

## RESEARCH ARTICLE

# Widespread changes in UK air quality observed from space

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Previous studies have used surface observations of pollutants (e.g., nitrogen dioxide [NO<sub>2</sub>] and aerosols) to evaluate improvements in United Kingdom (UK) air quality over recent decades. However, surface monitoring networks provide limited spatial coverage and are not always representative of air quality over a wider region. Satellite observations, such as tropospheric column NO<sub>2</sub> (TCNO<sub>2</sub>), aerosol optical depth (AOD) and sub-column (0–6 km) ozone (SCO<sub>3</sub>), provide widespread monitoring of air quality on a national scale. In this study, we use such observations to analyse trends in UK air quality, between 2005 and 2015, finding significant decreases in TCNO<sub>2</sub> and AOD over UK pollution hotspots. The largest changes in short-lived NO<sub>2</sub> are located over populated (i.e., source) regions, associated with large emissions of nitrogen oxides (NO<sub>x</sub>). AOD changes are more spread out, and less strongly associated with source regions, due to a longer aerosol lifetime, secondary aerosol formation and non-urban aerosol sources. SCO<sub>3</sub> shows no significant trends over England/Wales, but significant positive trends over Scotland, which is consistent with previous studies of changes in background tropospheric ozone in other remote regions of north-western Europe.

**KEYWORDS**

air quality, earth observation, tropospheric column NO<sub>2</sub>

## 1 | INTRODUCTION

Degradation in air quality in the United Kingdom (UK) is an important public health concern, estimated to result in approximately 40,000 premature deaths per year (Holgate, 2017) and an average reduction in life expectancy of 7–8 months (House of Commons, 2010). Exposure to pollutants such as ozone (O<sub>3</sub>), nitrogen dioxide (NO<sub>2</sub>) and particulate matter (PM<sub>2.5</sub> & 10—atmospheric particles with diameters less than 2.5 and 10 μm, respectively) leads to health ailments such as lung disease and cancer, cardiovascular problems, asthma and eye irritation (WHO, 2014). In light of these concerns, government policy has led to the monitoring of many pollutants (e.g., Automated Urban and Rural Network [AURN], Department for Environment, Food and Rural Affairs [DEFRA], 2015) and efforts to reduce their emissions (e.g., the Environment Act 1995, the

National Emission Ceilings Regulations 2002 and the Air Quality Standards Regulations 2010; DEFRA, 2011). Multiple studies have highlighted the impact of emission control legislation on abundances of pollutants, such as nitrogen oxides (NO<sub>x</sub>) and aerosol, over the United Kingdom (e.g., Carslaw *et al.*, 2011; Turnock *et al.*, 2015). Trends between 2000 and 2014 in UK summer-time surface concentrations of the secondary pollutant tropospheric O<sub>3</sub>, show significant variability but an overall increase (Chang *et al.*, 2017). Munir *et al.* (2013) found positive trends in mean surface O<sub>3</sub> at urban and rural AURN sites, 1993–2011, but a decrease in peak O<sub>3</sub> concentrations. Since surface monitoring sites are point measurements, they are often unrepresentative of pollution over a wider region given inhomogeneities in air pollutant concentrations that are produced by strong gradients in surface emissions and complex meteorology. Satellite observations, given their wide spatial

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coverage, may offer an opportunity to assess changes in UK air quality on a national scale.

Only a few studies have used satellite data to evaluate UK or north-western Europe air quality. For instance, van der A *et al.* (2008) and Zhou *et al.* (2012) used satellite tropospheric column NO<sub>2</sub> (TCNO<sub>2</sub>) to identify negative trends in NO<sub>2</sub> over the United Kingdom/Europe between 1996–2005 and 2004–2009, respectively. Pozzer *et al.* (2015) detected large negative trends in aerosol optical depth (AOD) over Europe (including United Kingdom), between 2001 and 2010, from the Moderate Resolution Imaging Spectroradiometer (MODIS), but this was much weaker for other instruments. Alpert *et al.* (2012) also found negative AOD trends over London, between 2002 and 2010, from MODIS-Terra, but MODIS-Aqua and the Multi-angle Imaging Spectroradiometer (MISR) observed positive trends. This study utilises satellite observations of TCNO<sub>2</sub> and AOD to investigate more recent changes in UK pollution, building upon the previous studies mentioned above. However, by using higher resolution satellite data, we are able to investigate UK pollution hotspots, such as Manchester, Birmingham and the Yorkshire power stations, which have not been investigated before in significant detail. We also utilise satellite measurements of sub-column (0–6 km) O<sub>3</sub> (SCO<sub>3</sub>) for the first detailed look at tropospheric O<sub>3</sub> trends over the United Kingdom from space. Section 2 discusses the satellite data and gridding methodology used to obtain satellite data at a high resolution, section 3 presents our results and section 4 highlights our conclusions.

## 2 | SATELLITE DATA AND HIGH RESOLUTION GRIDDING ALGORITHM

### 2.1 | Satellite data

TCNO<sub>2</sub> and SCO<sub>3</sub> data (Miles *et al.*, 2015) come from the Ozone Monitoring Instrument (OMI), on-board NASA's EOS-Aura satellite, which are provided by the Tropospheric Emissions Monitoring Internet Service and the Rutherford Appleton Laboratory (RAL), respectively. AOD data is from MODIS, on-board NASA's EOS-Aqua satellite, which has been obtained directly from NASA. Both Aura and Aqua are polar orbiting with approximate equator crossing times of 13:30 local time (LT). OMI and MODIS have nadir-viewing spectral ranges of 270–500 nm (Boersma *et al.*, 2007) and 0.41–15 μm (Remer *et al.*, 2005), respectively, with pixel sizes in the order of ~10–100 km × ~10–100 km depending on viewing angle. All data sets have been quality controlled for geometric cloud fraction less than 0.2, good quality flags and the OMI row anomalies (Braak, 2010) where applicable.

### 2.2 | Gridding algorithm

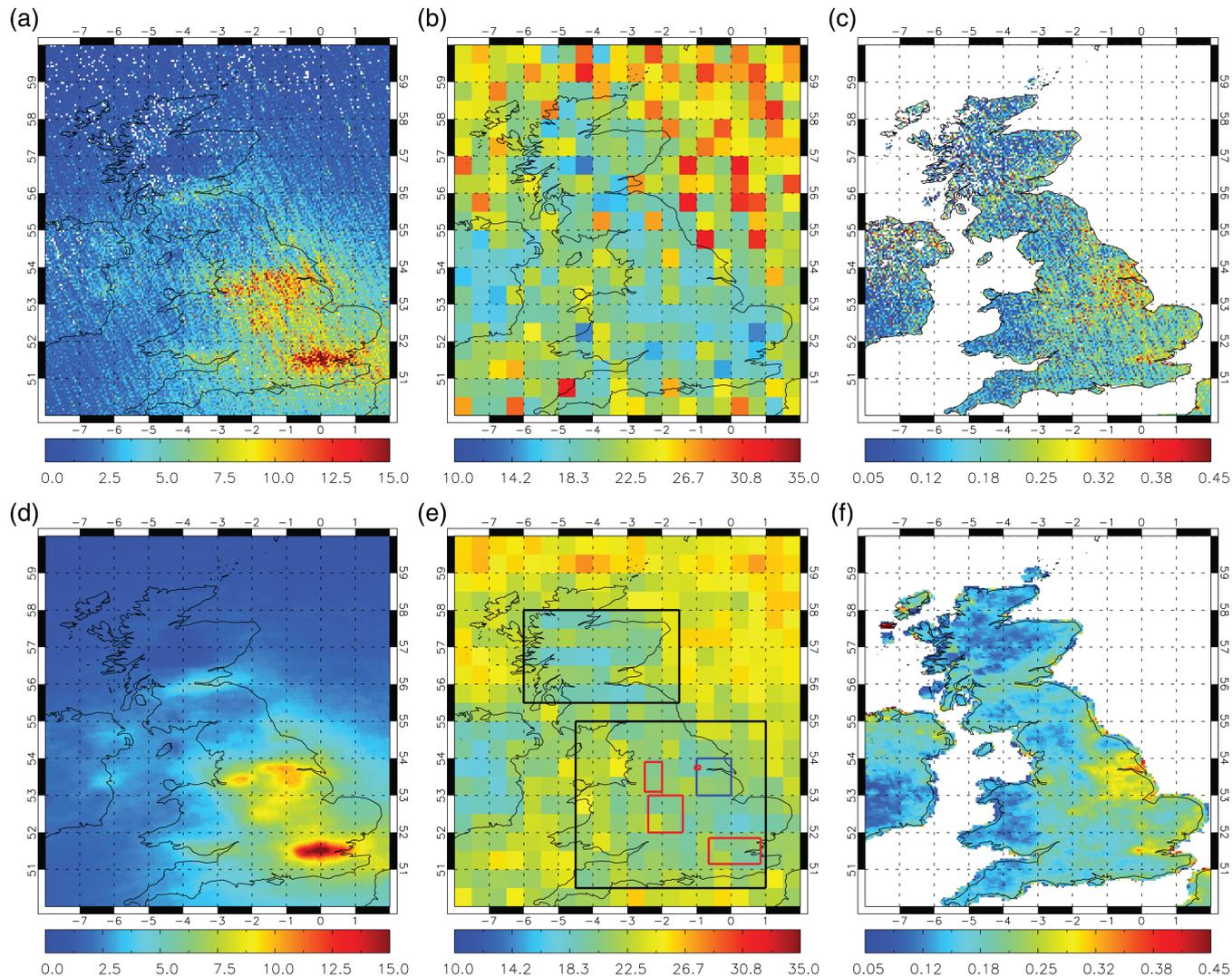
Several studies have applied complex methods to map air quality satellite data onto high resolution regular grids (e.g., 0.05° × 0.05°). This provides much more spatial detail than the resolution of 0.25° × 0.25° used in many studies such as Pope *et al.* (2014) and Zhou *et al.* (2012). Russell *et al.* (2011) use higher resolution inputs than the standard TCNO<sub>2</sub> products to generate the air mass factors (AMFs) in the retrieval process for the Berkeley High Resolution (BEHR) TCNO<sub>2</sub> product. Barkley *et al.* (2017) used the area-weighting tessellation algorithm of Spurr (2003) to investigate air quality over the Middle East. In this study we use a simpler methodology of slicing up satellite pixels and mapping them onto a higher resolution grid, which is less computationally expensive, while maintaining a robust data product. Often, only the central position of the retrieval is used to map satellite data onto a regular grid. This can result in missing information if the retrieval pixel overlaps several grid boxes on a higher resolution regular grid (e.g., 0.05° × 0.05° instead of 0.25° × 0.25°). By using the central and corner positions of the satellite pixel, this study slices the pixel into many sub-pixels, which are mapped onto the higher resolution grid retaining much of the information and yielding more detailed spatial patterns. This is shown in Figure 1 for TCNO<sub>2</sub> (a,d), SCO<sub>3</sub> (b,e) and AOD (c,f), where TCNO<sub>2</sub> and AOD are mapped onto a 0.05° × 0.05° grid and SCO<sub>3</sub> is mapped onto 0.5° × 0.5° grid. Even though TCNO<sub>2</sub> and SCO<sub>3</sub> are both retrieved from OMI with the same horizontal resolution, multiple swath pixels have to be merged together in the RAL O<sub>3</sub> retrieval process to get a robust tropospheric signal-to-noise ratio due to stratospheric O<sub>3</sub> interference. Therefore, the horizontally coarser OMI SCO<sub>3</sub> data is mapped onto a lower resolution grid. This gridding technique is discussed in more detail in File S1, Supporting information.

## 3 | RESULTS

We examined the 2005–2015 TCNO<sub>2</sub>, SCO<sub>3</sub> and AOD trends using the methodology of van der A *et al.* (2006, 2008). Here the monthly species' time-series is represented by the function:

$$Y_t = C + BX_t + A \sin(\omega X_t + \phi) + N_t, \quad (1)$$

where  $Y_t$  is the monthly species column for month  $t$ ,  $X_t$  is the number of months after January 2005,  $C$  is the January 2005 species concentration,  $B$  is the monthly linear trend and  $A \sin(\omega X_t + \phi)$  is the seasonal model component (Weatherhead *et al.*, 1998).  $A$  is the amplitude,  $\omega$  is the frequency (set to 1 year;  $\omega = \pi/6$ ) and  $\phi$  is the phase shift.  $C$ ,  $B$ ,  $A$  and  $\phi$  are the fit parameters based on a nonlinear least squares fit.  $N_t$  represents the model errors/residuals. The linear trend,  $B$ , is determined to be significant at the 95%



**FIGURE 1** OMI TCNO<sub>2</sub> ( $\times 10^{15}$  molecules/cm<sup>2</sup>), SCO<sub>3</sub> (Dobson units, DU) and MODIS AOD data for 2005–2006 have been mapped onto the high resolution grid ( $0.05^\circ \times 0.05^\circ$  for NO<sub>2</sub>,  $0.5^\circ \times 0.5^\circ$  for O<sub>3</sub> and  $0.05^\circ \times 0.05^\circ$  for AOD) using only the pixel centre point information (a, b and c) and then using the pixel sub-points (d, e and f). The red and black boxes in panel (e) represent the regions the TCNO<sub>2</sub> and SCO<sub>3</sub> trends are calculated for, respectively. AOD trends are calculated over the same regions as TCNO<sub>2</sub> except for the Yorkshire region, shown by the blue box

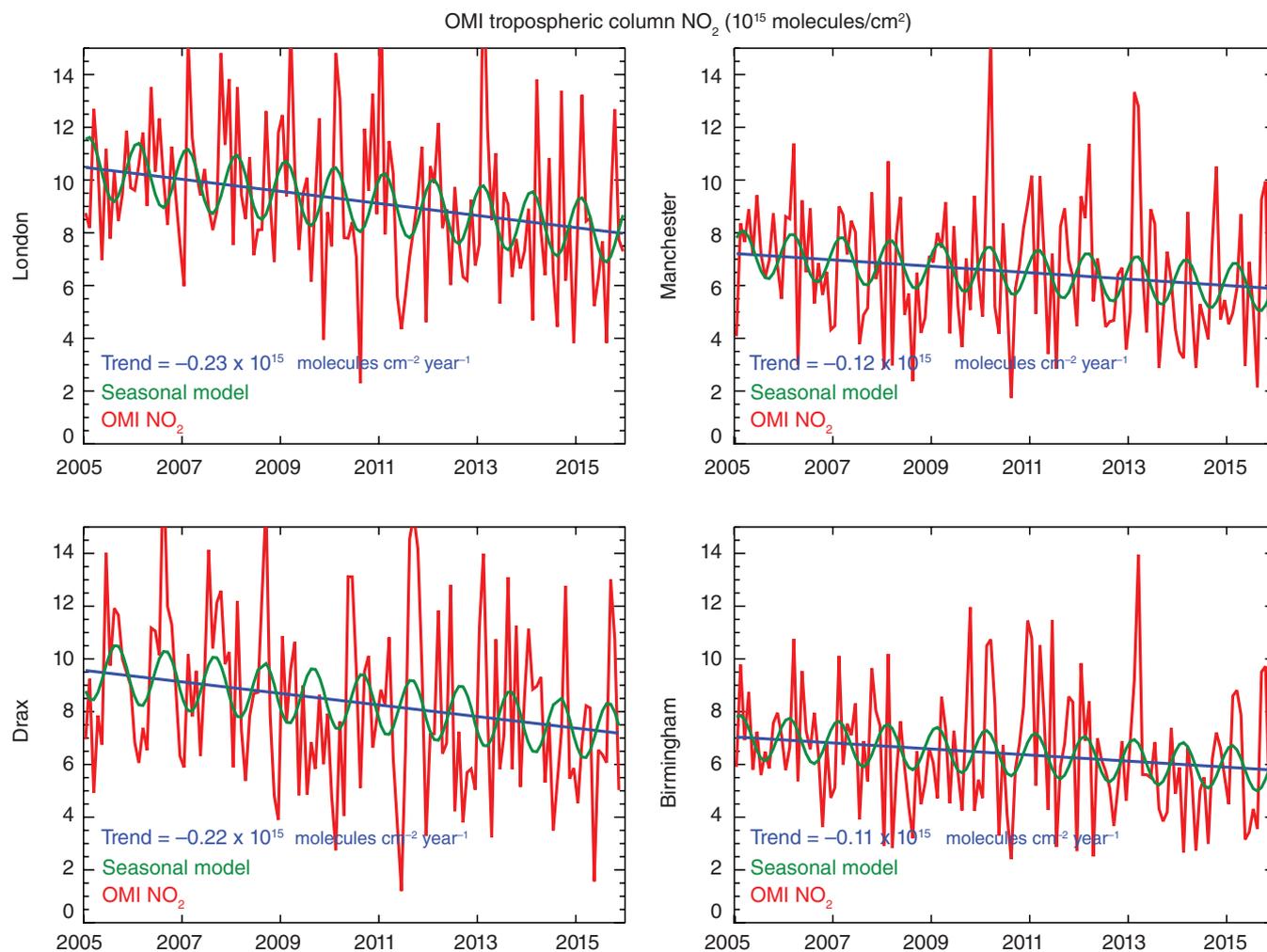
confidence level if  $|B/\sigma_B| > 2.0$  (Weatherhead *et al.*, 1998).  $\sigma_B$  represents the precision of the trend. This is calculated by:

$$\sigma_B \approx \left[ \frac{\sigma_N}{n^{3/2}} \sqrt{\frac{1+\alpha}{1-\alpha}} \right], \quad (2)$$

where  $n$  is the number of years,  $\alpha$  is the autocorrelation in the residuals ( $N_t$ ) and  $\sigma_N$  is the standard deviation in the residuals. As in van der A *et al.* (2006), we calculate the autocorrelation for each time-series using a lag of one time step (i.e., 1 month). Autocorrelation in the species time-series is not accounted for in Equation 1, so is factored into the trend uncertainty (Equation 2), as used and discussed by van der A *et al.* (2006) and Weatherhead *et al.* (1998), respectively. Like van der A *et al.* (2006), we typically find autocorrelation values of 0.1 for TCNO<sub>2</sub>, while similarly small autocorrelations of approximately 0.1 and 0.05–0.2 for AOD and SCO<sub>3</sub>. Monthly time-series for all pollutants were constructed

applying the quality criteria stated in section 2.1. In addition, for each OMI and MODIS grid box the mean and standard deviation were calculated from the daily species time-series, between 2005 and 2015, and where individual day values were greater than the mean plus 3.0 standard deviations (outliers), they were removed from the time-series when calculating the monthly means used for the trend analysis (Equation 1).

For TCNO<sub>2</sub> and AOD four hotspot regions were investigated defined as London, Manchester, Drax (Yorkshire/Humberside for AOD) and Birmingham from the pollutant spatial distributions in Figure 1d,f, respectively. London, Manchester and Birmingham represent large populous urban regions, while Drax is a large power station in Yorkshire. Pope and Provod (2016) identified Drax, along with the nearby Eggborough and Ferrybridge C power stations, as the source of the North-East Yorkshire TCNO<sub>2</sub> hotspot. In each of the regions, we see significant negative linear trends (blue lines, Figure 2) over the 11-year period in TCNO<sub>2</sub>; London ( $-0.23 \pm 0.05 \times 10^{15}$  molecules cm<sup>-2</sup> year<sup>-1</sup>), Manchester



**FIGURE 2** OMI TCNO<sub>2</sub> trends (10<sup>15</sup> molecules cm<sup>-2</sup> year<sup>-1</sup>), 2005–2015, at London (0.65°W–0.85°E, 51.15–51.85°N), Manchester (2.50–2.00°W, 53.10–53.90°N), Drax (1.05–0.90°W, 53.70–53.80°N) and Birmingham (2.40–1.40°W, 52.00–53.00°N). Red is the OMI TCNO<sub>2</sub> monthly time-series, blue is the OMI NO<sub>2</sub> linear trends (term *BX*, in Equation 1) and green is the seasonal component (Equation 1) of the OMI NO<sub>2</sub> trends. All significant OMI TCNO<sub>2</sub> trends have insignificant trends in the stratospheric slant columns and the tropospheric AMFs

( $-0.12 \pm 0.06 \times 10^{15}$  molecules cm<sup>-2</sup> year<sup>-1</sup>), Drax ( $-0.22 \pm 0.10 \times 10^{15}$  molecules cm<sup>-2</sup> year<sup>-1</sup>) and Birmingham ( $-0.11 \pm 0.05 \times 10^{15}$  molecules cm<sup>-2</sup> year<sup>-1</sup>). We calculate insignificant trends in the stratospheric slant columns and tropospheric AMFs, both used to derive the tropospheric column (Boersma *et al.*, 2004), giving confidence to our interpretation of the negative TCNO<sub>2</sub> trends found here. Similar negative trends were seen in the AOD (Figure 3); London ( $-0.0061 \pm 0.0026/\text{year}$ ), Manchester ( $-0.0057 \pm 0.0017/\text{year}$ ), Yorkshire/Humberside ( $-0.0050 \pm 0.0019/\text{year}$ ) and Birmingham ( $-0.0052 \pm 0.0024/\text{year}$ ). However, it should be noted that the AOD time-series over Manchester has a large proportion of missing data in the winter-time. The specific longitude–latitude grid boxes (shown in Figure 1e) used to define these locations are described in the captions of Figures 2 and 3. Some of the TCNO<sub>2</sub> and AOD boxes (Figure 1e) are slightly larger than the hotspot domain to obtain sufficient data (grid boxes) to identify significant (non-significant) trends in the TCNO<sub>2</sub> (stratospheric slant column NO<sub>2</sub> and AMFs). Maps showing the spatial

distribution of all grid box trends for NO<sub>2</sub> and AOD are shown in File S1.

Our London TCNO<sub>2</sub> trend is nearly four times larger than that found by van der A *et al.* (2008), between 1996 and 2006 (GOME/SCIAMACHY TCNO<sub>2</sub>) ( $-0.06 \times 10^{15}$  molecules cm<sup>-2</sup> year<sup>-1</sup>), most likely since they are from different time periods. Zhou *et al.* (2012), between 2004 and 2009 (OMI TCNO<sub>2</sub>), found trends of approximately  $-5.0\%/ \text{year}$ ,  $-12.0\%/ \text{year}$ ,  $-7\%/ \text{year}$  and  $-12\%/ \text{year}$  over London, Manchester, Drax and Birmingham, respectively. The trends we present here are smaller, typically  $-2.0$  to  $-1.0\%/ \text{year}$ , and represent 11 years rather than 5 years. This suggests that there has been a significant decrease in TCNO<sub>2</sub> over the past 20 years (i.e., 1996–2015), but with peak decreases between 2004 and 2009. Negative AOD trends range between  $-3.3$  and  $-2.8\%/ \text{year}$  across the regions studied here. This is similar to MODIS-Terra AOD trends reported by Pozzer *et al.* (2015) and Alpert *et al.* (2012) between 2001/2002 and 2010. However, Alpert *et al.* (2012) found positive trends from MODIS-Aqua of  $8.4\%/ \text{year}$  and  $7.9\%/ \text{year}$  over London and Birmingham, respectively.

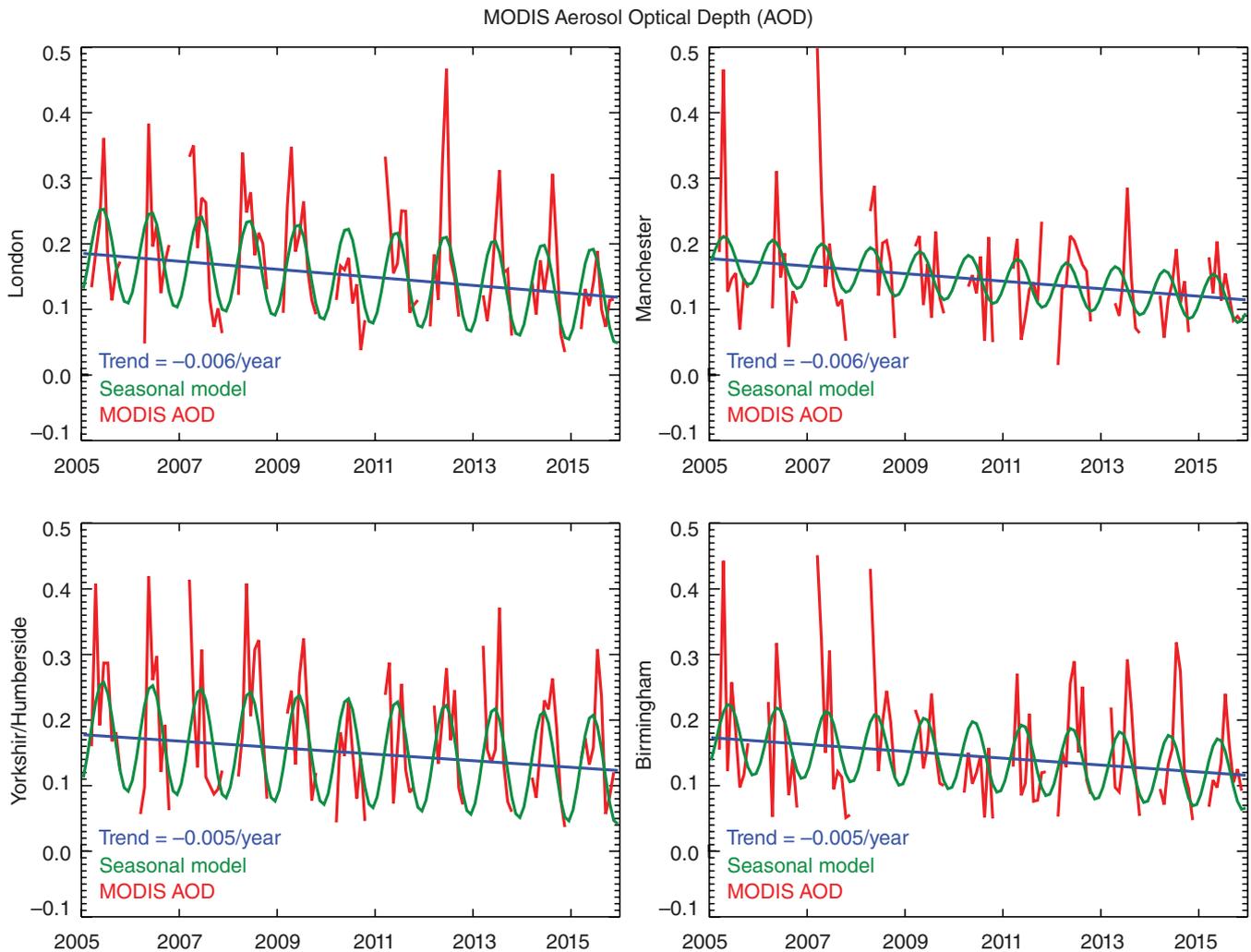
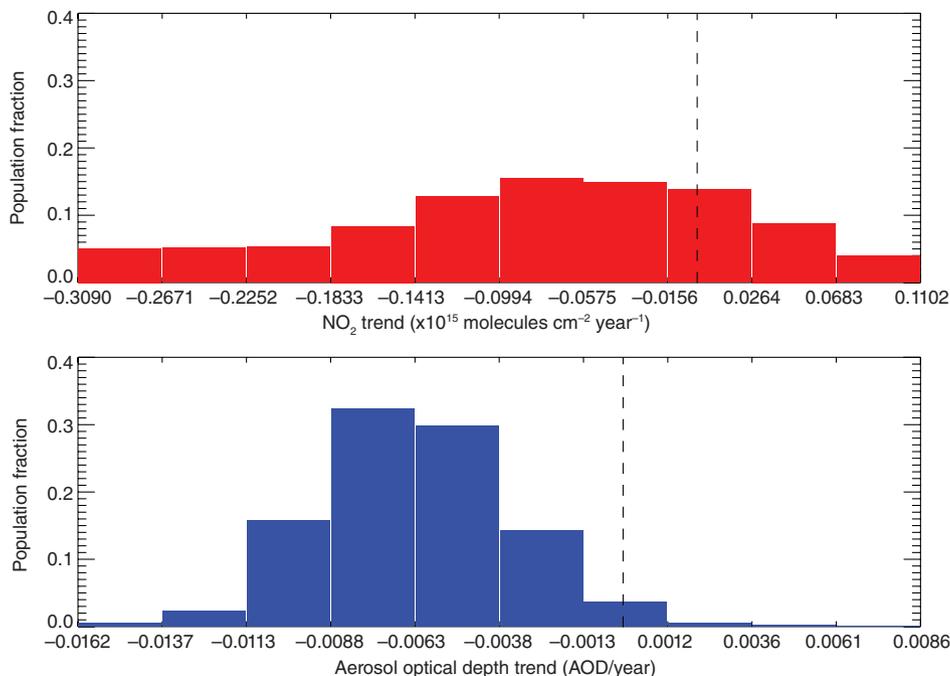


FIGURE 3 Same as Figure 2, but for MODIS AOD and Yorkshire/Humberside (1.00–0.00°W, 53.00–54.00°N) instead of Drax

Significant negative pollutant trends in the major cities between 2005 and 2015 suggest some success for efforts to reduce emissions (e.g., the Air Quality Standards Regulations 2010; DEFRA, 2011) to improve urban air quality. The National Atmospheric Emissions Inventory (NAEI) demonstrates decreases in total UK  $\text{NO}_x$  emissions from 1,608 kt (2005) to 918 kt (2015) over the period analysed (National Atmospheric Emissions Inventory, 2017). Despite this, urban  $\text{NO}_2$  concentrations remain in breach of levels permitted by European Union Air Quality Directives in most UK regions (DEFRA, 2011), and the United Kingdom has received a final warning to address unsafe levels of  $\text{NO}_2$  or face legal action from the European Commission (2017). Though UK  $\text{NO}_x$  emissions have decreased, long-range transport of pollution from continental Europe, as shown by Pope *et al.* (2014), will contribute significantly to the total  $\text{NO}_2$  concentrations, especially in urban regions. The NAEI  $\text{NO}_x$  emissions also do not account for the under-representation of diesel car sources (Jonson *et al.*, 2017), which potentially weakens the reported decreasing NAEI UK  $\text{NO}_x$  emissions trend.

$\text{NO}_2$  has a relatively short lifetime compared with aerosol (Seinfeld and Pandis, 2006), so changes in  $\text{NO}_2$  are more concentrated over populated regions (anthropogenic emission sources). Primary aerosol, due to its longer life-time (several weeks), is more susceptible to long-range transport from source regions yielding a more homogeneous spatial distribution than that of  $\text{NO}_2$ . Therefore, changes in source region aerosol emissions will result in satellite observed AOD changes over a wider region with a lower average population density. This is highlighted in Figure 4 where the population fraction, subject to different changes in air pollution, is distributed against these trends represented by equally spaced bins during the 2005–2015 period. Year 2010 population data from the Socioeconomic Data and Applications Centre (SEDAC) is used. Ten equally spaced bins are used for both pollutants ranging between the trend's 0.5th and 99.5th percentiles. Approximately, 90% of the population have benefited from negative AOD trends (bottom panel) between  $-0.0113/\text{year}$  and  $-0.0013/\text{year}$  (i.e., distributed across four bins with peak population exposure of 32%). However, the  $\text{TCNO}_2$  trends (top panel) are distributed more evenly with 15% peak



**FIGURE 4** The UK population fraction, subject to changes in air pollution, distributed against OMI TCNO<sub>2</sub> ( $\times 10^{15}$  molecules  $\text{cm}^{-2}$  year<sup>-1</sup>) and MODIS AOD (AOD/year) trends, between 2005 and 2015, represented by 10 evenly spaced bins. The summation of all the pollutant bins results in a population fraction of 1.0. The x-axis bins are evenly spaced, but rounding of the x-axis labels results in slightly different size bins. The black dashed vertical lines show the location of the 0.0 trend line

population exposure for the  $-0.0994$  to  $-0.0575 \times 10^{15}$  molecules  $\text{cm}^{-2}$  year<sup>-1</sup> bin. Approximately, 10% and 2.5% of the total population have benefited from the largest TCNO<sub>2</sub> and AOD negative trends, respectively.

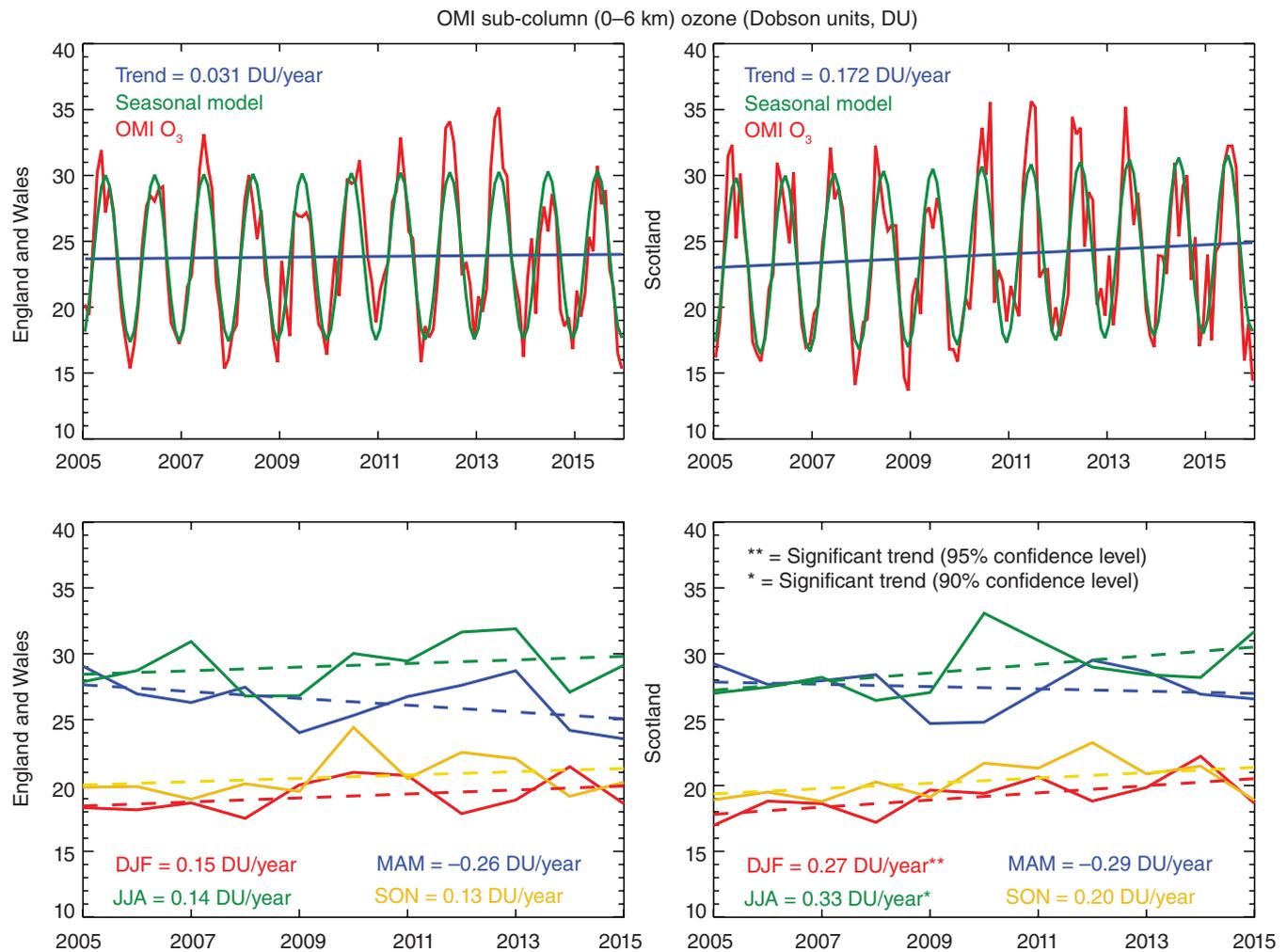
We detect a negative TCNO<sub>2</sub> trend from Drax, linked to several factors including the conversion of several of Drax's stacks from coal to biomass burning (Drax Power, 2016), the reduced activity of Eggborough and Ferrybridge C (closed in 2016) and the Large Combustion Plants Regulations 2007 (Large Combustion Plants, 2007), which resulted in a large NO<sub>x</sub> emissions drop between 2007 and 2008. There is no noticeable step change in OMI TCNO<sub>2</sub> between 2007 and 2008 related to the Large Combustion Plants regulations, but other processes such as meteorological variability could be masking this sharp emissions drop. Instead, there is a steady decrease in TCNO<sub>2</sub> throughout the remaining time-series. Figure 1f highlights a larger AOD hotspot over Yorkshire/Humberside, potentially linked to the accumulation of pollution in the Vale of York, a low lying region flanked by the uplands of the Pennines on the west and the North York Moors to the north. A negative trend here may be explained by reduced in situ aerosol/precursor gas emissions and transport of cleaner air masses accumulating in the region.

As the spatial resolution of SCO<sub>3</sub> is coarse relative to the TCNO<sub>2</sub> and AOD, we focus on regional average trends (Figure 5a,b) over England/Wales and Scotland. England/Wales ( $0.031 \pm 0.115$  DU, Dobson units) and Scotland ( $0.172 \pm 0.089$  DU) have insignificant and significant positive trends at the 95% confidence level, respectively. Seasonal trends (Figure 5c,d) also show increasing trends in SCO<sub>3</sub> in these regions, except for spring (negative trends). Chang *et al.* (2017) found several surface sites in Scotland and northern England had strong increasing summer-time (April–September, 2000–2014) daily average O<sub>3</sub> trends, but England

typically had weaker insignificant trends. Karlsson *et al.* (2017) highlighted significant increasing winter-time (1990–2014) surface O<sub>3</sub> trends in northern Europe, but were insignificant in summer. Ordóñez *et al.* (2007) associated increases in background tropospheric O<sub>3</sub>, using models and ozonesondes, with downwelling of lower stratospheric O<sub>3</sub>, especially in winter and spring. The England/Wales and Scotland December–January–February (DJF) trends support this where there is an insignificant and significant (95% confidence level) increase in SCO<sub>3</sub>, respectively. However, the spring (March–April–May, MAM) trends show insignificant negative trends (potentially linked to different conflicting processes and large meteorological inter-annual variability). The England/Wales DJF trend is possibly insignificant as the region is more polluted (Figure 1d) with larger emissions of nitric oxide (NO), which can act as a sink of O<sub>3</sub> weakening the winter-time trend. Over Scotland, there is also an increasing June–July–August (JJA) trend (90% confidence level) which supports the results of Chang *et al.* (2017). Both of DJF and JJA seasons appear to be driving the positive annual SCO<sub>3</sub> trend over Scotland, despite the large SCO<sub>3</sub> inter-annual variability. The investigation of upper tropospheric O<sub>3</sub> trends (450–100 hPa) in the OMI data set highlighted increases in tropospheric O<sub>3</sub> over the whole UK, but these annual trends are not significant (not shown).

## 4 | CONCLUSIONS

By using high resolution data sets of satellite-observed air quality, this study has identified recent (2005–2015) widespread changes in UK tropospheric column NO<sub>2</sub> (TCNO<sub>2</sub>), sub-column ozone (SCO<sub>3</sub>) and aerosol optical depth (AOD). Over the UK pollution hotspots (e.g., London), there have been significant decreases in TCNO<sub>2</sub> and AOD resulting from



**FIGURE 5** Same as Figure 2, but for OMI  $\text{SCO}_3$  (DU) for England/Wales ( $4.50^\circ\text{W}$ – $1.00^\circ\text{E}$ ,  $50.50$ – $55.00^\circ\text{N}$ ) and Scotland ( $6.00$ – $1.50^\circ\text{W}$ ,  $55.5$ – $58.00^\circ\text{N}$ ). The seasonal trends (dashed lines) between 2005 and 2015 for December–January–February (DJF, red), March–April–May (MAM, blue), June–July–August (JJA, green) and September–October–November (SON, yellow) have been calculated for England/Wales and Scotland in the bottom two panels. \* and \*\* represent significant trends at the 90% and 95% confidence levels. These trends are calculated using a linear version (i.e., no seasonal term) of Equation 1

reduced emissions linked to government policy (e.g., the Air Quality Standards Regulations 2010 and the Large Combustion Plants Regulations 2007). When pollution trends, both significant and non-significant, across the United Kingdom are weighted by population fraction exposed, it suggests that the general public are benefiting more from larger changes in  $\text{NO}_2$  than aerosols. The lifetime of  $\text{NO}_2$  is shorter than that of aerosol, so any changes in emissions (i.e., anthropogenic activities) are more strongly co-located with populated regions. Finally, trends in  $\text{SCO}_3$  are significant over Scotland, a more remote and cleaner region of the United Kingdom, which are consistent with other studies showing increasing background tropospheric  $\text{O}_3$  over north-western Europe.

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and NASA's MODIS AOD (Aerosol 5-min L2 Swath 10km V006) data (<https://reverb.echo.nasa.gov/reverb>). Population data comes from the Socioeconomic Data and Applications Centre (SEDAC: <http://sedac.ciesin.columbia.edu/data/collection/gpw-v4>). This work was supported by the UK Natural Environment Research Council (NERC) by providing funding for the National Centre for Earth Observation (NCEO).

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## SUPPORTING INFORMATION

Additional Supporting Information may be found online in the supporting information tab for this article.

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