Supporting Information: Widespread changes in UK air quality observed from space

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S1. Satellite Data Gridding Algorithm

Figure S1 shows an example Ozone Monitoring Instrument (OMI) pixel (pale blue) over the UK and pixel corners (black circles). The blue circle represents the centre of the OMI pixel and this is often used to interpolate the data onto a regular grid shown by the purple grid box at a 0.25° × 0.25° resolution ($Grid_{low}$; black lines). In such methods, a large proportion of the OMI pixel information is lost from $Grid_{low}$ as the proportion of the pixel outside the purple box is not included on the grid. To avoid this, we split the OMI pixel into many of sub-points, which are mapped onto a higher resolution 0.05°×0.05° grid (green dashed lines; $Grid_{high}$). In the **Figure S1a** example, the OMI pixel is split into 20 points (n_slice) along each axis leading to 400 points (red crosses). These sub-points are then mapped onto $Grid_{high}$. Now all grid boxes on $Grid_{high}$ which contain red crosses are given the same value as that of the OMI pixel. Therefore, a much larger proportion of the OMI pixel will be transformed onto a regular grid where the daily (13.30 LT) average is determined for each $Grid_{high}$ grid box if there is overlapping of OMI pixels. Averaging the OMI tropospheric column NO_2 ($TCNO_2$) $Grid_{high}$ data to get e.g. monthly composites results in significantly higher resolution spatial distributions necessary for detailed assessment of UK air quality. This is shown for 2005-2006 in **Figure S1b**) and **c)** where the OMI $TCNO_2$ is respectively mapped onto $Grid_{high}$ using only the centre point of the pixel and all the sub-points considered by our gridding algorithm. In **Figure S1b**), there are areas of missing data (white grid boxes) and the spatial pattern, especially over the hot spots, is extremely noisy. However, in **Figure S1c**), as more of the OMI pixel is mapped onto $Grid_{high}$, the hot spots are clearly defined and there is more data available for statistical analysis. The same is true for the Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol optical depth (AOD) data in **Figures S1f)** and **1g)**, respectively.

OMI sub-column (0-6 km) O_3 (SCO_3) data, was mapped onto a 0.5°×0.5° grid. In the Rutherford Appleton Laboratory (RAL) product, the spatial resolution is reduced to improve the signal-to noise ratio when retrieving tropospheric O_3 to approximately 50 km × 50 km (i.e. the across-along track pixels are merged at a ratio of 2:4 pixels). Therefore, in the processing of the OMI data, there are fewer O_3 than NO_2 pixels. Due to the homogeneity of O_3 compared with NO_2, we find that a 0.5° × 0.5° regular grid is satisfactory for the gridding algorithm. If the OMI O_3 data is directly mapped onto the same grid using only the pixel centre point, the spatial maps become noisy so data have to be gridded to a coarser resolution (e.g. 1.0° × 1.0° or 2.0° × 2.0°). This is highlighted in **Figures S1d**), where only using the pixel centre point to map onto the 0.5° × 0.5° grid results in a noisy spatial pattern. Using the algorithm presented here (**Figure S1e**), the availability of extra pixel information yields a clearer spatial pattern (i.e. the land-sea O_3 gradient).

S2. Spatial Trend Maps

Figure S2 shows the OMI $TCNO_2$ ($\times 10^{15}$ molecules/cm^2/year) and MODIS AOD (AOD/year) trends for 2005 to 2015. $TCNO_2$ negative trends peak at -0.5 $\times 10^{15}$ molecules/cm^2/year over London and -0.3 to -0.1 $\times 10^{15}$ molecules/cm^2/year over the other urban hotspots. AOD trends are more homogeneous spatially with negative trends of -0.001 to 0.0 /year across the majority of the UK. The stronger $TCNO_2$ trend gradient will be linked to the larger emission of NO_2 over the hotspots and its short lifetime. However, aerosol has a longer lifetime (e.g. several weeks), which results in a more uniform spatial distribution of the pollution. Also, natural aerosol will be emitted in the UK rural regions further enhancing the AOD spatially homogeneous distribution.

$TCNO_2$ positive trends in parts of the UK rural background regions have been removed from the left panel in Figure S2, where the 2005-2015 average $TCNO_2$ concentration is greater than 5.0×10^{15} molecules/cm^2, as pixels affected by the OMI row anomaly (Braak, 2010), which have managed to pass our quality assessment of the OMI $TCNO_2$ data, can lead to artificial positive trends in regions with low NO_2 concentrations. This artificial NO_2 enhancement with time has negligible impact though on trends calculated over source regions as the concentrations are too large and any trend (e.g. London decreasing trend) will mask this artificial positive trend.

References:

Braak R. 2010. Row Anomaly Flagging Rules Lookup Table, KNMI Technical Document TN-OMIE-KNMI-950, *KNMI*, Netherlands.

Figures:

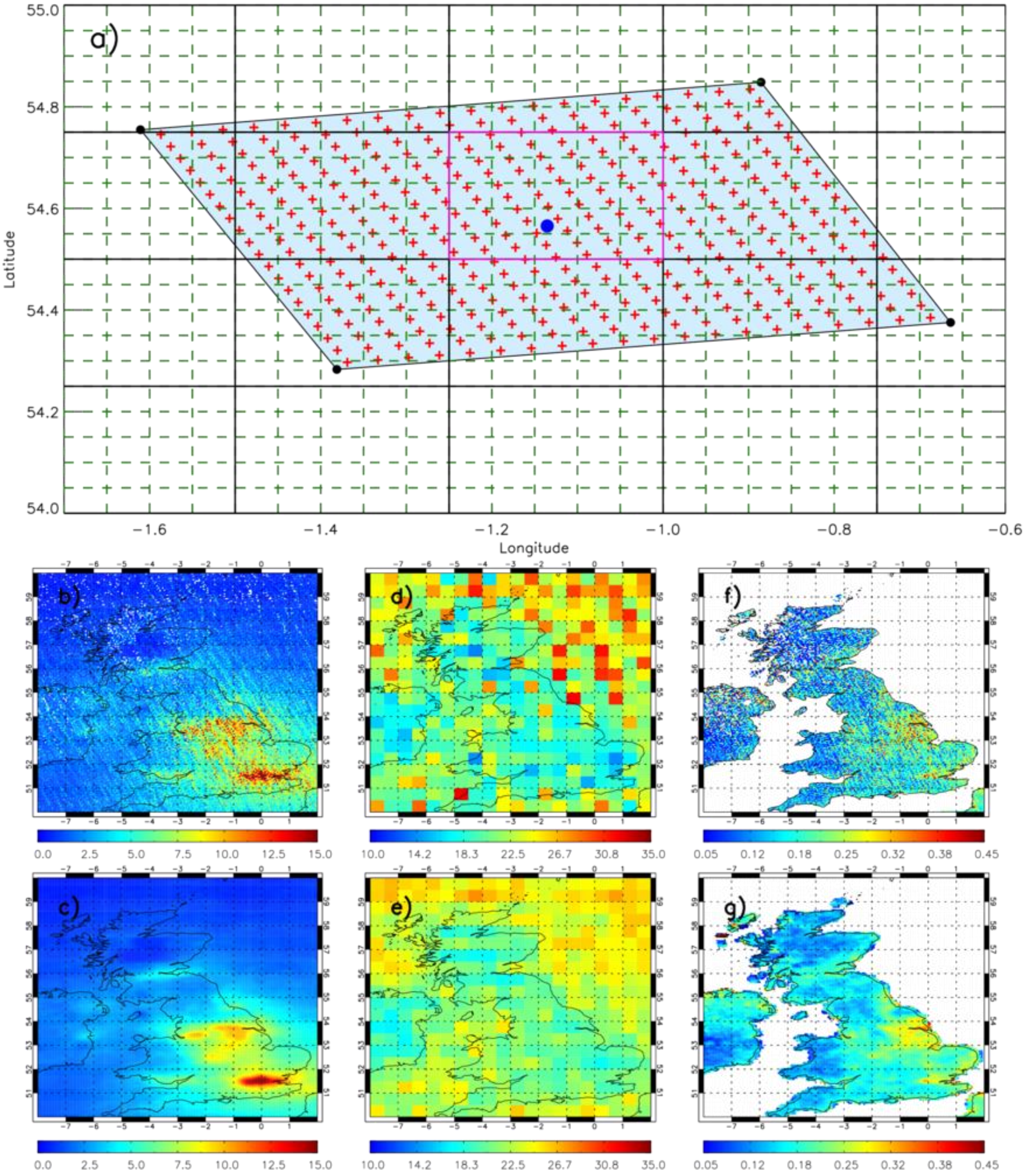

Figure S1: a) Schematic of an OMI pixel (pale blue) over the UK with the centre point (blue) plotted with multiple sub-pixel points (red crosses) which are mapped to a high resolution (0.05° x 0.05°) grid (green dashed lines) compared with the coarser grid (0.25° x 0.25°; black lines). The purple grid box indicates where the entire OMI pixel information, using the centre point, would have been mapped onto the regular grid resulting in a loss of information. By using the many sub-pixel points, the entire pixel information is mapped onto the higher resolution grid. OMI $TCNO_2$ ($\times 10^{15}$ molecules/cm^2), SCO_3 (Dobson Units - DU) and MODIS AOD data for 2005-2006 have been mapped onto the high resolution grid (0.05° x 0.05° for NO_2, 0.5° x 0.5° for O_3 and 0.05° x 0.05° for AOD) using only the pixel centre point information (b), d) and f)) and then using the pixel sub-points (c), e) and g)).

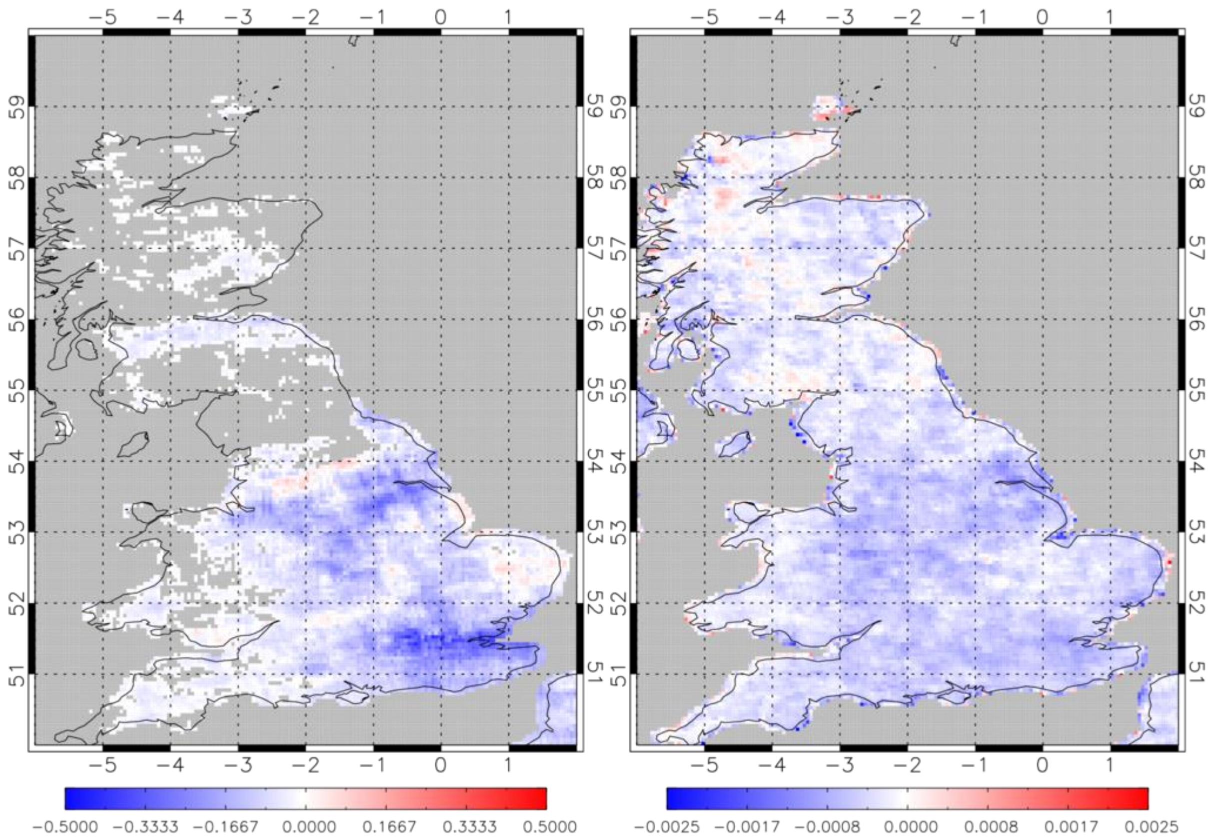

Figure S2: OMI $TCNO_2$ (left) and MODIS AOD (right) trends ($\times 10^{15}$ molecules/cm^2/year and AOD/year) for 2005 to 2015. Significant and insignificant trends are plotted on the grid resolution of 0.05°×0.05°.