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Performance characteristics of MM5–SMOKE–CMAQ for a summer photochemical episode in southeast England, United Kingdom

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Abstract

In this study the Mesoscale Model (MM5)–Sparse Matrix Operator Kernel Emissions (SMOKE)–Community Multiscale Air Quality (CMAQ) ~~modelling system~~ has been applied to a summer photochemical period in southeast England, UK. Ozone (O_3), nitrogen dioxide (NO_2) and particulate matter ($PM_{2.5}$) concentrations modelled with different horizontal grid resolutions (9 and 3 km) were evaluated against available ground-level observations from the UK Automatic Urban and Rural Network (AURN) and London Air Quality Network (LAQN) for the period of 24–28 June 2001 with a focus on O_3 predictions. This effort, which represents the first comprehensive performance evaluation of the modelling system over a UK domain, reveals that CMAQ's ability to reproduce surface O_3 observations varies with O_3 concentrations. It underpredicts O_3 mixing ratios on high-O_3 days and overpredicts the maximum and minimum hourly O_3 values for most low-O_3 days. Model sensitivity analysis with doubled anthropogenic NO_x or volatile organic compounds (VOC) emissions and ~~analysis~~ of the daylight-averaged levels of OX (sum of O_3 and NO_2) as a function of NO_x revealed that the underepredicition of peak O_3 concentrations on high-O_3 days is caused by the underprediction of regional contribution and to a lesser extent local production, which might be related to the underestimation of European emissions in EMEP inventory and the ~~lacked~~ reactivity of the modelled atmosphere. CMAQ systematically underpredicts hourly NO_2 mixing ratios but captures the temporal variations. The normalized mean bias for hourly NO_2, although much larger than that for O_3, falls well within the generally accepted range of −20% to −50%. CMAQ with both resolutions (9 and 3 km) significantly underpredicts $PM_{2.5}$ mass concentrations and fails to reproduce its temporal variations. While model performance for O_3 and $PM_{2.5}$ are not sensitive to model grid resolutions, a better agreement between modelled and measured hourly NO_2 mixing ratios was achieved with higher resolution. Further investigation into the uncertainties in

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meteorological input, uncertainties in emissions, as well as representation of physical and chemical processes (e.g. chemical mechanism) in the model is needed to identify the causes for the discrepancies between observations and predictions.
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Keywords: Model evaluation; CMAQ; $PM_{2.5}$; Ozone; UK

1. Introduction

Despite the substantial efforts in reducing pollutant emissions in the last decades, especially in transportation and in industry, pollutant concentrations in many European cities are still likely to exceed ambient air quality standards and guidelines. Elevated concentrations of tropospheric ozone (O_3), nitrogen dioxide (NO_2) and fine particulate matter are known to have negative effects on human health and the environment (e.g. Englert, 2004; Koop and Tole, 2006), and to a larger extent have important influence on the global atmospheric chemistry and climate change (Jenkin and Clemitshaw, 2000). In order to minimize the environmental and health impacts of pollutants such as O_3, organizations such as the United Nations Economic Commission for Europe (UNECE) and the Commissions for Europe Communities (EC) have proposed and agreed to protocols designed to reduce concentrations of these pollutants in Europe (e.g. UNECE, 1999; CEC, 1999, 2002). In the United Kingdom, air pollutants are also subject to standard specified by the National Air Quality Strategy and European Air Quality Framework Directives.

Air quality models are now widely used to estimate the spatial distribution and evolution of tropospheric pollutant concentrations, resulting from both local emissions and long-range transport. They are also valuable tools for the exploration of emission control strategies to mitigate elevated concentrations of pollutants such as O_3, NO_2 and particulate matter (e.g. PM_{10} and $PM_{2.5}$). During the last two decades different air quality models, ranging from simple statistical models to fully three-dimensional (3-D) comprehensive Eulerian models, have been developed in Europe and elsewhere. The Community Multiscale Air Quality (CMAQ) model developed by the US Environment Protection Agency (US EPA), have been increasingly used in North America (e.g. Hogrefe et al., 2004; Eder and Yu, 2006; Smyth et al., 2006; Byun et al., 2007) and Asia (e.g. Zhang et al., 2006a; Chen et al., 2007) for both scientific studies and regulatory assessment. However its application and validations for European domains are very limited (e.g. Sokhi et al., 2006; Vautard et al., 2007; Jiménez et al., 2007). In fact, apart from the reported works of Sokhi et al. (2006) and Cocks et al. (2003) there are no published studies on the use of CMAQ for studying air pollution episodes for the UK. Dispersion models with simplified treatment for meteorology and/or chemistry, such as Trajectory Model with Atmospheric Chemical Kinetics (TRACK) (Lee et al., 2000), Ozone Source–receptor Model (OSRM; Hayman et al., 2002) and ADMS (CERC, 1998), have been adopted as policy tools in the UK. These approaches are not adequate for cases where complex multi-pollutants and multiscale interactions and coupling between atmospheric chemistry and dynamics are involved.

The purpose of the present study is to apply CMAQ to the UK domain and evaluate its ability to simulate ambient concentrations of O_3, NO_2 and $PM_{2.5}$ in the southeast of England during a summer (June 2001) air pollution episode. The modelled concentrations have been compared with observations from several ground-based monitoring stations. The paper focuses on O_3 by undertaking further sensitivity studies and analysis of OX (the sum of O_3 and NO_2) to help understand the O_3 behaviour during the period and possible sources of model discrepancies.

The rest of the paper is structured as follows. The modelling system and its configuration are briefly introduced in Section 2, along with details of model input preparation. Evaluation results are presented in Section 3 focusing on O_3, NO_2 and $PM_{2.5}$, with discussions given in Section 4. Conclusions are presented in Section 5.

2. Model set-up and input preparation

2.1. Modelling period and CMAQ configuration

The surface O_3 distribution in the UK is characterized by a marked gradient from south to north with the highest concentrations in the south

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and east of the British Isles (UK PORG, 1997). The degree of severity of summertime photochemical episodes largely depends on daytime air temperature, and high summertime air pollution events are almost always associated with anticyclonic conditions and temperatures in excess of 28–30 °C (UK PORG, 1997). Long-range transport of O_3 and its precursors from the European continent may contribute significantly to the elevated O_3 concentrations during these photochemical episodes (Derwent et al., 2003). Although the general synoptic causes of episodes are known, there is an important scientific and policy need to be able to explain the behaviour of pollutants, spatially and temporally, under such meteorological conditions. One ~~of~~ such event occurred during 24–26 June 2001 when warm weather (with maximum temperatures reaching 30 °C on 26 June 2001) prevailed over much of the south and east of the UK. The UK Automatic Urban and Rural Network (AURN) recorded a peak O_3 concentration of 198 $\mu g\,m^{-3}$ (~99 ppb) at Lullington Heath near the south coast of England on 26 June and a peak NO_2 concentration of 161 $\mu g\,m^{-3}$ (~84 ppb) at an urban site on the same day. The UK Air Quality Expert Group (AQEG, 2004) has identified this episode as an example of a NO_2 episode related to a summertime photochemical episode and it was therefore selected for evaluating the Mesoscale Model (MM5)–Sparse Matrix Operator Kernel Emissions (SMOKE)–CMAQ modelling system in the present study. The modelling time period began at 12 UTC 22 June and ended at 12 UTC 28 June 2001. The first two simulation days were used as a 'spin-up' period as recommended by Berge et al. (2001) and Jiménez et al. (2007) and the analyses focus on the following 4 days after which the episode dissipated as a low pressure system brought relatively cooler and more changeable weather with occasional outbreaks of rain or showers.

In this study, the US EPA's CMAQ version 4.4 was used with a modified version of the Carbon-Bond Mechanism version IV (CB-IV) chemical mechanism. Fig. 1a shows the CMAQ modelling domain that consists of four nested domains with resolutions of 81, 27, 9 and 3 km. The coarsest domain covers most of the Europe and the finest 3 km-grid domain covers the southeast of England. Vertically there are 26 σ-levels extending from the surface to an altitude of about 14 km. Vertical layers were unevenly distributed with fifteen layers in the lowest kilometre and a surface layer of approximately 14 m above ground level (AGL). Fig. 1b shows the enlarged 3 km-grid domain and the locations of measurement sites referred to in this paper.

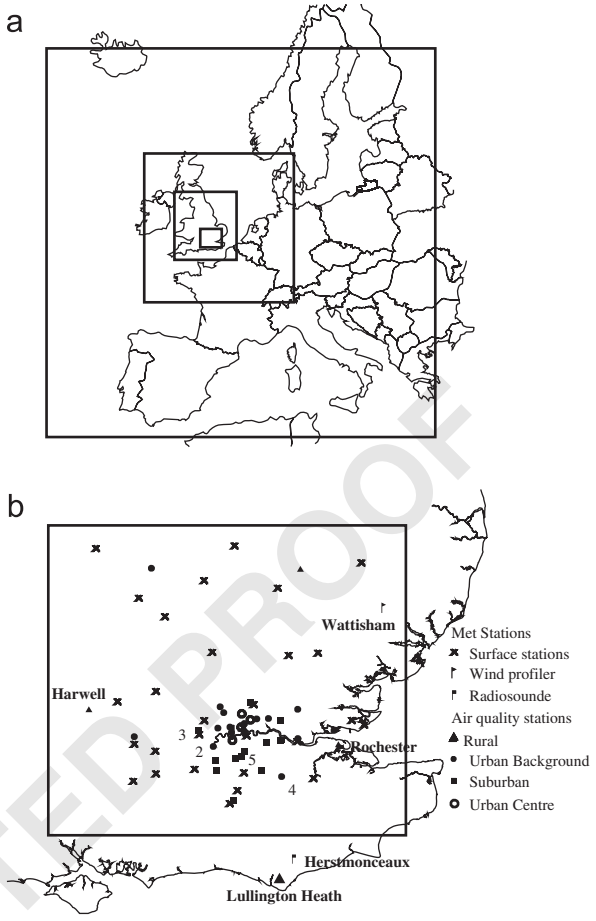

Fig. 1. (a) Four nested CMAQ modelling domains; (b) the 3 km-grid CMAQ domain marked with locations of UK Hourly Weather Observation sites, UK Automatic Urban and Rural Network (AURN) sites and London Air Quality Network (LAQN) observation sites used for model evaluation. 1—London Bloomsbury (Urban Centre), 2—London Teddington (Urban Background), 3—London Hillingdon (Suburban), 4—Sevenoaks (Urban Background), 5—Croydon (Suburban).

2.2. Model input preparation

2.2.1. Meteorology

The Fifth-generation Pennsylvania State University–National Center for Atmospheric Research (NCAR) MM5, version 3 (Dudhia et al., 2004) was used to generate meteorological fields for CMAQ. The MM5 was configured to have four nested domains, covering and aligning with the

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CMAQ domains shown in Fig. 1a with each of the MM5 domain being at least five grid cells larger than the corresponding CMAQ domains. The European Centre for Medium-Range Weather Forecasts (ECMWF) $1^\circ \times 1^\circ$ reanalysis data available at every 6 h were used to provide initial and boundary conditions for the coarsest MM5 domain. The physical options used in MM5 include the Medium Range Forecast (MRF) PBL scheme, the Dudhia simple ice microphysics scheme, the cloud radiation scheme and the five-layer soil model. The Anthes–Kuo cumulus parameterization scheme was used for the coarsest model domain, the Grell cumulus parameterization scheme was used for the 27- and 9-km grid domains and no cumulus scheme was used for the 3-km resolution domain.

2.2.2. Emissions

Annual anthropogenic emissions for six pollutants, i.e. NO_x, non-methane volatile organic compounds (NMVOCs), sulphur dioxide (SO_2), carbon monoxide (CO), ammonia (NH_3) and fine and coarse particulate matter (i.e. $PM_{2.5}$ and PM coarse), were taken from the European Monitoring and Evaluation Programme (EMEP) for year 2002 (http://www.emep.int) and used for all the CMAQ domains except model grid cells covering the UK (including North Ireland, Scotland, England and Wales), where the 1-km spatial resolution National Atmospheric Emissions Inventory (NAEI) data (http://www.naei.org.uk/) were used. Point source emissions were extracted from the European Pollutant Emission Register (EPER, http://www.eper.cec.eu.int/) and NAEI database, for non-UK and UK point sources, respectively. The SMOKE model (Carolina Environmental Program, 2003) was used to process these annual emissions to a temporally resolved, spatially distributed and speciated model-ready emissions data for CMAQ. NMVOC emissions were split into model species represented in the CB-IV chemical mechanism. Different speciation profiles were derived for different activity sectors based on the detailed UK volatile organic compounds (VOC) speciation given in Dore et al. (2004). It is assumed that the speciation profile for the UK could be applied across Europe without further adjustment. This assumption was considered to be reasonable as vehicle exhaust emissions, fuel evaporative emissions and solvents are likely to have similar profiles across north west Europe, although uncertainty in the spatial distribution of individual VOC emissions may be large for other parts of Europe. Temporal profiles were developed, taking into account monthly, weekday/weekend and hourly variations, for each country, activity sectors and pollutant using information provide by the Institute for Energy Economics and Rational Use of Energy, University of Stuttgart (IER, private communication) and information available in Jenkin et al. (2000).

Biogenic emissions of isoprene and monoterpenes were calculated based on the following formulate (Guenther et al., 1995; Sanderson, 2002):

$$F_i = \varepsilon_i D \gamma_i$$

where F_i is the emission flux ($\mu g\,m^{-2}\,h^{-1}$), ε_i is an ecosystem (i)-dependent emission factor ($\mu g\,C\,g^{-1}\,h^{-1}$) and D is the foliar density. Values of ε_i and D are taken from Sanderson (2002); γ_i is the environmental correction factor accounting for the dependence on temperature and radiation (Guenther et al., 1995). The spatial distribution of ecosystems was established by firstly aggregating the 100 m resolution Coordination of Information on the Environment (CORINE) Land Cover data for Europe (CLC2000, http://dataservice.eea.europa.eu/dataservice/) to the CMAQ model grids. Then the 44 CORINE land use classes were aggregated into four ecosystems (i.e. grass, broadleaf forest, needle leaf forest and shrub) and the fraction of the area of each grid cell covered by each ecosystem class and the associated emissions of isoprene and monoterpenes was calculated using the hourly temperature and solar radiation values from MM5. Nitrogen oxide (NO_x) released from soil and formed by lightning are not included in the present study. The possible effect on model results will be discussed later.

2.2.3. Initial and boundary conditions

The initial and boundary conditions for the coarsest CMAQ domain were generated based on monthly mean data from the UK Met Office global 3-D Lagrangian tropospheric chemistry model (STOCHEM). This model outputs concentrations of 26 species with a horizontal resolution of 5° latitude $\times$ 5° longitude and nine vertical layers extending from surface up to 150 hPa (Collins et al., 1997). The initial and boundary conditions for the inner three domains are provided by the coarser domain.

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3. Model evaluation and simulation results

3.1. Meteorological predictions

The MM5 modelled near-surface temperature, wind speed and wind direction were compared to hourly weather observations from 29 land surface stations archived at the Met Office Integrated Data Archive System (MIDAS; UK Meteorological Office, 2006). These quantities were selected because they reflect the nature of the local thermodynamic circulation and govern contaminant distributions in air quality models. Several standard statistical measures were employed for the evaluation. These include the mean observed and modelled values, the mean bias (MB), the normalized mean bias (NMB), the mean error (ME), the normalized mean error (NME), the root-mean-square error (RMSE) and the index of agreement (IA). While calculation of these statistics is straightforward for wind speed and temperature, it poses problem for circular data, i.e. wind direction. To get around this problem, a 'modified' wind direction, following Lee and Fernando (2004), was used, where a 360° is either added to or subtracted from the predicted wind direction to minimize the absolute difference between the observed and predicted wind direction. Table 1 summarizes the performance statistics for MM5 calculated based on near-surface data from the 29 stations displayed in Fig. 1b, along with definitions of statistical measures. These values reflect averages over space (all monitoring stations within the 3 km-grid CMAQ domain) and time (all hours in the simulation period). The table shows small MEs (and biases) for the 2-m temperature and 10-m wind speed and direction, and low RMSE. In general the model captured the observed near-surface temperatures and winds quite well. Overall IA values of 0.97 for 2-m temperature and 0.75 and 0.93 for 10-m wind speed and direction, were achieved. The IA, which is a measure of how well the solutions represent the spatial variability (Willmott et al., 1985), indicates a good overall agreement between observations and model predictions.

A qualitative comparison of the modelled meteorological fields with observations is shown in Fig. 2. The figure shows the vertical profiles of wind speed, wind direction and temperature at Herstmonceaux (see Fig. 1b for location) on 26 June 2001 when the highest temperature was experienced. Herstmonceaux is the only radiosounde station within the 3-km grid MM5 domain. The model is able to reproduce the major features of the observed wind and temperature fields and the modelled profiles show a generally good agreement with measurements.

3.2. O_3, NO_2 and $PM_{2.5}$ predictions

Hourly surface concentrations of O_3, NO_2 and $PM_{2.5}$ obtained from the UK AURN (http://www.airquality.co.uk/archive/index.php) and London Air Quality Network (LAQN, http://www.londonair.org.uk/london/asp/default.asp) were used in the evaluation. The locations of air quality monitoring sites used in the evaluation are shown in Fig. 1b. Only monitoring sites reporting measurements for at least 75% of the hours in the studied period were included in the analysis. Monitoring sites are presented for four categories as used in the networks, i.e. rural, urban background, suburban and urban centre. Traffic monitoring sites are not

Table 1
Performance statistics of modelled temperature and wind speed and direction

Variables	Temp. (K)	WS ($m\,s^{-1}$)	WD (deg)	Statistic definition[a]
Mean obs. $\bar{O}$	18.2	3.4	155	$(1/N)\sum_1^N C_o$
Mean sim. $\bar{M}$	18.9	3.0	158	$(1/N)\sum_1^N C_m$
Total N	4599	4414	4391	
MB	0.7	−0.3	7.3	$(1/N)\sum_1^N (C_m - C_o)$
NMB (%)	3.7	−8.8	4.7	$((1/N)\sum_1^N (C_m - C_o)/\bar{O}) \times 100\%$
ME	1.4	1.2	28.2	$(1/N)\sum_1^N \lvert C_m - C_o\rvert$
NME (%)	7.6	36.6	18.2	$((1/N)\sum_1^N \lvert C_m - C_o\rvert/\bar{O}) \times 100\%$
RMSE	1.7	1.5	42.6	$\sqrt{(1/N)\sum_1^N (C_m - C_o)^2}$
Index of agreement	0.97	0.75	0.93	$1 - \sum_1^N (C_m - C_o)^2 / \sum_1^N (\lvert C_m - \bar{O}\rvert + \lvert C_o - \bar{O}\rvert)^2$

[a]m = modelled, o = observed.

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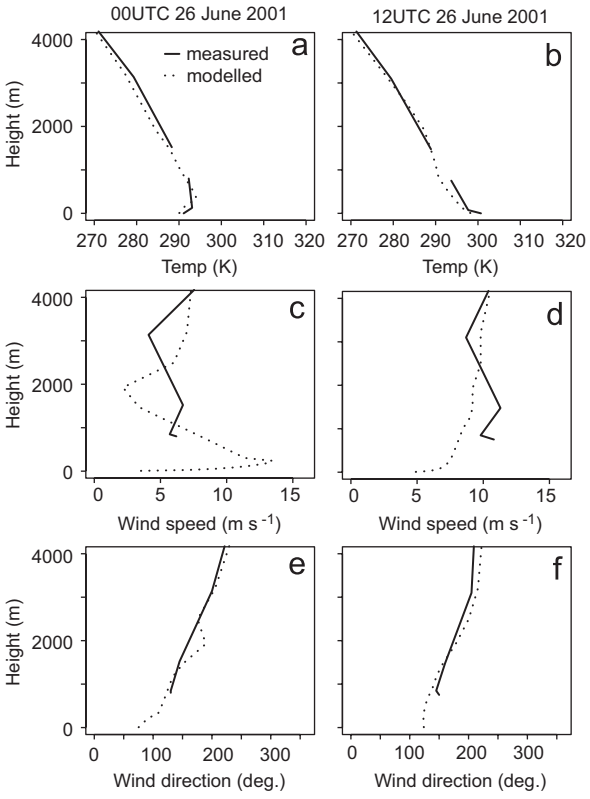

Fig. 2. Comparison of modelled and measured vertical profiles of temperature, wind speed and wind direction at radiosounde station Herstmonceaux on 26 June 2001.

considered due to their poor representativeness in the model resolution. The model evaluation focuses on the 3-km grid domain. The results from a coarser grid domain, i.e. 9 km, are, therefore, only compared against the observations obtained for locations within the 3-km grid domain. Model values used for evaluation were extracted from the first model level (about 14 m AGL). In addition to the statistical measures used in Section 3.1, the correlation coefficient R was also calculated to quantify the model performance. Although no single set of evaluation techniques is universally recommended, the statistical measures used here have been widely used in recent regional air quality model evaluations (Hogrefe et al., 2004; Eder and Yu, 2006).

3.2.1. O_3

The modelled O_3 time series from both 3- and 9-km resolution simulations are compared with the measured values in Fig. 3 at six 'representative' sites chosen to cover the different types of monitoring sites. The model is in general able to capture the diurnal O_3 variations for most of the days and exhibits overall good agreement with measurements (with $R = 0.7$, see Table 2). The modelled O_3 mixing ratios are in close agreement with observations during most of the nighttime hours at the rural site. While CMAQ underpredicts the maximum O_3 mixing ratios on high-O_3 days, for example, 25 and 26 June, it tends to overpredict the maximum and minimum O_3 mixing ratios for most low-O_3 days, especially at urban centre and suburban sites, with most of the overpredictions occurring during night and early morning hours, indicating the underestimaion of O_3 titration, which is consistent with the underprediction of NO_2 mixing ratios shown in Fig. 6 and Table 3. Overall, the 3 km-grid simulation gives comparable or slightly better predictions than the 9 km-grid simulation, especially at urban and suburban sites.

Fig. 4 presents a scatter plot of modelled versus measured hourly O_3 mixing ratios for all the modelling hours and sites. The most important feature of model errors revealed by this plot is the overprediction of hourly O_3 concentrations in the lower range and underprediction in the higher range of O_3 concentrations. Table 2 summarizes the corresponding O_3 performance statistics for the 3 km-grid simulation. The statistics for 9 km-grid simulation are similar to those for 3 km-grid simulation and are not shown for brevity. The MB (−2.0 ppb) and NMB (−5.3%) show an underprediction of hourly O_3 mixing ratios. For different categories of sites (not shown for brevity), NMB ranges from −7.8% at urban background sites to −2.7% at suburban sites. The ME and NME are 11.9 ppb and 32.4%, respectively, with values ranging from 23.7% at rural sites to 40.1% at urban centre sites. In total, about 83% of all modelled values are within a factor of two of the corresponding measured O_3 concentrations. The statistical measures for daily maximum 8-h average O_3 mixing ratios a also calculated and summarized in the last column of Table 2. The daily maximum 8-h average O_3 mixing ratios were underpredicted by 14% based on NMB. The values of MB and NMB are higher than those for hourly O_3, indicating that the model is less accurate in reproducing the highest hourly O_3 values experienced during the episode. US EPA has suggested informal criteria for regulatory modelling practices of ±5–15% for NMB and 30–35% for NME (Russell and Dennis, 2000). It is seen from Table 2 that our evaluation statistics fall well within the suggested values. The

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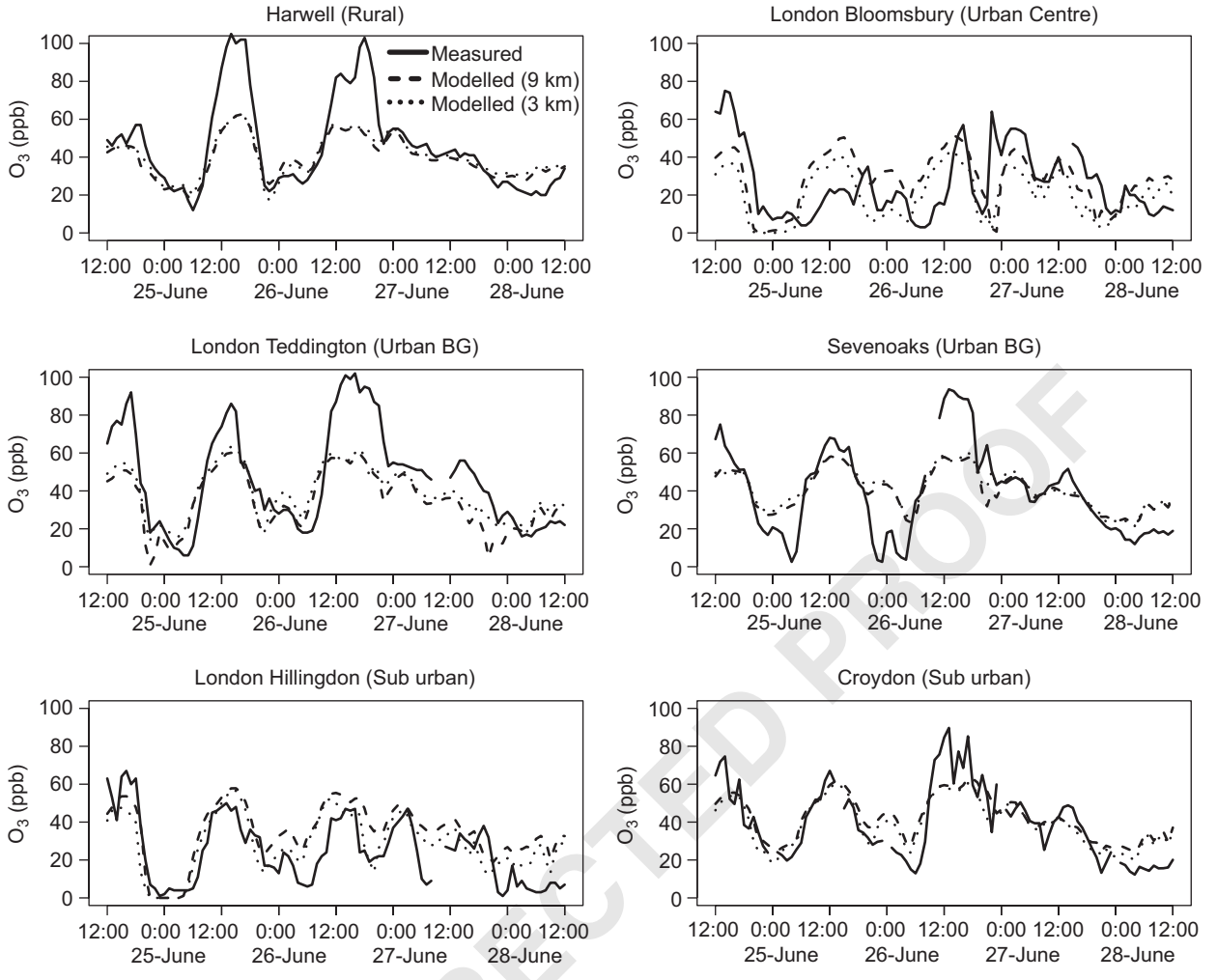

Fig. 3. Comparison of measured and modelled O_3 mixing ratios at a selection of six 'representative' sites, chosen to cover the different types of monitoring sites within the 3 km-grid domain (see Fig. 1b for locations).

Table 2
Performance statistics for modelled surface ozone concentrations (ppb)

Statistics	Hourly O_3	Max. 8-h mean O_3
Mean obs. $\bar{O}$	37.0	54.4
Mean sim. $\bar{M}$	35.0	46.8
Total N	2128	110
MB	−2.0	−7.6
NMB (%)	−5.3	−14.0
ME	11.9	14.6
NME (%)	32.4	26.9
RMSE	15.4	18.2
R	0.70	0.40
IA	0.79	0.53
% Within factor of 2 of measured	83.3	94.5

Table 3
Performance statistics of modelled surface NO_2 concentrations (ppb)

Statistics	3 km	9 km
Mean obs. $\bar{O}$	21.3	
Mean sim. $\bar{M}$	15.1	14.2
Total N	2762	
MB	−6.2	−7.1
NMB%	−28.9	−33.5
ME	9.7	10.5
NME%	45.5	49.2
RMSE	13.5	13.0
R	0.64	0.58
IA	0.75	0.70
% Within factor of 2 of measured	62	59

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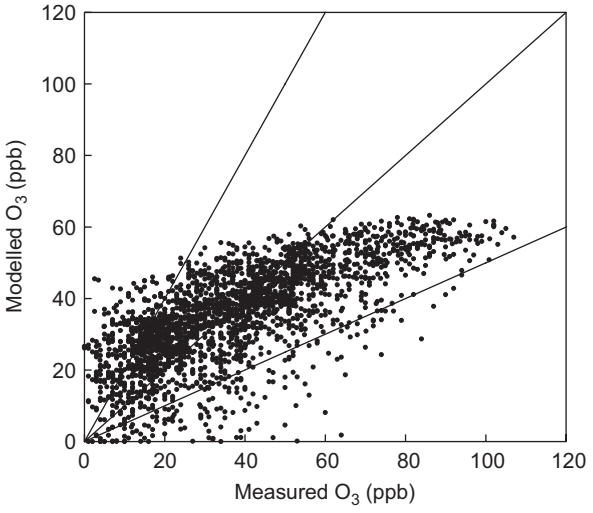

Fig. 4. Measured versus modelled hourly O_3 mixing ratio. Modelled values were extracted from the 3 km-grid simulation; 1:2, 1:1 and 2:1 reference lines are provided.

performance of our modelling system ~~is also~~ comparable ~~to~~ or better than other air quality models used in Europe. For example, Schmidt et al. (2001), using the European scale Eulerian chemistry transport model CHIMERE, obtained a R of 0.58–0.81 for daily maximum O_3 concentrations at representative rural sites in the UK and Dufour et al. (2005), using the new MOdèle de Chimie Atmosphérique à Grande Echelle (MOCAGE) 3-D multiscale chemistry and transport model, reported a R of 0.44–0.86 for hourly O_3 concentrations in two summer episodes that occurred during the Experience sur Site pour COntraindre les Modèles de Pollution atmosphérique et de Transport d' Emission (ESCOMPTE) field programme. In general, CMAQ performs better at rural sites than at urban sites, where O_3 prediction is more sensitive to the representation of mixing near sources, errors in meteorological parameters (e.g. boundary layer height) and titration by local emissions.

From the above analyses it is seen that under moderate photochemical activity, CMAQ can reproduce the observed O_3 concentrations, but the model tends to underpredict peak O_3 mixing ratios (>55 ppbv) during this typical summer episode. The underprediction of peak O_3 concentrations on high-O_3 days indicates that the O_3-production chemistry may not be sufficiently reactive. A further examination of the surface O_3 time series shown in Fig. 3 reveals that urban background (suburban) sites located upwind of the London metropolitan area (see Fig. 5a for the wind field), for example Sevenoaks (Croydon), experienced a nighttime overprediction (peak O_3 underprediction) that are not seen for sites located downwind, indicating that long-range transport of O_3 and its precursors from the European continent may contribute to the elevated O_3 concentration. This is also indicated by the more significant underprediction of NO_2 for urban background/suburban sites located upwind of the London metropolitan area (see Fig. 6). Some early studies (e.g. AQEG, 2004; Derwent et al., 2003) have also suggested that the contribution from European emissions could be an important factor in high-O_3 episodes that occur in summer in the UK. Surface O_3 distribution from the 3 km-grid simulation at 15 UTC is shown in Fig. 5a. The modelled spatial distributions of O_3 were similar to that measured for 25 and 26 June, but the model did not reproduce the O_3 mixing ratios that exceeded 55 ppb at sites affected by urban plume on 25 June and at most of the observational sites on 26 June. It can be inferred from the wind fields that both London emissions and European emissions may have contributed to the spatial distribution of O_3 on 25 June while the transport into the UK of already polluted air from the European boundary layer may be more important on 26 June. To investigate the possible causes of the underestimations, sensitivity studies with doubled anthropogenic NO_x or VOC emissions were carried out. However, it should be noted that there are several other sources of uncertainties in the model, including inaccurate meteorological predictions (e.g. cloud cover, PBL height) and less well represented physical/chemical processes (e.g. entrainment of regionally polluted air from aloft, enhanced VOC reactivity), which also influence the prediction of photooxidants concentration and should be addressed in future studies. For example, a recent study by Lee et al. (2006) suggests that entrainment of regionally polluted air from aloft may contribute to the rapid increase of O_3 in the morning on high-O_3 days. In addition, chemical processes leading to O_3 production under episodic high temperature conditions may be substantially different from that occurring at normal conditions. It was found that the total VOC reactivity to OH could be doubled under high-temperature conditions such as those that occurred during the August 2003 heatwave (Lee et al., 2006). It is seen from Fig. 5b that doubling the anthropogenic NO_x emission leads to less O_3 over most of the domain on 25 June and over northeast part of

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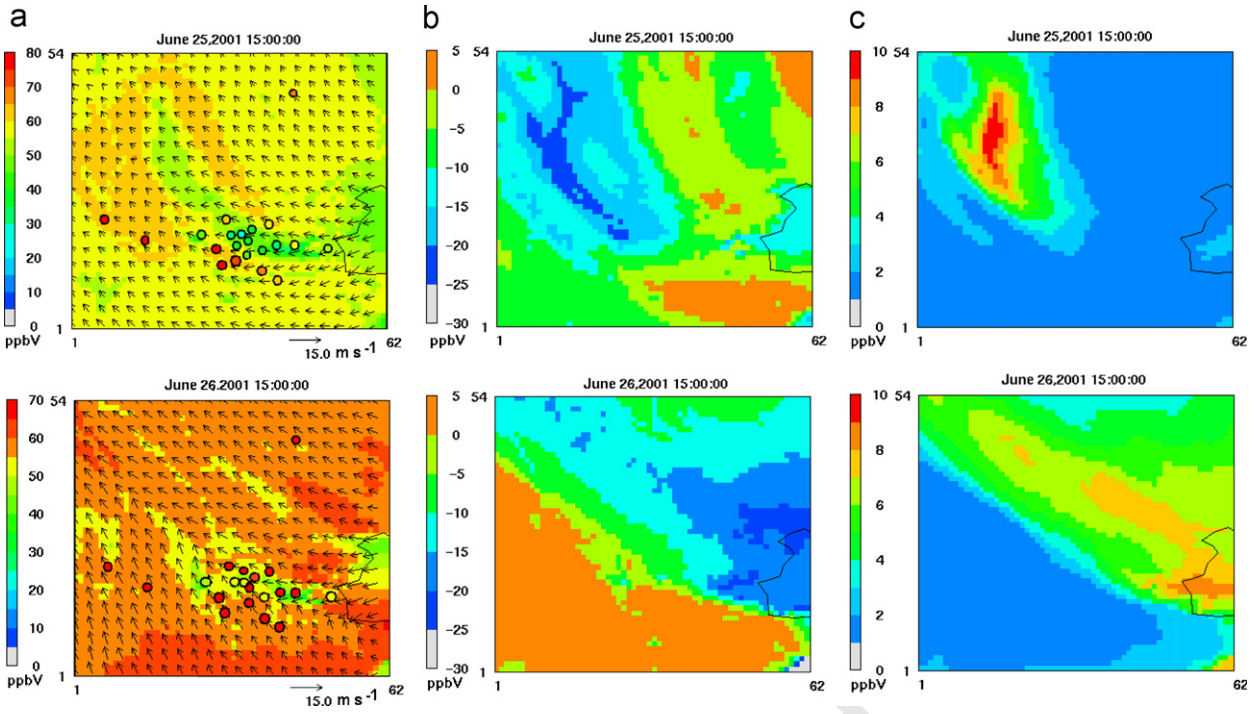

Fig. 5. (a) Spatial distribution of ozone for two consecutive days at 15 UTC for base case simulation; (b) difference in ozone between cases with doubled anthropogenic NO_x emission and base emission and (c) difference in ozone between cases with doubled anthropogenic VOC emission and base emission. The coloured points (same colour scale as model results) indicate the measured values.

the domain on 26 June but it increases O_3 concentration over the southwest domain due to the transport of European-derived O_3 from the south and east model boundaries on 26 June. On 25 June, doubling anthropogenic VOC emission results in increasing O_3 concentrations in urban plume, where high NO_x are present and oxidation is VOC limited, while on 26 June it increases the O_3 transported from the east model boundary. The above analyses indicate that both NO_x and VOC emissions may be underestimated in the EMEP inventory, which leads to the underprediction of O_3 and its precursors transported into the studied domain under episodic conditions. Although doubling of NO_x or VOC emissions reduced the low bias in the modelled O_3 concentration compared to the observations, the model still could not reproduce the high O_3 observed during the episode, indicating the lack of reactivity of the modelled atmosphere.

3.2.2. NO_2

Fig. 6 compares the measured NO_2 time series with the modelled results at the same six sites as for O_3. Both the diurnal variations and magnitudes of NO_2 mixing ratios are well captured by the model at the rural site. The model performs better at London Teddington (located near the London metropolitan area) than at Sevenoaks (located upwind of the London metropolitan area), suggesting the underprediction of O_3 and its precursors transported from the European continent as discussed in Section 3.2.1. The NO_2 mixing ratios were appreciably underpredicted at suburban and urban centre sites most of the time, indicating the underestimation of NO_x emissions in these areas. The appreciable difference of predicted NO_2 mixing ratios with different horizontal grid resolutions indicates that the nonlinearity of chemical reactions and heterogeneity associated with precursor emissions have a significant impact on model predictions.

Fig. 7 shows a scatter plot of the modelled versus measured hourly NO_2 mixing ratios for all hours and sites. Overall, the model reproduced about 62% of the hourly NO_2 mixing ratios within a factor of two of the measurement. Table 3 summarizes the hourly NO_2 performance statistics for both the 3 km- and the 9 km-grid resolutions for all the sites. The statistics for NO_2 show much larger bias and error when compared to the same statistics for O_3

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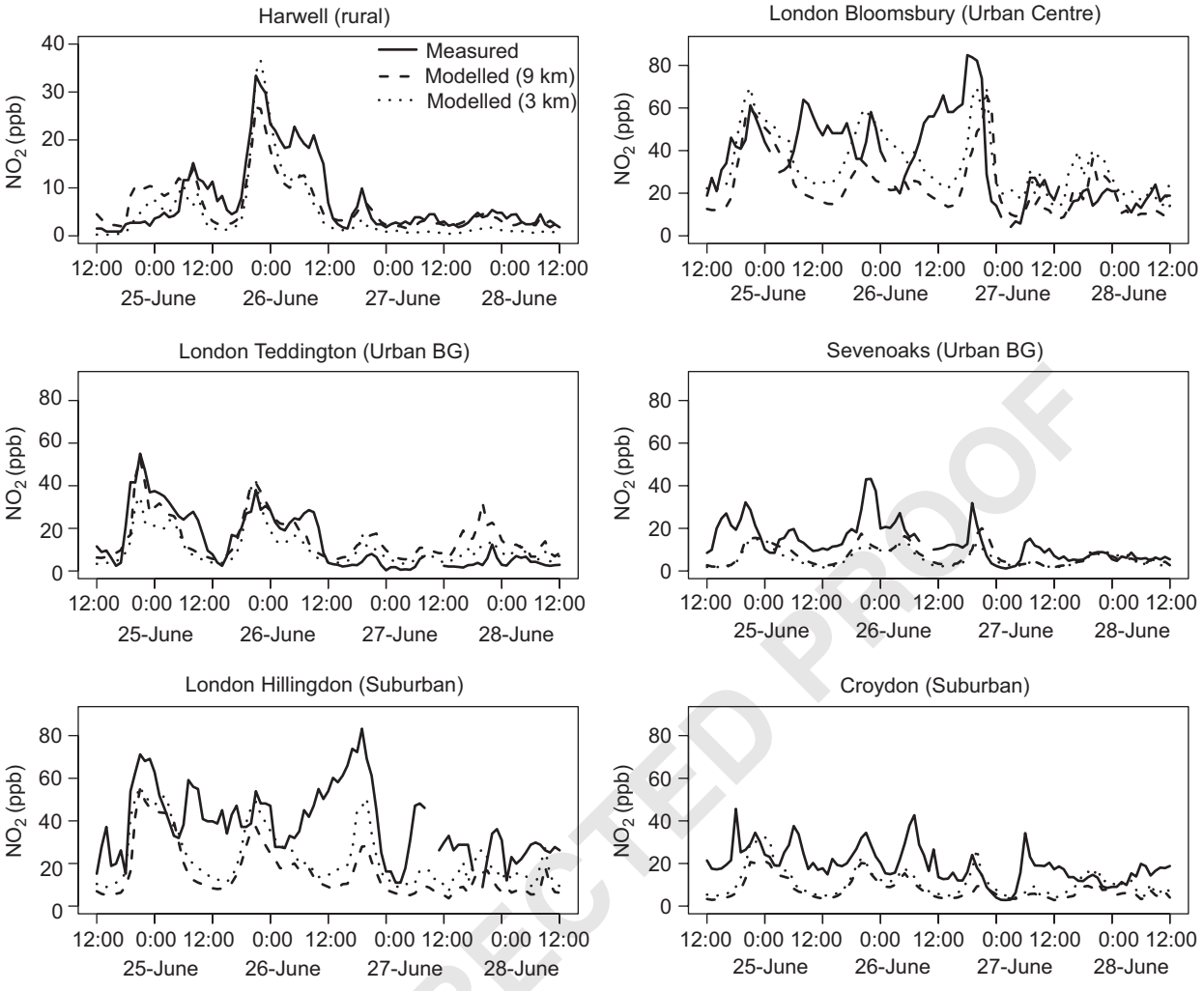

Fig. 6. Comparison of measured and modelled NO_2 mixing ratios at the same six sites as shown in Fig. 3.

due to the generally ~~more~~ sensitivity of NO_2 to errors in emissions and meteorology, especially under stagnant conditions. Overall, the model underpredicted NO_2 concentrations with a MB of −6.2 ppb and a NMB of −28.9% for the 3 km-grid simulation and a MB of −7.1 ppb and a NMB of −33.5% for the 9 km-grid simulation. The ME and NME values over all hours and sites are 9.7 ppb and 45.5%, respectively, for the 3 km-grid simulation and 10.5 ppb and 49.2%, respectively, for the 9 km-grid simulation. Underprediction of O_3 precursors is experienced by many currently used photochemical models (Russell and Dennis, 2000). The negative NMB of NO_2 from our study falls well within the range of −20% to −50% inferred from other studies (Hanna et al., 1996).

3.2.3. *Fine particulate matter ($PM_{2.5}$)*

Hourly measurements of $PM_{2.5}$ are available at four sites within the evaluation domain (one of them is located at roadside and is not included in the present study). Fig. 8a compares the measured and modelled time series of $PM_{2.5}$ concentrations at Harwell, Rochester and London Bloomsbury. Both the 9 km- and 3 km-grid resolution simulations failed to reproduce the temporal variations and magnitude of the measured $PM_{2.5}$ mass concentrations. Overall, the model tends to underpredict the $PM_{2.5}$ mass concentrations with a MB of $-8.7\,\mu g\,m^{-3}$ and a NMB of −45% over the three sites. The ME and NME are $9\,\mu g\,m^{-3}$ and 49.3%, respectively. These values indicate an overall significant underprediction of $PM_{2.5}$ mass concentrations. A scatter plot of the modelled versus

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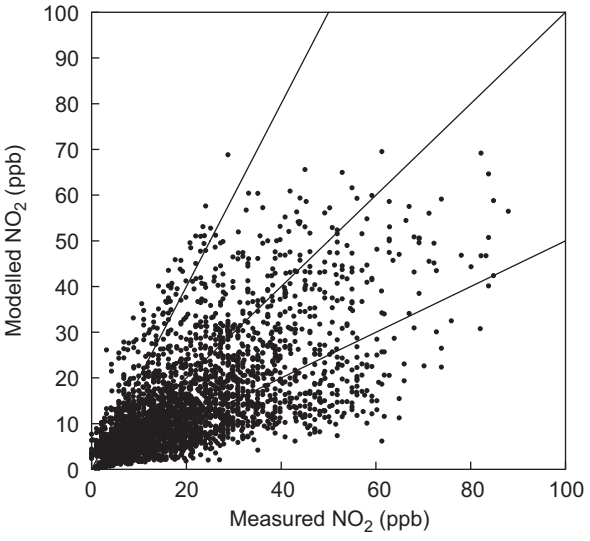

Fig. 7. Measured versus modelled hourly NO_2 mixing ratio. Modelled values were extracted from the 3 km-grid simulation; 1:2, 1:1 and 2:1 reference lines are provided.

measured $PM_{2.5}$ mass concentrations for all hours and sites is shown in Fig. 8b. Only 45% of all modelled $PM_{2.5}$ concentrations are within a factor of two of the corresponding measurement. These statistics are consistent with the current performance expected from most air quality models for particulate matter (e.g. Seigneur, 2001; Bessagnet et al., 2004; Zhang et al., 2006b; Vautard et al., 2007). The limited availability of $PM_{2.5}$ mass concentration and composition data makes the results of the model performance analysis less conclusive and robust; however, the analysis still provides a general indication of the model performance. Several factors could contribute to the underprediction of $PM_{2.5}$ mass concentrations, including uncertainties in emissions of particulate matter precursor gases and primary particulate matter and uncertainties in the model treatment of chemistry and thermodynamics of aerosols. For example, sea salt and the interactions between the fine- and coarse-mode particles are not treated in CMAQ v4.4 (e.g. Zhang et al., 2006c); emissions inventory (e.g. EMEP) may be deficient, as some biogenic sources are missing and emissions of re-suspension related to traffic on paved or dirt roads or related to wind are not included. Further studies are needed to evaluate the relative importance of these parameters on overall model performance for $PM_{2.5}$ prediction.

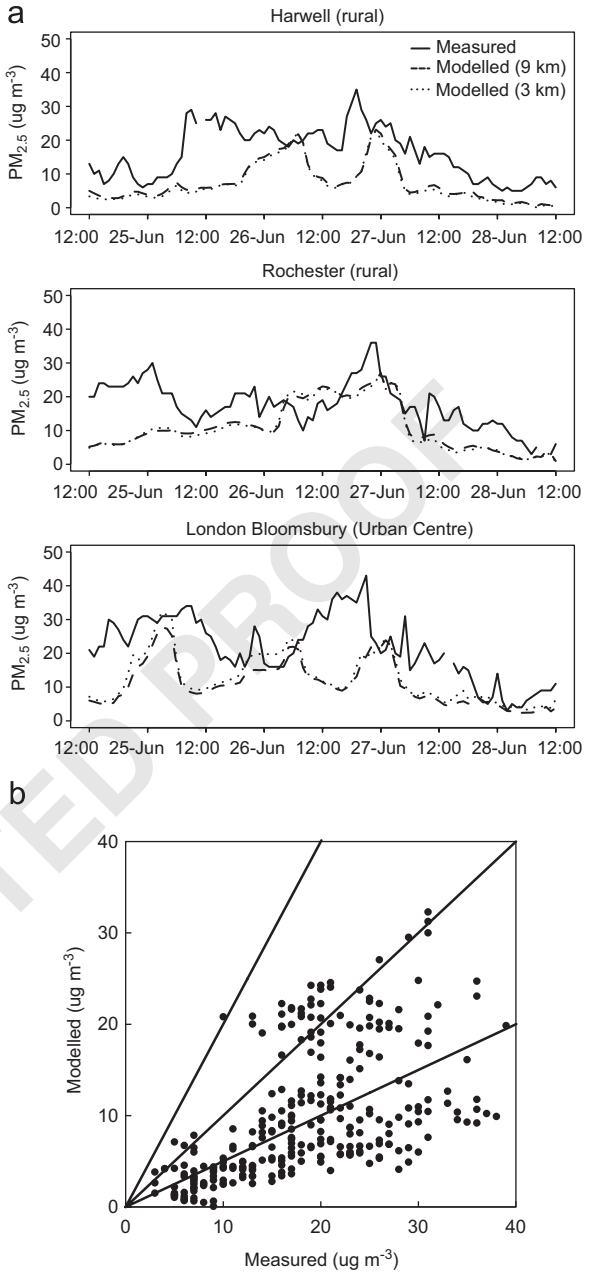

Fig. 8. (a) Comparison of measured and modelled time series of $PM_{2.5}$ mass concentrations at Harwell, Rochester and London Bloomsbury sites; (b) measured versus modelled hourly $PM_{2.5}$ mass concentration; 1:2, 1:1 and 2:1 reference lines are provided.

4. Discussions

The above results on O_3 predictions reveal a general underprediction of the hourly and daily maximum 8-h mean O_3 mixing ratios by CMAQ for the studied area, especially on high-O_3 days. Underprediction of daily maximum O_3 mixing ratios for

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most UK rural sites was also reported by Schmidt et al. (2001) and was attributed to the underestimation of boundary concentrations. Previous studies on regional O_3 distribution across the British Isles using both the EMEP and Edinburgh Lancaster Model for Ozone (ELMO) models also indicated the difficulty for these models to capture the high O_3 levels occurring in the southern England (Metcalfe et al., 2002). Additional sensitivity study with modified boundary conditions (not shown) indicates that boundary condition has very limited effect on surface O_3 prediction, while correct emission input is more important for a better model performance as shown in Section 3.2.1.

Previous study by Clapp and Jenkin (2001) shows that the level of OX (the sum of O_3 and NO_2) at a given location is made up of NO_x-independent regional contribution (the intercept) and NO_x-dependent local contribution (the slope). It is thus possible to estimate which part was underestimated by CMAQ by comparing measured and modelled OX versus NO_x relationship. Daylight average analyses were carried out for June 2001 using data from 16 monitoring sites, where O_3, NO and NO_2 were simultaneously measured within the innermost model domain. Following Clapp and Jenkin (2001), the data were separated as 'episode' days (with daylight-averaged OX mixing ratio at Teddington >50 ppb) and 'non-episode' days, which resulted in 24, 25 and 26 being selected as 'episode' days. It is seen from Fig. 9 that the level of OX was significantly higher on 'episode' days (black open dots), as a result of the increased regional contribution (the intercept increased from 41 ppb on 'non-episode' days to 73 ppb on 'episode' days) and a 3% higher local contribution. Thus the enhanced OX levels occurring during this episode is a contribution of regional transport in a combination of local processes, which is consistent with the analyses shown in Section 3.2.1. CMAQ significantly underpredicted the regional contribution during the episode. The model also underpredicted the local contribution indicated by the smaller increase of OX with NO_x than is observed (16.9% versus 18.4%). These analyses further demonstrate the importance of long-range transport of O_3 and its precursors from the European continent to the elevated O_3 concentrations during the episode.

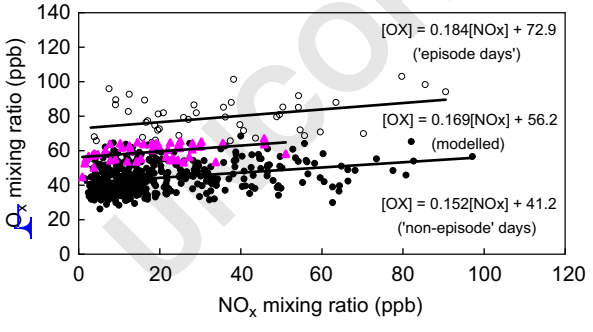

Fig. 9. Variation of daylight-time-averaged mixing ratios of OX with the level of NO_x. Data are presented for each day of June 2001 at 16 sites. The lines were defined by regression analysis of observed 'non-episode', 'episode' and modelled 'episode' days. Black solid dots, black open dots and triangles are for 'non-episode' days, 'episode' days and modelled 'episode' days, respectively.

As mentioned in Section 2, NO_x emissions from soil are not included in the present study, which may also lead to underprediciton of O_3 mixing ratios. Stohl et al. (1996) studied the importance of NO_x emissions from soil on O_3 production in Europe and argued that although on European average, biogenic NO emissions account for only 4% of anthropogenic NO emissions, they can be relevant in rural areas. The inclusion of NO_x emissions from soil and their possible effect on O_3 predictions will be a subject of future work.

5. Conclusions

A performance evaluation of MM5–SMOKE–CMAQ modelling system for southeast England, UK, for a summer photochemical episode has been presented. The simulated concentrations of O_3, NO_2 and $PM_{2.5}$ were compared with ground-level observations from the AURN and LAQN. The evaluation shows that CMAQ tends to underpredict hourly O_3 mixing ratios on high-O_3 days and overpredict the maximum and minimum O_3 mixing ratios for most low-O_3 days. Sensitivity studies and analysis of ambient OX levels (sum of O_3 and NO_2) as a function of NO_x reveal that the transport of O_3 and its precursors from the European continent was significantly underpredicted by CMAQ, which likely resulted from the underestimation of European emissions, except other physical/chemical processes, e.g. entrainment of regionally polluted air from aloft and enhanced VOC reactivity under episodic conditions, that are not well represented in the model.

In terms of NO_2, the model captured the magnitudes and temporal variations generally well, but produced much larger bias and error than those for O_3 with significant underpredictions at urban

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centre and suburban areas, which may be due to the inaccurate meteorological field (e.g. PBL height) and missing or incorrect emissions. For example, NO_x emissions from soil were not accounted for in the present study.

For $PM_{2.5}$, CMAQ with both resolutions (9 and 3 km) significantly underpredicted the mass concentrations and failed to reproduce the temporal variations, with only 45% of all modelled $PM_{2.5}$ mass concentrations falling within a factor of two of the corresponding measured values. While CMAQ performance for O_3 and $PM_{2.5}$ was not sensitive to model resolutions, higher resolution was found to be beneficial for properly simulating NO_2 mixing ratios.

The overall performance of MM5–SMOKE–CMAQ modelling system is comparable to or better than similar model predictions by other models used ~~in the~~ ~~Europe~~. Further studies, however, are needed to explore how the model response to uncertainties linked to different processes ~~of tropospheric chemistry modelling,~~ namely large-scale pollution transport, refinement of emission inventory, as well as representation of the physical and chemical processes (e.g. enhanced VOC reactivity under episodic conditions). Notably, photochemical episode may vary from event to event and the evaluation of one episode ~~is by no means comprehensive~~. More cases need to be conducted to increase the confidence in these results.

Acknowledgements

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