Impact of SARS-CoV-2 lockdown and de-escalation on air-quality parameters.

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Abstract

The SARS-CoV-2 health crisis has temporarily forced the lockdown of entire countries. This work reports the short-term effects on air quality of such unprecedented paralysis of industry and transport in different continental cities in Spain, one of the countries most affected by the virus and with the hardest confinement measures. The study takes into account sites with different sizes and diverse emission sources, such as traffic, residential or industrial emissions. This work reports new field measurement data for the studied pandemic period and assesses the air quality parameters within the historic trend of each pollutant and site. Thus, 2013-2020 data series from ground-air quality monitoring networks have been analysed to find out statistically significant changes in atmospheric pollutants during March-June 2020 due to this sudden paralysis of activity. The results show substantial concentration drops of primary pollutants, including NO_x, CO, BTX, NMHC and NH_3. Particulate matter changes were smaller due to the existence of other natural sources. During the lockdown the ozone patterns were different for each studied location, depending on the VOCs-

NOx ratios, with concentration changes close to those expected from the historical series in each site and not statistically attributable to the health crisis effects. Finally, the gradual de-escalation and progressive increase of traffic density within cities reflects a slow recovery of primary pollutants. The results and conclusions for these cities, with different sizes and population, and specific emission sources, may serve as a behavioural model for other continental sites and help understand future crises.

1 Introduction

The coronavirus disease SARS-CoV-2 has caused an unprecedented public health emergency worldwide and it will have huge social and economic long-term impacts. The efforts to mitigate the spread of the virus have led to the confinement of the population of entire countries, the cessation of industrial and commercial activities considered as not essential and a drastic drop in transport. Due to the direct correlation between anthropogenic activity and the emissions to the atmosphere (Omri, 2013; Kang et al., 2019), air pollution levels have been affected by the current health crisis.

Air pollution is a mix of gases, particulate matter (PM), and biological materials, which constitutes a critical global health issue affecting the population massively. A report from the WHO (WHO, 2016) stated that around 3 million deaths per year were attributable solely to outdoor air pollution and billions of people were being harmed. Atmospheric pollutants contribute to breathing problems, cardiovascular diseases, and premature mortality (Sunyer, 2001; Mafrici et al., 2008; Song et al., 2014; Abdo et al., 2016; Wang et al., 2019; Wu et al., 2019).

The improvement of given air-quality parameters during the last two decades may be explained as a result of the environmental policies adopted to abate pollution and their effects (Rexeis and Hausberger, 2009; Gaba and Iordache, 2011; Querol et al., 2014; Alves et al., 2015; Roy and Sandar, 2015; Euro 6, 2020). However, it is in atmospheric PM and tropospheric O_3 that less important reductions have been achieved (EPA, 2020). These minimum decreases may be due to the stationary natural contributions to PM and to the lack of regulation on the concentrations of volatile organic compounds, VOCs, which are precursors of both tropospheric ozone and ultrafine particles.

During the past few years, several studies have reported the effect of economic downturns on both the emissions and concentrations of atmospheric pollutants (Hilboll et al., 2013, Sánchez de la Campa and de la Rosa, 2014; Cuevas et al., 2015, Zyrichidou et al., 2019; Pacca et al., 2020). Although previous financial crises had an immediate impact (during the year of the crisis) on the emissions of CO_2, SO_2 and NOx, the data reflect that these beneficial impacts on air quality were short-lived and insignificant in the medium-term (over ten years since the onset of the crisis) (Du and Xie, 2017, Pacca et al., 2020). Since the SARS-CoV-2 pandemic has led to the steepest drop in economic activity at a global level so far, it provides a unique scenario to assess the changes in atmospheric pollution and the derived health benefits during this new and unexpected crisis. On the other hand, it is also paramount to study the role of atmospheric pollution in the spread of diseases and their effects. In this sense, recent works suggest that long-term exposure to air pollution, especially PM and NO_2, increases vulnerability to SARS-CoV-2 (Wu et al., 2020; Ogen, 2020; Coccia, 2020; Zhu et al. 2020).

Images from the Copernicus Sentinel-5P satellite have recently shown strong reductions in NO_2 concentrations over several cities across Europe, Asia and America (Bauwens et al., 2020;

Muhammad et al., 2020; van Geffen et al., 2020; Wang and Su, 2020; Zambrano-Monserrate et al., 2020, Vadrevu et al, 2020). Nevertheless, even though satellite measurements display a fast and global image of the concentrations over large areas, they may not provide enough resolution for local effects and they may bias the data when comparing different regions since satellites overpass them at different local times. Furthermore, surface equipment can provide measurements for a greater number of pollutants. Thus, the use of data from ground-air quality local networks is the ideal tool to assess the surface concentrations in given sites, such as individual cities.

Following this methodology, recent works have shown local decreases of several atmospheric pollutants concentrations (NO, NO_2, SO_2, O_3 and PM) in the biggest Spanish cities (Barcelona, Madrid and Valencia) during the lockdown period, (last fortnight of March - first fortnight of April) (Baldasano, 2020; Sicard et al., 2020; Tobias et al., 2020). Likewise, some studies have involved the effect of the lockdown on air quality in megacities across the globe (Kumari and Toshniwal, 2020, Connerton et al., 2020). Being Spain one of the countries most affected by the SARS-CoV-2 and where the hardest confinement measures have been taken to control the propagation of the virus, it may be adopted as a reference model for the behaviour of air-quality parameters. However, in this country, as in many others, the larger portion of population (71%) lives in small cities or villages (INE, 2019), where the effect of pollutants emissions in the surrounding areas may be very important as well. Thus, additional studies of representative sites are also required to know the overall exposure of citizens to air pollutants during this health crisis. In addition, due to the existence of diverse sources, the emission of each pollutant may drop differently. The effect on atmospheric concentrations may be quite different considering the pre-existing levels of each species in a specific place and their different atmospheric chemistry mechanisms and lifetimes. Thereby, the assessment of the greatest possible number of pollutants is necessary to quantify the real impact of the health crisis on air quality.

For these reasons, this work aims to study the short-term effect of the sudden stop of industrial and transport activities on air quality parameters based on data provided by local monitoring networks. Unlike most previous studies where the effects of lockdown in large cities were evaluated, this work covers four different size continental measurement areas, including one as a model for large cities (Madrid), one for medium-size cities (Albacete), one small city as a model for industrial places (Puertollano) and, finally, a remote site (San Pablo de los Montes, S. Pablo). In contrast to previous works, numerous pollutants were analysed (NOx; O_3; SO_2; CO; $PM_{2.5}$; PM_{10}; BTXs (Benzene, Toluene and Xylenes); NMCH (non-methane hydrocarbons); CH_4 and NH_3), covering not only the lockdown but also the de-escalation period (14 March - 30 June). In addition, the 2020 measurements were compared with the corresponding values of the last seven years, which allows to detect short-term emissions changes within the long time series.

As in the case of the study of carbon dioxide changes due to the COVID-19 crisis and related prospects (Wang and Wang, 2020), understanding previous and current evolution of pollutants may be useful for developing new strategies addressed to improve air quality in the future.

2 Materials and Methods

2.1 Area of Study. The study is focused on the south-central plateau of the Iberian Peninsula (Fig. 1). It is a zone with Mediterranean-continental climate, moderately industrialized, and whose most relevant pollution episodes are usually due to traffic emissions in cities under stagnation and high irradiance conditions (MITECO, 2018).

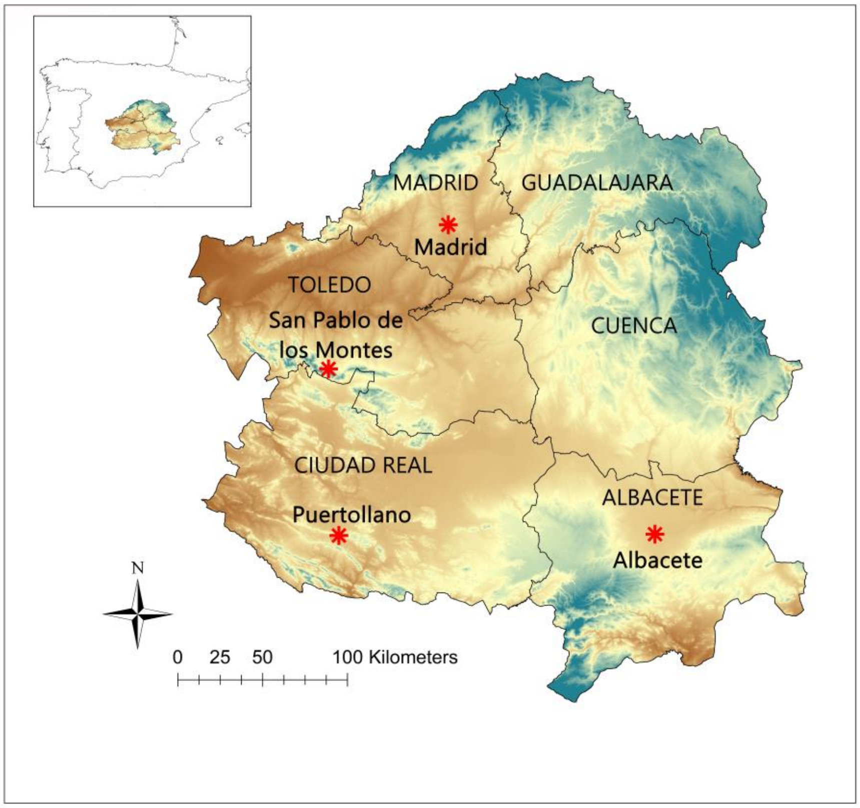

Figure 1. Map of the study area and locations of air quality monitoring stations.

Madrid is the most densely populated city in Spain with 3.3 million inhabitants (INE, 2019). Road transport is the most important source of urban pollution, although other sources such as residential heating also contribute to emissions. In this sense, road transport in Madrid was roughly responsible for 58% of NOx emissions and 52% of $PM_{2.5}$ emissions (DGSEC, 2018). Castilla-La Mancha is located in the South-East, (Fig. 1), where Albacete is the largest city (173,000 inhabitants, INE, 2019) and it is placed 250 km south-east from Madrid. Puertollano, with 47,000 inhabitants (INE, 2019), is located 240 km south-west of Madrid, and it has the highest concentration of heavy industry in the centre of the Iberian Peninsula with a refinery, petrochemical installations, a fertilizer factory and two power plants. Lastly, S. Pablo is a small village (1,800 inhabitants, INE, 2019) at 170 km south-west of Madrid. It is a rural area with agriculture as the main activity and is considered as a remote continental area of central Spain.

2.2. Data source and analysis. Datasets of air-quality monitoring networks were used to study the evolution of air pollutants during the pandemic. In contrast to the earlier studies in the field, local monitoring stations have been chosen to characterize changes in surface air quality since they provide data for a large number of pollutants. A representative monitoring station was selected in each of the above-mentioned places. The urban-traffic station of "Escuelas Aguirre" is sited in Madrid city downtown and therefore its location is directly influenced by intense traffic emissions. The suburban background station of Albacete is located in a residential and commercial area and, consequently, the main source of emission that affects this station is traffic and combustion from both sectors. On the other hand, the station "Campo de Fútbol", in Puertollano, covers both industrial and residential sectors. With regard to S. Pablo, its station provides national coverage of the background atmospheric pollution network. The hourly average level of pollutants measured were SO_2, CO, NO_2, PM_{10}, $PM_{2.5}$, and O_3, however, in S. Pablo PM_{10} and $PM_{2.5}$ were daily measured by a gravimetric sampling method. Other pollutants, such as BTXs, were measured only in Madrid and Puertollano. Likewise, NMCH were available only for Madrid, and NH_3 was only measured in the industrial site, Puertollano.

The methods used to analyse the different air pollutants are defined by the European Directive (2008/50/CE) as reference or equivalent methods. BTXs and NMHC were measured by gas chromatography with flame ionization detectors and NH_3 through chemiluminescence.

All statistical analyses were done using SPSS (IBM SPSS Statistics 23). In the case of variables normally distributed, Student's t-test was performed for the comparisons among monthly average concentrations of the different pollutants measured. If the measures were not normally distributed, then a nonparametric Mann-Whitney U test was performed. P-values less than 0.05 were considered to be statistically significant.

2.3. Chronology. On 14 March 2020 it was declared the state of alarm in Spain to control the spread of SARS-CoV-2 (BOE, 2020) introducing measures such as the restriction on the freedom of movement of people, the suspension of public activities, the closure of cultural and recreational facilities, etc. On 30 March, the Spanish government introduced even stricter measures to the confinement. Non-essential activities, including industrial and construction sectors, were suspended, beginning a temporary period of "economic hibernation". On 13 April, both sectors were again allowed to return to activity with the initial restrictions. The de-escalation started on 4 May 2020, with the gradual reduction of the restrictions. Mobility and commercial activities progressively increased until 21 June, the date on which the alarm state ended.

3 Results and discussion

For each studied site, a short description of air quality parameters just before the lockdown is introduced. Then, the data from the period March-June 2020, which cover both lockdown and de-escalation, are discussed taking into account the pandemic chronology and are compared with those obtained from previous months (January-February, 2020) and with those from the same months interval (March-June) since 2013.

3.1. Madrid. Figure S1 (Supplementary material) shows the monthly average hourly data for January-June 2020 for Madrid. In January and February 2020, before the SARS-CoV-2 crisis, the time profiles in Madrid for NOx had two significant maxima corresponding with local rush hours and kept very high (most time during the day above 100 µg/m^3) due to the dense traffic in the city. CO showed a very similar profile with concentrations around 0.5 mg/m^3, see for example January or February 2020. At the same time the ozone concentrations were very low, with maximum values below 40 µg/m^3 (at 16:00h, local time) and minimum levels as low as 10 µg/m^3 (at 9:00h/21:00h) before the lockdown. The surface concentrations profile of BTXs also resembled those of NOx, with, for example, toluene values around 3 µg/m^3. SO_2, with average concentration around 7µg/m^3, showed a smoother variability peaking during the daylight hours. The daily trend of NMHC was also similar to that of NOx, with values ranging from 60 to 150 µg/m^3 (minimum and maximum, respectively). The time behaviour of all these gas pollutants seems to be driven by traffic emissions. Concerning particulate matter, both PM_{10} and $PM_{2.5}$ showed lowering trends from midnight to 7:00h and raised smoothly during the day, showing the contribution from vehicles.

In contrast, if we consider the average daily data for the months with harsh lockdown, March and April 2020, Figure S1, NOx maximum values fell below 70 µg/m^3 and 40 µg/m^3, respectively, toluene to 1.5 µg/m^3 and 1 µg/m^3; and CO also lowered to values around 0.3 mg/m^3 for both months. A significant decrease, although lower than that of NO_2, was also observed in particulate matter when comparing January averages with those of March and April, with values during the lockdown below 15 µg/m^3 and 10 µg/m^3 for PM_{10} and $PM_{2.5}$, respectively (Table 1). However, changes in SO_2 concentrations were hardly noticeable.

Figure 2 shows the monthly average profiles for the period January-June 2020 for all the studied pollutants. In general, a clear decrease can be observed comparing the weeks before and after the beginning of the lockdown. The results were confirmed analysing statistically the average concentrations of each pollutant in January-March and January-April, and finding statistically significant differences (p-value < 0.05) for all of them except for SO_2. Similar results have been reported for the Spanish largest cities, Madrid, Barcelona and Valencia, with the highest reductions for NO_2 (-62%; -50% and -70%, respectively), lower decreases for PM (−30% in Barcelona and Valencia) and no noticeable changes for SO2 (in Barcelona) (Baldasano, 2020; Sicard et al., 2020; Tobias et al., 2020).

Considering the de-escalation period (May - June) (Figure 2a and S1), the observed pollutant concentrations increased slightly with respect to April's values, mainly in June, but the concentrations didn´t return to the starting values measured in January for any pollutants except to PM and NMHC. Indeed, the restrictive measures reduced emissions of these pollutants from transport and fuel combustion in institutional and commercial buildings, but these decreases were partly counterbalanced by the increase in household emissions (e.g. domestic heating, biomass burning).

The exception was observed for ozone, whose values increased progressively throughout the analysed months (Figure 2a).The rise of solar radiation intensity in spring months and the decrease of the titration effect under lower NO concentrations may explain the rise of ozone despite the fact that emissions of O_3 precursors such as NO_2 or organic compounds were low. Moreover, the decrease of fine particulate matter during this period may have slowed down the aerosol sink of hydroperoxy radicals (HO_2), stimulating ozone production (Li et al., 2019). This rise has been also observed in others Spanish large cities, such as Barcelona and Valencia (Sicard et al., 2020; Tobias et al., 2020).

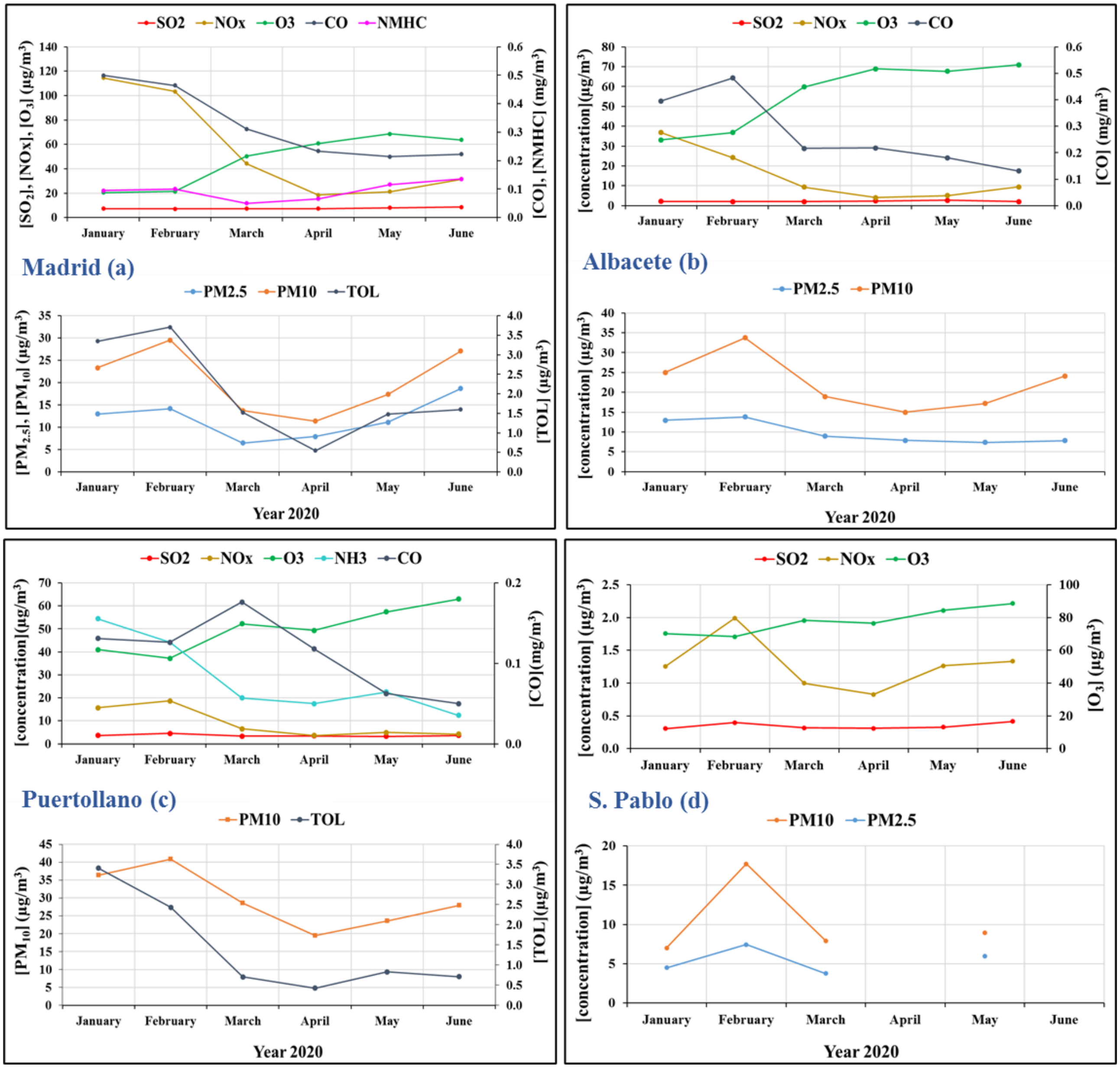

Figure 2. Monthly average profiles for the period January-June 2020. In Figure 2d, the March average only covers the period 1-14 March, just before the declaration of the state of alarm and lockdown beginning.

Although the effects of the lockdown are visible, the changes in pollutants are seasonally dependent (Notario et al., 2012, Notario et al., 2013) and so, the observed behaviour is not solely attributable to the loss of anthropogenic activity. For that reason, a larger database under similar seasonal conditions is required to assess the real effect of this massive confinement. In this sense, we have used 2013-2020 raw data from the studied sites to derive monthly averaged values for the available pollutants. Thus, Table 1 shows the average surface concentrations during the months March-June since 2013 and the reduction percentage during 2020 with respect to the average values obtained for the previous seven years. Likewise, the 2013-2020 data series are displayed in graphical format in Figure S5 and the change percentage during the lockdown and de-escalation with respect to 2013-2019 averages are shown in Figure 3.

Table 1. Monthly average concentrations for March, April, May, and June in Madrid since 2013 and deviation of the 2020 value from the corresponding 2013-2019 average. Metadata from national and regional repositories: MITECO, 2020a, Madrid 2020. *Data not available.

(mg/m³)	Year → Month ↓	2013	2014	2015	2016	2017	2018	2019	Average 2013-2019	2020	Variation %
NO_x	March	68.2	87.3	108.3	90.6	111.2	78.3	85.2	89.9 ± 15.4	44.2	-50.8
	April	60.8	76.5	68.4	80.0	72.8	76.8	67.5	71.8 ± 6.7	18.5	-74.2
	May	55.5	60.9	71.3	80.0	77.9	67.2	56.3	67.0 ± 9.9	21.0	-68.6
	June	60.2	62.3	66.9	80.6	79.7	65.7	54.9	67.2 ± 9.7	31.4	-53.3
SO_2	March	7.5	7.9	11.2	16.9	7.2	4.8	9.1	9.2 ± 3.9	7.3	-20.9
	April	5.3	6.2	7.7	15.5	5.0	1.4	8.7	7.1 ± 4.4	7.3	2.7
	May	4.8	6.4	9.2	15.0	6.1	4.0	9.1	7.8 ± 3.7	8.0	2.3

	June	3.9	6.5	11.7	17.3	8.0	4.0	9.6	8.7 ± 4.7	8.5	-2.4
CO	March	352.0	330.0	683.0	423.0	372.0	340.0	171.0	381.6 ± 154.0	311.0	-18.5
	April	371.0	301.0	581.0	362.0	472.0	336.0	144.0	366.7 ± 136.6	234.0	-36.2
	May	342.0	368.0	406.0	352.0	378.0	311.0	144.0	328.7 ± 86.7	214.0	-34.9
	June	337.0	354.0	395.0	341.0	307.0	290.0	284.0	329.7 ± 39.2	222.0	-32.7
O_3	March	39.7	42.6	37.2	44.3	41.7	49.4	50.6	43.6 ± 4.9	50.3	15.3
	April	49.9	43.9	55.4	46.9	59.9	50.9	59.2	52.3 ± 6.1	60.7	16.1
	May	53.1	60.7	58.9	53.1	56.0	61.3	60.4	57.6 ± 3.6	69.1	19.8
	June	55.5	61.2	70.0	55.0	58.4	64.3	64.5	61.3 ± 5.4	63.7	4.0
$PM_{2.5}$	March	9.1	12.0	12.2	9.9	9.9	5.1	15.2	10.5 ± 3.1	6.5	-38.2
	April	10.2	10.4	9.7	7.4	8.4	7.0	7.5	8.7 ± 1.4	7.9	-9.0
	May	9.1	10.3	10.6	7.8	8.8	7.4	6.4	8.6 ± 1.5	11.1	28.4
	June	11.9	11.9	13.6	10.5	11.8	11.5	9.0	11.4 ± 1.4	*	*
PM_{10}	March	11.6	23.2	22.2	16.6	19.1	7.0	25.5	17.9 ± 6.7	13.7	-23.2
	April	20.3	20.2	19.8	12.2	19.3	11.9	14.6	16.9 ± 3.9	11.4	-32.5
	May	16.9	19.2	23.5	14.7	19.8	10.3	14.6	17.0 ± 4.3	17.4	2.2
	June	29.4	24.8	25.3	23.1	26.7	22.6	25.4	25.4 ± 2.3	*	*
TOL	March	0.7	3.0	3.1	2.4	3.5	1.4	2.1	2.3 ± 1.0	1.5	-34.2
	April	0.9	2.8	2.1	2.0	2.4	1.8	1.5	1.9 ± 0.6	0.5	-73.8

	May	0.8	2.7	2.3	1.9	2.1	2.0	2.8	2.1 ± 0.7	1.5	-29.8
	June	0.9	2.8	2.1	2.3	2.3	1.9	2.5	2.1 ± 0.6	1.6	-24.3
NMHC	March	*	*	146.0	136.0	167.0	39.0	72.0	112.0 ± 54.1	50.0	-55.1
	April	*	158.0	118.0	124.0	143.0	43.0	57.0	107.2 ± 46.7	66.0	-38.4
	May	*	153.0	145.0	121.0	116.0	48.6	65.1	108.1 ± 42.4	116.7	7.9
	June	*	144.0	164.0	175.0	54.0	86.0	66.0	114.8 ± 52.5	135.0	17.6

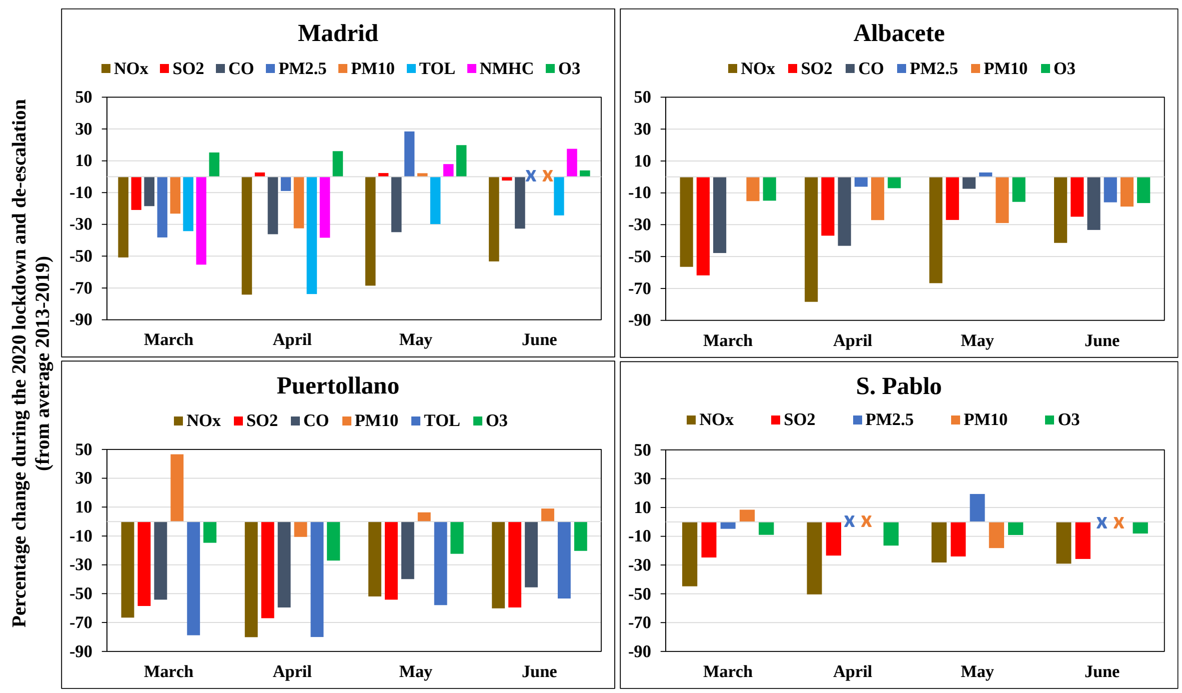

Figure 3. Monthly average percentage change during lockdown and de-escalation with respect to 2013-2019 average. [X]For Madrid, 3a, June PM data were not available in 2020. [X]In S. Pablo, 3d, the 2020 March average only covers the period 1-14 March, just before the declaration of the state of alarm and April PM data were not available in 2020.

The results show clear drops for the primary pollutants NOx, CO and toluene (the BTXs with the highest concentration) during the state of alarm, with a maximum decrease in April, since this month was the period under the hardest lockdown measures (See Figure 3a). An analysis of their monthly means shows statistically significant differences (95.0% confidence interval) between the means measured in 2020 and those of 2013-2019. Similar results have been reported for NO_2, when the March averages are compared with those of 2018 (decreasing of 46%) and 2019 (decreasing of 56 %) for 23 air quality monitoring networks in Madrid (Baldasano, 2020). The decrease in NO_2 measured by local surface stations (this work, Baldasano 2020) are significantly larger than those based on satellite observation, in the order of 20-30%, (Muhammad et al., 2020) probably due to a lack of sufficient resolution for relatively small sites such as cities.

The change in these primary pollutants in Madrid may be explained from the decrease of local and national transport all around the country during the lockdown. In this sense, Figure 4 shows the trend of traffic accessing cities (DGT, 2020). The level of traffic dropped during given days down to 80% with respect to any equivalent day under normal activity conditions and the decrease was even more pronounced during weekends and Easter Holidays, down to 90%. The level of traffic started to rise slightly since April 13 due to the lifting of some restrictions addressed to enable the recovery of economic activity, still within the alarm state. A higher rise was observed since 4 May, when the de-escalation started, until the end of the state of alarm, on 21 June. Thus, the average decreases for March, April, May and June were 41, 76, 53 and 24 %, respectively. The monthly average profiles of NOx, CO and toluene (Figure 2a) for the period January-June 2020 are consistent with these traffic reductions.

For SO_2, the beginning of the lockdown involved a -21% decrease with respect to the 2013-19 March average, while the changes were smaller (+3, +2 -2%) for April, May and June, respectively (Figure 3a). The data reflect no statistically significant changes in the studied period, with measurements for 2020 similar to the averages for the period 2013-2019 within the uncertainty interval (Table 1). The regulation banning sulphur in fuels has reduced drastically the SO_2 levels in cities during the last two decades and, so, the drop in traffic has not had a net effect on SO_2 concentrations.

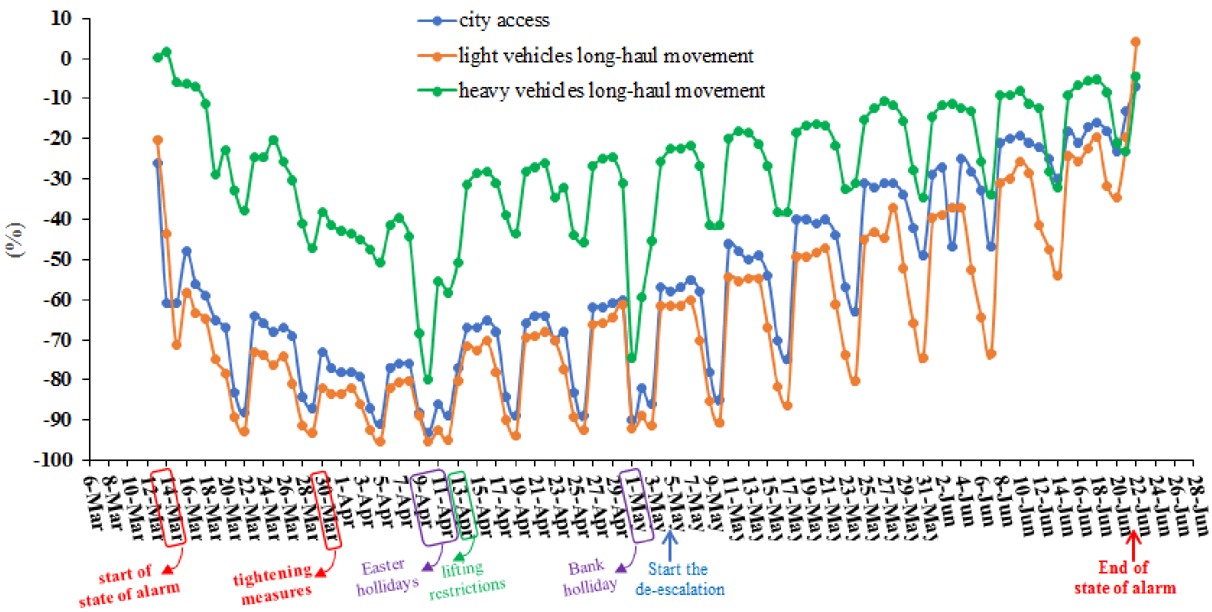

Figure 4. Average percentage decrease of traffic density relative to an equivalent day (before the crisis) in Spain. Data from DGT repository (DGT, 2020).

The relative percentages of change for $PM_{2.5}$ were -38, -9 and +28%; and -23, -33 and +2 % for $PM_{10,}$ in March, April and May, respectively. For June, the month averages could not be obtained since experimental measurements were only carried out during six days (Figure 3a).The highest PM decrease corresponds to the stricter period of the state alarm (March and April), although the decrease for $PM_{2.5}$ in April was lower than expected. In May, also unexpectedly, a significant PM increase was observed with respect to average 2013-2019, which indicates that the effect of the lockdown on PM is not equal to that observed for NOx, and that traffic density cannot be the only influential parameter in their behaviour. In this sense, the existence of other sources, some of them natural, which lead to a regional background also affected by Saharan intrusions, could be the reason for the unexpected increases in the PM concentrations (Salvador et al., 2013). In this sense, Table S1 shows the dates or periods with contributions of such intrusions on particulate matter. In addition, PM rise from domestic heating (higher due to the lockdown of people at home) and garden activities (e.g. biomass burning) should also be considered (Sicard et al., 2020). Thus, PM emission from the residential sector, including household and gardening combustion, means 41 % of the total in Spain. These emissions have risen by 23.8% since 2000, despite the decrease in total fuels consumption (-1.3% since 2000), mainly due to the increase in biomass consumption (+27.5% since 2000) (MITECO, 2020b).

The results obtained in this study for $PM_{2.5}$ are consistent with those reported in a recent work involving the 50 most polluted capital cities in the world (Rodríguez-Urrego and Rodríguez-Urrego, 2020). Although an average 12% reduction was found, a unique behaviour was not observed, with some cities keeping or even increasing PM levels during the lockdown, showing that local or regional factors contribute significantly to PM pollution.

For NMHCs, Figure 3a, a significant decrease from the 2013-2019 average was observed in March and April (-55 and -38 %, respectively), while the concentration rose in May and June (+8 and +18 %), reflecting a lack of correlation with the reported density of vehicles during the studied period. These results show that the lockdown had not a clear effect on NMHCs levels, probably due to the contribution of additional sources different from traffic such as agriculture and livestock (accounting for 13% of total NMHC emissions in Spain), use of domestic solvents (10%), and residential stationary combustion (6%) linked to wood used as a fuel (MITECO, 2020b). A similar behaviour has been found recently in Rio de Janeiro, Brazil (Siciliano et al., 2020).

The case of ozone, with an increase in surface concentrations around 15 % during the harsh lockdown, is also clearly different to the rest of primary pollutants and is discussed below.

3.2. Albacete. For Albacete, model for a medium-size city, the concentrations of NOx were between three and four-fold lower than in Madrid for winter time before the SARS-CoV-2 crisis (see January and February 2020 in Figure S2) and also showed two maxima (around 70µg/m^3) corresponding to rush hours. Nevertheless, from 12:00h to 18:00h there is a significant decrease to concentrations below 30 µg/m^3. Under such lower NO concentrations, the titration of ozone is reduced and the effect of irradiation during the daytime and photolysis of NO_2 results effective in the production of surface ozone. Thus, the concentrations of ozone (ranging from 17 to 63 µg/m^3) were higher than those measured in Madrid during the same months and under similar meteorological conditions. SO_2 and CO showed very slight increases during the day, matching up in time with those of NOx. On the other hand, the mass particle concentrations for PM_{10} and $PM_{2.5}$ were very similar to those reported for Madrid in the same months. Finally, the monitoring system does not provide data concerning organic compounds.

During lockdown and de-escalation months (See Figure 2b and S2) the reduction in the concentrations was notable and statistically significant ($p < 0.05$) for all pollutants, although the concentration values increased slightly for NOx and PM_{10} in June. Ozone was again the exception with a progressive rise during the spring months.

Regarding the annual comparison, the reduction percentages during March-June 2020 with respect to previous years were significant for NOx (-57, -58, -67and -41%, respectively) (Figure 3b). Likewise, for CO, concentrations lowered in -48, -43, -8 and -33%. For both NOx and CO the analysis of their average monthly concentrations shows statistically significant differences between 2020 and the period 2013-2019 (p-value <0.05), except for CO during the de-escalation (May and June). As it can be seen in Table S2 and Figure S5, the historical data for CO show a higher variability than those of NOx and precludes the confirmation of the effect on CO in these two months. Comparing the average data (2013-2019) with Madrid, the concentration of CO was unexpectedly higher in Albacete during March and April and lower in May and June. A previous work about the assessment of air quality in Spain had also reported such behaviour for CO in this city in 2013, (MITECO, 2013). Moreover, in this study the ratio [CO]/[NOx] was always higher in Albacete and the time profiles of these two primary pollutants show a poor correlation in this city (see Figures S5 and S6), which suggests the existence of additional sources on CO, other than traffic. Up to our knowledge, the origin of these high unusual levels of CO in Albacete has not been confirmed.

In the case of SO_2, substantial and statistically significant (p-value< 0.05) changes from the averages (-63, -36, -27 and -26%) were also observed for March-June respectively during 2020 (Figure 3b). Nevertheless, since 2015 the levels of SO_2 in this city were already very low and stable. In this sense, the decrease of SO_2 with respect to the previous five-year period was in fact negligible (Table S2 and Figure S5).

For PM_{10} and $PM_{2.5}$, the changes with respect to the averages for the same months in the period 2013-19 were low (Figure 3b) and their absolute concentrations were similar to the averages considering the uncertainty ranges (Table S2). So, the effect of the lockdown has been minor and lower than in the case of Madrid. These results suggest that the contribution of traffic emissions to particulate matter in this medium size city is not dominant.

According to the monthly average profiles for the period January-June 2020 of this city (Figure 2b), in June a significant increase of PM_{10} was observed compared to $PM_{2.5}$. The increase of coarse particles over fine particles may be due to the dry and hot conditions in this site during June which may have enhanced the release of mineral dust, with a higher contribution to the PM_{10} fraction. Furthermore, as it can be seen in Figure 2, the $PM_{2,5}/PM_{10}$ ratio is smaller in Albacete and correlates worse than in Madrid, showing a lower contribution of traffic to air particulate matter.

3.3. Puertollano. For Puertollano, model for small-size industrial towns, in January and February 2020, before the Covid-19 crisis, the concentrations of NOx were even lower than in Albacete, with minimum and maximum values of 3 and 40 µg/m^3 respectively (Figure S3). The maximum concentrations were found at 9:00h and 21:00h local times, showing that the main emissions of NOx also come from the traffic. The peak concentration of CO occurs simultaneously with those of NOx.

Ozone concentrations during the day ranged from 16 to 70 µg/m^3, higher than Albacete and Madrid, with a wider span between max and min data. Concerning SO_2, a peak at 13:00h-14:00h (12 µg/m^3 and 21 µg/m^3 for January and February, respectively) showed an additional, yet low, source other than traffic. The same behaviour was also observed for aromatic compounds. Their peak was at a different time (13:00h) than NOx revealing industrial sources. Toluene was, as in Madrid, the BTXs species with the highest concentration (13 µg/m^3 and 9 µg/m^3 in January and February, respectively, followed by xylenes). Also, and even more noticeable, during the day (from 9:00h to 17:00h) the concentration of NH_3 increased significantly, peaking at 13:00h, 200 µg/m^3. This peak of ammonia was observed in all the months of the study previous to the SARS-

CoV-2 crisis (Figure S3). The most probable source of NH_3 is the fertilizer factory placed in the town. PM_{10} showed maximum levels at midday, simultaneously with the industrial emissions.

During the lockdown (March - April) (Figure S3), the daily maxima of NOx fell below 16 µg/m^3, SO_2 below 9 µg/m^3, and toluene below 2.5 µg/m^3. The concentration of NH_3 also decreased substantially with respect to January and February, with a maximum value of 58 µg/m^3 and 33 µg/m^3, at 13:00h in March and April, respectively.

Figure 2c shows the average evolution of pollutants during the months January-June 2020. All of them show statistically significant differences at 95.0% confidence level comparing the average concentration between January and the months with the harsh restrictions. As in the other studied locations, the pollutant concentrations increased slightly during the de-escalation, but never reached the pre-lockdown values.

On the other hand, comparing the period March-June 2020 with respect to previous years (2013-2019) the concentration reduction percentages were significant for NOx (-66, -80,-52 and -60%); SO_2 (-59 to -67, -54 and -59 %); CO (-40, -46, -64 and -75 %), and toluene (-78, -79, -56 and -52 %) (Figure 3c). Nevertheless, for carbon monoxide and toluene the lack of data for several years makes these results less reliable (Table S3 and Figure S5).

For NH_3 there is also some data unavailable from the air-monitoring networks. Furthermore, the existing data for March-June (2013-2019) span from 0.3 to 94 µg/m^3, which suggests that the industrial source of ammonia is sporadic. For those reasons, the data from previous years have not been used to calculate the percentage change in 2020. Nevertheless, as it was stated above, the 2020 data for the period January-June (Figure 2c) show a significant decrease of NH_3 after the start of the crisis. Similar behaviour is also observed for NOx, CO, and toluene.

All these parameters reflect a direct effect of the drop of traffic and loss of industrial activity in the city due to the SARS-CoV-2 health crisis.

For particulate matter, PM_{10}, slight relative changes were observed in April 2020 (-11 %), May (+7 %) and June (+9 %) while a significant increase was registered in March (+47 %) (Figure 3c). The abnormally high value for March comes from the contribution of an intense Saharan intrusion event (18-22 March), Table S1, with daily average concentrations up to 80 µg/m^3. Similar PM_{10} values were measured in Ciudad Real, a city 40 km distant from Puertollano and so the high average PM_{10} concentration during March was not attributable to local industrial emissions or to the paralysis of activity.

3.4. San Pablo de los Montes. In the case of the monitoring site for a rural background reference, the data were expected to be little affected by traffic. In wintertime, the level of NOx generally remains below 4 µg/m^3 and below 1µg/m^3 for SO_2, close to the detection limit of the analyser. Thus, during the months before lockdown, (Fig S4), under such low concentrations of NOx, the ozone concentration remained high during the whole day with a very slight gap between night and daytime (ranging from 64 to 72 µg/m^3).

Shortly after the start of the crisis, in March (Figure S4), O_3 concentrations rose higher than during winter months, ranging from 72 to 85 µg/m^3 at 8:00h and 16:00h respectively and continued increasing during spring months,(Figure 2d). NOx concentrations fell below 1 µg/m^3 in March and April, increasing slightly (up to 1.3 µg/m^3) during the de-escalation. As it is shown in Figure 3d, the percentage changes during state of alarm are lower than those of other studied cities. In addition, for this remote placement, the NOx and SO_2 concentrations during March-June 2020 are essentially similar to those measured in the previous three years, 2017-2019 (Table S4 and Figure

S5). These results show the minimum effect of changes in local emissions due to the SARS-CoV-2 crisis and the negligible effect of remote emissions.

In S. Pablo, PM is measured through gravimetric methods which require more on-site technical support compared to unattended automatized methods, and no samples could be collected during the interval 14 March - 30 April. Moreover, the June data were not available during the writing process of this paper. The May data show an increase (+19 %) in $PM_{2.5}$ and a decrease in PM_{10} (-18 %), but the lack of information for the whole March-June period does not allow to derive a behaviour pattern.

3.5. Ozone behaviour. There are many factors involved in the ozone dynamics, making it more complex to observe a direct effect of lockdown compared to the case of primary pollutants. Thus, for example in Madrid, Table 1, ozone concentration was 14% higher for March-June 2020 than the corresponding average for the period 2013-2019. Nevertheless, during such years, O_3 had shown rising trends for March-June with average increases of 1.8, 1.7, 0.7 and 0.8 $\mu g/m^3$ per year, respectively. The percentages of change observed for these four months in 2020 (Figure 3a), are small and consistent with the trend of the previous years (Table 1, Figure S5). In fact, the results obtained for March, April and June 2020 are essentially equal to those measured in 2019. So, a clear effect of the alarm state and confinement on ozone concentration cannot be stated.

Similar rates of increase had been observed for S. Pablo in the period 2013-2019 (1.5, 1.8, 0.3 and 0 $\mu g/m^3$ per year for March -June, respectively) (Table S4, Figure S5). Nevertheless, for this remote site ozone decreased 9, 17, 9 and 8% in March-June 2020, respectively (Figure 3d).

On the other hand, for the period 2013-2019, ozone falls had been found for Albacete (-3.9, -3, -1.3 and -1.6 µg/m^3 per year, March-June respectively) and Puertollano (-2.6, -1.6, -2.2 and -2.3 µg/m^3 per year, March-June respectively). Although the 2020 drop compared with the average 2013-2019 (Tables S2 and S3) may suggest noticeable changes, the data from this year fits the decreasing trends for both sites (Figure S5). So, the effect of the crisis on ozone surface concentrations is not obvious. Mixed behaviour trends have also been reported for monthly mean ozone concentrations in major cities (Kumari and Toshniwal, 2020).

These results are consistent with previous studies. Thus, the EPA data reflect a very slight decrease of ozone for the period 2010-2018. Likewise, in Europe (EEA, 2018) the decline of ozone for the period 2001-2012 was below 10 %, despite the fact that NOx and VOCs emissions decreased about 40% between 2000 and 2016. On the other hand, some data from remote locations worldwide indicate that current ozone concentration is greater than during the 1980s (Gaudel et al., 2018) and there is no clear global pattern for surface ozone changes since 2000.

The fact that local concentrations of ozone have not changed significantly right after the start of the confinement of population is in part due to the high tropospheric ozone concentrations worldwide. The existence of local natural and anthropogenic surface sources of ozone, the transport due to exchange with the stratosphere and the long-range horizontal transport from highly polluted areas (Monks et al., 2015) keep the tropospheric ozone's level high. In this sense tropospheric ozone is a global issue, as is CO_2. On the other hand, for the rest of atmospheric pollutants studied in this work, they are not uniformly distributed in the atmosphere, showing high concentrations only near the sources and in time scales relatively short from their emissions. Thus, mixing with surrounding air masses leads effectively to the dispersion and fast decrease in the local concentrations of the emitted compounds. Also the reactivity and lifetime of each pollutant influence the time profiles. Since ozone concentration is also high in rural or areas surrounding the sources, mixing with surrounding air masses tend to keep ozone nearly constant. This mechanism is expected to delay the long-term effects of environmental policies or even dispel short–term events of economic and health crises.

Furthermore, ozone is a secondary pollutant involved in different atmospheric reactions mechanisms that act as sources and sinks. Thus for example, for high NO levels, such as in Madrid, especially under temperature inversions and stagnation events, surface O_3 concentration is largely VOCs-limited (Monks et al., 2015). High NO concentrations tend to consume ozone through titration ($NO+O_3=NO_2+O_2$) leading to low concentrations of O_3. In this sense, a local decrease of NO concentration was expected to result in an increase of ozone in Madrid, similar to those observed in other large cities (Sicard et. al., 2020; Tobias et al., 2020). Nevertheless, NO_2 and VOCs, which are ozone precursors, have experienced similar drops since the start of the SARS-CoV-2 crisis, which may have counteracted the effect of NO, softening the rise of ozone.

On the contrary, in non-polluted sites, such as in S. Pablo, the local production of ozone is NOx-limited. Since most VOCs come from natural local sources and are not affected by the lockdown in remote cities, a small decrease in NOx may explain the decrease in ozone in March, April, May and June 2020. On the other hand, Albacete and Puertollano have intermediate environments. Their decreases in ozone concentration during the lockdown suggest that ozone generation is mainly NOx-limited. Nevertheless, given the previous decreasing ozone trends in these two sites (2013-2019) other factors different from the lockdown may have contributed to the net change in O_3.

4 Conclusions

What is unique in this pandemic-driven economic crisis is the instantaneous standstill of entire countries (within hours) compared to the gradual loss of activity observed in previous "conventional" economic crises. As a result, a fast decrease of surface atmospheric primary pollutants (within days) has been observed associated with the confinement of population and the sudden stop of the economy, including productive systems and transport. This short-term effect on the concentrations of air pollutants has been deeper than for any other economic previous crises, including the 2008 recession.

Nevertheless, the atmospheric levels of air pollutants are highly variable, seasonal and strongly dependent on meteorological conditions and, so, some previous assessments based on too short periods databases (in the order of weeks) may result misleading. Thus, in this work a wide database (2013-2020) has been used to infer changes in atmospheric parameters due to the SARS-CoV-2 lockdown. As shown in the results and discussion section, the conclusions would be different if we just compared with the previous months or year ignoring the historic trend of atmospheric pollutants. The results show statistically significant drops of NO_x, CO, BTXs, NMHC and NH_3, during March and April compared to the same months in the period 2013-2019. In May and June, with the gradual de-escalation, traffic inside cities recovered up to 80% of the average before the crisis, increasing emissions and inducing the rise of air pollutants with respect to the previous months. Nevertheless, the levels of air pollutants during May and June 2020 were still below the average for the period 2013-2019.

Although the three studied cities are very different in size and population, similar changes in primary pollutants have been found for these sites which share similar continental climate conditions and traffic as the main air pollution source. Additionally, for Puertollano, the industrial sources were also affected by the confinement. In the case of the rural background site, San Pablo de los Montes, data show slight changes in local emissions due to the SARS-CoV-2 crisis and the negligible effect of remote emissions. Up to our knowledge this is the first work reporting the effect of the pandemic on a remote background area.

For all the studied sites, PM changes were small due to the existence of other natural sources, such as the Sahara dust intrusions or emissions from the residential sector, including household and gardening combustion, and a clear correlation of $PM_{2.5}$ or PM_{10} with traffic emissions, could not be stated. Likewise, the changes observed in ozone concentrations have been assessed comparing with months previous to the crisis and with the same month interval (March-June) for the period 2013-19. Each studied site shows different ozone patterns, but in all cases the observed changes in March-June are statistically within the historical trends and so they are not clearly attributable to the lockdown. The results found in this work show that local and regional tools are needed to understand the specific environment of given cities.

The fast response of atmospheric pollutants to the measures to mitigate the spread of SARS-CoV-2 provides interesting conclusions which may be useful in the implementation of future actions or technologies addressed to improve air quality. Thus for example, in cities, the massive use of electric cars replacing fossil- fuel technology is expected to have a similarly fast and beneficial effect on local environments such as those exemplified in this study. On the other hand, environmental policies are not expected to have an immediate response on secondary pollutants such as surface ozone or on atmospheric aerosols.

The de-escalation of people's lockdown and the progressive return to activity will lead slowly to the resurgence of air quality problems in cities. The preferential use of private cars to the detriment of public transport to avoid new infections may also lead to the increase of traffic emissions.

Furthermore, the International Monetary Fund' prospects foresee for 2020 the worst financial crisis since the Great Depression. Consequently, the current crisis, like others before (Pacca et al., 2020), is expected to have medium and long-term effects on air quality. The major risk is that environmental policies may result shelved to balance budgets and face other urgent needs, as it happened in previous economic crises (Botetzagias et al., 2018).

Finally, the lack of essential supplies in the global market during the hardest weeks of the SARS-CoV-2 crisis may result in the generalized local production of critical and strategic goods to face alike future challenges. This partial de-globalization of the economy may imply the appearance of new industries worldwide with new consequences for local or regional environments. So, further studies will be required to assess the long-term effect of SARS-CoV-2 on air quality, and vice versa. Thus, for example it has been found that in sites with high levels of particulate matter (Ciencewicki and Jaspers, 2007; Wu et al., 2020) the spread of virus is faster than in cleaner environments and population is affected in a greater extent by respiratory diseases which also means greater dangers under SARS-CoV-2 infection.

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Conflicts of interest. There are no conflicts to declare.

The databases used in this work are all available and accessible through the links in references: DGT, 2020; Castilla-La Mancha 2020; Madrid, 2020 and MITECO, 2020a.

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