The final editorial comment was "The figures need final checks regarding layout and readability". We have done this and uploaded the figures as separate, high resolution 'png' files in accordance with submission instructions.

We also noticed three minor errors in the text that needed correcting.

On Line 404 the sentence should read:

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..

On line 432 there was a very slight error in the number of NO_2 exceedances. This is corrected and the text now reads:

At roadside sites, exceedances dropped consistently from 275 in 2016 to 13 in 2019, with 9 of these 13 at a site in Wandsworth (Putney High Street).

On line 449 was also a very slight error in the number of O_3 exceedances. This is corrected and the text now reads:

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.

The changes are also marked in the document below

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 (23rd March—31st May, 2020, inclusively), and compare them with values from an equivalent period over the previous five years. Data are from the Automatic Urban and Rural Network (AURN) that form 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 Coronavirus SARS-CoV-2 (COVID-19) outbreak was first identified in Wuhan, China, in December 2019, and was recognised 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). 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 NO_2 and 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, e.g.

meteorology, using atmospheric chemistry transport models and weather normalisation 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) and 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 NO. Further afield, in India 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 Sao 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% decrease across the whole country, relative to a similar period in 2015-2019 (Bao and Zhang, 2020). In situ measurements in cities in northern China measurements 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 1-24th January 2020 and 26th January - 17th 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 on 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 O3 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 nitrogen dioxide (NO_2) across the UK and discuss them in context of observed changes in surface ozone (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). 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)e.g. 47±6 % comes from road transport, , with 17±5 % from domestic combustion, 15±6 % from non-road transport sources (mainly rail), and 14±10 % from energy industries and 6±2 % from industrial combustion. In recent years, there has been a pronounced reduction in NO_x emission (Defra, 2018a) that 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, and the number of ultra-low emission vehicles (e.g. electric, hybrid cars) has increased its market share from 0.59% in 2014 to achieved 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 microns ($PM_{2.5}$) 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 two 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/year in the UK, equivalent to 340,000 life years across the population in any one year and costs the UK economy between £10 billion and £20 billion/year (Royal College of Physicians, 2016). To meet the UK Government's clean air strategy (UK Government, 2019) and its commitment to achieve 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-NOx 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 that we use as a proxy for a future low-NO_x vehicle fleet. Figure 1 summarises 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 was used as a proxy for traffic counts as it is readily accessible, however for any quantitative analysis of the effect of reduction in traffic on pollution levels, real traffic counts or flow data would be required. The UK Foreign and Commonwealth Office issued a travel advisory on 28th January not to travel to mainland China. The first two UK cases of COVID-19 were confirmed on 31st January, with a third case confirmed on 6th February. As the number of cases continues to rise, the first UK death from COVID-19 was confirmed on the 5th 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 three weeks later on 23rd March, with citizens instructed to stay at home with the exception of shopping for basic necessities and 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 section 3 we describe our results for NO_2 that we put into context in section

4 with the observed changes in surface O_3, as well as comparing our results with other studies. We conclude the paper in section 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, currently consisting of 150 active sites across the UK (Figure S1 and Tables S1 and S2) 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), sulphur 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. 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 stations. 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 1,000 km^2.

The AURN network uses standardised 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/ but a brief description will be given here. Nitric oxide (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 (un-activated) 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 (PMT) 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 oxidised 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 254nm, 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 that have measurements between 2015 to the end of May 2020.

2.2 Correlative Meteorological Data

Measured meteorological data (wind direction, wind speed and temperature) is not available at most AURN sites so modelled data, based on the position of the site, from the UK Met Office unified model is used. UV-A irradiance data is taken from measurements made by the Public Health England (PHE) 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 de-seasonalise NO_2 data at each AURN site. To deseasonalise the data, we determine the climatology based on the mean annual cycle of the previous five years (from January 1st 2015 to December 31st 2019) which is then repeated to match the length of the time series, subtracted from the mean to standardise the data, and then subtracted from the original time series to produce a time series of the residuals. This five-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 calculated 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 (Figure 2) and calculated 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 to determine BAU that, for example, explicitly take into account local changes in meteorology (Grange and Carslaw, 2019) will also be subject to uncertainties, e.g. the extent which regional-scale meteorological fields can describe smaller-scale variations in atmospheric pollutants. We define the start of the UK lockdown period as the 23th 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 the 9^{th} 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^{st} 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 same period for the past five 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 five 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 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) μg 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 five years of -5.5 μg m^{-3} yr^{-1} (-6.7 % yr^{-1}), whereas eight urban sites show a small increasing trend in NO_2 between 0.1 - 0.6 μg m^{-3} yr^{-1} (0.5 – 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 (10th January – 10th March) and post (23rd March – 31st May) lockdown 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 named storms 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 five years. The six cities saw an average wind speed in 2020 of (6.5±1.2) ms^{-1}, which was 33.5% higher than the average of the previous five 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 southern and western UK. Average wind speeds across the six cities was (4.1±0.4) ms^{-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 five years (16.8%), with Bristol showing a 10% increase. The other cities all saw slight (<5%) decreases in wind speed in 2020. Typically, lower wind speed meteorological conditions are associated with higher levels of air pollution due to increased atmospheric stability and transport of pollution from mainland Europe in the UK, respectively, and so care must be taken when comparing pre and post-lockdown levels of air pollution, as described in section 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 NO_2

Measurements in 2020 from 65 urban traffic (figure S2a) and 61 urban background (figure S2b) AURN measurement sites across the UK show clear reductions in NO_2 concentrations across all sites since the lockdown. Some of these differences are due to the natural 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±2.1) $\mu g\ m^{-3}$ in NO_2 over the lockdown period compared with the same period over the previous five 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 Marylebone Road (London) 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 (Northern Ireland) is the only urban traffic site to show a mean increase in NO_2 (1.3±1.1) $\mu g\ m^{-3}$ (6.7±6.2) %. Urban background sites show a smaller mean reduction of (4.9±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±3.3) $\mu g\ m^{-3}$ was observed in 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 (Figure S3(b), there is very little correlation between the observed change in NO_2 between 2020 and the previous five years and any change in wind speed (Figure S3(a)).

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 five years, for NO_2 measurements during the lockdown period and measurements in 2020 immediately prior to the lockdown, and for NO_2 measurements immediately prior to lockdown in 2020 with the same period for the previous five years. We find that using these tests that 115 out of the 128 (89%) urban sites show a statistically significant ($p < 0.01$) difference in NO_2 between the mean observations during lockdown to the mean of the same period during the past five years. We also find 112 sites (from a possible 128 sites, 88%) show a significant difference between NO_2 measurements made in 2020 immediately prior to the lockdown to the mean of the same period from the previous five years, with urban background sites showing a -6.3±1.5 $\mu g\ m^{-3}$ change and urban traffic 9.2±1.9 $\mu g\ m^{-3}$) We attribute this difference to changes in meteorology during January and February 2020 (Figure 3), in particular wind speed, which was on average 33% higher than the average of for 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 (Figure S4) typical pre-lockdown diurnal cycles are driven by emission peaks in the morning and evening rush hours, with the evening peaks supressed due to the higher mean boundary layer that is grown 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 S5 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 section 2.3, we find that O_3 at urban background sites increased by a mean value of (7.2±2.6) μg m^{-3} during the lockdown period when compared with the previous five years equating to a percentage increase of (11±3.2) %. Leamington Spa (West Midlands) and London Hillingdon observed the largest mean increases of (21.3±2.7) μg m^{-3})(35±3.6) %) and (21.6±3.6) μg m^{-3} ((54±7.5) %) respectively. Three sites observed a decrease during the lockdown with Aberdeen seeing a large decrease in O_3 of (24.0±4.5) μg m^{-3} ((36±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 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 Marylebone Road (London) 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±5.5) %) and Birmingham A4540 Roadside (West Midlands) with an increase of (13.3±2.5) μg m^{-3} ((25±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 that 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±3.4) %) and

Aberdeen where we find the largest O_X decrease of (-27.6±0.4) µg m^{-3} ((58±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±0.4) %) at Birmingham A4540 Roadside, (-7.9±1.8) µg m^{-3} ((-11±3.1) %) at Exeter Roadside and –(20.5±4.7) µg m^{-3} ((-15±3.1) %) at London Marylebone Road.

Following our approach for NO_2, we use independent the Mann-Whitney U-test to determine the significance of changes in O_3 pre-lockdown and lockdown periods in 2020 and in the previous five 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 to 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 has been around a 75% reduction in road traffic across the UK, (using Google activity data as a proxy for traffic) (see Figure 1). According to the NAEI, road transport is estimated to make up 53% of NO_x emissions in the 1km x 1km 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, 40% at urban background – see figure S6), 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 most reduction in passenger cars and less reduction in high emitters like HGVs. There is 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 Figure S7), suggesting that the change in traffic flow near to individual sites is variable and will be to 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 (Figure 6(b)). 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 five years(Figure 6(a)). 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 – 18:00) 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 UVA at eight sites in the UK and compared data from 2020 to the mean of the previous five years (Figure S8). We find levels of UV across the UK were higher in 2020 compared to previous years, with the largest increases in 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 southern UK, some of the O_3 increase is not solely attributable to reduced NO_x, but also 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 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 that have a largely traffic source and saw a decrease of 23±5.1% and 29±6.5%, respectively, compared to previous years. At 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 is 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 / 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 shows total VOC loading in ppb and total OH reactivity for each day in 2020, with colours showing the percentage change from the previous three 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 while 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 Marylebone Road were 17% lower and the corresponding k' 15% lower than the 2017-2019 mean. At 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) isoprene data and its contribution to k' at two sites in 2019 and 2020 (the only years where reliable isoprene data is available). Observed isoprene was a factor of two higher at both 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 Marylebone Road, but at 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 2ºC higher in 2020 compared to 2019 for the lockdown period.temperature increases described above. 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 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, 52 31 roadside, and 2514 background sites for NO_2 and 1 kerbside, 87 roadside and 15 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 one-hour mean was calculated and each hour greater than 200 ug m^{-3} was counted as an exceedance. We calculated a rolling mean value for O_3, using a window of eight hours and a step size of one hour. If a given calendar day saw this rolling mean exceed 100 ug 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 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, the number of exceedances dropped quickly from 1154 in 2015 to only 17 in 2019. At roadside sites, exceedances dropped consistently from 221 275 in 20165 to 13 in 2019, with almost all9 of these remaining 13 at a site in 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 restrictions were relaxed, is required to put 2020 into context of 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 % (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 less 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-hour 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 ~~consistent~~ increase from ~~5~~ 6 in ~~2015~~ 2016 to ~~27~~ 12 in 201~~8~~9, followed by a drop to 18 in 2019 and an increase to ~~35~~ 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 (23rd March – 31st May). We compared data to the detrended average from the previous five 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 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 show 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 is all available for public download from the UK-AIR website (uk-air.defra.gov.uk). The LAQN data is available from the LondonAir website (londonair.org.uk). UVA data is available on request from Public Health England.

Author contribution

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.

Competing interests

The authors declare no conflict of interest.

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Figures

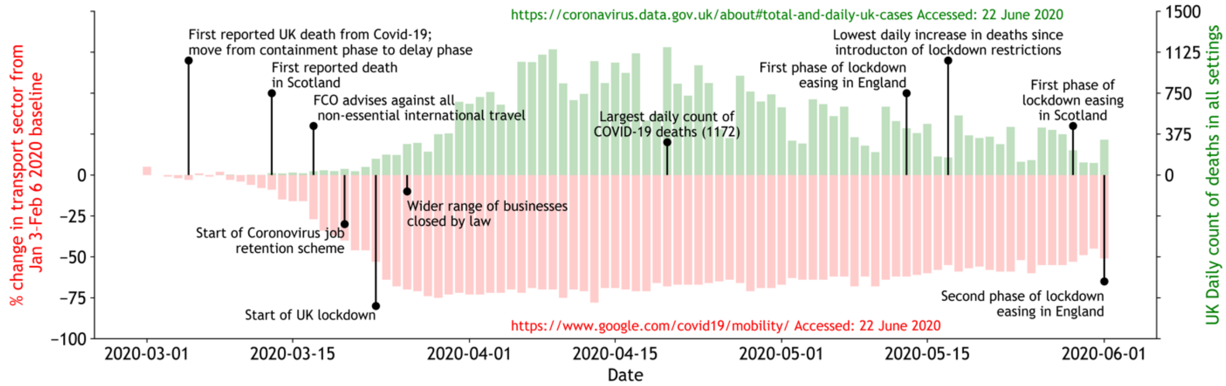


Figure 1: Schematic of the timelines involved with daily mortality values attributed by the UK government to COVID-19, and changes in mobility from the transport sector (inferred from Google location data on smartphones) compared to a reference period (3 Jan- 6 Feb, 2020) before the lockdown period. Also included are key dates that describe the run up and evolution of the UK lockdown. Data acknowledgements are shown inset of the plot.

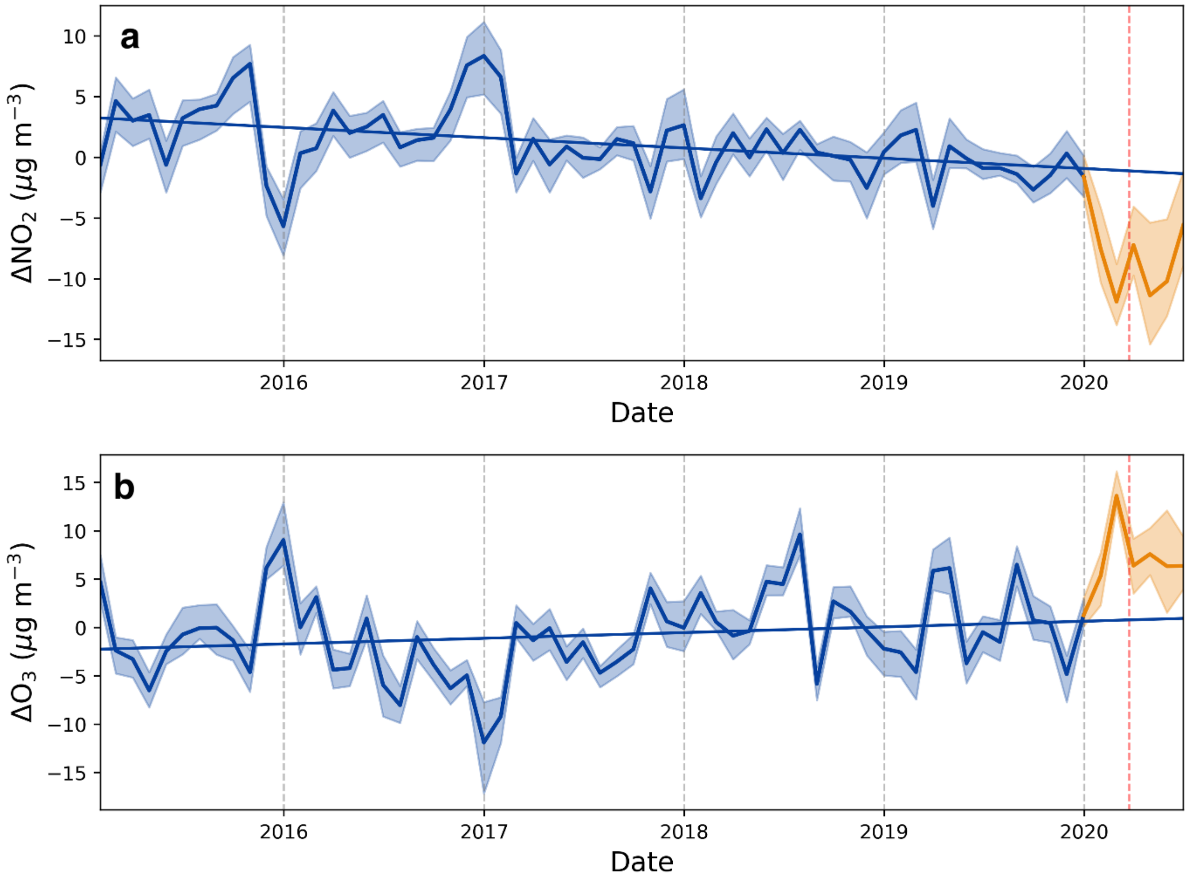


Figure 2: Mean relative change of deseasonalized UK values of a) NO_2 and b) O_3 for all urban background and traffic sites from 2015 to 2020, with the mean 2015 -2019 trend superimposed. Data from 2020 is shown in orange, with the red dashed line denoting the start of the lockdown on 23th March 2020. The 25^{th} – 75^{th} percentile is shown by the shaded area.

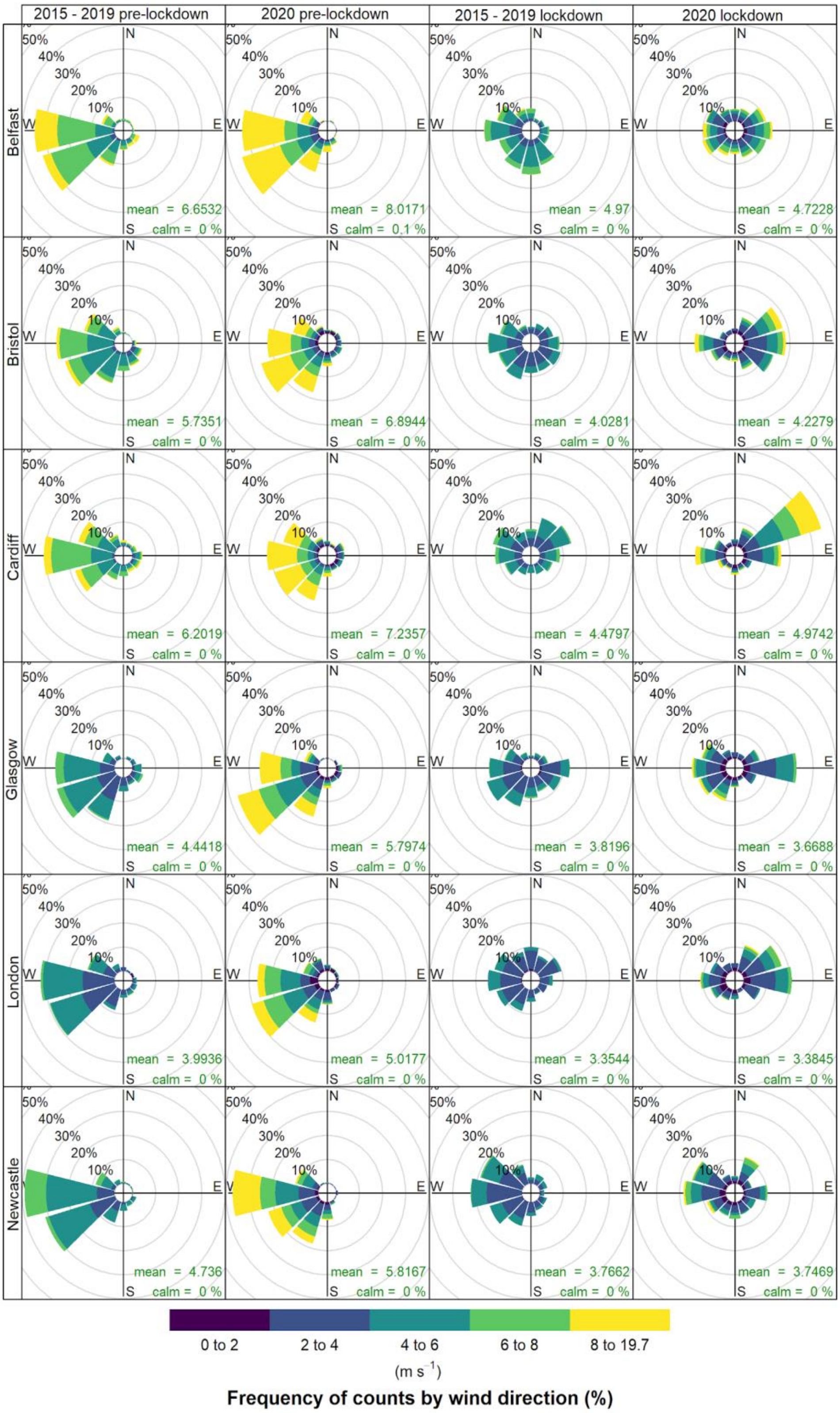


Figure 3: Average wind roses for 6 cities for pre and post lockdown period and lockdown period 2015-2019 and 2020. Data used is modelled using the UK Met Office Unified Model.

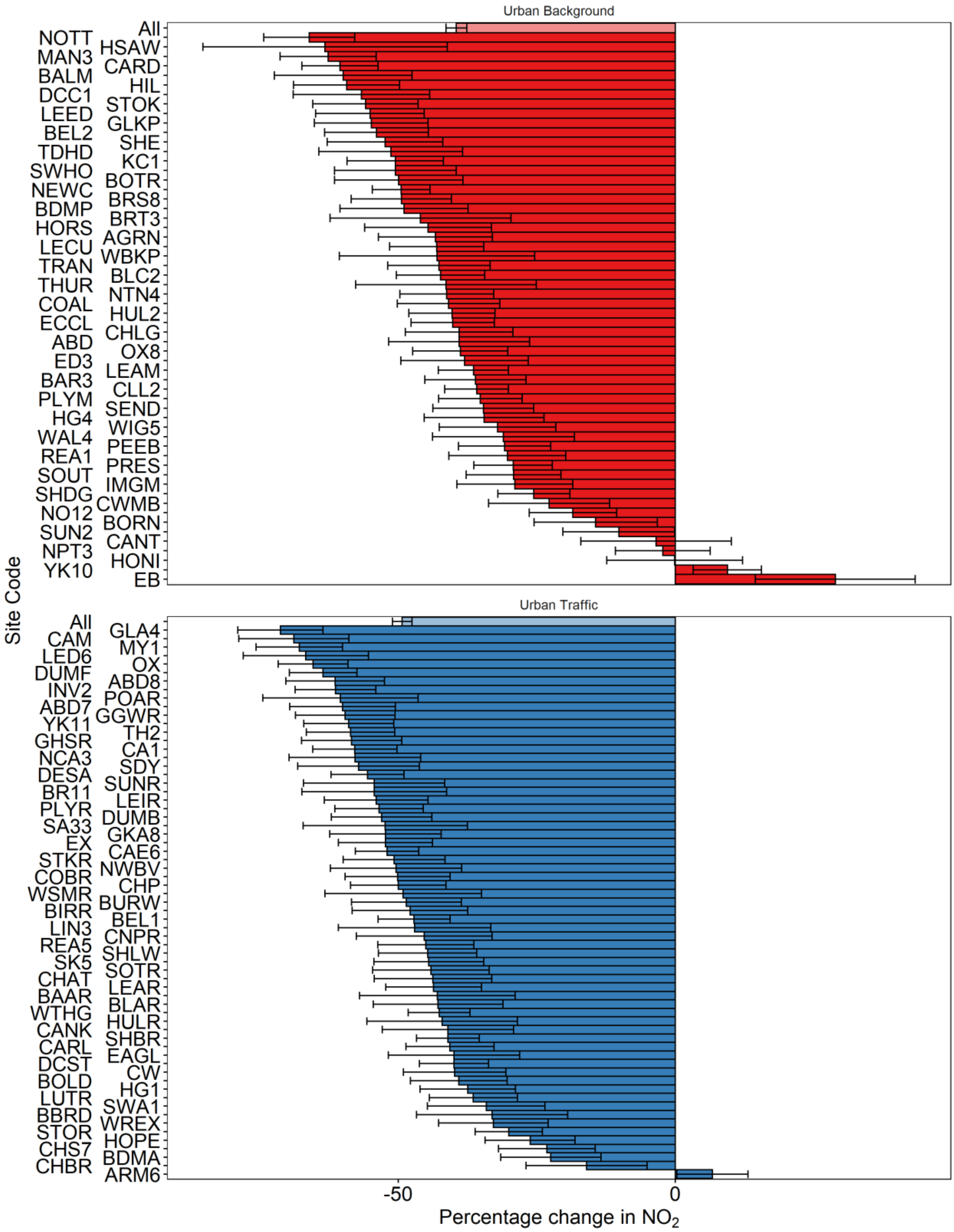
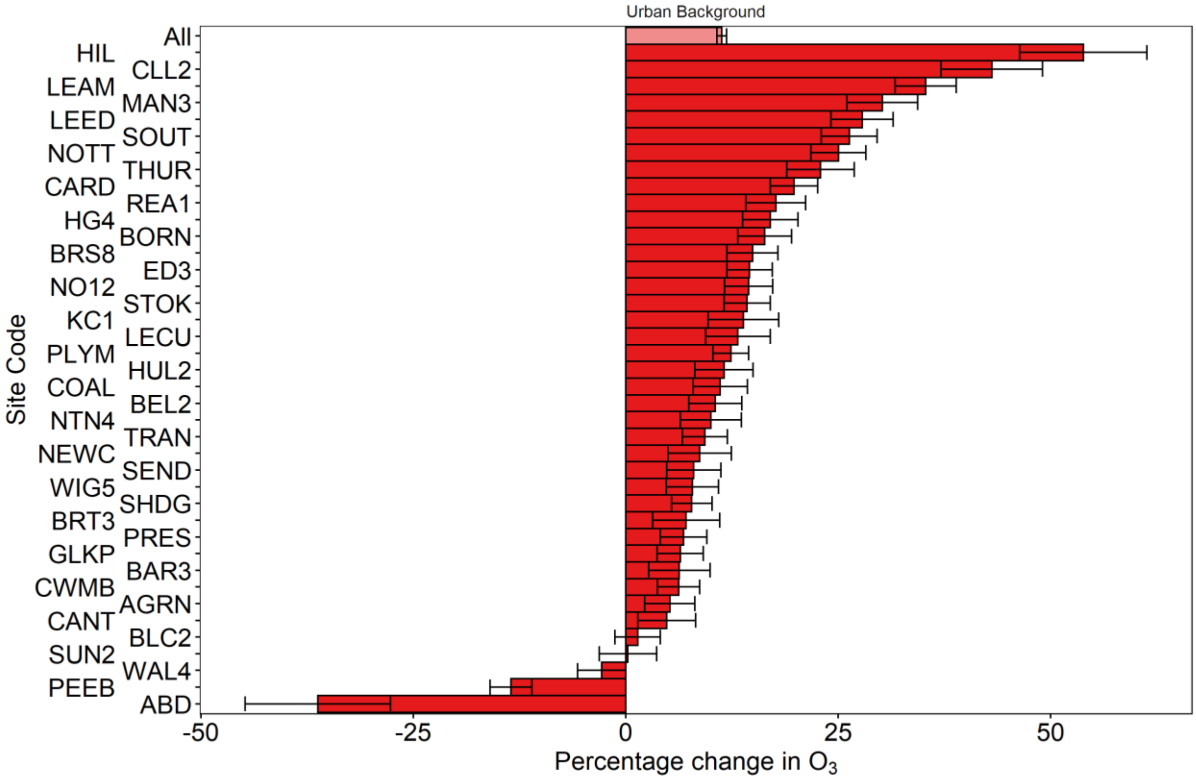


Figure 4: Percentage change in NO_2 at all urban background and urban traffic sites for the lockdown period (23rd March – 31st May) in 2020 compared to the same period averaged across the previous 5 years, after removing site-dependent trends. The lighter coloured bar at the top shows the average of all sites. Site acronyms can be found in the SI.



675

Figure 5: Percentage change in O_3 at all urban background sites for the lockdown period (23rd March – 31st 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 SI.

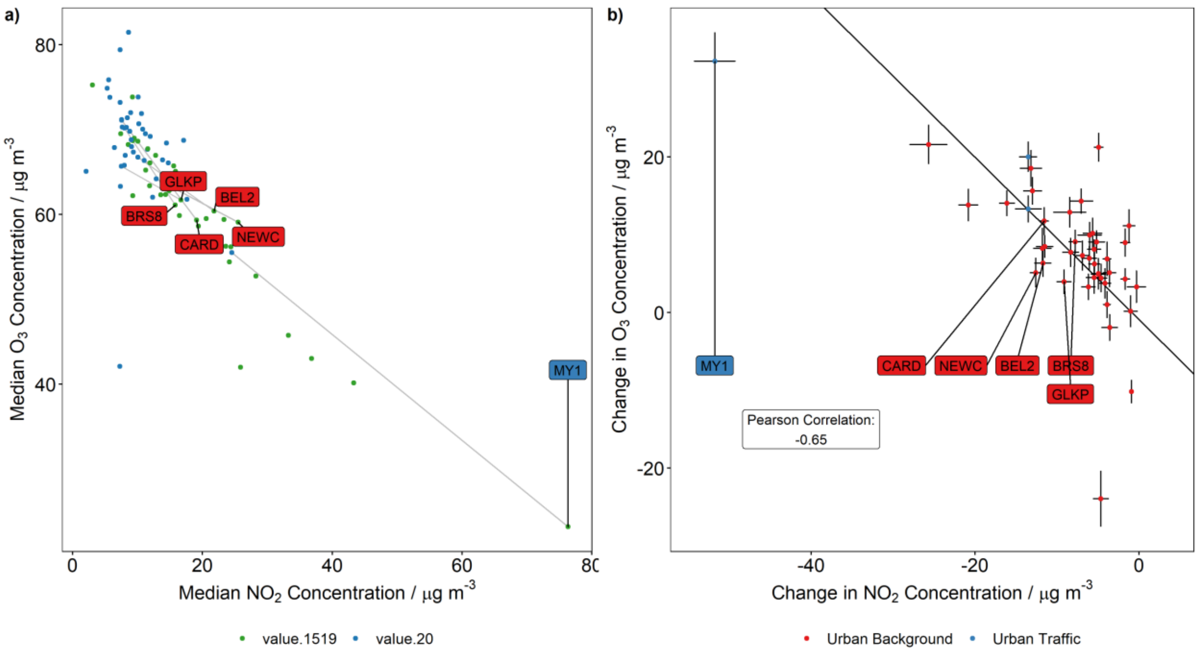


Figure 6: (a) shows median O_3 concentration plotted against median daily NO_2 concentration for each site. 2020 and the average of 2015-2019 data are coloured blue and green respectively. (b) shows change in O_3 concentration between 2020 and the average of 2015-2019 plotted against the change in NO_2 concentration for the same time period. Labelled are sites from six cities across the UK.

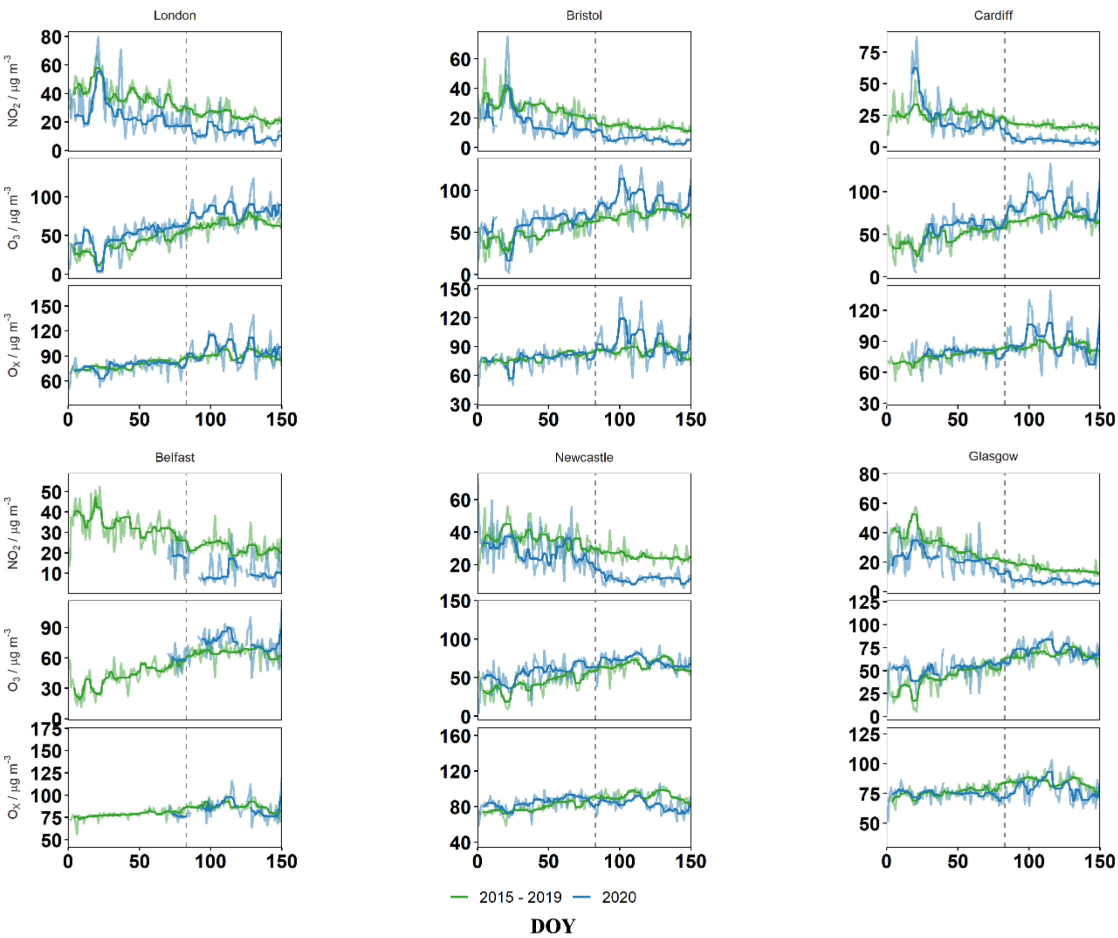


Figure 7: Daily median time series of NO_2, O_3 and O_x (NO_2 + O_3) for 2020 and the average of 2015-2019 at urban background sites in 6 cities representing a geographical and political spread across the UK. The thick line represent 7 day rolling mean. The hashed grey line indicates the start of the lockdown period.

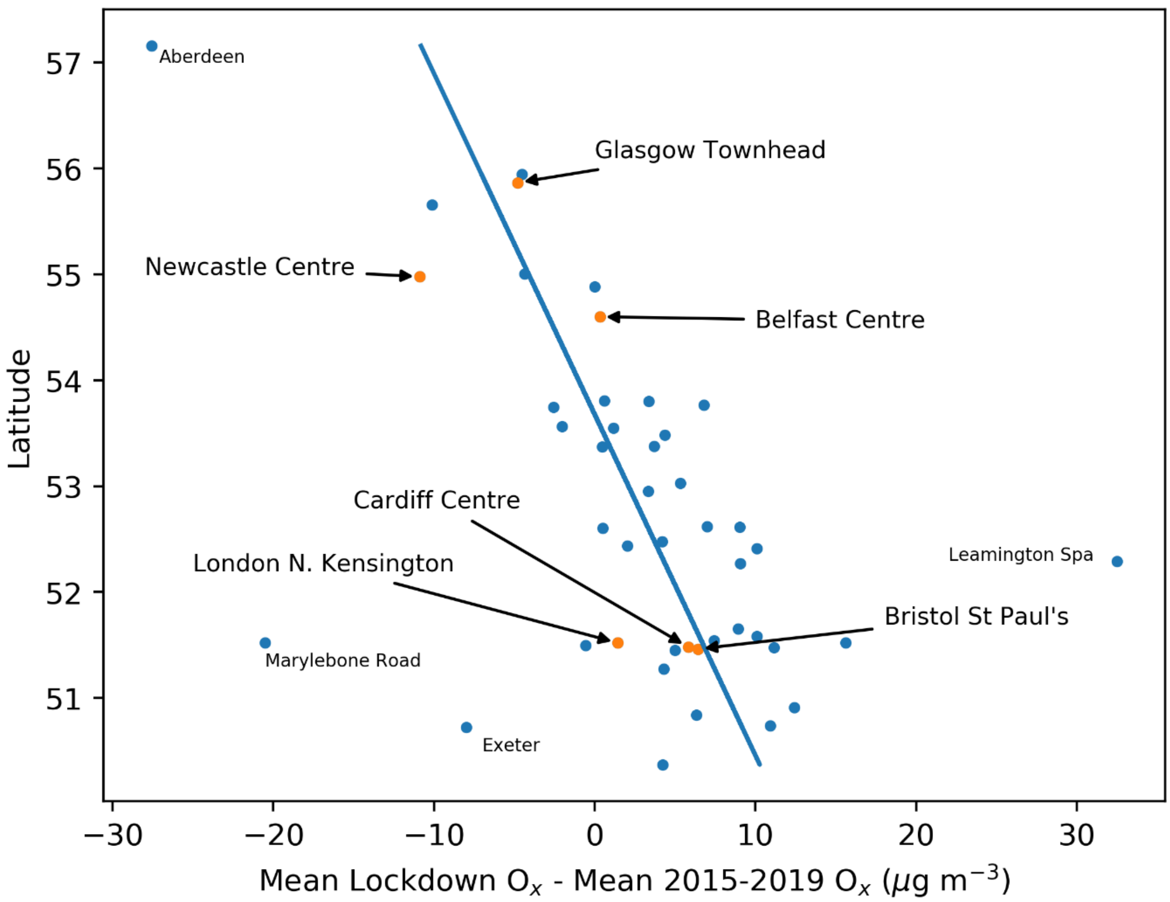


Figure 8: Difference in mean (a) O_x ($\mu g\ m^{-3}$) and between the lockdown period and the detrended mean of the same period from 2015 to 2019 for urban background sites as a function of latitude. Sites examined in figure 6 are highlighted in orange.

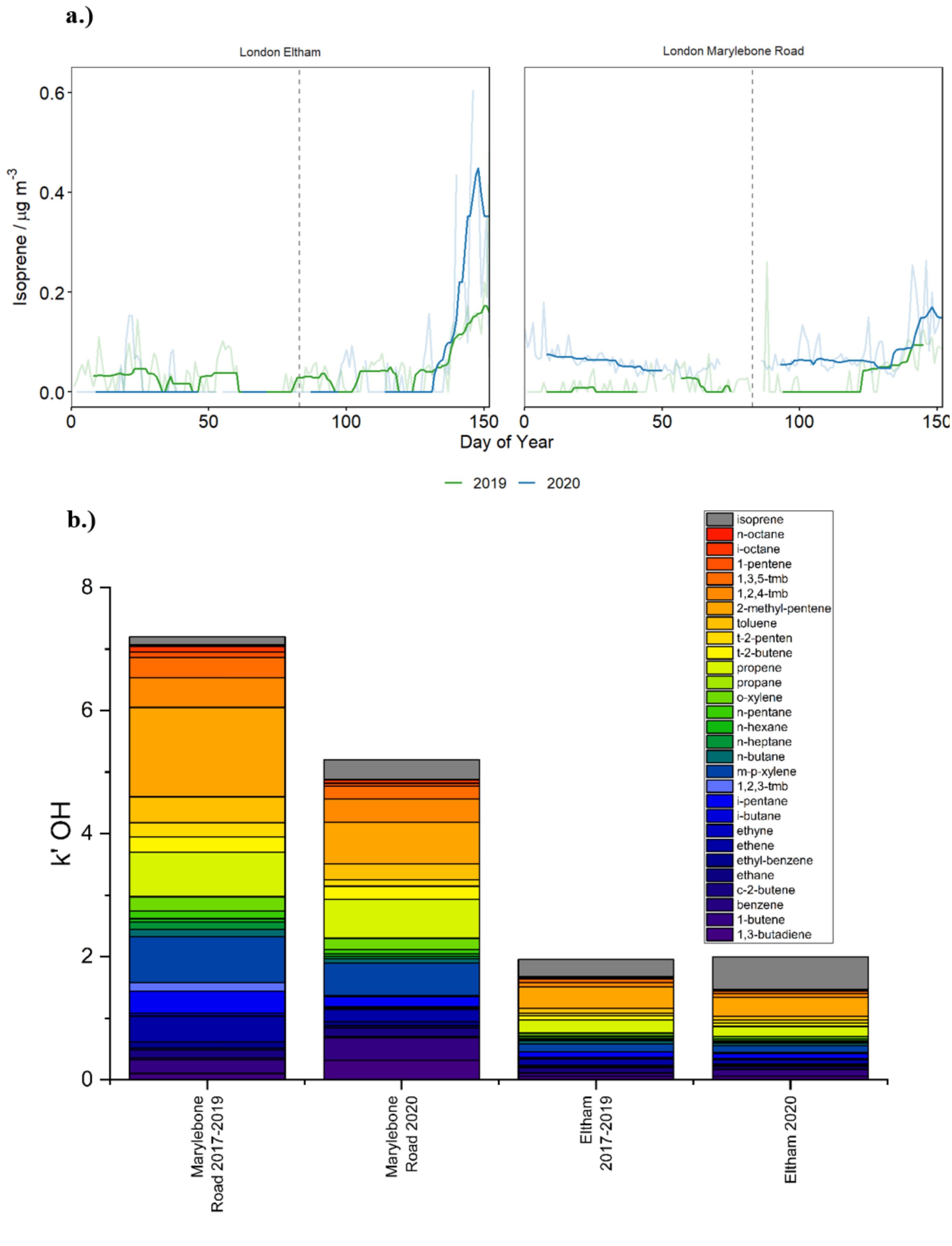


Figure 9: (a) Levels of isoprene at the London Eltham (urban background) and London Marylebone Road (urban traffic) sites in 2020 and 2019. (b) shows the contribution of isoprene (grey slice) and other VOCS to total OH reactivity (k') for each site for 19 compared to 2020.

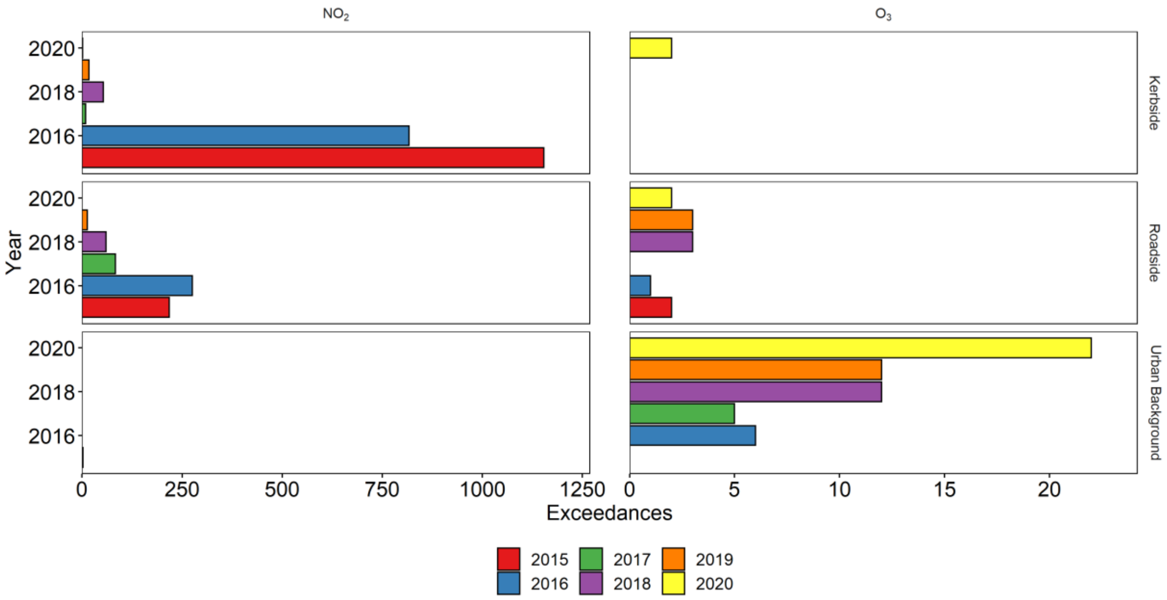


Figure 10: Exceedances of the UK air quality objectives for NO_2 and O_3 across the London Air Quality Network.

Tables

Table 1: Summary of previous measurements.

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	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 30th April 2020	https://uk-air.defra.gov.uk/library/reports.php?report_id=1005
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%) Nice	+24% Nice		Sicard et al. (2020)
Italy	-46% (-69%) Rome, -30% (-53%) Turin	+14% Rome, +27% 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.	--	Used a ML approach to determine deviation from BAU NO2, trained using 2017-2019 data from background and traffic surface AQ monitoring sites. Study considers all three phases of lockdown up to 24th April 2020.	Petetin et al. (2020)
Spain	-69% Valencia	+2.4% 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	-24-33% compared to 2019	--	Study over Rio de Janeiro used data from automatic monitoring station run by Municipal Department of the Environment. Study period is from March 2nd to April 16th 2020, with lockdown on 23rd March 2020.	Dantas et al. (2020)
Brazil	-54% (-77%) on urban roads	+30%	Study over Sao Paulo using three in situ AQ sites. Changes relative to similar periods from previous five-year mean.	Nakada and Urban (2020)
China	-25%	--	Study of data from 44 cities in northern China from 1st January to 21st March 2020. Lockdowns started on 23rd January in Wuhan with other cities following soon afterwards.	Bao and Zhang (2020)

			Linear regression was used to determine BAU.	
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 1-24th January and 26th January-17th February 2020.	Huang et al. (2020)
China	-71.9%	+25.1%	TROMPI measurements over Eastern China and Wuhan, compared to previous 5 years.	Le et al. (2020)
India	-18% from previous 5-year mean; over New Delhi -54%	--	Used satellite observations of NO2 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 compare pre-lockdown period 3rd-24th March and during lockdown period 24th March-14th April 2020.	Mahato et al. (2020)
Kazakhstan	-35%	+15%	Study over Akmaty using data from a similar previous period from 2018-2019. Data from the Airkaz publics AQ monitoring network.	Kerimray et al. (2020)
Morocco	-96%	--	Study over Salé City, Morrocco using urban in situ data.	Otmani et al. (2020)
USA	-30%	Weak, inconsistent response	Used EPA data.	Bekbulat et al. (2020)

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 ug m^{-3} more than 25 times per year in a given 3 year window (EEA, 2016).

Pollutant	Limit / ug m^{-3}	Measured as	Allowed annual exceedances
NO_2	200	1 hour mean	18
O_3	100	8 hour mean	10