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A Lagrangian model of the evolution of the particulate size distribution of vehicular emissions

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Abstract

The emission inventory for London indicates that nearly 80% of the particulate emissions derive from vehicular sources. Most of this mass is in the form of ultrafine submicrometer particles which are of concern because of their influence on lung function. The prediction of their dispersion in the atmosphere coupled to the physical and chemical transformations which affect their size distribution and concentration are of great importance. This paper reports the first results from a new meso-scale Lagrangian model which follows the particulate emissions and the evolution of their size distribution across the city. The vehicular emissions are based on the published inventory, corrected to time of day, while other emissions are assumed steady. The initial size distributions of background and emitted particles are represented by the sum of three lognormal distributions. Meteorological data are derived from Meteorological Office reports and are preprocessed to obtain the hourly values of boundary layer depth, Monin–Obukov (MO) length, friction velocity, etc., needed for the computation of the vertical dispersion process via eddy diffusivities and the aerodynamic component of the dry deposition process. In the vertical direction, three layers are assumed—surface layer (typically 50 m), canopy layer and one further layer up to the prevailing boundary layer depth. Currently, the model includes wet and dry deposition and coagulation but not chemical reaction, nucleation or deliquescence. Trajectories are evolved for several hours across the city and the number size distributions and mass concentrations (PM10, PM2.5, PM1 and PM0.1) output at each step. This enables the vehicular contributions over and above the background concentration in each size range to be studied in detail. Data from the model have been compared with experimental data for one of the London background sites where particle number size distribution up to 450 nm (SMPS), plus PM10 and PM2.5 (TEOM) data are available.
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1. Introduction

Concerns about urban air pollution tend to be dominated by discussions of vehicular emissions and their effects. Both gaseous and particulate emissions can, in principle, lead to damage to human health, but in this paper, the focus will be exclusively towards urban particles. Vehicles can contribute a large proportion of urban particulate matter. For example, the emissions inventory for London suggests that vehicles contribute nearly 80% of the emissions of the city, with diesel vehicles contributing 67% and petrol

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vehicles 11.5% (Buckingham et al., 1997a). The figures for different cities vary quite significantly. Glasgow has nearly as high a vehicular contribution to PM10 emissions as London (73%; Buckingham et al., 1998), whilst areas such as Middlesborough, Swansea and Merseyside where there are large industrial emissions have vehicular contributions below 20% (Buckingham et al., 1997b, 1998). London is probably the UK city with the highest proportion of vehicular emissions.

Particle size is a key factor in determining the potential health effects. Fine particles penetrate more deeply into the lungs and cause irritation or more specific effects which could lead, for example, to cancer. Most of the particles emitted by vehicles are in the 'ultrafine range' < 100 nm (0.1 μm), and it is these which give rise to most concern (Seaton et al., 1995) although, as yet, the national requirements for air quality are expressed in terms of PM10 (Europe) or PM10 + PM2.5 (USA). The UK Expert Panel on Air Quality Standards (EPAQS, 2001) recently addressed the question as to whether the current PM10 standard should be supplemented or replaced by a fine particle mass or particle number standard but concluded that there was insufficient evidence on which to base such a change at the present time. The health effects have been reviewed in the UK by the Committee on Medical Effects of Air Pollutants (COMEAP, 1995, 1998, 2001), and their assessments have been used in the drawing up of Air Quality Standards and in the UK Air Quality Strategy (AQS, 2000). Proposals for new objectives for particles (PM10) to be achieved by 2010 have been published (AQS, 2001), which set a national target of 50 μg/m[3] 24 h average not to be exceeded more that seven times per year and an annual average of 20 μg/m[3]. However, slightly more lenient objectives have been set for London 50 μg/m[3] 24 h average not to be exceeded more that 10–14 times per year and an annual average of 23–25 μg/m[3]. Particulate pollution is worse in London than in other cities and this gives an added reason for seeking to understand the prevailing levels by modelling studies.

Vehicles also contribute to coarse particle concentrations by raising dust from the streets. This process is very poorly quantified and the amounts are excluded from emission inventories, as are the dusts that arise from general human activities such as building. Dusts from these sources contribute to PM10 and probably have their largest contribution in the 2.5–10-μm range. The proportion of mass in this size range in urban areas can be 20–40% (APEG, 1999; AQS, 2001). For example, at the Bloomsbury site in London, the annual average PM10 in 2000 was 21 μg/m[3] and the PM2.5 was 14 μg/m[3]—one third of the particulate mass is between 2.5 and 10 μm in this annual average. Because PM10 must be controlled under the UK National Air Quality Strategy and Local Air Quality Management plans, it is important that all sources of coarse particles should be considered including the vehicular contribution.

Clearly, the assessment and modelling of particulate pollution in urban areas must not be restricted to the total mass of particles—TSP or PM10—but must explicitly detail the particle size distribution and the physical processes which affect this distribution. The most commonly available models for dispersion and transport of pollutants in urban areas such as ADMS Urban (CERC, 1999) cannot adequately take into account particle size distributions and the size dependence of aerosol processes. Lagrangian trajectory models do not have this limitation, and therefore, a prime objective of this project was to develop a trajectory model which could be applied to the evolution of vehicular particulates emitted into a preexisting background aerosol. No attempt has been made to model street-canyon situations or the first few seconds of dilution in the vehicle wake. Rather, the approach addresses such questions as ''what peak particle number concentration may be reached in the city air away from the roadside?'', ''what changes take place in the transport of city centre vehicular emissions to the suburbs?'' or ''how persistent is the ultrafine mode downwind of the emissions?''.

A completely new computer program has been developed and is being applied to London. It provides a platform from which to study the combined effects of emissions, dispersion and transport, dry and wet deposition, coagulation, etc., as an air parcel moves across the city. It explicitly allows for the effects of particle size on all the processes. Output files give the particle mass concentration (as PM10, PM2.5, PM1 and PM0.1) at each point along the trajectory and the particle number in each of a large number of size fractions.

The work reported here examines the effects of the individual aerosol processes on the observed size dis-

tributions and mass concentrations and the diurnal variations of these parameters. The model allows comparison of the predicted results with the data obtained in London for PM10 (TEOM data) and the particle size obtained by the DEFRA (formerly DETR)-funded monitoring project using SMPS instruments.

2. Model description

2.1. Emissions

The emissions data are drawn from the published inventory for London (Buckingham et al., 1997a) giving annual mass emissions on a 1-km grid square basis, Fig. 1. Two categories of emissions—'vehicular' and 'other'—are considered separately. Vehicle emissions are assumed to be at ground level. Other emissions are assumed also to be near the surface, except for large point sources (arbitrarily taken as sources emitting over 10 tonnes per annum of particles) when the emissions are injected into the canopy layer (>50 m) rather than the surface layer (see below). There are only about 20 such sources in the London inventory. Vehicle emissions are adjusted for time of day, day of week, month of year. Time of day factors allow for central, inner and outer London cordons. Since trajectories extend over several hours,

the vehicular emissions factors vary with actual time of day during the run. 'Other' emissions are assumed to remain constant with time.

2.2. Size distributions

The size range is usually taken to be 10 to 10 000 nm (10 μm) diameter and is subdivided into (typically) 30 bands defined logarithmically ($\mathrm{dlog}D = 0.1$). Emissions and initial background size distributions are take to be the sum of 3 lognormal distributions, each described by a geometric mean diameter D_g and σ_g. The first mode represents ultrafine particles typical of vehicular emissions. The second is for accumulation mode particles. The third is for coarse mode particles. The vehicular, 'other' and background aerosols can be represented as a single mode or a mixture defined on a mass percent basis. The coarse mode can be introduced as an additional percentage of the vehicular emissions to represent resuspended road dust and/or be included in the background and 'other' emissions'. Mass concentration or mass emission values are converted to the corresponding number of particles. The lognormal distributions are truncated at the size limits (usually 10 and 10 000 nm) and the numbers scaled up to correct for particles which a lognormal distribution would place outside the limits, so that the correct mass to number conversion is achieved.
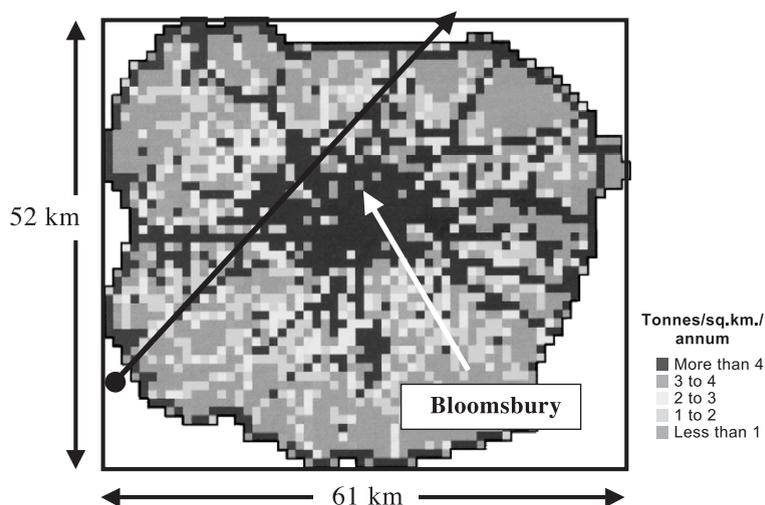

Fig. 1. The PM10 emissions inventory for London (1-km grid square).

2.3. Vertical structure

A four-layer structure is assumed. The lowest, surface layer is usually taken as 50 m, followed by a canopy layer (e.g., 100-m thick). The third layer stretches to the top of the boundary layer which is variable in height as determined from the meteorology. The boundary layer is allowed to drop to 150 m, thereby potentially eliminating layer 3, and in these circumstances, the particles within them disappear into the reservoir layer above. In situations of a rising boundary layer, reservoir air with a defined mass concentration is drawn in and layer 3 can be reestablished. Concentrations within each layer are assumed uniform.

The vertical dispersion is described by a simple transfer term between layers based on the eddy diffusivity K_z at the height of the layer interface and the concentration gradient estimated from the difference in concentration of the adjacent layers and the interlayer spacing. The K_z values are calculated using relations for neutral, stable and unstable conditions derived by Businger et al. (1971) which involve the friction velocity and the Monin–Obukov (MO) length.

2.4. Meteorology

Hourly met data is drawn from the Meteorological Office via the British Atmospheric Data Centre for one of the London sites (e.g., London Airport). This is then edited to exclude unwanted data and preprocessed to produce the necessary parameters for dispersion calculation. The preprocessor associated with ADMS 3 (CERC, 1999) has been used rather than writing a new routine. The algorithms follow those derived by Holtslag and Van Ulden (1983). The parameters include the boundary layer thickness, friction velocity and Monin–Obukov length. These hourly data are then linearly interpolated to the actual time of day in the trajectory. (MO length is interpolated on an inverse basis because it could go to infinity). As will be described below, considerable difficulties were encountered in obtaining realistic modelling results with this approach, and in some cases, typical values of the parameters for stable, neutral or unstable conditions were set rather than the parameters from the preprocessor.

2.5. Trajectories

As the program is currently written, the trajectories are either assumed to be linear or can follow the prevailing 10-m wind direction. Use of a numerically defined wind flow field from separate calculations could be envisaged in the future. The starting point for any trajectory is specified in terms of the ordnance-survey grid coordinates at an upwind point on or near the limit of the emissions inventory area (see Fig. 1). When linear trajectories are assumed, it is possible to ensure that the trajectory passes over a particular receptor point.

2.6. Background aerosol

The background PM10 mass concentration must be defined. This could come from a wider area model [e.g., using Met. Office NAME model] or based on the experimental data at rural sites adjacent to the city under study. For London, the only relevant PM10 sites deemed rural are at Rochester and Harwell. The background aerosol in layers 2, 3 and the reservoir will generally not be known, and the program allows these to be defined as a percentage (%) of the ground-level background (e.g., 100%, 80% and 50%). The background aerosol is usually assumed to be dominated by accumulation mode particles with smaller ultrafine and coarse contributions.

2.7. Dry deposition

Dry deposition is particle size-dependent and is described by deposition velocity v_d which is a combination of sedimentation velocity v_s and terms involving an aerodynamic resistance r_a (defined by the meteorology) and the laminar sublayer resistance r_b: $v_d = v_s + (r_a + r_b + r_a r_b v_s)^{-1}$ (Seinfeld and Pandis, 1996).

Sedimentation of coarse particles is allowed to contribute to transfer between layers 3 to 2 and 2 to 1.

2.8. Wet deposition

Wet deposition is particle size-dependent and increases with precipitation intensity which must be user-supplier (Seinfeld and Pandis, 1996).

2.9. Coagulation

The coagulation kernels $K(i,j)$ are calculated for all pairs of size bands i,j using conventional theory with the Fuchs gas kinetic correction factor in the form used by Ström et al. (1992) in their work on Brownian coagulation. When particles collide, the size of the aggregated particle is estimated using a user-defined Fractal Dimension rather than FD = 3 (liquid drop model). Because coagulation only affects ultrafine particles significantly, a value FD = 2, typical of diesel particles, is more appropriate (Gorbunov et al., 2002). Coagulation is only included in the surface layer closest to the emissions where the concentrations are at their highest.

Processes which are not currently included in the model are the effects of humidity on deliquescent particles, gas–surface reactions and nucleation from gaseous precursors (e.g., sulfuric acid, primary or secondary organics). These processes could be added at a later date, although they involve handing various types of aerosol in parallel, together with gas-phase species, whereas in the simple version of the model currently operated, a single type of aerosol is assumed.

2.10. Computation, input and output

Most of the input data required are held in separate files and only a few parameters are input from the terminal at run time. A typical setup file with the default values of the parameters used to obtain the results reported below is shown in the Appendix. After setting up the initial conditions, the trajectory is evolved with a time step of 30–60 s. The concentrations are solved using a forward linear approximation

$$c(\text{time } n+1) = c(n) + dt^*[dc(\text{time } n)/dt].$$

Improved numerical schemes could easily be incorporated but results showed virtually no change in output (maximum change along trajectory of 0.1 μg/m³) for change in time step to 10 or 90 s.

The output is directed to two files. One has time/location and mass concentration data for each layer at the end of each step (PM10, PM2.5, PM1.0 and PM0.1). The other has the particle numbers in each size band at each step. No graphical processing of the

data is incorporated into the programme but the files can be read into EXCEL or other packages for processing. For example, the number data can be plotted as number size distributions on a log–log scale or can be converted to mass distributions and then plotted.

3. Results and discussion

Fig. 2 shows the results for the mass concentrations of a typical run Jan 8, 2000 starting at 7 A.M. In 5 h, the trajectory covered about 45 km from southeast to northwest, passing over central London. The initial dip in PM10 is caused by dilution due to upward dispersion in the absence of significant emissions. The sharp rise after 30 km corresponds to an area of heavy traffic near Hyde Park. The PM10, PM2.5 and PM1 values track one another, and the PM0.1, starting from a very low background in this case, rises slowly to a few microgrammes per cubic metre. Overall, the gain in PM10 is only about 5 μg/m³.

The results are extremely sensitive to the assumptions made about the meteorology affecting vertical dispersion. Fig. 2 was actually run with assumed
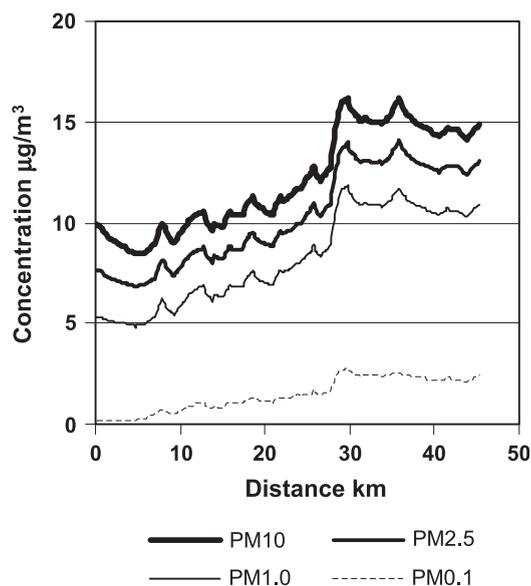

Fig. 2. Concentrations of PM10, PM2.5, PM1 and PM0.1 along a trajectory. Constant neutral stability assumed.

constant neutral stability conditions. Fig. 3 shows the same trajectory (same emissions, wind speed and direction) run with the data derived from the preprocessor (which are strongly stable with a low-boundary layer height for much of the trajectory but become neutral with elevated boundary layer later). Also shown are the data for neutral and slightly stable conditions. Unstable conditions would not occur on a mid-winter's morning. The corresponding parameters are neutral ($u^* = 0.5$, $1/LMO = 0.0001$, $H = 500$ m) and slightly stable ($u^* = 0.25$, $1/LMO = 0.005$, $H = 500$ m). The change is roughly equivalent to moving from Pasquill–Gifford stability class D to E. This change results in more than doubling of the additional PM10 concentration above background. The preprocessed actual meteorological data would suggest even larger increases in concentration early on but a rapid fall-off when the mixing depth increases and the stability becomes less stable in the middle of the day.

The effect of the assumptions about meteorological conditions at a particular receptor site can be seen in Fig. 4. Here trajectories have been started at one hour intervals through the day and the starting points chosen to ensure that the air mass passes over the Bloomsbury monitoring site (based on the initial wind
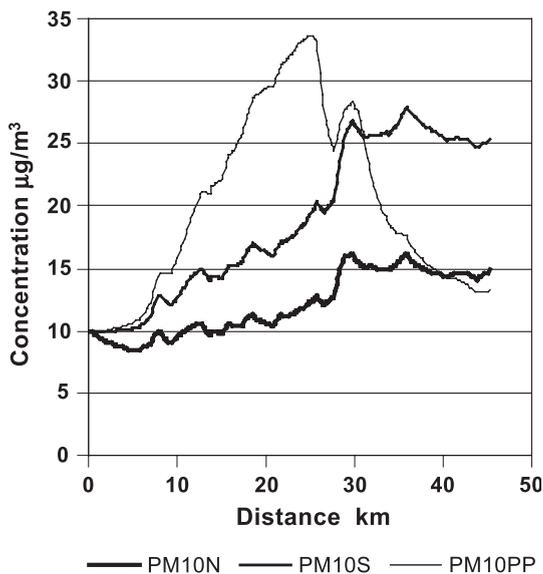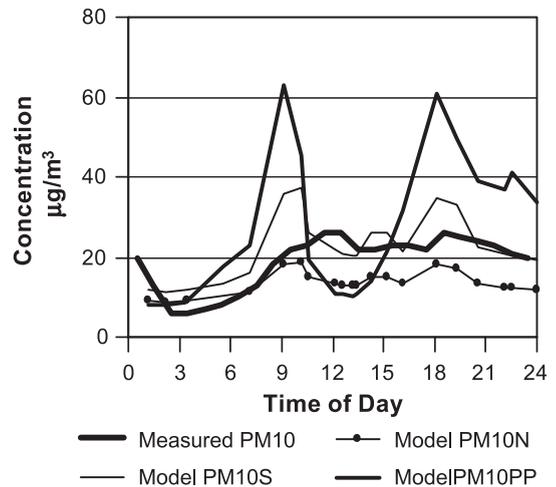

Fig. 4. Measured and estimated diurnal PM10 concentrations at Bloomsbury assuming neutral (N), slightly stable (S) and preprocessed actual hourly met data (PP).

direction). The measured PM10 concentrations for that day are shown together with model predictions with three different meteorological assumptions. The most realistic results are obtained by assuming constant, slightly stable conditions. Neutral conditions underestimate the actual concentrations. The preprocessed meteorological data leads to excessively high concentrations in the morning and early evening, with a sharp reduction near midday when dispersion is most effective. This graph summarises a general feature of our studies to date namely that dispersion over the city is most frequently best represented by nearly neutral stability and that the predictions made for supposedly very stable or very unstable conditions do not agree well with the measured experimental data. Clearly, the combination of preprocessor output and eddy diffusivity formulae that has been used so far does not give a good representation of the urban atmospheric conditions and needs to be completely reviewed.

Fig. 5 shows the diurnal variation of the various mass fractions for the slightly stable assumption which is in best agreement with the measured PM10. The spacing of the modelled points is not exactly hourly because each trajectory may have a different start location and wind speed conditions, and therefore, the air mass reaches the receptor location at different times after the start time. Unfortunately,
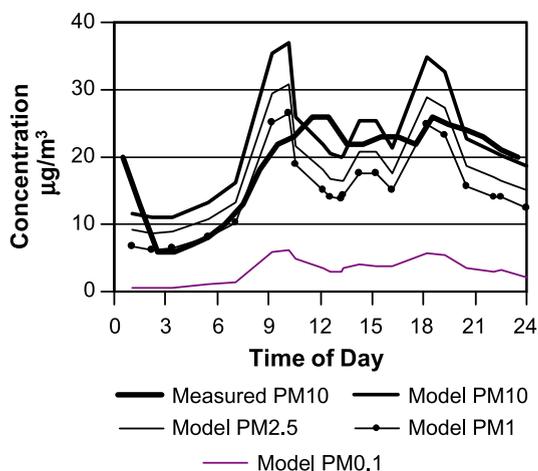

Fig. 3. PM10 concentrations along trajectory assuming neutral (N), slightly stable (S) and preprocessed actual hourly met data (PP).

Fig. 5. Measured and estimated particle mass fractions at Blooms-bury; slightly stable assumption.

although PM2.5 is monitored at this site, no data is available for this particular day. As mentioned above the annual averages at this site are for PM2.5 to be 66% of PM10, whereas in this model run, the proportion is slightly higher, around 80%.

Fig. 6 shows the effect of heavy rain on the particle concentrations (rainfall intensity, 10 mm/h) for the slightly stable assumption. The predominant effect is on the PM10 values because the scavenging coefficients are largest for large particles. Over the 5 h of this trajectory, the effect on PM2.5 is not discernible on the figure. Assumptions made in the model may
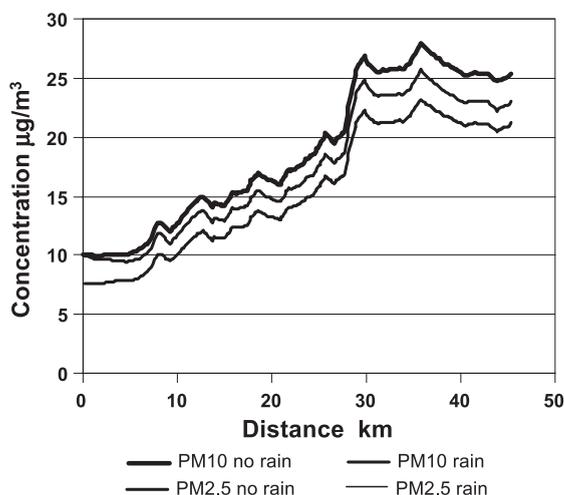
lead to an underestimate of the rate of particle scavenging. Water-soluble particles deliquesce and grow in size significantly at the 100% relative humidity conditions of rainfall and will then be more efficiently scavenged. The accumulation mode from 0.1 to 2.5 μm is often dominated by water-soluble particles such as sulphates and nitrates, whilst there will be a sea salt contribution in the coarse mode. Particle growth is not yet included in the model.

Fig. 7 shows that the effect of dry deposition is similar to that of rain, in that, coarse particles are most affected. The initial background concentration of the PM2.5 to the PM10 band is increased due to the coarse mode emissions included in the model. The final concentration of this band is less than it would otherwise be due to deposition. The effect on the particles below 2.5 μm is very small, not visible on the scale of the figure.

Fig. 8 shows the evolution of the size distribution by mass) with time along the trajectory. Fig. 8 shows mass concentration per size band; $dM/dlogD$ values are a factor of 10 higher because $dlogD = 0.1$. The three lognormal contributions are clearly visible. The ultrafine mode is the one which changes most markedly because this is where the bulk of the emissions occur. The peaks on the mass distribution are, of course, at larger diameters than the peaks of the number distribution which are the parameters set for the model (see Appendix). Note that half of the 'ultrafine' mode mass is above the PM0.1 size range (>100 nm) although the peak of the number distribution is only 60 nm.

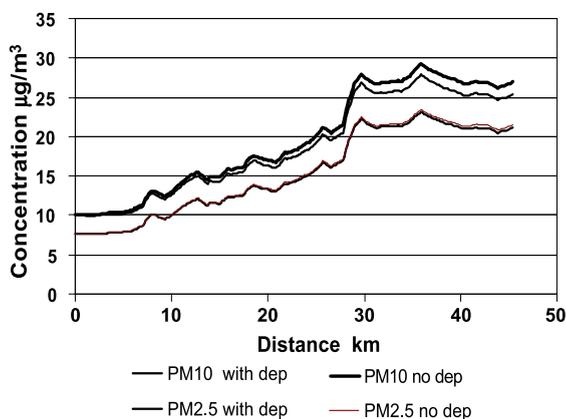Fig. 6. Effect of 10 mm/h of rain on estimated PM10 and PM2.5.

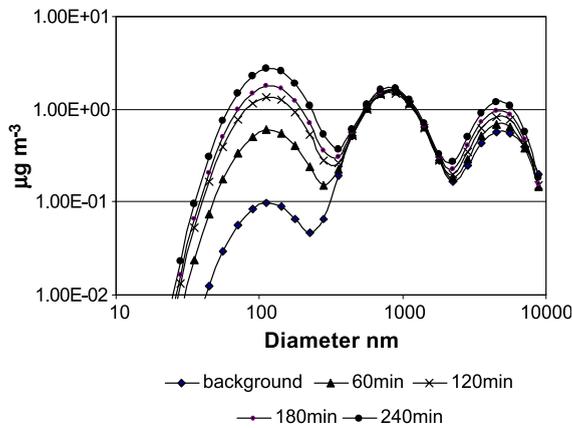Fig. 7. Effect of dry deposition on estimated PM10 and PM2.5.

Fig. 8. Size distribution by mass at various times along a trajectory.

Fig. 9 shows the size distributions by number (number concentration per size band, dN/dlog$D$ values 10 times higher). The particle number is dominated by the ultrafine mode particles derived from vehicles. The peak of the particle number distribution for this mode was taken to be 60 nm. This is rather smaller than the peak diameters found when monitoring diesel emissions where values of 70–90 nm are more common. The lower value was adopted partly to reflect the fact that we have a mix of diesel- and petrol-derived particles and that the particles from petrol engines are generally smaller than those from diesel engines (40–50 nm). The other factor is that the measured atmospheric particle size distributions in London frequently have peaks significantly smaller than the usual peak expected from diesel. This is
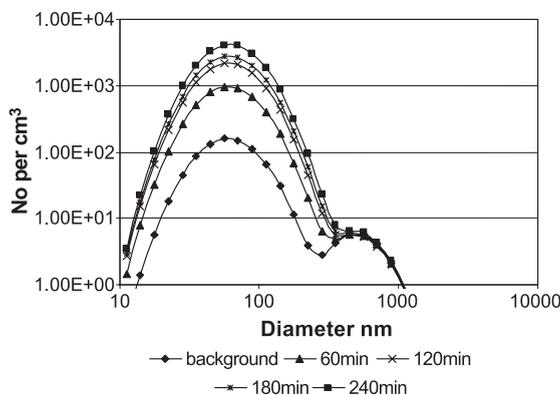
illustrated in Fig. 10 which shows the previous background and model data, together with the Bloomsbury SMPS data for 11 A.M., Jan 8, 2000. The experimental data have a much lower peak in the distribution (at about 30 nm) and the distribution is much flatter. To refine the model to give a better representation of the data, we have assumed our three modes to be defined as:

- Petrol: 20% of vehicular mass emissions with $D_g = 30$ nm, $\sigma_g = 1.8$,
- Diesel: 80% of vehicular mass emissions with $D_g = 100$ nm, $\sigma_g = 1.8$,
- Accumulation (background and nonvehicular emissions): with $D_g = 500$ nm, $\sigma_g = 1.6$.

The modelled result shown in the figure is in very good agreement with the experimental data, showing that the representation of the size distribution in this way is adequate for modelling purposes.

Runs with and without coagulation revealed only small differences in the size distributions. The particles most affected by coagulation are those below 50 nm but, as has been seen, the mass concentrations in this size range are very small—only a few $\mu g/m^3$. Depending on the relative concentrations in different modes, the predominant effect may be self-coagulation of ultrafine particles or scavenging of ultrafine particles by larger (accumulation mode particles). However, at the low concentrations in this model (less than 50 $\mu g/m^3$), neither effect has a major impact on the size distribution over the time scale of a few hours.

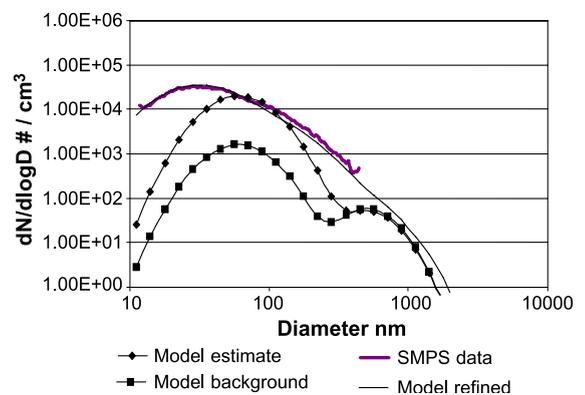Fig. 9. Size distribution by number at various times along a trajectory.

Fig. 10. Experimental and modelled size distributions for Bloomsbury.

For the trajectory with assumed slightly stable conditions (for which the PM10 concentrations are depicted in Fig. 3), the reductions in ultrafine mode particle numbers after the trajectory has evolved for 3 and 5 h are as follows:

Size band, %	Reduction in particle no.	
Mean diameter, nm	3 h	5 h
11.2	31	55
14.1	23	46
17.8	18	36
22.4	13	29
28.2	10	23
35.5	8	19
44.7	6	15

Total particle numbers are reduced by 5% after 3 h and 12 % after 5 h as a result of coagulation in this particular case where vehicle emissions are added to a low background concentration of 10 μg/m^3.

4. Conclusions

The model allows detailed analysis of the factors affecting particulate number and mass concentration in a large urban area for which the emissions inventory is available.

The dominant process affecting vehicular emissions is upward dispersion. Both coagulation and deposition have only small effects on the size distributions during mixing and dilution in the urban atmosphere.

Comparison with experimental size distribution data shows that the model needs to represent the vehicular emissions explicitly as a sum of the lower mass and smaller-sized petrol emissions together with the higher mass, larger-sized diesel emissions. A single lognormal distribution for vehicular particles is inadequate.

Coagulation is unimportant at the low concentrations (< 50 μg/m3) modelled but could be significant in the near field situation (tailpipe → street canyon) not covered by the model.

The model needs to be nested within a regional-scale model to provide appropriate background input.

The particular method of handling the urban meteorology in the model was found to be inadequate and requires revision.

Appendix A. Setup file and default parameters

Particle characteristics	
Lowest diameter (nm)	10
Highest diameter (nm)	10 000
No. of intervals	30
Fractal dimension	2

Log-normal Modes (1, ultrafine; 2, accumulation; 3, coarse)

Mode No.	Density (g/cm^3)	Diameter Dpg (nm)	Sigma
1	2	60	1.6
2	2	500	1.5
3	2	3000	1.5

Emissions

Percent in each mode based on inventory mass values (not restricted to total 100%)

	Mode 1	Mode 2	Mode 3
Background	5	70	25
Vehicular	100	0	20
Nonvehicular	0	80	20

Initial vertical concentration profile	
Canopy layer (% of background surface layer)	100
Upper layer (% of background)	80

Vertical structure	
Surface layer (m)	50
Canopy layer thickness (m)	100

Trajectory	
Length (h)	5
Time step (s)	60

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