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Mechanisms responsible for the build-up of ozone over South East England during the August 2003 heatwave

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ABSTRACT

The Community Multiscale Air Quality (CMAQ) model is used in order to quantify reasons for the build-up of ozone over South East England during the August 2003 heatwave. Unlike previous studies, the effects of individual meteorological and chemical processes on the temporal evolution of the episode are assessed quantitatively in the present work. The performance of the modelling system was briefly evaluated. The modelling system was able to capture the evolution of the episode, with increasing ozone levels during the period 1–4 August 2003, and maximum values afterwards. Analysis of the results of the CMAQ model indicates that three mechanisms were mainly responsible for the episode: (i) horizontal transport from mainland Europe in the presence of a long-lived high-pressure system, (ii) convergence of westerly and easterly near-surface winds, and (iii) downward entrainment of ozone-rich air from residual layers in the free troposphere. The downward entrainment of ozone from residual layers in the morning is found to be key to enhancing ozone levels during the day. The relevance of this mechanism is supported by the good agreement of the model vertical ozone distribution with that derived from Light detection and ranging (Lidar) measurements. The process analysis of the rate of change of ozone concentration shows that both horizontal transport and vertical transport were equally important in explaining the variability of ozone. The contribution of chemical processes to the increase of ozone concentration as simulated by the modelling system is relatively small close to the surface. However, its contribution to the decrease of ozone concentration there becomes as important as that of meteorological processes. By investigating the role of separate meteorological and chemical mechanisms, this study hopes to add to the current understanding of the evolution of air pollution episode.

1. Introduction

The summer 2003 heatwave was one of the hottest periods ever recorded in Europe with record-breaking temperatures across Europe. The heatwave was associated with unusual weather conditions and unprecedented air pollution events in Europe and worldwide. During the period June–August 2003, observed temperatures were about 20–30% above the seasonal average over most parts of Europe. The average near-surface temperature during the heatwave period was about 3 °C higher than that of the equivalent periods in 1961–1990 (Schär et al., 2004). The period broke temperature records dating back to the year 1500. Heatwave periods are becoming more frequent in the present climate. A study, reconstructing monthly and seasonal temperature fields in Europe, indicated that the European climate is becoming warmer, especially from early 21st century (Luterbacher et al., 2004). This trend in warming of the European climate is attributed to the increase in frequency of summer heatwaves.

The unusual hot and dry summer triggered several prolonged air pollution episodes over Europe. Several studies indicated exceptionally intense, long-lasting, and spatially extensive episodes of high ozone concentration over the regions with the highest temperatures, especially during the first two weeks of August 2003 (e.g. Grynszpan, 2003). During this period, the limit value of 120 μg m^{-3} (about 60 ppb) for ozone concentration was repeatedly breached in the UK, especially in South East (SE) England.

Severe socio-economic effects, in relation to the summer 2003 heatwave, were observed in most parts of western Europe. The most affected sector was public health, and especially the elderly population, who were not only exposed to high temperatures, but also long exposures to high concentration of pollutants, notably ozone and particulate matter. The associated total death toll across

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Europe was estimated to be about 35,000 (Vandentorren et al., 2004). The European countries strongly affected were France, Germany, Spain, Italy, the UK, the Netherlands, Portugal, and Belgium, with France reporting the highest number of deaths (De Bono et al., 2003). In the UK, especially in SE England, between 21 and 38% of the excess mortality during the summer 2003 heatwave was estimated to be attributable to exposure to high concentrations of ozone and particulate matter (Johnson et al., 2005). Stedman (2004) investigated the air pollution related deaths in the UK during August 2003 and found a 45 μg m^{-3} increase in population-weighted mean ozone concentration, as compared with the same period in 2002.

The heatwave during the first two weeks of August 2003 resulted primarily from a high-pressure ridge located over western Europe holding back the rain bearing low-pressure systems that usually enter the continent from the Atlantic Ocean. An analysis of ozone simulations (Vautard et al., 2005) suggested that, for most of the period, the associated anticyclonic wind re-circulated the warm air throughout Europe and over the Mediterranean region, leading to a build-up of pollutants together with a rise in temperature.

Most tropospheric ozone is formed and destroyed through reactions involving nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in the presence of sunlight (Sillman, 1999; Jenkin and Clemitshaw, 2000). A reduction of 30%, in peak ozone concentration at the European Monitoring and Evaluation Programme (EMEP) stations in the UK, was identified over the period 1986–1999 (NEGTAP, 2001). This downward trend in episodic peak ozone levels is attributed to the effective reduction in emissions of ozone precursors, notably NO_x and some VOCs, during that period (Derwent et al., 2003). This correlates well with a reduction, in annual emissions of NO_x and VOCs in western and central Europe, in the range 23–32% during the period 1991–2002 reported by Vestreng et al. (2004). However, in spite of the overall reduction in precursors of tropospheric ozone, air pollution events are still often observed during heatwave periods.

A number of factors, which contributed to the prolonged heatwave in Europe and associated degradation of air quality, have been discussed in the literature (Solberg et al., 2008, and references therein). Less attention has been paid to the ozone smog episode in SE England. Lee et al. (2006) suggested that the initial morning increase of ozone concentration was caused primarily by entrainment of air from higher levels, further enhanced by increased emissions of isoprene in the afternoon. The high levels of peroxy radicals observed during the episode indicated a high level of local photochemical activity. A sensitivity study using the EMEP unified model for the UK (EMEP4UK) indicated that meteorology, boundary conditions, and chemistry all played significant roles in contributing to the magnitude of the UK surface ozone concentration during the heatwave period (Vieno et al., 2010).

The accumulation of tropospheric ozone over the UK is often attributed to transport of pollutants from adjacent European countries. Jenkin et al. (2002) analyzed back trajectories to identify the origin and day-of-week dependence of photochemically active ozone episodes in the UK and found that the highest ozone concentrations generally occur under summertime anticyclonic conditions, when air masses from mainland Europe overlapped the UK. Derwent et al. (2004) conducted a model study using a global three-dimensional Lagrangian chemistry-transport model and showed that intercontinental transport can have a significant impact on ozone levels at ground surface sites in Europe. Li et al. (2002) found that transport in the boundary layer and subsidence from the free troposphere enhanced ground surface ozone concentrations over mainland Europe by 2–4 ppb during summertime and 5–10 ppb during transatlantic transport events from North America.

The meteorological and chemical mechanisms contributing to the high-ozone episode over SE England during the August 2003 heatwave have not been quantified so far. The primary focus of the present study is to quantify the contributions of the key meteorological and chemical mechanisms to the build-up of ozone over SE England. Results from this study are expected to provide a greater appreciation of the processes responsible for the build-up of ozone associated with summer heatwaves, which is needed for reliable air quality predictions and to make effective control strategies for episodic conditions.

The Community Multiscale Air Quality (CMAQ) model coupled with the Advanced Research core of the Weather Research and Forecasting (WRF) model is used to characterize the build-up of ozone, during the first two weeks of August 2003. Section 2 gives a brief description of the setup of the modelling system and the observation data used for this study. The synoptic situation associated with the high-ozone episode and reasons for the build-up of

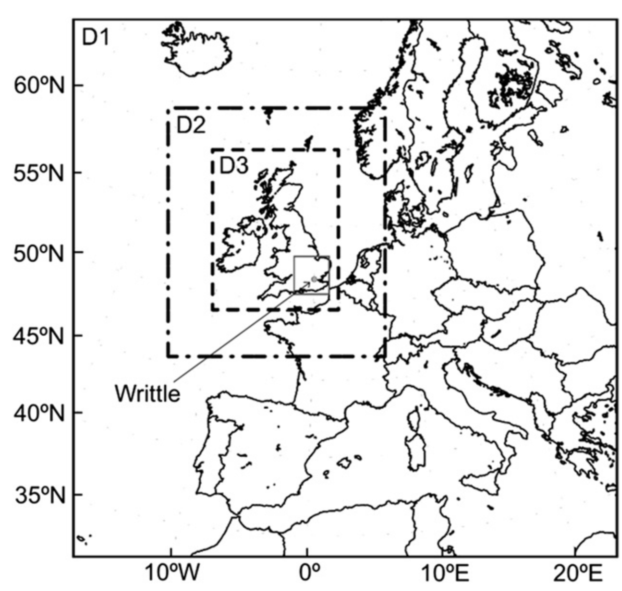

Fig. 1. CMAQ model domains (D1: 45 km, D2: 15 km, and D3: 5 km horizontal resolutions). The rectangular box inside D3 delineates the 'SE England domain'.

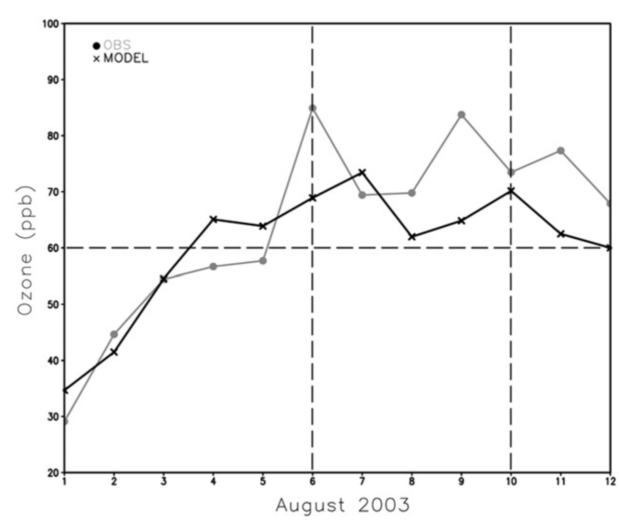

Fig. 2. Time series of observed and predicted maximum daily running 8-h mean ozone concentration (ppb) averaged over 18 ozone monitoring sites in SE England for the period 1–12 August. The horizontal dashed line indicates the limit value of 120 μg m^{-3} (about 60 ppb).