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The effect of COVID-19 lockdown on atmospheric total particle numbers, nanoparticle numbers and mass concentrations in the UK

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ABSTRACT

The main aim of the COVID-19 lockdown was to curtail the person-to-person transmission of COVID-19. However, it also acted as an air quality intervention. The effect of the lockdown has been extensively analysed on NO_2, O_3, PM_{10} and $PM_{2.5}$, however, little has been done on how total (TPN) and nanoparticle numbers (NPN) have been affected by the lockdown. This paper quantifies the effect of the lockdown on TPN and NPN in the UK, and compares how the effect varies between rural, urban background and traffic sites. Furthermore, the effect on particle numbers is compared with particle mass concentrations, mainly PM_{10} and $PM_{2.5}$. Two approaches are used: (a) comparing measured levels of the pollutants in 2019 with 2020 during the lockdown periods; and (b) comparing the predictions of machine learning with measured concentrations using business as usual (BAU) scenario during the lockdown period. P_{100} (particle size $\leq$100 nm) increased by 39% at Chilbolton Observatory (CHO) and decreased by 13% and 14% at London Honor Oak Park (LHO) and London Marylebone Road (LMR), respectively. Particles from 101 to 200 nm (P_{200}) showed a similar trend to P_{100}, however, average levels of particles 201–605 nm (P_{605}) decreased at all sites. TPN, PM_{10} and $PM_{2.5}$ concentrations decreased at LMR and LHO sites. Estimated PM_{10}, $PM_{2.5}$ and TPN decreased at all three sites, however, the amount of change varied from site to site. Pollutant concentrations increased back the to pre-pandemic levels, suggesting more sustainable interventions for permanent air quality improvement.

1. Introduction

Air pollution is a serious environmental issue affecting human health globally, especially in large urban areas. According to Lelieveld et al. (2020), air pollution contributed to 8.8 million deaths worldwide in 2015. Air pollution is reported to affect lung function, cause respiratory infections, aggravate asthma, and result in ischaemic heart disease, stroke and neurodegenerative conditions (WHO, 2021; Landrigan, 2017). Long-term exposure to air pollution affects people of all ages, however, it is particularly harmful to the young, elderly and people with existing health conditions (Khallaf, 2011).

To decrease the adverse effect of air pollution on human health, various interventions are implemented from time to time to cut emissions and reduce atmospheric concentrations of air pollutants in urban areas. Normally, in the UK air quality interventions are implemented at the scales of local authorities. According to the Department for Environment, Food and Rural Affairs (DEFRA, 2020), an air quality intervention is a deliberate measure aimed at improving air quality. An intervention may be primarily aiming at other outcomes but it could also indirectly affect air pollution. The COVID-19 pandemic provided an opportunity for such an intervention. To curtail the person-to-person transmission of COVID-19, the UK government implemented a lockdown on March 26, 2020, which acted as a national intervention for air quality in the UK. The lockdown restrictions included: (a) people to stay at home, except for limited purposes; (b) closing of all non-essential businesses and public venues (including community centres, libraries, and places of worship); and (c) stopping all gatherings of more than two people in public (Jephcote et al., 2021). Due to restrictions on mobility road traffic decreased abruptly and monthly-average daily traffic counts on English A-roads and motorways in April 2020 were down by 69% compared with April 2019, with a 74% reduction in light and a 35% reduction in heavy vehicles (coaches and HGVs) (Jephcote et al., 2021). Therefore, the COVID-19 lockdown improved air quality in major cities in the UK and elsewhere (e.g., Shi et al., 2021; Dacre et al., 2020; Munir

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et al., 2021; Potts et al., 2021; Ropkins and Tate, 2021).

The focus of most of the studies analysing the effect of COVID-19 lockdown on air quality has been on NO_2. Brown et al. (2021) analysed the levels of NO_2 concentrations at air quality monitoring stations within a distance of 500 m of nurseries, primary schools, secondary schools and colleges in England. They reported that NO_2 concentrations had decreased during lockdown by 35% and 41% in the UK at background and traffic sites, respectively. According to Dacre et al. (2020) the largest decreases in NO_2 concentrations associated with reduced emissions were found in urban traffic (27%) and urban background sites (14%), where NO_2 concentrations were representative of local areas and thus dominated by local reduction in emissions from vehicles. Dacre et al. (2020) also reported that NO_2 levels even increased at some rural and remote sites, where NO_2 measurements were representative of large areas and thus dominated by the regional advection of secondary NO_2 from Europe. Mohajeri et al. (2021) analysed the concentrations of NO_2 and $PM_{2.5}$ in four cities in the UK and reported that NO_2 levels decreased by 21% in Greater London, 19% in Cardiff, 27% in Belfast and 41% in Edinburgh. The levels of $PM_{2.5}$ increased by 7% in Greater London, and decreased by 1% in Cardiff, by 15% in Edinburgh, and by 14% in Belfast. The reported reduction was greater in NO_2 than in $PM_{2.5}$ levels, which was probably because $PM_{2.5}$ was also affected by non-exhaust regional sources. Also, the lockdown period was accompanied by a period of hot dry weather and easterly winds, which was likely to increase resuspension and import of PM from Europe (Munir et al., 2021). They attributed the reductions in pollutant levels to the mobility restrictions imposed by the COVID-19 lockdowns. Similarly, Higham et al. (2021) reported that NO_2 concentration decreased by about 50%, whereas O_3 concentration increased by 10% during the lockdown period. Jephcote et al. (2021) analysed measurements from 129 monitoring stations in the UK and reported mean reductions of 38.3% in NO_2 and 16.5% in $PM_{2.5}$, whereas a positive gain of 7.6% was reported in O_3 concentrations. According to their analysis, the reduction in NO_2 and $PM_{2.5}$ concentrations was largest at urban traffic sites and modest at the background and rural sites across the UK.

Solberg et al. (2021) analysed the effect of the lockdown measures on NO_2 in Europe and reported the largest NO_2 reductions in Spain, France, Italy, Great Britain and Portugal and the smallest in eastern countries (Poland and Hungary). In addition to NO_2, several authors have studied the lockdown effect on ground levels ozone (O_3) and $PM_{2.5}$. Previously, Dumka et al. (2020), Jain and Sharma (2020), Ganguly et al., (2021), and Kumar et al. (2020) analysed the effect of lockdown on several air pollutants including PM_{10} and $PM_{2.5}$ in India and reported considerable reduction in their concentrations. Similarly, Donzelli et al. (2021) analysed the effect of lockdown on particulate matter in Italy, Grivas et al. (2020) in Greece, Tobías et al. (2020) in Spain, and Shi et al. (2021) compared the effect of lockdown in Italy, Spain, UK, France and Germany.

Although several studies have analysed mass concentrations of PM_{10} and $PM_{2.5}$, no study was found in the UK that focused on the effect of COVID-19 lockdown on total particle numbers (TPN) and nanoparticle numbers (NPN). This is probably due to two main reasons: (a) the data availability problem – hourly concentrations of gaseous pollutants (e.g., NO_2, NO, CO, and O_3) and mass concentrations of PM_{10} and $PM_{2.5}$ are available abundantly throughout the UK, however, the data of TPN and NPN is very limited. Hourly data of TPN and monthly data of NPN were only available from three sites for the lockdown period in the UK. (b) According to the UK Air Quality Standards Regulations 2010, NO_2, O_3, PM_{10} and $PM_{2.5}$ are among the regulated outdoor air pollutants, whereas TPN and NPN are not regulated. Therefore, measurements of particle number concentration are not mandatory. Although evidences suggest that particle number concentrations are probably more relevant from public health perspectives (AQEG, 2012). Nanoparticles (particle diameter <100 nm), also known as ultrafine particles are smaller in size with a much larger surface area per unit mass than the fine and course particles ($PM_{2.5}$ and PM_{10}). The human body inflammatory response to particles has a strong correlation with particle surface area, rather than with particle mass (Sager and Castranova, 2009). Nanoparticles can pass through the lungs alveolar membrane and reach the cardiovascular system, where they may cause toxic effects (Oberdorster et al., 2004). Therefore, nanoparticles are considered more harmful to human health due to their large surface area and tiny size. Unlike $PM_{2.5}$ and PM_{10}, the mass of a sample of nanoparticle is too small to be measured accurately by normal PM measuring methods. Ultrafine particles are, therefore, not generally measured as a mass concentration "$PM_{0.1}$" in μg/m^3 and are rather typically characterised by their particle number concentrations or particle number counts in units of particles per cubic centimetre (particles/cm^3) or p/cm^3 or ppcm^{-3}). The main objective of this paper are: (a) To quantify the effect of COVID-19 lockdown on TPN and NPN in the UK. (b) Compare the effect of COVID-19 lockdown on NPN and TPN with PM_{10} and $PM_{2.5}$. (c) How the effect of COVID-19 lockdown on particulate matter varies between rural, urban background and traffic sites. In this study, we used two main approaches: (a) comparing measured levels of PM_{10}, $PM_{2.5}$, TPN and NPN in 2019 with 2020 for the months when COVID-19 lockdown was in place; and (b) Employing a machine learning approach, which was used to predict the levels of these pollutants for the lockdown period in Business As Usual (BAU) scenario, and the predicted concentrations were compared with the measured levels. The model predictions are basically weather corrected pollutant concentrations, which are important for understanding the real change caused by changed in emission after removing the effect due to changes in meteorological conditions.

2. Methodology

The main aim of this paper is to analyse the effect of COVID-19 lockdown on NPN (particles/cm^3), TPN (particles/cm^3) and particle mass concentration of PM_{10} (μg/m^3) and $PM_{2.5}$ (μg/m^3) at several air quality monitoring stations using measured and weather corrected concentrations. Meteorological data of several parameters were used from the air quality monitoring stations and from London Heathrow Airport meteorological station. The meteorological parameters used were: air temperature (temp, ^oC), wind speed (ws, m/s), wind direction (wd, degrees from the north), relative humidity (rh, %), atmospheric pressure (P, millibars), visibility (vis, m), precipitation (precip, mm), ceiling height (m) and dew point (dp, ^oC). Hour of the day (hour), day of the week (wday) and day of the year (yday) were also used to account for diurnal, weekly and seasonal variations. Julian day was used in the model to account for long term change in pollutant levels.

2.1. Air quality and meteorology data

Data of NPN was available only in monthly resolution from five air quality monitoring stations: Chilbolton Observatory, Harwell, London Honor Oak Park, London N. Kensington, and London Marylebone road. Hourly data of TPN was also available from these five sites plus Birmingham Tyburn as shown in Table 1. However, three of the sites, namely, Birmingham Tyburn, Harwell and London N. Kensington did not have data of both NPN and TPN for 2019 and 2020, and therefore were not considered for further analysis in this paper. Hourly data of mass concentrations of PM_{10} and $PM_{2.5}$ were available for over 60 sites in the UK. All these sites also provide modelled temperature, modelled wind speed and modelled wind direction data derived from the WRF model. However, PM_{10} and $PM_{2.5}$ data were only analysed for the sites where NPN and TPN data was available. Data of the other weather parameters (rh, P, dp, precip, vis, and ceiling height) was obtained from the London Heathrow Airport (51.479, −0.449, 25 m above mean sea level (amsl)). More details of the monitoring sites are provided in Table 1. Fig. 1 shows the air quality and meteorological stations from which data was used in this study.

Table 1
Air quality monitoring stations, their environmental types and data availability. NPN, TPN, $PM_{2.5}$, and PM_{10} stand for nanoparticle numbers, total particle numbers, particle of 2.5-μm diameter and particle of 10-μm diameters, respectively.

Site (lat, long)	Environment type	Pollutants monitored	data availability
Chilbolton Observatory (51.149617, −1.438228)	Rural Background	NPN, TPN, $PM_{2.5}$, PM_{10}	2016–2020
Birmingham Tyburn (52.512194, −1.830861)	Urban Background	TPN, $PM_{2.5}$, PM_{10}	2010–2013
Harwell (51.571078, −1.325283)	Rural Background	NPN, TPN, $M_{2.5}$, PM_{10}	2010–2015
London Honor Oak Park (51.449674, −0.037418)	Urban Background	NPN, TPN, $M_{2.5}$, PM_{10}	2018–2020
London Marylebone Road (51.522530, −0.154611)	Urban traffic	NPN, TPN, $M_{2.5}$, PM_{10}	2010–2020
London N. Kensington (51.521050, −0.213492)	Urban Background	NPN, TPN, $M_{2.5}$, PM_{10}	2010–2018
Heathrow airport (51.479, −0.449)	Airport (25 m amsl)	Meteorological data	1948 - to date

2.2. Measurement techniques

Hourly TPN concentrations were measured using condensation particle counters (CPCs), TSI model 3772-CEN at Chilbolton, Honor Oak Park and Marylebone Road (NPL, 2021; AQEG, 2018). These CPCs are sensitive to particles from about 7 nm up to several μm in size and have a concentration measurement range from zero to 50,000 particles/cm^{-3}. The TSI model 3773-CEN has been developed to comply with the requirements of CEN/TS 16976:2016. At all concentrations, each particle is counted individually. Particle size distributions were measured using a Scanning Mobility Particle Sizer (SMPS). This consists of a CPC (TSI model 3775) combined with an electrostatic classifier (TSI model 3080). Reference equivalent method Filter Dynamic Measurement System (FDMS) was used for measuring PM_{10} and $PM_{2.5}$ at these sites, which is allowed for regulatory purposes (AQEG, 2012).

2.3. Data analysis and modelling

2.3.1. Nanoparticles, total particle numbers and mass concentrations

Monthly NPN data were downloaded from the three air quality monitoring stations: London Marylebone Road, London Honor Oak Park, and Chilbolton Observatory (Fig. 1). NPN were measured in the particle size diameter (nm) of 16.6, 17.8, 19.2, 20.6, 22.1, 23.7, 25.4, 27.4, 29.5, 31.7, 34.9, 36.6, 39.3, 42.2, 45.3, 48.7, 52.4, 56.3, 60.5, 65.0, 69.8, 75.1, 80.6, 86.7, 93.1, 107.5, 115.5, 124.1133.4, 143.3, 154.0, 165.5, 177.9, 191.2, 205.4, 220.7, 237.2, 254.9, 273.9, 294.3, 316.3, 339.9, 365.3, 392.5, 421.8, 453.3, 487.1, 523.4, 562.4, and 604.3. For convenience the different size ranges were aggregated into three main groups: (1) particle size ≤100 nm (P_{100}), (2) particle size between 101 and 200 nm (P_{200}), and (3) particle size between 201 and 605 nm (P_{605}). As given in Table 2, the number of particles was greatest in P_{100} range at all three sites with mean levels (particles/cm^3) of 8236, 3291 and 2156 at London Marylebone road, London Honor Oak Park and Chilbolton Observatory, respectively. The mean P_{200} ranged from 399 to 972 and P_{605} ranged from 121 to 287 (Table 2). This clearly shows that most of the particles exist in the smaller particle size ($P_{100)}$. A summary of the NPN, TPN, PM_{10} and $PM_{2.5}$ is presented in Table 2, showing mean

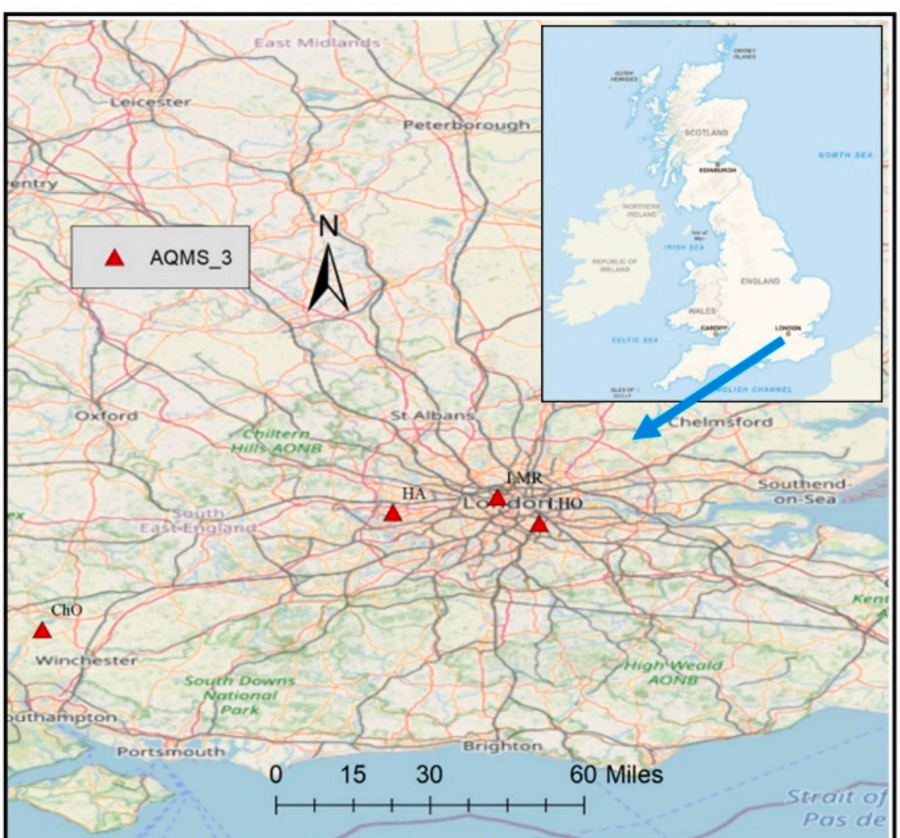

Fig. 1. Location of the air quality and meteorology monitoring stations. Here HA stands for Heathrow Airport, LMR stands for London Marylebone Road, LHO stands for London Honor Oak Park, and ChO stands for Chilbolton Observatory.

Table 2
A summary of the nanoparticle numbers (NPN) (particles/cm^3), total particle numbers (TPN) (particles/cm^3), PM_{10} (μg/m^3) and $PM_{2.5}$ (μg/m^3) for 2019 at the three sites, where LMR stands for London Marylebone Road, LHO for London Honor Oak Park, and CHO for Chilbolton Observatory. The values are mean levels and their standard deviations.

Pollutant	LMR	LHO	CHO
TPN	28,505 ± 12,468	10,613 ± 5509	4979 ± 3290
PM_{10}	22.24 ± 12.56	14.67 ± 11.86	11.89 ± 10.06
$PM_{2.5}$	14.35 ± 10.96	9.90 ± 9.97	8.03 ± 7.90
P_{100}	8236 ± 1801	3291 ± 784	2156 ± 624
P_{200}	972 ± 303	518 ± 182	399 ± 157
P_{605}	287 ± 100	181 ± 83	121 ± 57

concentrations along with their standard deviations for 2019. Concentrations of all three pollutants were highest at London Marylebone Road, followed by London Honor Oak Park. The lowest levels were observed at the Chilbolton Observatory site. London Marylebone road is a busy urban traffic (roadside) site, London Honor Oak Park is an urban background site, and Chilbolton Observatory is a rural background site, which explains why particles pollution was highest at London Marylebone road and lowest at the Chilbolton Observatory site.

2.3.2. Machine learning techniques

Machine learning approaches are frequently used to evaluate the effect of different interventions on air quality in urban areas (Jephcote et al., 2021; Solberg et al., 2021). Since the beginning of the COVID-19 pandemic in late 2019, numerous studies using machine learning approaches have been published, which assessed the effect of the lockdown on air quality in urban areas in the UK and worldwide (e.g., Jephcote et al., 2021; Solberg et al., 2021; Shi et al., 2021; Collivignarelli et al., 2020). Several researchers have preferred to employ Generalise Additive Model (GAM) for such analysis as it is an interpretable supervised machine learning technique, easy to apply, and able to address the nonlinear association between the response and predictor variables (Wood, 2006, 2020; Hastie and Tibshirani, 1990). The performance of GAM is comparable to any other advanced machine learning technique, for example, Boosted Regression Trees, Random Forest and neural network (Solberg et al., 2021; Ropkins and Tate, 2021; Munir et al., 2021; Carslaw et al., 2007). Therefore, here we employed a GAM to assess how the COVID-19 lockdown has affected the levels of TPN, PM_{10} and $PM_{2.5}$ in the UK. No hourly data of NPN was available, therefore, we could not model the levels of NPN.

2.3.3. Model development and validation

To model PM_{10}, $PM_{2.5}$ and TPN, here we used several predictors including both meteorological and temporal parameters, following the methodology of the previously published papers (e.g., Jephcote et al., 2021; Solberg et al., 2021; Shi et al., 2021). Meteorological parameters used in the models were temperature (temp, ^{o}C), wind speed (ws, m/s), win direction (wd, degree from the north), relative humidity (rh,%), atmospheric pressure (P, millibars), visibility (vis, m), dew point (dp, ^{o}C), precipitation (precip, mm), and ceiling height (ceil_hgt, m), which is the height above ground level of the lowest cloud or any other obscuring phenomena layer. Temporal parameters were hour of the day (hr), day of the week (dweek), and day of the year (yday) to represent daily, weekly and annual variations in pollutant levels. Julian day was included to account for long-term temporal trend.

It should be noted that ws, wd and temp data was available from the air quality monitoring stations, whereas the data of the other meteorological parameters were obtained from the Heathrow airport meteorological station.

$$\text{GAM}(PM_{10} \sim s1(ws) + s2(wd) + s3(temp) + s4(P) + s5(rh) + s6(vis) + s7(dp) + s8(precip) + s9(ceil\_hgt) + s10(hour) + s11(dweek) + s12(dyear) + s13(jday)) \quad (1)$$

$$\text{GAM}(PM_{2.5} \sim s1(ws) + s2(wd) + s3(temp) + s4(P) + s5(rh) + s6(vis) + s7(dp) + s8(precip) + s9(ceil\_hgt) + s10(hour) + s11(dweek) + s12(dyear) + s13(jday)) \quad (2)$$

$$\text{GAM}(TPN \sim s1(ws) + s2(wd) + s3(temp) + s4(P) + s5(rh) + s6(vis) + s7(dp) + s8(precip) + s9(ceil\_hgt) + s10(hour) + s11(dweek) + s12(dyear) + s13(jday)) \quad (3)$$

The above three GAM models (1, 2 and 3) are used to model PM_{10}, $PM_{2.5}$ and TPN, respectively using several predictors. In equations (1)–(3), the terms s1 to s13 are the smoothing nonparametric functions, which relate the modelled variables with the explanatory variables. The models were fitted on randomly selected 75% training data and validated on 25% randomly selected testing data from hourly measurements of 2018–2019. Correlation coefficients (r - values) and Root Mean Squared Error (RMSE) for both trained (fitted) and cross-validated models were calculated for assessing the model goodness of fit.

2.3.4. Modelling BAU scenario

For predicting the levels of PM_{10}, $PM_{2.5}$ and TPN in the BAU scenario for the lockdown period, the models were fitted on 2018–2019 data and then used to predict these pollutants for March, April and May 2020. In the BAU scenario, local emission sources continue to operate as normal under the meteorological conditions observed during the lockdown period. These forecasts allow for a direct comparison with the measured data. The estimated concentrations could be considered as weather corrected, deweathered, normalised or adjusted for meteorological variability concentrations.

2.3.5. Software and packages

All data analysis and the implementation of machine learning models were performed in R programming language (R Core Team, 2021) using its core packages and several independent packages. The package 'mgcv' (Wood, 2020) was used to implement the GAM model, 'openair' (Carslaw, 2021a) was used to download air quality data and calculate correlation coefficient and RMSE for model assessment, 'lubridate' (Spinu, 2021) was used to edit date and add temporal variables to the dataset, and 'worldmet' (Carslaw, 2021b) was used to download meteorology data from the London Heathrow Airport weather station.

3. Results and discussion

3.1. Comparison of the measured data

To calculate how NPN was affected during the COVID-19 lockdown, NPN in 2019 was subtracted from those in 2020. Therefore, a positive difference indicated an increase, whereas a negative difference indicated a decrease in the number of nanoparticles during the lockdown period. Change in the number of nanoparticles during March, April and May are shown in Table 3 for the three monitoring sites. P_{100} have increased by 13%, 46%, and 58% in March, April, and May, respectively at Chilbolton Observatory. In contrast, P_{100} have decreased at London Honor Oak Park by 29%, 2%, and 7% and at London Marylebone Road by 18%, 15% and 9% in March, April and May, respectively. P_{200} have shown a similar pattern to P_{100} i.e. their number has increased at Chilbolton Observatory and decreased at London Honor Oak Park and London Marylebone Road sites. However, P_{605} have shown slightly different results than P_{100} and PM_{200}, as shown in Table 3 that in addition to London Hoor Oak Park and London Marylbone Road, P_{605} have decreased even at the Chilbolton Observatory site in April and May. On average, the number of particles has increased at the rural sites and decreased at the urban background and urban traffic sites. When we averaged changes in particle numbers for the three sites, the rural sites showed positive gains, background site showed reductions, and urban traffic showed higher reductions. The reason for this is that particle numbers at urban traffic (roadside) sites are controlled by the local

Table 3
Difference in Nanoparticle numbers (particles/cm^3) between 2019 and 2020 during the COVID-19 lockdown.

Month	Size	Site	y2019	y2020	Diff	Diff (%)
March	P_{100}	CHO	1495	1682	187	13
April	P_{100}	CHO	2399	3492	1093	46
May	P_{100}	CHO	2361	3741	1380	58
March	P_{100}	LHO	2824	2257	−567	−20
April	P_{100}	LHO	3638	3550	−88	−2
May	P_{100}	LHO	3848	3562	−286	−7
March	P_{100}	LMR	8515	6964	−1551	−18
April	P_{100}	LMR	6407	5459	−948	−15
May	P_{100}	LMR	6637	6021	−616	−9
March	P_{200}	CHO	295	326	31	11
April	P_{200}	CHO	585	753	168	29
May	P_{200}	CHO	405	426	21	5
March	P_{200}	LHO	415	379	−36	−9
April	P_{200}	LHO	706	635	−71	−11
May	P_{200}	LHO	544	463	−81	−15
March	P_{200}	LMR	949	729	−220	−23
April	P_{200}	LMR	1039	796	−243	−23
May	P_{200}	LMR	789	641	−148	−19
March	P_{605}	CHO	107	137	30	28
April	P_{605}	CHO	249	234	−15	−6
May	P_{605}	CHO	138	97	−41	−30
March	P_{605}	LHO	167	158	−9	−5
April	P_{605}	LHO	375	246	−129	−34
May	P_{605}	LHO	195	129	−66	−34
March	P_{605}	LMR	272	224	−48	−18
April	P_{605}	LMR	423	247	−176	−42
May	P_{605}	LMR	251	153	−98	−39

emission sources, for example, emissions from the road traffic and because road traffic flow was decreased on roads in London, this directly affected the levels of NPN. Particle numbers at urban background sites are affected by the city scale emissions, whereas particle numbers at rural sites are affected by the regional scale emissions. Therefore, particle numbers at different types of environment demonstrated different changes during the COVID-19 lockdown.

Fig. 2 shows the ratio of different particle size at (a) London Marylebone road to Chilbolton Observatory (LMR/CHO), (b) London Marylebone to London Honor Oak (LMR/LHO), and (c) London Honor Oak to Chilbolton Observatory (LHO/CHO) for the period of COVID-19 lockdown. Fig. 2 clearly showed that the ratios at all sites decreased during 2020 for the lockdown period. Reduction in the ratio of LMR/CHO showed that NPN has decreased more at urban traffic (LMR) site than at the rural site (CHO). Furthermore, P_{100} demonstrated a greater reduction at urban traffic and urban background sites than at rural sites as shown by the ratios of LMR/CHO and LHO/CHO. This is because the levels of P_{100} are strongly related to the direct emissions from road traffic, whereas relatively larger size particles are of secondary nature i.e. secondary particles formed in the atmosphere (Rivas et al., 2020). Furthermore, the average ratios of P_{100}/P_{200} (10.10) and P_{100}/P_{605} (37.05) were highest at London Marylebone Road, followed by London Honor Oak Park, where the ratios of P_{100}/P_{200} and P_{100}/P_{605} were 6.8 and 22.25, respectively, whereas the lowest ratios of P_{100}/P_{200} (6.00) and P_{100}/P_{605} (21.45) were observed at Chilbolton Observatory sites. Again, this shows that P_{100} are in much larger proportion at urban traffic sites than at urban background and rural sites (Fig. 3), indicating emissions from road traffic.

Changes in TPN, PM_{10} and $PM_{2.5}$ during the lockdown period were calculated by subtracting their levels in 2019 from those in 2020 (Table 4). Both PM_{10} and $PM_{2.5}$ levels have decreased in all 3 months at London Marylebone road and London Honor Oak Park sites, except $PM_{2.5}$ in May at London Honor Oak Park site. Chilbolton Observatory showed slightly different results, where both $PM_{2.5}$ (22.80%) and PM_{10} (9.10%) levels decreased in April and increased in March and May (Table 4). TPN also decreased in April at London Marylebone road and London Honor Oak Park sites. Overall, PM_{10} and $PM_{2.5}$ decreased 30%

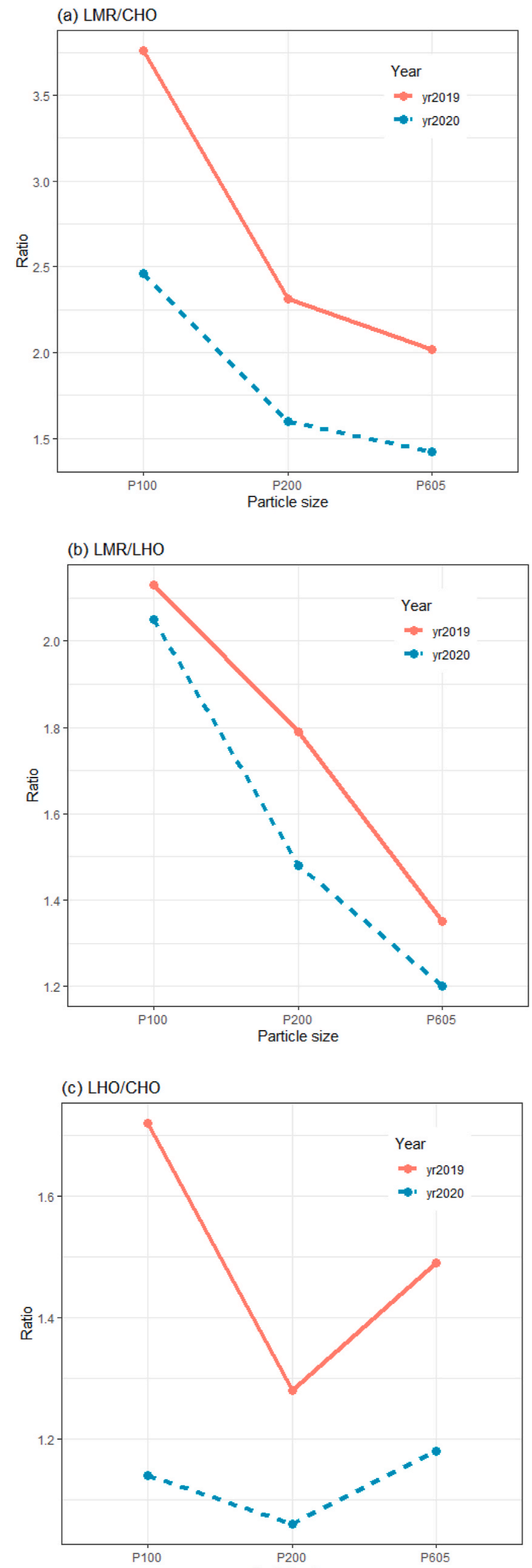

Fig. 2. Ratios of nanoparticle numbers: (a) LMR/CHO; (b) LMR/LHO; (c) LHO/CHO, where LMR stands for London Marylebone Road, LHO for London Honor Oak, and CHO for Chilbolton Observatory, P_{100} for particles of size up to 100 nm, P_{200} for particle size 101–200 nm, and P_{605} for particle size 201–605 nm.

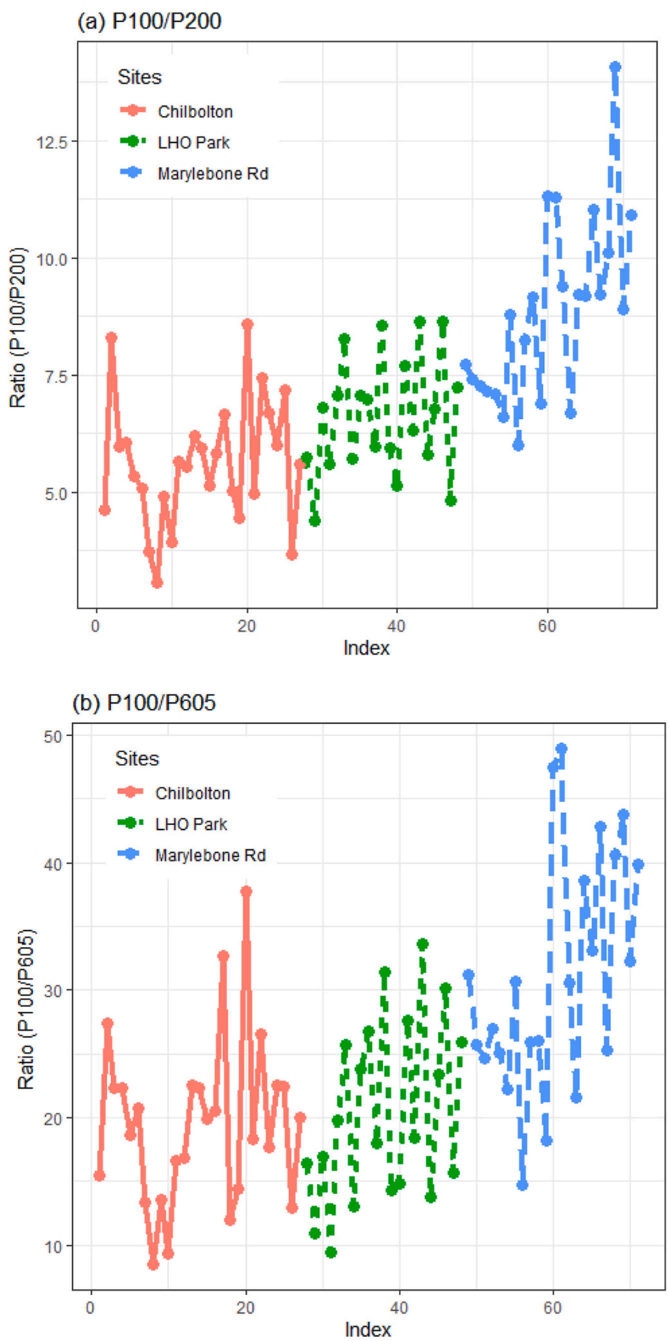

Fig. 3. Ratios of the different size of nanoparticle numbers (a) P_{100}/P_{200}, and (b) P_{100}/P_{605} at Chilbolton Observatory, London Honor Oak and London Marylebone Road.

and 50% at London Marylebone road, and 18% and 10% at London Honor Oak Park, respectively. However, PM_{10} increased by 8% and $PM_{2.5}$ decreased by 4% at Chilbolton Observatory. So, London Marylebone road (roadside) showed greater reduction compared to London Honor Oak Park (urban background), and no or little reduction was observed at Chilbolton Observatory rural site. TPN have decreased 53% and 30% at London Marylebone road and London Honor Oak Park sites, respectively. Due to missing data, the change in TPN at Chilbolton Observatory site was not calculated. The reason for this is that particle pollution at urban traffic (roadside) sites are controlled by the local emission sources, for example, emissions from the road traffic and because road traffic flow was decreased on roads in London, this directly affected the levels of particulates. Particulate matter at urban

Table 4
Total particle numbers (TPN) and PM_{10} and $PM_{2.5}$ mass concentrations and their difference between 2020 and 2019 (2020–2019). No TPN data was available for Chilbolton Observatory for year 2019. LMR stands for London Marylebone Road, LHO for London Honor Oak Park and CHO for Chilbolton Observatory.

Month	Pollutant	Site	2019	2020	diff	Diff (%)
March	PM_{10}	LMR	23.71	16.61	−7.10	−29.96
April	PM_{10}	LMR	32.26	22.01	−10.25	−31.77
May	PM_{10}	LMR	19.59	14.12	−5.48	−27.95
March	$PM_{2.5}$	LMR	16.13	8.45	−7.68	−47.60
April	$PM_{2.5}$	LMR	25.82	13.67	−12.15	−47.06
May	$PM_{2.5}$	LMR	15.14	7.04	−8.10	−53.52
March	TPN	LMR	26,249	NA	NA	NA
April	TPN	LMR	25,534	12,108	−13426	−53
May	TPN	LMR	NA	11,718	NA	NA
March	PM_{10}	CHO	11.77	13.15	1.38	+11.74
April	PM_{10}	CHO	24.69	22.44	−2.25	−9.10
May	PM_{10}	CHO	9.78	11.87	2.09	+21.33
March	$PM_{2.5}$	CHO	8.25	8.51	0.27	+3.26
April	$PM_{2.5}$	CHO	18.85	14.56	−4.30	−22.80
May	$PM_{2.5}$	CHO	6.60	7.05	0.44	+6.73
March	PM_{10}	LHO	9.99	8.83	−1.17	−11.66
April	PM_{10}	LHO	22.24	14.61	−7.63	−34.32
May	PM_{10}	LHO	8.32	7.56	−0.77	−9.24
March	$PM_{2.5}$	LHO	14.72	13.80	−0.92	−6.25
April	$PM_{2.5}$	LHO	29.56	21.86	−7.70	−26.06
May	$PM_{2.5}$	LHO	12.59	13.05	0.46	3.62
March	TPN	LHO	NA	11,703	3066	NA
April	TPN	LHO	12,096	8447	−3649	−30
May	TPN	LHO	NA	8189	NA	NA

background sites are affected by the city scale emissions, whereas particulate matter at rural sites are affected by the regional scale emissions. Therefore, particle pollution at different types of environment demonstrated different changes during the COVID-19 lockdown.

3.2. Machine learning results

The model results could be considered as weather-corrected air pollution changes. Firstly, the model was validated by using an independent dataset, which was not included in training the model. Correlation coefficient and RMSE values were calculated for the model assessment using both training and tested data. The model performance did not get worse for the independent dataset, showing a good model transferability. The values of correlation coefficients and RMSE showed acceptable model performance (Table 5). Table 6 presents the difference between predicted and observed values.

After validation, the model was retrained using 2018 and 2019 data and then used to predict PM_{10}, $PM_{2.5}$ and TPN for the month of March, April and May 2020 for the three sites. Predicted and measured levels of PM_{10}, $PM_{2.5}$ and TPN are compared. As an example, a comparison of the three species at London Marylebone Road is depicted in Fig. 4, which shows that predicted levels of PM_{10}, $PM_{2.5}$ and TPN are higher than the measured levels, indicating that pollutants levels have decreased during the lockdown period.

The modelling results (Table 6) showed that the concentrations of PM_{10}, $PM_{2.5}$ and TPN decreased at all three sites, however, the amount of change varied from site to site. PM_{10}, $PM_{2.5}$ and TPN levels decreased

Table 5
Comparison of fitted and cross-validated models for predicting PM_{10}, $PM_{2.5}$ and TPN.

Modelled pollutants	Fitted/Cross-validated	R - value	RMSE
PM_{10}	Fitted	0.83	8.55
	Cross-validated	0.83	7.36
$PM_{2.5}$	Fitted	0.84	7.05
	Cross-validated	0.82	6.08
TPN	Fitted	0.88	7852
	Cross-validated	0.88	7975

Table 6
Difference and percent difference between the observed and predicted PM_{10}, $PM_{2.5}$ and TPN using BAU scenario at different monitoring sites.

Site	Pollutant	Observed (min, max)	Predicted (min, max)	Difference (min, max)	Diff (%) (min, max)
London Marylebone Road	PM_{10}	17.58 (0, 79.8)	26.26 (0, 56)	−8.68 (−36, 51)	−33.05 (−321, 499)
	$PM_{2.5}$	9.73 (0, 72.3)	17.50 (−2, 44.55)	−7.76 (−31, 35)	−43.42 (−123, 675.23)
	TNC	11,958 (1186, 30,107)	24,073 (0, 46,858)	−12115 (−37273, 11,253)	−50.32 (−912, 529)
Chilbolton Observatory	PM_{10}	15.84 (0.58, 130)	16.16 (−2, 36)	−0.32 (−22, 117)	−1.98 (−98, 236)
	$PM_{2.5}$	10 (0.24, 76)	11.91 (−3, 30)	−1.85 (−20, 47)	−15.53 (−222, 350)
	TNC	6334 (260, 27,081)	NA	NA	NA
London Honor Oak Park	PM_{10}	16.29 (0.73, 82)	20.49 (−5, 52)	−4.20 (−30, 32)	−20.49 (−132, 775)
	$PM_{2.5}$	10.37 (0.35, 67)	14.20 (−4, 40)	−3.83 (−26,33)	−26.19 (−77, 546)
	TNC	9479 (681, 57,197(	11,309 (−3626, 22,085)	−1830 (−14913, 41,718)	−16.18 (−360, 854)

by 33.05%, 43.42% and 50.32%, respectively at London Marylebone Road site. PM_{10} and $PM_{2.5}$ decreased by 1.98%, 15.53% at Chilbolton Observatory, however, due to missing data of TPN in 2018 and 2019, the model could not predict TPN for the lockdown period. PM_{10}, $PM_{2.5}$ and TPN decreased by 20.49%, 26.19% and 16.18%, respectively at London Honor Oak Park. The reductions in all three pollutant concentrations were highest at the London Marylebone Road site, followed by the London Honor Oak Park. Chilbolton Observatory experienced the least reduction in PM_{10} and $PM_{2.5}$ concentrations. The model results showed that the increase in TPN and mass concentrations at Chilbolton Observatory when comparing measured data in 2019 and 2020 were probably caused by the changes in meteorological conditions.

4. Discussion

In London during the first lockdown, on average private driving, public transport and walking trips decreased by 69%, 86% and 78%, respectively (Mohajeriet al., 2021). However, the levels of pollutants did not experience similar reductions, which showed nonlinear relationship between mobility and pollutant concentrations. In addition to emission sources, pollutant levels are affected by the meteorological conditions, which vary from time to time. To normalise for the variations in meteorological conditions, in machine learning techniques several meteorological parameters were used. So, basically the model predicts that if there was no lockdown (BAU), what the pollutant levels would have been. Fig. 5 shows the Hysplit back trajectory during the lockdown period for April in 2019 and 2020. Back trajectories are used for understanding the origins of air masses, which can significantly affect the type and levels of air pollutants. This shows that it is not just the local reduction in the emission amount that affects the air pollutants but also the wind direction, speed and other meteorological parameters. An increase in particles pollution at rural sites could be partly related to sudden changes in wind speed, direction and their origin that bring different air masses from outside over large parts of the UK or even from the EU countries (Fig. 5). However, there was a positive association between mobility reduction and the levels of mass and particle concentrations at the roadside and urban background concentrations.

Jephcote et al. (2021) reported an average reduction of 12.6% in $PM_{2.5}$ concentrations in London. Furthermore, they reported higher reductions of $PM_{2.5}$ at urban background and urban traffic sites than the suburban and rural sites. According to Jephco te et al. (2021), four out of 47 sites experienced an overall rise in $PM_{2.5}$ concentrations. The four sites included the Chilbolton Observatory site, which is a rural background site and experienced 4% increase during the lockdown period. Overall, the findings of Jephcote et al. (2021) are in agreement with the current study. It was previously reported that Easterly winds from Central and Eastern Europe were loaded with high pollutant emissions and caused pollution episodes in the UK, especially in London and southern England (Pope et al., 2016; Graham et al., 2020). Episodes of $PM_{2.5}$ concentrations from 8th to 12th and 15th to April 17, 2020 were associated with easterly air flow from Eastern and Central Europe (Jephcote et al., 2021). This showed that meteorology plays an important role in controlling the levels of air pollution and therefore has the following implications:

1. Even if emissions are cut from road traffic, emissions from area and point sources and changes in meteorological conditions may still contribute to air pollution episodes;
2. The effect of changes in weather needs to be carefully considered, when quantifying long term trends, especially in view of the climate change and global temperature rising;
3. For a sustainable air quality improvement, the contribution of continental air masses to UK air quality needs to be considered, which emphasises the need for national and international collaboration. Only local efforts alone will have limited impacts on air quality.

No paper was found on the effect of lockdown intervention on particle numbers in the UK. However, several studies were conducted in other countries. For example, Shen et al. (2021) analysed the impact of lockdown measures on particle numbers in Italy. They reported that during the lockdown period the primary particle numbers of size 10–25 and 25–50 nm reduced by 66% and 34%, respectively at a regional background site in Ispra, Italy. However, they reported that lockdown had no effect on particle numbers at an urban background site in Modena, Italy. New particle formation frequency slightly increased compared to the same period in 2016–2019 in Ispra. However, the particle growth rates were lower during the lockdown at both sites compared to other periods. Shen et al. (2021) showed that significant decrease in traffic flow had little influence on particulate pollution levels in the Po Valley in Italy, which probably suggested that in addition to road traffic, other sources and processes also impacted particle numbers and mass concentrations. According to Shen et al. (2020) the particle number concentrations of nanoparticles decreased by approximately 40% during the lockdown period compared to the pre-lockdown in Beijing, China. However, they reported that accumulation mode particles increased by approximately 20% as several polluted episodes contributed to secondary aerosol formation. Similarly, Kanawade et al. (2020) analysed the effect of COVID-19 lockdown on particles number in India and reported that the number concentrations of particles $<$3 nm did not decrease, however, the number concentrations of particles $>$10 nm diameter decreased by 85% during the lockdown compared to the BAU scenario. It probably showed that the reduction in primary anthropogenic emissions did not inhibit the formation of particles $<$3 nm. Dinoi et al. (2021) analysed submicron particle concentrations, in the size range from 10 nm to 800 nm and found that particle numbers decreased by 19% in Lecce and 7% in Lamezia Terme, Italy during the lockdown period. The above studies were carried out in different cities around the world having different weather and emission conditions and therefore direct comparison with the UK would not make much sense. Rivas et al. (2020) carried out a source contribution in four European Cities including London, and concluded that the main emission sources were photonucleation, traffic emissions and secondary particle formation, however, the contribution of each source differed at each site. Therefore, the change in particle concentrations reported in different

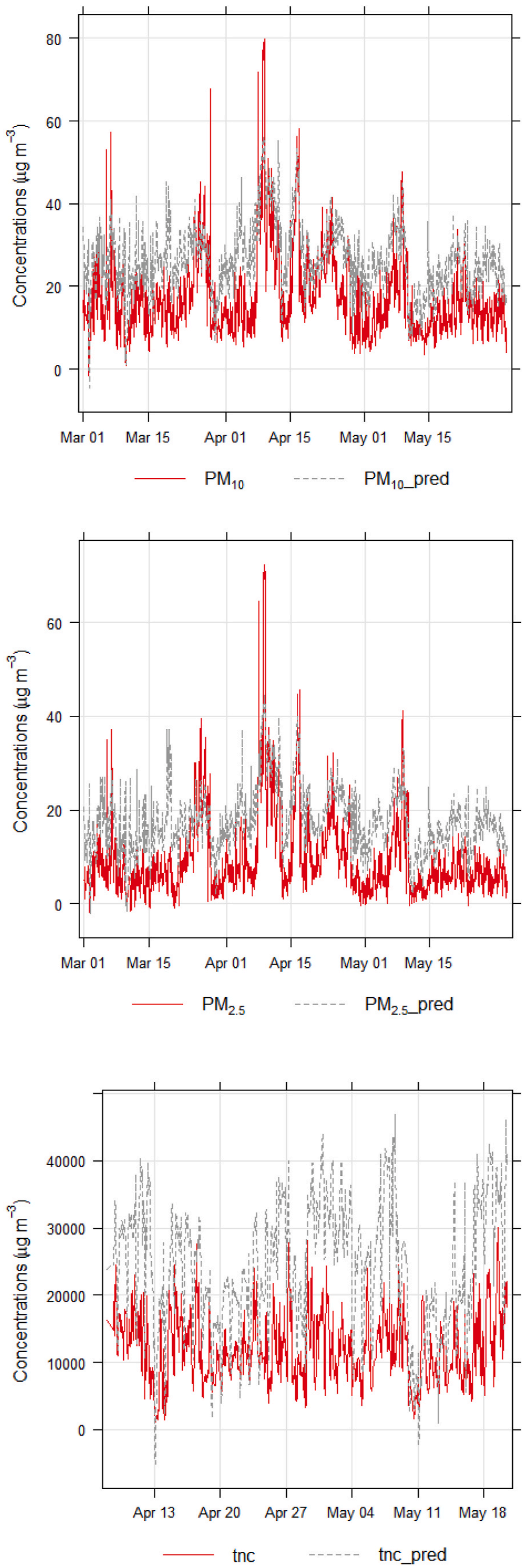

Fig. 4. Comparing predicted and measured PM_{10}, $PM_{2.5}$ and TPN concentrations for the lockdown period at Marylebone Road.

cities during the lockdown was different quantitatively, however, they all agreed that COVID-19 lockdown changed air quality worldwide, acting as a global air quality intervention. Such global interventions are very uncommon in recent history.

Several processes affect particle number counts and size such as nucleation, evaporation, condensation, deposition, and coagulation (Harrison et al., 2018). As freshly emitted particles move away from the emissions source their particle number and size distribution change due to these processes. Therefore, primary particles normally affect air quality near the emission source, however, as they travel away from the emission source secondary particles are formed, which affect air quality at the urban background and rural sites (Morawska et al., 2008). This is probably the main reason that both particle numbers and their composition are different at urban traffic, background and rural sites and they demonstrated different change during the lockdown period. Rivas et al. (2020) have provided a detailed analysis on how particle numbers in different size range are affected by local, city scale and regional scale emission sources and how the effect varies at different monitoring sites with different environment types. They have particularly analysed the effect of emissions from the Heathrow Airport on urban background monitoring sites in London.

Here it should be noted that this study has only considered the first lockdown for three reasons: (1) The data of the pollutants considered in this study was only available for the first lockdown period; (2) The first lockdown had a much greater impact on the mobility and air quality than the second and third lockdowns; and (3) the first lockdown was implemented simultaneously in all cities in the UK, whereas the later lockdowns were implemented at different times. Mohajeri et al. (2021) analysed the effect of COVID-19 lockdown on air quality in Greater London, Cardiff and Belfast and reported that the effect of the first lockdown on mobility was more abrupt and much stronger than the second lockdown. Quinio and Enenkel (2020) reported that the improvement in air quality was short lived and pollution levels increased again after the lockdown was relaxed. The recovery in pollution level was of three types: (a) V-shaped recovery, when the pollution levels returned back to their previous levels; (b) Plateau-shaped recovery, when the pollution increased but it didn't reach the previous levels observed before the lockdown period; and (c) Tick-shaped recovery, when pollution levels increased even more than those observed before the lockdown period. This shows that the effect of lockdown was not sustainable and hence the implementation of Clean Air Zones in large cities in the UK should go ahead as planned.

5. Conclusions

The novelty of the study is that it focuses on particle numbers considering both total particle and nanoparticle numbers. This study uses more meteorological parameters than previous studies, which include wind speed, wind direction, temperature, relative humidity, atmospheric pressure, dew point, visibility, and ceiling height. Furthermore, the study considers both raw data and weather corrected data to quantify the effect of the lockdown on air quality. In this paper, the effect of COVID-19 lockdown on the levels of PM_{10}, $PM_{2.5}$, TPN and NPN is analysed using measured data from a rural, an urban background and an urban traffic site during the first lockdown in the UK. It is shown that although COVID-19 lockdown was implemented to curtail person-to-person transmissions of the virus, it acted as an air quality intervention and improved air quality in urban areas, particularly at urban traffic sites. Two approaches are employed to extract the effect of lockdown: (1) Comparing the measured concentrations for the lockdown equivalent months in the year 2019 and 2020; (2) Comparing the prediction of machine learning with the measured concentration for the lockdown period of 2020. Overall, according to measured concentrations particle numbers including both nanoparticles and total particles increased at Chilbolton Observatory (a rural site) and decreased at both London Honor Oak Park (urban background site) and London Marylebone Road

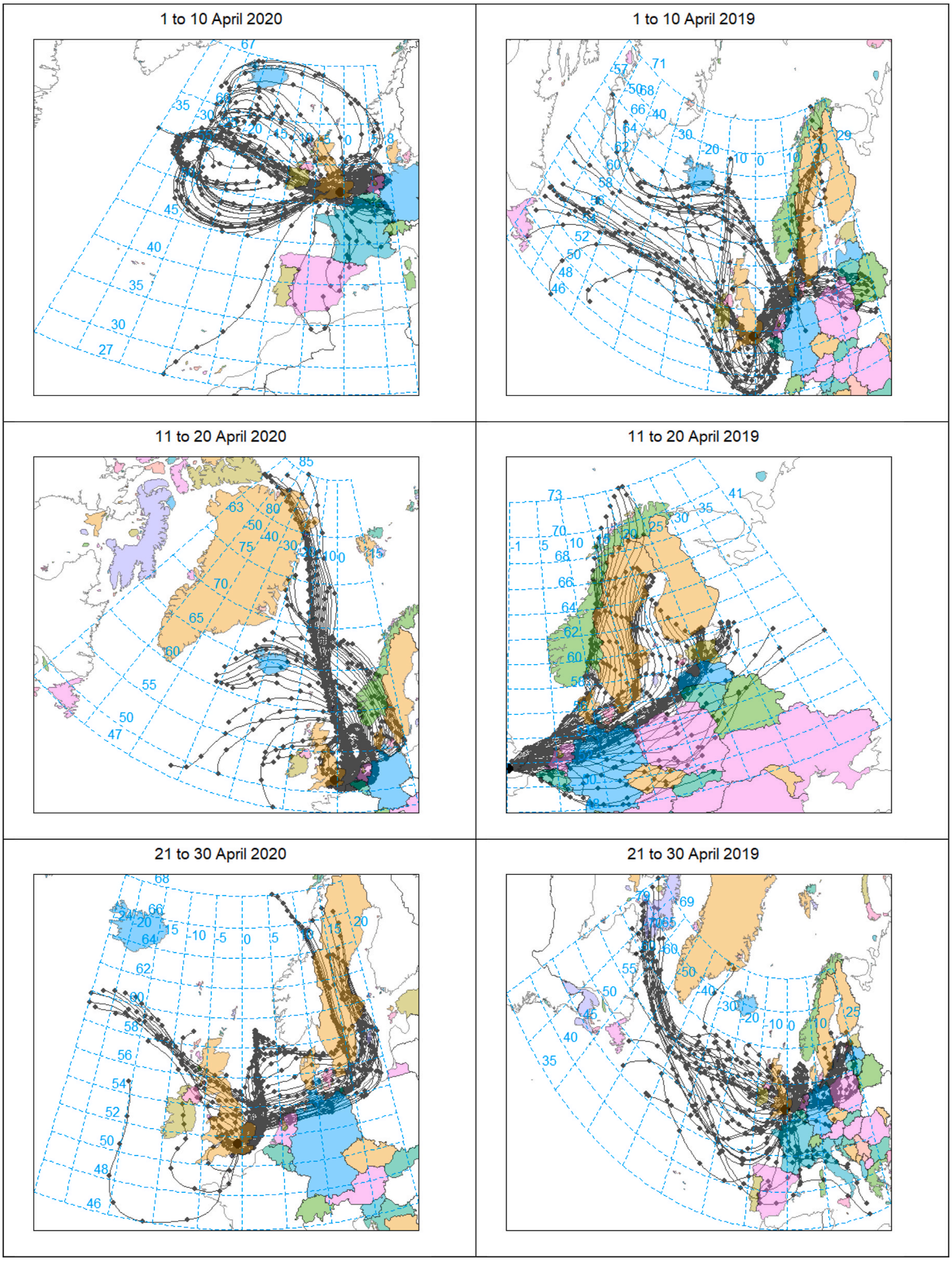

Fig. 5. Hysplit back trajectories centred on London UK for the month of April in 2019 and 2020.

(urban traffic site). Likewise, the mass concentrations of PM_{10} and $PM_{2.5}$ demonstrated reductions at London Honor Oak Park (18% and 10%) and London Marylebone Road (30% and 50%) and increase at Chilbolton Observatory (8% and 4%), respectively. Using measured data, the average for all three sites demonstrated increase in P_{100} and reductions for all other metrics i.e. PM_{10}, $PM_{2.5}$, TPN, P_{200} and P_{605}. However, quantitatively the amount of change was significantly different for each species. This shows that different metrics used for the expression of particles pollution are affected differently by their emission sources, meteorological parameters and policy interventions. The machine learning techniques results, however, showed that the concentrations of PM_{10}, $PM_{2.5}$ and TPN decreased at all three sites, but the amount of change varied from site to site. London Marylebone Road showed a significantly greater reduction than the other two sites.

Different changes in pollutant levels at different environment types demonstrate the complexity of the atmospheric system. On average, road traffic flow decreased by about 70% during the lockdown period in the UK, however, reductions in pollutant concentrations are relatively much milder. This probably shows the nonlinear association between the emissions and atmospheric concentrations of various pollutants. It is shown by several studies that after the COVID-19 pandemic air pollution levels increased back to the levels observed before the lockdown period, which demonstrated that the reductions in pollutant levels were not sustainable. Therefore, sustainable interventions are required to cut emissions, induce behaviour change and introduce legal policies at local, national and international levels.

The major limitation of this study is the data availability of NPN and TPN only from a few monitoring stations in the UK. Gaseous pollutants (e.g., NO_2) and mass concentrations of $PM_{2.5}$ and PM_{10} are measured at more than 150 air quality monitoring stations, which are part of the automatic urban and rural network (AURN) in the UK. In contrast, particles number concentrations, especially of nanoparticles are measured only at three monitoring stations. This is probably the main reason why very few studies have focused on particles numbers concentrations.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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