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To cite this article: Daniel A Potts *et al* 2021 *Environ. Res. Lett.* **16** 054031

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ENVIRONMENTAL RESEARCH
LETTERS

LETTER

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RECEIVED

6 November 2020

REVISED

11 January 2021

ACCEPTED FOR PUBLICATION

21 January 2021

PUBLISHED

29 April 2021

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Keywords: air quality, COVID-19, satellite NO₂, atmospheric chemistry modelling

Supplementary material for this article is available [online](#)

Abstract

The dramatic and sudden reduction in anthropogenic activity due to lockdown measures in the UK in response to the COVID-19 outbreak has resulted in a concerted effort to estimate local and regional changes in air quality, though changes in underlying emissions remain uncertain. Here we combine satellite observations of tropospheric NO₂ from TROPOMI and the Goddard Earth Observing System (GEOS)-Chem 3D chemical transport model to estimate that NO_x emissions declined nationwide by ~20% during the lockdown (23 March to 31 May 2020). Regionally, these range from 22% to 23% in the western portion of the country to 29% in the southeast and Manchester, and >40% in London. We apply a uniform 20% lockdown period emission reduction to GEOS-Chem anthropogenic emissions over the UK to determine that decline in lockdown emissions led to a national decline in PM_{2.5} of 1.1 μg m⁻³, ranging from 0.6 μg m⁻³ in Scotland to 2 μg m⁻³ in the southwest. The decline in emissions in cities (>40%) is greater than the national average and causes an increase in ozone of ~2 ppbv in London and Manchester. The change in ozone and PM_{2.5} concentrations due to emission reductions alone is about half the total change from 2019 to 2020. This emphasizes the need to account for emissions and other factors, in particular meteorology, in future air pollution abatement strategies and regulatory action.

1. Introduction

The first cases of COVID-19 were reported in December 2019 in Wuhan, China, the initial epicentre of the outbreak, and rapidly spread to the US and Europe. By mid-March, all European countries had confirmed cases, leading to the implementation of travel restrictions and local, regional, or national lockdown measures. The UK government imposed strict lockdown measures on 23 March 2020 in response to model simulations that showed that the rate of spread of the virus would overwhelm the healthcare system [1].

The movement restrictions imposed by the UK and other governments led to an unprecedented

decline in road traffic and slowdown or shutdown of intensive industries. The associated reduction in air pollutant emissions has led to a flurry of research to determine the short-term response in air quality and atmospheric composition. A contemporary record of these is maintained by the Health Effects Institute (<https://tinyurl.com/y3xyty6x>). The general pattern that emerges from these studies comparing air pollutant concentrations before and after lockdown measures is widespread regional decline in nitrogen oxide (NO_x ≡ NO + NO₂) concentrations. Also observed is the anticipated increase in ozone due to reduced titration of ozone by NO_x in cities where this titration is prevalent [2–12]. Studies that use models to interpret

air quality observations have also diagnosed the contribution of meteorology to the observed changes in atmospheric composition and air quality [13], estimated the influence of emission changes on the oxidizing capacity of the atmosphere [5], and quantified regional decline in emissions due to lockdown measures [14]. Across China, for example, NO₂ concentrations declined by 40%–60% [2–4], whereas PM_{2.5} (particles with aerodynamic diameter < 2.5 μm) increased in heavily populated and very polluted northern China. This was due to a combination of stagnant conditions, unusually high humidity promoting heterogeneous chemistry, sustained industrial emissions, and an increase in the oxidizing capacity of the atmosphere [5]. In India, an increase in fire activity in 2020 masked the anticipated decline in NO_x due to lockdown measures in cities close to these fires [15].

In the UK, preliminary reports on changes in air quality were commissioned by the national Department for the Environment, Food and Rural Affairs. These mostly compared surface network observations in 2020 before and during the lockdown. Reported changes in air pollutants included an average 21% decline in NO₂ from roadside reference network monitors [16, 17], though this percent decline includes ordinary seasonal decrease in NO_x due to changes in photochemistry during the shift from winter to spring [18]. A surprising change in air quality in London was an observed increase in PM_{2.5}, despite the decline in NO_x, that was attributed to long-range transport of PM_{2.5}-laden air from mainland Europe [16]. The impact of the lockdown on air quality throughout the UK was later diagnosed by Lee *et al* [6] using surface data from 126 *in situ* measurement sites distributed across the UK. They found from comparison of observations averaged over 23 March to 31 May that surface NO₂ was 42% less and surface ozone 11% more in 2020 than the mean of the 5 preceding years. They speculated that routine exceedances of NO₂ air quality limits in London would halve in 2020 relative to 2019 as a result of the lockdown. They also identified a marked increase in ozone exceedances during the lockdown, but suggested that the influence of this on the rest of 2020 depends on emissions and meteorological conditions in summer when surface ozone concentrations in the UK peak [6].

Changes in air quality by comparison of the lockdown period to previous weeks or years will be impacted by interannual variability due to extreme weather or shifts in transport patterns, seasonality in photochemistry affecting formation and persistence of air pollution, and ongoing decline in emissions due to air quality policies. As a result, it is challenging to disentangle the contribution of changes in emissions and other factors to changes in air quality with observations alone. A more suitable approach is to use an explicit and detailed atmospheric chemical transport

model (CTM) [10, 13, 14, 19] or to account for confounding effects of weather using appropriate statistical methods [7, 11, 13, 20]. The UK network of reference monitors, while relatively extensive compared to most countries, has large monitoring gaps, so may not be appropriate for diagnosing national and regional changes in air quality.

Here we use satellite observations of tropospheric column NO₂ from the TROPospheric Monitoring Instrument (TROPOMI) to quantify national, regional and large city (London, Manchester) changes in NO₂ in the UK due to the lockdown. Analysis of the satellite observations is supported by observations of surface concentrations of NO₂ from the air quality networks in London and Manchester and interpreted with targeted Goddard Earth Observing System (GEOS)-Chem CTM simulations. The model is used to account for non-linearities in atmospheric chemistry driving changes in air quality [21], relate changes in the tropospheric column to changes in emissions at the surface, discern the role of meteorology and emission changes, and quantify the effect of lockdown measures on regional and national surface concentrations of the criteria pollutants ozone and PM_{2.5} that adversely affect our health and the environment.

2. Data and methods

2.1. Space-based TROPOMI tropospheric column observations of NO₂

We use tropospheric NO₂ column densities from TROPOMI onboard the Sentinel-5 Precursor (S5P) satellite [22, 23]. S5P was launched on 13 October 2017 and has an overpass time of 13:30 local solar time (LST). TROPOMI achieves daily global coverage with a swath width of 2600 km and a ground pixel resolution of 7.2 km × 3.5 km at nadir (along track × across track) until 5 August 2019, refined thereafter to 5.6 km × 3.5 km [24]. We use TROPOMI NO₂ over the UK from the offline Level 2 product (version 01–03–02) from the S5P Pre-Operations Data Hub (<https://s5phub.copernicus.eu/dhus/>; last accessed 20 July 2020) for 1 January to 30 June 2019 and 2020. TROPOMI data are filtered to remove poor quality retrievals with a quality flag ('qa_value' in the data file) >0.5 and cloudy scenes identified with a TROPOMI cloud radiance fraction ≥0.5, as suggested by Eskes *et al* [25] for comparison of TROPOMI to models. TROPOMI validation studies generally find that TROPOMI overestimates NO₂ at rural and remote locations and underestimates NO₂ over polluted scenes from comparison to ground-based total and tropospheric column measurements [26–30].

2.2. Surface air quality measurements

Surface hourly *in situ* observations that we use to assess temporal variability of TROPOMI NO₂ in Manchester and London and the ability of

the model to reproduce changes in surface concentrations of ozone and PM_{2.5} across the UK are from the Automatic Urban and Rural Network (AURN). All data are downloaded from the UK-AIR data portal (<https://uk-air.defra.gov.uk/networks/network-info?view=aurn>; accessed 11 August 2020 for NO₂ and 25 September 2020 for ozone and PM_{2.5}). We use NO₂ data from eight sites in London and three sites in Manchester. City-wide average NO₂ is estimated for each city from observations obtained during the satellite overpass (13:00–14:00 LST). Ozone data are available from 71 monitors in both 2019 and 2020 and 80 monitors in 2019 and 81 in 2020 for PM_{2.5}. These are used to calculate 24-hour averages at each site to compare to coincident GEOS-Chem data.

2.3. The GEOS-Chem CTM

We use GEOS-Chem version 12.1.0 (<https://doi.org/10.5281/zenodo.1553349>) nested over Europe (32.75–61.25° N, 15° W–40° E) at 0.25° (~25 km latitude) × 0.3125° (~31 km longitude) horizontal resolution and extending from the Earth's surface to 0.01 hPa. Dynamic boundary conditions are from a global simulation at 4° × 5°. The model is driven with NASA Global Modelling and Assimilation Office GEOS—Forward Processing assimilated meteorology. The model also includes detailed gas- and aerosol-phase chemistry and physical loss processes (wet and dry deposition). Emission inventories are updated in this work to include anthropogenic emissions over the UK and mainland Europe from the gridded European Monitoring and Evaluation Programme inventory for 2016 scaled to the simulation year of interest using reported rates of annual emission reductions [31]. The model also includes natural emissions from vegetation, soils, seabirds, lightning and volcanoes.

The model is simulated in 2019 as a reference year in terms of anthropogenic emissions and in 2020 using five emission scenarios. These include business-as-usual (BAU) using normal annual decline in anthropogenic emissions due to air quality policies, and lockdown emission reduction scenarios that, in addition to the normal annual decline in emissions, include anthropogenic emission reductions of NO_x, SO₂, non-methane volatile organic compounds, and primary PM_{2.5} in the UK and mainland Europe. We refer to these throughout as GC_BAU for the BAU scenario, GC_x for *x*% emission reduction, where *x* is 15, 30, 45, and 60. In the GC_x simulations, emissions reductions are applied to mainland Europe on 15 March and the UK on 23 March to represent the earlier lockdown measures across mainland Europe. Sectors that are reduced include industry, transport, and other residential and commercial activity. The instantaneous reduction in emissions in the model is approximate, as some countries gradually transitioned from local to national lockdown

measures (such as in Italy). There was also a range in the timing of national lockdowns (17 March for France to 23 March for Germany) and severity of measures. The Netherlands and Greece, for example, did not impose a national lockdown [32]. The model is sampled from 1 February to 30 June for the reference year (2019) and the GC_BAU emission scenario. The GC_x simulations are sampled from 15 March to 30 June 2020.

3. Results and discussion

3.1. Changes in column densities of NO₂ in the UK

Figure 1 shows TROPOMI monthly mean tropospheric NO₂ columns over the UK for 2019 and 2020. The decline in TROPOMI tropospheric column NO₂ from January to June is expected, due to seasonal variability in photochemistry leading to a longer lifetime and greater abundance of NO_x in winter than in spring and summer. The location of cities with large traffic emissions of NO_x, such as London in the south-east and Manchester in the north-west, are particularly apparent in winter 2019.

TROPOMI NO₂ over the UK is in general lower in 2020 than 2019, even for the months preceding the lockdown. Lower concentrations of NO₂ in January and February for 2020 compared to the mean of the five preceding years has also been observed for surface NO₂ and attributed to unusually high wind speeds and frequent storms in 2020 [6]. In April, the peak of the lockdown period, the relative decrease from 2019 to 2020 is 36 ± 13% (median ± standard error) for the SE, 31 ± 11% for the SW, 19 ± 11% for the NE, 38 ± 11% for the NW, 31 ± 11% for SC, 40 ± 12% for London, and 31 ± 14% for Manchester. In May, when there was a gradual transition to more lenient lockdown measures in the UK, there is generally a smaller reduction in TROPOMI NO₂ in 2020 relative to 2019 (29 ± 15% for the NW, 17 ± 11% for SC, and 31 ± 17% for London). The SE (36 ± 9%) and Manchester (34 ± 14%) show greater decline in May than April. The decline in the SW in May is not significant (20 ± 19%) and there is a non-significant increase in the NE of 9 ± 18%.

Figure 2 assesses the skill of TROPOMI at reproducing day-to-day variability in surface NO₂ in London and Manchester which have been used in the study by Lee *et al* [6]. City-average daily mean surface observations are compared to TROPOMI daily mean tropospheric columns for TROPOMI pixel centres within the city domains shown in figure 1. The range in surface NO₂ concentrations is similar for the two cities (5–75 μg m⁻³), declining from winter to spring, as is expected from seasonal changes in photochemistry. There is also large day-to-day variability related to weather and weekend–weekday variability in traffic activity [34, 35], though midday TROPOMI and surface observations will be less sensitive to this effect than observations during rush hour [36]. TROPOMI

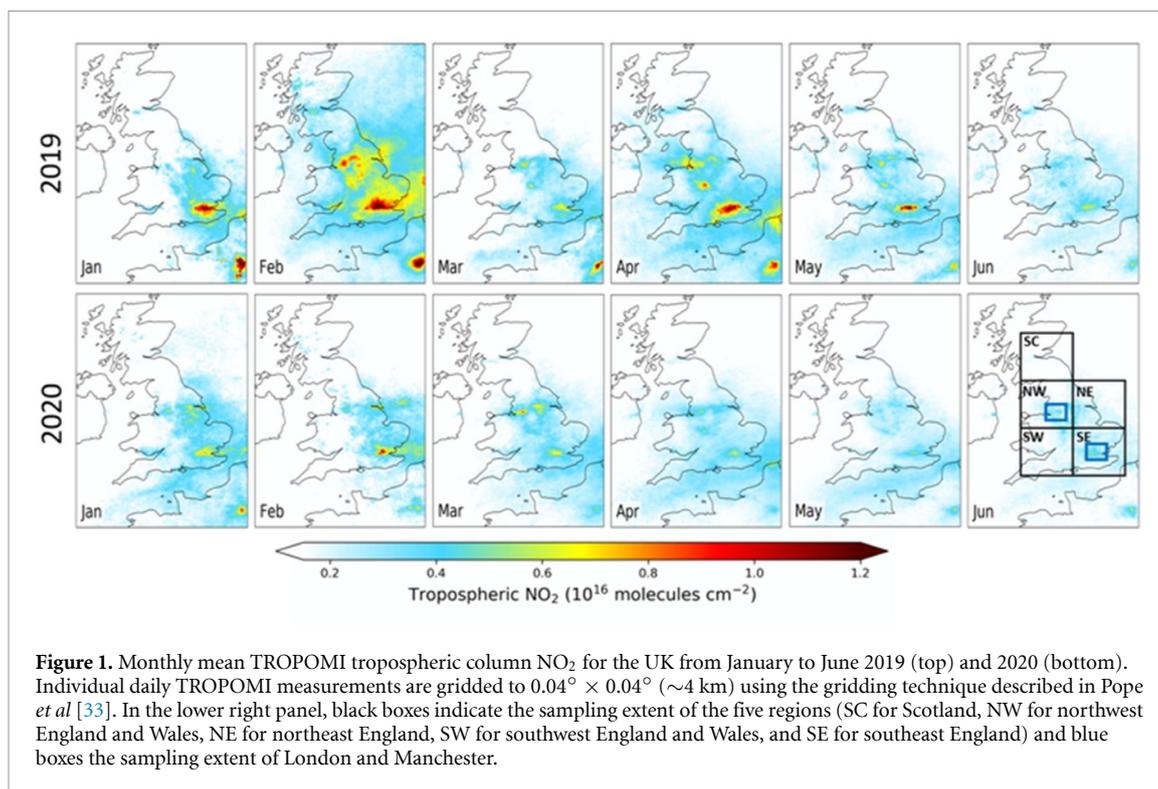


Figure 1. Monthly mean TROPOMI tropospheric column NO_2 for the UK from January to June 2019 (top) and 2020 (bottom). Individual daily TROPOMI measurements are gridded to $0.04^\circ \times 0.04^\circ$ (~ 4 km) using the gridding technique described in Pope *et al* [33]. In the lower right panel, black boxes indicate the sampling extent of the five regions (SC for Scotland, NW for northwest England and Wales, NE for northeast England, SW for southwest England and Wales, and SE for southeast England) and blue boxes the sampling extent of London and Manchester.

NO_2 are sparse in winter due to persistent clouds. A relatively shallow planetary boundary layer in winter also leads to reduced sensitivity to surface NO_2 and degrades temporal consistency between the column and surface [37]. The surface *in situ* observations are also more prone to positive interference in winter from thermal decomposition of reservoir compounds to NO_2 [38]. Despite these issues, the temporal correlation between the surface and satellite observations is reasonably strong. For the full record shown in figure 2, the Pearson's correlation coefficient, R , is 0.78 for London and 0.51 for Manchester using a 75% spatial coverage threshold for TROPOMI. Similarly, during just the lockdown period and again using a 75% spatial coverage threshold, R is 0.71 for London and 0.70 for Manchester. The increase in R for Manchester during the lockdown period is because the pre-lockdown period has fewer coincident observations. The small decrease in R for London may be due to less dynamic variability in NO_2 during the lockdown period.

3.2. Interpretation of observed changes in NO_2 with GEOS-Chem

We use GEOS-Chem to determine the contribution of anthropogenic emissions changes associated with lockdown measures to the observed changes in TROPOMI NO_2 from 2019 to 2020. We do this by identifying the emission scenario in 2020 that yields results consistent with regression of GEOS-Chem versus TROPOMI tropospheric NO_2 columns in 2019. This approach is conceptually similar to the standard approach of estimating emissions by

applying the modelled ratios of tropospheric column NO_2 to surface emissions of NO_x to TROPOMI tropospheric NO_2 columns to derive NO_x emissions [39] for two different time periods and estimating the relative change in satellite-derived emissions. Our regression approach eliminates unnecessary steps in the calculation and also minimizes influence of systematic biases in the model and observations. In all comparisons of the model to TROPOMI we apply the TROPOMI averaging kernels to the model to ensure the comparison is independent of the TROPOMI *a priori* [25, 40, 41]. City and regional averages for the domains shown in figure 1 are obtained using the Level 2 pixels and coincident GEOS-Chem grids. The model is sampled during the satellite overpass (12:00–15:00 LST), as is standard. The difference in modelled fields obtained for a narrower sampling range (13:00–14:00 LST) is negligible.

Figures 3 and 4 compare temporal variability in daily mean TROPOMI and GEOS-Chem regional (SE, figure 3) and city (London, figure 4) tropospheric columns. Only the results for the GC_BAU, GC_30, and GC_60 scenarios are shown, as the response of the model to emission changes is approximately linear. Comparisons for the other regions and Manchester for all scenarios are in supplementary figures S1–S5 (available online at stacks.iop.org/ERL/16/054031/mmedia). We use 2019 to quantify how the model compares to the satellite observations during a normal year. This accounts for systematic biases in the modelled tropospheric NO_2 column that may result from model representation of emissions, chemistry, and loss pathways,

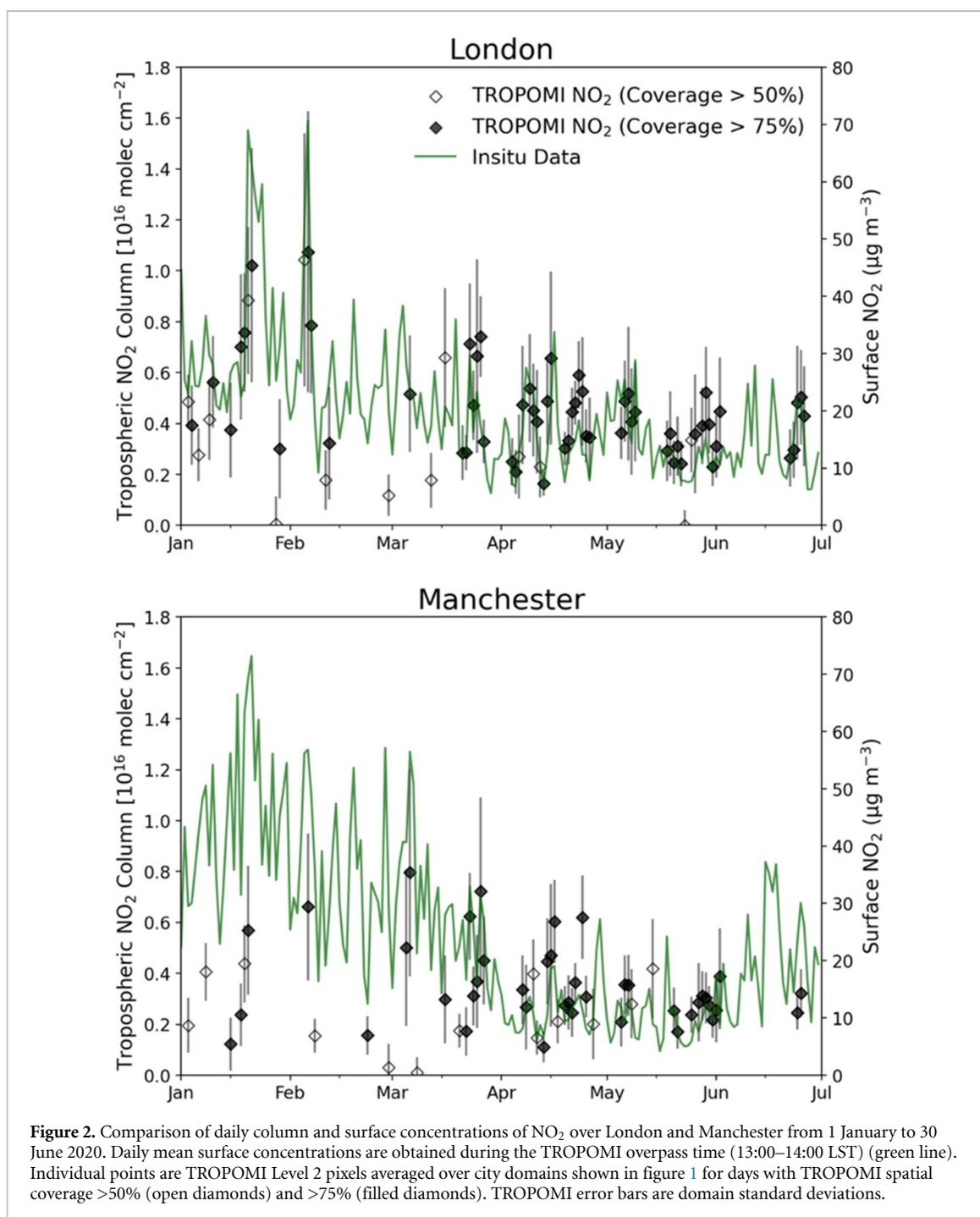
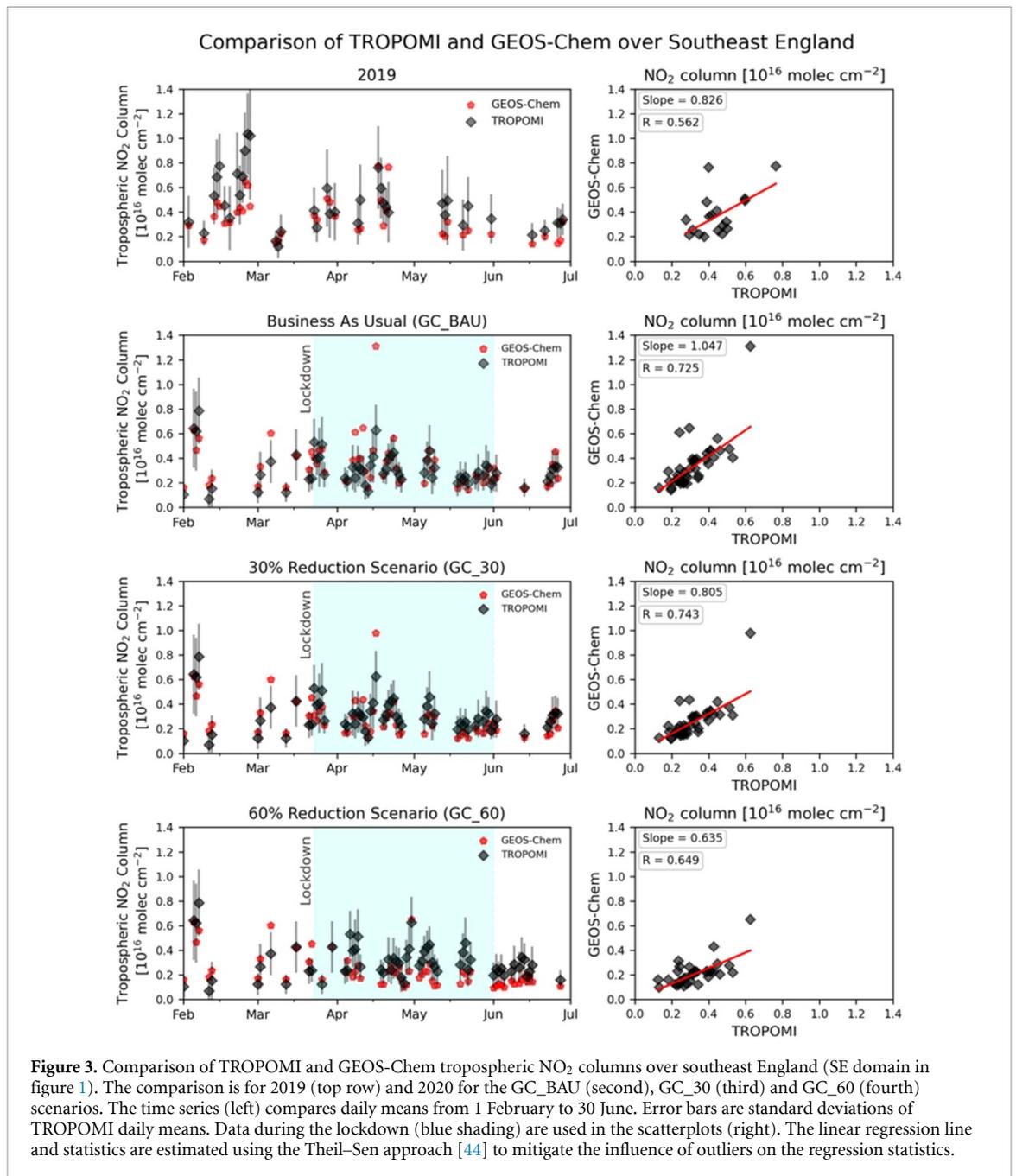


Figure 2. Comparison of daily column and surface concentrations of NO_2 over London and Manchester from 1 January to 30 June 2020. Daily mean surface concentrations are obtained during the TROPOMI overpass time (13:00–14:00 LST) (green line). Individual points are TROPOMI Level 2 pixels averaged over city domains shown in figure 1 for days with TROPOMI spatial coverage $>50\%$ (open diamonds) and $>75\%$ (filled diamonds). TROPOMI error bars are domain standard deviations.

and also systematic biases in TROPOMI [26–30]. To do this, we regress TROPOMI columns against co-located GEOS-Chem columns for 2019. We find that the model underestimates variance in tropospheric column NO_2 . Regression slopes for 2019 are less than unity in all regions (0.70–0.94) and cities (0.70 for London, 0.84 for Manchester). This may be due to a model underestimate in free tropospheric NO_2 that will be pronounced in comparison to TROPOMI due to its enhanced vertical sensitivity to NO_2 in the free troposphere [42, 43]. We would then expect that the 2020 model scenario that most closely matches air pollutant emissions during the lockdown is the one

that yields a slope similar to that obtained with the 2019 data.

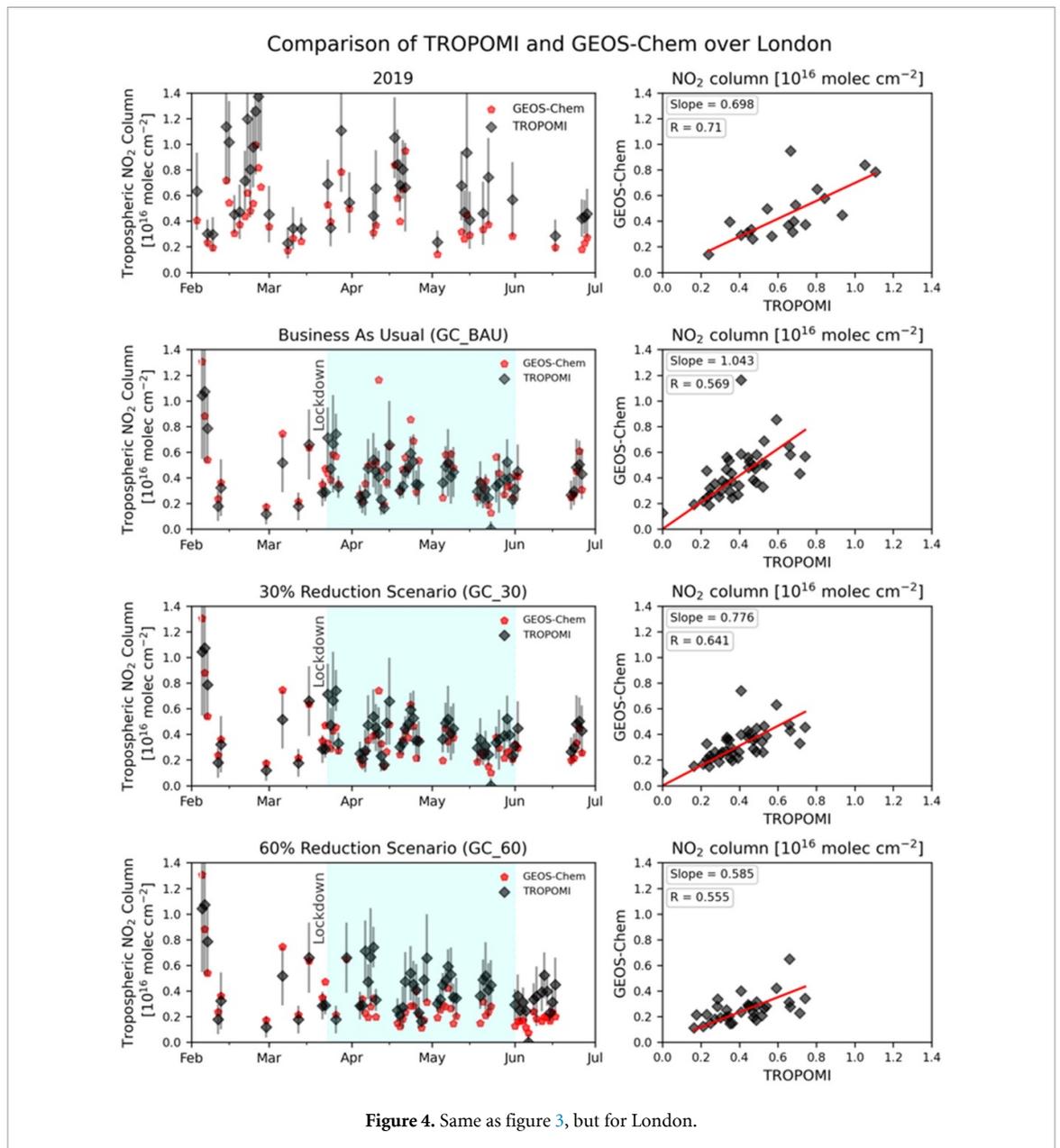
The slopes in figures 3 and 4 and S1–S5 for the GC_BAU scenario are 21%–49% larger than the reference year (2019) slopes everywhere except SC and the NE. In SC, GC_BAU is 7% less than 2019 and in the NE GC_BAU is consistent with ($<1\%$ difference) that for 2019, suggesting either no appreciable change in emissions during the lockdown or limited regional sensitivity of tropospheric columns to changes in surface emissions. Regional GEOS-Chem NO_x emissions in SC in 23 March to 31 May 2019 are relatively low. NO_x emissions in SC are 10% of



national total emissions. This is similar to the contribution from just the London domain. NO_x emissions are also limited to a few large cities such as Edinburgh and Glasgow (figure 1). Tropospheric columns of NO₂ over SC will be more influenced by factors such as long-range transport and meteorology than perturbations in local sources. NO_x emissions in the NE are 20% of the national total, but this includes a large contribution from industrial activity and power generation. According to the UK Office of National Statistics, activity from these decreased less dramatically (20% decline in April 2020 relative to April 2019) than road traffic. Road traffic in April 2020 was 35% of typical conditions for April and 50% of typical conditions in May [45, 46]. By interpolation of slopes for the GEOS-Chem 2020 emission scenarios,

we estimate that emission reductions that would yield a slope that best matches the slope for 2019 include 42% for London, 29% for Manchester and the SE, 23% for the NW, and 22% for the SW. There is no detectable change for the NE and SC.

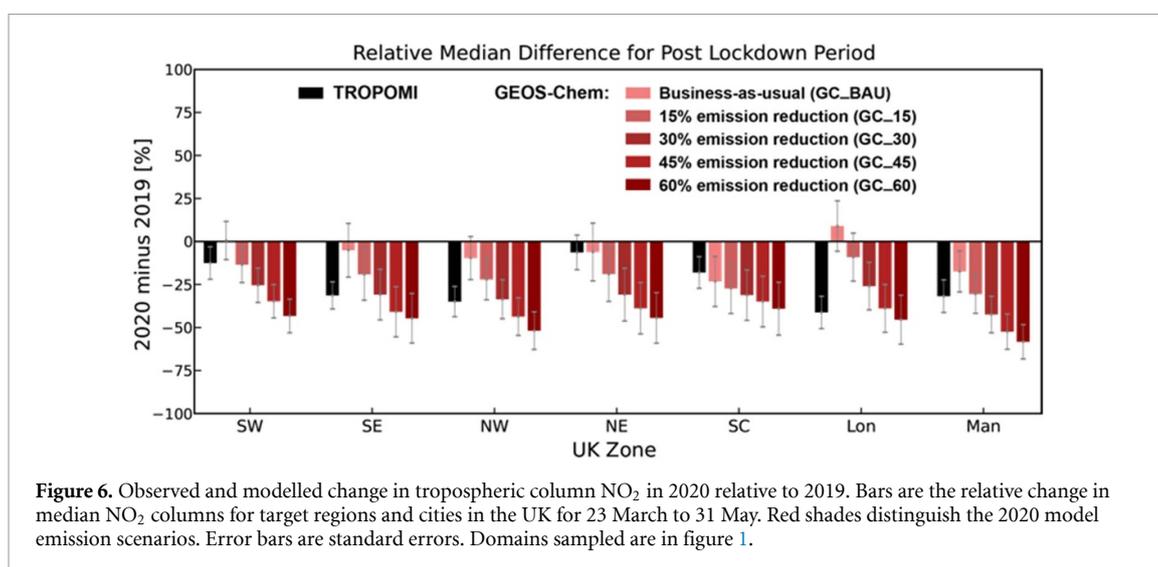
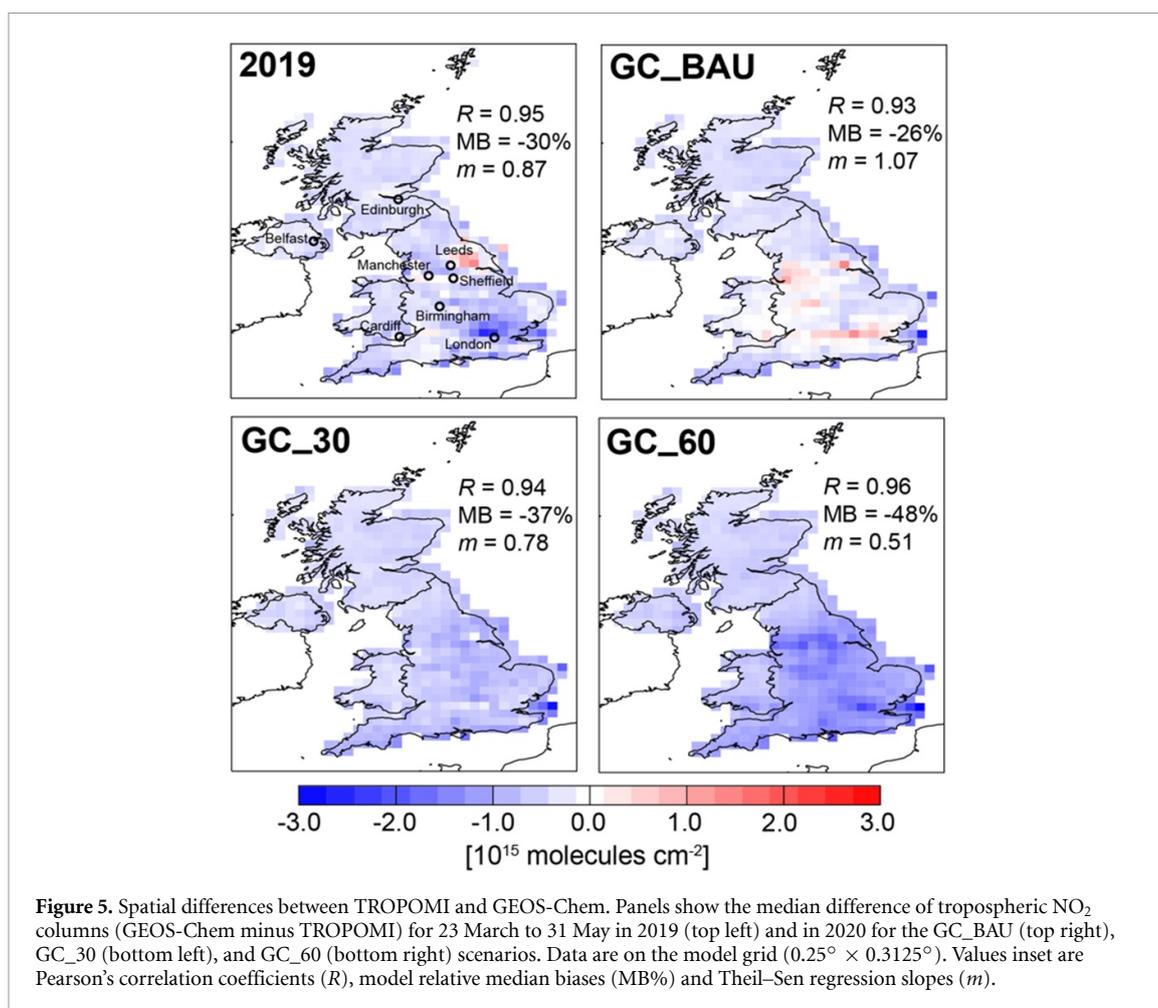
Figure 5 compares the spatial distribution of TROPOMI and GEOS-Chem tropospheric NO₂ columns at the national level for 2019 and the GC_BAU, GC_30, and GC_60 emission scenarios. TROPOMI and GEOS-Chem are spatially consistent in 2019 ($R = 0.95$) and in 2020 for all emission scenarios ($R = 0.93$ – 0.96). Results for GC_15 and GC_45 also yield $R > 0.9$. Regression slopes are $m = 0.92$ for GC_15 and $m = 0.63$ for GC_45. By interpolation, the nationwide decrease in anthropogenic NO_x emissions is $\sim 20\%$.



The bar chart in figure 6 shows the relative change in city and regional median TROPOMI and GEOS-Chem tropospheric NO_2 columns from 2019 to 2020 as an alternate approach to the regression method to identify the scale of emission reductions required to reproduce the observations. The decline in TROPOMI NO_2 in 2020 relative to 2019 is 41% over London and 32% over Manchester. This is greater than the decline reported by Barré *et al* [20] for London (30%) and Manchester (27%), though within the margin of error for Manchester. They also used TROPOMI, but for a lockdown period that starts earlier than the UK lockdown (15 March) and so includes more days in March than us when 2019 and 2020 TROPOMI concentrations are relatively similar (figure 1). The regional decline we obtain with TROPOMI ranges from 6% over the NE to 35% over the

NW. The relative change in the model from 2020 to 2019 due to normal emission mitigation measures and meteorology (GC_BAU) ranges from +9% for London to -23% for SC. The emission reduction estimates obtained with the relative difference approach are 46% for London, 18% for Manchester, 15% for the SW, 32% for the SE, 33% for the NW, and BAU for the NE and SC. These are similar to the regression approach for London, the SE, the SW, the NE and SC, but less than the regression approach for Manchester and more than the regression approach for the NW, as the relative difference is more influenced by extreme values than the regression approach.

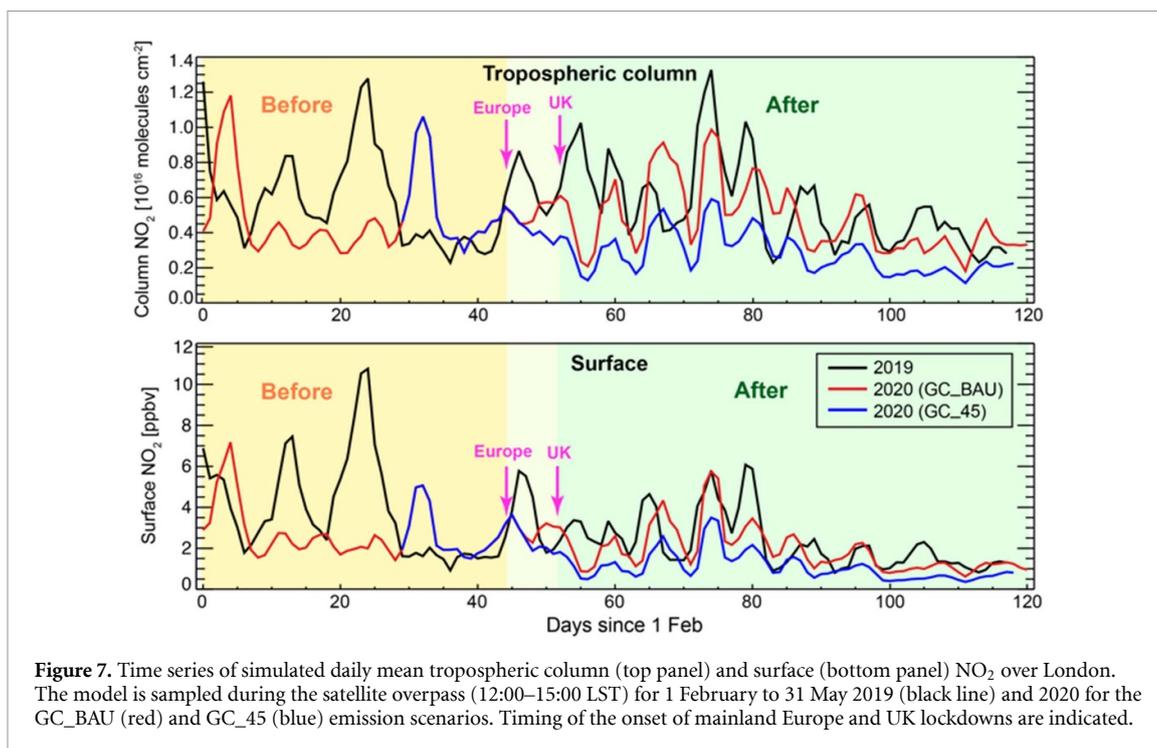
The size of the errors are relatively large in figure 6, so we use the two-sample *t*-test to determine whether the differences in TROPOMI and GEOS-Chem



tropospheric NO_2 columns for 2019 and 2020 are statistically significantly different. The t -scores and p -values from this analysis are in the supplementary (table S1). The model emission reduction scenarios for which the differences between GEOS-Chem and TROPOMI are inconsistent with those for 2019 have a t -score significantly different from zero (and p -value < 0.05). The t -test confirms a 30% reduction

for the NW and Manchester and no emissions reduction for the NE and SC, but yields larger reduction for London (GC_60). The results are ambiguous for the SW (GC_15 and GC_30) and the SE (GC_30 and GC_45).

Figure 7 shows the complete GEOS-Chem time series of tropospheric column and surface NO_2 for 1 February to 31 May in 2019 and 2020 for the GC_BAU



scenario and the emission scenario that best fits the observations over London according to the regression slope approach (GC_45). Time series for the SW, SE, NW and Manchester are in the supplementary (Figures S6–S9). Modelled columns and surface NO₂ before the lockdown in 2020 are much less than NO₂ in the previous year, as was observed for TROPOMI NO₂ (figure 1). According to the model, this is due to buildup of pollution in 2019 due to stagnant conditions, and less precipitation and colder temperatures in 2019 than 2020. This is consistent with the interpretation by Lee *et al* [6] that lower pre-lockdown surface concentrations of NO₂ in 2020 than 2019 were caused by dissipation of pollution by unusually high wind speeds and frequent storms in early 2020.

The change in NO₂ concentrations due to a 45% reduction in emissions in figure 7 during the lockdown (GC_45 minus GC_BAU for 23 March to 31 May 2020) is 42% at the surface and for the column, supporting sensitivity of the column to changes in local emissions. There is evidence of influence of air pollution from mainland Europe, as GC_BAU and GC_45 surface and column NO₂ diverge after the mainland Europe lockdown and before the UK lockdown. This influence is apparent in all regions (figures S6–S8).

3.3. Lockdown impact on surface ozone and PM_{2.5}

We now use the model to determine the contribution of emission reductions due to lockdown measures to changes in ozone and PM_{2.5}. We use results from a model simulation with a uniform 20% reduction in UK anthropogenic emissions, as this is representative of nationwide decline in NO_x emissions (figure 5).

We first assess the ability of the model to reproduce AURN ozone and PM_{2.5} in 2019 and the observed change in PM_{2.5} and ozone in 2020 relative to 2019. AURN PM_{2.5} are obtained at standard atmospheric pressure, 20 °C and 50% relative humidity [47], so GEOS-Chem PM_{2.5} is calculated for the same conditions. Additional details of the GEOS-Chem simulation of PM_{2.5} components and calculation of total PM_{2.5} is in the supporting information.

Figure 8 compares modelled and observed ozone and PM_{2.5} for 23 March to 31 May 2019 and 2020. The comparison of the difference in these is in the supplementary (figure S10). The relatively low ozone concentrations in 2019 (<30 ppbv) in the model and observations occur in and around cities due to titration by NO_x. Ozone increases to 32–35 ppbv in 2020. Average ozone for the observations and coincident model grid squares is similar in 2019 (31.7 ppbv for AURN, 31.5 ppbv for the model), increasing by 2.2 ppbv in the observations and 1.5 ppbv in the model in 2020 (34.3 ppbv for AURN, 32.9 ppbv for the model). The increase in modelled ozone is more consistent with AURN using a 45% emission reduction scenario (2.2 ± 1.9 ppbv), as the effect of NO_x on ozone is local to cities where NO_x emission reductions exceed 40%, according to our emission reduction estimates for London using TROPOMI and GEOS-Chem.

PM_{2.5} is less spatially heterogeneous than ozone, as its physical removal from the atmosphere (wet deposition) can take days to weeks, whereas persistence of ozone in cities is determined by its reaction with short-lived (seconds to minutes) NO. Modelled and observed PM_{2.5} are similar in 2019 ($13.5 \mu\text{g m}^{-3}$

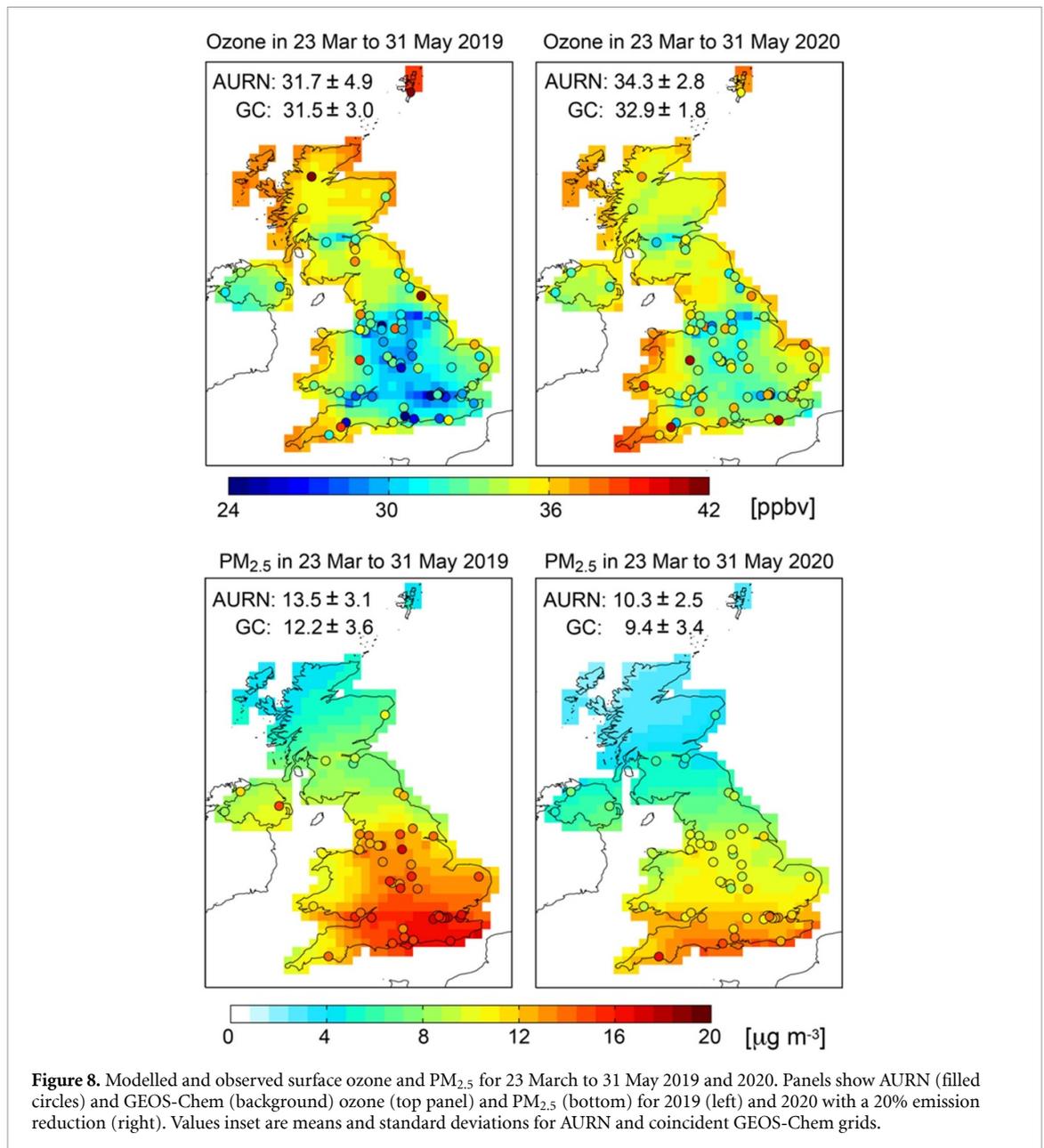
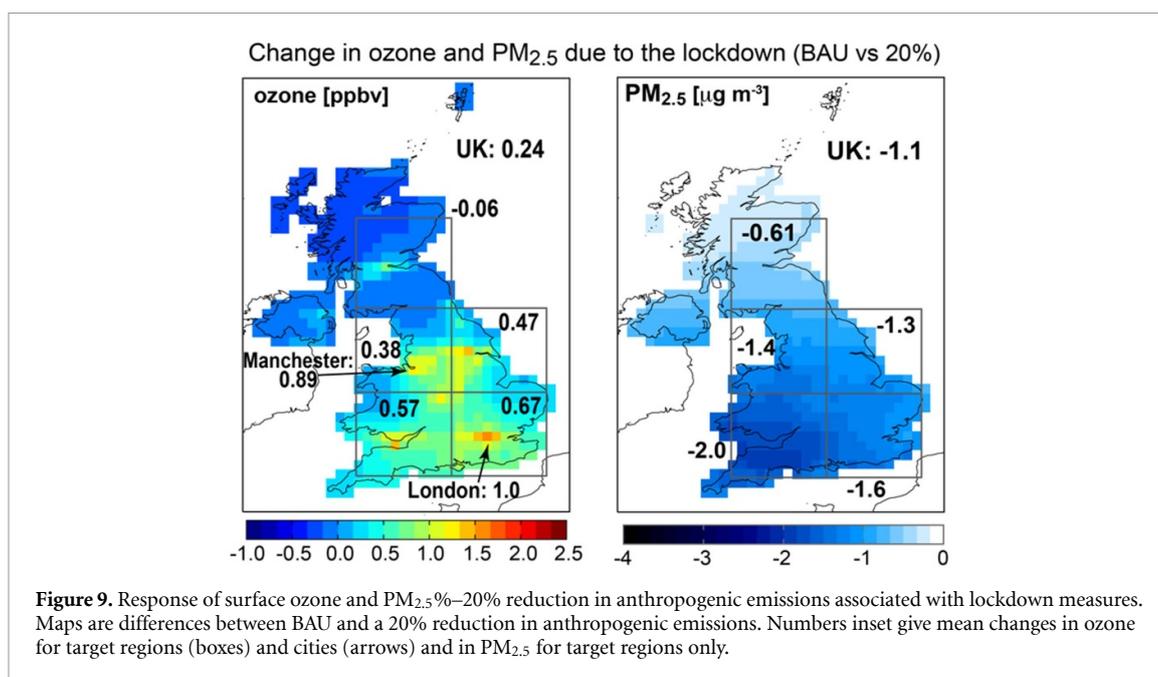


Figure 8. Modelled and observed surface ozone and PM_{2.5} for 23 March to 31 May 2019 and 2020. Panels show AURN (filled circles) and GEOS-Chem (background) ozone (top panel) and PM_{2.5} (bottom) for 2019 (left) and 2020 with a 20% emission reduction (right). Values inset are means and standard deviations for AURN and coincident GEOS-Chem grids.

from AURN, 12.2 $\mu\text{g m}^{-3}$ from the model) and in the decline in PM_{2.5} in 2020 relative to 2019 (3.2 $\mu\text{g m}^{-3}$ from AURN, 2.8 $\mu\text{g m}^{-3}$ from the model). According to AURN, PM_{2.5} declines at all except two sites in Cornwall where PM_{2.5} increases by 0.4 $\mu\text{g m}^{-3}$ and 2.1 $\mu\text{g m}^{-3}$ (figure S10). The model underestimates the observed 6–7 $\mu\text{g m}^{-3}$ decline in PM_{2.5} in cities, but captures the decline in rural and remote regions (figure S10). The aerosol nitrate component of PM_{2.5} is from oxidation of NO_x and the decline in emissions of these is constrained in this work with TROPOMI, whereas emissions of other precursors and primary PM_{2.5} are not. The spatial distribution of the modelled decline in nitrate and associated ammonium and aerosol water is consistent with decline in AURN PM_{2.5} in rural and remote areas (figure S11). The observed change in ozone and PM_{2.5} in cities is, in general, greater than in the model using 20% emission

reduction (figure S10), as the measurements are influenced by local effects not captured at the spatial resolution of the model and the regional analysis using TROPOMI does not capture local emission reductions in cities.

Figure 9 shows the effect of 20% emission reductions alone on ozone and PM_{2.5}. The national increase in ozone is small (0.24 ppbv) and ranges from increases in ozone over cities of 0.89 ppbv for Manchester and 1.0 ppbv for London to regional decline in ozone in remote and rural areas. The SE experiences the largest regional increase in ozone of 0.67 ppbv. A simulation with 45% lower emissions, likely more representative of the decline in emissions in cities, leads to an increase in surface ozone of 1.9 ppbv for Manchester and 2.1 ppbv for London. The national decline in PM_{2.5} for 20% emission reduction is 1.1 $\mu\text{g m}^{-3}$, ranging from a decrease of



0.61 $\mu\text{g m}^{-3}$ in SC to 2.0 $\mu\text{g m}^{-3}$ in the SW. The largest response in the SW is due in part to decline in primary and secondary sources of PM_{2.5} in the SE that would ordinarily be transported by prevailing southeasterlies. The response in simulated ozone and PM_{2.5} averaged across the UK to emissions alone (figure 9) is about half the combined response of meteorology and emissions (figure S10).

4. Conclusion

We combined data from TROPOMI and the GEOS-Chem model to determine that lockdown measures in the UK in response to the rise in COVID-19 cases in March 2020 led to a nationwide decline in NO_x emissions of ~20%. These range from being indistinguishable from BAU in Scotland and northern England to >40% reduction in London. Emission changes only account for half the national mean increase in surface ozone and decline in PM_{2.5} in 2020 relative to 2019. The remaining contribution is from meteorological differences between the 2 years, emphasizing the need to account for meteorology and pollution sources in future abatement strategies and in imposing regulatory action.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: <https://s5phub.copernicus.eu/dhus/#/home>.

Acknowledgments

HB, DAP, RJP, and MC acknowledge funding as part of NERC's support of the National Centre for Earth Observation (NCEO Grant Numbers: nceo020005

and NE/R016518/1). EAM acknowledges funding from NERC/EPSC (Award Number EP/R513465/1) and from the European Research Council under the European Union's Horizon 2020 research and innovation programme (through a Starting Grant, Grant Number 851854_UPTROP). The University of Leicester High Performance Computing Facility ALICE was used to conduct data analysis and model simulations. The manuscript was written through contributions of all authors. HB and EAM designed the research. DP and RP conducted the data analysis comparing GEOS-Chem and TROPOMI, and EAM simulated and analyzed model output. EAM and HB with contributions from RP and DP wrote the manuscript. JL and WD provided surface in-situ data. JR, DPM, MC, BK and RS contributed to initial discussions on the study and MC reviewed the manuscript.

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