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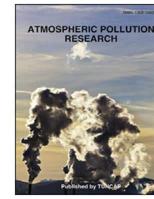

Original Article

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

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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.

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.