Atmos. Chem. Phys. Discuss., 9, 19509–19544, 2009
www.atmos-chem-phys-discuss.net/9/19509/2009/

Atmospheric Chemistry and Physics Discussions

This discussion paper is/has been under review for the journal *Atmospheric Chemistry and Physics (ACP)*. Please refer to the corresponding final paper in *ACP* if available.

Modelling surface ozone during the 2003 heat wave in the UK

M. Vieno[1,2], A. J. Dore[2], D. S. Stevenson[1], R. Doherty[1], M. R. Heal[3], S. Reis[2], S. Hallsworth[2], L. Tarrason[4], P. Wind[4], D. Fowler[2], D. Simpson[4,5], and M. A. Sutton[2]

[1]School of GeoSciences, The University of Edinburgh, UK
[2]Centre for Ecology and Hydrology, Penicuik, UK
[3]School of Chemistry, The University of Edinburgh, UK
[4]Norwegian Meteorological Institute, Oslo, Norway
[5]Dept. Radio and Space Sci., Chalmers University of Technology, Gothenburg, Sweden

Received: 15 July 2009 – Accepted: 26 August 2009 – Published: 18 September 2009

Correspondence to: M. Vieno (mvieno@staffmail.ed.ac.uk)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

A high resolution ($5\times5\,km^2$) UK-scale chemistry-transport model (EMEP4UK) is used to study ground-level ozone (O_3) during the August 2003 heat-wave. Meteorology is generated by the Weather Research and Forecast (WRF) model, nudged every six hours with reanalysis data. We focus on SE England, where hourly average O_3 reached up to 140 ppb during the heat-wave. EMEP4UK accurately reproduces observed annual and diurnal cycles of surface O_3 at urban and rural sites. Elevated O_3 and much of its day-to-day variability during the heat-wave are well captured. Key O_3 precursors, nitrogen dioxide and isoprene (C_5H_8), are less well simulated, but show generally accurate diurnal cycles and concentrations to within a factor of ~2–3 of observations. The modelled surface O_3 distribution has an intricate spatio-temporal structure, governed by a combination of meteorology, emissions and photochemistry. A series of sensitivity runs with the model are used to explore the factors that influenced O_3 levels during the heat-wave. Various factors appear to be important on different days and at different sites. Ozone imported from outside the model domain, especially the south, is very important on several days during the heat-wave, contributing up to 85 ppb. Dry deposition of O_3, when completely switched off, elevated simulated O_3 by up to 50 ppb, and this may have been an important factor on several days. Modelled C_5H_8 concentrations are generally best simulated if C_5H_8 emissions are changed from the base emissions: typically doubled, but elevated by up to a factor of five on some days. Accurately modelling the exact positions of individual plumes of anthropogenically emitted nitrogen oxides and volatile organic compounds is crucial for the successful simulation of O_3 at a particular time and location. Variations in surface temperature of ±5 K were found to have impacts on O_3 of typically less than ±10 ppb.

ACPD

9, 19509–19544, 2009

1 Introduction

In the UK, episodes of increased concentrations of ground level ozone are often caused by elevated temperatures associated with summertime anticyclonic conditions (e.g. Jenkin et al., 2002). During the first two weeks in August 2003, a blocking area of high atmospheric pressure centred over Scandinavia caused very high temperatures (>35°C) for several consecutive days over parts of the UK and central Europe (Schar and Jendritzky, 2004; Solberg et al., 2008; Trigo et al., 2005; Vautard et al., 2005). This was associated with a series of afternoon ozone peaks, reaching above 90 ppb, in the south of England (Lee et al., 2006). The heat-wave period was coincident with the Tropospheric ORganic CHemisty (TORCH) field campaign (Lee et al., 2006), which provided detailed measurements of ozone concentrations and its precursors, including isoprene, at a site in Writtle about 70 km NE of London. The high temperatures and high levels of ozone experienced during the 2003 heat wave had a substantial effect on human health (Stedman, 2004).

In this study we investigate the causes of the elevated ozone levels using a high resolution ($5\times5\,km^2$ grid) chemical transport model over the UK domain. We first show that the model is able to simulate hourly ozone measurements realistically from a range of sites over SE England during August 2003, including measurements made as part of the TORCH campaign. We then conduct a series of sensitivity runs to investigate the influences of a variety of different meteorological and chemical factors (emissions of biogenic isoprene, emissions of anthropogenic NO_x ($NO+NO_2$) and volatile organic compounds (VOCs), temperature, ozone dry deposition, and transport) that contributed to the high ozone episodes in this region during the August 2003 heatwave.

2 Model description and set-up

The EMEP4UK model framework is a nested regional chemistry transport model (CTM) driven by high-resolution meteorology and national emissions that is used to produce

ACPD
9, 19509–19544, 2009

a detailed representation of the physical and chemical state of the atmosphere over Europe and, in particular, over the UK (Vieno et al., 2009). The underlying CTM is the EMEP Unified Model (Simpson et al., 2003a), which has been modified in recent years to enable application on spatial scales ranging from the $5\times5\,km^2$ grid used here for the UK to the global scale (Jonson et al., 2007).

For this study, the EMEP4UK model was driven by the Weather Research Forecast (WRF) model (http://www.wrf-model.org) with a resolution of $5\times5\,km^2$. The WRF model included data assimilation (Newtonian nudging) of the numerical weather prediction model meteorological reanalysis from the US National Center for Environmental Prediction (NCEP)/National Center for Atmospheric Research (NCAR) Global Forecast System (GFS) at 1° resolution, every 6 h.

WRF was applied here using a nested domain approach, with an outer domain resolution of $50\times50\,km^2$ (approx.), an intermediate domain resolution of $10\times10\,km^2$ (only needed by the WRF model) and an inner domain with a resolution of $5\times5\,km^2$, to provide meteorological data at the required horizontal and vertical resolution. Simulations were performed over each of these domains; the results from the outermost domain being used as boundary conditions to the intermediate domain and so on.

The innermost domain covers the British Isles plus parts of France, Denmark, Holland and Belgium. As in the standard EMEP model, the model is everywhere divided vertically into 20 layers using terrain following coordinates with resolution increasing towards the surface. The vertical column extends from the surface (centre of the surface layer ~45 m) up to 100 hPa (~16 km). Modelled concentrations are calculated at 3 m above the surface plant or other canopy by making use of the constant-flux assumption and definition of aerodynamic resistance (Simpson et al., 2003b). The WRF coarse grid of $50\times50\,km^2$ resolution was used to drive the EMEP Unified Model across the European domain to calculate the chemical initial conditions and boundary conditions for the EMEP4UK model (driven by the inner WRF domain $5\times5\,km^2$). The EMEP Unified Model itself ($50\times50\,km^2$) was initialised with a climatologically-derived ozone boundary and initial conditions (Logan, 1999). To more accurately simulate the import

ACPD
9, 19509–19544, 2009

of ozone in a specific year, the so called "Mace Head" adjustment was applied (Simpson et al., 2003a). This adjustment uses monthly "clean-air (Atlantic)" observations from the Mace Head site on the west coast of Ireland, adjusting the monthly Logan climatology to match Mace Head data, and it was only applied to the EMEP Unified Model at $50 \times 50\,km^2$ resolution.

The current EMEP Unified model is a development of the 3-D chemical transport model of Berge and Jakobsen (1998), extended with photo-oxidant chemistry (Simpson et al., 1993; Andersson-Sköld and Simpson, 1999) and the EQSAM gas/aerosol partitioning model (Metzger et al., 2002). Two types of emissions are present in the model: anthropogenic and natural. For the UK, anthropogenic emissions of NO_x, NH_3, SO_2, $PM_{2.5}$, PMCO (coarse particulate matter), CO, and non-methane VOC (NMVOC) are derived from the UK's National Atmospheric Emissions Inventories (NAEI; Dore et al., 2008; Hellsten et al., 2008). Elsewhere and for international shipping, EMEP emissions are used (www.emep.int). NMVOC are speciated into 10 reactive and one unreactive species, using emission-sector specific values as shown in Simpson et al. (2003a). Biogenic emissions of isoprene are based on Guenther et al. (1993) and Simpson et al. (1999), driven by land-use, temperature and light. Biogenic emissions of dimethylsulphide (DMS) are input as monthly average emission data, derived from Tarrason et al. (1995), and treated as SO_2 on input to the calculations. Emissions of NO_x from lightning are included as monthly averages (Köhler et al., 1995). Seasonally averaged aircraft emissions are included for NO_x from Gardner et al. (1997). Both aircraft and lightning emissions are provided as 3-D fields for the whole model domain. Natural soil NO_x emissions and non-anthropogenic biomass burning are not included. For CH_4 a constant mixing ratio over the whole domain is prescribed (Simpson et al., 2003a).

Sixteen basic land-use classes are used in the deposition module of the EMEP4UK model. For those vegetative landuse categories for which stomatal modelling is undertaken, the start and end of the growing season is specified and the development of leaf area index within this growing season is also modelled (Simpson et al., 2003a, b).

ACPD
9, 19509–19544, 2009

Dry deposition is calculated using a resistance analogy combined with stomatal and non-stomatal conductance algorithms (Emberson et al., 2000; Simpson et al., 2003a, b), whereas wet deposition uses scavenging coefficients applied to the 3-D rainfall.

Full details of the EMEP model are given in Simpson et al. (2003a) and Fagerli et al. (2004).

3 Methods

A full year simulation was performed for 2003 using EMEP4UK/WRF in the configuration described in Sect. 2. Ten further one-month sensitivity experiments were carried out to investigate the contributing factors to the elevated ozone in the southern UK during the 2003 August heat-wave. These were identical to the base experiment in all respects except that in each case a single meteorological or chemical variable was changed in the EMEP4UK 5×5 km^2 grid inner domain.

The first factor investigated was surface temperature, which was either increased or decreased by 5 K. This affected ozone by changing the chemical reaction fluxes in the lowest model layer, temperature-sensitive emissions of biogenic isoprene, and also dry deposition of ozone through the surface exchange scheme. The imposed change in temperature did not affect the dynamic meteorology, as the influence was limited to the EMEP4UK part of the code, and no feedbacks operate from EMEP4UK to WRF. Several experiments then varied emissions of specific species: anthropogenic VOC (±50%), anthropogenic NO_x (−50%), or biogenic isoprene (zero – no emissions–, 2×, and 5×). The focus of the isoprene experiments, generally the most important biogenic VOC with regard to ozone formation, was to investigate the importance of UK-generated isoprene on surface ozone formation. As an extreme test of the importance of dry deposition, a further experiment was conducted in which ozone dry deposition (both stomatal and non-stomatal) was entirely switched off. The final experiment fixed ozone at the EMEP4UK boundary to the monthly climatological value from Logan (1999) rather than using 3-hourly values from the EMEP 50×50 km^2 model. This

ACPD
9, 19509–19544, 2009

allowed the influence of import from the outer domain to be isolated. The ten sensitivity experiments are summarised in Table 1.

4 Results

4.1 Base 2003 simulation

4.1.1 Surface temperature

To demonstrate that surface temperatures simulated by WRF with data assimilation produce a realistic representation of the August 2003 heat-wave, we compare model output with independent measurements (i.e. data that were not used in the WRF assimilation) during the TORCH campaign. Figure 1a shows hourly surface temperatures calculated by WRF from the $5 \times 5\,km^2$ grid cell containing Writtle (51°44′12″ N, 0°25′28″ E), together with data from two instruments deployed during the TORCH campaign. WRF is able to simulate the diurnal and longer timescale variations quite well, although the model underestimates some peak temperatures. However temperatures during the heat-wave are generally well simulated with an overall, R^2 value of 0.9 (square of the correlation coefficient) for both sensor for modelled versus measured hourly temperature for August 2003 (University of Leicester sensor: slope 0.9 intercept of 0.7; University of Leeds sensor: slope 0.8 and intercept of 0.6). The UK Met Office weather station Wattisham (52°07′22″ N, 0°57′43″ E) observed temperature versus WRF model predicted temperature is shown in Fig. 1b (R^2=0.8, slope 0.9 and intercept 0.8).

4.1.2 Surface ozone

Observed, from the UK Automatic Urban and Rural monitoring Network (AURN), and model-simulated surface ozone for the hour 14:00–15:00 GMT for each day of 2003 is shown for two sites in SE England; Wicken Fen, a rural site (52°17′54″ N, 0°17′28″ E), and London Eltham, an urban background site (51°27′09″ N, 0°04′14″), in Fig. 2a and

ACPD
9, 19509–19544, 2009

b, respectively. The model closely simulates the seasonal variation of surface ozone at the two sites. Moreover the model is able to capture ozone peaks (>50 ppb) for the whole of 2003. The August episode is not exceptional – there are several episodes of similar magnitude, from late March to mid-September.

Simulated hourly surface ozone concentrations for August 2003 were compared with observations. Observation and model results from two AURN stations Wicken Fen, and London Eltham, are shown in Fig. 3a and b. Similarly, the model simulations are compared with observations at Writtle (TORCH) in Fig. 3c. Scatter plots of these data are shown in Fig. 4. In terms of R^2 for all the hourly August data, the model performs best at Wicken Fen (R^2=0.7), London Eltham (R^2=0.6) and worst at Writtle (R^2=0.5). Nevertheless, the model accurately simulates many of the high ozone days during the heat-wave in comparison to cooler days with lower ozone, and the typical diurnal variation of ozone at the three sites. Some of the reasons for discrepancy between the model and observations at Writtle are discussed in Sect. 4.2.

The spatial and temporal variability of simulated surface daily maximum ozone for the first 15 days of August 2003 is shown in Fig. 5. During this period a clear feature of elevated ozone building up after the 3rd of August, is visible across the south of England. The feature shows strong spatial and day-to-day variability. The detailed structure in the model O_3 field clearly illustrates how difficult it is to simulate this accurately at some sites, particularly those near sources or with other significant heterogeneous factors.

Modelled monthly mean concentrations of ozone for August 2003 are shown in Fig. 6 together with the location of the observation sites included in this study. The influence of surface NO_x emissions on these ozone fields is clearly evident along road corridors and over centres such as London, Birmingham and Manchester. This highlights the importance of detailed emissions to simulate properly the spatial pattern of ozone over the UK and, more generally, wherever discrete emissions are present (i.e. road, point sources etc).

ACPD
9, 19509–19544, 2009

4.2 Attribution of high surface ozone over Writtle during the 2003 heat wave

Figure 7 shows the results of the temperature sensitivity experiments on model-simulated surface ozone for the first 15 days in August 2003 at the Writtle site. Increasing and decreasing the surface temperature by 5°C increases and decreases the surface ozone concentrations by approximately similar amounts, up to 9 ppb.

The effects of the NMVOC sensitivity experiments on modelled surface ozone are shown in Fig. 8. In contrast to the sensitivity to temperature, the response of ozone to changes to NMVOC emissions varies over the 15 days in August. Indeed, halving the UK anthropogenic NMVOC emissions has only a small effect on simulated surface ozone concentration on most days, whereas for a few days (2nd, 6th, 9th and 13th of August) the effect of the NMVOC emissions reduction is to decrease simulated surface ozone concentrations by as much as 16 ppb. Similarly, increasing NMVOC emissions by 50% increases O_3 by up to 30 ppb.

Figure 9 shows the comparison between observed isoprene and model-simulated isoprene at Writtle for the base case run and for a 2× and 5× increase in UK emissions of biogenic isoprene. The model-simulated isoprene is, in general, in better agreement with observations for the model simulation with double isoprene emissions. In terms of impact on surface ozone (Fig. 10), the model indicates that UK biogenic isoprene emissions contribute up to ~10 ppb ozone on some days in the base run case. Doubling UK isoprene emissions enhances surface ozone concentrations by up to 10 ppb, and with 5× emissions the effect is ~5 times higher (up to ~45 ppb). An approximately linear dependency of surface ozone to zero, 2× and 5× UK biogenic emissions during this period is found with this experiment (Fig. 10).

The impact of decreasing UK anthropogenic NO_x emissions by 50% on modelled surface ozone at Writtle is shown in Fig. 11. This change has a strong effect on surface ozone at Writtle, though this effect varies substantially for the 15 days in August. As for the sensitivity towards UK NMVOC emissions, a decrease in UK NO_x emissions affects a few days on which high ozone concentrations were simulated (Fig. 3c), with

ACPD
9, 19509–19544, 2009

particularly strong effects on the 2nd and 9th of August, increasing ozone by as much as 65 ppb. The high sensitivity on some days is related to the model's ability to capture plume transport of NO_x accurately, the sensitivity to which is illustrated by the ability to model surface NO_2. The comparison between modelled and observed hourly NO_2 concentrations at Writtle is shown in Fig. 12, and this point is considered further in Sect. 5.

The spatial variation in the response of modelled maximum daily ozone to halving UK NO_x emissions is shown in Fig. 13 for the first 15 days of August 2003. The red areas of Fig. 13 are where O_3 increases when NO_x emissions are halved. Typically, the red areas are regions of high NO_x concentration, such as downwind of London and other major NO_x source areas. The position of the London plume is clear on some days (4th and 5th of August), as depicted by strong ozone increases when NO_x emissions are halved, where predominant SE winds transport it toward the NW. On other days (e.g. 8th, 9th of August), with lower wind speed, the plume is less coherent and NO_x accumulates near the source areas.

The impact of switching off UK ozone dry deposition (both stomatal and non stomatal) is shown in Fig. 11. This model change has a comparatively large impact on surface ozone throughout the simulation period, particularly at night time when surface ozone increase by 50 ppb.

In the final sensitivity experiment, the ozone boundary conditions for the EMEP4UK model were fixed to a climatological value (Logan, 1999) for the whole month. The results of this sensitivity test (Fig. 14) show that in August 2003 surface ozone concentrations are strongly controlled by import, especially in SE England. Ozone changes that range between +10 and −85 ppb on different days in the 15 day period. For example, most of the ozone present on the 6th of August was generated and imported from outside the EMEP4UK inner domain. The spatial variation in the deviation of simulated maximum daily surface ozone from the control run is shown for this boundary condition experiment in Fig. 15 (note the different numerical scale for this figure compared with the analogous Fig. 13). This illustrates how the imported contribution to surface ozone

ACPD
9, 19509–19544, 2009

concentrations in the base run is largest in the south and east of England.

A summary of some of the key results from the ten sensitivity experiments is also included in Table 1.

5 Discussion

A discrepancy of up to 5 K is present in peak temperatures calculated by the WRF model when compared with TORCH observations at Writtle. Similar results are found in other station for example Fig. 1b shows Wattisham (~60 km NE of Writtle). Discrepancies between the model and observations may be caused by several factors. Of course, uncertainties are present in all numerical weather prediction models, and this study represents one of the first attempts to apply WRF to the UK for this period. In addition, the driving analysis and the assimilated data may be insufficient to fully describe the local meteorology at the 5 km grid scale. Also, there may be significant sub-grid variation that the model cannot resolve i.e. for scales less than 5 km in the horizontal or less than 90 m in the vertical. Given the limited spatial resolution of the model, it will not be possible to simulate the expected variation in temperature related to landscape structure. The temperature sensors used may also have accuracy limitations. The fact that there is some disagreement between the two sensors at Writtle (TORCH) indicates that at least one of the latter two factors introduces uncertainty of typically 1–3 K. Nevertheless, the WRF application used in EMEP4UK simulates the heat-wave and diurnal variations of temperature with sufficient accuracy to be of use in simulating photochemistry.

When the UK emissions of NMVOC were modified by ±50%, the model response was to change surface ozone at Writtle by up to ±4 ppb. The exceptions were for the 2nd, 6th, 9th and 13th of August, when an increase in NMVOC increased surface ozone by as much as 30 ppb, while reduced NMVOC emissions decreased it by as much as 16 ppb (Fig. 8). The model overestimates surface NO_2 (Fig. 12) especially for the 2nd and 9th of August providing an artificial abundance of NO_2 which reacts with

ACPD

9, 19509–19544, 2009

NMVOC oxidation products, thereby allowing the production of more ozone. For the 6th of August the absolute difference of maximum surface ozone is ~5 ppb (Fig. 8a), but the timing of the peak is altered by perturbing emissions of NMVOC by ±50%. Surface ozone decrease is limitated to −16 ppb when the NMVOC emissions have been reduced by half and this is caused by the model using all of the available NMVOCs to produce ozone. The implication is that a possible policy aiming to decrease ozone by controlling NMVOC emissions will have a non-linear effect on lowering the production of ozone.

Emissions of biogenic VOC are notoriously uncertain, with isoprene emissions estimates for the UK exhibiting substantial variability. The emissions estimates of Guenther et al. (1995), Simpson et al. (1999), which are used in this work and Stewart et al. (2003), suggested annual European biogenic isoprene emissions of 110, 48 and 8 Gg C y^{-1}, respectively. There are many reasons for the large differences in inventories and their underpinning emission factors, including limitations in the number of measurements, assumptions concerning extrapolation of emission data and characterisation of the effects of environmental and biogeophysical variables (temperature, light, soil moisture, canopy-effects, diversity between and among species). Uncertainties for short time-periods and at specific locations can be expected to be larger than for national averages, and the suggestion of Simpson et al. (1999) that overall biogenic isoprene emissions may be uncertain to within a factor of 3 to 5 may even underestimate the uncertainty of UK emissions during this episode. Moreover, due to the high reactivity of isoprene within this intense photochemical episode, a strong vertical gradient of isoprene is present, as shown in Fig. 16, and the vertical resolution of the model may not be adequate to simulate fully the vertical distribution of isoprene. Despite the above-noted difficulties and complexities, Fig. 9 shows that the EMEP4UK model was able to simulate isoprene at the Writtle site to within a factor of 3 with respect to observations. It should also be noted that the EMEP4UK model currently does not include estimates of any anthropogenic emissions of isoprene.

ACPD
9, 19509–19544, 2009

An interesting feature of surface isoprene was a double peak in the morning and evening, with the latter peak generally higher. OH is understood to be the cause of mid-day dip in isoprene concentrations while the increased afternoon temperatures are thought to be the cause the evening peak in isoprene concentrations. This feature was present in both observations and simulated isoprene concentrations, as seen in Figs. 9 and 16. Overall, the model simulations show that the magnitude of UK emissions of isoprene has only a relatively small impact on surface ozone during this period compared with the other factors investigated. The largest difference in hourly average simulated surface ozone for this experiment is found on the 6th of August (40 ppb), however, the difference for the model daily maximum ozone is 23 ppb. Curci et al. (2009) estimated that BVOC emissions contribute 0–4 ppb towards the maximum daily ozone for the summer (June-July-August) of 2003 in the UK. This is reasonably consistent with our results: we find an EMEP4UK domain average contribution for August of ~1 ppb for the base simulation, and ~3 ppb for the case with 5× isoprene emissions (Fig. 10).

Halving NO_x emissions enhances the surface concentration of ozone by up to 65 ppb on 9th August and up to 32 ppb on 2nd August, whilst on other days it has little impact (Fig. 11). This sensitivity test further supports the conclusion that the deviations between modelled and observed O_3 were particularly related to uncertainties in local patterns of calculated NO_x concentrations, which may be related to local uncertainties in the NO_x emission data. Figure 13 highlights the area affected by the London plume. The location of Writtle is on the edge of the plume and therefore also highly sensitive to small errors in modelled location of the plume. This is illustrated by the fact that on 9th of August the model performed well for the two sites Wicken Fen and London Eltham, which were well outside and inside the London NO_x plume, respectively. When NO_x emissions are reduced, NO_x does not accumulate as fast during the night time and late afternoon as it does during the morning and through midday and therefore ozone production is not limited or depleted as fast by direct reaction of ozone with NO.

The model does not agree well with observed NO_2 at Writtle for the first week of August, but a better agreement is present for the second week (Fig. 12). The discrepancy

ACPD
9, 19509–19544, 2009

between modelled and observed is consistent with the larger standard deviation of the observed averaged ozone concentrations from the high frequency (~minute) observations during the first week compared with the second week of August (data not shown). Large standard deviations (minute data over hourly averages) imply the existence of fast small scale variations of concentration due to local factors which cannot be represented by a 5 km model.

Suppressing dry deposition in the model generally increased the surface ozone concentration (Fig. 11), especially at night-time, when a shallow boundary layer provides a smaller reservoir of ozone, such that a larger depletion by dry deposition takes place. The night between the 10th and 11th of August was the only episode of this month when the measured nocturnal surface concentration of ozone is as high as daytime concentrations (~80 ppb). The dry deposition of ozone is a major factor controlling the magnitude of surface concentrations of ozone. Ozone dry deposition shows a large diurnal cycle. This is because stomatal deposition is a strong function of temperature, humidity, and sunlight. Vautard et al. (2005) found that due to the exceptionally hot weather of August 2003 over Europe, dry deposition calculations in their model needed to be modified to reduce dry deposition of ozone. This study however retains the unmodified dry deposition calculation for the full year simulation of 2003 suggesting that for the UK the parameterisation used in the EMEP4UK model for dry deposition is, in general, adequate for the range of temperature and extreme weather modelled here. Nevertheless, we do find on some specific occasions (e.g. night of 10/11th August) that switching off deposition improves the comparison with observations.

Correct boundary conditions are very important to accurately calculate surface ozone and previous work has demonstrated that European transport and cross-Atlantic transport are well simulated by the EMEP model (e.g. Simpson et al., 2005). Figures 14 and 15 show that the import of ozone from outside the UK was the factor contributing most to the very high ozone in the SE of UK during August 2003. Import is important on different days at different locations (Fig. 15). This is evident for the 5th, 6th, 10th and 11th of August (Fig. 15) when localised incursions of European-emitted precursors and/or

ACPD
9, 19509–19544, 2009

ozone itself were present. This agrees with Solberg et al. (2008), who found that the higher values of ozone observed over SE England were often the result of import from the continent. However, one of the conclusion in Solberg et al. (2008) was that Portuguese forest fire where a possible cause for the unusually high surface ozone over Europe. The EMEP4UK model was able to simulate high surface ozone without emissions from forest fires. Further works need to be done with the EMEP4UK to include forest fire emissions to properly assess their impact on ozone over the UK. It has been shown that, during the TORCH campaign at Writtle, the high level of ozone observed was not created within the model domain of the British Isles, but imported from continental Europe. Figure 15 highlights this clearly on the 6th of August where an incursion of European ozone was present in SE England. However, high resolution modelling is also critical as this type of incursion may influence a small area (<100 km^2), as can be seen in Fig. 15 especially for the 6th, 10th, 11th, and 12th of August.

6 Conclusions

This paper has discussed the performance of the EMEP4UK model during the August 2003 episode. Particular attention has been given to the site at Writtle, where the TORCH campaign made extensive atmospheric measurements, and at two nearby rural and urban background sites. The main conclusions of these analyses are that:

EMEP4UK simulated ozone well through the whole of 2003, including the high O_3 events of August, at two of the sites.

Ozone was fairly well reproduced at the site Writtle for most days, but the peak ozone events were underpredicted, and the sensitivity analysis discussed in this work suggested important reasons for these discrepancies.

Even using a coarse grid (50 km^2) for UK emissions of isoprene, daily and hourly variations of isoprene are captured by the model within a factor 2–3, a satisfactory result given the uncertainty of isoprene emissions, and expected sub-grid variations. The strong vertical gradient modelled for isoprene shows that high vertical resolution

ACPD
9, 19509–19544, 2009

is required.

UK produced isoprene has been found not to be a major driver in the simulations for UK surface ozone during the August 2003 UK heat wave.

Import from outside the British Isles was the cause of the highest levels of ozone in the south of England during the 2003 heat wave.

Different causes of high level of surface ozone have been found for different days.

Surface temperature was exceptionally high in August 2003; however, surface ozone levels were less extraordinary, with similar concentrations occurring in episodes from May to August of 2003.

Meteorology, boundary conditions, and chemistry all play an important part in controlling the magnitude of UK surface ozone.

While import into the UK of ozone and its precursors provides the largest controls on overall ozone concentrations during the episode of August 2003, high resolution (5 km) modelling allows the local distribution of ozone production and consumption to be simulated, allowing improved comparison with site based ozone measurements.

The largest uncertainties identified by this study involved sub-grid NO_x plumes, biogenic VOC (BVOC) emissions and the dry deposition scheme. Further measurements to verify the model predictions concerning BVOC and dry deposition are clearly required.

Acknowledgements. This work is supported jointly by the UK Department for Environment Food and Rural Affairs (Defra), the Centre for Ecology and Hydrology (CEH), the National Environment Research Council (NERC) under its Environment and Health Programme (Grant NE/E008593/1), the EMEP programme under the UNECE LRTAP Convention, and the Norwegian Meteorological Institute (Met. No), and provides a contribution to the work of the NitroEurope Integrated Project (Verification Component) funded by the European Commission. We gratefully acknowledge travel support from the COST 729 action and the European Science Foundation NinE programme.

References

Andersson-Skold, Y. and Simpson, D.: Comparison of the chemical schemes of the EMEP MSC-W and IVL photochemical trajectory models, Atmos. Environ., 33, 1111–1129, 1999.

Berge, E. and Jakobsen, H. A.: A regional scale multi-layer model for the calculation of long term transport and deposition of air pollution in Europe, Tellus, 50, 205–223, 1998.

Curci, G., Beekmann, M., Vautard, R., Smiatek, G., Steinbrecher, R., Theloke, J., and Friedrich, R.: Modelling study of the impact of isoprene and terpene biogenic emissions on European ozone levels, Atmos. Environ., 43, 1444–1455, 2009.

Dore, C. J., Murrells, T. P., Passant, N. R., Hobson, M. M., Thistlethwaite, G., Wagner, A., Li, Y., Bush, T., King, K. R., Norris, J., Coleman, P. J., Walker, C., Stewart, R. A., Tsagatakis, I., Conolly, C., Brophy, N. C. J., and Hann, M. R.: UK Emissions of Air Pollutants 1970 to 2006, AEA Energy and Environment, Harwell, Oxfordshire, 2008.

Emberson, L. D., Ashmore, M. R., Cambridge, H. M., Simpson, D., and Tuovinen, J. P.: Modelling stomatal ozone flux across Europe, Environmental Pollution, 109, 403–413, 2000.

Fagerli, H., Simpson, D., and Tsyro, S.: Transboundary acidification, eutrophication and ground level ozone in Europe, EMEP Status Report 1/2004, Unified EMEP model: Updates, The Norwegian Meteorological Institute, Oslo, Norway, pp. 11–18, 2004.

Gardner, R. M., Adams, K., Cook, T., Deidewig, F., Ernedal, S., Falk, R., Fleuti, E., Herms, E., Johnson, C. E., Lecht, M., Lee, D. S., Leech, M., Lister, D., Masse, B., Metcalfe, M., Newton, P., Schmitt, A., Vandenbergh, C., and VanDrimmelen, R.: The ANCAT/EC global inventory of NO_x emissions from aircraft, Atmos. Environ., 31, 1751–1766, 1997.

Guenther, A. B., Zimmerman P. R., Harley P. C., Monson R. K. and Fall, R.: Isoprene and monoterpene emission rate variability: model evaluations and sensitivity analyses, J. Geophys. Res., 98(D7), 12609–12617, 1993.

Guenther, A., Hewitt, C., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W., Pierce, T., Scholes, R., Steinbrecher, R., Tallamraju, R., Taylor, J., and Zimmerman, P.: A global model of natural volatile organic compound emissions, J. Geophys. Res., 100(D5), 8873–8892, 1995.

Hellsten, S., Dragosits, U., Place, C. J., Vieno, M., Dore, A. J., Misselbrook, T. H., Tang, Y. S., and Sutton, M. A.: Modelling the spatial distribution of ammonia emissions in the UK, Environ. Pollut., 154, 370–379, 2008.

ACPD

9, 19509–19544, 2009

Jenkin, M. E., Davies, T. J., and Stedman, J. R.: The origin and day-of-week dependence of photochemical ozone episodes in the UK, Atmos. Environ., 36, 999–1012, 2002.
Jonson J. E., Tarrason, L., Wind, P., Gauss, M., Valiyaveetil, S. S., Tsyro, S., Klein, H., Isaksen, S. A. I., and Benedictow, A.: First evaluation of the global EMEP model and comparison with the global OsloCTM2 model, EMEP/MSC-W Technical Report 2/07, Norwegian Meteorological Institute, Blindern, Norway, 2007.
Köhler, I., Sausen, R., and Klenner, G.: NO_x production from lightning, The impact of NO_x emissions from aircraft upon the atmosphere at flight altitudes 8–15 km ($AERONO_x$), edited by: Schumann, U., final report to the Commission of the European Communities, Deutsche Luft und Raumfart, Oberpfaffenhofen, Germany, 1995.
Lee, J. D., Lewis, A. C., Monks, P. S., Jacob, M., Hamilton, J. F., Hopkins, J. R., Watson, N. M., Saxton, J. E., Ennis, C., Carpenter, L. J., Carslaw, N., Fleming, Z., Bandy, B. J., Oram, D. E., Penkett, S. A., Slemr, J., Norton, E., Rickard, A. R., Whalley, L. K., Heard, D. E., Bloss, W. J., Gravestock, T., Smith, S. C., Stanton, J., Pilling, M. J., and Jenkin, M. E.: Ozone photochemistry and elevated isoprene during the UK heatwave of August 2003, Atmos. Environ., 40, 7598–7613, 2006.
Logan, J. A.: An analysis of ozonesonde data for the troposhere: Recommendations for testing 3-d models and development of a gridded climatology for troposheric ozone, J. Geophys. Res., 104(D13), 16115–16149, 1999.
Metzger, S., Dentener, F., Pandis, S., and Lelieveld, J.: Gas/aerosol partitioning: 1, A computationally efficient model, J. Geophys. Res.-Atmos., 107(D16), 4312, doi:10.1029/2001JD001102, 2002.
Schar, C. and Jendritzky, G.: Climate change: Hot news from summer 2003, Nature, 432, 559–560, 2004.
Simpson, D., Andersson-Sköld, Y., and Jenkin, M. E.: Updating the chemical scheme for the EMEP MSC-W oxidant model: current status, Norwegian Meteorological Institute, EMEP MSC-W Note 2/93, 1993.
Simpson, D., Winiwarter, W., Borjesson, G., Cinderby, S., Ferreiro, A., Guenther, A., Hewitt, C. N., Janson, R., Khalil, M. A. K., Owen, S., Pierce, T. E., Puxbaum, H., Shearer, M., Skiba, U., Steinbrecher, R., Tarrason, L., and Oquist, M. G.: Inventorying emissions from nature in Europe, J. Geophys. Res.-Atmos., 104(D7), 8113–8152, 1999.
Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J. P.:Transboundary Acidification, Eutrofication and Ground Level Ozone in Europe, Part 1, Unified EMEP Model

Description, EMEP/MSC-W Report 1/03, Norwegian Meteorological Institute, Blindern, Norway, 2003a.
Simpson, D., Tuovinen, J.-P., Emberson, L. D., and Ashmore, M. R.: Characteristics of an ozone deposition module II: Sensitivity analysis, Water, Air Soil Pollut., 143, 123–137, 2003b.
Simpson, D., Jonson, J. E., and Fagerli, H.: Transboundary Acidification, Eutrofication and Ground Level Ozone in 2003, EMEP Status Report 1/2005, Norwegian Meteorological Institute, Blindern, Norway, 2005.
Solberg, S., Hov, O., Sovde, A., Isaksen, I. S. A., Coddeville, P., De Backer, H., Forster, C., Orsolini, Y., and Uhse, K.: European surface ozone in the extreme summer 2003, J. Geophys. Res.-Atmos., 113, D07307, doi:10.1029/2007JD009098, 2008.
Stedman, J. R.: The predicted number of air pollution related deaths in the UK during the August 2003 heatwave, Atmos. Environ., 38, 1087–1090, 2004.
Stewart, H. E., Hewitt, C. N., Bunce, R. G. H., Steinbrecher, R., Smiatek, G., and Schoenemeyer, T.: A highly spatially and temporally resolved inventory for biogenic isoprene and monoterpene emissions: Model description and application to Great Britain, J. Geophys. Res.-Atmos., 108, 4644, doi:10.1029/2002JD002694, 2003.
Tarrason, L., Turner, S., and Floisand, I.: Estimation of Seasonal Dimethyl Sulfide Fluxes over the North-Atlantic Ocean and Their Contribution to European Pollution Levels, J. Geophys. Res.-Atmos., 100, 11623–11639, 1995.
Trigo, R. M., Garcia-Herrera, R., Diaz, J., Trigo, I. F., and Valente, M. A.: How exceptional was the early August 2003 heatwave in France?, Geophys. Res. Lett., 32, L10701, doi:10.1029/2005GL022410, 2005.
Vautard, R., Honore, C., Beekmann, M., and Rouil, L.: Simulation of ozone during the August 2003 heat wave and emission control scenarios, Atmos. Environ., 39, 2957–2967, 2005.
Vieno, M., Dore, A. J., Wind, P., Di Marco, C., Nemitz, E., Phillips, G., Tarrason, L., and Sutton, M. A.: Application of the EMEP Unified Model to the UK with a Horizontal Resolution of $5\times5\,km^2$, Atmospheric Ammonia – Detecting Emission Changes and Environmental Impacts, edited by: Sutton, M. A., Reis, S., and Baker, S. M. H., Springer, Dordrecht, The Netherlands, 367–372, 2009.

Table 1. Summary of the maximum change in model-simulated ozone at the Writtle site during the first 15 days August 2003 induced by each of the various sensitivity model experiments described in this work.

Sensitivity test	+ (ppb)	– (ppb)	Effect on:
$\frac{1}{2}$ UK NO_x emissions	65	5	Chemistry
No dry deposition of O_3	50	2	Dry deposition
+50% UK NMVOCs emissions	30	0	Chemistry
5× UK isoprene emissions	45	0	Chemistry
Fix boundary conditions	10	85	All
+5°C Temperature (2 m)	9	0	Biogenic emission and dry dep.
2× UK Isoprene emissions	10	0	Chemistry
$\frac{1}{2}$ UK NMVOCs emissions	0	16	Chemistry
−5°C Temperature (2 m)	0	9	Biogenic emission and dry dep.
No Isoprene emissions	0	10	Chemistry

ACPD
9, 19509–19544, 2009

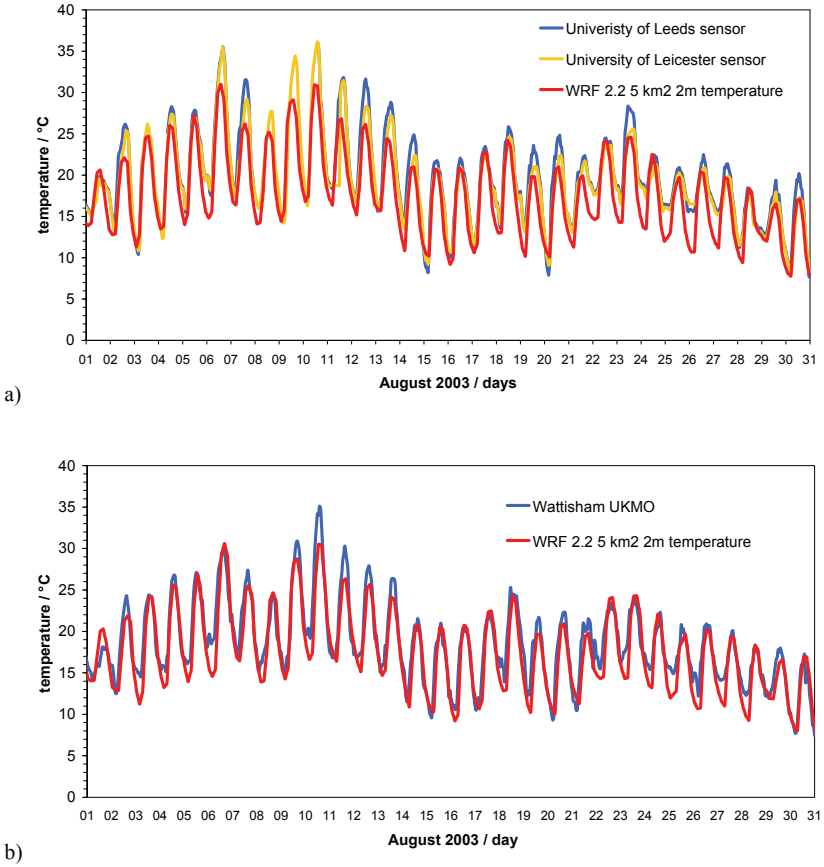


Fig. 1. Hourly surface temperature as measured at **(a)** Writtle TORCH campaign, and **(b)** Wattisham UK Met Office weather station. For August 2003, as compared with the present simulations using the WRF model application for the surrounding $5 \times 5\,km^2$ gridsquare.

ACPD
9, 19509–19544, 2009

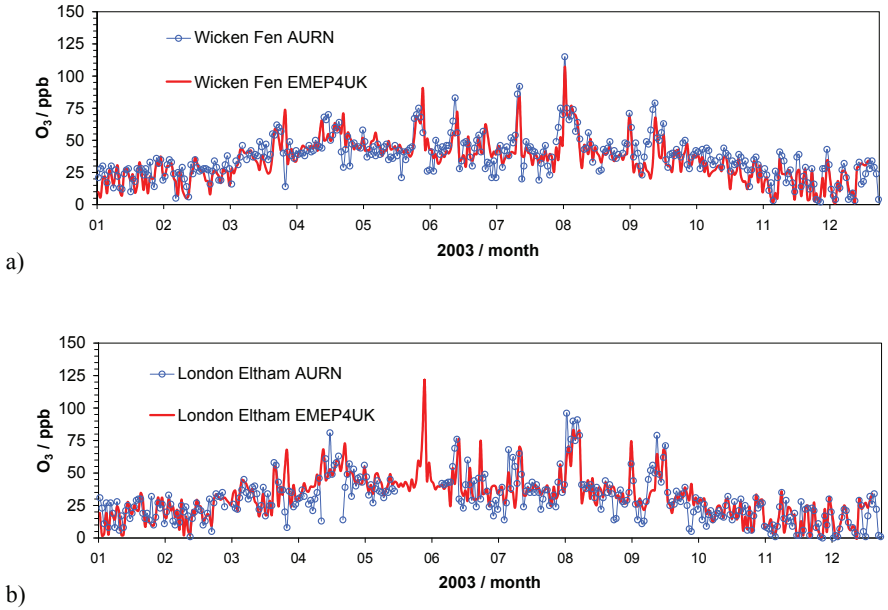


Fig. 2. 14:00–15:00 hourly value for each day of 2003 of modelled (red) and measured (blue) surface ozone at: **(a)** Wicken Fen and **(b)** London Eltham (some missing data in the observations between May and June). Units are ppb.

ACPD
9, 19509–19544, 2009

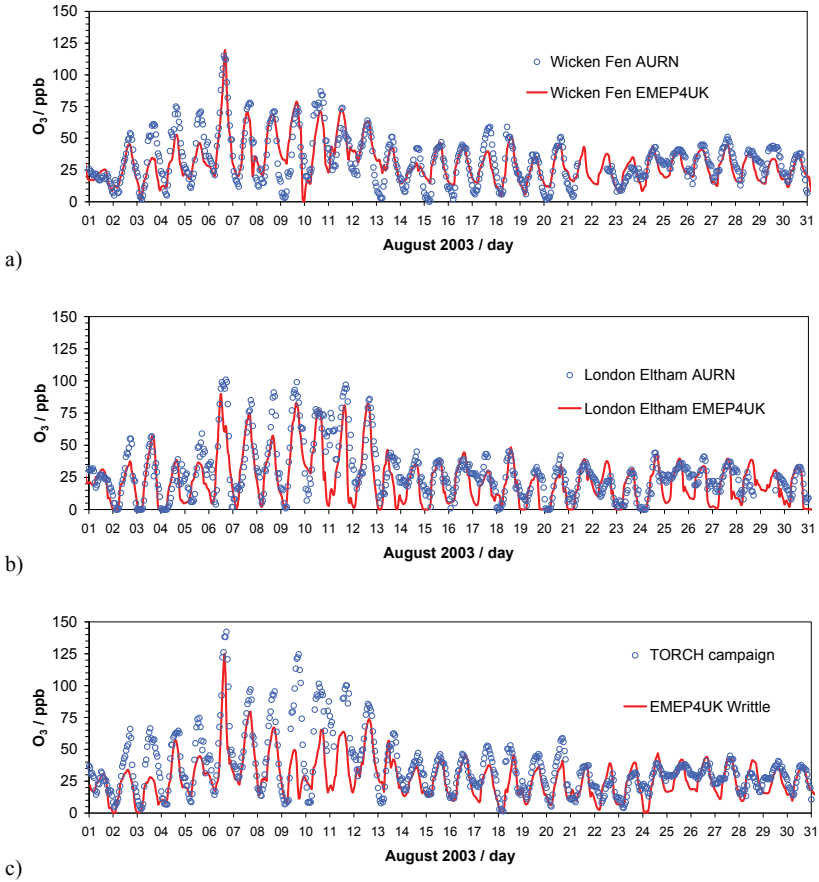


Fig. 3. Hourly time-series of modelled (red) and measured (blue) surface ozone during August 2003 at: **(a)** Wicken Fen, **(b)** London Eltham, and **(c)** TORCH campaign (Writtle). Units are ppb.

ACPD
9, 19509–19544, 2009

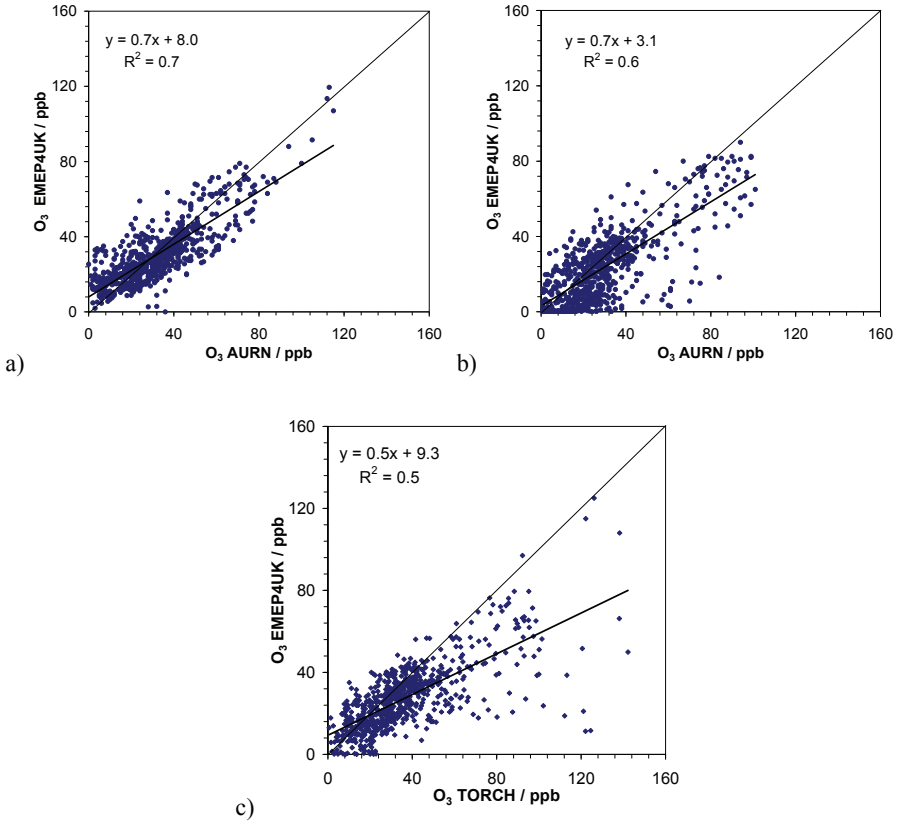


Fig. 4. Scatter plots of modelled vs. observed hourly August 2003 surface ozone concentrations at **(a)** Wicken Fen (AURN), **(b)** London Eltham (AURN), and **(c)** Writtle (TORCH). The 1:1 line extends to the full scale where the best fit line finishes with the maximum modelled or observed value.

ACPD
9, 19509–19544, 2009

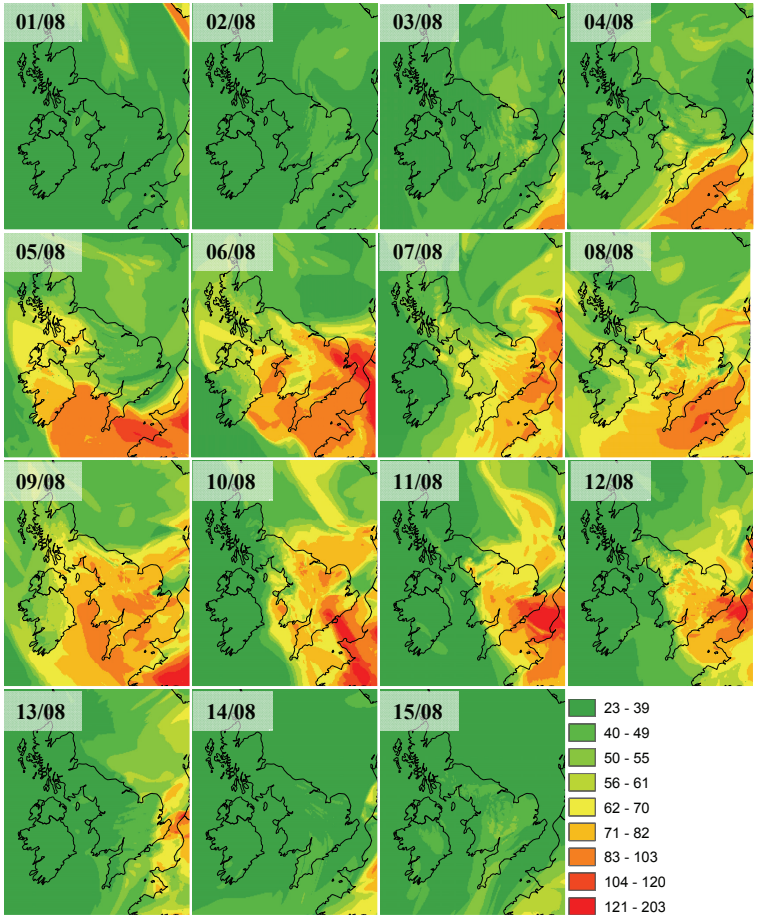


Fig. 5. EMEP4UK surface daily maximum ozone concentration for the first 15 days of August 2003. Units are ppb.

ACPD

9, 19509–19544, 2009

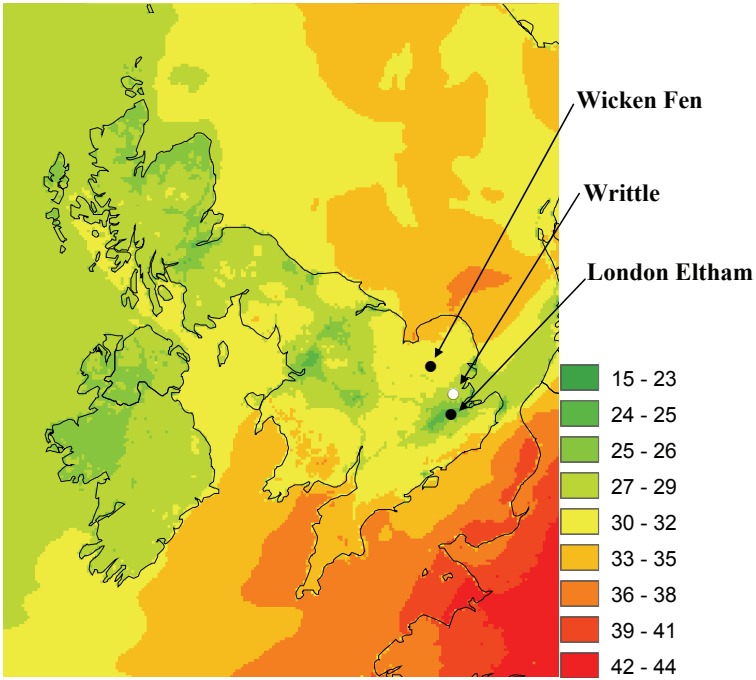


Fig. 6. Monthly mean (August 2003) surface ozone concentration calculated by the EMEP4UK model. The black dots indicate the AURN sites included in this work and the white dot indicates the location of Writtle (TORCH).Units are ppb.

ACPD
9, 19509–19544, 2009

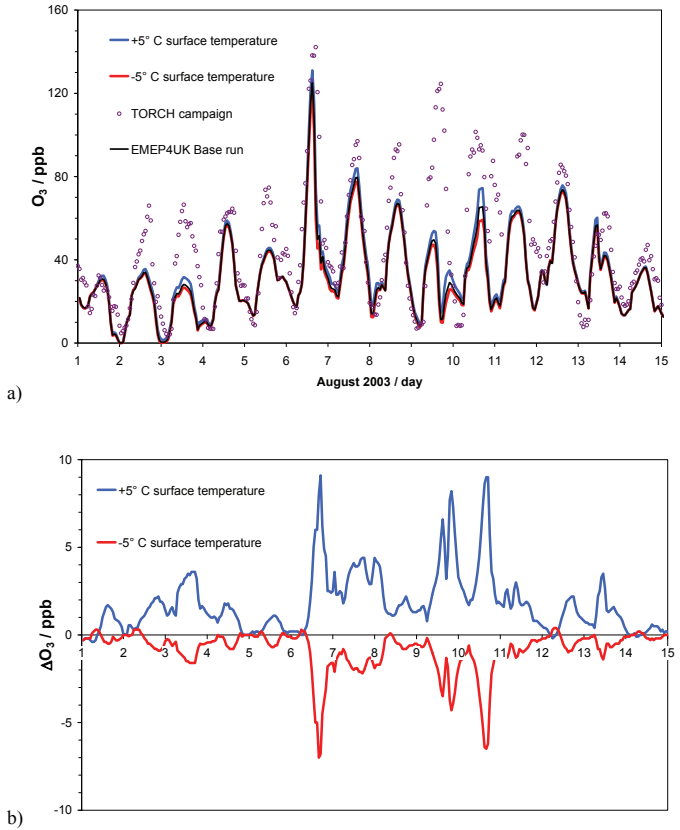


Fig. 7. Hourly time-series of modelled and measured surface ozone for 1–15 August 2003 at the TORCH campaign site at Writtle. EMEP4UK baseline simulation (black line) of hourly ozone together with the effects on simulated ozone of model sensitivity experiments for surface temperature: **(a)** surface concentration of O_3 and **(b)** experiment minus base run.

ACPD
9, 19509–19544, 2009

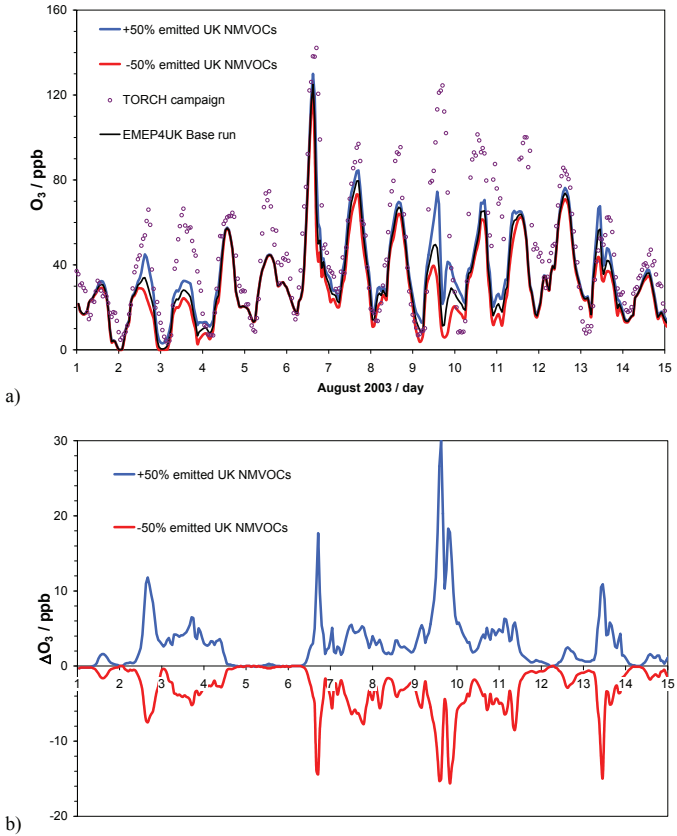


Fig. 8. As for Fig. 7 but for model sensitivity experiments on emissions of UK NMVOC.

ACPD
9, 19509–19544, 2009

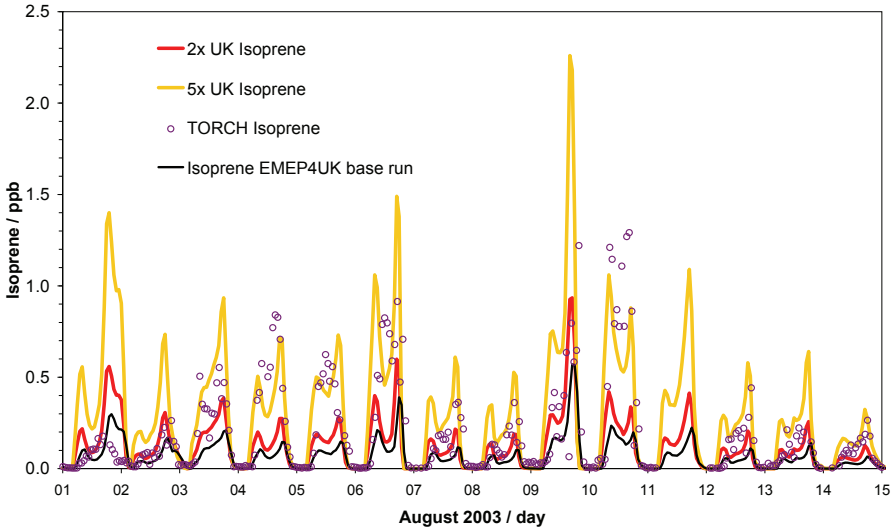


Fig. 9. Time-series of modelled and measured hourly surface isoprene concentrations during August 2003 at Writtle (TORCH).

ACPD

9, 19509–19544, 2009

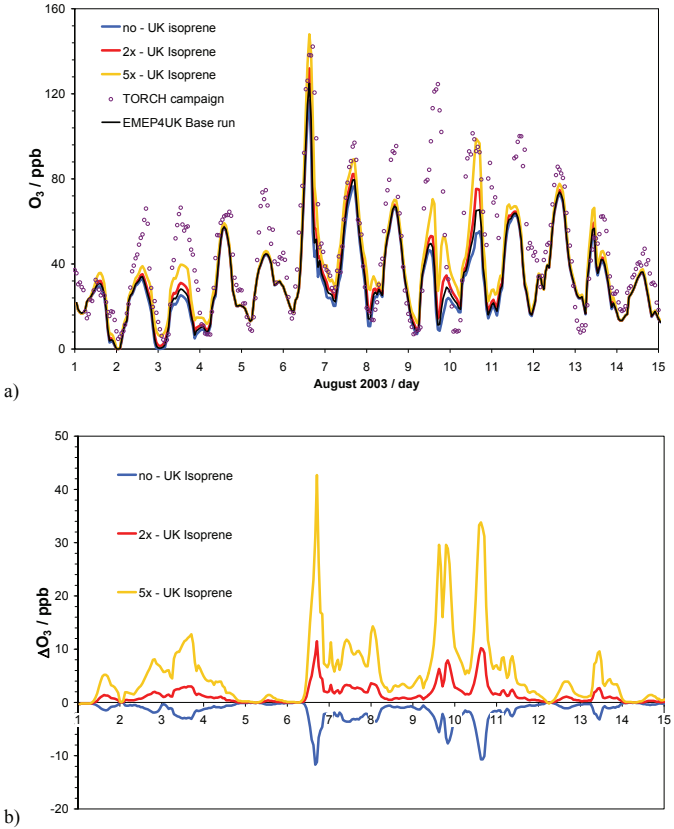
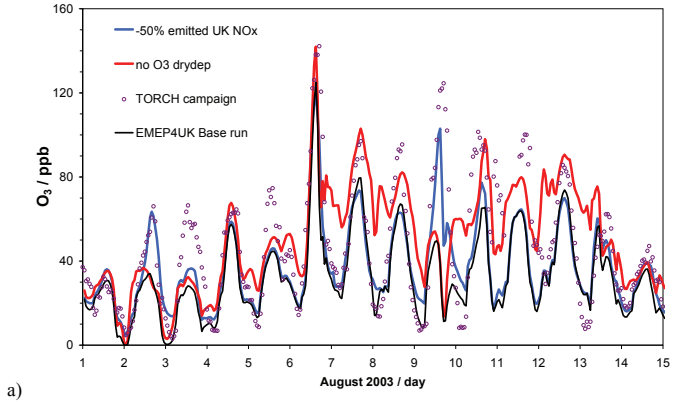
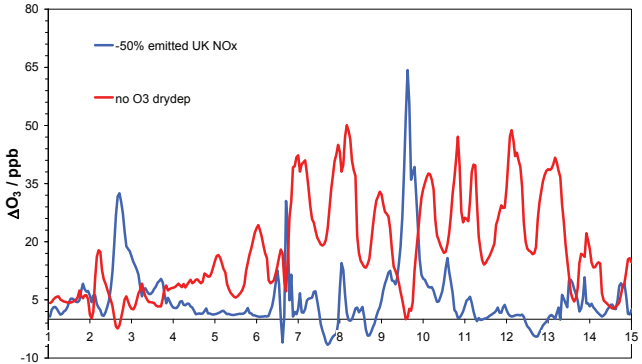


Fig. 10. As for Fig. 7 but for model sensitivity experiments on emissions of biogenic isoprene.

ACPD
9, 19509–19544, 2009



a)



b)

Fig. 11. As for Fig. 7 but for model sensitivity experiments with −50% UK NO_x and with zero O_3 dry deposition.

ACPD

9, 19509–19544, 2009

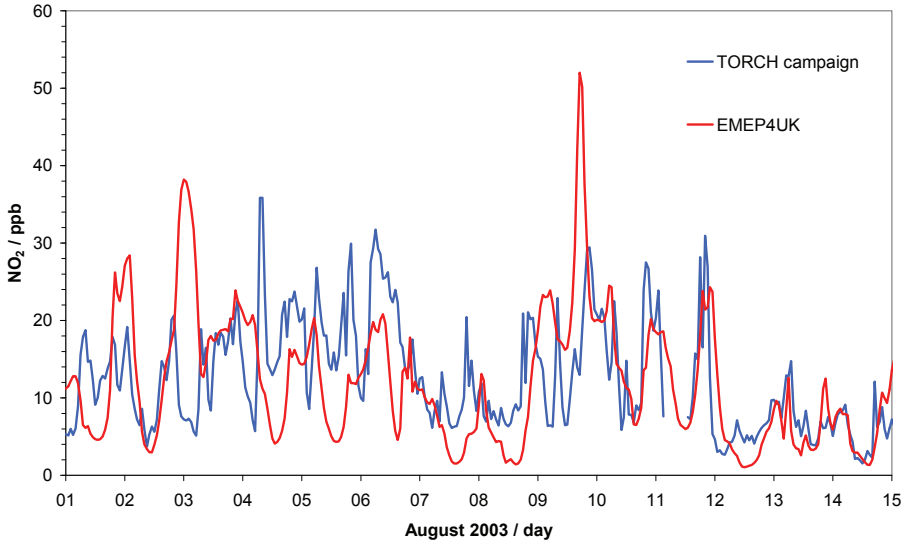


Fig. 12. Hourly time-series of modelled surface NO_2 during August 2003 at Writtle. Units are ppb.

ACPD

9, 19509–19544, 2009

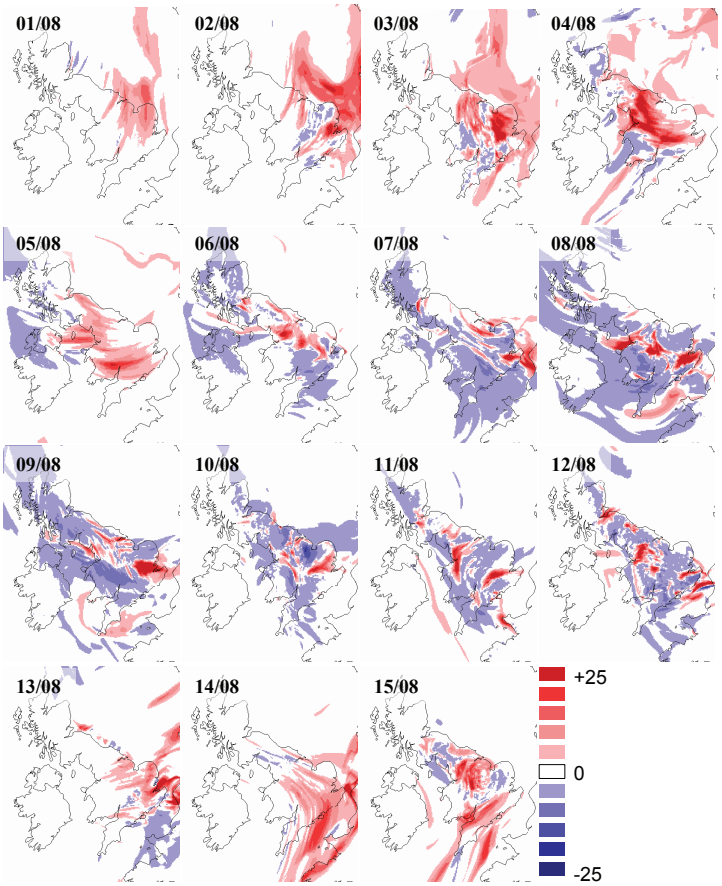


Fig. 13. Change in August 2003 simulated surface daily max ozone concentration relative to the base case scenario for the sensitivity experiment with half UK NO_x emissions: Units are ppb.

ACPD

9, 19509–19544, 2009

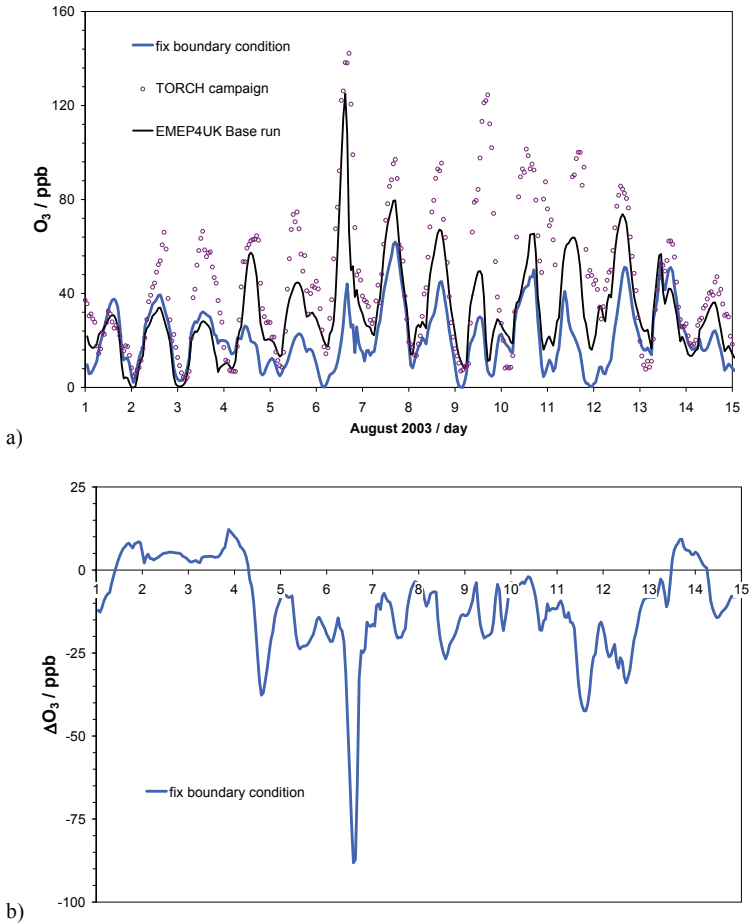


Fig. 14. As for Fig. 7, but for model sensitivity experiments with a fixed O_3 climatologically averaged concentration at the boundary of the EMEP4UK inner domain. Units are ppb.

ACPD
9, 19509–19544, 2009

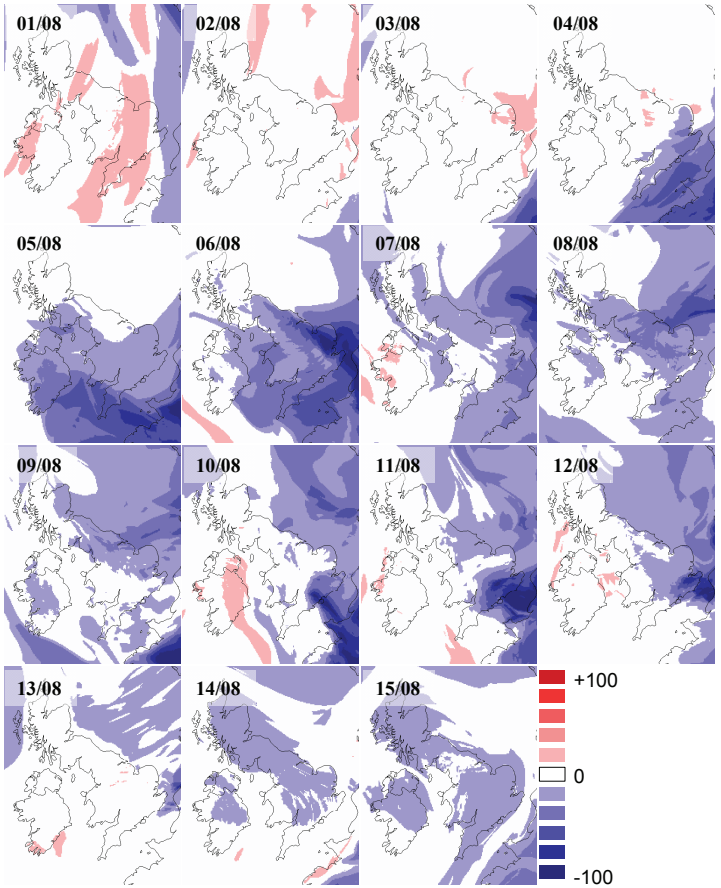


Fig. 15. Change in August 2003 simulated surface daily max ozone concentration relative to the base case scenario for the sensitivity experiment with fixed boundary condition (climatologically) for O_3. Units are ppb.

ACPD
9, 19509–19544, 2009

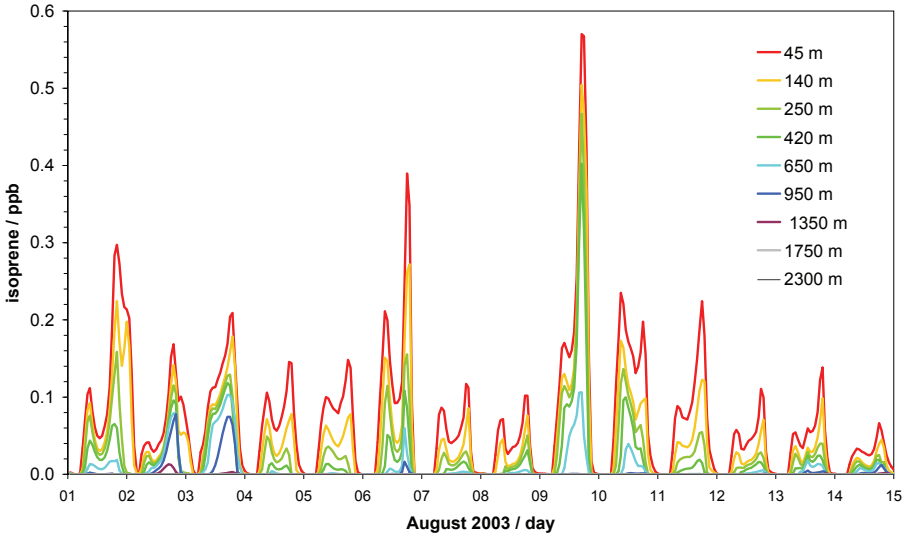


Fig. 16. Time series of modelled hourly isoprene concentrations for the lowest 9 EMEP4UK model levels (mid point approx. altitudes indicated in the legend) at the Writtle site for the first 15 days of August 2003.

ACPD
9, 19509–19544, 2009