.

Visibility and its squared term are in the model. Visibility might be considered more a consequence of high particulates than a cause, and that it should be excluded, especially since visibility is at a minimum when TWD is SE. But although the ANOVA finds visibility to be informative in interpreting PM_{10} variation, it still leaves TWD as highly significant, and so the inclusion of visibility is a conservative assumption and strengthens the argument that the SE peak is wind related. If visibility is totally excluded it is found for the 48hTWD45 solution that the estimated mean PM_{10} of the highest data point on the SE peak rises from 49.8 to 54.9 μ g m⁻³.

If the model showing the largest sensitivity is correct (48hTWD45) and visibility is excluded, then the southeasterly increment advected rises to ca. $30 \ \mu g \ m^{-3}$ and there is a probability of the statutory limit being exceeded whenever TWD is in the above sector. In the raw data, the geometric mean PM₁₀ at 3 sites when 24hTWD is from 120–160° is 40.6 $\mu g \ m^{-3}$, and at Haringey, the site with the highest PM₁₀, the geometric mean is 43.8, with exceedance of the 50 $\mu g \ m^{-3}$ limit on 46% of days. Stedman (1998) has already described the contribution of secondary particulates from the SE to the episode of high PM_{10} in London in March 1996. Since the TEOM does not retain the important secondary particulate constituent of ammonium nitrate, it follows that the SE peak could be higher still if a manual gravimetric method such as the Partisol (Green et al., 2001) were used to measure PM_{10} . In other parts of the world the advection of aerosol pollution from one conurbation to another has also been reported, e.g. Mukai and Suzuki (1996), in Japan. Richards et al. (1999) showed from source apportionment in San Joaquin Valley aerosol that light extinction was dominated by emission sources contributing to the formation of secondary species, especially nitrate.

3.6. Influence of factors and covariates

3.6.1. TWS

This covariate approximates to the geostrophic wind speed. Both it and TWS squared are significant in the 8 solutions in Table 1. Fig. 5 shows that TWS has a U shaped relationship with estimated geometric mean PM₁₀. APEG (1999) presents data which show that PM_{2.5} decreases rapidly with increasing wind speed (R = 0.59), but that coarse particulates (i.e. PM₁₀–PM_{2.5}), rise with increasing wind speed due to resuspension (R = 0.34), which needs two components, traffic to make particles airborne, and wind to maintain them aloft. Lam et al. (1999) in Hong Kong studied the 3 parameters of wind speed, solar radiation and relative humidity in relation to PM₁₀, and found the highest correlation between wind speed and PM₁₀, (R = 0.49).

ANOVA produces one constant for the whole model, but none for individual covariates. In order to plot an individual covariate against $Log_e PM_{10}$ (or after conversion, against PM_{10}), a constant can be estimated from

Overall Mean $Log_e PM_{10} = Constant + coeff_1$

 \times (Mean TWS) + coeff₂ \times (Mean TWS)².

Coefficients are calculated when factors and covariates are at their means, and the above constant derives from assuming that the variation in $\text{Log}_{e} PM_{10}$ by one covariate leaves mean $\text{Log}_{e} PM_{10}$ unchanged.

There is a highly significant interaction between TWD and TWS, (48hTWD45 × TWS). The model produces coefficients for each of the 45 categories of the interaction. The interpretation of these coefficients is that they are modifications to the overall mean slope between $\text{Log}_{e} \text{PM}_{10}$ and TWS, and that for each value of TWD45, the coefficient changes the mean slope by an amount according to its sign.

In the linear model without TWS², the overall slope of $Log_e PM_{10}$ against TWS is negative, but when TWD is between 90° and 200° the slope modified by the interaction coefficients becomes positive. So within this sector the faster is TWS, the greater are particulates, presumably because they are advected in. For the base case, which has TWS in both linear and squared forms, the relationship between TWS and $Log_e PM_{10}$ is a flat U shape, see Fig. 5. The effect of the wind speed/wind direction interaction is to modify the position of the turning point (minimum) in the curve, to higher or lower values of wind speed. At values of wind speed above this minimum, the gradient is positive, implying that PM₁₀



Fig. 5. Relationships from ANOVA on base case. Effect of three London meteorological parameters on PM_{10} (other influences at mean values).

are advected in. Fig. 6 shows the calculated position of the minimum as estimated by the 48hTWD45 model. The graph shows individual turning points as a function of TWD45 and a smoothed plot from them using a Minitab statistical smoothing procedure (Kleinbaum and Kupper, 1978). Between 90° and 190° the minimum in the Log_e PM₁₀ versus TWS plot is at negative wind speed, so for all positive wind speeds PM₁₀ rises with wind speed, qualitatively the same conclusion as from the earlier linear model. This is selective directional advection and not simply re-suspension, because the latter would not be directional, nor happen at low wind speeds, which are at a minimum in the SE direction.

3.6.2. Evidence for re-suspension

The results of ANOVA have three covariates which at high values demonstrate an increase in calculated geometric mean PM_{10} . These are mean temperature (*T*-mean), temperature range (*T*-range) and TWS (Fig. 5), of which the two latter are present as both linear and squared terms. Days with low mean temperatures may show higher PM_{10} due to inversion conditions, and at high temperatures thermals are created which could maintain dust in suspension. High wind speed supports re-suspension, since for example, Harrison et al. (1997) found that coarse particles from re-suspension showed a positive dependence on wind speed.

The friction velocity (u^*) is a function of the turbulence caused by surface interference. It is related to local wind speed at 10 m height and to roughness length. Smooth surfaces have a low roughness length while a city has a high value. Here roughness length is constant, so u^* is proportional and interchangeable with

local wind speed, which is not in the model apart from the TWS. The plot of u^* (significant as both linear and quadratic terms), shows a concave curve with PM₁₀, but flattens at higher values of u^* . It appears to be modelling inversion conditions (see Pasquill classes below). If TWS² is omitted from the model, then u^* rises more sharply at high values, again reinforcing the re-suspension concept.

3.6.3. Site differences

The factor discriminating between sites is very significant (see Tables 1 and 2), but its interaction with TWD is very non-significant (p > 0.995), so even when TWD puts the centre of London between one site and another, there is still no rise in PM₁₀ at the downwind site. This is strong evidence for a remote source for the south-easterly peak. It is possible that for small distances between sites within London the TWD is less appropriate than the surface wind direction, which is not in the model. Scaperdas and Colvile (1999) have found big differences in CO concentrations in London as a result of surface wind variations.

3.6.4. Long term trend

The inclusion of computer date number in the model as a covariate allows the calculation that over nearly 3 years (1047 days) of monitoring the long-term reduction in PM₁₀ is 7.0 µg m⁻³ (95% confidence interval 6.0–8.0 µg m⁻³), which is presumably attributable to abatement measures and vehicle and fuel technical improvements. A 14 day running mean of raw PM₁₀ data from the 3 sites over the period has also been calculated, and linear regression carried out ($R^2 = 0.036$, p < 0.0005). The relationship explains



Fig. 6. Results from ANOVA on base case. TWS at which Log, PM₁₀ against TWS gradient becomes zero, (minimum).

negligibly the variation that is occurring, but confirms the downward trend (because p the probability of the null hypothesis is so low), and that over the 1047 days it predicts 4.8 µg m⁻³ of reduction, rather lower than that estimated from ANOVA.

3.6.5. Pasquill stability classes

Atmospheric stability has been classified in terms of categories following the work of Pasquill (1961). Classes A-C denote unstable atmospheres, E-G a stable nocturnal atmosphere, and D a neutral atmosphere is produced by dawn, dusk, overcast skies or stronger winds. Class 'D' (PSC-D) is significant in the base case, with a positive coefficient, showing that a neutral atmosphere promotes PM₁₀. Under neutral conditions PM₁₀ may be held in suspension, with little tendency to either rise or deposit. The Pasquill stability classes which relate to high stability (inversions) and high instability are not found significant, probably because other significant covariates (e.g. T-range, T-mean, u*) are collinear. When u^* and u^{*2} are removed and replaced by PSC-F&G, and its squared term, these then become significant.

Dupont et al. (1999) in a comparative study between the boundary layer of Paris and a suburb, found that at night the heat island effect gives a more neutral atmosphere than in the suburbs, where the ground is cooler. Oke (1995) provides more examples of this effect in large North American conurbations. Tsuang and Chao (1999) in their modelling of Taipei, found that fine particulates are major contributors to hazardous air quality occurring under meteorological conditions of stable air, low wind speed, and no rain. Under these conditions, the dry deposition velocities of fine particulates become 1–2 orders slower than normal, and high concentrations accumulate.

3.6.6. Other covariates and factors

As noted above, the monitoring periods for rainfall and PM_{10} coincided poorly. Rainfall corresponding to the covariate "Yesterday's Rain" was within a period of 24 h which began 15 h prior to the start of PM_{10} monitoring. The "Today's Rain" covariate totalled rainfall from 9 h after the start of PM_{10} monitoring until 9 h after its cessation. Both measures were in the base case (linear terms only), but yesterday's rain was the more significant, presumably because rainfall in 9 h of today's rain would be without effect on PM_{10} .

The increment in rain needed to achieve a reduction, (say 10%), in PM_{10} level can be estimated. It can be shown that

 $\frac{-\text{Log}_{e}(1/0.9)}{\text{coeff}} = \text{increase in rain.}$

From the coefficients in Table 1 extra rainfall needed to achieve a 10% reduction in TEOM PM_{10} can be shown

to be 6.5 mm (95% confidence limits 5.2–8.6) for yesterday's rain and 10.0 mm (7.2–16.3) for today's rain. In a theoretical study, Mircea and Stefan (1998) showed that light rain was much more effective at scavenging aerosols from the atmosphere than heavy rain. Beverland et al. (1998) looked at the ion content of rainfall in relation to back trajectories and found marked chemical differences according to origin.

The following were also found significant in the base case: hours of daylight (linear and squared terms); vapour pressure (VP, squared term only). VP was collinear with relative humidity and displaced it from the model, and higher values are associated with lower levels of PM_{10} indicating particle removal from the atmosphere. This is in line with increased aerosol particle size and deposition rates at high relative humidity (Quinn and Ondov, 1998; Harrison et al. 1999).

4. Conclusions

Calculated wind directions and wind speeds from BADC wind trajectories have been used, with the relevant meteorological data, to interpret 3 years of TEOM PM_{10} data in London. It was found from a simple plot of geometric mean PM_{10} against TWD that there is a clear peak in PM_{10} in the south-easterly direction. An ANOVA using the Minitab statistical software has been carried out to test the statistical significance of this peak, and to de-couple it from the meteorological effects in London.

TWD was calculated from positions on air mass back trajectories of 12, 24, 48 h and 5 days prior to London approach. It was found that the peak in calculated PM_{10} from the ANOVA was highly statistically significant in all three shorter descriptors of TWD, but the lower peak from 5 day TWD data on a smaller data base was only marginally or not significant. The peak was found to be at its largest and most significant when the 48 h trajectories were used.

Since the peak in the PM_{10} against TWD plot calculated by the ANOVA is when all other modelled influences are at their means, it follows that local London weather effects are not the prime cause.

There is a statistical probability that whenever the TWD sector is 120–160°, the geometric mean PM₁₀ is at least 15–20 μ g m⁻³ higher than from other directions, and that this occurred on 6–7% of the days during 1995–97. The above conclusions are significant at the 95% level. The 6 ANOVA model variants based on the 12, 24, and 48 h TWD assumptions, and on 45 and 91 groupings of wind rose sectors, all explained 60–65% of the observed variation in PM₁₀.

Within the SE sector, but from about $90-200^{\circ}$, the higher is the TWS, the greater are PM_{10} . This is

additional strong evidence that the PM_{10} are advected in. Outside this sector TWS has a flat U shaped relationship with PM_{10} . Additionally within the SE sector, mean TWS is at a minimum.

The model provides evidence of re-suspension of particulates contributing to elevated PM_{10} . Thus higher PM_{10} in London result at high mean temperatures, on days of high temperature range and at high wind speeds.

Other conclusions from the ANOVA are

- there is reduction of 7.0 μ g m⁻³ (95% confidence 6.0–8.0) in calculated PM₁₀ over the 1047 day monitoring period.
- an atmosphere of neutral stability in London seems to promote higher PM₁₀.

By removing each factor or covariate in turn, the model allows the relative importance of the causes of variation in PM_{10} to be assessed. TWD and its interaction with TWS, TWS itself and u^* in that order were shown to have the greatest contribution to explaining the variation in PM_{10} .

Acknowledgements

The authors acknowledge British Atmospheric Data Centre, Rutherford Laboratory, Harwell, London Borough of Greenwich, London Borough of Haringey, Royal Borough of Kensington & Chelsea, and K.R. Stribley for computer programming.

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