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UK surface NO$_2$ levels dropped by 42 % during the COVID-19 lockdown: impact on surface O$_3$

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Abstract. We report changes in surface nitrogen dioxide (NO$_2$) across the UK during the COVID-19 pandemic when large and rapid emission reductions accompanied a nationwide lockdown (23 March–31 May 2020, inclusively), and compare them with values from an equivalent period over the previous 5 years. Data are from the Automatic Urban and Rural Network (AURN), which forms the basis of checking nationwide compliance with ambient air quality directives. We calculate that NO$_2$ reduced by 42 % ± 9.8 % on average across all 126 urban AURN sites, with a slightly larger (48 % ± 9.5 %) reduction at sites close to the roadside (urban traffic). We also find that ozone (O$_3$) increased by 11 % on average across the urban background network during the lockdown period. Total oxidant levels (O$_x$ = NO$_2$ + O$_3$) increased only slightly on average (3.2 % ± 0.2 %), suggesting the majority of this change can be attributed to photochemical repartitioning due to the reduction in NO$_x$. Generally, we find larger, positive O$_x$ changes in southern UK cities, which we attribute to increased UV radiation and temperature in 2020 compared to previous years. The net effect of the NO$_2$ and O$_3$ changes is a sharp decrease in exceedances of the NO$_2$ air quality objective limit for the UK, with only one exceedance in London in 2020 up until the end of May. Concurrent increases in O$_3$ exceedances in London emphasize the potential for O$_3$ to become an air pollutant of concern as NO$_x$ emissions are reduced in the next 10–20 years.

1 Introduction

The current SARS-CoV-2 (COVID-19) outbreak was first identified in Wuhan, China, in December 2019 and was recognized as a pandemic by the World Health Organization (WHO) on 11 March 2020 (WHO, 2020). As of early August 2020, there have been almost 18 million confirmed cases and over 700 000 deaths reported across the world (https://coronavirus.jhu.edu/map.html, last access: 15 December 2020). Efforts to prevent the virus spreading have included severe travel restrictions and the closure of workplaces, inevitably leading to a significant drop in emissions of primary air pollutants from several important sectors. This has provided a unique opportunity to examine how air pollutant concentrations respond to an abrupt and prolonged perturbation, followed by policy-relatable increases as restrictions are incrementally relaxed.

The effects of the change of emissions on nitrogen dioxide (NO$_2$) and ozone (O$_3$) have been observed using satellite and in situ measurements in several studies. Table 1 summarizes studies from a growing body of work that report changes in NO$_2$ and other air pollutants in countries across the world that are associated with the global COVID-19 lockdown, including satellite observations (Liu et al., 2020) and in situ measurements. These studies have used various methods to isolate the impact of the COVID-19 lockdown on changes in air pollutants from confounding factors, for instance meteorology, using atmospheric chemistry transport models and weather normalization techniques based on machine learning (ML) algorithms. In Europe, reductions of NO$_2$ are typically slightly larger than we have seen in the UK in our study, perhaps reflecting more stringent lockdown policy. In

Spain, NO_2 was reduced by 50 % at both urban traffic and urban background sites (Petetin et al., 2020); in Rome, Turin and Nice, NO_2 was reduced by 46, 30 and 63 %, respectively (Sicard et al., 2020). In all of these studies similar-magnitude increases of O_3 were observed, mainly attributed to the decreased nitric oxide (NO). Further afield, in India the TROPOspheric Monitoring Instrument (TROPOMI) satellite measurements showed that during the COVID-19 lockdown there was a 18 % decrease in NO_2 over the whole country, with a 54 % decrease over New Delhi compared to the same period in 2015–2019 (Pathakoti et al., 2020). In situ measurements in New Delhi showed a 53 % decrease in NO_2 and a 0.8 % increase in O_3 for the lockdown period compared to the 2 weeks immediately preceding it. In Rio de Janeiro, Brazil, there was a 24–33 % decrease in NO_2 during the lockdown compared to the week before (Dantas et al., 2020), and in São Paulo data from urban roadside sites showed a 54 % decrease in NO_2 compared to the previous 5 years (Nakada and Urban, 2020). In China, satellite observations showed a mean NO_2 decrease of 21 % across the whole country, relative to a similar period in 2015–2019 (Bao and Zhang, 2020). In situ measurements in cities in northern China before and after lockdown showed a 53 % decrease in NO_2 (Shi and Brasseur, 2020), and in situ measurements in cities across the whole of China showed a 60 % decrease in NO_2 comparing 1–24 January 2020 and 26 January–17 February 2020 (Huang et al., 2020). Both these two studies also reported a > 100 % increase in O_3. These studies were both during wintertime, so the O_3 increase was largely attributed to the reduction in NO emissions, reducing titration of O_3 to NO_2; however the possible effect of reduced particles on UV radiation and hence O_3 production was also considered to have led to some of the increased O_3. Le et al. (2020) use satellite data to show a 71.9 % decrease in NO_2 and 93 % decrease in Wuhan at the peak of the outbreak. They also report a 25.1 % increase in O_3 in Wuhan, largely attributed to a reduction in titration with NO. In the USA one study using EPA data showed a mean decrease of 30 % of NO_2 in urban areas of Seattle, Los Angeles and New York during the lockdown. The study did not show any consistent change in O_3 levels (Bekbulat et al., 2020). Here, we report changes in NO_2 across the UK and discuss them in context of observed changes in surface O_3.

In 2018 the road transport sector accounted for 37 % of UK NO_x (sum of NO and NO_2), the largest emission from a single sector, followed by energy industries (21 %), non-road transport (mainly rail and aviation) (15 %), manufacturing industries and construction (10 %) and domestic combustion (9 %) (https://www.gov.uk/government/statistical-data-sets/env01-emissions-of-air-pollutants, last access: 15 December 2020). In major cities, the contribution from road transport is typically much higher. On average across six cities in the UK (London, Bristol, Cardiff, Newcastle, Glasgow and Belfast) for example 47 % ± 6 % comes from road transport, with 17 % ± 5 % from domestic combustion, 15 % ± 6 % from non-road transport sources (mainly rail), 14 ± 10 % from en-

ergy industries and 6 % ± 2 % from industrial combustion. In recent years, there has been a pronounced reduction in NO_x emission (Defra, 2018a), which largely reflects lower transport emissions, with NO_2 showing an average decrease of 3.3 % per year since 2015. Since 2014, Euro 6 standards for light passenger diesel vehicles reduced the maximum permitted NO_x emission from 0.18 to 0.08 $g\,km^{-1}$, and the number of ultra-low emission vehicles (e.g. electric, hybrid cars) has increased its market share from 0.59 % in 2014 to 2.6 % in 2018. Despite these developments, air pollution is still currently the largest environmental health stressor on the UK population (Public Health England, 2019).

At present the main pollutants of concern are NO_2 and particulate matter with diameter smaller than 2.5 μm ($PM_{2.5}$) in urban centres, and O_3 in urban, suburban and rural environments with exposure to excess levels of these species is known to have a negative effect on human health (An et al., 2018; Kurt et al., 2016; Mannucci et al., 2015). O_3 is a secondary air pollutant formed photochemically by the oxidation of volatile organic compounds (VOCs) in the presence of NO_x (Monks et al., 2015). It is generally lower in urban areas due to reactions with NO_x, but in the past 2 decades over the UK (Finch and Palmer, 2020), and across the world (Fleming et al., 2018; Lefohn et al., 2018; Ma et al., 2016; Paoletti et al., 2014; Sicard et al., 2013; Sun et al., 2016), there have been large mean surface O_3 increases in urban centres driven by reduced NO_x emissions. In more rural environments, the opposite has been observed, with O_3 decreasing with decreasing NO_x emissions (Cooper et al, 2012; Cooper et al., 2014; Strode et al., 2015). Air pollution has led to an estimated 29 000 premature deaths per year in the UK, equivalent to 340 000 life years across the population in any 1 year, and costs the UK economy between GBP 10 billion and GBP 20 billion yr^{-1} (Royal College of Physicians, 2016). To meet the UK government's clean air strategy (UK Government, 2019) and its commitment to achieve its zero-carbon-emission target by 2050, sales of non-zero-emission cars, vans and motorcycles will end by 2035. One of the challenges associated with the progressive move to a low-NO_x vehicle fleet in the UK is to understand the impacts on surface air pollution if other emissions are not reduced commensurately.

The widespread and rapid reduction in UK transport activity (and therefore the associated emissions) from the COVID-19 lockdown represents a natural experiment to study air pollution with a greatly reduced volume of NO_x-emitting vehicles, which we use as a proxy for a future low-NO_x vehicle fleet. Figure 1 summarizes the timeline of events associated with COVID-19 in the UK, including Google mobility data that describe the percentage changes in transport from a pre-lockdown baseline and daily mortality values reported by the UK Office of National Statistics. Google mobility data were used as a proxy for traffic counts as they are readily accessible; however for any quantitative analysis of the effect of reduction in traffic on pollution levels, real

Table 1. Summary of previous measurements. AQ: air quality.

Focus region	Observed change in NO$_2$ (NO)	Observed change in O$_3$	Comments	Reference
UK				
UK-wide	−48 % at urban traffic, −41 % at urban background sites	11 % increase at urban background sites	Changes relative to detrended lockdown period 2015–2019.	This study
UK-wide	−30 % to −50 % in urban areas	Increase mostly explained by reduced NO	UK government (Defra) synthesis report describing contributions from 50 individual responses. Data submitted up to 30 April 2020.	https://uk-air.defra.gov.uk/ library/reports.php?report_ id=1005 (last access: 15 December 2020)
Europe				
Greece	−22 % for March and April 2020 compared to 2019		TROPOMI monthly mean tropospheric nitrogen NO$_2$ observations used.	Koukouli et al. (2020)
France	−63 % (−71 %) in Nice	+24 % in Nice		Sicard et al. (2020)
Italy	−46 % (−69 %) in Rome, −30 % (−53 %) in Turin	+14 % in Rome, +27 % in Turin		Sicard et al. (2020)
Spain	Mean changes over all three phases of the lockdown: −4.1 ppb (−50 %) for background urban and −6.3 (−50 %) for traffic sites	–	ML approach used to determine deviation from BAU NO$_2$, trained using 2017–2019 data from background and traffic surface AQ monitoring sites. Study considers all three phases of lockdown up to 24 April 2020.	Petetin et al. (2020)
Spain	−69 % in Valencia	+2.4 % in Valencia	Hourly data provided by local and regional agencies. Changes relative to 2017–2019. All sites noted a decrease before the lockdown. Larger reductions observed at traffic sites.	Sicard et al. (2020)
International				
Brazil	−24 % to −33 % compared to 2019	–	Study over Rio de Janeiro using data from automatic monitoring station run by Municipal Department of the Environment Brazil. Study period is from 2 March to 16 April 2020, with lockdown on 23 March 2020.	Dantas et al. (2020)
Brazil	−54 % (−77 %) on urban roads	+30 %	Study over São Paulo using three in situ AQ sites. Changes relative to similar periods from previous 5-year mean.	Nakada and Urban (2020)
China	−25 %	–	Study of data from 44 cities in northern China from 1 January to 21 March 2020. Lockdowns started on 23 January in Wuhan, with other cities following soon afterwards. Linear regression was used to determine BAU.	Bao and Zhang (2020)
China	−21 %		Study used satellite observations of tropospheric NO$_2$ data over China. Decrease relative to similar period during 2015 to 2019.	Liu et al. (2020)
China	−53 %	+100 %	Study focused on northern China using in situ measurements. Data compared before and after lockdown.	Shi and Brasseur (2020)
China	−57 % (−62 %)	+36.4 %	Study over Wuhan.	Sicard et al. (2020)
China	−60 %	> +100 %	Used in situ measurements across China. Differences between 1–24 January and 26 January–17 February 2020.	Huang et al. (2020)
China	−71.9 %	+25.1 %	TROPOMI measurements over eastern China and Wuhan, compared to previous 5 years.	Le et al. (2020)
India	−18 % from previous 5-year mean; −54 % over New Delhi	–	Used satellite observations of NO$_2$ from TROPOMI, relative to same period 2015–2019.	Pathakoti et al. (2020)
India	−53 % over New Delhi compared to before lockdown	+0.8 % over New Delhi compared to before lockdown	Using 34 monitoring in situ monitoring stations over New Delhi, study compared pre-lockdown period 3–24 March and lockdown period 24 March–14 April 2020.	Mahato et al. (2020)
Kazakhstan	−35 %	+15 %	Study over Almaty using data from a similar previous period from 2018–2019. Data from the AirKaz public AQ monitoring network.	Kerimray et al. (2020)
Morocco	−96 %	–	Study over Salé, Morocco, using urban in situ data.	Otmani et al. (2020)
USA	−30 %	Weak, inconsistent response	Used EPA data.	Bekbulat et al. (2020)

traffic counts or flow data would be required. The UK Foreign and Commonwealth Office issued a travel advisory on 28 January not to travel to mainland China. The first two UK cases of COVID-19 were confirmed on 31 January, with a third case confirmed on 6 February. As the number of cases continued to rise, the first UK death from COVID-19 was confirmed on 5 March. On that same day, the UK government moved from the "containment" to the "delay" phase of addressing COVID-19, which included, for example, social distancing. A UK-wide lockdown was announced nearly 3 weeks later on 23 March, with citizens instructed to stay at home with the exception of shopping for basic necessities, one form of exercise per day, medical needs and travel associated with key workers. An immediate effect of these restrictions in movement was a large and progressive drop in transport use, with an associated reduction in motor vehicles throughout the lockdown period. We use in situ measurements collected across the UK to examine how these reductions (and other changes) have affected NO$_2$ in the UK, with a discussion on how this could have, in turn, affected O$_3$. We also examine the changes in exceedances of limit values for NO$_2$ and O$_3$ and assess whether the COVID-19 lockdown can provide useful information on how air pollution will respond to future changes in emissions due to the move to a low-carbon economy. In the next section we discuss the data we use. In Sect. 3 we describe our results for NO$_2$, which we put into context in Sect. 4 with the observed changes in surface O$_3$, as well as comparing our results with other studies. We conclude the paper in Sect. 5.

2   Data and methods

2.1   In situ measurements of NO$_2$ and O$_3$

We use data collected as part of the Defra Automatic Urban and Rural (AURN) network, which currently consists of 150 active sites across the UK (Fig. S1 and Tables S1 and S2 in the Supplement) and is the main network used for compliance reporting against the Ambient Air Quality Directives. It includes automatic air quality monitoring stations measuring oxides of nitrogen (NO$_x$), sulfur dioxide (SO$_2$), ozone (O$_3$), carbon monoxide (CO) and particles (PM$_{10}$, PM$_{2.5}$). Online measurements of VOCs are available at a small number of sites. These sites provide hourly information, which is communicated rapidly to the public, using a wide range of electronic media and web platforms. More detail can be found at https://uk-air.defra.gov.uk (last access: 15 December 2020). Three different site types are used in this analysis. Urban traffic sites are defined as being in continuously built-up urban areas, with pollution levels predominantly influenced by emissions from nearby traffic. Urban background sites are located such that pollution levels are not influenced significantly by any single source or street, but rather by the integrated contribution from all sources upwind of the sta-

tions. These can be considered more representative of residential areas. Rural background sites are sited more than 20 km away from agglomerations and more than 5 km away from other built-up areas, industrial installations or motorways or major roads, so that the air sampled is representative of air quality in a surrounding area of at least 1000 km^2.

The AURN network uses standardized techniques and operating procedures to ensure data are comparable. Full details can be found at https://uk-air.defra.gov.uk/assets/documents/reports/empire/lsoman/ (last access: 15 December 2020), but a brief description will be given here. NO in the sample air stream reacts with O$_3$ in an evacuated chamber to produce activated nitrogen dioxide (NO$_2$*). This then returns to its ground (unactivated) state, emitting a photon (chemiluminescence). The intensity of the chemiluminescent radiation produced depends upon the amount of NO in the sampled air. This is measured using a photomultiplier tube or photodiode detector, so the detector output voltage is proportional to the NO concentration. The ambient air sample is divided into two streams. In one stream, the ambient NO$_2$ is reduced to NO (with at least 95 % efficiency) using a molybdenum catalyst converter before reaction. The molybdenum converter should be at least 95 % efficient at converting NO$_2$ to NO. External gas cylinders or an internal permeation oven and zero air scrubber are used to provide daily automatic check calibrations for NO. The NO$_2$ conversion efficiency is checked every 6 months using either an NO$_2$ calibration cylinder or gas phase titration of the NO with O$_3$. In recent years it has become well established that NO$_2$ measurements using molybdenum converters can overestimate NO$_2$ due to interferences from other oxidized nitrogen species (e.g. HNO$_3$, PAN, HONO) (Steinbacher et al., 2007). However, in urban environments the interferences are often minimal compared to the levels of NO$_x$ (Villena et al., 2012). In addition, as we are looking at a change in NO$_2$, it is likely that any interference that is present will be there in very similar amounts in both the 2020 and 2015–2019 data. Ozone is measured by UV absorption at 254 nm, with concentrations calculated using the Beer–Lambert law (Parrish and Fehsenfeld, 2000). An O$_3$-removing scrubber is used to provide a zero-reference intensity. An internal ozone generator and zero air scrubber are used to provide daily automatic check calibrations, and instruments are calibrated with a primary ozone standard every 12 months. Whilst the accuracy of the measurement will vary on a site-by-site basis, the maximum allowed uncertainty for the AURN network is 15 % for NO$_2$ and O$_3$ measurements. To study the effect of the lockdown on NO$_2$ levels in the UK, we use measurements from 66 urban traffic and 62 urban background sites across the UK, all of which have measurements between 2015 and the end of May 2020.
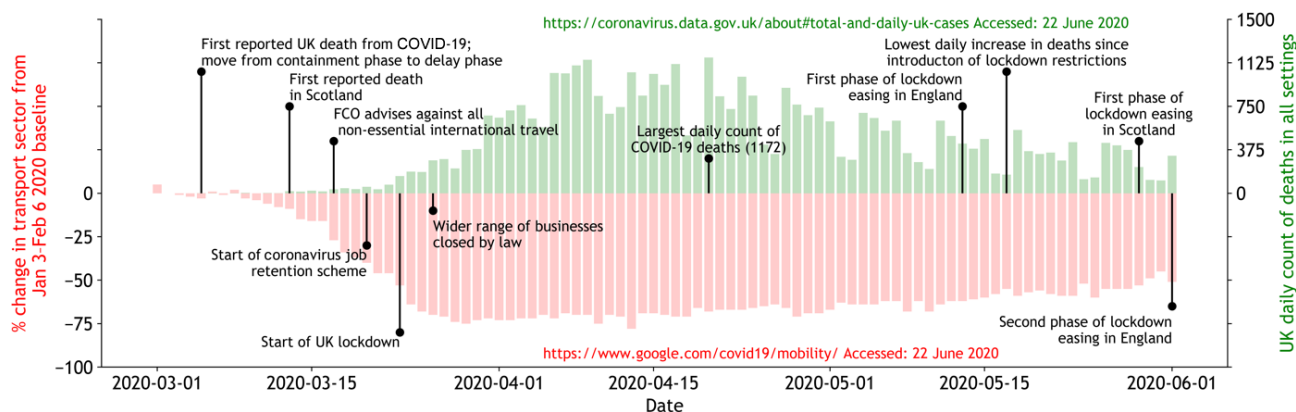
Figure 1. Schematic of the timelines involved with daily mortality values attributed by the UK government to COVID-19, and changes in mobility from the transport sector (inferred from Google location data on smartphones) compared to a reference period (3 January–6 February 2020) before the lockdown period. Also included are key dates that describe the run-up and evolution of the UK lockdown. Data acknowledgements are shown inset on the plot.

2.2 Correlative meteorological data

Measured meteorological data (wind direction, wind speed and temperature) are not available at most AURN sites, so modelled data, based on the position of the site, from the UK Met Office Unified Model are used. UV-A irradiance data are taken from measurements made by the Public Health England solar network.

2.3 Statistical methods

To quantify the impact of the COVID-19 lockdown on atmospheric levels of NO$_2$ and O$_3$, we compare measurements during the lockdown with values corresponding to "business as usual" (BAU), i.e. what we would have expected in the absence of the pandemic. To determine our BAU scenario, we first linearly detrend and deseasonalize NO$_2$ data at each AURN site. To deseasonalize the data, we determine the climatology based on the mean annual cycle of the previous 5 years (from 1 January 2015 to 31 December 2019), which is then repeated to match the length of the time series, subtracted from the mean to standardize the data and then subtracted from the original time series to produce a time series of the residuals. This 5-year period is sufficiently long to take into account year-to-year variations in meteorology but short enough to reduce the impact of any longer-term trends driven by earlier changes in emission standards. We then calculate the difference between a linear regression model of the previous data, projected forward to June 2020 to predict BAU values of NO$_2$ and O$_3$ (Fig. 2), and calculate the difference between this and the measured values. We acknowledge there are uncertainties associated with our approach, but this method offers simplicity and straightforward error propagation. Other more complex methods of determining BAU that, for example, explicitly take into account local changes in meteorology (Grange and Carslaw, 2019) will also be subject to

uncertainties, for example the extent to which regional-scale meteorological fields can describe smaller-scale variations in atmospheric pollutants. We define the start of the UK lockdown period as 23 March 2020, when the lockdown was advised by the UK government. Figure 1 shows that a decrease in mobility in the transport sector is already evident from 9 March, which in the absence of any obvious change in law is perhaps influenced by the emerging crises in nearby European countries. Our analysis concludes on 31 May 2020, the day before the first phase of lockdown easing in England.

We use independent sample Mann–Whitney U tests to test the significance of changes in mean concentration for each site between the lockdown period and the mean of the same period for the past 5 years, the lockdown period and measurements in 2020 prior to the lockdown, and measurements from prior to lockdown in 2020 with the same period for the previous 5 years. This test indicates how likely the observed changes in mean concentration between the different time periods are due to chance and noise in the data or whether they are statistically significant and can be attributed to a real signal, which in our work is the start of the lockdown. We use this test rather than a t test or z test due to the large sample size and non-normal distribution of the data.

3 Results

Figure 2 shows the mean relative change of UK deseasonalized NO$_2$ and O$_3$ observations from all urban sites from 2015–May 2020 and the mean trend from 2015 to 2019. The mean NO$_2$ linear trend across all AURN urban traffic (background) sites is -1.4 $(-0.6)\,\mu g\,m^{-3}\,yr^{-1}$ $(-4.5$ $(-2.1)\,\%\,yr^{-1})$. The urban traffic site at London Marylebone Road shows the largest decreasing trend over the past 5 years: $-5.5\,\mu g\,m^{-3}\,yr^{-1}$ $(-6.7\,\%\,yr^{-1})$; in contrast, eight urban sites show a small increasing trend in NO$_2$ between
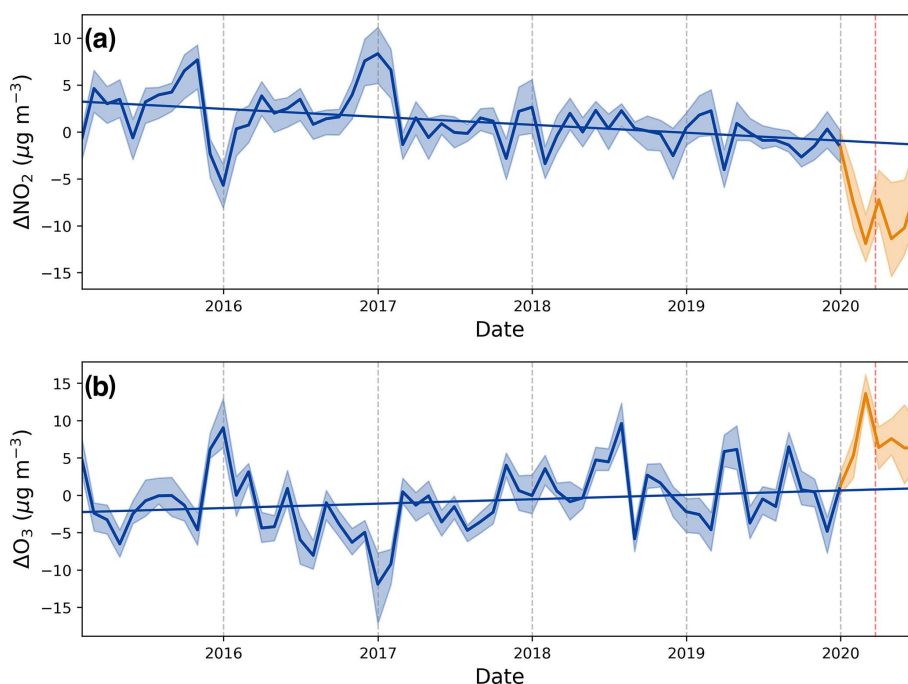
Figure 2. Mean relative change of deseasonalized UK values of **(a)** NO$_2$ and **(b)** O$_3$ for all urban background and traffic sites from 2015 to 2020, with the mean 2015–2019 trend superimposed. Data from 2020 are shown in orange, with the red dashed line denoting the start of the lockdown on 23 March 2020. The 25–75 % range is shown by the shaded area.

0.1 and 0.6 μg m^{-3} yr^{-1} (0.5 % yr^{-1}–1.2 % yr^{-1}). The mean standard error of the NO$_2$ trend for all sites is 0.002 μg m^{-2}. The mean O$_3$ linear trend across all urban traffic (background) sites is 2.4 (1.3) μg m^{-3} yr^{-1} (5.5 (3.1) % yr^{-1}), and the mean standard error of the fit for all sites is 0.003 μg m^{-2}.

3.1 Meteorological context

It is well understood that ambient concentrations of air pollutants are greatly affected by meteorology, with low wind speeds causing a build-up of pollutants, and over the UK easterly flow is often accompanied by pollution from mainland Europe. Figure 3 shows surface wind data from six cities across the UK (London, Bristol, Cardiff, Newcastle, Glasgow and Belfast), providing information from a wide geographical range across the country. Wind roses for the pre- (10 January–10 March) and post-lockdown (23 March–31 May) periods of 2020 and the mean of 2015–2019 show that all cities during the pre-lockdown period in 2020 were dominated by strong westerly winds across all of the UK, with successive low-pressure systems across the UK, including the storms named Ciara, Dennis and Jorge through the month of February and early March. The winter season (January–February) was the fifth wettest on record and the fifth warmest. February 2020 was the wettest-ever February recorded in the UK. The wind roses also show that 2020 saw much stronger winds than the mean of the previous 5 years. The six cities saw an average wind speed in 2020 of

(6.5 ± 1.2) m s^{-1}, which was 33.5 % higher than the average of the previous 5 years. Since the beginning of the COVID-19 lockdown, meteorological conditions have been much more settled, with high pressure and easterly winds dominating UK weather since mid-March, especially in the southern and western UK. Average wind speed across the six cities was (4.1 ± 0.4) m s^{-1}, although this is still an increase of 7.5 % compared to the previous 5 years. Of the cities analysed, Cardiff saw the largest increase in wind speed for 2020 compared to the previous 5 years (16.8 %), with Bristol showing a 10 % increase. The other cities all saw slight (< 5 %) decreases in wind speed in 2020. Typically, meteorological conditions of lower wind speed are associated with higher levels of air pollution due to increased atmospheric stability and transport of pollution from mainland Europe in the UK, and so care must be taken when comparing pre and post-lockdown levels of air pollution, as described in Sect. 2.3, and comparing to the average of the previous 5 years is a better measure of the changes.

3.2 Observed changes in daily mean and diurnal variations of NO₂

Measurements in 2020 from 65 urban traffic (Fig. S2a in the Supplement) and 61 urban background (Fig. S2b in the Supplement) AURN measurement sites across the UK show clear reductions in NO$_2$ concentrations across all sites following the lockdown. Some of these differences are due to the nat-
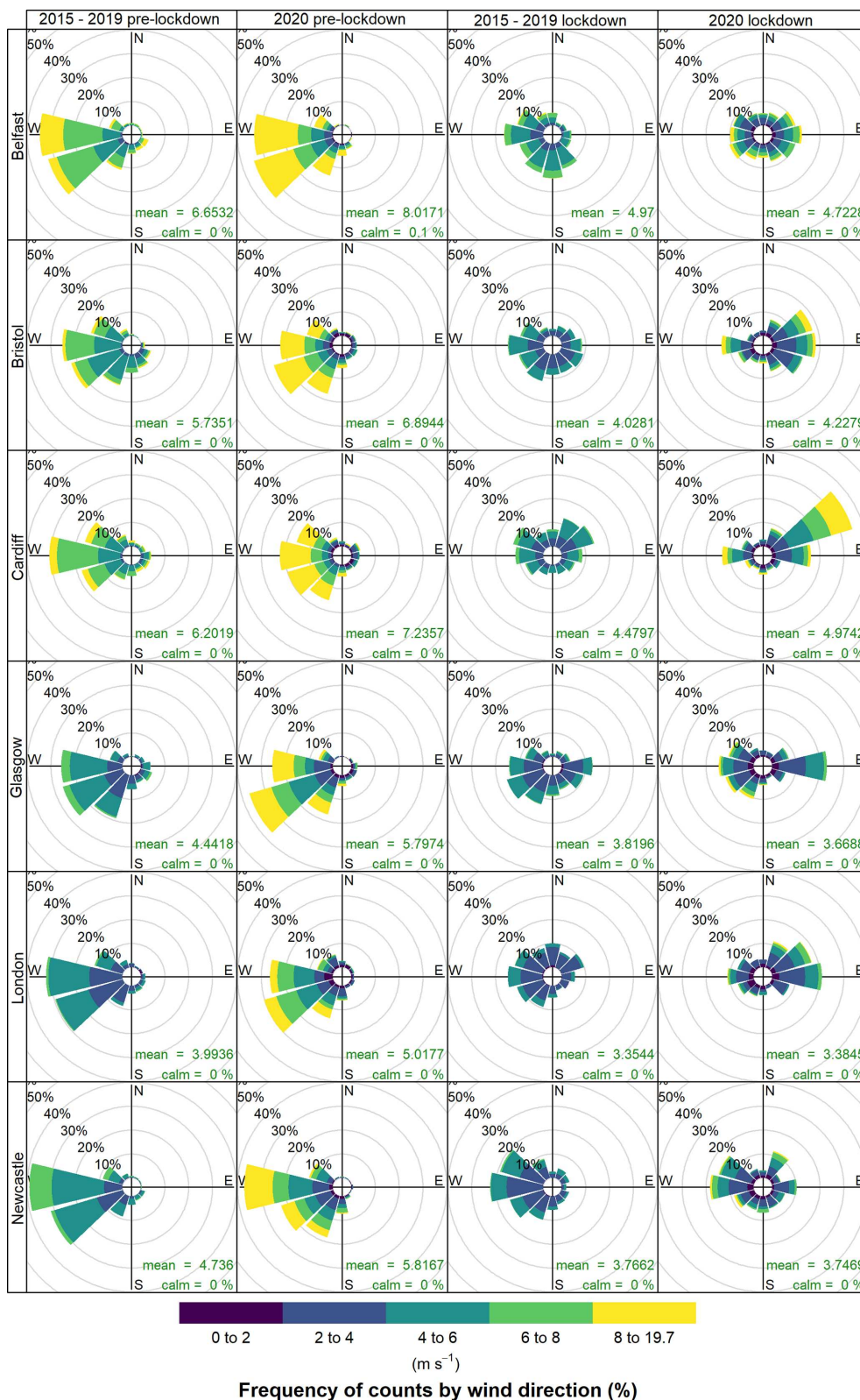
Figure 3. Average wind roses for six cities for pre- and post-lockdown period and lockdown period 2015–2019 and 2020. Data used are modelled using the UK Met Office Unified Model.

ural seasonal variation in NO$_2$ and meteorology. To account for these expected variations, we calculate the daily difference of NO$_2$ values from 2020 with mean NO$_2$ values from detrended values from 2015 to 2019 for the appropriate day of year. This approach allows us to emphasize the difference of NO$_2$ values in 2020 from previous years. During the lockdown period, we find that 83 % of days in 2020 at urban traffic sites have lower NO$_2$ values, far outnumbering those with higher NO$_2$ values (17 %). During the pre-lockdown period, we find 76 % of days at urban traffic sites in 2020 are below the 2015–2019 mean. We find a similar situation for urban background sites, with 73 % of days below the 2015–2019 mean, but the decrease during the lockdown period is not as dramatic.

Figure 4 shows the percentage difference for all urban traffic and urban background sites for the lockdown period in 2020 compared to the same period averaged across 2015–2019. To assess the error, we combined the standard error in the median of the daily median concentrations for the lockdown period in 2015–2019 and 2020, with error bars shown on the graph. After removing site-dependent trends, it is observed that urban traffic sites have a mean decrease of $(13.4 \pm 2.1) \, \mu g \, m^{-3}$ in NO$_2$ over the lockdown period compared with the same period over the previous 5 years. This mean decrease approximately equates to a (48 ± 9.5) % drop in NO$_2$ levels across the UK. The AURN site Glasgow Kerbside observed the largest mean percentage decrease of (71.2 ± 7.7) % during the lockdown period, closely followed by Cambridge Roadside (68.8 ± 9.9) % and London Marylebone Road with a decrease of (67.8 ± 7.8) %. In total, 32 of the 65 urban traffic sites saw a decrease in NO$_2$ of greater than 50 %. Armagh Roadside (Northern Ireland) is the only urban traffic site to show a mean increase in NO$_2$ $(1.3 \pm 1.1) \, \mu g \, m^{-3}$ (6.7 ± 6.2) %. Urban background sites show a smaller mean reduction of $(4.9 \pm 1.1) \, \mu g \, m^{-3}$, equating to a decrease of (40.6 ± 10.1) % in NO$_2$ levels across the UK. The largest decrease of $(25.7 \pm 3.3) \, \mu g \, m^{-3}$ was observed at London Hillingdon, corresponding to (59.3 ± 9.6) %. Small increases $(< 3 \, \mu g \, m^{-3} \, (< 10 \, \%))$ were seen in York Bootham (Yorkshire and Humberside) and Eastbourne (South East). On average across all urban sites (traffic and background), a decrease in NO$_2$ of (42 ± 9.8) % is observed. We see that, whilst NO$_2$ concentrations do tend to be higher at lower wind speed (Fig. S3b in the Supplement), there is very little correlation between the observed change in NO$_2$ between 2020 and the previous 5 years and any change in wind speed (Fig. S3a in the Supplement).

We perform independent Mann–Whitney U tests on NO$_2$ measurements during the lockdown period and the mean from the same period from the previous 5 years, on NO$_2$ measurements during the lockdown period and measurements in 2020 immediately prior to the lockdown, and on NO$_2$ measurements immediately prior to lockdown in 2020 with the same period for the previous 5 years. Using these tests, we find that 115 out of the 128 (89 %) urban sites show a sta-

tistically significant ($p < 0.01$) difference in NO$_2$ between the mean observations during lockdown and the mean of the same period during the past 5 years. We also find that 112 sites (from a possible 128 sites, 88 %) show a significant difference between NO$_2$ measurements made in 2020 immediately prior to the lockdown and the mean of the same period from the previous 5 years, with urban background sites showing a $-6.3 \pm 1.5 \, \mu g \, m^{-3}$ change and urban traffic a $9.2 \pm 1.9 \, \mu g \, m^{-3}$ change. We attribute this difference to changes in meteorology during January and February 2020 (Fig. 3), in particular wind speed, which was on average 33 % higher than the average of the previous 5 years. Finally, we find that 94 sites (75 %) show a statistically significant difference between mean NO$_2$ observations during lockdown and immediately prior to lockdown in 2020, implying there was a significant drop in NO$_2$ across the UK as a direct result of the lockdown.

We also examine mean changes in the diurnal cycles of NO$_2$ at urban traffic and urban background sites in London, Bristol, Cardiff, Newcastle, Glasgow and Belfast during the lockdown period compared to the same periods from the 2015–2019 mean. Based on the diurnal profile of NO$_2$ levels, we find that (Fig. S4 in the Supplement) typical pre-lockdown diurnal cycles are driven by emission peaks in the morning and evening rush hours, with the evening peaks suppressed due to the higher mean boundary layer that grows during the day. In general, we find that the evening rush hour peaks at urban traffic sites across all cities during the lockdown period are suppressed compared to previous years, potentially due to changing working patterns. A notable exception is in Cardiff, where the morning rush hour peak is suppressed. In contrast, at urban background sites in Cardiff the diurnal cycle of NO$_2$ is very similar in 2020 to the previous years, with rush hour peaks of similar magnitude in the morning and evening. A reason for these observed diurnal cycles could be domestic combustion, which typically makes up around 17 % of urban NO$_x$ emissions (compared to 47 % for road transport and 15 % for other transport, e.g. rail). We do not expect domestic combustion to have changed much during the lockdown; therefore its contribution to the total (and the diurnal cycle) will be greater.

3.3 Observed changes in daily mean O$_3$ and O$_x$ (O$_3$ + NO$_2$)

Typically, close to sources of NO$_x$, O$_3$ is suppressed due to the reaction of high levels of NO with O$_3$. Further away from the sources, O$_3$ can reform through the oxidation of NO to NO$_2$ with peroxy radicals (formed from the reaction of VOCs with OH) and subsequent photolysis of NO$_2$ to form O$_3$. To account for this photochemistry, we also report changes in the total oxidant, O$_X$, the sum of O$_3$ and NO$_2$, which should be approximately conserved in the absence of any change in the source strength of NO$_x$ or VOCs or a change in OH.
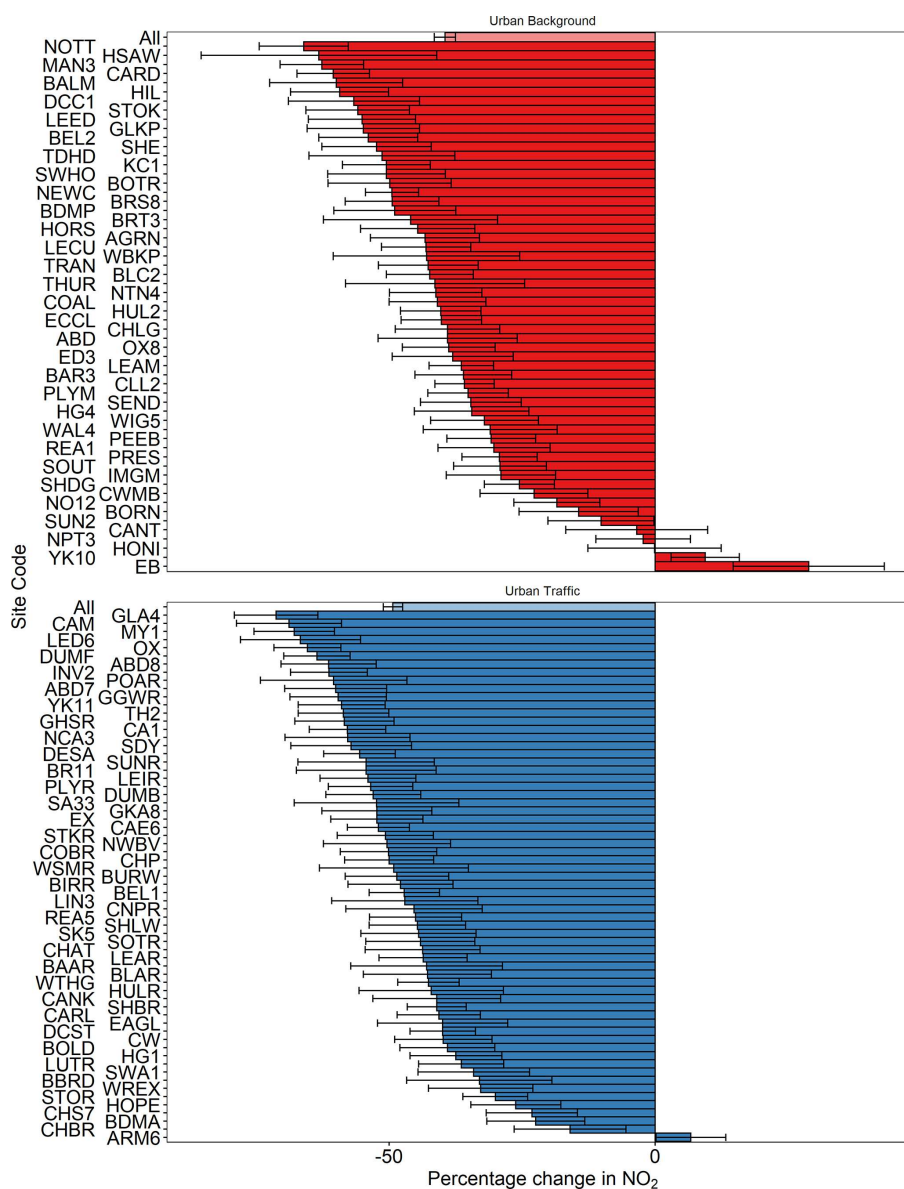
Figure 4. Percentage change in NO_2 at all urban background and urban traffic sites for the lockdown period (23 March–31 May) in 2020 compared to the same period averaged across the previous 5 years, after removing site-dependent trends. The lighter-coloured bar at the top shows the average of all sites. Site acronyms can be found in the Supplement.

Figure S5 in the Supplement shows measurements of O_3 in 2020 from 46 urban traffic and background AURN measurement sites across the UK, along with the daily difference of NO_2 values from 2020 with mean O_3 values from detrended values from 2015 to 2019. It shows the opposite trend to NO_2, with clear increases across the majority of the sites. Figure 5 shows the percentage difference for all urban traffic and urban background sites for the lockdown period in 2020 compared to the same period averaged across 2015–2019. After we remove site-specific trends, as described in Sect. 2.3, we find that O_3 at urban background sites increased by a mean value of $(7.2 \pm 2.6)\,\mu g\,m^{-3}$ during the lockdown

period when compared with the previous 5 years, equating to a percentage increase of $(11 \pm 3.2)\,\%$. Leamington Spa (West Midlands) and London Hillingdon observed the largest mean increases of $(21.3 \pm 2.7)\,\mu g\,m^{-3}$ $((35 \pm 3.6)\,\%)$ and $(21.6 \pm 3.6)\,\mu g\,m^{-3}$ $((54 \pm 7.5)\,\%)$, respectively. Three sites observed a decrease during the lockdown, with Aberdeen seeing a large decrease in O_3 of $(24.0 \pm 4.5)\,\mu g\,m^{-3}$ $((36 \pm 8.6)\,\%)$ even though this site also experienced a substantial decrease in NO_2. We do not have a definitive explanation for this result, but it is consistent with a NO_x-limited photochemical environment in which a decrease in NO_2 would reduce O_3 production. This could be achieved by
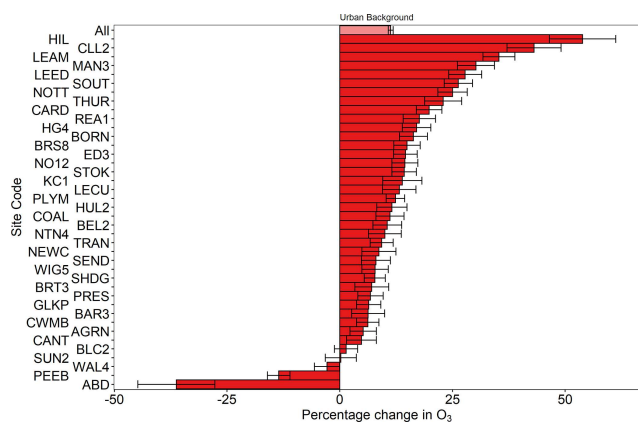
Figure 5. Percentage change in O$_3$ at all urban background sites for the lockdown period (23 March–31 May) in 2020 compared to the same period averaged across the previous 5 years, after removing site-dependent trends. The lighter-coloured bar at the top shows the average of all sites. Site acronyms can be found in the Supplement.

possible fugitive emissions from the onshore gas terminals near Aberdeen, although we have no VOC measurements to confirm this, so the hypothesis is entirely speculative. Only three urban traffic sites measured O$_3$ during our study period. Of those London Marylebone Road saw the largest increase ((32.0 ± 4.8) µg m^{-3} or (104 ± 10.1) %), followed by Exeter Roadside (South West) with an increase (20.0 ± 2.8) µg m^{-3} $((47 \pm 5.5)$ %) and Birmingham A4540 Roadside (West Midlands) with an increase of (13.3 ± 2.5) µg m^{-3} $((25 \pm 3.9)$ %).

A similar statistical analysis has also been carried out for daytime (10:00–18:00 UTC) O$_x$ (NO$_2$ + O$_3$), and we find a mean increase of O$_x$ at urban background sites of (3.5 ± 0.3) µg m^{-3} or (3.2 ± 0.2) %. The two outliers are Leamington Spa (West Midlands), where we find the largest O$_x$ increase of (32.5 ± 1.2) µg m^{-3} $((18 \pm 3.4)$ %), and Aberdeen, where we find the largest O$_X$ decrease of (-27.6 ± 0.4) µg m^{-3} $((58 \pm 5.4)$ %). The three urban traffic sites measuring both O$_3$ and NO$_2$ show a large range in observed differences of (4.2 ± 1.1) µg m^{-3} $((+3 \pm 0.4)$ %) at Birmingham A4540 Roadside, (-7.9 ± 1.8) µg m^{-3} $((-11 \pm 3.1)$ %) at Exeter Roadside and (-20.5 ± 4.7) µg m^{-3} $((-15 \pm 3.1)$ %) at London Marylebone Road.

Following our approach for NO$_2$, we use independent Mann–Whitney U tests to determine the significance of changes in O$_3$ in pre-lockdown and lockdown periods in 2020 and in the previous 5 years. We find that 36 out of 46 urban sites (78 %) show a statistically significant ($p < .01$) difference between the mean O$_3$ observations during lockdown and the mean of the same period from 2015 to 2019. However, we also find that 41 of those sites (83 %) show a statistically significant difference between O$_3$ measurements immediately prior to the lockdown compared to the mean of the same period from 2015 to 2019. Finally, we find that 40 sites (95 %) show a statistically significant difference between O$_3$

observations during the lockdown period and values taken from the period immediately prior to the lockdown.

3.4 Relationship to emissions

During the lockdown period there was around a 75 % reduction in road traffic across the UK (using Google activity data as a proxy for traffic) (see Fig. 1). According to the National Atmospheric Emissions Inventory (NAEI), road transport is estimated to make up 53 % of NO$_x$ emissions in the 1 km × 1 km grid square that both urban background and urban traffic sites are situated in (Defra, 2018b). Therefore, we might expect there to be a reduction in NO$_2$ of around 40 % across all sites. Mean decreases in NO$_x$ are very similar to those for NO$_2$ described above (47 % at urban traffic sites and 40 % at urban background sites – see Fig. S6 in the Supplement), which is in line with the 40 % reduction figure. However, it is clear that individual sites have very different behaviour, and the 75 % traffic reduction may not necessarily equate to 75 % reduction in emissions because different types of vehicle were affected differently, with the greatest reduction in passenger cars and less reduction in high emitters like HGVs. There are a wide range of contributions of NO$_x$ emissions from road traffic across the sites, and there does not appear to be much correlation between this and the reduction seen during lockdown (see Fig. S7 in the Supplement), suggesting that the change in traffic flow near to individual sites is variable and will be the largest contributing factor to NO$_2$ reductions.

4 Discussion

4.1 Surface O$_3$

The COVID-19 lockdown has resulted in a significant decrease in NO$_2$ in cities across the UK, largely caused by the reduction of NO$_x$ emissions due to reduced traffic, and a concurrent increase in O$_3$. NO$_x$ and O$_3$ are closely linked through their photochemistry, and here we examine the reasons for the O$_3$ increase (Lelieveld and Dentener, 2000). Figure 6 shows the relationship between NO$_2$ and O$_3$ during the lockdown period, across all the AURN sites we examined. There is a clear anti-correlation between median NO$_2$ and median O$_3$ for all data, with data from 2020 tending towards lower NO$_2$ and high O$_3$ (Fig. 6b). We also see that there is a correlation between the change in NO$_2$ and the change in O$_3$ between 2020 and the previous 5 years (Fig. 6a). Another key factor that plays a role in O$_3$ concentrations is meteorology (Monks, 2000). High levels of actinic radiation cause the photochemistry involved in O$_3$ formation to happen faster, and low wind speed conditions allow precursor species such as NO$_x$ and VOCs to build up and react to form O$_3$. Therefore, observed variations of O$_3$ in different UK cities will be influenced by a number of processes to varying degrees. Figure 7 examines NO$_2$, O$_3$ and total daytime (10:00–
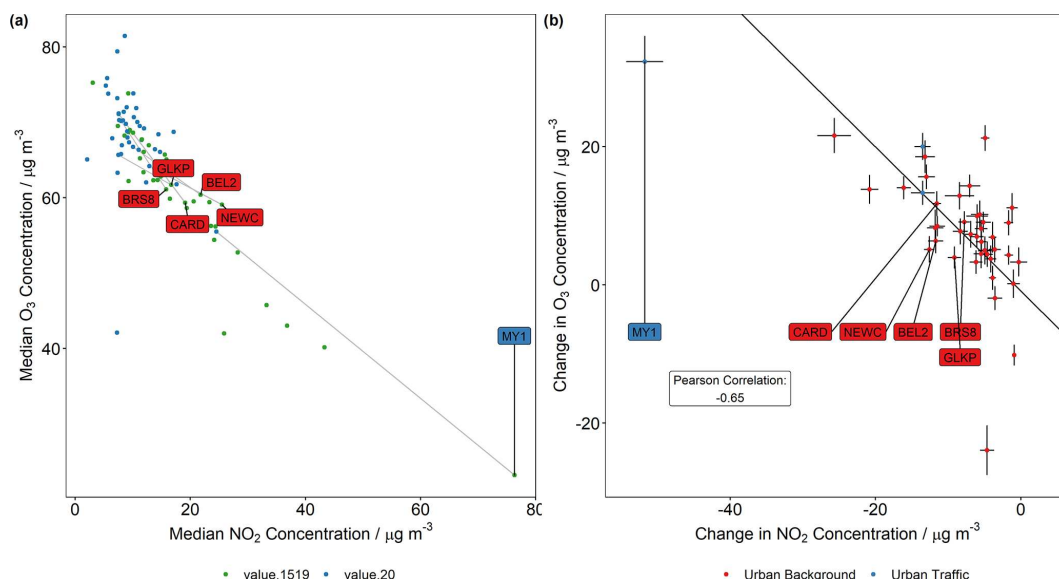
Figure 6. Panel (**a**) shows median O$_3$ concentration plotted against median daily NO$_2$ concentration for each site. Data for 2020 and the average of 2015–2019 are coloured blue and green, respectively. Panel (**b**) shows change in O$_3$ concentration between 2020 and the average of 2015–2019 plotted against the change in NO$_2$ concentration for the same time period. Labelled are sites from six cities across the UK.
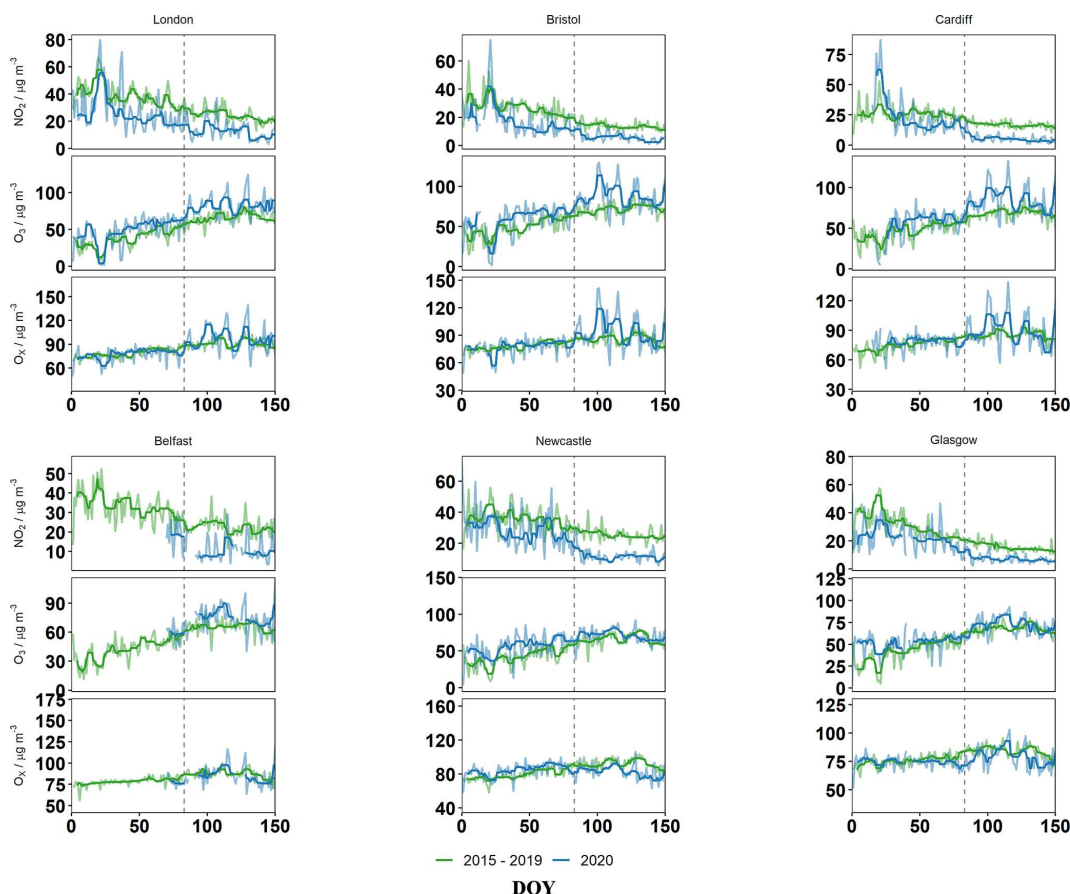

Figure 7. Daily median time series of NO$_2$, O$_3$ and O$_x$ (NO$_2$ + O$_3$) for 2020 and the average of 2015–2019 at urban background sites in six cities representing a geographical and political spread across the UK. The thick line represents the 7 d rolling mean. The dashed grey line indicates the start of the lockdown period.
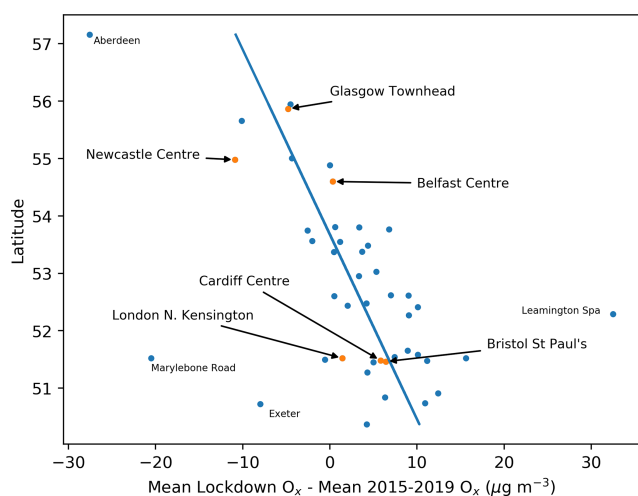
Figure 8. Difference in mean O$_x$ (µg m^{-3}) and between the lockdown period and the detrended mean of the same period from 2015 to 2019 for urban background sites as a function of latitude. Sites examined in Fig. 6 are highlighted in orange.

18:00 UTC) O$_x$ during the lockdown period for urban background sites in six different cities across the UK. Any change in O$_x$ can be thought of as a change in the abundance of oxidants, taking into account the repartitioning of NO$_2$ and O$_3$ caused by changes in NO$_x$ emissions. Whilst all cities have seen an increase in O$_3$ in the urban background compared to previous years, only southern UK cities saw a significant increase in total O$_x$, with London, Bristol and Cardiff showing increases of 5.1 % ± 0.3 %, 5.8 % ± 0.6 % and 5.6 % ± 1.2 %, respectively. In contrast, O$_x$ slightly decreased in Newcastle (-3.2 % ± 0.3 %), Glasgow (-2.8 % ± 0.2 %) and Belfast (-1.4 % ± 0.2 %). To assess if OH is the cause of changes in O$_x$, we examine six measurements of total UV-A at eight sites in the UK and compare data from 2020 to the mean of the previous 5 years (Fig. S8 in the Supplement). We find levels of UV across the UK were higher in 2020 compared to previous years, with the largest increases in the southern UK. London, Chilton and Camborne saw increases of around 50 % compared to previous years, with Glasgow and Inverness showing smaller increases of around 30 %. Figure 8 shows a summary of the O$_x$ change in 2020 compared to 2015–2019 from individual sites across the UK as a function of latitude. We find a positive trend in O$_x$ towards lower latitudes, consistent with the higher excess UV levels further south. Therefore we conclude that in the cities in the southern UK some of the O$_3$ increase is attributable not solely to reduced NO$_x$ but also to an increase in photochemistry related to the hot, sunny weather experienced in 2020.

Observed variations in O$_3$ may also be affected by changes in precursor VOCs. Online measurements of VOCs are only available at a small number of sites, and here we consider measurements made at London Marylebone Road (an urban traffic site) and London Eltham (an urban background site).

Figure S9 in the Supplement shows measurements of a range of different VOCs for each site during 2020 and mean values for 2017–2019 when data are available at both sites. The data show most VOCs decrease in concentration during the post-lockdown period in 2020 compared to previous years. This is particularly true at the urban traffic site and for species such as benzene and toluene, which have traffic emissions as a main source and saw a decrease of 23 % ± 5.1 % and 29 % ± 6.5 %, respectively, compared to previous years. At London Eltham the decreases were both around 12 %. VOCs have a wide range of lifetimes and emissions sources, and they can be transported large distances, meaning their concentrations at a given site are much more affected by meteorology and chemistry than NO$_2$. Indeed, in London according to the NAEI (in 2018), road transport only contributes 11 % to sources of benzene, with other major sources being domestic or commercial combustion (69 %), other transport (10 %) and offshore oil and gas production (6 %) (Vaughan et al., 2016). Therefore it is not surprising that VOCs show less of a reduction during the lockdown than NO$_2$. When examining O$_3$, it can be useful to look at the total VOC loading and OH reactivity (k'). Figure S10 in the Supplement shows total VOC loading in parts per billion and total OH reactivity for each day in 2020, with colours showing the percentage change from the previous 3 years for that day. A full analysis of the behaviour of different VOCs during the lockdown period is beyond the scope of this work, and it is unlikely that the measurements made at the AURN sites cover all VOCs that contribute to OH reactivity (e.g. few oxygenated compounds or larger VOCs are measured). Our focused analysis shows that, whilst the picture is not straightforward, there is an apparent decrease in total VOCs at both sites compared to previous years. Mean values for total VOCs at London Marylebone Road were 17 % lower, and the corresponding k' 15 % lower, than the 2017–2019 mean. At London Eltham total VOCs saw a decrease of 10 %, with a slight increase in the total k', largely driven by an increase in biogenically emitted isoprene.

Figure 9 shows daytime mean (10:00–18:00 UTC) isoprene data and its contribution to k' at two sites in 2019 and 2020 (the only years where reliable isoprene data are available). Observed isoprene was a factor of 2 higher at both London Marylebone Road and London Eltham during April and May 2020 compared to those months in previous years. Isoprene represents only a small contributor to OH reactivity at London Marylebone Road, but at London Eltham in 2020 it represents around 25 % of total k'. Biogenic emissions of isoprene, originating from a variety of trees and shrubs, are driven in part by temperature, and so it is perhaps not surprising that isoprene levels at the London sites were higher in 2020 compared to 2019 due to the fact that temperature was approximately 2 °C higher in 2020 compared to 2019 for the lockdown period. Further detailed chemical modelling studies, beyond the scope of this study, are required to assess in detail the chemistry behind O$_3$ formation and how this has
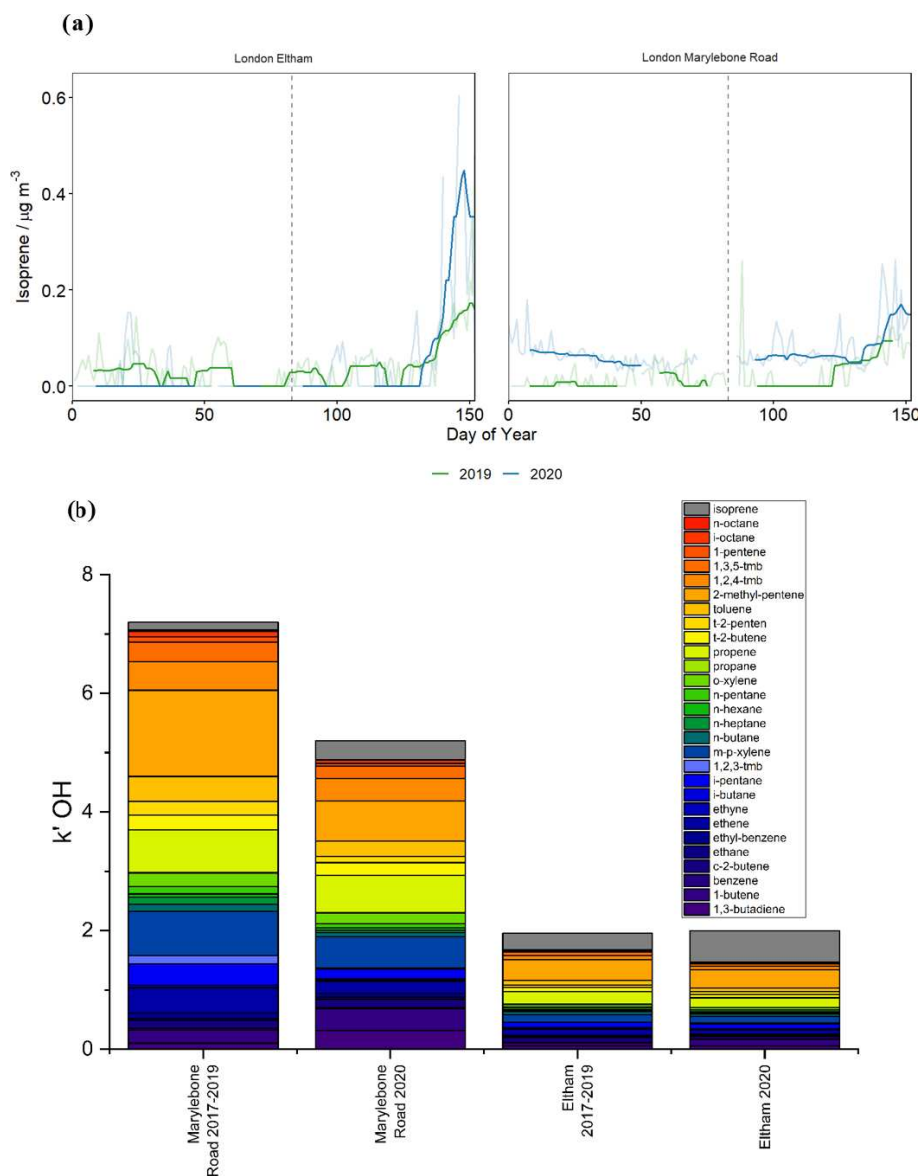
Figure 9. (a) Levels of isoprene at the London Eltham (urban background) and London Marylebone Road (urban traffic) sites in 2020 and 2019. Panel **(b)** shows the contribution of isoprene (grey slice) and other VOCs to total OH reactivity (k') for each site for 2019 vs. 2020.

been affected by the lockdown; however we observe that O_3 has increased across the UK and see a clear anti-correlation with a decrease in NO_2 across the sites. We also see an increase in total O_x at urban background sites in the south of England, likely due to increased radiation and biogenically emitted VOCs compared to previous years, things that are unlikely to be linked to the COVID-19 lockdown.

4.2 Exceedances

To put the changes in air pollutants in context with human health effects, we have examined the number of exceedances of UK air quality objectives (Defra, 2019) and EU directive limits (EEA, 2016) for both NO_2 and O_3 in 2020 compared to

previous years (see Table 2). For this analysis, we have used data from the London Air Quality Network (LAQN) consisting of 9 kerbside, 31 roadside and 14 background sites for NO_2 and 1 kerbside, 7 roadside and 5 urban background sites for O_3 in the Greater London area. London has historically had by far the largest number of air quality exceedances in the UK, so this analysis allows us to see the effect of the lockdown on the city with the most acute air pollution problems.

Exceedances were calculated on a per-site basis and then summed across all sites of a given type. For NO_2 a simple 1 h mean was calculated, and each hour greater than $200 \, \mu g \, m^{-3}$ was counted as an exceedance. We calculated a rolling mean
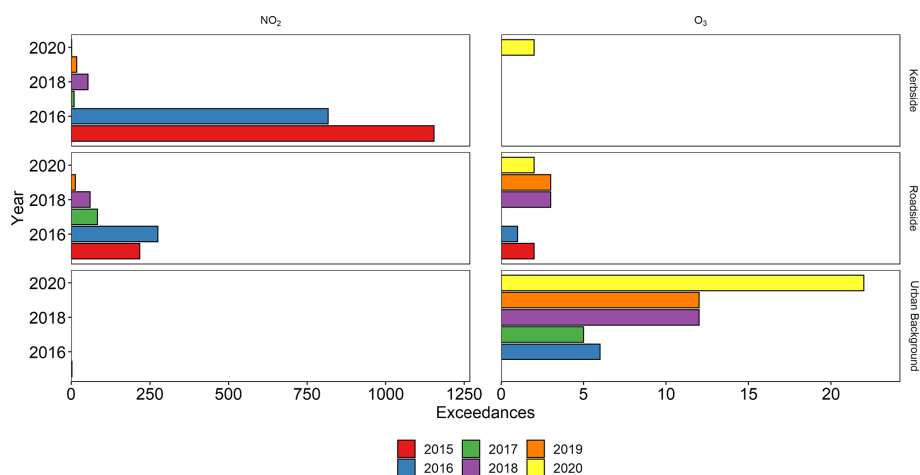
Figure 10. Exceedances of the UK air quality objectives for NO$_2$ and O$_3$ across the London Air Quality Network.

Table 2. UK air quality objectives (Defra, 2019). Note that the UK has adopted the EU NO$_2$ limit as a part of its air quality objectives but improves upon the O$_3$ obligations, where O$_3$ must not exceed 120 µg m^{-3} more than 25 times per year in a given 3 yr window (EEA, 2016).

Pollutant	Limit/ µg m^{-3}	Measured as	Allowed annual exceedances
NO$_2$	200	1 h mean	18
O$_3$	100	8 h mean	10

value for O$_3$, using a window of 8 h and a step size of 1 h. If a given calendar day saw this rolling mean exceed 100 µg m^{-3}, an exceedance was counted. Using this method, multiple exceedances (contiguous or separated in time) were only counted as one to avoid ambiguity in their definition, and therefore they can be thought of as "days when an O$_3$ exceedance occurred".

Figure 10 shows the results for the lockdown period in 2020 and comparisons to the same time period in 2015–2019. We find a general downward trend of NO$_2$ exceedances at roadside and kerbside sites in London, due to the continued reduction in NO$_x$ emissions from the vehicle fleet. At kerbside sites, the number of exceedances dropped quickly from 1154 in 2015 to only 17 in 2019. At roadside sites, exceedances dropped consistently from 275 in 2016 to 13 in 2019, with 9 of these 13 at the site London Wandsworth (Putney High Street). In 2020, up until the end of May, there was only one NO$_2$ exceedance at sites across the LAQN network, again at Putney High Street. Because we have only analysed data up until the end of May 2020, we do not know the cumulative effect on exceedances for the year or how many exceedances will breach the 18 allowed by the air quality objective. Consequently, further analysis on data collected for the whole of 2020, including the period when lockdown restric-

tions were relaxed, is required to put 2020 into the context of the previous year. As an estimate of the effect the lockdown may have on total exceedances in 2020, we replaced the number of exceedances during the lockdown period in 2019 with the number from 2020. This resulted in a 47 % decrease (34 to 18) in total exceedances of NO$_2$ at kerbside sites and a 12 % decrease (76 to 67) at roadside sites. As the effects of lockdown certainly extend beyond the end of the time period explored by this study, we would expect there to be fewer exceedances still during the remainder of 2020.

When considering any health benefits to this apparent improvement in air quality due to reduced NO$_2$, we should also consider exceedances to O$_3$ limits. The WHO has set a guideline value for ozone levels at 100 µg m^{-3} for an 8 h daily mean. Figure 10 also shows the total number of exceedances of this limit across kerbside, roadside and urban background sites in the LAQN network for March–May in all years from 2015–2020. Urban background sites have seen an increase from 6 in 2016 to 12 in 2019, followed by a drop to 18 in 2019 and an increase to 22 exceedances up until the end of May in 2020. Peak O$_3$ in the UK often occurs in June and July, so it will be necessary to analyse data from the whole year, alongside NO$_2$, in order to fully assess the importance, but it is clear that any perceived benefits of reduced NO$_2$, both during the lockdown and in a lower-NO$_x$ future, should be considered alongside any concurrent increase in O$_3$.

5 Summary and conclusions

We examined NO$_2$ and O$_3$ measurements from urban traffic and urban background sites across the UK during the COVID-19 lockdown period in 2020 (23 March–31 May). We compared data to the detrended average from the previous 5 years in order to assess how these air pollutants have changed as a result of the reduced activity caused by the nationwide lockdown. NO$_2$ decreased by an average of 48

and 40 % at urban traffic and urban background sites, respectively. This is in broad agreement with the expected reduction based on the reduction in traffic and the proportion of NO$_x$ in the UK that comes from vehicles. For O$_3$, we find that values increased on average by 11 % at urban background sites and by 48 % at the three urban traffic sites. Total O$_x$ increased by 3 % on average, suggesting the majority of the increase in O$_3$ is due to photochemical repartitioning as NO$_x$ is decreased. However there are difference across the UK, with the southern cities London, Bristol and Cardiff showing a 5 % increase in O$_x$ and Newcastle, Belfast and Glasgow showing only a slight decrease in O$_x$. Whilst anthropogenic VOCs are slightly decreased during the lockdown, we find some evidence that suggests that biogenic VOCs such as isoprene are higher due to warmer temperatures and higher UV levels across the southern UK in 2020 compared to previous years; we find no evidence to suggest that higher UV levels were due to cleaner skies related to air pollution changes due to the lockdown. Analysis of exceedances of air quality objectives in London for NO$_2$ and O$_3$ shows that, whilst there has been a decrease in exceedances of the NO$_2$ objective, this has come alongside an increase in O$_3$ exceedances. If we are to take the COVID-19 lockdown as an analogue of how air quality will respond to future reductions in emissions from vehicles (e.g. over the next 10–20 years), then observations show that there could be a corresponding increase in O$_3$, which should be considered in any air quality abatement strategy. In China, NO$_x$ reductions have led to increases in O$_3$ (Li et al., 2019a; Ma et al., 2016; Sun et al., 2016), and therefore air quality abatement strategies are being developed in order to offset this, largely by also controlling VOCs (Li et al., 2019b; Le et al., 2020). These changes are attributable to photochemical processes (e.g. the reduction in particles causing increased radiation and photochemistry); however our study shows that a large reduction in NO$_x$ directly causes an increase in O$_3$ due to a reduction in titration with NO. In addition, a warming climate may lead to increased emissions of biogenic VOCs, further adding to the O$_3$ burden. Therefore it will be vital to control anthropogenic VOCs in the UK to avoid any health gains made by the reduction of NO$_2$ being offset by O$_3$ increases.

Data availability. The AURN data are all available for public download from the UK-AIR website (https://uk-air.defra.gov.uk, last access: 15 December 2020). The LAQN data are available from the LondonAir website (https://londonair.org.uk, last access: 15 December 2020). UV-A data are available on request from Public Health England.

Supplement. The supplement related to this article is available online at: https://doi.org/10.5194/acp-20-15743-2020-supplement.

Author contributions. WSD and DPF carried out the data analysis and designed the figures. JDL and PIP wrote the manuscript with input from WSD and DPF. SEW designed and created figures and reviewed the manuscript.

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