Supplementary Material

1. Health Burden Metrics:

The relative risk (RR) is used to derive the attribution fraction (AF), which represents the fraction of the disease burden attributable to the RR. The RR is represented by (see Silva et al., 2013):

\[ RR = e^{\beta(X - X_0)} \]

where \( \beta \) is the concentration response function (CRF), \( X \) is the maximum daily 8-hour running mean ozone concentration and \( X_0 \) is the threshold value (e.g. recommended safe health threshold).

The AF can then be determined from:

\[ AF = \frac{RR - 1}{RR} \]

The AF is combined with the baseline mortality rate (all-cause mortality, \( \phi \)) and the population sample (pop) to yield the excess mortality:

\[ \text{Excess mortality} = AF \cdot \phi \cdot \text{pop} \]

The excess mortality was calculated for each grid cell and then summed over all the grid-boxes that covered the UK.

2. Air Quality in the Unified Model (AQUM) Validation:

Figure SM1 shows the evaluation of AQUM (sampled at the observation sites) against AURN for 2006. AQUM captures the observational seasonal cycle over the UK sites within the observational monthly variability (i.e. standard deviation) (apart from March). Both AQUM and AURN have an ozone peak in June-July (70-80 µg/m\(^3\)) with minimum ozone in winter (30-50 µg/m\(^3\)). In July 2006 the UK was subject to conditions conducive to enhanced ozone production (Rebetez et al., 2008). Overall, AQUM falls within the observational uncertainty range. There is a good correlation of 0.83 and low mean bias (MB) of -6.03 µg/m\(^3\) (within the error range of ±9.68 µg/m\(^3\)) between the two seasonal cycles. The uncertainty range in the MB is based on the standard error with the influence of autocorrelation removed (Bence, 1995).

AQUM and AURN midday surface ozone were sampled under anticyclonic and easterly/southerly (NE, E, SE and S) conditions in summer 2006. The surface ozone anomalies for anticyclonic conditions for AURN (Figure SM2a) and AQUM (Figure SM2c) show positive anomalies of 0-10 µg/m\(^3\). The AQUM signal is similar but slightly weaker by 2-3 µg/m\(^3\) on average. Under easterly conditions (Figure SM2b & d), the surface ozone anomalies are larger (10-20 µg/m\(^3\)), but again AQUM is lower by 2-3 µg/m\(^3\). The spatial correlations between AQUM and AURN anomaly fields for both synoptic conditions are 0.55
and 0.62 (both significant at the 95% confidence level). Overall, AQUM has very similar anomaly fields to AURN and we therefore have confidence in the model and use the full AQUM domain maximum daily 8-hour mean surface ozone fields to calculate the health burden per day for the different synoptic conditions over the UK.

3. Tropospheric Emissions Spectrometer (TES) Carbon Monoxide (CO):

Figure SM3a shows UK TES CO profiles sampled under cyclonic, anticyclonic and south-easterly conditions with peak CO concentrations (in the peak AK sensitivity range) at 950 hPa of 125-135 ppbv. CO concentrations then decrease with altitude to approximately 100-120 ppbv at 600 hPa. In Figure SM3b, anticyclonic and cyclonic conditions lead to significant (>90% confidence level) increases or decreases in CO of 2-5 and 5-7 ppbv, respectively, consistent with the trapping and transport of CO. South-easterly conditions lead to the largest CO concentrations with significant positive anomalies of >5 ppbv between 950-600 hPa. This is consistent with Figure 3, which shows enhanced TES ozone under south-easterly conditions as well, supporting the argument of long-range pollution transport from continental Europe.

![Figure SM1](image)

**Figure SM1:** The seasonal cycle of surface ozone (µg/m³) in 2006 for the Air Quality in the Unified Model (AQUM, blue) and AURN (red). The uncertainty range (vertical bars) is based on the standard deviation. The MB, RMSE and correlation (R) are shown in the top right.
Figure SM2: AURN (top) and AQUM (bottom) summertime surface ozone anomalies (µg/m³), relative to the seasonal average, for a), c) anticyclonic conditions and b), d) S, SE, E, NE flows in 2006.
Figure SM3: TES summer time vertical CO profiles (ppbv) over the UK (12°W-6°E, 48°-62°N) between 2005-2011 sampled under anticyclonic (red), cyclonic (blue) and south-easterly (green) conditions. In panel a) the black line is the summer time average. The dotted lines show the variability (standard deviation) in the composite profiles. In panel b) the profiles are shown relative to the seasonal average and the black dashed line is the zero bias line. Horizontal solid lines represent the profile uncertainty. The yellow region in both panels is the approximate region of peak TES sensitivity to retrieving lower tropospheric CO. Squares and diamonds signify where the profiles are significantly different to the summer time average at the 90% and 95% confidence levels.

References:

