Changes in air pollution during COVID-19 lockdown in Spain: A multi-city study

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ABSTRACT

The COVID-19 pandemic has escalated into one of the largest crises of the 21st Century. The new SARS-CoV-2 coronavirus, responsible for COVID-19, has spread rapidly all around the world. The Spanish Government was forced to declare a nationwide lockdown in view of the rapidly spreading virus and high mortality rate in the nation. This study investigated the impact of short-term lockdown during the period from March 15th to April 12th 2020 on the atmospheric levels of CO, SO2, PM10, O3, and NO2 over 11 representative Spanish cities. The possible influence of several meteorological factors (temperature, precipitation, wind, sunlight hours, minimum and maximum pressure) on the pollutants’ levels were also considered. The results obtained show that the 4-week lockdown had significant impact on reducing the atmospheric levels of NO2 in all cities except for the small city of Santander as well as CO, SO2, and PM10 in some cities, but resulted in increase of O3 level.

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Introduction

The global spread of COVID-19 has escalated into one of the largest health and economic crises of the 21st Century (Yu et al., 2020). Spain has recorded one of the highest mortality rates with 266,194 confirmed cases and 28,424 confirmed deaths (ISCIII, n.d.) (as on 22nd July 2020) in a population of about 47 million people. Experts (Verity et al., 2020) have warned that there could be many more cases due to the existing uncertainty of the data, especially in the developing countries. This life-threatening pathogen has been identified as a zoonotic coronavirus closely related (88%–89% similarity) to two bat-derived severe acute respiratory syndrome-like coronaviruses, bat-SL-CoVZC45, and bat-SL-CoVZXC21 (Lai et al., 2020). Other close known human coronavirus are SARS-CoV (79% similarity) and Middle East respiratory syndrome coronavirus (MERS-CoV) (50% similarity) (Jiang et al., 2020; Lu et al., 2020; Ren et al., 2020).

Cultural variations have been evident in people’s reactions to state control measures in response to the growing COVID-19 pandemic (Chiu et al., 2020). In Spain, the government was one of the first to declare a lockdown on March 14th, 2020 (starting midnight March 15th). From then on, unprecedented containment and mitigation policies were taken to limit the spread of COVID-19, including working from home when possible.
sible, travel restrictions, confinement and quarantine, cancel-
lization of mass events, online education for schoolchildren and
university students, closure of bars, restaurants and clubs or
prohibition of public gatherings. Only shops selling essentials,
such as supermarkets, pharmacies and a few industrial ac-
tivities were allowed. On March 30th, 2020, the Spanish Gov-
ernment, forced by the overburdened intensive care units and
health systems and encouraged by the scientific community
(Mitjá et al., 2020), reinforced the lockdown measures to a
complete lockdown for two weeks more until April 13th, lead-
ing to one of the world’s major social distancing measures,
in which only essential work was permitted and many indus-
trial activities such as construction were forbidden. After one
month of lockdown, positive results were obtained in flattening
the epidemic’s curve (Tobías, 2020).

This pandemic has forced behavioral changes in our so-
ciety in contradiction to the common routines which could
provide a useful insight into more sustainable supply and produc-
tion patterns (Sarkis et al., 2020). The importance of reduc-
ing air pollution is understood on account of its well-
known impact on climate change and its influence on health
due to increased morbidity and mortality (Manisalidis et al.,
2020; Xiao et al., 2018). Previous studies suggested that amb-
ient air pollutants are risk factors for respiratory infections
(Becker and Soukup, 1999; Horne et al., 2018; Xie et al., 2019;
Xu et al., 2016) such as COVID-19. And even though there are
contradictory opinions on the transmission of SARS-CoV-2, it
seems that people can acquire this coronavirus through the
air (Morawska and Cao, 2020) due to its stability in aerosols
(van Doremalen et al., 2020) and the fact that the gas cloud
and its payload of pathogen-bearing droplets of all sizes can
travel 7–8 m (Bourouiba, 2020; Morawska and Cao, 2020). Also,
a significant proportion of confirmed cases have recently been
associated with air pollution in 120 Chinese cities (Zhu et al.,
2020).

Particulate matter, NO₂, CO, O₃, and SO₂ are among the
most frequently monitored pollutants when assessing air qual-
y. Particulate matter (PM) in the form of small parti-
cles of 10 µm or less (PM₁₀) can penetrate the respiratory
system via inhalation, causing cancer and respiratory dis-
eases, among many other health problems (Chen et al., 2012;
Wu et al., 2019). Some authors have attributed five specific fac-
tors to PM₁₀: secondary inorganic aerosol, combustion, crustal
dust, vehicle exhaust, and biomass burning (Liu et al., 2020).
Dust resuspension has been also pointed out as a source of particulate matter (Fenech and Aquilina, 2020). Although
stratospheric O₃ plays crucial role in protecting ecosystem at
earth’s surface from ultraviolet irradiation, the presence of
certain O₃ concentrations at ground level is harmful to the
respiratory and cardiovascular system due to its high reac-
tivity as an oxidative gas (Nuvolone et al., 2018). Nitrogen
oxide (NOₓ), sulfur dioxide (SOₓ), and carbon monoxide (CO)
are all considered air pollutants that are harmful to humans,
being both short and long-term exposure-related with pul-
monary diseases (Manisalidis et al., 2020), for example, their
current pollution levels in developed European countries such
as Spain have been found to have an important health im-
 pact (Arroyo et al., 2019). Air pollution is known to cost Euro-
pean countries hundreds of millions of euros in health needs,
the environment (European Environment Agency, 2014), loss
of productivity and associated co-morbidities (Trasande et al.,
2016).

The aim of this study was to analyze and quantify the changes in air quality achieved after lockdown in Spain using
CO, SO₂, PM₁₀, O₃, and NO₂ data from March 15th to April
12th 2020, including what is hereafter referred to as the mi-
nor lockdown (from March 15th to March 29th) and major lock-
down (from March 30th to April 12th), for which pollution
data from the normal (non-lockdown) and lockdown periods
were collected and analyzed. Meteorology can play an impor-
tant role in air pollution, transport, deposition and transfor-
mation and can bring severe pollution on days even when
the total emission is reduced (Li et al., 2017; Shi et al., 2018;
Wang et al., 2019, 2020). In this regard, it is worth noting
that Spain, despite lying in a temperate zone (Briz-Redón and
Serrano-Aroca, 2020), is the most climatically diverse country
in Europe and one of the ten most climatically diverse coun-
tries in the world (Ministerio de Medio Ambiente y Medio Rural
y Marino. AEMET, 2011). The changes in diverse meteorological
factors (temperature, humidity, wind, pressure, and sunlight)
were registered during the study.

1. Data collection

1.1. Pollution data

A total of 11 of Spain’s largest cities were considered in the
study (see Fig. 1).

The population of these cities (on 1st January 2019) ranged
from 97,260 to 3266,126 inhabitants (see Table 1) (INE,
2019), all subject to a high human impact in terms of
pollution. Pollution data was downloaded from several region-
al and local webpages: Barcelona (Catalunya, n.d.); Bilbao
(Euskadi.eus, 2020); Lleida (Catalunya, n.d.); Madrid
(Ayuntamiento de Madrid, 2020); Pamplona (“Calidad del aire
- navarra.es,” 2020); Santander (Gobierno de Cantabria.
Consejería de Medio Ambiente. Ordenación del Territorio y
Urbanismo, 2020); Santiago de Compostela (Xunta de Galicia,
2020); Sevilla (CMACOT, 2020); Valencia (GVA, 2020); Vigo
(Xunta de Galicia, 2020); Zaragoza (Ayuntamiento de Zaragoza,
2020). One traffic station was selected for data collection and
analysis of the cities studied (see Table 1).

The traffic stations’ pollution level was determined pre-
dominantly by the emissions from nearby traffic (UK Depart-
ment for Environment Food and Rural Affairs, n.d.). Daily lev-
els of CO (in mg/m³), NO₂ (in µg/m³), PM₁₀ (in µg/m³), O₃ (in
µg/m³), and SO₂ (in µg/m³) were collected for each station and
day from March 4th - April 14th 2019 and March 2nd-April 12th
2020, i.e. the first 28 days of lockdown, two weeks before the
start of the lockdown and the same period of 2019.

1.2. Meteorology data

Meteorological data was downloaded from the OpenData plat-
form of the State Meteorological Agency (AEMET) via its API.
The meteorological stations closest to the cities were chosen
to obtain the meteorological variables of interest: temperature
(°C), precipitation (mm), wind speed (km/hr), sunlight hours
Fig. 1 – Locations of the 11 cities considered for the analysis. The coordinates used were those of the cities’ pollution stations. The borders of the different Spanish provinces are plotted in gray.

### Table 1 – City, province and inhabitants (as on 1st January 2019) and positions of air quality stations selected for the 11 cities.

<table>
<thead>
<tr>
<th>City</th>
<th>Province</th>
<th>Population</th>
<th>Station</th>
<th>Longitude</th>
<th>Latitude</th>
</tr>
</thead>
<tbody>
<tr>
<td>Barcelona</td>
<td>Barcelona</td>
<td>1636,762</td>
<td>Eixample</td>
<td>2.1538</td>
<td>41.3853</td>
</tr>
<tr>
<td>Bilbao</td>
<td>Biscay</td>
<td>346,843</td>
<td>María Díaz de Haro</td>
<td>−2.9457</td>
<td>43.2588</td>
</tr>
<tr>
<td>Lleida</td>
<td>Lleida</td>
<td>138,956</td>
<td>Lleida</td>
<td>0.6157</td>
<td>41.6158</td>
</tr>
<tr>
<td>Madrid</td>
<td>Madrid</td>
<td>3266,126</td>
<td>Escuelas Aguirre</td>
<td>−3.6823</td>
<td>40.4217</td>
</tr>
<tr>
<td>Pamplona</td>
<td>Navarre</td>
<td>201,653</td>
<td>Plaza de la Cruz</td>
<td>−1.6402</td>
<td>43.4685</td>
</tr>
<tr>
<td>Santander</td>
<td>Cantabria</td>
<td>172,539</td>
<td>Tetuán</td>
<td>−3.7904</td>
<td>42.8113</td>
</tr>
<tr>
<td>Santiago de Compostela</td>
<td>A Coruña</td>
<td>97,260</td>
<td>San Caetano</td>
<td>−8.5311</td>
<td>42.8878</td>
</tr>
<tr>
<td>Sevilla</td>
<td>Seville</td>
<td>688,592</td>
<td>Torneo</td>
<td>−6.0028</td>
<td>42.2224</td>
</tr>
<tr>
<td>Valencia</td>
<td>Valencia</td>
<td>794,288</td>
<td>Avenida Francia</td>
<td>−0.3428</td>
<td>39.4575</td>
</tr>
<tr>
<td>Vigo</td>
<td>Pontevedra</td>
<td>295,364</td>
<td>Lope de Vega</td>
<td>−8.7111</td>
<td>41.6559</td>
</tr>
<tr>
<td>Zaragoza</td>
<td>Zaragoza</td>
<td>674,997</td>
<td>Avenida Soria</td>
<td>−0.8992</td>
<td>41.6559</td>
</tr>
</tbody>
</table>

(number of hours over irradiance threshold of 120 W/m²), and min and max atmospheric pressure (hPa).

## 2. Method

### 2.1 Software

The statistical analysis was carried out on R programming language (Team, 2020). The R packages `effects` (Fox and Weisberg, 2018), `ggplot2` (Wickham, 2016), `lubridate` (Glehemund and Wickham, 2011), `RCurl` (Lang and the CRAN team, 2015), `sjPlot` (Ludecke, 2016) and `XML` (Lang, 2020) were specifically required for some parts of the study.

### 2.2 Statistical model

Since the effects of weather patterns (temperature, rain, wind speed, sunlight hours, or atmospheric pressure) can significantly affect ground-level pollutant concentrations and thereby compromise the observable effects of the lockdown (Venter et al., 2020), these changing meteorological factors were taken into account in the statistical model. Weekends were also included as there was less traffic and lower pollutant levels. The daily pollutant levels (CO, SO₂, PM₁₀, O₃, NO₂) were fitted through five statistical models (one per pollutant) accounting for meteorological conditions, weekends and lockdown periods. The level of each pollutant in a city i on date t was modeled through the following linear regression shown in Eq. (1).
Pollutant_{it} = \alpha + \beta_1 \text{Temperature}_{it} + \beta_2 \text{Rain}_{it} + \beta_3 \text{Wind}_{it} + \beta_4 \text{Sunlight}_{it} + \beta_5 \text{Max\_pressure}_{it} + \gamma \text{Weekend}_{i} + \delta_1 \text{City}_{i} + \delta_2 \text{Minor\_lockdown}_{i} + \delta_3 \text{Major\_lockdown}_{i} \tag{1}

where, \( \alpha \) is the global intercept of the model, \( \beta_k (k = 1, 2, 3, 4, 5) \) measures the effect of the corresponding meteorological covariate, measured for every city and day, on the value of Pollutant_{it}; \( \gamma \) measures the effect of weekend days on the value of Pollutant_{it}; \( \delta_1 \) represents the overall city-specific effect (normal pollutant levels in the absence of lockdown), \( \delta_1 \) and \( \delta_2 \) measure the overall effect of the minor and major lockdown, respectively, on the values of Pollutant_{it}; \( \delta_1 \) and \( \delta_2 \) measure the city-specific effect of the minor and major lockdown, respectively, on the value of Pollutant_{it}. The variables Weekend, Minor\_lockdown, and Major\_lockdown are all binary (0/1), allowing each date within the period under study to be fully characterized. Several bank holidays in the study periods were also considered as weekend days.

The regression model can estimate the magnitude of the variation of each pollutant due to lockdown in each of the cities considered while accounting for meteorological and weekday effects. The results of this model therefore provide information on the different pollutant levels between the lockdown (major and minor) and normal (no lockdown) periods.

3. Results and discussion

3.1. Exploratory analysis of pollutant levels across cities and periods

Before applying the regression models, which were the main analytical tools, percentage variations in pollutant levels between 2019 and 2020 were computed for each city. This exploratory analysis showed remarkable variations in pollutant levels and inter-city heterogeneity, although the regression models allowed us to better assess the lockdown effect.

Fig. 2 shows the percentage variations in pollutant levels between March 17th-March 31st 2019 and March 15th-March 29th 2020 (Fig. 2a), and between April 1st-April 14th 2019 and March 30th-April 12th 2020 (Fig. 2b). In general, pollutant levels decreased in 2020, especially for NO\textsubscript{2}. The rest of the pollutants also present lower concentration levels in many cities, with certain increases. The following sections focus on the results provided by the linear regression model expressed in Eq. (1).

3.2. Correlation analysis of the meteorological variables

The correlation analysis of the meteorological variables shows that all the pairwise correlation values between the meteorological variables were moderate to low (see Table 2).

Multicollinearity issues were thus discarded except for atmospheric pressure. Only maximum pressure was finally considered because of the high correlation found between the maximum and the minimum pressure.

3.3. Meteorological, weekend and overall lockdown effects on pollutant levels

Table 3 shows the overall coefficients estimated for the models fitted with Eq. (1), considering the five pollutants under analysis.

The effect of meteorological covariates on the pollutant values indicates that the concentration of pollutants increases with sunlight hours. The mean temperature also shows a positive association with the concentration of pollutants, except for O\textsubscript{3}. Precipitation values show no association with any of the pollutants, and wind speed has a negative association with all pollutant concentrations but O\textsubscript{3}. The maximum pressure is more unstable because its effect varies with pollutant type. The major lockdown days clearly show a negative correlation with all the compounds studied except ozone, which can be expected.

In general, the results obtained are quite consistent with the literature. On the one hand, dry and sunny weather leads to thermal inversion, which prevents vertical pollutant dispersion, so that their concentration increases, and they generate significant smog episodes. On the other hand, higher wind speed can reduce air pollution (Mao et al., 2020; Radzka, 2020; Yousefian et al., 2020). Nevertheless, it is important to highlight that very low wind speed can also be associated with reduced PM in the air due to higher deposition (Xu et al., 2020). High pressures seem to have a positive effect on the formation of pollution, especially PM, as particulates cannot disperse properly and tend to concentrate, which contributes to high atmospheric particulate loads (Czerwinska and Wielgosinski, 2020; Hoque et al., 2020).

The case of O\textsubscript{3} should be analyzed separately. While the UV radiation produced in higher sunlight hours showed positive correlations with O\textsubscript{3} in different studies (Mao et al., 2020), which is consistent with our results, higher temperatures are usually associated with higher O\textsubscript{3} pollution (Dong et al., 2020; Mao et al., 2020). The mismatch of this association with the results in Table 3 should be highlighted and can be attributed to the short period of the data analyzed, i.e. low representativeness, or the capability of the meteorological variables to affect the formation of O\textsubscript{3}, among others.

3.4. Analysis of coefficients of determination ($R^2$)

The capability of the meteorological variables to explain data variability was generally low, as can be seen from the analysis of the coefficients of determination ($R^2$) associated with the models (see Table 4).

This analysis shows that including city-level effects and the lockdown period are essential to achieve reasonably high $R^2$ values, while the meteorological covariates only explain a small percentage of data variability, which explains some mismatches between our results and the literature.

3.5. City-specific marginal effects for pollutant levels

The results shown in Table 3 suggest that both the minor and major lockdown led to overall CO, NO\textsubscript{2}, and PM\textsubscript{2.5} reductions, while SO\textsubscript{2} was only reduced during the major lockdown and O\textsubscript{3} increased during lockdown. However, a full analysis of
Fig. 2 – Percentage variations in pollutant levels in each of the cities under study between March 17th-March 31st 2019 and March 15th-March 29th 2020 (a), and between April 1st-April 14th 2019 and March 30th-April 12th 2020 (b).

Table 2 – Correlation matrix for the meteorological variables considered for the analysis.

<table>
<thead>
<tr>
<th></th>
<th>Temperature</th>
<th>Precipitation</th>
<th>Wind speed</th>
<th>Sunlight hours</th>
<th>Min pressure</th>
<th>Max pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature</td>
<td>1.000</td>
<td>−0.211</td>
<td>−0.031</td>
<td>0.264</td>
<td>0.414</td>
<td>0.405</td>
</tr>
<tr>
<td>Precipitation</td>
<td>−0.211</td>
<td>1.000</td>
<td>0.157</td>
<td>−0.411</td>
<td>−0.136</td>
<td>−0.096</td>
</tr>
<tr>
<td>Wind speed</td>
<td>−0.031</td>
<td>0.157</td>
<td>1.000</td>
<td>−0.062</td>
<td>0.056</td>
<td>0.103</td>
</tr>
<tr>
<td>Sunlight hours</td>
<td>0.264</td>
<td>−0.411</td>
<td>−0.062</td>
<td>1.000</td>
<td>0.103</td>
<td>0.081</td>
</tr>
<tr>
<td>Min pressure</td>
<td>0.414</td>
<td>−0.136</td>
<td>0.056</td>
<td>0.103</td>
<td>1.000</td>
<td>0.990</td>
</tr>
<tr>
<td>Max pressure</td>
<td>0.405</td>
<td>−0.096</td>
<td>0.103</td>
<td>0.081</td>
<td>0.990</td>
<td>1.000</td>
</tr>
</tbody>
</table>

Table 3 – Meteorological, weekend and overall lockdown effects on pollutant levels analyzed by Eq. (1). The estimated coefficients of the model are shown along with the associated p-values.

<table>
<thead>
<tr>
<th></th>
<th>CO</th>
<th>NO₂</th>
<th>O₃</th>
<th>PM₁₀</th>
<th>SO₂</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intercept(α)</td>
<td>−0.0676</td>
<td>0.8900</td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
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<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature (β₁)</td>
<td>0.0021</td>
<td>0.1151</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
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<td></td>
</tr>
<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Precipitation (β₂)</td>
<td>−0.0001</td>
<td>0.7932</td>
<td></td>
<td></td>
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<tr>
<td>Estimate</td>
<td></td>
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<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>Wind speed (β₃)</td>
<td>−0.0066</td>
<td>0.0000</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
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</tr>
<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sunlight hours (β₄)</td>
<td>0.0026</td>
<td>0.0033</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Max pressure (β₅)</td>
<td>0.0004</td>
<td>0.3730</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Weekend day (γ)</td>
<td>−0.0213</td>
<td>0.0009</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Minor lockdown day (δ₁)</td>
<td>−0.1610</td>
<td>0.0000</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>p-value</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Major lockdown day (δ₂)</td>
<td>−0.1629</td>
<td>0.0000</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Estimate</td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>p-value</td>
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</tr>
</tbody>
</table>
the model proposed for each pollutant requires the estimated city-specific coefficients, which are omitted in Table 4. Indeed, the overall city-specific effects, total period (no lockdown, minor lockdown, or major lockdown) effects and city-specific effects corresponding to city-period interactions were used to estimate the city-specific marginal effects of the pollutants considered. In brief, these city-specific marginal effects can be interpreted as the mean contribution of each period to the level of a pollutant for a given city. In other words, the computation of these effects is helpful to appreciate the mean variation in pollutant levels that can be attributed to each lockdown period for each city. The function plot_model of the R package sjPlot (see Section 3.1) specifically designed to compute the marginal effects associated with the regression model (Eq. (1)) was used. The marginal effects and associated 95% confidence intervals are shown in Figs. 3 to 7. Statistically significant differences between marginal effects can be identified when the associated confidence intervals do not overlap.

### Table 4 - Coefficients of determination ($R^2$) obtained from modeling the values measured for each of the five pollutants under study, considering an increasing subset of the variables included in the statistical model. The values of the last row are those of the complete model provided by Eq. (1).

<table>
<thead>
<tr>
<th>Model</th>
<th>CO</th>
<th>NO$_2$</th>
<th>O$_3$</th>
<th>PM$_{10}$</th>
<th>SO$_2$</th>
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<td>0.7969</td>
<td>0.4871</td>
<td>0.5204</td>
<td>0.7762</td>
</tr>
</tbody>
</table>

### Fig. 3 – City-specific marginal effects estimated by Eq. (1) for NO$_2$ levels in each of the three study periods. Differences between marginal effects are statistically significant when the associated confidence intervals do not overlap.

However, due to being comparatively small, Santander did not show significant changes after the COVID-19 lockdown, with lower NO$_2$ pollution (see no lockdown level in Fig. 3). However, the differences between the reduced NO$_2$ concentrations during the major and minor lockdown were not statistically significant for most cities. Only Bilbao, which has a lot of industry, showed significant differences. NO$_2$ is mainly produced from oxidation of NO by O$_3$ and peroxyl radicals, while NO is mainly from direct emissions from various combustion processes, including vehicles, industrial boilers, power plants and ships (Tobias et al., 2020). Therefore, the significant reduction of NO$_2$ accompanied with rising O$_3$ concentration (see Fig. 4) during the lockdown period can be mainly attributed to the reduction of NO emission sources.

#### 3.5.1. City-specific marginal effects for NO$_2$ levels

Fig. 3 shows that the estimated reductions of NO$_2$ were statistically significant in 10 cities during the minor and major COVID-19 lockdown with respect to the no lockdown levels. These reductions are large in the most heavily populated cities: Madrid, Barcelona, Sevilla and Valencia as expected.

#### 3.5.2. City-specific marginal effects for O$_3$ levels

The result obtained for the NO$_2$ levels contrasts with those found for O$_3$ (see Fig. 4). An increasing trend of O$_3$ levels during lockdown can thus be seen, although there are only statistically significant differences for Barcelona and Valencia. Most O$_3$ increases are not significant or are moderate compared to NO$_2$ reductions (Fig. 3). Since weather changes were taken into account in the statistical model, this phenomenon can be explained as a decline in anthropogenic-origin NO$_2$ levels in urban areas in VOC-limited environments, which leads to reduced local titra-
tion of NO\textsubscript{x} and thus to a more moderate increase of O\textsubscript{3} levels (Souza and Ozonur, 2019; Tobias et al., 2020). It is important to mention that NO\textsubscript{x} concentrations can fall faster than those of other pollutants because they can also participate in other atmospheric reactions, among other causes (Dadashi et al., 2020).

3.5.3. City-specific marginal effects for PM\textsubscript{10} levels
Lockdown periods also led to a general reduction in particulate matter (PM\textsubscript{10}) levels (Fig. 5).

Significant reductions of PM\textsubscript{10} levels were found during lockdown mainly and more clearly in three cities: Barcelona, Valencia and Sevilla, although Barcelona did not show significant reductions. This effect can be attributed to the size of the cities and the activities carried out in them: the larger the city, the higher the anthropogenic influence (combustion, vehicle exhaust, etc.) due to transport, agriculture and industry (Wang et al., 2020). However, this cannot be deduced in all cases, as in Madrid, the biggest city in Spain. In fact, the differences in PM\textsubscript{10} concentrations were not significant in the rest of the cities. These results make sense, since the decrease was not expected to be very high since these inhalable particles persist in the air for long periods and are emitted in large quantities (Xiao et al., 2018). The lower percentage of particulate matter from natural sources such as the sea (as in the case of coastal cities such as Vigo, Valencia or Barcelona) or wind-blown dust (Madrid or Santiago de Compostela) cannot be ignored (Cellis et al., 2004).

3.5.4. City-specific marginal effects for SO\textsubscript{2} levels
The lockdown periods also led to a reduction in SO\textsubscript{2} levels in some of the cities (Fig. 6).

Significant reductions of SO\textsubscript{2} levels were found during the lockdown in Bilbao, Zaragoza, and Santiago de Compostela, with more discreet variations in the others. In many cities there were either no significant differences in SO\textsubscript{2} levels or in-
significant variations, e.g. in Vigo and Pamplona. As in the case of NO$_2$ and PM$_{10}$, this pollutant has an anthropogenic origin mostly attributed to traffic and shipping.

### 3.5.5. City-specific marginal effects of CO levels

The lockdown periods also reduced CO levels in some of the largest cities (Fig. 7).

The significant reductions of CO levels during the lockdown in Barcelona, Santiago de Compostela and Sevilla can be attributed to the already mentioned reduction of their anthropogenic activity. However, many of the other cities had no significant differences in CO levels, although robust conclusions cannot be extracted. The reduced CO, which is also a precursor of O$_3$ like NO and NO$_2$ (Yousefian et al., 2020) in Barcelona and Sevilla are in agreement with the higher O$_3$ level found there during the lockdown (see Figs. 3, 4 and 7).

To sum up, the minor and major Spanish COVID-19 lockdown periods were not long enough to reduce air pollution in all its forms, although some initial differences were found. Firstly, a significant reduction of NO$_2$ was achieved in most cities, mainly due to the significant reduction in transport and industry. Secondly, SO$_2$, CO and PM$_{10}$ were only reduced in some of the cities, which is beneficial because these chemical compounds can be transformed into H$_2$SO$_4$ and HNO$_3$ through acid rain, which pollute air and water (Gerhardsson et al., 1997). The ozone concentration increased in some cities during lockdown, associated with the reduced NO$_2$ and CO levels, in agreement with previous studies (Tobías et al., 2020; Uttamang et al., 2020; Wang et al., 2020). Air pollutant levels can be expected to fall further in the forthcoming weeks because of the longer-term effects of the restrictions and the lifestyle changes during the global health and economic crises. These reduced air pollutants can
be expected to improve the prognosis of patients with respiratory diseases, as has been suggested in previous studies (Becker and Soukup, 1999; Horne et al., 2018; Xie et al., 2019; Xu et al., 2016).

4. Conclusions

The variations of air pollutants during the COVID-19 lockdown in Spain were studied as a unique opportunity to evaluate the effects of the reduction of emission sources for consideration in future air quality policies. Previous studies on this health crisis found that changes in atmospheric pollutant levels may not be directly attributed to the lockdown due to the variety of factors involved in their causes, including the weather and their regional or long-distance transport. A regression model was used in this study to measure pollutant variations during the Spanish lockdown while considering the influence of the main meteorological factors involved. The results indicate that the major and minor COVID-19 lockdowns were not long enough to significantly improve the air quality as regards all the analyzed pollutants (CO, SO2, PM10, O3, and NO2). However, some improvements in NO2, CO, SO2, and PM10 levels were found in some cities, while the O3 pollution level was found to increase. However, it should be noted that the confidence intervals of the results obtained advise treating them with a certain amount of caution. An in-depth analysis of the long-term effects should be carried out to gain further information on this subject.

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