TRENDS IN GROUND-LEVEL OZONE CONCENTRATION AND THEIR IMPLICATIONS FOR THE UK

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ABSTRACT
In the UK there is evidence of an increase in annual mean concentrations of between 0.1 and 0.3 ppb yr^{-1}. This is consistent with the impact of global increases in precursor (NO_x and VOC) emissions on background ozone concentrations. Results from a compilation of global ozone models predict a steady rise in background ozone concentrations over the northern hemisphere (1 to 16 ppb by 2050 for the UK). These data have been used, along with current ozone measurements to simulate changes in the UK ozone climate during the next few decades. The method used predicts hourly ozone concentrations at rural monitoring sites, allowing the calculation of critical levels and maps. These data are then used to assess the possible effects of ozone on human health and vegetation.

At present, the effect of ozone on human health is generally restricted to sensitive individuals, although increases in hospital admissions for respiratory illnesses are observed during episodes of high ozone. The results of this analysis show that by ~2030 people will be exposed to larger ozone concentrations more often and so sensitive individuals may have to restrict time spent outdoors. Ozone has adverse impacts on vegetation in the UK at current levels and the results indicate that vegetation will be exposed to higher ozone concentrations, for more of the time in the future, emphasising that vegetation is at particular risk.

INTRODUCTION
A number of important changes in the pollution climate of the UK have been identified in recent years. In the 1960s the long-range transport of SO_2 and the “acid-rain” it subsequently produced (Sweden, 1971), led to a series of agreements on pollutant emission control amongst European countries, the USA and Canada. Significant reductions in the UK’s emission of pollutants such as SO_2, VOCs and NOx have occurred since then. Ground-level ozone is mainly produced by reactions of NOx and VOCs in the presence of sunlight, and so its peak concentrations have also declined (NEGTAP, 2001). However, there is evidence of an increase in annual mean concentrations, of between 0.1 to 0.3 ppb yr^{-1} in the UK (Ashmore*, et al.*, 2003), Europe and N America (Bronnimann*, et al.*, 2002;Vingarzan, 2004). This is consistent with the impact of global increases in precursor emissions. Several modelling studies have predicted this trend and results from a compilation of global models (*Prather et al.*, 2003) anticipate a steady rise in background ozone over the UK of 1 to 16 ppb by 2050. Between 1995 and 2003 annual mean ozone concentrations in the rural areas of the UK ranged from 10 to 40 ppb, with peaks of 60 and on occasion over 100 ppb. Thus by 2050 annual average concentrations could be ~30 to 60 ppb with peaks of 160 ppb or more.

Ozone is toxic to plants as they take in ozone through their stomata as they respire; above concentrations of ~20 ppb the plants detoxification mechanisms are compromised and effects occur. Most studies have focused on the impact of peak exposures but there are a few experiments using mean concentrations in the range 20-50 ppb. These demonstrate the potential for adverse effects with concentrations in the range 40-50 ppb.

Different measures of the effective ozone dose can be used to assess vegetation's exposure, based on either atmospheric concentrations (eg AOT40) or the accumulated stomatal flux (AFst6, 6 nmol-O_3 m^{-2} s^{-1}). Concentration based studies show that concentrations are already causing visible injury and crop loss across much of the northern hemisphere (Fuhrer*, et al.*, 1997; Buse*, et al.*, 2003). However, these indices can only indicate a risk of damage, not quantify crop yield loss, for example. Flux indices for Europe, based on the actual plant dose, have recently been agreed by the ICP-Vegetation (ICP, 2004) and are expected to provide more realistic assessments (Pleijel*, et al.*, 2004). Mapping studies of ozone exposure across Europe show quite different patterns when AOT40 and stomatal flux are compared (Emberson*, et al.*, 2000). Although the AFst6 has yet to be mapped for the UK it has been evaluated for a typical wheat crop, showing that ozone dose already exceeds the critical level, even though the site's AOT40 does not.

Ozone also affects human health by causing inflammation in the lungs. At present, the effects are restricted to sensitive individuals, although increases in hospital admissions for respiratory illnesses are observed during episodes of high ozone. In the UK, an air quality objective of no more than 10 days per year having an 8-hour running mean ozone concentration above 50 ppb, by 2005, has been set. This level is based on the assumption that there is a threshold concentration for human health effects. COMEAP (the Committee on the Medical Effects of Air Pollution) have recommended a new metric without a threshold, to be related to health impacts: the annual mean daily maximum 8-hour running mean (Mudway and Kelly, 2003).

DATA and METHODS

Future Predictions

Prather *et al.*, (2003) compiled the results from 10 global ozone models that used a range of emission scenarios from the IPCC (Nakicenovic*, et al.*, 2000). Their meteorology and spatial resolution range widely but results for the present-day atmosphere (2000) simulate observations of O_3 and CO at remote sites quite well. Although varied, future predictions were consistent in magnitude and basic spatial distribution, with an increase in average surface ozone concentrations, mainly in the N hemisphere. Figure 1 shows the average, maximum and minimum monthly changes for the S/SE UK in 2050, using the A1B, A1FI, A2 and B1 emission scenarios (balanced growth/control, fossil fuel intensive, business as usual and sustainable, respectively). Ozone concentrations in the UK consist of the background with episodic peak values and other variations superimposed. The effects of increasing the background concentration have been simulated at all 18 rural monitoring sites in the UK national network (http://www.airquality.co.uk/) and 3 additional sites using the following methodology. Recent years, 1998 to 2002/03, were selected with all sites having >75% data capture for every year:

1. Calculate daily average 1000 to 1800 hr UTC (10-18h davg) from each hourly time series.
2. Smooth the 10-18h davg series using three passes of a 7 day-running mean (7drm 10-18h davg).

3. Calculate a 5 year average of the 7drm 10-18h davg, which is the daily average background series then allocate this to each hour of a day (ie 0100 to 2400 hours), to give an hourly average background time series.
4. Enhance these hourly values using the predicted change, in the 5^o x 5^o grid square containing the site, for that month.
5. Scale the future average background series by the relative difference between the current average background cycle and hourly average, ie re-impose typical hourly variations on the time series. The structure of the diurnal cycle and the amount of nocturnal ozone depletion is maintained, so the daily range (max – min) increases with time.

Effects Indices

The effects indices used in the analysis are calculated are follows:

1. AOT40 for crops and semi-natural vegetation, 3000 ppb h^{-1}

Accumulated ozone concentrations above 40 ppb, summed during May-July daylight hours where the ozone concentrations are for the canopy top and daylight is when solar radiation exceeds 50 Wm^{-2} (ICP, 2004). The AOT40 is very variable from year to year and so a 5-year mean is used for comparison with the critical level.

2. AFst6 for wheat, 1 mmol m^{-2} PLA*

Accumulated flux above a flux threshold of 6 nmol m^{-2} s^{-1}, accumulated during daylight hours. The time period for wheat is 970˚C days, starting 270˚C days before mid-anthesis (flowering). For *spring wheat*, mid-anthesis can be estimated using a temperature sum value of 1075^{o}C days calculated from plant emergence. For *winter wheat*, starting date for the accumulation of the effective temperature sum to mid-anthesis is the first date after 1 January when the temperature exceeds 0°C.

*PLA - projected leaf area

3. Number of days per year the 8 hour running mean (8hrm) exceeds 50 ppb (8hrm50), ≤ 10 days by 2005

The 8hrm's are calculated as hour-ending averages only where there are at least 6 hourly averages, ie the 1000-hour 8hrm includes 0300 – 1000 hours.

Ozone concentrations at the canopy height and stomatal flux

At most monitoring sites, ozone concentrations are measured at a fixed height of ~3 m, rather than at the canopy top. The dry deposition of ozone at the surface causes a large gradient in concentration between canopy top and 3 m therefore the data needs adjustment. For this analysis, measurements from Sutton Bonington during May-July 1999 to 2003 have been compiled, to provide "typical" data for a cereal crop: wind speed, temperature, ozone concentration (all at 3 heights), solar radiation, surface wetness, rainfall and wind direction are continuously monitored. Canopy top ozone concentrations and stomatal flux are then calculated using the standard resistance analogy (Monteith and Unsworth, 1990, equations 1 & 2) and a Jarvis type stomatal model (Tuovinen*, et al.*, 2004, equation 3 & 4), as follows:

$$\text{Total Ozone Flux } (\mu g\ m^{-2}\ s^{-1}),\ FO_3 = -|O_3|/(R_a + R_b + 1/(R_{st}^{-1}+R_{ct}^{-1}+R_g^{-1})) \quad (1)$$

Measurements of ozone, windspeed and temperature profiles are used to calculate the total O_3 flux, R_a, R_b and R_c (see refs. for a full description), from this the canopy concentration can be estimated using:

$$|O_3(z_0')| = |O_3(z)| (1 - (R_a(z) + R_b)/R_c) \text{ where } z_0' = \text{aerodynamic canopy height} \quad (2)$$

$$R_{st} = 1/(g_{max} \times f_{age} \times MAX(f_{min}, (f_{light} \times f_{temperature} \times f_{vapour\ pressure\ deficit} \times f_{soil\ water\ potential}))) \quad (3)$$

$$\text{Stomatal flux, } F_{st} = -|O_3(z_0')|/R_{st} \quad (4)$$

Full details of how the various factors in (3) are calculated can be found in Tuovinen*, et al.*, 2004 and references therein.

RESULTS

Future Trends

Summary statistics for the current and predicted hourly time series show that:

- past trends in the annual average concentrations are in line with predictions from the A1B (balanced growth/controls) scenario.
- the high emission scenarios (A1FI, A2) predict large increases in the annual mean by 2100, whereas A1B predicts a rise then decline to about current levels and, the most optimistic scenario, B1 predicts a decrease (Figure 2 for example).

Vegetation Effects

The two effects indices for vegetation are calculated using the Sutton Bonington summary data and the ozone time series from Somerton, as this should be representative of a typical cereal crop in Southern England. They give quite different pictures of exposure (Figure 3):

- AOT40 does not exceed the critical level at present and only significantly exceeds in the high emission scenarios, by 2030 in A1FI and A2 scenarios.
- $AF_{st}6$ currently exceeds the critical level and increases in the future.

However R_{st} will increase as CO_2 concentrations rise and decrease as temperature increase. This will affect surface deposition and so, AOT40 and $AF_{st}6$. This has been modelled for 2050 by decreasing g_{max} (equation (3)), and increasing ambient temperatures: decreasing g_{max} reduces AFst6, although it is still above the critical level, and increases AOT40 whereas increasing temperature does the opposite.

Human Health

The number of days the 8hrm exceeds 50 ppb has declined slightly, from around 22 on average in the 1980s to 20 at present; Figure 4 shows the spatial distribution in 2001, estimated using the methodology in Coyle*, et al.*, 2002. Assuming the downward trend at rural monitoring sites of ~1 day y^{-1} continues, there will still be over 10 days of exceedance across a significant area of the UK by 2005 (~30%). The future predictions for 2010, 2050, 2060, 2100 indicate that this trend will continue until 2010 after which the number of days exceeding 50 ppb starts to increase again. The average changes for the A1B and B2 scenarios give a decrease in 2100 but only B1 predicts exceedance on less than 10 days y^{-1}; the plot in Figure 5 illustrates these changes.

DISCUSSION

The data set of predicted hourly mean ozone concentrations for individual monitoring sites makes it possible to examine many aspects of the future ozone climate in the UK. In this paper we have focused on changes that could have significant impacts on our well-being and that of natural ecosystems:

Vegetation Effects
The AOT40 critical level for effects on crops and semi-natural vegetation is only exceeded at a few sites in the UK at present, however it is likely that the $AF_{st}6$ is exceeded across most of the country, assuming our results are representative. In the future, levels of both indices increase, and other changes in the ozone climate such as an earlier spring-time rise will occur. These could have significant impacts on crops yields and semi-natural communities, particularly in upland areas where the diurnal cycle in ozone is small. However it is difficult to quantitatively assess current or future effects of background ozone as modelling tools to calculate AF_{st} for the UK are still being developed. Other factors, such as land-use change, increases in CO_2 and temperature will also influence the severity of any ozone effects, particularly by 2100; different crops may be planted and CO_2 and temperature have opposing effects on stomatal conductance.
Human Health
The 8-hour running mean is currently used, by WHO (the World Health Organisation) and the UK government, to assess human health impacts. It exceeds 50 ppb on at least 10 days per year across most of the UK, generally only having effects sensitive individuals. Although the number of exceedance days is declining at present they will increase after 2010, and there could be more than 200 in some areas by 2100. This could have significant implications for sensitive people and those working or exercising outdoors.

In conclusion, although regional ozone-precursor emission controls have reduced peak ozone concentrations in the UK, global increases in emissions will lead to a rise in background ozone and hence concentrations across the UK. Increases will be greatest during the spring and summer, particularly in upland areas. These changes will have significant impacts on vegetation (natural and agricultural) and possibly on human health, by as soon as 2030. However, further work is needed to fully examine the data set of future concentrations and develop the tools to make quantitative predictions of impacts.

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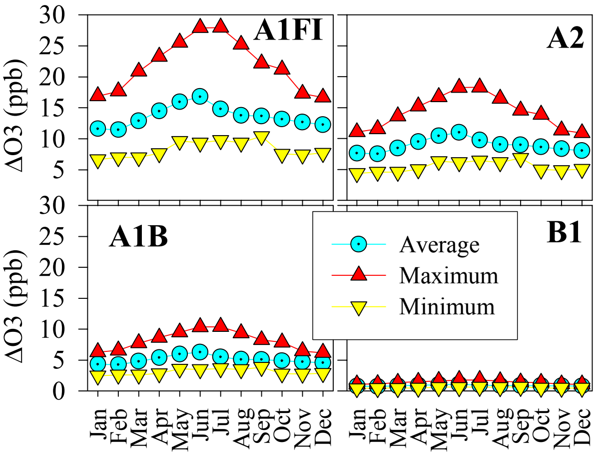

Figure 1. Average, maximum and minimum changes in ozone concentration by 2050 in S/SE England (5° x 5° grid square) from Prather, *et al.*, 2003 for 4 emissions scenarios (A1B, A1FI, A2 and B1,

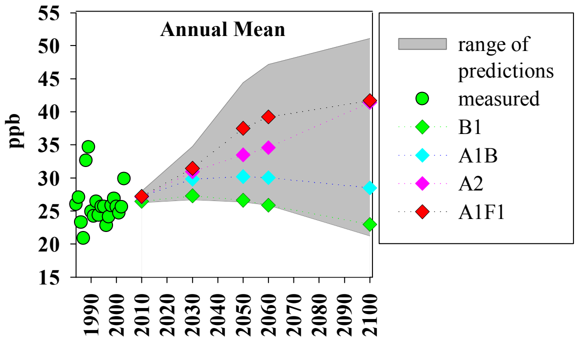

Figure 2. Measured data and predicted annual mean at Harwell in central England, using the average predicted change. The grey shaded area shows the full range from the maximum and minimum predicted changes.

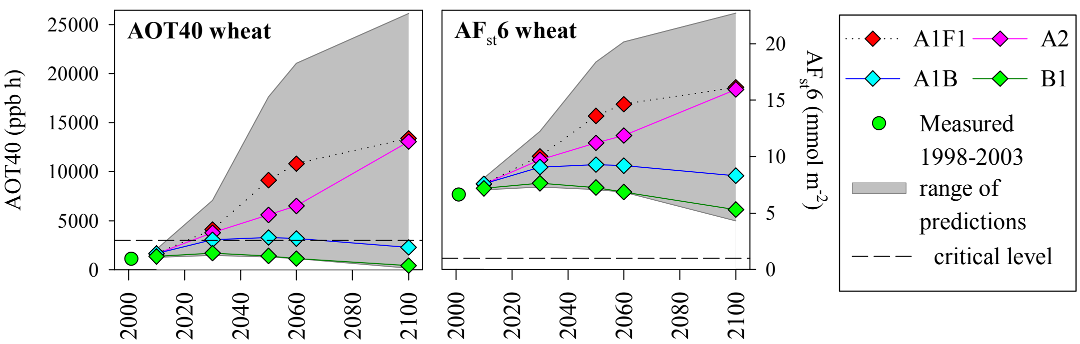

Figure 3. Trends in vegetation effects indices, the dashed line shows the critical level, modelled using micrometeorological data from Sutton Bonington (1.3 °W 52.8 °N) and ozone concentrations at Somerton (2.7 °W 51.0 °N).

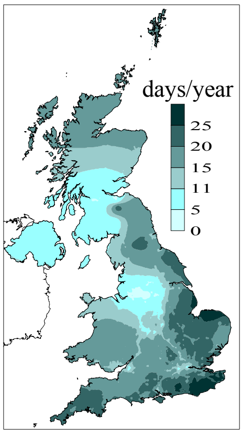

Figure 4. Number of days in 2001 that the 8-hour running mean exceeded 50 ppb (62% of the UK).

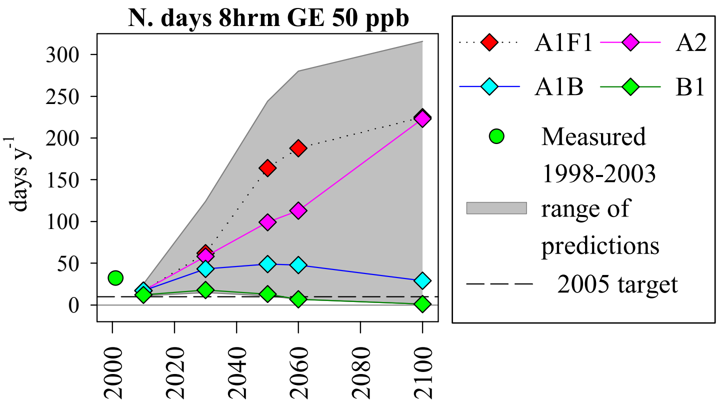

Figure 5. The current and future number of days per year the 8hrm exceeds 50 ppb at Ladybower in the English Midlands (1.8 ^{o}W 53.4 ^{o}N).