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Original Article

Evaluation of an urban modelling system against three measurement campaigns in London and Birmingham

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ABSTRACT

The results of three measurement campaigns are presented in this study. The campaigns have been undertaken at an urban roadside site in London, for more than a year and three months in 2003–2004 and for a year in 2008, and at an urban background site in Birmingham, U.K, for about four months in 2002. The concentrations of $PM_{2.5}$, PM_{10}, NO_x and NO_2 were predicted using the roadside dispersion model CAR-FMI, combined with a national U.K. emission model, a meteorological pre-processor, and measured values at urban background stations. The agreement of the predicted and measured hourly and daily time-series has been assessed statistically for all of the campaigns and pollutants. For instance, the Indices of Agreement (IA) in all the campaigns ranged from 0.68 to 0.78, 0.87, from 0.70 to 0.80, and from 0.61 to 0.83 for $PM_{2.5}$, PM_{10}, NO_x and NO_2, respectively. However, in case of the campaigns in London, both the PM fractions and the nitrogen oxide concentrations were under-predicted. The model performance in terms of atmospheric stability, wind speeds and other factors was analysed, and reasons for the disagreement of predictions and measurements have been discussed. It is useful to consider the model performance statistics for several measurement campaigns simultaneously as some of the results were found to be specific only to one or two campaigns. The spatial concentration distribution of NO_x in London for 2008 has also been presented.

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1. Introduction

Health effects of airborne particulate matter are widely recognized. For example, Delamater et al. (2012) found that asthma hospitalization rate in Los Angeles County of California has a significant positive relationship with ambient levels of carbon monoxide, nitrogen dioxide and fine particulate matter ($PM_{2.5}$). Based on an analysis of 22 European cohorts within the multicentre ESCAPE project, Beelen et al. (2014) found that the long-term exposure to fine particulate air pollution was associated with natural-cause mortality. Their study identified a significantly increased hazard ratio (HR) for $PM_{2.5}$ of 1.07 (95% CI 1.02–1.13) per 5 $\mu g/m^3$.

Particulate matter (PM) may be formed via gas-to-particle conversion, and is constantly transformed and depleted during atmospheric transport by various physical and chemical processes, including coagulation, condensation and evaporation, chemical transformation, and dry and wet deposition (for example, Pohjola et al., 2003; Ketzel and Berkowicz, 2004). Based on the 419 source apportionment studies undertaken after 1990 using the data monitored at sites across the world (except rural and remote sites), Karagulian et al. (2015) identified that from global averages of source contributions, 25% of urban ambient air $PM_{2.5}$ was contributed by vehicular traffic. DEFRA (2013) identifies that modelling of $PM_{2.5}$ remains a substantial challenge owing to uncertainties in and lack of measured data, understanding of the dynamic, physical and chemical processes, and uncertainties in the

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emission data and their projections. In many major European cities, no reliable PM emission inventories are available. In particular, the formation of vehicle non-exhaust emissions that originate from break, tyre, and road wear and that from resuspension of dust is poorly understood (for example, Kauhaniemi et al., 2011, 2014).

Stocker and Carruthers (2007) reported that nearly 24% of the $PM_{2.5}$ concentrations in the UK was contributed by brake, tyre and road wear. At roadside locations in urban areas, the $PM_{2.5}$ fraction was found to be as high as 90% of the PM_{10} fraction, whilst a lower 57% of PM_{10} fraction was found at other UK locations (Williams et al., 2006). Regarding the secondary PM components, this fraction is higher, as expected.

The available monitoring data in the UK indicates that regional background is responsible for 60–80% of the urban background concentrations of $PM_{2.5}$ in southern England (DEFRA, 2013). The primary emissions from road traffic, including the non-exhaust component, are responsible for a significant (about 30–50%) contribution to the urban background increment of $PM_{2.5}$ above rural concentrations in the UK. Williams et al. (2006) evaluated using source apportionment that the percentage of $PM_{2.5}$ in PM_{10} was up to 90% in elemental carbon component. Barlow et al. (2007) identified that non-exhaust processes are an important source of particulate matter in the U.K., constituting approximately 25%, 50% of and 90% of the emissions of $PM_{2.5}$, PM_{10} and coarse PM, respectively. Neglecting the non-exhaust emissions of $PM_{2.5}$ in the U.K. would therefore cause an under-prediction of the local contribution of approximately one quarter (Stocker and Carruthers, 2007; Barlow et al., 2007).

The aims of this study are (i) to present two air quality measurement campaigns in London and one in Birmingham, and (ii) to evaluate the performance of an urban modelling system against the results of these campaigns. In particular, an evaluation and analysis of model performance against three campaigns in two cities allows one to draw more general conclusions. A new particulate matter monitoring system that can measure aerosol number and mass size distributions in the ultrafine to coarse size ranges (called Ambicount) was also described.

2. Materials and methods

2.1. Experimental methods and data

2.1.1. The measurement campaigns

This study addresses the results from three targeted measurement campaigns at two urban locations in the UK. Maps of the measurements sites have been presented in Figs. 1 and 2. These specific locations were selected as they represent (i) in case of London, a fairly polluted urban environment, intensively influenced by the pollution originated from vehicular traffic, and (ii) in case of Birmingham, urban background concentrations within an urban centre, i.e., the exposure of population to air pollution in a fairly wide urban region. There is also a wide range of air pollutant measurements at both sites.

The site of Cromwell Road is located in the Wildlife Garden of the Natural History Museum, London, near a densely trafficked junction of streets. The site is designated as a roadside site as per the UK Site Classification; this is equivalent to 'urban, traffic' type, as referred by the European Directive on ambient air quality. The site of Centenary Square is located within the pedestrianised area of the Birmingham city centre. This site was designated as an urban centre site as per the UK Site Classification; this is equivalent to 'urban background', as defined in the Directive 2008/50/EC (2008).

The measurement sites were operated and maintained by the respective UK city councils, Royal Borough of Kensington and Chelsea in case of London, and Birmingham City Council. The traffic count data was also provided by the local councils. Selected key characteristics of the measurement campaigns are provided in Table 1. Further details of the measurement campaigns are discussed later in this section.

2.1.2. Measurement instruments

Several different instruments were used to measure PM at the various sites as described in Table 1.

The condensation particle counter TSI CPC 3022 system has a lower detection limit of 7 nm. The inlet of TSI CPC was connected to a copper tube (7 mm internal diameter) extended to about 1.5 m long (1.0 m inside cabin and 0.5 m outside above roof with a bend). This instrument was installed and maintained by Birmingham City Council and Bureau Veritas (formerly Stanger Science and Environment), U.K.

Ambicount is a near real-time particle monitoring system developed by the University of Hertfordshire and Casella CEL Ltd. Ambient air is sampled through a sharp-cut cyclone with a 10 μm cut-off enabling sampling of the PM_{10} fraction. The flow rate has been set at 5.0 L per minute. The instrument incorporates an Optical Particle Counter (OPC) and a Condensation Particle Counter (CPC). Both OPC and CPC sub-sample a 10 ml per minute flow isokinetically from the main sample air stream. This enables to count particles in the whole size range from 10 nm to 10 μm, in four size fractions of 10 nm–360 nm; 500 nm–1.0 μm; 1.0 μm–2.5 μm; and 2.5 μm–10 μm, at a resolution of 15 s. The remainder of the 5.0 L per minute sample flow passes through filter, which collects the particles, enabling gravimetric analysis of the PM_{10} fraction.

Ambicount measured particle number concentrations have been converted to particle mass concentrations using an apparent density of particles calculated by dividing the measured average particle mass concentration by the average particle volume concentration.

Performance evaluation of Ambicount has been presented by Kuhn et al. (2003), Srimath et al. (2003) and Srimath (2006).

Partisol is a Rupprecht and Patashnick (R&P) gravimetric particle sampling system fitted with a PM_{10} or $PM_{2.5}$ inlet head. Partisol sampled ambient air at a rate of 16.67 L per minute. This instrument was installed and maintained by Birmingham City Council and Bureau Veritas, U.K.

During a separate monitoring systems comparison campaign carried out in Hatfield, UK (Srimath (2006)), the PM_{10} concentrations gravimetrically measured by Ambicount were found to be well correlated with those measured by Partisol. The outcomes of this campaign are presented in Appendix A of this manuscript.

TEOM (Tapered Element Oscillating Microscope) Rupprecht and Patashnick (R&P) Particle Counter fitted with a PM_{10} or $PM_{2.5}$ inlet head was used. TEOM sampled ambient air at a rate of 16.67 L per minute. This version of TEOM preheats the air sample to eliminate interference from water molecules. However, in doing so, TEOM evaporates volatile component of particles sampled and hence correction factors need to be applied to derive true particle mass concentration. This instrument was installed and maintained by Birmingham City Council and Bureau Veritas, U.K.

NO_x analyser instrument measures nitrogen oxides (NO_x) continuously by chemiluminescence method. Ozone analyser measures ozone continuously by ultraviolet absorption method.

2.1.3. Details of the measurement campaigns in London

The site is adjacent to the traffic light controlled junction of two major roads, Cromwell Road and Queen's Gate. The curb of the nearest road, Queen's Gate is at a distance of 5 m from the site. The traffic flows on the nearest busy roads, Queen's Gate and Cromwell Road, were 25 000 and 45 000 vehicles per day, respectively. The measurement height is 2.0 m.

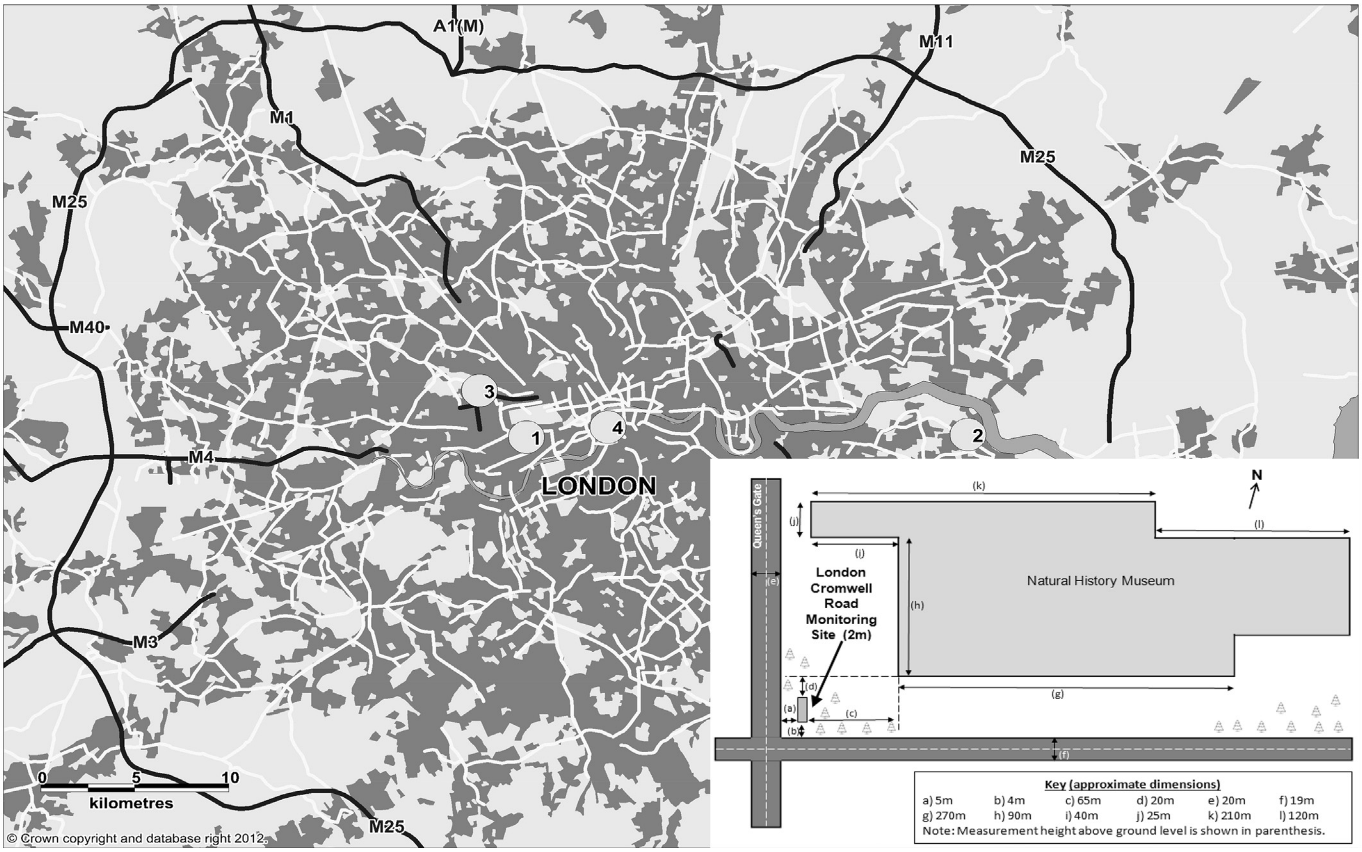


Fig. 1. Locations of the monitoring stations that were used in both of the measurement campaigns in London. Numbers indicated within the circles identify the following monitoring stations: (1) the roadside air quality measurement station at Cromwell Road, (2) the meteorological station, London Weather Centre (in eastern London), (3)the urban background air quality monitoring station at North Kensington, and (4) the urban background air quality monitoring station (in case of $PM_{2.5}$) at Belvedere. The average height of the Natural History Museum is 12 m.

As part of the present study, Ambicount has been deployed at the site along with another TEOM for measuring $PM_{2.5}$ and a meteorological station to measure wind speed and direction. The nearest urban background site for this measurement site is North Kensington, which is approximately at a distance of 3.7 km from the site. Monitored pollutants at this background site include PM_{10}, CO, O_3, NO_2 and SO_2. However, $PM_{2.5}$ is not measured at this background site. The nearest urban background site monitoring for $PM_{2.5}$ is Belvedere, which is approximately at a distance of 23.4 km from the Cromwell Road site. Therefore the urban background concentrations measured at North Kensington were used for all the other pollutants, except for $PM_{2.5}$, the urban background of which was extracted from the data of the station at Belvedere.

The meteorological data from the site of London Weather Centre (LWC) were used, and the global radiation data were extracted from the British Atmospheric Data Centre.

2.1.4. *Details of the measurement campaign in Birmingham*

The nearest busy urban street, Broad Street, is approximately at a distance of 60 m from the station. The traffic flow on Broad Street was 54 000 vehicles per day. The busy motorway M6 is at a radial distance of 4 km in the northerly to northeasterly direction from this site. The surrounding area is generally open and comprises urban retail and business outlets.

This site contains also TSI CPC to count total particle number concentration in the size range of 7 nm to 1 μm. Ambicount was co-located with the above instruments. The inlet head of Ambicount was kept at approximately the same level as that of the inlets of other instruments, i.e., 3.5 m above ground level. The urban background concentrations of NO_x, NO_2 and O_3 were obtained from the urban background site of Birmingham East, located at a distance of 5.5 km from the site of Centenary Square. The urban background concentrations of $PM_{2.5}$ were measured at the urban background site of Hodgehill, situated at a distance of 6.3 km from the site. Ambicount has been deployed to measure $PM_{2.5}$ concentrations.

The meteorological parameters (wind speed, wind direction, surface temperature, atmospheric pressure, precipitation) were obtained from the British Atmospheric Data Centre, UK, measured at the Elmdon station.

2.1.5. *Data quality assurance and quality control*

The following data quality assurance and control (QA/QC) procedures were followed during each of the field measurement campaigns. These are listed for each of the instruments deployed.

Ambicount and its inlet head have been calibrated separately. Ambicount calibration procedures have been presented by Kuhn et al. (2003). Flow calibration was conducted on the day of instrumentation setup and every time on the day of data download, once in a month. Ambicount counted particles every second; but for deriving hourly averages, only those datasets available for the whole hour were averaged. Similarly, flow and temperature data logged to the Ambicount data logger was checked and any abnormal data was excluded from the data analyses. Pre-weighed filters were used in Ambicount; the filters have been conditioned, weighed using a balance with sensitivity of 1 μg.

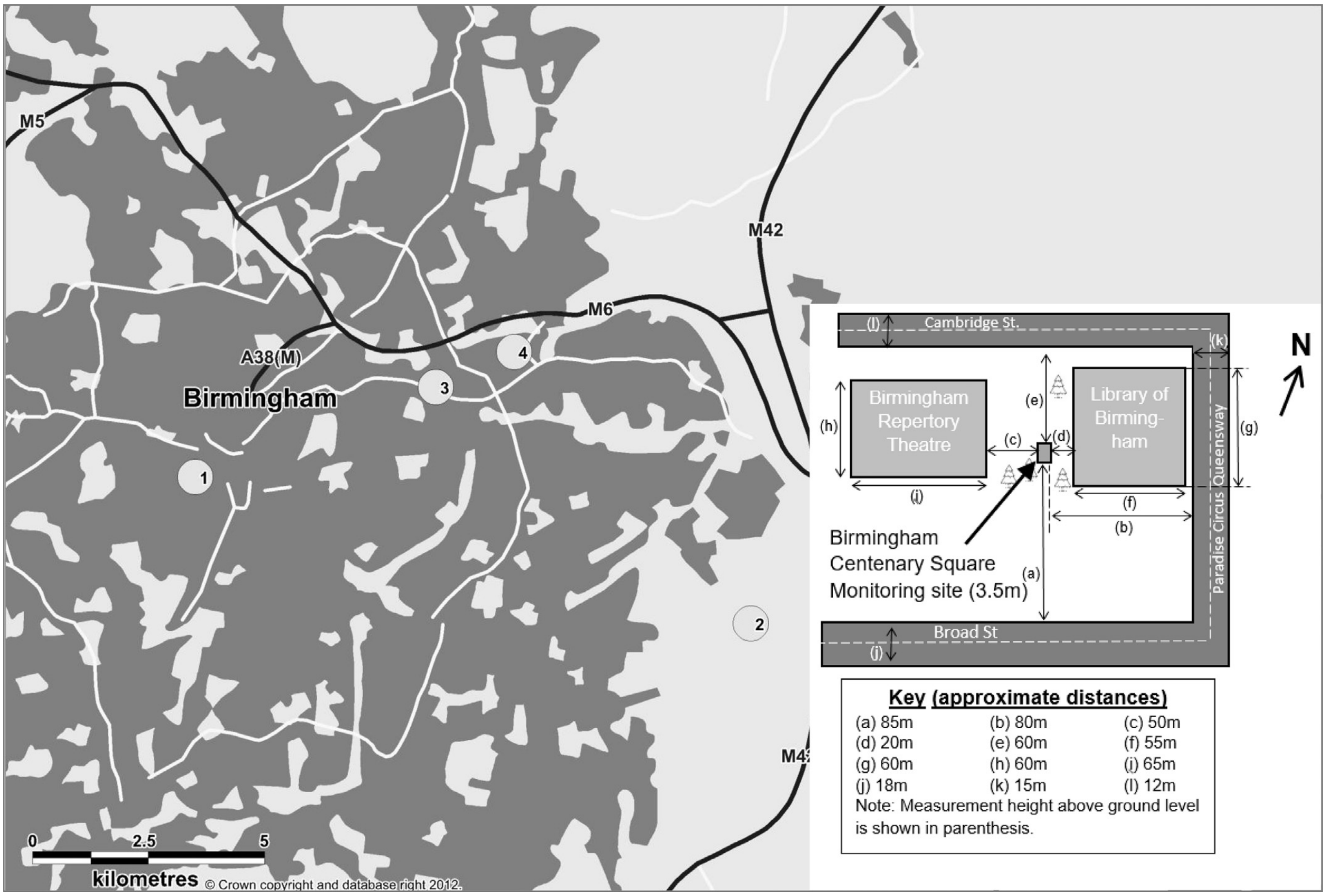


Fig. 2. Locations of the monitoring stations that were used in the measurement campaigns in Birmingham. Numbers indicated within the circles identify the following monitoring stations: (1) the urban centre air quality measurement station at Centenary Square, (2) the meteorological station, Elmdon Weather Centre, (3) the urban background air quality monitoring station, Birmingham East and (4) the urban background monitoring station (in case of $PM_{2.5}$) at Hodgehill. The average heights of the Birmingham Repertory Theatre and the Library of Birmingham are 10 m and 15 m, respectively.

A TEOM real-time analyser, which preheats the air sample to 50 °C was used. Before deploying TEOM real-time analyser, flow calibration and leak test were conducted. Onsite flow audit was conducted every time on the day of data download. In addition, parameters such as main flow, bypass flow and temperatures were checked against the operational ranges and any abnormal data was removed. Ratified hourly PM_{10} mass concentrations recorded by TEOM, as published by the Department for Food and Rural Affairs, were used in the analyses of the 2008 data at the Cromwell Road site, and hence the correction factor to account for the loss of volatile component of the particulate matter was already accounted.

The hourly average $PM_{2.5}$ concentrations measured by TEOM were used in the data analyses without using any correction factor for the loss of volatile component of the particulate matter. This is likely to result in the understatement of $PM_{2.5}$ concentrations. The data analyses were also undertaken with Ambicount measured hourly average $PM_{2.5}$ concentrations. No preheating of the sample is done by Ambicount and hence no loss of volatile component of

Table 1
Summary information of the measurement campaigns in London and Birmingham.

City and the name of station	EU classi-fication	Duration of measurements	Instruments in operation	Urban background sites	Meteorological data
London, Cromwell Road	Urban roadside	From 15.7.2003 to 2.11.2004 (One year, three months and 19 days)	Ambicount, TEOM for $PM_{2.5}$, TEOM for PM_{10}, gaseous pollutant analysers (NO_x, NO_2, CO, SO_2).	North Kensington (NO_x, NO_2, O_3, CO, SO_2) and Belvedere ($PM_{2.5}$)	London Weather Centre, measured on site, and British Atmospheric Data Centre (global radiation)
	Urban roadside	From 1.1.2008 to 31.12.2008 (One year)	TEOM for PM_{10}, gaseous pollutant analysers (NO_x, NO_2)	North Kensington (PM_{10}, NO_x, NO_2)	London Weather Centre
Birmingham, Centenary Square	Urban back-ground	From 30.4.2002 to 10.9.2002 (4 months and 11 days)	Ambicount, TEOM for PM_{10}, Partisol for $PM_{2.5}$ and PM_{10}, TSI- CPC for particle number concentration, gaseous pollutant analysers (NO_x, NO_2, O_3, CO, SO_2).	Birmingham East (NO_x, NO_2 and O_3) and Hodgehill ($PM_{2.5}$)	British Atmospheric Data Centre, measured at the station of Elmdon

the particulate matter is anticipated in the Ambicount measurements.

Flow and leak tests were conducted before deploying Partisol gravimetric analyser. Onsite flow audit was conducted once in a week.

Only those datasets were used for data analyses, for which readings from all the instruments were available.

2.2. The modelling of traffic flows, emissions and atmospheric dispersion

2.2.1. Meteorological pre-processing model

The atmospheric dispersion model, CAR-FMI requires a range of hourly meteorological parameters, including wind speed, wind direction, solar radiation, friction velocity and Monin-Obukhov length. These were provided by the meteorological pre-processor GAMMA-MET, described by Bualert (2002).

The meteorological pre-processor needs the following input parameters: wind speed, wind direction, ambient temperature, cloud cover and global radiation. The model is then used to evaluate atmospheric stability parameters, including the Monin-Obukhov lengths and mixing heights. The effects of land use characteristics on parameters, such as surface roughness, Bowen ratio, albedo and anthropogenic heat flux, are taken into account.

2.2.2. Evaluation of the traffic flows and vehicular emissions

The traffic flow data were provided by the Royal Borough of Kensington and Chelsea, and Birmingham City Council. These data have been computed using traffic planning models, calibrated according to measured traffic flow values. Traffic classification, speed, and temporal traffic profiles were taken from the Department for Environment, Food and Rural Affairs (DEFRA), U.K.

Two kinds of road and street inventories were used in this study. For the computations in London in 2008, a network of 63726 road links in London was used, which includes motorways, and major and minor roads. The emission factors and functions to calculate the vehicular emissions are based on the 2008 London Atmospheric Emissions Inventory (LAEI). For the computations in London in 2003–2004, and for those in Birmingham, more limited emission inventories were used within a square area of 1×1 km^2, centred on the measurement sites. The vehicular emissions were computed using the emission factors and functions in the National Atmospheric Emissions Inventory (NAEI, 2003).

Both kinds of inventories included the emissions from vehicular traffic, for the network of roads and streets within the area. The influence on concentrations near the ground level originated from all the other local sources (such as marine traffic, major stationary sources and small-scale combustion) is substantially smaller, compared with that of vehicular emissions.

The vehicular $PM_{2.5}$ emissions due to tyre and brake -wear have also been included from National Atmospheric Emissions Inventory (NAEI) for the computations in London for 2008, but not for the computations for the other two campaigns. The resuspension of material deposited on the road surface has not been taken into account.

2.2.3. Atmospheric dispersion modelling

The atmospheric dispersion of vehicular emissions has been evaluated using a roadside dispersion model, CAR-FMI (Kukkonen et al., 2001a,b; Öttl et al., 2001; Härkönen, 2002; Levitin et al., 2005). The dispersion equation is based on a semi-analytic solution of the Gaussian dilution equation for a finite line source (Luhar and Patil, 1989). CAR-FMI model requires five categories of input: (i) details of roads in the area of interest, (ii) temporal traffic profiles, (iii) emission factors for the pollutants to be studied, (iv) meteorological parameters at the site and (v) background pollutant concentrations for the pollutants of interest.

The dispersion parameters are modelled as a function of the Monin-Obukhov length, the friction velocity and the mixing height (Gryning et al., 1987); these quantities are computed by the GAMMA-MET model. Traffic-originated turbulence is modelled with a semi-empirical treatment (Petersen, 1980). The model includes a treatment for the basic reactions of nitrogen oxides, oxygen and ozone, using a receptor-oriented discrete parcel method, and the dry deposition of the fine particles.

Two kinds of atmospheric dispersion computations were performed in this study. For the computations in London for 2008, the vehicular emissions originating from an extensive network of streets and roads in the whole of London were included. The OSCAR Air Quality Assessment System (Sokhi et al., 2008) has been set up to model traffic-related $PM_{2.5}$ at high resolution near the whole road network across London for the year 2008. The models within the OSCAR system consist of an emission model, a meteorological preprocessing model, and a line source Gaussian dispersion model (CAR-FMI). Singh et al. (2014) have previously presented in detail the domain, the applied road and street network and the computational method. The OSCAR system calculates emissions from line sources to predict hourly concentration at the defined high resolution receptor points placed at varying distance of 10, 40, and 90 m near both sides of the roads, and with 100 m distance apart in outskirts.

Regional and urban background levels were taken from Grice et al. (2009). These contain regional and urban background annual mean concentrations on a 1-km $\times$ 1-km grid resolution in 2008 for the whole of the United Kingdom. The background concentrations were calculated using the National Atmospheric Emissions Inventory (Dore et al., 2008). This includes regional and urban emissions from domestic heating, agriculture, combustion, and processes in industries, construction, energy production, quarries, solvents, waste, shipping, and aviation. Also, secondary $PM_{2.5}$ is included. Urban traffic increments in London were excluded from these background values to avoid double counting.

For the computations in London for 2003–2004, and those for Birmingham, a simpler modelling set-up was used. A model domain of 1×1 km^2, centred at the measurement location was used. The predicted concentration was then assumed to be equal to the combined contribution from all the sources in the above mentioned domain, added to the measured urban background concentration. The concentrations were computed at the two monitoring sites, and in addition, at receptors spaced at regular intervals of 50 m, covering the respective modelling domains.

3. Results

First, the street increments, i.e., the differences of roadside and urban background concentrations were analysed. This is useful for methodological reasons. Second, statistical parameters were defined for the comparison of predictions and measurements. Third, the differences of model predictions and measured data were analysed.

Two kinds of dispersion computations were performed. For the computations in London for 2008, the vehicular emissions originated from an extensive network of streets and roads in the whole of London were included. For the computations in London for 2003–2004, and those for Birmingham, a simpler modelling set-up was used. For the simpler computations, the predicted concentrations were computed as a sum of (i) the contribution from vehicular sources in the vicinity of the site (defined here as an area of 1 km^2) and (ii) the measured urban background concentration.

The simpler approach was adopted for part of the computations, as it is much less resource-consuming. However, that approach has two significant limitations: (i) the definition of the computational domain is somewhat arbitrary, and (ii) the modelling relies on the proper representativity of the urban background measurements.

3.1. The street increments

For methodological purposes, it is therefore useful to analyse, how much higher the measured roadside concentrations are, compared with the corresponding urban background values. The average values of the concentrations at the urban background and roadside sites are shown in Table 2. A normalised roadside increment was defined as the difference of the measured roadside and urban background values, divided by the measured concentration at the urban background site. Clearly, all such increments should be positive, to be physically reasonable.

The normalised roadside increments were as follows: in London, 0.59, 1.0 and 1.8–2.0, for $PM_{2.5}$, PM_{10} and for NO_x, respectively, and in Birmingham, 0.17 and 0.51, for $PM_{2.5}$ and NO_x, respectively. In both cities, the roadside increments were smaller in case of $PM_{2.5}$, compared with the corresponding increment for NO_x, due to the more significant regional background for fine PM. Clearly, the exact values of these increments also depend on the selection of the urban background and street stations. The increments for both $PM_{2.5}$ and NO_x were substantially smaller in case of Birmingham, compared with the corresponding values in London. This is probably mainly due to the location of the selected urban background stations in closer vicinity of the local traffic in Birmingham, compared with the selected corresponding urban background stations in London.

3.2. Statistical parameters for the comparison of predictions and measurements

Five statistical parameters were computed: the index of agreement (IA), the coefficient of determination (R^2), the factor-of–two (F2), the fractional bias (FB) and the root mean square error (RMSE). The parameters IA, R^2 and RMSE are measures of the correlation of the predicted and observed time series of concentrations, while FB is a measure of the agreement of the mean concentrations. F2 is a measure of how many predictions are within a factor of two compared with the observations.

The index of agreement (IA) varies from 0.0 (theoretical minimum) to 1.0 (perfect agreement between the observed and predicted values) and is calculated using the formula stated below.

$$IA = 1 - \left[\frac{\sum_{i=1}^{n} (x_i - y_i)^2}{\sum_{i=1}^{n} \left[\left| x_i^1 \right| + \left| y_i^1 \right| \right]^2} \right]$$

where n is the number of data points, and x and y refer to the predicted and measured pollutant concentrations, respectively;

$x_i^1 = x_i - \bar{x}$; and

$y_i^1 = y_i - \bar{y}$

where the overbar refers to an average value.

However, it can be shown numerically that even for a predicted random distribution of concentration values, extending from zero to twice the measured average concentration, the IA value will be approximately 0.40 (Karppinen et al., 2000). This implies that all reasonable models should produce IA values that are higher than approximately 0.40.

Fractional bias (FB) ranges from −2.0 to + 2.0 in cases of extreme under- and over-prediction, respectively. Values of the FB that are equal to −0.67 and + 0.67 are equivalent to under-and over-prediction by a factor of two, respectively.

3.3. Evaluation of the model predictions against the measured data

Ambicount measured hourly average concentrations were used for comparing the predicted hourly average $PM_{2.5}$ concentrations at all measurement sites. Ambicount measured daily average concentrations in 2004 were used to compare with the predicted daily average $PM_{2.5}$ concentrations at the Cromwell Road roadside site.

Partisol measured daily average $PM_{2.5}$ concentrations were used to compare the daily average predicted $PM_{2.5}$ concentrations at the Birmingham Urban Centre site. For comparing the predicted and the measured hourly and daily average PM_{10} concentrations in 2008 at Cromwell Road roadside site, TEOM measured concentrations were used.

Comparison of the predicted hourly average $PM_{2.5}$ concentrations at the Cromwell Road site in 2004 was also made using the TEOM measured hourly average $PM_{2.5}$ concentrations. As no correction for the volatile component of the particulate matter was made in the TEOM measured $PM_{2.5}$ concentrations, the differences between the predicted and the TEOM measured $PM_{2.5}$ concentrations are partly attributed to the loss of volatile matter in the measurements.

Table 2
Statistical analysis of the observed (Co) and predicted (Cp) daily and hourly concentrations of PM_{10}, NO_x and NO_2 at Cromwell Road site in London for 2008. The observed values were measured using TEOM. C_{bg} is the measured urban background concentration.

Statistical parameter	PM_{10}		NO_x		NO_2	
	Daily	Hourly	Daily	Hourly	Daily	Hourly
Average C_{bg} ($\mu g\ m^{-3}$)	20.3	20.1	50.1	50.2	33.6	33.4
Average C_o ($\mu g\ m^{-3}$)	28.8	28.7	150.8	151.3	68.0	68.2
Average C_p ($\mu g\ m^{-3}$)	23.1	23.0	94.8	95.2	52.7	52.6
Index of agreement (IA)	0.91	0.87	0.68	0.70	0.63	0.61
Coefficient of determination (R^2)	0.87	0.68	0.50	0.36	0.28	0.21
Factor-of-two (F2; %)	99	90	63	50	86	72
Fractional bias (FB)	0.22	0.22	0.46	0.46	0.25	0.26
Root Mean Square Error (RMSE) ($\mu g\ m^{-3}$)	7.1	10.3	70.9	96.5	24.9	36.8
Number of data points (N)	363	8400	291	6392	307	6770

3.3.1. *Statistical analysis of the agreement of the predicted and measured data*

The above mentioned statistical parameters were computed for three pollutants for each campaign.

The statistical parameters for the PM_{10}, NO_x and NO_2 concentrations in Cromwell Road, London in 2008 are presented in Table 2. Example scatter plots of the predicted and measured concentrations are presented in Figs. 3 and 4, in case of Cromwell Road, London for 2008 and Centenary Square, Birmingham for 2002.

The evaluation of model performance was focused on the PM fractions and nitrogen oxides. The model performance for the O_3 concentrations was also computed. However, the detailed results for O_3 have not been presented here, as predicting ozone was not the main focus of this study. In summary, the model predictions for O_3 were fairly good both in terms of the temporal variation and the agreement of the average concentrations.

The scatter plots presented in Fig. 3 identify that the concentrations of PM_{10}, NO_x and NO_2 were slightly under-predicted; however, the majority of the predicted concentrations are within a factor-of-two of the measured concentrations. For instance, there were approximately ten cases, in which the NO_2 concentrations were substantially over-predicted (i.e., the highest predicted values of NO_2). A more detailed analysis shows that these cases are associated with very low wind speed or calm conditions (less than 0.6 m/s) under stable atmospheric stratification (the inverse of Monin-Obukhov length was of the order of 0.02 m^{-1}).

The scatter plots presented in Fig. 4 show no systematic under- or over-prediction of pollutant concentrations for the majority of the predicted concentrations. However, the model under-predicted especially on some occasions, in which the measured $PM_{2.5}$ concentrations were in excess of 100 $\mu g/m^3$. A more detailed analysis shows that these cases correspond to very low traffic counts (less

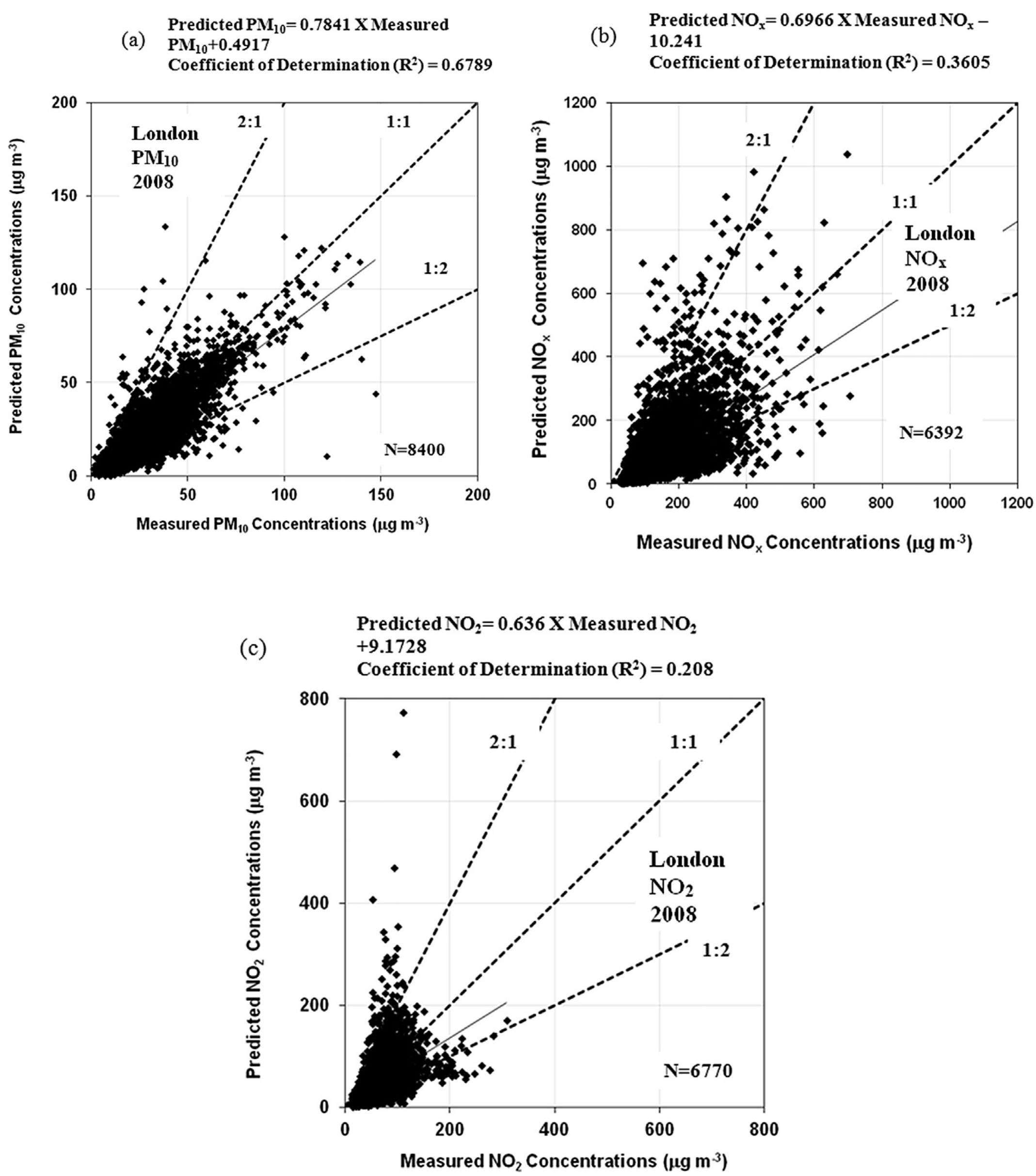


Fig. 3. a–c. Scatter plots of the predicted and measured hourly concentrations of PM_{10} (upper left panel), NO_X (upper right) and NO_2 (lower panel) at the Cromwell Road Site in London in 2008.

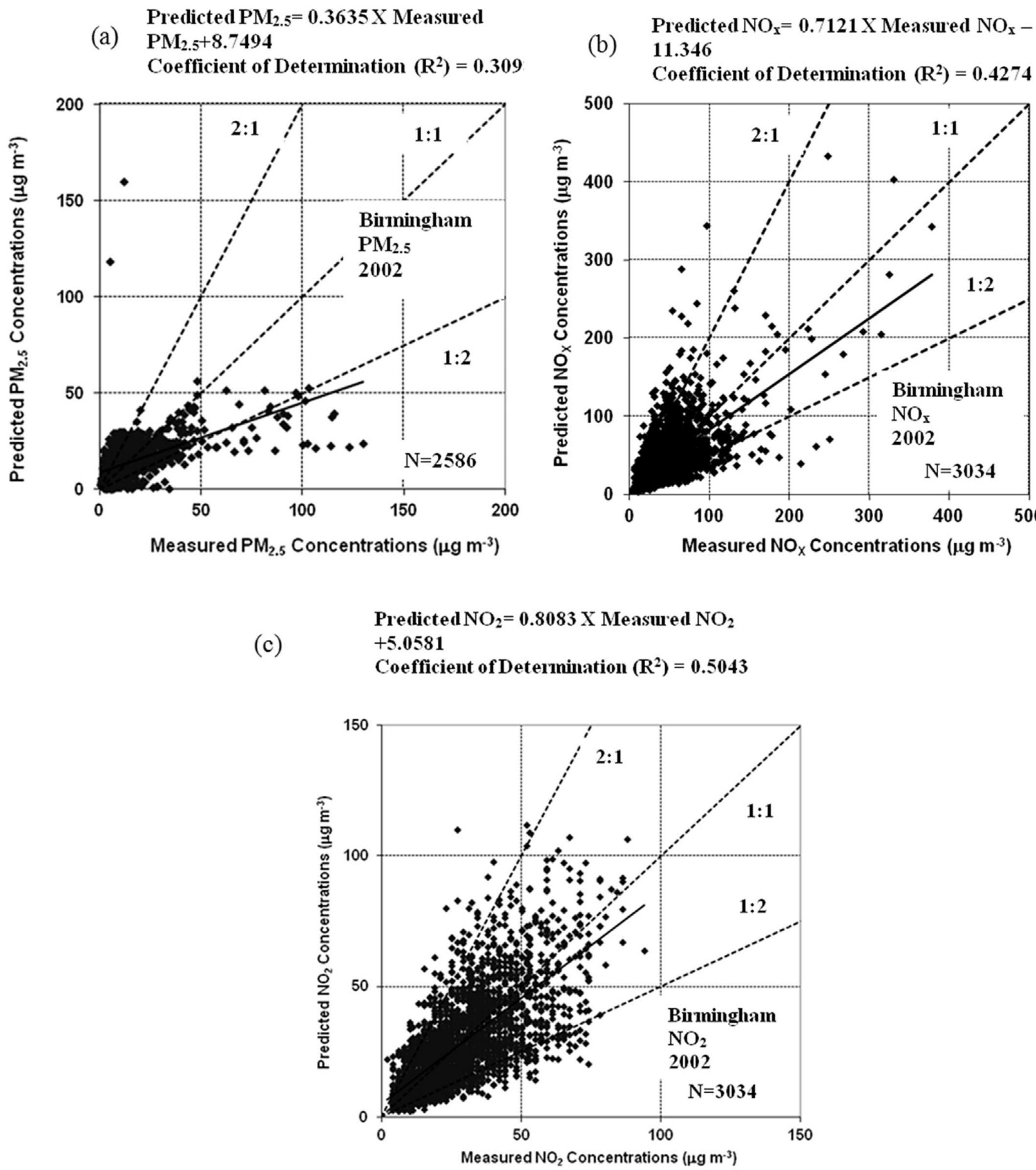


Fig. 4. a–c. Scatter plots of the predicted and measured hourly concentrations of $PM_{2.5}$ (upper left panel), NO_X (upper right) and NO_2 (lower panel) at the Centenary Square Site in Birmingham in 2002.

than 500 vehicles per hour). These cases may be influenced by the uncertainty associated with the Ambicount data (more specifically, the uncertainty associated with the particle number to mass conversion algorithm) in such conditions.

The fairly high or high IA values in Tables 3, 5 and 6 indicate that the temporal variation of the predicted $PM_{2.5}$ concentrations agrees fairly well with the observed data. The ranges of IA values for different pollutants are not substantially different, except for

Table 3
Statistical analysis of the observed (C_o) and predicted (C_p) daily (D) and hourly (H) concentrations of $PM_{2.5}$, NO_x and NO_2 at Cromwell Road site in London for 2004. The observed values of $PM_{2.5}$ were measured using TEOM and Ambicount. C_{bg} is the measured urban background concentration.

Statistical parameter	$PM_{2.5}$ using TEOM				$PM_{2.5}$ using Ambicount				NO_x				NO_2			
	D	H	$u \le 2$	$u > 2$	D	H	$u \le 2$	$u > 2$	D	H	$u \le 2$	$u > 2$	D	H	$u \le 2$	$u > 2$
Average C_{bg} ($\mu g\ m^{-3}$)	12.7	12.7	18.8	12.0	12.7	12.7	18.8	12.0	67.9	67.9	138.0	60.5	45.1	45.1	68.6	42.6
Average C_o ($\mu g\ m^{-3}$)	20.2	20.2	27.4	19.4	12.1	12.1	16.2	12.1	192.6	193.7	273.3	185.4	73	73	91.9	71.0
Average C_p ($\mu g\ m^{-3}$)	13.2	13.2	20.1	12.5	8.9	8.7	13.1	8.9	90.7	91.5	190.4	81.2	48.6	48.8	73.2	46.2
Index of agreement (IA)	0.80	0.78	0.78	0.77	0.75	0.72	0.93	0.70	0.64	0.70	0.77	0.65	0.61	0.66	0.62	0.65
Coefficient of determination (R^2)	0.81	0.57	0.59	0.55	0.75	0.44	0.87	0.42	0.69	0.47	0.46	0.45	0.53	0.36	0.19	0.35
Factor-of-two (F2; %)	78.3	67	87	64.8	94.1	87.5	87.5	87.5	31	36	71.6	32.3	82.6	71	88.3	69.2
Fractional bias (FB)	0.42	0.42	0.31	0.43	0.3	0.3	0.22	0.3	0.72	0.72	0.36	0.78	0.40	0.40	0.22	0.42
Root Mean Square Error (RMSE) ($\mu g\ m^{-3}$)	7.9	10.1	11	10.0	4.0	5.6	4.5	5.6	111.1	135.8	153.8	133.8	27.8	34.6	36.5	32.4
Number of data points (N)	157	3690	354	3336	17	407	8	399	155	3617	342	3275	155	3617	342	3275

Notes: D = daily average; H = hourly average; u = wind speed in m/s.

Table 4
The definitions of the relations of the atmospheric stability and the inverse Monin-Obukhov length, used in this study.

Atmospheric stability	Range of the inverse of Monin-Obukhov length (1/L), metre^{-1}	
	Lowest value	Highest value
Extremely unstable	- Infinity	−0.01
Unstable	−0.01	−0.00001
Neutral	Absolute value of 1/L < 0.00001	
Stable	0.00001	0.1
Extremely stable	0.1	+ Infinity

somewhat higher values in case of PM_{10}. For the campaigns in London, the temporal agreement of the predicted and measured NO_x and NO_2 concentrations was worse compared with those for $PM_{2.5}$ (in 2003–2004) or PM_{10} (2008). However, this situation was vice versa in case of the campaign in Birmingham (in case of $PM_{2.5}$). Therefore it can not be concluded that the modelling system would perform clearly better or worse for the various considered pollutants.

Considering the results for London, the model agreement with measured values was better for 2008, compared with those for 2003–2004, for most of the statistical parameters. This is expected and is partly caused by the more thorough modelling approach for 2008. The whole of the city was modelled for 2008, whereas for 2003–2004 simulations, a more limited modelling domain, combined with urban background concentration values was selected. The vehicular $PM_{2.5}$ emissions due to tyre and brake wear have also been included for the computations in London for 2008, but not for those in 2003–2004. However, there are obviously other factors also, such as meteorological conditions, which were different for these two sets of data.

As expected, the statistical parameters regarding the corresponding hourly concentrations indicate the same agreement of the average values (FB) as for the daily data, but slightly worse agreement of the temporal variations (measured using IA, R^2, F2 and RMSE). The data was segmented to two wind categories, to light wind speeds (defined as smaller than or equal to 2 m/s) and all the other wind speeds. There was some indication that the model performance was relatively slightly worse for the light wind speed category (especially for the R^2 values).

Both the PM mass fractions, and NO_x and NO_2 were substantially under-predicted in the campaigns in London, but not in those in Birmingham. For the computations in London for 2003–2004, and for those for Birmingham, the agreement of the average concentrations partly depends on how the urban background concentrations have been determined. In this study, the urban background for these two campaigns was evaluated based on the available urban background measurement stations.

The vehicular $PM_{2.5}$ emissions due to tyre and brake wear have been included for the computations in London for 2008, but not for the computations for the other two campaigns. The resuspension of material deposited on the road surface has not been taken into account for any of the campaigns. It is considered that the under-predictions of PM fractions were partly caused by the omission of the vehicular non-exhaust emissions.

Barlow et al. (2007) identified that non-exhaust processes in the U.K. constitute approximately 25% and 50% of the emissions of

Table 5
Statistical analysis of the observed (C_o) and predicted (C_p) daily (measured by Partisol) and hourly $PM_{2.5}$ (measured by Ambicount) concentrations; the latter presented also in two wind speed classes ($u \le 2$ and $u > 2$ m/s), from 30 April 2002 to 10 September 2002 at the Birmingham Centre site.

Statistical parameter	$PM_{2.5}$ using Partisol (D)/Ambicount (H)				$PM_{2.5}$ using Ambicount				NO_x				NO_2			
	D	H	$u \le 2$	$u > 2$	D	H	$u \le 2$	$u > 2$	D	H	$u \le 2$	$u > 2$	D	H	$u \le 2$	$u > 2$
Average C_{bg} (μg m^{-3})	11.2	11.2	12.9	10.8	11.2	11.2	12.9	10.8	25.9	25.9	43.7	22.4	21.3	21.3	33.6	18.9
Average C_o (μg m^{-3})	13.1	10.2	11.6	9.9	10.2	10.2	11.6	9.9	39.1	39.2	53.4	36.2	26.4	26.4	35.3	24.6
Average C_p (μg m^{-3})	12.3	12.5	15.1	11.9	12.4	12.5	15.1	11.9	38.7	39.1	67.9	33.3	26.2	26.4	42.5	23.1
Index of agreement (IA)	0.67	0.68	0.62	0.70	0.77	0.68	0.62	0.7	0.84	0.80	0.79	0.76	0.87	0.83	0.78	0.83
Coefficient of determination (R^2)	0.30	0.31	0.27	0.32	0.54	0.31	0.27	0.32	0.51	0.43	0.46	0.36	0.58	0.50	0.46	0.49
Factor-of-two (F2; %)	81	65.4	66.2	65.2	78.9	65.4	66.2	65.2	97.8	80	76.3	80.5	98.5	83.8	84.8	83.5
Fractional bias (FB)	0.06	−0.20	−0.26	−0.18	−0.2	−0.2	−0.26	−0.18	0	0	−0.24	0.08	0	0	−0.19	0.06
Root Mean Square Error (RMSE) (μg m^{-3})	7.2	10.2	11.5	9.9	6.8	10.2	11.5	9.9	15.3	26.4	42.9	21.4	8.1	12.9	18.2	11.6
Number of data points (N)	113	2586	441	2145	109	2586	441	2145	134	3034	519	2515	134	3034	519	2515

Notes: D = daily average; H = hourly average; u = wind speed in m/s.

Table 6
Summary of selected statistical parameters for the hourly data in the three considered measurement campaigns, in case of various pollutants. IA is the index of agreement, determined based on hourly or daily data, and FB is the fractional bias.

Measurement site and period		Pollutant	Statistical Parameter		
			IA, hourly	IA, daily	FB
London, Cromwell Road	2003–2004, more than 1 year, 3 months	$PM_{2.5}$	0.78	0.80	0.42
		NO_x	0.70	0.64	0.72
		NO_2	0.66	0.61	0.40
	2008, 1 year	PM_{10}	0.87	0.91	0.22
		NO_x	0.70	0.68	0.46
		NO_2	0.61	0.63	0.26
Birmingham, Centenary Square	2002, more than 4 months	$PM_{2.5}$	0.68	0.67	−0.20
		NO_x	0.80	0.84	0.00
		NO_2	0.83	0.87	0.00
Range of values at all sites and years		$PM_{2.5}$	0.68–0.78	0.67–0.80	−0.20–0.42
		PM_{10}	0.87	0.91	0.22
		NO_x	0.70–0.80	0.64–0.84	0.00–0.72
		NO_2	0.61–0.83	0.61–0.87	0.00–0.40

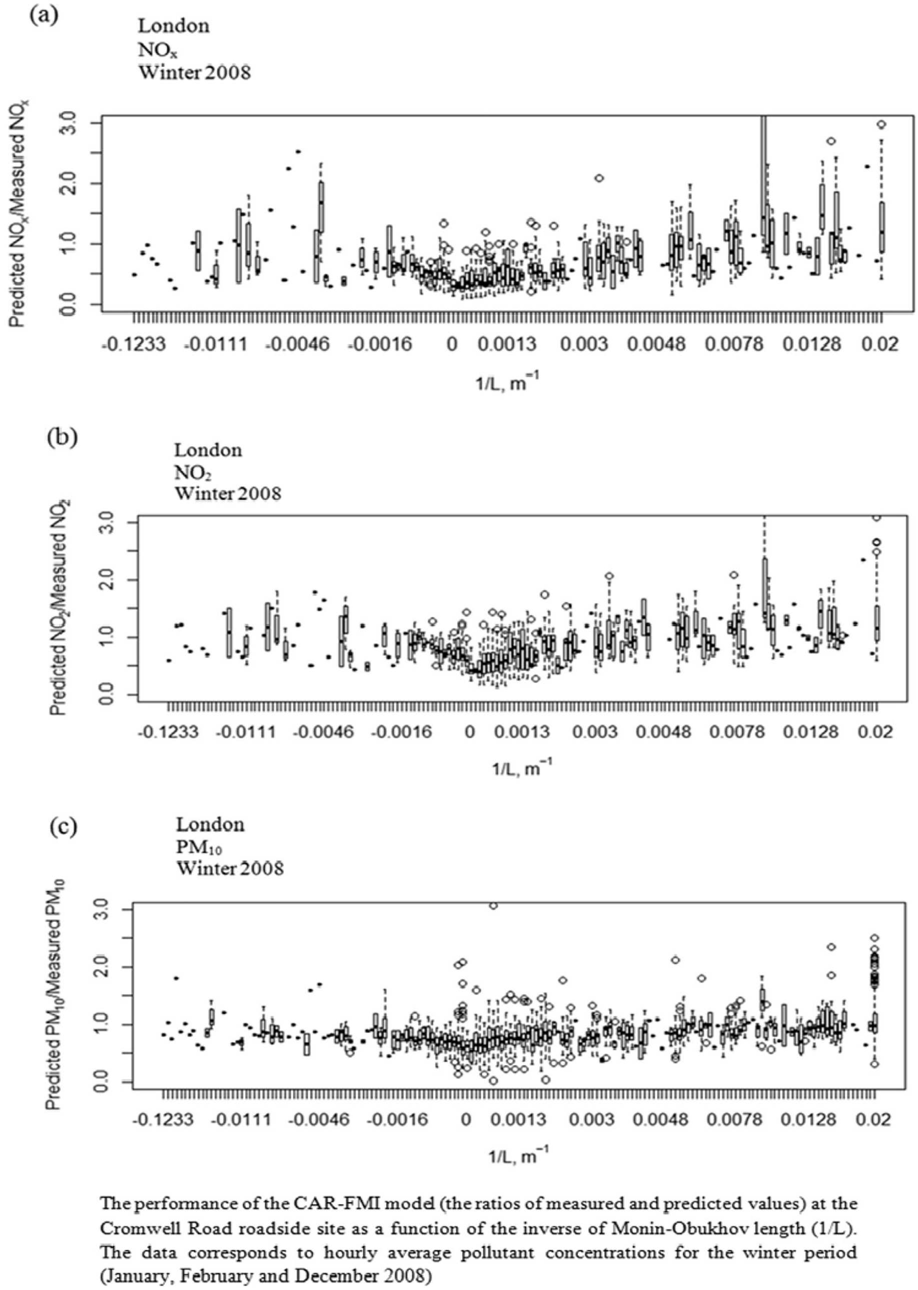


Fig. 5. a–c. Box and whisker plots of the predicted and measured hourly concentrations of NO_x (top panel), NO_2 (middle panel) and PM_{10} (lower panel) as factor of the inverse of Monin-Obukhov length at the Cromwell Road roadside site in London in winter 2008. The median of the data (indicated by a line within the box), the lower and upper quartiles (25% and 75% respectively, indicated by edges of the box), the minimum and maximum values (indicated by horizontal lines outside the box), and the outliers (indicated by circles).

$PM_{2.5}$ and PM_{10}, respectively. According to this estimate, neglecting the non-exhaust emissions of $PM_{2.5}$ would cause an under-prediction of the local contribution of approximately one quarter. However, the local contribution is commonly responsible for less than a half of the total $PM_{2.5}$ concentration. Only a minor fraction of the regional background $PM_{2.5}$ is originated from non-exhaust. In conclusion, the influence of the exclusion of the non-exhaust contribution would be on the average of the order of magnitude of one half of a quarter or less (i.e., <13%), for $PM_{2.5}$.

Especially in case of London, the under-predictions of both PM fractions and NO_x could also be caused by the commonly occurring severe congestion situations in the capital city.

A detailed analysis of the $PM_{2.5}$ data in the campaign in Birmingham shows that the model predicts the smaller concentrations fairly well, but under-predicts most of the highest concentrations measured by Ambicount (higher than 30 μg/m^3). The vehicular emission factors were based on the National Atmospheric Emissions Inventory, UK (NAEI, 2003), which is based on nationally conducted vehicle emission measurements. However, the contribution of non-exhaust emissions, and the suspended particulate matter from street surfaces was not allowed for the campaign in Birmingham. The influence of suspension emissions is expected to be relatively smaller than that of non-exhaust emissions in the UK conditions. The non-exhaust emissions could therefore be the reason for an under-prediction of the highest concentrations. Another reason could potentially be the emissions in severely congested conditions, which are challenging to model accurately.

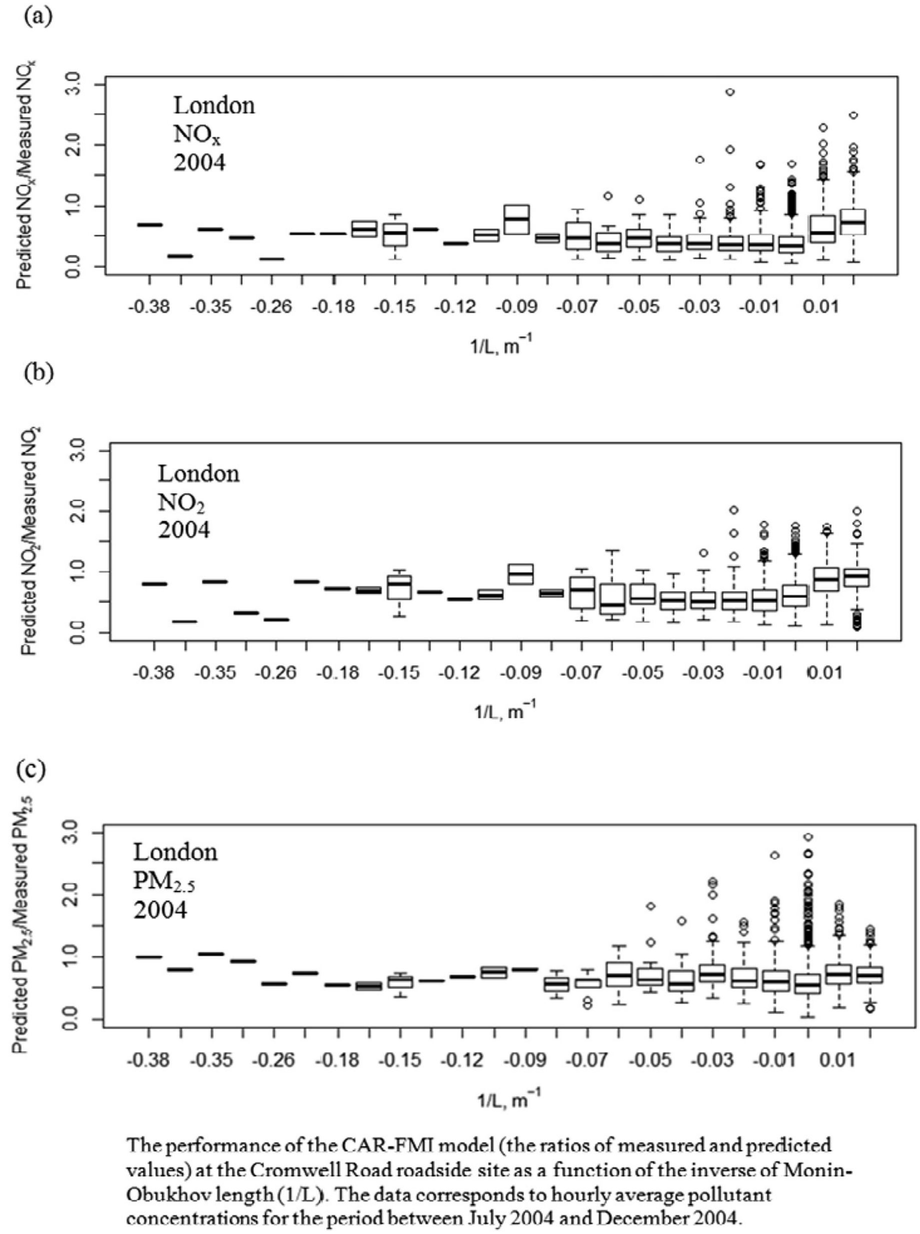


Fig. 6. a–c. Box and whisker plots of the predicted and measured hourly concentrations of NO_x (top panel), NO_2 (middle panel) and $PM_{2.5}$ (lower panel) as factor of the inverse of Monin-Obukhov length at the Cromwell Road roadside site in London in 2004. The median of the data (indicated by a line within the box), the lower and upper quartiles (25% and 75% respectively, indicated by edges of the box), the minimum and maximum values (indicated by horizontal lines outside the box), and the outliers (indicated by circles).

3.3.2. Evaluation of model performance with respect to atmospheric stability and road traffic conditions

The model performance with respect to a selected stability parameter, Monin-Obukhov length, was analysed. The values of the inverse of Monin-Obukhov length are associated to atmospheric stability conditions, as presented in Table 4.

The box and whisker plots showing the model performance, evaluated in terms of this stability parameter, are presented in Figs. 5–7 for all three monitoring campaigns (Cromwell Road, 2008 campaign, Cromwell Road, 2004 campaign and Birmingham, 2002 campaign, respectively).

In general, considering the results from all the measurement campaigns (in Figs. 5–7), the model performance does not show any substantial, systematic dependency with respect to atmospheric stability. However, in closer inspection of the individual campaigns, there are some slight dependencies.

The results in Figs. 5 and 6a–c showed that the modelling system mostly slightly under-predicted the concentrations in unstable and neutral atmospheric conditions, and slightly over-predicted in part of the stable conditions. The 'predicted/measured' concentration ratio also varied more significantly under stable and extremely stable conditions, compared with more neutral atmospheric stability conditions.

The results in Fig. 5a–c show the model performance in London in 2008 during the winter season; the fraction of stable conditions was therefore substantially higher, compared with those in Fig. 6a–c. The median value of the 'predicted/measured' ratio is close to 1.0 for the majority of the presented stability conditions, although the model over-predicted during part of the extremely unstable atmospheric conditions.

The performance of the modelling system for nitrogen oxides in relation to the average diurnal profiles of road traffic data and the

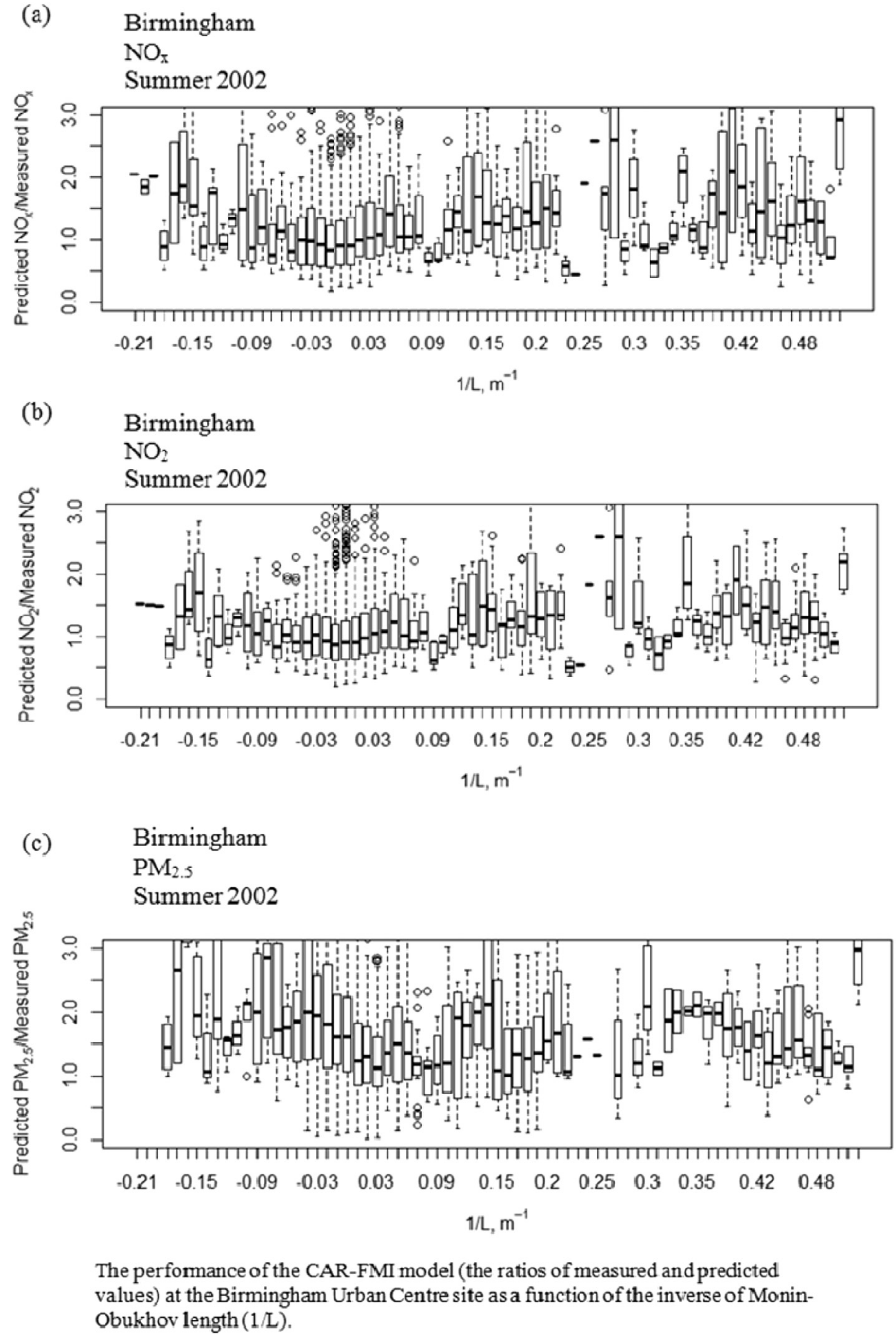


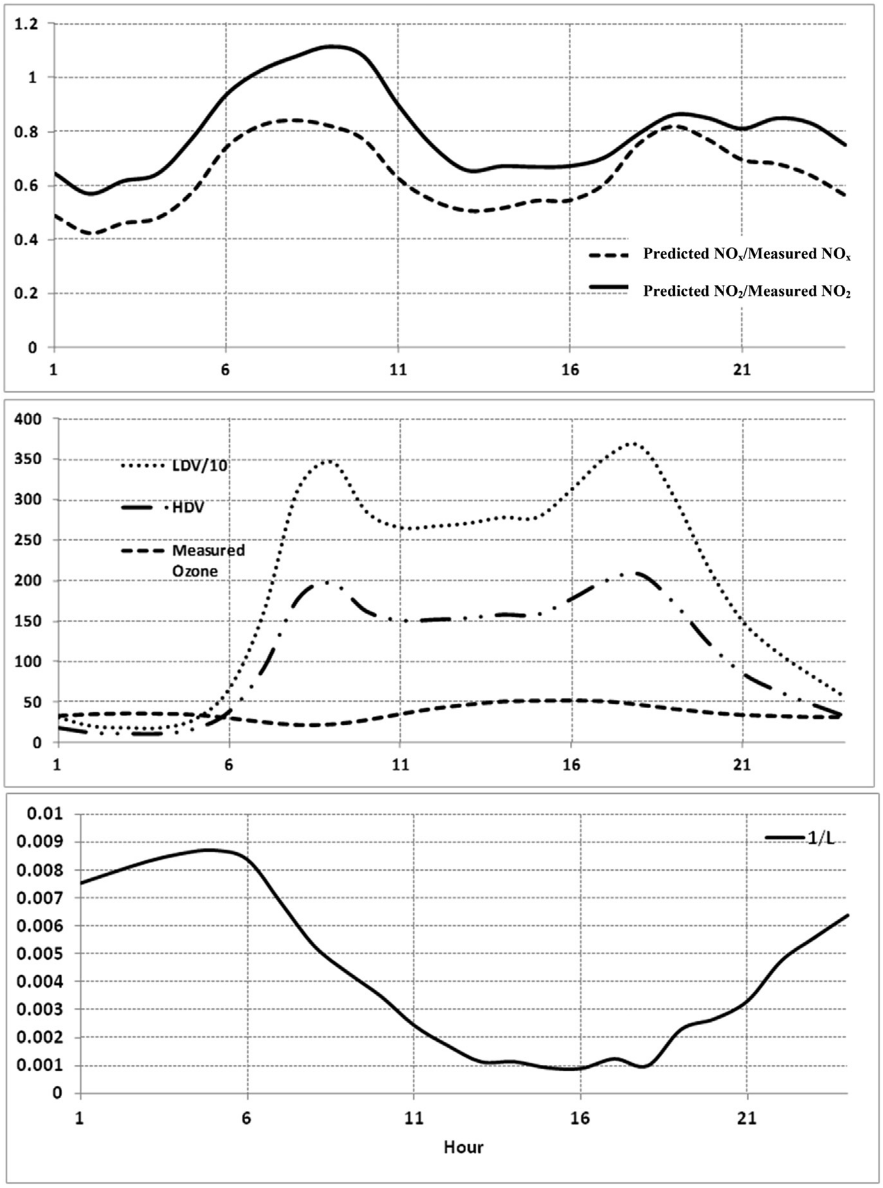
Fig. 7. a–c. Box and whisker plots of the predicted and measured hourly concentrations of NO_x (top panel), NO_2 (middle panel) and $PM_{2.5}$ (lower panel) as factor of the inverse of Monin-Obukhov length at the Centenary Square Site in Birmingham in 2002. The median of the data (indicated by a line within the box), the lower and upper quartiles (25% and 75% respectively, indicated by edges of the box), the minimum and maximum values (indicated by horizontal lines outside the box), and the outliers (indicated by circles).

inverse of Monin-Obukhov length is presented in Figs. 8–10 for all three monitoring campaigns (Cromwell Road campaign in 2008, Cromwell Road campaign in 2004 and Birmingham campaign in 2002, respectively).

The results in Figs. 8 and 10 show that the modelling system under-predicted the NO_x and NO_2 concentrations during the midday and in the afternoon, compared with the other times of the day. However, considering the results in Fig. 8, the model performance does not show any clear systematic diurnal dependency. These diurnal dependencies of the model performance could be caused either by the diurnal cycle of the meteorological parameters, or more severe traffic congestion at certain times of the day.

3.4. Spatial concentration distribution of NO_x in London for 2008

The spatial distribution of the modelled annual mean NO_x concentrations for the year 2008 is presented in Fig. 11. The highest concentrations occur near busy roads and motorways, at their junctions, and in the centre of London. The modelled annual mean NO_x concentration in central London ranges approximately from 50



The average diurnal profiles during weekdays for various parameters at the roadside monitoring site of Cromwell Road during the period between January 2008 and December 2008. The concentrations of NO_x, NO_2 and O_3 are presented in $\mu g/m^3$. LDV and HDV represent light and heavy duty vehicles, respectively. 1/L is the inverse of Monin-Obukhov length in m^{-1}. Measured ozone concentrations at the urban background site of North Kensington are also presented.

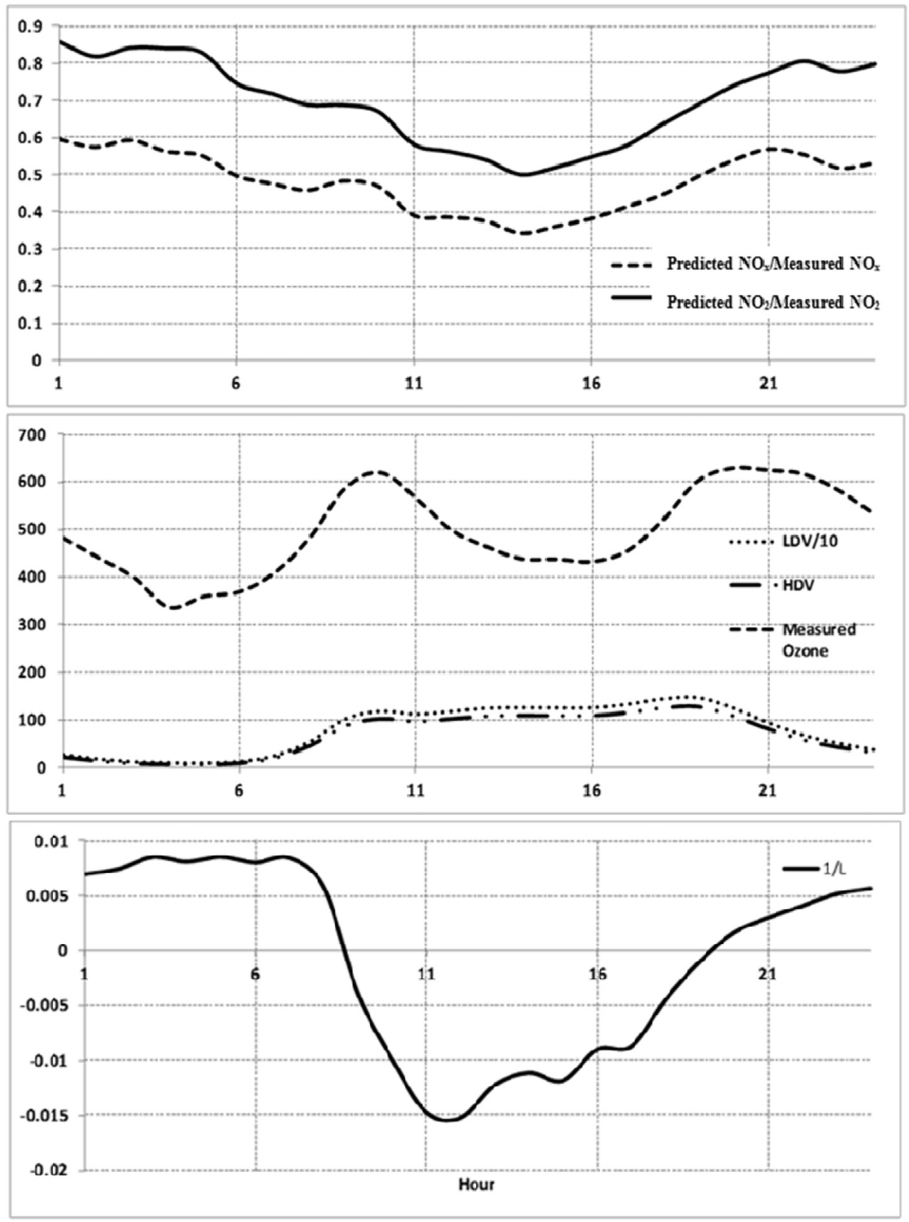
Fig. 8. Weekday diurnal profiles showing the CAR-FMI model performance with reference to the road traffic and stability conditions at the Cromwell Road roadside site in London in 2008.

to 150 $\mu g/m^3$, which decreases to a couple or a few tens of $\mu g/m^3$ in the suburban area. Annual mean NO_x concentration near motorways can reach over 400 $\mu g/m^3$. A corresponding concentration distribution of $PM_{2.5}$ for 2008 has previously been presented by Singh et al. (2014).

4. Conclusions

The results of three measurement campaigns in the two most populous cities in the U.K., London and Birmingham have been presented. The concentrations of $PM_{2.5}$, PM_{10}, NO_x and NO_2 were predicted using the OSCAR air quality modelling system. Two versions of the urban scale modelling system were used: one that applied the emissions from a network of streets and roads in the whole of London for 2008, and a simpler configuration that considered local vehicular concentrations within a limited domain in both cities, in combination with urban background concentrations.

A normalised roadside increment was defined as the difference of the measured roadside and urban background values, divided by the measured concentration at the urban background site. The normalised roadside increments (compared with the corresponding urban background values) for $PM_{2.5}$ and NO_x were estimated to be approximately 0.59 and 1.9 in London, and 0.17 and 0.51 in Birmingham, respectively. As expected, the roadside increments were smaller for $PM_{2.5}$, compared with the corresponding



The average diurnal profiles during weekdays for various parameters at the roadside monitoring site of Cromwell Road during the period between July 2004 and December 2004. The concentrations of NO_x, NO_2 and O_3 are presented in μg/m³. LDV and HDV represent light and heavy duty vehicles, respectively. 1/L is the inverse of Monin-Obukhov length in m^{-1}. Measured ozone concentrations at the urban background site of North Kensington are also presented.

Fig. 9. Weekday diurnal profiles showing the CAR-FMI model performance with reference to the road traffic and stability conditions at the Cromwell Road roadside site in London in 2004.

increments for NO_x, due to the more significant regional background for $PM_{2.5}$. The increments for both $PM_{2.5}$ and NO_x were substantially smaller in case of Birmingham, probably due to the location of the selected urban background stations in closer vicinity of the local traffic in case of Birmingham.

The emission and dispersion computations contain several inherent uncertainties. The modelling system did not allow for the influences of individual buildings and obstacles on atmospheric dispersion. The computations for two of the measurement campaigns neglected vehicular non-exhaust emissions. The emission modelling also does not sufficiently allow for the increased emissions during severe traffic congestion. The computations for two of the campaigns relied on measured or modelled urban background concentrations; the accuracy of the former depends on the representativity of the selected urban background stations.

Despite the several potential sources of uncertainty, the agreement of the predicted and measured hourly and daily time-series can be considered to be fairly good or good, when compared to model responses reported in other similar studies (e.g., Karppinen et al., 2000) for all of the considered campaigns and pollutants. Considering all results from various campaigns, the modelling system performed approximately equally well for $PM_{2.5}$, compared with NO_x and NO_2. In case of the campaigns in London, both the PM fractions and the nitrogen oxide concentrations were under-

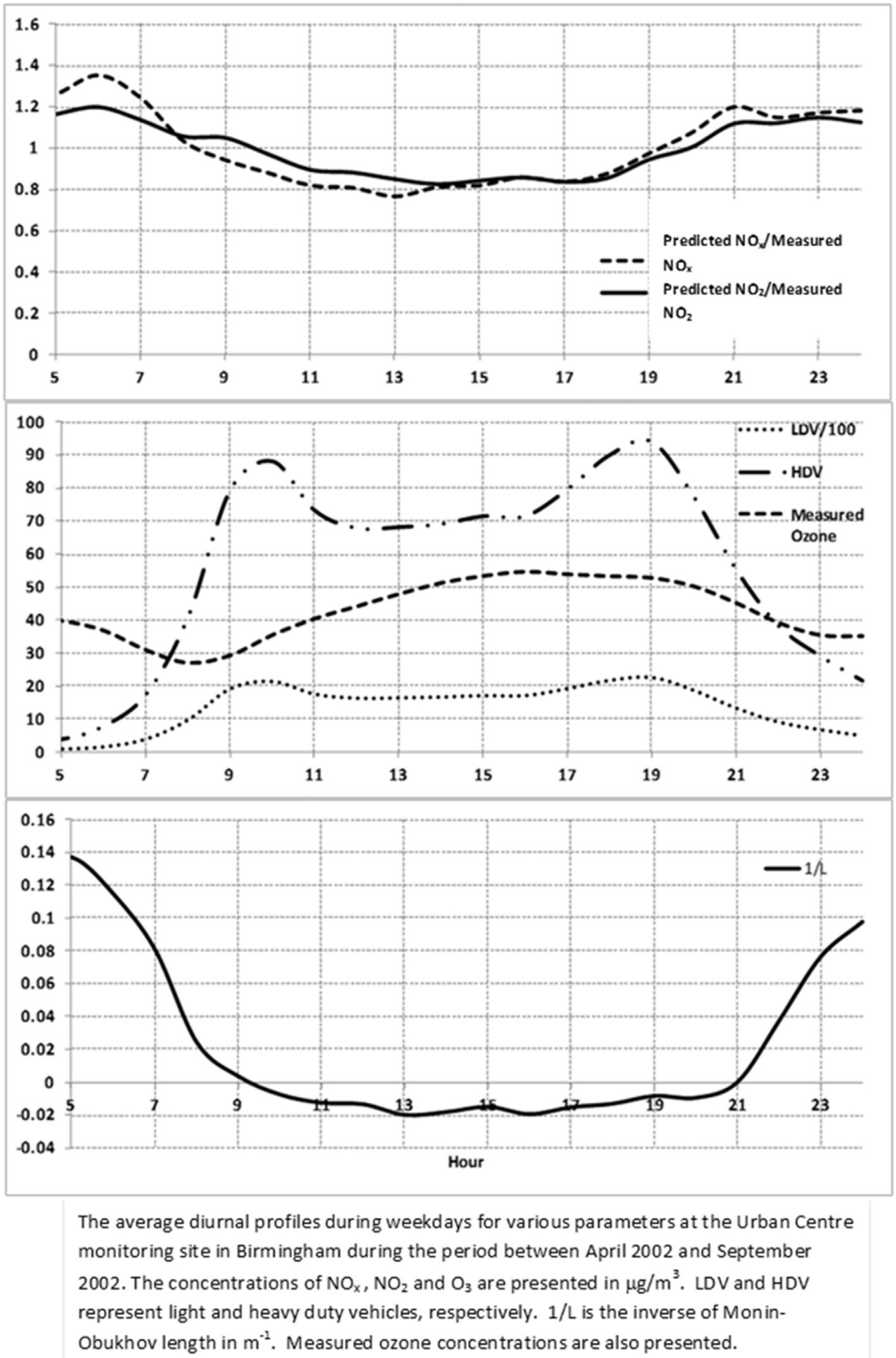


Fig. 10. Weekday diurnal profiles showing the CAR-FMI model performance with reference to the road traffic and stability conditions at the Birmingham Urban Centre site in 2002.

predicted. Considering the results from all measurement campaigns, the model performance did not show any systematic dependency with respect to atmospheric stability. Generally, it is beneficial to have performance statistics corresponding to several measurement campaigns, as the details of model performance can be specific to any single campaign.

The model agreement with measured values was better for the results in London in 2008, compared with those in London in 2003–2004 (in terms of both the IA and FB values). This could partly be caused by the more thorough modelling approach: vehicular traffic within the whole of the city was modelled for 2008, whereas for 2003–2004, a substantially smaller explicit modelling domain for vehicular traffic was selected. The non-exhaust share of the $PM_{2.5}$ emissions was also taken into account for the computations for 2008.

The highest NO_x concentrations occurred near busy roads and motorways, at their junctions, and in the centre of London. The modelled annual mean NO_x concentration in central London ranged approximately from 50 to 150 µg/m^3, whereas annual mean NO_x concentrations near motorways reached over 400 µg/m^3.

The OSCAR air quality modelling system used in this study can be applied to any location worldwide, assuming that the necessary traffic, emission and meteorological input data are available. More generally, this study has shown that ideally a more detailed

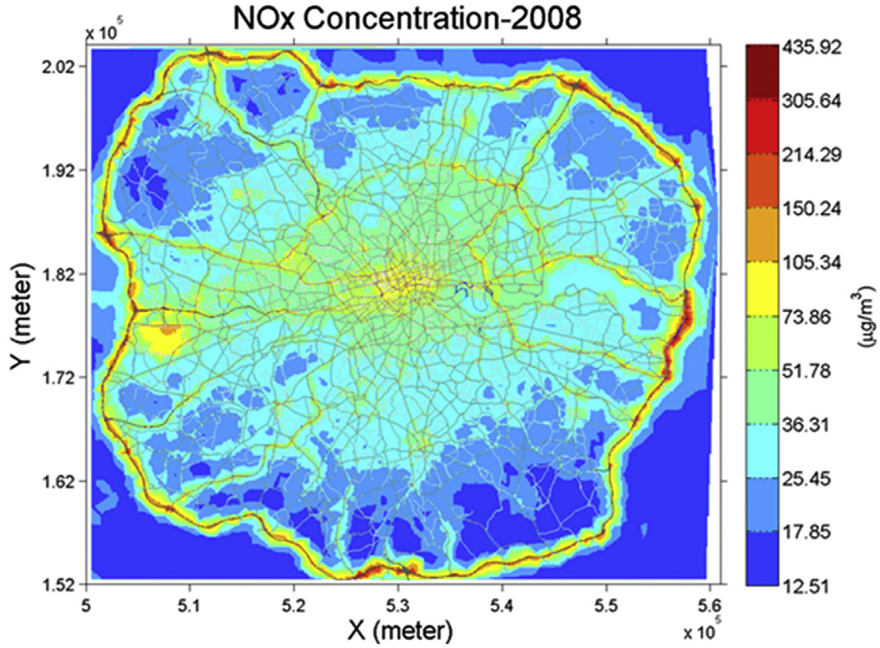


Fig. 11. Predicted spatial distribution of the annual mean NO_x concentrations in London in 2008.

emission inventory for a larger proportion of the urban road network and urban background contributions is needed for more accurate simulations. Clearly, the accurate representation of the urban background concentrations is an additional source of uncertainty.

Acknowledgements

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Appendix A

Gravimetric particle mass concentrations measured by two prototypes of Ambicount

Fig. A.1 shows PM_{10} concentrations measured gravimetrically from filters of two prototypes of Ambicount (AC1 and AC2). PM_{10} concentrations were very well correlated ($R^2 = 0.98$), with an average difference of 0.01% with AC1 measuring more than AC2.

Gravimetric particle mass concentrations measured by Partisol and two prototypes of Ambicount

PM_{10} concentrations measured by Partisol (a Rupprecht and Patashnick (R&P) gravimetric particle sampling system fitted with a PM_{10} inlet head) and two prototypes of Ambicount are shown in Figs. A.2 and A.3.

Correlation between Partisol and AC1 PM_{10} concentrations was 0.91 and that between Partisol and AC2 was 0.87. Average difference between Partisol and AC1 was about 3% (Partisol measured higher concentrations than AC1). Average difference between Partisol and AC2 was 3% (AC2 measured higher concentrations than Partisol).

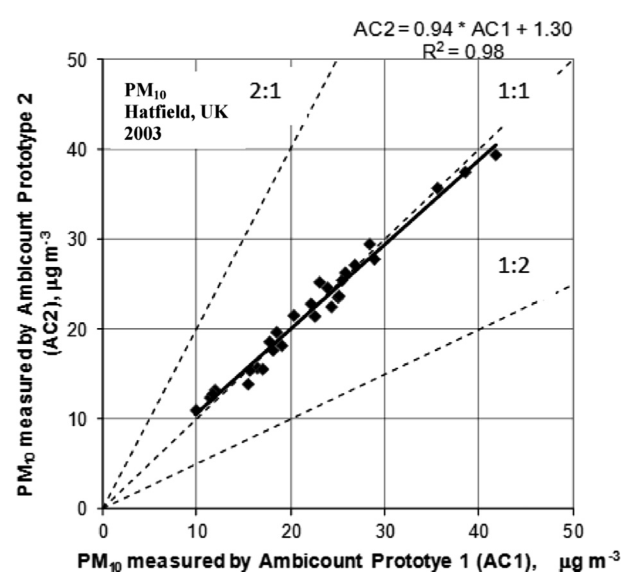

Fig. A1. Comparison of PM_{10} concentrations measured gravimetrically by two prototypes of Ambicount (AC1 and AC2) during the entire measurement period in June 2003 at a Suburban site in Hatfield, UK. Filters changed every 12 h (Data from 28 filters plotted). The dashed line (1:1) indicates 100% agreement.

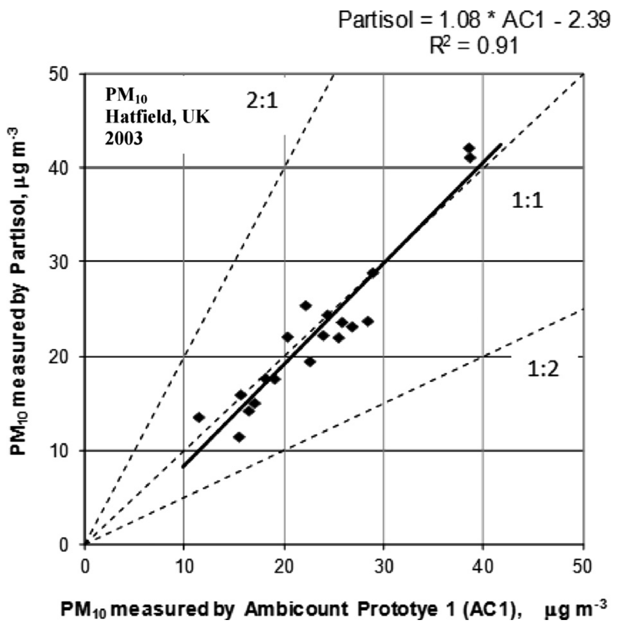

Fig. A2. Comparison of PM_{10} concentrations measured gravimetrically by Partisol and Ambicount Prototype 1 during the entire measurement period in June 2003 at a Suburban site in Hatfield, UK. Filters changed every 12 h (Data from 19 filters plotted).

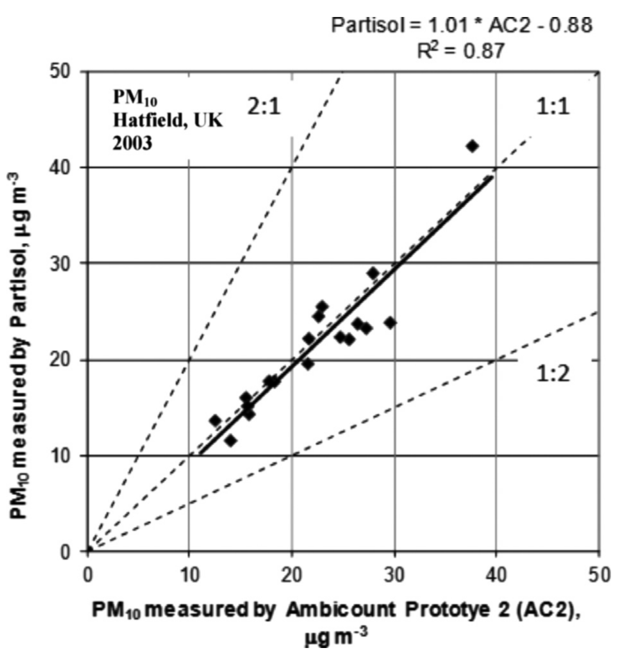

Fig. A3. Comparison of PM_{10} concentrations measured gravimetrically by Partisol and Ambicount Prototype 2 during the entire measurement period in June 2003 at a Suburban site in Hatfield, UK. Filters changed every 12 h (Data from 18 filters plotted).

References

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