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Scorched earth: how will changes in ozone deposition caused by drought affect human health and ecosystems?

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Abstract

This unique study investigates the effect of ozone (O_3) deposition on ground level O_3 concentrations and subsequent human health and ecosystem risk under hot summer "heat wave" type meteorological events. Under such conditions, extended drought can effectively "turn off" the O_3 vegetation sink leading to a substantial increase in ground level O_3 concentrations. Two models that have been used for human health (the CMAQ chemical transport model) and ecosystem (the DO_3SE O_3 deposition model) risk assessment are combined to provide a powerful policy tool capable of novel integrated assessments of O_3 risk using methods endorsed by the UNECE Convention on Long-Range Transboundary Air Pollution. This study investigates 2006, a particularly hot and dry year during which a heat wave occurred during the summer across much of the UK and Europe. To understand the influence of variable O_3 dry deposition three different simulations were investigated during June and July: (i) actual conditions in 2006; (ii) conditions that assume a perfect vegetation sink for O_3 deposition and (iii) conditions that assume an extended drought period that reduces the vegetation sink to a minimum. The risk of O_3 to human health, assessed by estimating the number of days during which running 8-h mean O_3 concentrations exceeded $100\,\mu g\,m^{-3}$, show that on average across the UK, there is a difference of 16 days exceedance of the threshold between the perfect sink and drought conditions. These average results hide local variation with exceedances reaching as high as 20 days in the East Midlands and Eastern UK. Estimates of acute exposure effects show that O_3 removed from the atmosphere through dry deposition during the June and July period would have been responsible for approximately 460 premature deaths. Conversely, reduced O_3 dry deposition will decrease the amount of O_3 taken up by vegetation and, according to flux-based assessments of vegetation damage, will lead to protection from O_3 across the UK. The study therefore emphasises the importance of accurate estimates of O_3 deposition both for human health and ecosystem risk assessments. Extended periods of drought and heat wave type conditions are likely to occur with more frequency in coming decades,

therefore understanding the importance of these effects will be crucial to inform the development of appropriate national and international policy to mitigate against the worst consequences of this air pollutant.

1 Introduction

Strong connections exist between ozone (O_3) dry deposition and atmospheric O_3 concentrations. Globally the loss of atmospheric O_3 concentrations through dry deposition processes is equivalent to ~20 % of tropospheric O_3 photochemical production (Royal Society, 2008) though the magnitude of the deposition term will vary with season and land cover. This is because dry deposition is determined by the sink strength of underlying vegetation, which is largely controlled by stomata accounting on average for approximately 40–60 % of total ecosystem O_3 uptake (Cieslik, 2004; Fowler et al., 2001; Fowler et al., 2009). During spring and summer periods, when vegetation is physiologically most active, high stomatal conductance (g_{sto}) will result in high dry deposition rates thereby increasing the O_3 loss from the lower atmosphere and decreasing atmospheric O_3 concentrations at ground level (Colbeck and Harrison, 1985). Conversely, during periods of extended hot, dry weather conditions the vegetation can become stressed by high temperatures and soil moisture deficits (SMDs), these conditions will see plants reduce g_{sto} in an effort to limit water loss with subsequent reductions in dry deposition which can lead to maintained high atmospheric O_3 concentrations (Pio et al., 2000). This effect has been investigated in a number of studies concerned with understanding the importance of O_3 deposition on atmospheric O_3 concentrations. Vieno et al. (2010) found that "turning-off" the dry deposition increased O_3 concentrations by ~20 to 35 ppb on most days during the August heat wave period of 2003. The Royal Society (2008) also investigated the effects of altered dry deposition on O_3 concentrations using bespoke European model simulations; they found that "turning-off" surface deposition caused a 31 % increase in episodic peak O_3 concentrations and a 19 % increase in annual mean daily maximum 1-h O_3 concentrations. These results

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were in broad agreement with the regional modelling of (Solberg et al., 2008) which determined the key drivers of peak O_3 concentration during the height of the 2003 European heat wave by assessing the sensitivity of the modelled results to various parameter changes. Reducing the O_3 deposition term to zero produced the largest effect with average maximum hourly O_3 concentrations of eight surface measurement sites across Europe increasing from ~ 75 ppb to over 90 ppb.

The conditions that are likely to reduce dry deposition (i.e. extended periods of hot dry sunny weather) are the same conditions that are likely to result in the build-up of high O_3 concentrations. The association of poor air quality and extremely warm weather is well established (Lee et al., 2006) and is due to a combination of meteorological effects, atmospheric chemical interactions and changes to both the rates and types of terrestrial emissions which occur at elevated temperatures. For example, in the case of UK O_3 concentrations, high summer time concentrations of > 90 ppb are almost always associated with anticyclonic conditions and temperatures in excess of 28–30 °C (Lee et al., 2006) when limited mixing and dilution along with synoptic transport pathways often brings already highly polluted air from mainland Europe to the UK (Jenkin et al., 2002). Increases in biogenic volatile organic carbon (VOC) emissions are also likely to occur with higher temperatures (although the more extreme levels of temperature and drought stress may lead to decreases in biogenic VOC emissions) with such changes being non-linear and species dependant. For example, Lee et al. (2006) found that during the European heat wave of 2003 daytime isoprene concentrations of greater than 1600 ppt were observed in South-East England; such concentrations are more typical of high emitting tropical forested regions and were considered likely to, at least in part, have been due to increases in biogenic emissions.

Such conditions were experienced during the late summer heat wave of 2003 which affected much of Western Europe, especially Switzerland, France and Southern England. In the UK the heat wave lasted for a 2 week period between 4 to 13 August, during which time temperatures peaked at a new record of 38.5 °C. Stedman (2004) investigated the association of this heat wave with excess deaths caused by air pollution

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using established dose-response relationships and found that between ∼225 and 595 excess deaths were associated with elevated O_3 in the UK during the August 2003 episode as compared with the same period in 2002 (with ∼95 % of deaths occurring in England and Wales). These figures represented ∼10 to 30 % of the UK Office for National Statistics reported total excess deaths. The ranges given are based on estimates made assuming either a zero threshold or a threshold of 50 ppb for O_3 effects on human health respectively, based on COMEAP (1998). Across Europe, it was estimated that the 2003 heat wave was responsible for 22 000 premature deaths (Schär and Jendritzky, 2004) leading to losses of an estimated £7bn. Of the deaths occurring in European cities between ∼2.5 to 80 % could be attributed to O_3 based on data analysis from a study of major French cities (Filleul et al., 2006). The occurrence of such heat wave conditions is likely to increase in the future with climate models suggesting the probability of exceeding 35 °C in the UK will increase from 0.6 % under current conditions to 6 % by 2080 (Schar et al., 2004; Stott et al., 2004). The likelihood that such conditions will co-occur with high O_3 concentrations will depend on how UK and European air pollution emission reduction policy develops in coming years.

At the same time as increased atmospheric O_3 concentrations may be causing impacts on human health, the reduction of O_3 deposition to vegetation may be viewed as protecting ecosystems from O_3 damage (Fuhrer, 2009; Matyssek et al., 2007). However, the assessment of vegetation risk will be extremely dependent upon the metric used to estimate effects (Ashmore et al., 2004). In Europe, within the United Nations Economic Commission for Europe (UNECE) Convention on Long-Range Transboundary Air Pollution (LRTAP), use of the flux based metric to assess O_3 impacts on ecosystems is now firmly accepted (LRTAP Convention, 2010). This metric is capable of taking into account the influence of environmental conditions on the sensitivity of O_3 to vegetation and hence is suitable for risk assessment under conditions representative of future, warmer and drier, climates (Harmens et al., 2007).

The likely increases in the occurrence of future heat wave events across Europe will lead to a combination of events (increases in biogenic O_3 precursor emissions;

conditions favouring O_3 formation and conditions likely to reduce O_3 deposition) that are very likely to substantially increase ground level O_3 concentrations. To understand the complex interactions of these factors requires the development and application of models capable of predicting fine resolution O_3 concentration fields coupled with equally well resolved O_3 deposition estimates. In the study presented here this is achieved by combining the Community Multiscale Air Quality (CMAQ) chemical transport model (CTM) with the Deposition of Ozone for Stomatal Exchange (DO_3SE) O_3 dry deposition model. The CMAQ model can be applied using nested grids down to rather fine horizontal resolutions, desirable for human health assessments; in this study we use a 9 km resolution determined by the spatial resolution of the input land cover dataset. The CMAQ model has been found to provide reliable estimates of O_3 concentrations in both rural and urban settings across the UK (Carslaw, 2012; Chemel et al., 2010; Yu et al., 2008; Sokhi et al., 2006).

The DO_3SE model is the only regionally parameterised O_3 dry deposition model that has been developed specifically to estimate damage to vegetation (Emberson et al., 2001; Simpson, et al., 2007). The advantage of this scheme is that the surface resistance component, and particularly the g_{sto} algorithm, have been parameterised, evaluated and used to assess damage for a wide range of species grown across Europe (Emberson et al., 2001; Tuovinen et al., 2004; Simpson et al., 2003; Emberson et al., 2007; LRTAP Convention, 2008). Therefore, as well as providing an estimate of O_3 deposition that is likely to better incorporate the influence of European vegetation on O_3 mass balance, the DO_3SE dry deposition module can also be used to estimate effects on vegetation across the region.

Due to these advantages, the DO_3SE deposition modelling scheme was integrated into CMAQ, thus creating the "CMAQ-DO_3SE" model. The resulting model, is used in this study to assess risk and impacts (i) to human health, based on UK air quality objectives and methods to estimate premature mortality due to acute exposure to O_3 (e.g., COMEAP, 1998) and (ii) to ecosystems following methods proposed by the LRTAP Convention (LRTAP Convention, 2008). The study focuses on the heat wave of

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2006 which was preceded by a hot dry summer causing extensive drought coupled with high O_3 concentrations across the UK and Europe (Doherty et al., 2009). This study provides a first quantification of the importance of the vegetation sink in determining O_3 dry deposition, atmospheric O_3 concentrations, and associated impacts on human health and ecosystems.

2 Methods

2.1 Modelling O_3 photochemistry and dry deposition

The UK O_3 concentration fields used in this study were generated using the USEPA Models-3/CMAQ model, central to this model-3 framework is CMAQ, a third-generation CTM (Byun and Ching, 1999; available online at http://www.cmaq-model.org). The other two components are: a meteorological model (the Weather Research and Forecasting model, WRF), and an emissions processor, typically used by CMAQ users called the Sparse Matrix Operator Kernel Emissions (SMOKE) model. These three components are coupled by an interface called the Meteorology-Chemistry Interface Processor (MCIP) model. CMAQ version 4.6 was used within this study.

CMAQ ordinarily uses one of two different O_3 dry deposition schemes within the MCIP: the surface exchange aerodynamic method (Pleim et al., 2001) or the RADM dry deposition algorithm (Wesely, 1989); both use an electrical resistance approach to estimate dry deposition. Since such an approach is common to most O_3 deposition modelling methods, including the DO_3SE model, it was relatively straightforward to substitute the DO_3SE deposition model, described in (Emberson et al., 2000, 2001; Simpson et al., 2003) into CMAQs MCIP.

A particularly unique feature of the DO_3SE model is the method for estimating surface resistance (R_{sur}) described in Eq. (1)

$$R_{sur} = \frac{1}{\frac{LAI}{r_{sto}} + \frac{SAI}{r_{ext}} + \frac{1}{R_{inc}+R_{gs}}} \tag{1}$$

R_{sur} is calculated as a function of stomatal and non-stomatal canopy resistances, the latter including external plant surface (r_{ext}), within-canopy (R_{inc}) and ground surface/soil resistances (R_{gs}) for which empirical methods and constants are employed based on published literature as described in Simpson et al. (2003). Stomatal (r_{sto}) and external resistances to O_3 deposition are defined at a leaf/needle level (denoted by a lower case r) and scaled according to leaf and surface area indices (LAI and SAI, respectively) to provide canopy scale estimates (denoted by an upper case R).

To estimate g_{sto} (the inverse of r_{sto}) which represents the stomatal control of O_3 uptake to the sites of O_3 damage within the leaves/needles of plants, the DO_3SE model employs a multiplicative algorithm, based on that first developed by Jarvis (1976) and modified for O_3 stomatal flux estimates (Emberson et al., 2000, 2007) according to Eq. (2).

$$g_{sto} = g_{max} \cdot (\min\{f_{phen,fO_3}\} \cdot f_{light} \cdot \max\{f_{min}, f_T \cdot f_D \cdot f_{SW}\}) \tag{2}$$

where the species-specific maximum g_{sto} (g_{max}) is modified within a limit set by a minimum daytime g_{sto} value (f_{min}) and by functions (scaled from 0 to 1) to account for g_{sto} variation with leaf/needle age over the course of the growing season (f_{phen}) and the functions f_{light} , f_T, f_D and f_{SW} relating g_{sto} to irradiance (PPFD), temperature (T), vapour pressure deficit (D), and soil water status (SW). The influence of SW on g_{sto} (f_{SW}) is modelled according to a new method described in Büker et al. (2012) which incorporates the energy balance terms of the Penman-Monteith model (Monteith, 1965) and hence allows an estimate of actual canopy transpiration that is driven by radiant energy as well as atmospheric D. For this study the SWP (Soil Water Potential) model

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was used with root depth varying between 0.1 and 1 m as appropriate for different land cover types. The model has been extensively evaluated for conditions across Europe and is shown to perform well against observed data (Büker et al., 2012). Further details of these methods are given in the Supplement (S1).

The DO_3SE model is parameterised for both broad land cover types (Simpson et al., 2003) and individual forest, crop and grassland species (LRTAP Convention, 2008) such that European parameterisation currently exists for 10 land cover types, 7 forest tree species, 5 crop species and 2 grassland species. Some of the forest species have climate-specific parameterisations that account for the different species ecotypes (LRTAP Convention, 2008). When DO_3SE is used in combination with regional photochemical models the broad land cover parameterisations are used to determine O_3 deposition whilst the species-specific parameterisations are used to assess O_3 risk and vegetation damage. The parameterisations of the cover types and species used in this UK based study are given in Table S1. Six broad land cover types: coniferous, deciduous and mixed forests; croplands; productive grasslands and heathlands were used for total deposition estimates since these were considered to represent dominate land cover types in the UK landscape; the ecosystem risk assessment investigated three species: beech, wheat and productive grasslands.

2.2 Land cover data

Land cover data for the UK were obtained from the UNECE LRTAP Convention harmonized land cover map (Cinderby et al., 2007). These data were compiled specifically for use in assessing the impacts of air pollutants on European ecosystems and agriculture from a mixture of existing digital and paper sources including the European Environment Agency (EEA) Corine Land Cover 2000, the SEI Land European Cover Map (2002 Revision), the FAO Soil Map of the World and the EEA European Biogeographical regions (2005). These land cover data were aggregated to the 9 × 9 km UK grid used by CMAQ.

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2.3 Meteorological and emissions data

The meteorological driver for CMAQ-DO$_3$SE is the WRF model (Skamarock et al., 2008). The lateral conditions for WRF are provided by the National Centres for Environmental Prediction (NCEP) FNL (Final) Global Tropospheric Analyses at 1° grid spacing and 6-h temporal resolution (http://dss.ucar.edu/datasets/ds083.2/). The initial and boundary conditions for CMAQ were derived from the UK Meteorological Office CTM (STOCHEM). The annual anthropogenic emissions (CO, NO_x, NH_3, SO_2, NMVOC and PM_{10}) data were obtained from a number of sources including the European Monitoring and Evaluation Programme (EMEP) at a grid resolution of 50 km, the UK National Atmospheric Emissions Inventory (NAEI) at a grid resolution of 1 km. The emissions from point sources were derived from the European Pollutant Release and Transfer Register (E-PRTR) and the NAEI databases. The emissions from EMEP were used in CMAQ-DO$_3$SE domain 1 (EU 81 km grid) and 2 (EU/UK 27 km) and domain 3 (UK 9 km grid) used the emissions from NAEI.

The annual primary emissions were disaggregated into model chemical species using source specific model species speciation profiles. The profiles for NMVOC were estimated by mapping the UK VOCs emissions (Passant, 2002) with the model chemical species in the USEPA emissions speciation database (http://www.epa.gov/ttn/chief/software/speciate).

These species were then disaggregated into hourly emissions using temporal profiles for 11 CORINAIR/UNECE emission source categories from the City-Delta project (http://aqm.jrc.ec.europa.eu/citydelta/). The biogenic emissions, isoprene and terpene, were estimated using 100 m grid resolution CORINE land cover data, incoming shortwave radiation and surface temperature, using methods described by Guenther et al. (1995) and Sanderson (2002).

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2.4 Estimating O_3 effects on human health

The human health risk from O_3 pollution was assessed according to (i) exceedances of the UKs national air quality objective (http://uk-air.defra.gov.uk/air-pollution/uk-eu-limits). This states that the 8-h mean O_3 concentration should not exceed $100\,\mu g\,m^{-3}$ (DM_{100}) on any more than 10 days per year; and (ii) estimates of the attributable mortalities (or deaths brought forward) due to short-term exposure to O_3. This was estimated using a concentration-response function derived from the WHO (2004) meta-analysis of time series studies of 15 cities in France, Italy, the Netherlands, Spain and the UK. The time series study reported a risk estimate of 0.3 % increase in daily all natural cause mortality (95 % confidence interval (CI) 0.1 to 0.4 %) per $10\,\mu g\,m^{-3}$ daily maximum 8-h O_3 concentration. This figure is without a threshold and can be translated into the relative risk (RR) of 1.003 (95 % CI 1.001 to 1.004). The concentration response coefficient (β) is a slope of the log-linear relationship between RR and concentrations ($RR = \exp^{\beta \Delta X}$) and is estimated as $\ln(RR)/10$.

To calculate the acute premature deaths, the fraction of the disease burden attributable to the risk factor (AF) when the daily maximum 8-h mean was greater than a threshold (a level below which O_3 has no effect on mortality) was calculated as shown in Eq. (3).

$$AF = 1 - \exp^{-\beta \Delta X} \tag{3}$$

where ΔX is the change in daily maximum 8-h O_3 concentration above a threshold.

The attributable deaths for the days that concentrations were above the threshold were then estimated using the following expression (Eq. 4):

$$\Delta Mort = AF \cdot y_o \cdot Pop/365 \tag{4}$$

where, y_o is the baseline mortality rate (i.e., annual natural cause mortalities per million population), Pop is the annual size of the exposed population. Due to the lack of daily population and mortality rate, these annual values were divided by 365. The annual

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attributable deaths were then estimated as an accumulation of ΔMort for the entire year. The attributable deaths during the June to July period were also calculated to quantify the contribution of the peak O_3 heat wave period.

The 2006 baseline natural cause mortality rate, derived from ONS (2008), was 11 581 per million population. Population data at a spatial resolution of 100 m was taken from the European Environment Agency web site (EEA, http://dataservice.eea.europa.eu/) and based upon the Eurostat census 2001. As a consequence we have assumed no significant changes in UK population between 2001 and 2006. These population data were aggregated to the 9 × 9 km CMAQ grid.

It is recognised that the confidence in the existence of associations between O_3 exposures and the health outcomes decreases as the concentrations decrease (IGBP, 2007). The estimates of effects were only made at concentrations greater than 35 ppb daily maximum 8-h mean based on recommendations by WHO (2004) and UNECE (2004). Using this cut-off is recognised to underestimate the O_3 effects; estimates made without a threshold were provided to indicate an upper estimate of the attributable effects of O_3 on mortality.

2.5 Estimating O_3 effects on ecosystems

The flux-based method recommended by the UNECE LRTAP Convention is used in this study to assess O_3 risk to ecosystems during 2006 (LRTAP Convention, 2008). This method requires an estimate of the flux metric POD_y (Phytotoxic Ozone Dose over a threshold y) for the three different representative species: beech, wheat and grasslands. The respective critical levels for the POD_y metric (CL) represent levels below which damage would not be expected to occur and are provided in Table 1 and based on data described in LRTAP Convention (2008).

The calculation of POD_y requires estimates of stomatal O_3 flux (F_{st}) which is perfromed according to the methods provided by LRTAP Convention (2008). F_{st} (nmol O_3 m^{-2} PLA s^{-1}) is calculated according to Eq. (5) which accounts for deposition to the cuticle through incorporation of the leaf surface resistance (r_c) and boundary layer

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resistance (r_b) terms:

$$F_{st} = c(z_1) \cdot g_{sto} \cdot \frac{r_c}{r_b + r_c} \tag{5}$$

where $c(z_1)$ is the concentration of O_3 at the top of the canopy (nmol m^{-3}) at height z_1 (m), g_{sto} is in m s^{-1}, r_b is the leaf quasi-laminar resistance and r_c the leaf surface resistance, both given in s m^{-1}. For further details on the resistance scheme see LRTAP Convention (2008). The accumulated F_{st} above an O_3 stomatal flux threshold of y, given in nmol m^{-2} s^{-1} provides the POD_y index and is calculated according to Eq. (6).

$$POD_y = \sum_{i=1}^{n} \left[F_{st_i} - y\right] \quad \text{for} \quad F_{st_i} \geq y \quad \text{nmol m}^{-2} \text{ PLA s}^{-1} \tag{6}$$

In Eq. (6), F_{st_i} is the hourly mean O_3 flux in nmol O_3 m^{-2} PLA s^{-1}, and n is the number of hours within the accumulation period. The values for the threshold y vary by species as described in Table 1.

The F_{st} accumulation period aims to define that part of the growth period when the species is most sensitive to O_3 and varies for different species. For all species except wheat this is assumed to be equivalent to the period between the start (SGS) and end (EGS) of the growing season, given in the S1. For wheat, the accumulation period is considered to be shorter than the entire growth period since evidence shows that the period around wheat grain-filling is more sensitive to O_3 (Younglove et al., 1994; Soja et al., 2000), again full methods are provided in S1.

2.6 CMAQ-DO$_3$SE model simulations

The CMAQ-DO$_3$SE model was applied to simulate O_3 concentrations and deposition across the UK for the year 2006; this year was chosen as during the last couple of weeks of July much of the UK experienced a heat wave that resulted in extreme weather conditions and high O_3 concentrations that came at the end of an extended period of drought.

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The CMAQ-DO$_3$SE model was run for three alternative model scenarios for the whole of 2006; a "reference" scenario for which O$_3$ deposition was modelled using the DO$_3$SE model method described above; and two scenarios based upon forced alterations to the dry deposition model during the months of June and July 2006. These were achieved by altering Eq. (2); the first of the two scenarios assumed a minimum limitation to g_{sto} calculated by $g_{max} \cdot f_{phen} \cdot f_{light}$ (referred to as the "no stress" scenario; for these calculations the irradiance used to estimate f_{light} was assumed to be that for clear sky to avoid limitations to g_{sto} from overcast conditions), and the second scenario assumed full limitation to g_{sto} calculated by $g_{max} \cdot f_{min}$ (referred to as the "stress" scenario).

3 Results

3.1 Performance of the CMAQ-DO$_3$SE model

3.1.1 O$_3$ concentrations and metrics

The predictive performance of the CMAQ-DO$_3$SE model was assessed by comparing the modelled and measured number of days with a daily 8h-mean O$_3$ concentration $> 100\,\mu g\,m^{-3}$. Measured values were derived from hourly O$_3$ concentrations collected from nine rural sites across the UK which had a capture of more than 90 % during 2006. Figure 1 shows that the CMAQ-DO$_3$SE model is able to describe this human health risk metric reasonably well at most sites with the exception of Aston Hill and Yarner Wood, both sites with particularly high O$_3$ concentrations, where the number of days in exceedance of the threshold is underestimated by a factor of two.

Additional quantification of the model performance was made using statistical measures described in Chang and Hanna (2004) and Builtjes (2005), including factor of 2 of the observations (FAC2), mean bias (MB), normalised mean bias (NMB), root mean square error (RMSE) and correlation coefficient (R)). The results, given in Table 2, show that for all rural sites, greater than 75 % of the modelled O_3 concentrations are within a factor of two of the measurements although there is a tendency for the model to over predict (indicated by the tendency for positive MB values). This tendency varies by site from a small mean over prediction of 2 ppb (0.03) at the Bush Estate site to a more significant overestimate of 14 ppb (0.29) at Ladybower.

3.1.2 Soil moisture status

Central to this study is the CMAQ-DO$_3$SE models ability to predict SMD and subsequent influence on stomatal O_3 flux and deposition. To assess the former the SMD results of CMAQ-DO$_3$SE are compared with equivalent estimates made by the Met Office Rainfall and Evaporation calculation system (MORECS) model described in the National Hydrological Monitoring programmes review for 2006. This review provides estimates of SMDs for the end of July for a grassland cover type over the UK (Marsh et al., 2008). These data showed widespread drought conditions, particularly across almost all of England and the south and east of Scotland (Northern Ireland not shown). Based on an average available water content (AWC) assumed in the MORECS model (Hough and Jones, 1997) these drought areas had less than 25 % of (AWC) remaining. By comparison, the CMAQ-DO$_3$SE model estimates a similar pattern of drought across England by the end of July 2006 (shown in Fig. 2) with soils having less than 30 % of AWC remaining. In Scotland CMAQ-DO$_3$SE estimates the drier areas to the west of the country rather than the east which seems most likely driven by soil texture (see also Fig. 2). However, the area of discrepancy is relatively small and on the whole the CMAQ-DO$_3$SE model simulates the same spatial distribution and relative magnitude of reduced plant water availability. Importantly, the CMAQ-DO$_3$SE model captured the high July SMDs. These were attributed to the very limited rainfall between November 2005 to February 2006 which allowed significant SMD to be carried through the winter in parts of eastern, central and southern England, this was followed by sharp increases in SMD in April 2006, which were unable to recover in spite of a wet May and which were followed by a steep and more sustained rise in SMD during June (Marsh et al., 2008).

3.2 O_3 effects on human health

Table 3 provides estimates for the whole of the UK of exceedance of the DM_{100}. Results show that exceedance can reach nearly 30 days under the "stress" or drought condition, more than double the number of day's exceedance under the "no stress" condition. For the "reference" and "stress" scenarios, 80 % of the exceedance occurring over the entire year can be attributed to the June and July period.

The exceedance of this air quality objective varies across the UK as seen in Fig. 3 which describes the DM_{100} during June and July period across the UK for all three scenarios. This variation is driven by the north/south gradient of O_3 concentrations with higher concentrations in the south resulting from stronger photochemistry, higher UK temperatures and increased long-range transport of O_3 from outside the UK in southern England (Lee et al., 2006). Differences in annual mean 24-h O_3 concentrations between the "no stress" and "stress" scenario were on average 2.5 ppb across the whole of the UK. However, for the June–July period these differences increased to 8 ppb (data not shown). These variations in O_3 concentration translate into geographically variable human health risk. The "reference" results in Fig. 3 show the existence of a north-to-south DM_{100} gradient, with south east England having the highest number of days (between 30 and 35), exceeding 100 µg m^{-3}. These results are mirrored by those for the full annual period (data not shown). By contrast the "no stress" scenario is very different, with the southern part of England tending to have only 12 days on average during which the DM_{100} is exceeded; for this scenario the DM_{100} exceedances for the June–July period are less important in relation to the total number of days of exceedance for the entire year (Table 3) which is likely due to less extreme conditions during this high summer period.

Table 3 shows the estimates of deaths brought forward due to acute O_3 exposure assuming with and without a threshold of 35 ppb for the "reference", "no stress", and

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"stress" scenarios. The upper level of premature annual mortalities for the "reference" case, assuming no threshold, is just above 16 000 in which approximately 20 % of excess deaths are attributed to the June–July period. In contrast, the estimate of premature annual mortalities with a 35 ppb threshold is much smaller at approximately 1800 with almost 50 % associated with the June–July period. The estimates with the threshold show greater differences between the annual and June–July periods due to the fact that outside of this high summer period, concentrations are more likely to drop below the threshold.

The influence of altered O_3 deposition on mortality can be inferred from the results for all three scenarios presented in Table 3 as the number of attributable deaths under the "no stress" compared to the "stress" scenario. The number of UK mortalities in 2006 estimated for the "reference" scenario assuming no threshold increases under the "stress" scenario by ~ 95 premature deaths and is reduced under the "no stress" scenario with ~ 410 premature deaths avoided. Similar results are found using the 35 ppb threshold with the number of premature deaths in 2006 being reduced by ~ 370 under the "no stress" condition and with ~ 90 additional premature deaths estimated under the "stress" scenario.

Figure 4 shows the June–July premature mortalities due to O_3 exposure under the "reference", "no stress", and "stress" scenario using the 35 ppb cut-off. The numbers of deaths brought forward are higher in and around urban areas associated with large human populations in polluted areas. This spatial distribution pattern is similar to that found for the annual assessment and that estimated without a threshold hence these data are not shown here. The main differences between perfect sink and drought conditions occur over populated areas with the "no stress" scenario reducing premature mortalities in and around these urban centres by approximately 40 % during the June–July period.

3.3 O_3 effects on ecosystems

The O_3 effects on ecosystems have been assessed using the flux based POD_y metric. Under the "reference" scenario described in Fig. 5 the modelling suggests that beech is at risk from O_3 as $POD_{1.6}$ values exceed the critical level across most of the country, with more than double the exceedance across approximately half the geographical area. For wheat, the POD_6 index is not exceeded under the "reference" situation (data not shown); a POD_0 value is shown in Fig. 5 to give an indication of the geographical variation in O_3 flux though the index suggests such low fluxes would not be damaging. For grasslands, there is currently no flux based critical level for the LRTAP Convention (2008) parameterisation used in this study although recently established flux based critical levels of 1 mmol O_3 m^{-2} (LRTAP Convention, 2010) would suggest likelihood of a threat from O_3 across all of the UK.

Under the different scenarios the values recorded for each of these vegetation indices changes quite substantially. The magnitude of risk between the "reference" and "scenario" runs associated with the flux metric can be most easily compared by showing relative exceedances for each, i.e. scaling exceedance by the respective critical level for each index as suggested in Simpson et al. (2007). Figure 6 shows these relative exceedances (RCLe) for beech; a RCLe > 1 denotes an exceedance of the POD_y critical levels. The POD_y RCLe shows most risk under the "no stress" condition with values between 5 and 8 times the CL across the UK, with the highest values in the south of England. By comparison, the "reference" scenario shows very low exceedances, predominantly between 0 and 3 extending to 4 times the CL in southern England; under the "stress" scenario the CL is not exceeded (i.e. values are less than 1) for the whole of the UK.

The variation in O_3 flux to vegetation (in part described by POD_y) is related to total O_3 deposition and hence O_3 loss from the atmosphere; higher fluxes will equate to higher O_3 deposition rates and reductions in atmospheric O_3 concentration. Deposition rates vary with land cover; the influence of vegetation type on deposition during the growing season can be seen in Fig. 7 which investigates the influence of the three scenarios on the monthly trends of daily maximum 1-h O_3 deposition for four different sites dominated by particular land cover types common in the UK: Easter Bush (grasslands), NE Cambridge (wheat), Alice Holt (forest) and London North Kensington (urban). The results are presented as monthly mean box and whisker plots showing the maximum and minimum O_3 deposition rate as well as the inter-quartile range.

At all locations for the "no stress" scenario O_3 deposition increases from a minimum in winter ($\sim 0.01\,kgO_3\,ha^{-1}$ for all land cover types) to a maximum during the summer; the influence of land cover is apparent for these maximum deposition rates with the urban locations showing the least O_3 deposition ($\sim 0.025\,kgO_3\,ha^{-1}$) compared with the agricultural land cover type of NE Cambridge which sees deposition of up to $0.06\,kgO_3\,ha^{-1}$. The effect of reduced sink strength is apparent during June and July where there is a notable difference in the O_3 deposition for both the "reference" and "stress" scenarios for the three vegetated land cover types. Forests show a reduced O_3 deposition already by June for the "reference" scenario which may be due to a more rapid drying of the soil for this vegetation type. By July, the period of hot dry sunny weather has continued for long enough that all land cover types have dried out the soil so that the "reference" and "stress" scenarios record the same level of reduced O_3 deposition. As such, the land cover type, and the degree of stress to which the land cover is exposed will both influence the O_3 deposition and hence remaining atmospheric O_3 concentration.

4 Discussion

The results presented in this paper have, for the first time, quantified the likely influence of extreme meteorological events on O_3 dry deposition, subsequent O_3 concentration and ultimately human health and ecosystem effects. This study focussed on a heat wave period that occurred across the UK and much of Europe during June–July of 2006. The study clearly demonstrated the substantial influence that a drought limited vegetation sink had on atmospheric O_3 concentrations, increasing 24-h mean values by approximately 8 ppb as an average over the entire UK over the June–July period. Under such conditions, the human health risk, quantified by the DM_{100} index, was increased by almost two thirds. The increased ground level O_3 concentrations resulted in ~460 additional premature deaths for the June to July period assuming a damage threshold under the "stress" drought scenario, with the largest differences in the estimated premature deaths of the different scenarios reaching 40 % in and around the urban centres. In contrast to the risk to human health, the occurrence of drought reduces stomatal O_3 flux to vegetation as plants close stomata in an attempt to conserve water loss. The resulting reduction in O_3 dose means that under the dry "stress" and "reference" conditions ecosystems were at less risk from O_3 damage with RCLe being halved across much of the UK when compared with the "no stress" conditions.

The focus of this work on the hot dry year of 2006 and the June–July period and heat wave is particularly pertinent since such conditions are likely to increase in future years at many locations across the globe as climate changes (Schär et al., 2004; Stott et al., 2004; Knowlton et al., 2004). The combination of hot dry sunny conditions, often associated with slow moving anti-cyclonic weather patterns (Miao et al., 2006) and stagnant air that traps emissions in the boundary layer (Doherty et al., 2009) combine to create conditions particularly conducive to high levels of O_3 pollution; atmospheric chemical formation of O_3 will be enhanced, continental O_3 transport will be heightened and O_3 loss, due to the likely occurrence of a drought stressed vegetation layer, will be reduced.

There are two main areas of uncertainty in the use of the dose-response approach employed in this study to assess premature mortalities under heat wave conditions: firstly the question of whether a threshold for O_3 effects on human health exists and secondly, what is the likely confounding influence of other co-occurring environmental (and socio-economic) factors on the health impact results.

Due to uncertainties in the shape of dose-response function at very low O_3 concentrations, a threshold for the effects of O_3 on mortality is likely to exist. Although there is

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insufficient evidence for identifying such a threshold, a range of values have been assumed in previous studies such as 50 ppb (COMEAP, 1998 and Stedman et al., 1997) and 35 ppb (WHO, 2004 and UNECE, 2004). The 35 ppb cut-off was recommended by UNECE, (2004) and was used in this study as it is more relevant to the European seasonal variation and geographical distribution of background O_3 concentrations and importantly, the range of concentrations for which most CTMs provide reliable data. It is recognised that the human health effects estimated using this threshold are likely to underestimate the real effects of O_3; as such, estimates without a threshold were also made to indicate the upper estimate of the attributable effects of O_3 on mortality. The choice of threshold makes a substantial difference to absolute estimates of premature deaths, for the full year, assuming a zero threshold gave estimates of ~16 140 premature deaths due to O_3 compared with the 35 ppb threshold value of ~1880. However, the difference in estimates of premature deaths between the "no stress" and "stress" scenarios was similar irrespective of the use of a zero or 35 ppb threshold with values of ~505 and 460 respectively. Therefore, the importance of the O_3 deposition term on the magnitude of the human health risk is largely independent of the threshold value chosen.

In terms of confounding effects it is widely recognised that due to the processes governing O_3 formation, it is likely that high O_3 concentrations will often co-occur with high temperatures. The singular effects of O_3 (e.g. Stedman, 2004) and temperature stress (e.g. Johnson et al., 2005) on human health have been investigated frequently with results clearly showing that both stresses can cause substantial impacts when acting individually. However, there have only been a small number of studies investigating both stresses acting together. Those studies that have been conducted have shown the importance of geographical variation (perhaps due to the frequency of air conditioning use, personal activity and pollution exposure levels, and environmental conditions) as determinants of the O_3-heat mortality effects (Filleul et al., 2006; Ren et al. 2008). Doherty et al. (2009) performed simulations for 3 yr (2003, 2005 and 2006) for 15 UK conurbations and found that overall the number of deaths associated with O_3

appeared to be slightly higher than those attributable to heat and that proportionately more O_3 deaths occurred during periods of very high temperature. The work presented here suggests that similar studies to that by Doherty et al. (2009), that also specifically address issues related to variable O_3 deposition under climate change, should be performed in the future.

In terms of O_3 impacts on vegetation our results suggest a reduced risk under the extreme meteorological events. The findings presented here for the "reference" scenario provide the first national level POD_y assessments that have incorporated the influence of drought (soil water stress) on stomatal O_3 flux; these follow the methods described in Büker et al. (2012) and are consistent with methods recommended by the LRTAP Convention, (2008). Previously, regional modelling of O_3 effects on vegetation has assumed non-limiting soil water and hence could only provide a "worst case" assessment of O_3 risk to vegetation (Simpson et al., 2007). The results of Simpson et al. (2007) showed a range of 4 to 12 mmol O_3 m^{-2} for $POD_{1.6}$ values for forests across the UK with a north-south gradient from low to high values. The "no stress" scenario of this study mimics this geographical pattern though the magnitude of O_3 flux is higher (with values frequently reaching the 25 mmol O_3 m^{-2} across parts of southern England) as would be expected given the higher O_3 levels of this 2006 study year and the near unlimited stomatal conductance to O_3 of the scenario run.

In contrast, the "reference" case has a similar range of $POD_{1.6}$ values (4 to 12 mmol O_3 m^{-2}) across the UK but the spatial gradient is the opposite with highest values occurring across Scotland and lowest values in southern England. This pattern is driven by soil moisture and highlights the importance of incorporating the influence of soil moisture in estimates of both stomatal O_3 flux for ecosystem risk assessment but also for accurate estimates of O_3 deposition. The study also highlights the inappropriateness of using the former LRTAP Convention recommended AOT40 index under conditions of meteorological stress. Using AOT40 in the "reference" case would cause two problems, firstly, the reduced O_3 uptake due to water stress conditions would not be accounted for and secondly, the higher atmospheric O_3 concentrations resulting from reduced O_3 deposition would increase the magnitude of the calculated index. Together this would lead to a serious overestimate of the damaging influence of O_3 on vegetation, for example comparing the AOT40 RCLe of the "no stress" and "stress" scenarios frequently causes increases of 1.5 to 2.5 times the AOT40 critical level with the higher values occurring in the south of the UK (data not shown). This is in contrast with the pattern of risk suggested by the POD_y index.

However, it should be noted that enhanced SMD will not only affect stomatal O_3 fluxes; increased SMD will also adversely impact vegetation growth reducing net primary productivity (NPP) (Ciais et al., 2005) and increase the likely frequency of outbreaks of forest fires, which in turn can influence air quality through biomass burn emissions (Lyamani et al., 2006). There is also evidence to suggest that SMD itself may influence the formation of extreme meteorological events such as heat waves through alterations to the balance between latent and sensible heat fluxes; this is due to the absence of soil moisture reducing latent heat cooling which amplifies surface temperature anomalies (Fischer et al., 2007). As such, it seems inappropriate in this study to make any attempt to quantify changes in yield or biomass loss that might result from altered O_3 flux due to changes in stomatal O_3 deposition; our study can only indicate changes in O_3 risk.

Although this is the first time that the influence of dry deposition has been related to health effects for ground level O_3, this is not the case for other pollutants. A number of studies have investigated the relationship between particulate matter deposition to vegetated surfaces and human health (Tiwary et al., 2009). Recognition of the important role that vegetation (especially trees in urban centres) plays in improving urban air quality has even led to the development of planning policies that encourage the planting of trees within heavily populated urban centres in an attempt to help control exposure of citizens to this toxic pollutant (McDonald et al., 2007). Such policies may be suitable for particulate matter, a pollutant that tends to have more localised emission sources and effects due to its more limited atmospheric transport. However, the nature of O_3 as a secondary pollutant, often forming some considerable distance downwind of precursor

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emission sources, means that dry deposition to vegetation occurring along potentially far more extended trajectories will be important in determining O_3 concentrations and hence human health effects, potentially even at the continental scale.

This study clearly shows that reliable modelling of the O_3 dry deposition term is crucial for accurate assessments of the threat posed to human health by ground level O_3 concentrations. Other studies have only gone as far as estimating the influence of deposition on atmospheric O_3 concentrations but have reached similar results showing the importance of capturing the seasonally variable nature of O_3 deposition which is dependent upon meteorological conditions for accurate estimates of year round O_3 concentrations (Strong et al., 2010). Accurate estimates of dry deposition are also important in determining the O_3 concentration gradient above the ground surface and hence converting modelled O_3 concentrations provided at a height within the planetary boundary layer to the ground surface where impacts on human health and vegetation will occur. This was highlighted by Hayman et al. (2010) who used a dry deposition model, largely derived from the DO_3SE algorithms, to estimate the surface correction for O_3 concentrations across the UK. They found that incorporation of the dry deposition term, coupled with modelled estimates of NO_x emissions, improved estimates of O_3 concentration particularly at urban locations in the UK.

However, to truly understand the implications of changes in O_3 deposition for human health and ecosystem risk assessment the tools employed in the assessment must be "fit for purpose". CMAQ has been used in a number of previous modelling studies across the UK and has shown good agreement with observations in both urban and rural areas (Carslaw, 2012; Chemel et al., 2010; Yu et al., 2008; Sokhi et al., 2006). In this study, good agreement between model results and measurements over rural locations is observed. Although there is a tendency for CMAQ to overestimate O_3 concentrations (as is the case for many O_3 CTMs, Carslaw, 2012), the positive biases at most sites are below 10 % and as such the estimates of the effects of O_3 on human health and ecosystems are expected to be overestimated by a small margin.

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The ability of the DO_3SE model to estimate SMD is an important component of the modelling performed here; comparisons with MORECS modelled estimates of SMD (Marsh et al., 2008) confirmed the DO_3SE models estimate of high SMD across most of England and Wales. The extent and magnitude of the SMD was a result of very limited rainfall during the 2005/06 winter; such situations may increase in frequency in the future since hot summers tend to be preceded by winter rainfall deficits over Southern Europe according to data from the observational record with consequences for less efficient O_3 dry deposition (Vautard et al., 2007). Although the model seems capable of estimating the geographical variation in SMD it is the influence of such SMD on stomatal O_3 flux that is the direct driver of O_3 deposition. A recent evaluation paper by Büker et al. (2012) provides some evidence that this term is being modelled with reasonable accuracy. However, more testing of the module is required to ensure appropriate capture of the influence of SMD on O_3 deposition to different land cover types over the course of the growing season. Further, the importance of accurate land cover and soil texture mapping should not be underestimated given the sensitivity of the deposition term to these factors; management of vegetation (e.g. harvesting of crops) will also need to be understood to accurately determine O_3 loss from the atmosphere.

In addition to SMD there are other recognised uncertainties in the estimates of O_3 deposition; which include aspects of the non-stomatal deposition with evidence that this can vary with environmental conditions such as surface wetness and temperature (Fowler et al., 2009). As such, the use of constant deposition terms to these non-stomatal sinks assumed in the DO_3SE model would benefit from a rigorous review. Additionally, efforts to re-formulate and parameterise the stomatal component of the DO_3SE model may benefit from a focus on the incorporation of more process orientated algorithms that can account for changes in photosynthetic capacity and hence atmospheric CO_2 concentrations, especially under changing climates; such methods have been developed and trialled (Büker et al., 2007) but are yet to be used to estimate critical levels and hence are not yet available for use in O_3 risk assessment.

Finally, it is also recognised that under extreme meteorological conditions mechanisms other than O_3 deposition will affect O_3 concentrations. For example, the NO_x or VOC limitation to O_3 formation was found to change during spring and summer O_3 episodes occurring in 1995 across the UK with implications for NO scavenging and subsequent O_3 assessments (Strong et al., 2010). Vieno et al. (2010) also described the importance of capturing changes in temperature and subsequent biogenic isoprene emission that will occur under heat wave conditions. This is important both due to the importance of isoprene as an O_3 radical source due to its short lifetime (~5 h) as well as the temperature dependence of its emissions, isoprene concentrations have been shown to increase rapidly according to a non-linear relationship with temperature during heat wave O_3 episodes (Lee et al., 2006). If these biogenic emissions are underestimated whilst NO_x emissions remain high then less local O_3 production would be simulated (Vieno et al., 2010). Similar effects would also be caused by underestimating temperature as this would favour peroxyacetyl nitrate (PAN) formation tying up NO_x that would otherwise lead to O_3 formation (Vieno et al., 2010). Further, capturing the wind direction accurately is extremely important for assessment of background O_3 concentrations upon which local O_3 production relies; for example in the UK many O_3 episodes can be attributed to long-range transport arising from precursors originating over continental Europe (Vieno et al., 2010). This also means that the trajectory pathway of these plumes will be affected by O_3 chemistry in the atmosphere and O_3 deposition; an understanding of the influence of these factors along trajectories is fundamental to accurate assessments of O_3 concentrations downwind of major sources.

5 Conclusions

The study has clearly shown the importance of the O_3 absorbing capacity of vegetation in determining human health risk through alterations in ground level O_3 concentrations. For extreme meteorological events characterised by heat wave conditions lasting only a few weeks, the effect of reducing O_3 dry deposition due to drought can lead to at least

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~ 460 excess deaths. At the same time, O_3 damage to vegetation will likely be reduced. As such, not only is it important to improve our understanding of how emissions and meteorology couple to influence O_3 formation, but also how seasonal environmental conditions will affect the physiological activity and hence O_3 sink strength of the underlying vegetation. Understanding how these factors are likely to interact under those conditions most likely to lead to high O_3 episodes in the future under changing climates will provide valuable information to help inform policy decisions on emission reductions that can alleviate the worst effects of O_3 pollution both to human health as well as vegetation and subsequent ecosystem services.

Supplementary material related to this article is available online at: http://www.atmos-chem-phys-discuss.net/12/27847/2012/acpd-12-27847-2012-supplement.pdf.

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References

Ashmore, M., Emberson, L., Karlsson, P. E., and Pleijel, H.: New Directions: A new generation of ozone critical levels for the protection of vegetation in Europe, Atmos. Environ., 38, 2213–2214, 2004.

Builtjes, P.: Review of Methods for Assessing Air Quality at the Regional/Continental Scale, The 6th Framework programme: Air quality assessment for Europe: from local to continental scale, Deliverable D5.1, 2005.

Byun, D. W. and Ching, J. K. S.: Science algorithms of the EPA models-3 Community Multiscale Air Quality (CMAQ) modelling system, available at: http://www.epa.gov/asmdnerl/CMAQ/CMAQscienceDoc.html, 1999.

Büker, P., Emberson, L. D., Ashmore, M. R., Cambridge, H. M., Jacobs, C. M. J., Massman, W. J., Müller, J., Nikolov, N., Novak, K., Oksanen, E., Schaub, M., and de la Torre, D.: Comparison of different stomatal conductance algorithms for ozone flux modelling, Environ. Poll., 146, 726–735, 2007.

Büker, P., Morrissey, T., Briolat, A., Falk, R., Simpson, D., Tuovinen, J.-P., Alonso, R., Barth, S., Baumgarten, M., Grulke, N., Karlsson, P. E., King, J., Lagergren, F., Matyssek, R., Nunn, A., Ogaya, R., Peñuelas, J., Rhea, L., Schaub, M., Uddling, J., Werner, W., and Emberson, L. D.: DO3SE modelling of soil moisture to determine ozone flux to forest trees, Atmos. Chem. Phys., 12, 5537–5562, doi:10.5194/acp-12-5537-2012, 2012.

COMEAP: Quantification of the effects of air pollution on health in Great Britain. Department of Health Committee on the Medical Effects of Air Pollutants, 1998.

Carslaw, D.: Defra Phase 2 regional model evaluation, 2012.

Chang, J. C. and Hanna, S. R.: Air quality model performance evaluation, Meteorol. Atmos. Phys., 87, 167–196, 2004.

Chemel, C., Sokhi, R. S., Yu, Y., Hayman, G. D., Vincent, K. J., Dore, A. J., Tang, Y. S., Parin, H. D., and Fisher, B. E. A.: Evaluation of a CMAQ simulation at high resolution over the UK for the calendar year 2003, Atmos. Environ., 44, 2927–2939, 2010.

Ciais, P., Reichstein, M., Viovy, N., Granier, a, Ogée, J., Allard, V., Aubinet, M., Buchmann, N., Bernhofer, C., Carrara, A., Chevallier, F., De Noblet, N., Friend, A. D., Friedlingstein, P., Grunwald, T., Heinesch, B., Keronen, P., Knohl, A., Krinner, G., Loustau, D., Manca, G., Matteucci, G., Miglietta, F., Ourcival, J. M., Papale, D., Pilegaard, K., Rambal, S., Seufert, G., Soussana, J. F., Sanz, M. J., Schulze, E. D., Vesala, T., and Valentini, R.: Europe-wide reduction in primary productivity caused by the heat and drought in 2003, Nature, 437, 529–533, 2005.

Cieslik, S.: Ozone uptake by various surface types: a comparison between dose and exposure, Atmos. Environ., 38, 2409–2420, 2004.

Cinderby, S., Emberson, L., Owen, A., Ashmore, M.: CCE Progress Report: LRTAP land cover map of Europe, available at: http://www.rivm.nl/bibliotheek/digitaaldepot/PBL_CCE_PR07_PartI_5.pdf, 2007.

Colbeck, I. and Harrison, R. M.: The photochemical pollution episode of 5–16 July 1983 in North-West England, Atmos. Environ., 19, 1921–1929, 1985.

Doherty, R. M., Heal, M. R., Wilkinson, P., Pattenden, S., Vieno, M., Armstrong, B., Atkinson, R., Chalabi, Z., Kovats, S., Milojevic, A., and Stevenson, D. S.: Current and future

climate- and air pollution-mediated impacts on human health, Environ. Health, 8(Suppl I), S8, doi:10.1186/1476-069X-8-S1-S8, 2009.
Emberson, L. D., Büker, P., and Ashmore, M. R.: Assessing the risk caused by ground level ozone to European forest trees: a case study in pine, beech and oak across different climate regions, Environ. Poll., 147, 454–466, 2007.
Emberson, L. D., Ashmore, M. R., Simpson, D., Tuovinen, J.-P., and Cambridge, H. M.: Modelling and mapping ozone deposition in Europe, Water Air Soil Pollut., 130, 577–582, 2001.
Emberson, L. D., Simpson, D., Tuovinen, J.-P., Ashmore, M. R., and Cambridge, H. M.: Towards a model of ozone deposition and stomatal uptake over Europe, available at: http://emep.int/publ/reports/2000/dnmi_note_6_2000.pdf, 2000.
Filleul, L., Cassadou, S., Médina, S., Fabres, P., Lefranc, A., Eilstein, D., Le Tertre, A., Pascal, L., Chardon, B., Blanchard, M., Declercq, C., Jusot, J.-F., Prouvost, H., and Ledrans, M.: The Relation Between Temperature, Ozone, and Mortality in Nine French Cities During the Heat Wave of 2003, Environ. Health Persp., 114, 1344–1347, 2006.
Fischer, E. M., Seneviratne, S. I., Vidale, P. L., Lüthi, D., and Schär, C.: Soil Moisture–Atmosphere Interactions during the 2003 European Summer Heat Wave, J. Climate, 20, 5081–5099, 2007.
Fowler, D., Pilegaard, K., Sutton, M., Ambus, P., Raivonen, M., Duyzer, J., Simpson, D., Fagerli, H., Fuzzi, S., Schjoerring, J. K., Granier, C., Neftel, A., Isaksen, I. S. A., Laj, P., Maione, M., Monks, P. S., Burkhardt, J., Daemmgen, U., Neirynck, J., Personne, E., Wichink-Kruit, R., Butterbach-Bahl, K., Flechard, C., Tuovinen, J. P., Coyle, M., Gerosa, G., Loubet, B., Altimir, N., Gruenhage, L., Ammann, C., Cieslik, S., Paoletti, E., Mikkelsen, T. N., Ro-Poulsen, H., Cellier, P., Cape, J. N., Horváth, L., Loreto, F., Niinemets, U., Palmer, P. I., Rinne, J., Misztal, P., Nemitz, E., Nilsson, D., Pryor, S., Gallagher, M. W., Vesala, T., Skiba, U., Bruggemann, N., Zechmeister-Boltenstern, S., Williams, J., O'Dowd, C., Facchini, M. C., de Leeuw, G., Flossman, A., Chaumerliac, N., and Erisman, J. W.: Atmospheric composition change: Ecosystems-Atmosphere interactions, Atmos. Environ., 43, 5193–5267, 2009.
Fowler, D., Flechard, C., Cape, J. N., Storeton-West, R., and Coyle, M.: Measurements of ozone deposition to vegetation quantifying the flux, the stomatal and non-stomatal components, Water Air Soil Pollut., 130, 63–74, 2001.
Fuhrer, J.: Ozone risk for crops and pastures in present and future climates, Die Naturwissenschaften, 96, 173–194, 2009.

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Scorched earth

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

Harmens, H., Mills, G., Emberson, L. D. and Ashmore, M. R.: Implications of climate change for the stomatal flux of ozone: a case study for winter wheat, Environ. Poll., 146, 763–770, 2007.

Hayman, G. D., Abbott, J., Davies, T. J., Thomson, C. L., Jenkin, M. E., Thetford, R., and Fitzgerald, P.: The ozone source–receptor model – A tool for UK ozone policy, Atmos. Environ., 44, 4283–4297, 2010.

Hough, M. N. and Jones, R. J. A.: The United Kingdom Meteorological Office rainfall and evaporation calculation system: MORECS version 2.0-an overview, Hydrol. Earth Syst. Sci., 1, 227–239, doi:10.5194/hess-1-227-1997, 1997.

IGBP: An Economic Analysis to inform the Air Quality Strategy, 2007.

Jarvis, P. G.: The interpretation of the variations in leaf water potential and stomatal conductance found in canopies in the field., Philos. T. Roy. Soc. Lond., B, 273, 593–610, 1976.

Jenkin, M. E., Davies, T. J., and Stedman, J. R.: The origin and day-of-week dependance of photochemical ozone episodes in the U.K., Atmos. Environ., 36, 999–1012, 2002.

Johnson, H., Kovats, R. S., McGregor, G., Stedman, J., Gibbs, M., Walton, H., Cook, L., and Black, E.: The impact of the 2003 heatwave on mortality and hospital admissions in England, Health Stat Quart., 25, 6–11, 2005.

Knowlton, K., Rosenthal, J. E., Hogrefe, C., Lynn, B., Gaffin, S., Goldberg, R., Rosenzweig, C., Civerolo, K., Ku, J.-Y., and Kinney, P. L.: Assessing Ozone-Related Health Impacts under a Changing Climate, Environ. Health Perspectives, 112, 1557–1563, 2004.

LRTAP Convention: Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends. Chapter 3: Mapping Critial Levels for Vegetation, available at: http://icpvegetation.ceh.ac.uk/manuals/mapping_manual.html, 2008.

LRTAP Convention: Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends. Chapter 3: Mapping Critial Levels for Vegetation, http://icpvegetation.ceh.ac.uk/manuals/, 2010.

Lee, J., Lewis, A., Monks, P., Jacob, M., Hamilton, J., Hopkins, J., Watson, N., Saxton, J., Ennis, C., and Carpenter, L.: Ozone photochemistry and elevated isoprene during the UK heatwave of august 2003, Atmos. Environ., 40, 7598–7613, 2006.

Lyamani, H., Olmo, F. J., Alcántara, A., and Alados-Arboledas, L.: Atmospheric aerosols during the 2003 heat wave in southeastern Spain I: Spectral optical depth, Atmos. Environ., 40, 6453–6464, 2006.

Marsh, T., Hannaford, J., and Sanderson, F.: 2006 UK hydrological review, http://www.ceh.ac.uk/data/nrfa/nhmp/annual_review.html, 2008.

Matyssek, R., Bytnerowicz, A., Karlsson, P.-E., Paoletti, E., Sanz, M., Schaub, M., and Wieser, G.: Promoting the O3 flux concept for European forest trees, Environ. Poll., 146, 587–607, 2007.

McDonald, A. G., Bealey, W. J., Fowler, D., Dragosits, U., Skiba, U., Smith, R. I., Donovan, R. G., Brett, H. E., Hewitt, C. N., and Nemitz, E.: Quantifying the effect of urban tree planting on concentrations and depositions of PM10 in two UK conurbations, Atmos. Environ., 41, 8455–8467, 2007.

Miao, J.-F., Chen, D., and Wyser, K.: Modelling subgrid scale dry deposition velocity of O3 over the Swedish west coast with MM5-PX model, Atmos. Environ., 40, 415–429, 2006.

Monteith, J. L.: Evaporation and environment, Symp. Soc. Exp. Biol., 19, 205–234, 1965.

ONS: Mortality statistics: Death registered in 2006, ISBN 978-1-85774-672-3, 2008.

Passant, N. R.: Speciation of UK emissions of non-methane volatile organic compounds, AEAT/ENV/R/0545 Issue 1, http://uk-air.defra.gov.uk/reports/empire/AEAT_ENV_0545_final_v2.pdf, 2002.

Pio, C. A., Feliciano, M. S., Vermeulen, A. T., and Sousa, E. C.: Seasonal variability of ozone dry deposition under southern European climate conditions, Atmos. Environ., 34, 195–205, 2000.

Pleim, J. E., Xiu, A., Finkelstein, P. L., and Otte, T. L.: A coupled land-surface and dry deposition model and comparison to field measurements of surface heat, moisture, and ozone fluxes, Water Air Soil Pollut., 1, 243–252, 2001.

Ren, C., Williams, G. M., Mengersen, K., Morawska, L., and Tong, S.: Does temperature modify short-term effects of ozone on total mortality in 60 large eastern US communities? An assessment using the NMMAPS data, Environ. International, 34, 451–458, 2008.

Royal Society: Ground-level ozone in the 21st century: future trends, impacts and policy implications, London, 2008.

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Sanderson, M. G.: Emission of isoprene, monoterpenes, ethene and propene by vegetation, Hadley, 2002.
Schar, C., Vidale, P. L., Luthi, D., Frei, C., Haberli, C., Liniger, M. A., and Appenseller, C.: The role of increasing temperature variability in European summer heatwaves, Nature, 427, 3926–3928, 2004.
Schär, C. and Jendritzky, G.: Hot news from summer 2003, Nature, 432, 2003–2004, 2004.
Simpson, D., Ashmore, M. R., Emberson, L., and Tuovinen, J.-P.: A comparison of two different approaches for mapping potential ozone damage to vegetation. A model study, Environ. Poll., 146, 715–725, 2007.
Simpson, D., Fagerli, H., Jonson, J. E., Tsyro, S., Wind, P., and Tuovinen, J.-P.: Transboundary acidification, eutrophication and ground level ozone in Europe, Part I. Unified EMEP model description, EMEP Status Report 1/2003, Norwegian Meteorological Institute, Oslo, 74 p. http://www.emep.int, 2003.
Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X.-Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF Version 3, http://www.mmm.ucar.edu/wrf/users/docs/arw_v3_bw.pdf, 2008.
Soja, G., Barnes, J. D., Posch, M., Vandermeiren, K., Pleijel, H., and Mills, G.: Phenological weighting of ozone exposures in the calculation of critical levels for wheat, bean and plantain, Environ. Pollut., 109, 517–524, 2000.
Sokhi, R. S., San José, R., Kitwiroon, N., Fragkou, E., Pérez, J. L. and Middleton, D. R.: Prediction of ozone levels in London using the MM5–CMAQ modelling system, Environ. Modell. Softw., 21, 566–576, 2006.
Solberg, S., Hov, O., Sovde, A., Isaksen, I. S. A., Coddeville, P., De Backer, H., Forster, C., Oorsolini, Y., and Uhse, K.: European surface ozone in the extreme summer 2003, J. Geophys. Res., 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.
Stedman, J. R., Anderson, H. R., Atkinson, R. W., and Maynard, R. L.: Emergency hospital admissions for respiratory disorders attributable to summer time ozone episodes in Great Britain, Thorax, 52, 958–963, 1997.
Stott, P. A., Stone, D. A., and Allen, M. R.: Human contribution to the European heatwave of 2003, Nature, 432, 610–614, 2004.

Strong, J., Whyatt, J. D., Hewitt, C. N., and Derwent, R. G.: Development and application of a Lagrangian model to determine the origins of ozone episodes in the UK, Atmos. Environ., 44, 631–641, 2010.

Tiwary, A., Sinnett, D., Peachey, C., Chalabi, Z., Vardoulakis, S., Fletcher, T., Leonardi, G., Grundy, C., Azapagic, A., and Hutchings, T. R.: An integrated tool to assess the role of new planting in PM10 capture and the human health benefits: a case study in London, Environ. Pollut., 157, 2645–2653, 2009.

Tuovinen, J.-P., Ashmore, M. ., Emberson, L., and Simpson, D.: Testing and improving the EMEP ozone deposition module, Atmos. Environ., 38, 2373–2385, 2004.

UNECE: Modelling and assessment of the health impact of particulate matter and ozone, p. EB.AIR/WG.1/2004/11, 2004.

Vautard, R., Beekmann, M., Desplat, J., Hodzic, A., and Morel, S.: Air quality in Europe during the summer of 2003 as a prototype of air quality in a warmer climate, Comptes Rendus Geoscience, 339, 747–763, 2007.

Vieno, M., Dore, A. J., Stevenson, D. S., Doherty, R., Heal, M. R., Reis, S., Hallsworth, S., Tarrason, L., Wind, P., Fowler, D., Simpson, D., and Sutton, M. A.: Modelling surface ozone during the 2003 heat-wave in the UK, Atmos. Chem. Phys., 10, 7963–7978, doi:10.5194/acp-10-7963-2010, 2010.

WHO: Meta-analysis of time-series studies of Particulate Matter (PM) and Ozone (O_3), Copenhagen., 2004.

Wesely, M.: Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models, Atmos. Environ., 41, 52–63, 1989.

Younglove, T., McCool, P. M., Musselman, R. C., and Kahl, M. E.: Growth-stage dependent crop yield response to ozone exposure, Environ. Pollut., 86, 287–295, 1994.

Yu, Y., Sokhi, R. S., Kitwiroon, N., Middleton, D. R., and Fisher, B.: Performance characteristics of MM5-SMOKE-CMAQ for a summer photochemical episode in southeast England, United Kingdom, Atmos. Environ., 42, 4870–4883, 2008.

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Table 1. POD_y based critical levels (CL) for O_3 risk on wheat, beech and grasslands (LRTAP Convention, 2008).

Vegetation	Canopy height (m)	POD_y value	Time period for index accumulation	Critical level (CL) (mmol O_3 m^{-2})	Effect	Note
Wheat	1	POD_6	55 days	1	Grain yield (5 %)	Based on wheat
Beech	20	$POD_{1.6}$	Latitude defined growth period	4	Biomass reduction (5 %)	Based on birch and beech
Grassland	1	$POD_{1.6}$	Year round	–	–	–

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Table 2. Statistical measures of model performance in predicting hourly O_3 concentrations at nine UK rural sites where data capture was more than 90 % over the course of the year 2006.

Date	Number of data points	% data capture	FAC2	MB ($\mu g\,m^{-3}$)	NMB	RMSE (%)	R
Aston Hill	8093	92.39	0.94	−6.39	−0.09	22.32	0.5
Bottesford	8659	98.85	0.82	2.72	0.05	21.9	0.65
Bush Estate	8580	97.95	0.89	1.78	0.03	20.66	0.39
Eskdalmuir	8659	98.85	0.83	11.87	0.2	24.98	0.42
Harwell	8196	93.56	0.83	3.72	0.07	21.94	0.62
Ladybower	8313	94.90	0.75	14.24	0.29	27.58	0.42
Rochester	8633	98.55	0.75	2.99	0.06	23.42	0.62
Sibton	8060	92.01	0.86	5.5	0.1	21.57	0.66
Yarner Wood	8446	96.42	0.88	4.07	0.06	24.14	0.47

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Table 3. Number of days when the 8-h mean O_3 concentration (DM_{100} index) was exceeded in the UK and the UK attributable mortalities due to short-term O_3 exposure for both the entire annual period and the June to July period in 2006 using two different thresholds (0 and 35 ppb) for health impacts.

Scenario	Reference	No stress	Stress
No. of days when 8-h mean O_3 concentration > 100 $\mu g\,m^{-3}$			
Annual	25	13	29
June to July	20	8	24
Deaths brought forward due to short-term exposure to O_3			
Annual (no threshold)	16 140	15 725	16 230
Annual (35 ppb threshold)	1880	1510	1970
June to July (no threshold)	3480	3070	3575
June to July (35 ppb threshold)	880	510	970

N.B. Estimates of the "deaths brought forward" are rounded to the nearest multiple of 5.

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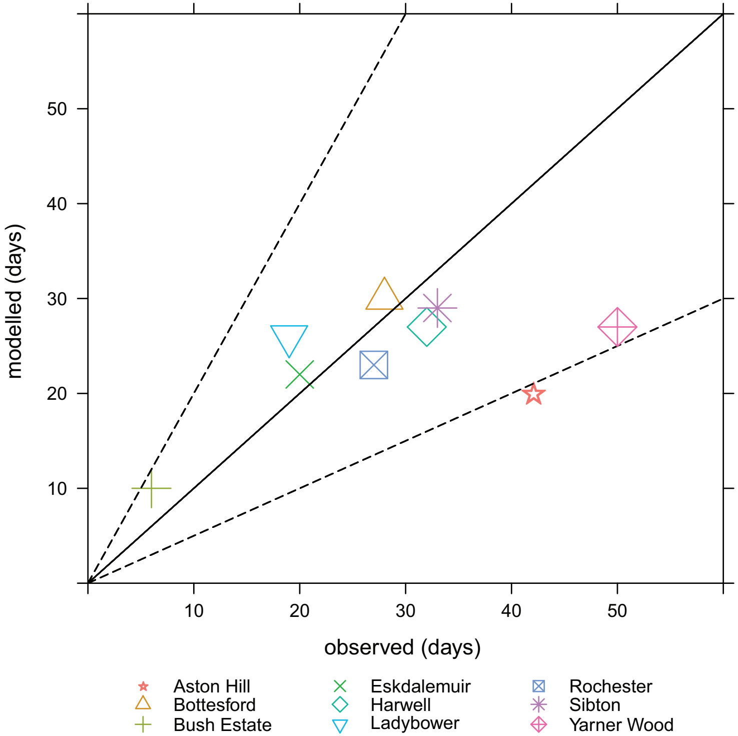

Fig. 1. Scatter plot of observed vs. modelled number days where the daily maximum 8-h mean O_3 concentration is > 100 µg m^{-3} (DM_{100}). Values shown are for nine rural sites with the calculations being made over a period between May and July 2006.

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(a) (b)

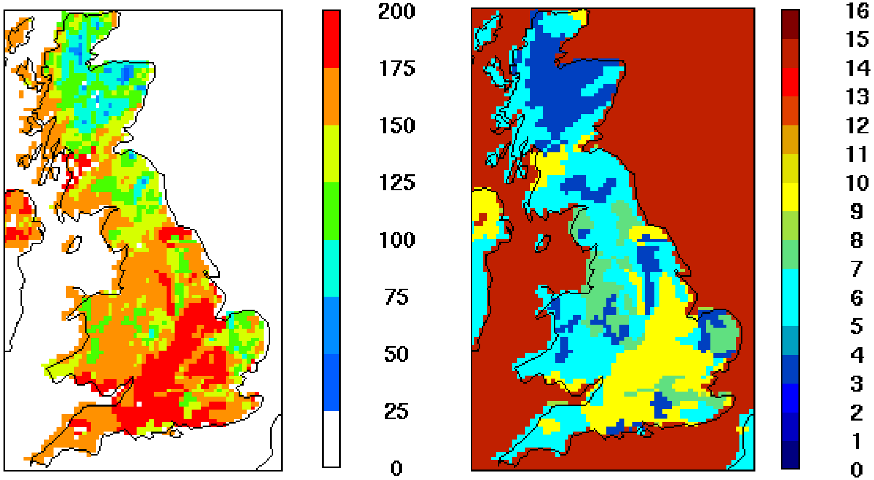

Fig. 2. (a) Soil moisture deficits (SMD, mm) estimated for the end of July for a grassland cover-type using the CMAQ-DO_3SE model and **(b)** the corresponding dominant soil type at the 9 × 9 km grid resolution used within the modelling. N.B. The major soil types in UK include clay loam (9), loam (6) and sandy loam (3).

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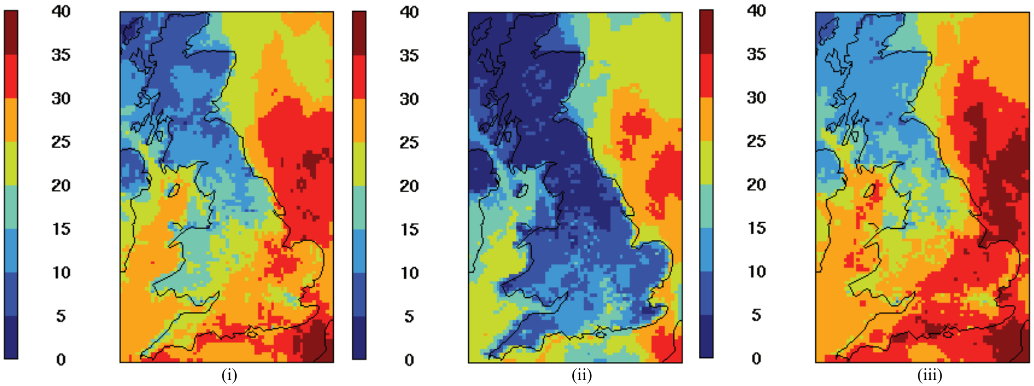

Fig. 3. Number of days when daily maximum 8-h average O_3 concentration > 100 µg m^{-3} (DM_{100}) during the June to July period in 2006 for **(i)** "reference"; **(ii)** "no stress" and **(iii)** "stress" scenarios.

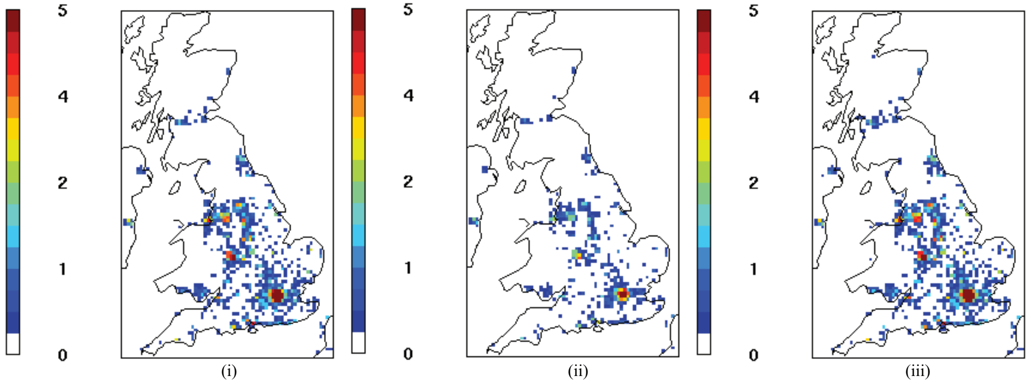

Fig. 4. Number of deaths brought forward due to short-term exposure to O_3 with a threshold value of 35 ppb for the June to July period for **(i)** "reference", **(ii)** "no stress" and **(iii)** "stress" scenarios.

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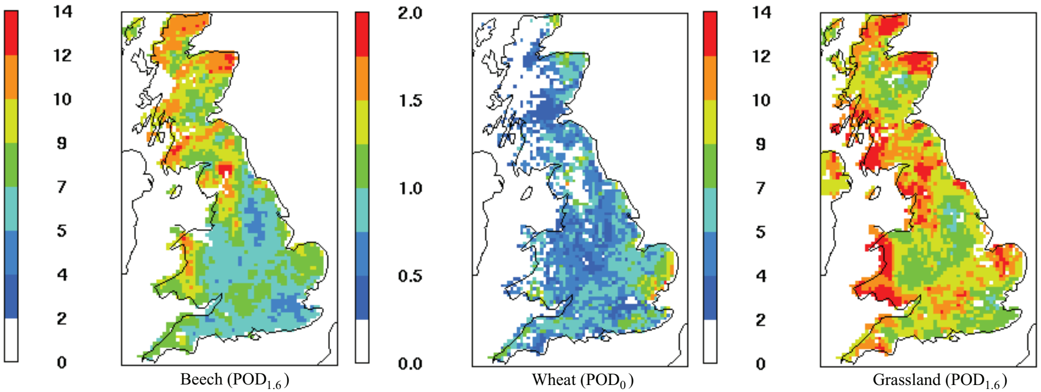

Fig. 5. The POD_y in mmol m^{-2} for beech, wheat and grasslands (with associated critical levels of 4 and 1 mmol m^{-2} respectively (currently there is no flux critical level established for grasslands) under the "reference" scenario for 2006.

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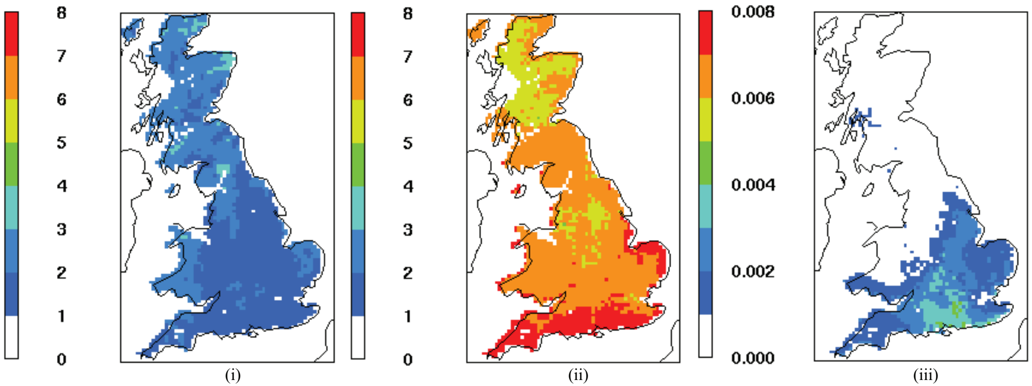

Fig. 6. The relative critical level exceedence (RCLe) using POD_y metrics for beech under the **(i)** "reference", **(ii)** "no stress" and **(iii)** "stress" scenario for 2006.

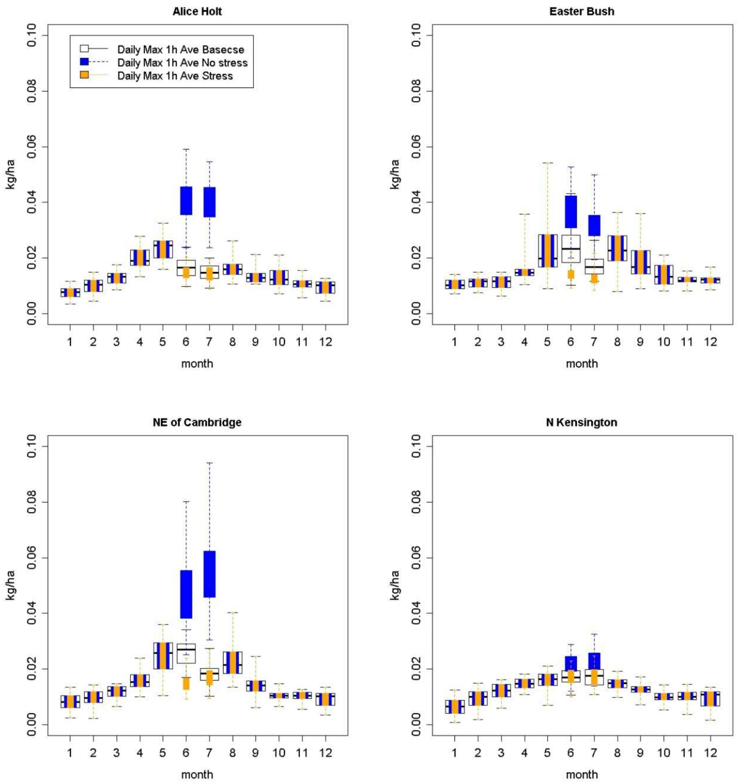

Fig. 7. Monthly variation of daily maximum 1-h average of O_3 deposition (kg O_3 ha^{-1}) at Alice Holt, Easter Bush, NE of Cambridge, and North Kensington.