

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

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

Álvaro Briz-Redón^{1,**}, Carolina Belenguier-Sapiña^{2,**},
Ángel Serrano-Aroca^{3,*}

¹Statistics Office, City Council of Valencia, c/Arquebisbe Mayoral, 2, 46002 Valencia, Valencia, Spain

²Department of Analytical Chemistry, Faculty of Chemistry, University of Valencia, c/Doctor Moliner 50, 46100 Burjassot, Valencia, Spain

³Centro de Investigación Traslacional San Alberto Magno, Universidad Católica de Valencia San Vicente Mártir, c/Guillem de Castro 94, 46001 Valencia, Valencia, Spain

ARTICLE INFO

Article history:

Received 15 May 2020

Revised 29 July 2020

Accepted 31 July 2020

Available online 4 August 2020

Keywords:

COVID-19

Lockdown

Air quality

Pollution

Meteorology

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.

© 2020 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences. Published by Elsevier B.V.

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 (ISCI, 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 coun-

tries. 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 pos-

* Corresponding author.

E-mail: angel.serrano@ucv.es (Á. Serrano-Aroca).

** These authors contributed equally

sible, travel restrictions, confinement and quarantine, cancellation 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 activities were allowed. On March 30th, 2020, the Spanish Government, forced by the overburdened intensive care units and health systems and encouraged by the scientific community (Mitjà et al., 2020), reinforced the lockdown measures to a complete lockdown for two weeks more until April 13th, leading to one of the world's major social distancing measures, in which only essential work was permitted and many industrial 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 society in contradiction to the common routines which could provide a useful insight into more sustainable supply and production patterns (Sarkis et al., 2020). The importance of reducing 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 ambient 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 quality. Particulate matter (PM) in the form of small particles of 10 µm or less (PM₁₀) can penetrate the respiratory system via inhalation, causing cancer and respiratory diseases, among many other health problems (Chen et al., 2012; Wu et al., 2019). Some authors have attributed five specific factors 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 reactivity as an oxidative gas (Nuvolone et al., 2018). Nitrogen oxide (NO₂), sulfur dioxide (SO₂), and carbon monoxide (CO) are all considered air pollutants that are harmful to humans, being both short and long-term exposure-related with pulmonary 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 impact (Arroyo et al., 2019). Air pollution is known to cost European 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 *minor lockdown* (from March 15th to March 29th) and *major lockdown* (from March 30th to April 12th), for which pollution data from the normal (non-lockdown) and lockdown periods was collected and analyzed. Meteorology can play an important role in air pollution, transport, deposition and transformation 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 countries 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 regional 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 (CMAOT, 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 predominantly by the emissions from nearby traffic (UK Department for Environment Food and Rural Affairs, n.d.). Daily levels 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 platform 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

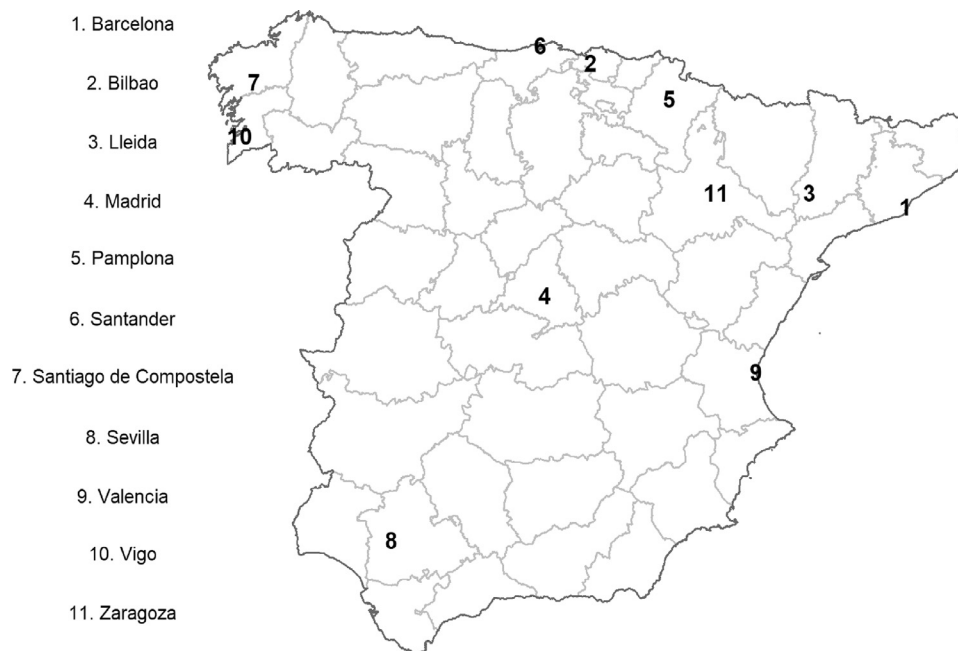


Fig. 1 – Locations of the 11 cities considered for the analysis. The coordinates used were those of the cities' pollution stations. The borders of the different Spanish provinces are plotted in gray.

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

City	Province	Population	Station	Longitude	Latitude
Barcelona	Barcelona	1636,762	Eixample	2.1538	41.3853
Bilbao	Biscay	346,843	María Díaz de Haro	-2.9457	43.2588
Lleida	Lleida	138,956	Lleida	0.6157	41.6158
Madrid	Madrid	3266,126	Escuelas Aguirre	-3.6823	40.4217
Pamplona	Navarre	201,653	Plaza de la Cruz	-1.6402	42.8113
Santander	Cantabria	172,539	Tetuán	-3.7904	43.4685
Santiago de Compostela	A Coruña	97,260	San Caetano	-8.5311	42.8878
Sevilla	Seville	688,592	Torneo	-6.0028	37.3953
Valencia	Valencia	794,288	Avenida Francia	-0.3428	39.4575
Vigo	Pontevedra	295,364	Lope de Vega	-8.7111	42.2224
Zaragoza	Zaragoza	674,997	Avenida Soria	-0.8992	41.6559

(number of hours over irradiance threshold of 120 W/m^2), 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` (Grolemund 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 , SO_2 , PM_{10} , O_3 , NO_2) 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).

$$\begin{aligned} \text{Pollutant}_{it} = & \alpha + \beta_1 \text{Temperature}_{it} + \beta_2 \text{Rain}_{it} + \beta_3 \text{Wind}_{it} \\ & + \beta_4 \text{Sunlight}_{it} + \beta_5 \text{Max_pressure}_{it} + \gamma \text{Weekend}_t \\ & + \rho_i \text{City}_i + (\delta_1 + \delta_{1i}) \text{Minor_lockdown}_t \\ & + (\delta_2 + \delta_{2i}) \text{Major_lockdown}_t \end{aligned} \quad (1)$$

where, α is the global intercept of the model, β_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} , γ measures the effect of weekend days on the value of Pollutant_{it} , ρ_i represents the overall city-specific effect (normal pollutant levels in the absence of lockdown), δ_1 and δ_2 measure the overall effect of the minor and major lockdown, respectively, on the values of Pollutant_{it} , and δ_{1i} and δ_{2i} measure the city-specific effect of the minor and major lockdown, respectively, on the value of Pollutant_{it} . The variables Weekend_t , Minor_lockdown_t , and Major_lockdown_t 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_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_3 . Precipitation values show no association with any of the pollutants, and wind speed has a negative association with all pollutant concentrations but O_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_3 should be analyzed separately. While the UV radiation produced in higher sunlight hours showed positive correlations with O_3 in different studies (Mao et al., 2020), which is consistent with our results, higher temperatures are usually associated with higher O_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_3 , among others.

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

The capability of the meteorological variables to explain data variability is 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_2 , and PM_{10} reductions, while SO_2 was only reduced during the major lockdown and O_3 increased during lockdown. However, a full analysis of

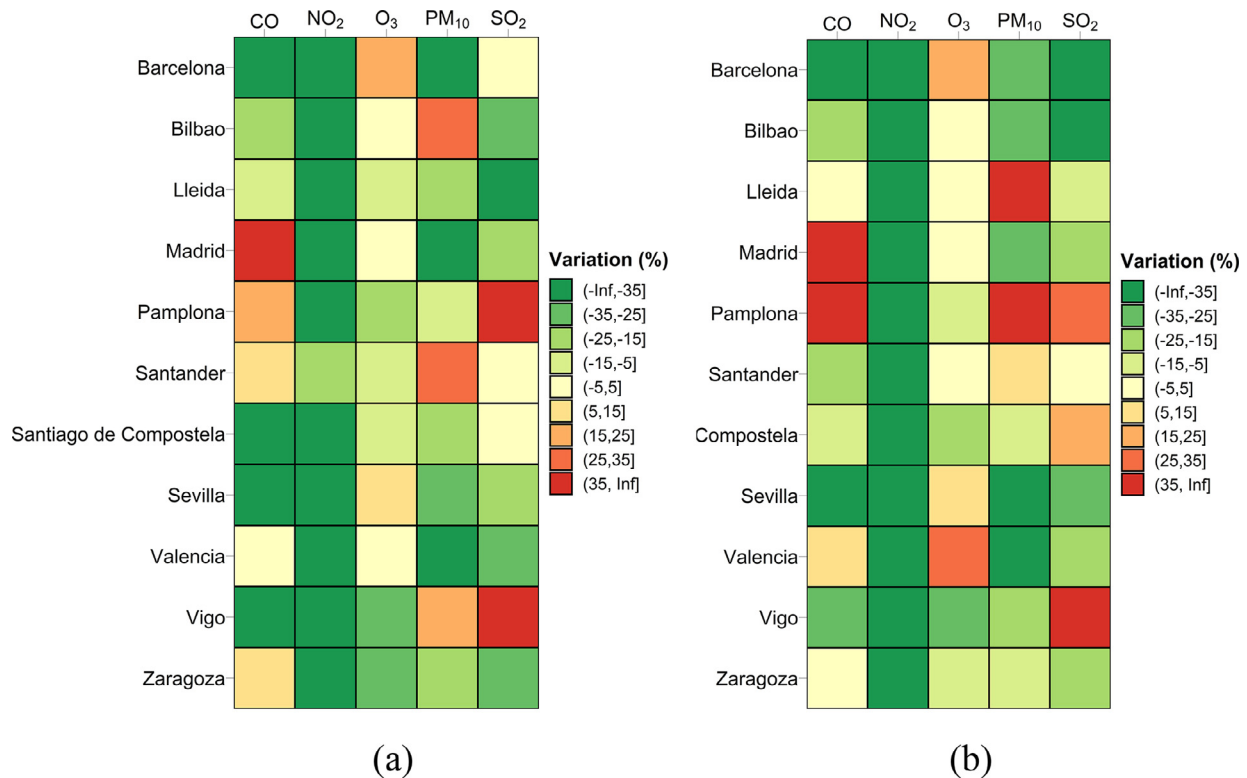


Fig. 2 – Percentage variations in pollutant levels in each of the cities under study between March 17th-March 31st 2019 and March 15th-March 29th 2020 (a), and between April 1st-April 14th 2019 and March 30th-April 12th 2020 (b).

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

	Temperature	Precipitation	Wind speed	Sunlight hours	Min pressure	Max pressure
Temperature	1.000	-0.211	-0.031	0.264	0.414	0.405
Precipitation	-0.211	1.000	0.157	-0.411	-0.136	-0.096
Wind speed	-0.031	0.157	1.000	-0.062	0.056	0.103
Sunlight hours	0.264	-0.411	-0.062	1.000	0.103	0.081
Min pressure	0.414	-0.136	0.056	0.103	1.000	0.990
Max pressure	0.405	-0.096	0.103	0.081	0.990	1.000

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.

	CO		NO ₂		O ₃		PM ₁₀		SO ₂	
	Estimate	p -value	Estimate	p -value	Estimate	p -value	Estimate	p -value	Estimate	p -value
Intercept(ω)	-0.0676	0.8900	-125.3590	0.0034	509.1870	0.0000	-233.0240	0.0000	7.8763	0.2780
Temperature (β_1)	0.0021	0.1151	0.0948	0.4143	-1.0103	0.0000	0.7410	0.0000	0.0695	0.0004
Precipitation (β_2)	-0.0001	0.7932	0.0301	0.5337	-0.0500	0.5208	-0.0507	0.2636	0.0068	0.4077
Wind speed (β_3)	-0.0066	0.0000	-2.1671	0.0000	2.5087	0.0000	-0.2017	0.1275	-0.1178	0.0000
Sunlight hours (β_4)	0.0026	0.0033	0.5182	0.0000	1.0020	0.0000	0.3530	0.0000	0.0779	0.0000
Max pressure (β_5)	0.0004	0.3730	0.1769	0.0000	-0.4596	0.0000	0.2448	0.0000	-0.0069	0.3292
Weekend day (γ)	-0.0213	0.0009	-5.2731	0.0000	5.2665	0.0000	-1.6488	0.0016	0.0463	0.6227
Minor lockdown day (δ_1)	-0.1610	0.0000	-25.7832	0.0000	16.5292	0.0000	-6.2502	0.0022	0.3310	0.3680
Major lockdown day (δ_2)	-0.1629	0.0000	-24.4542	0.0000	17.2843	0.0000	-9.9300	0.0000	-0.2761	0.4631

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).

Model	CO	NO ₂	O ₃	PM ₁₀	SO ₂
Meteorological	0.0713	0.0958	0.1179	0.1321	0.2130
Meteorological + Weekend	0.0799	0.1320	0.1460	0.1382	0.2131
Meteorological + Weekend + City	0.6066	0.5253	0.3787	0.4028	0.7285
Meteorological + Weekend + City * Lockdown	0.7331	0.7969	0.4871	0.5204	0.7762

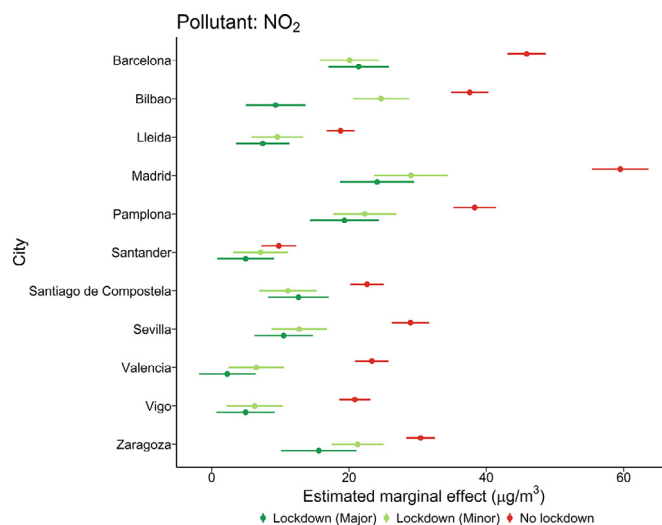


Fig. 3 – City-specific marginal effects estimated by Eq. (1) for NO₂ levels in each of the three study periods. Differences between marginal effects are statistically significant when the associated confidence intervals do not overlap.

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.

3.5.1. City-specific marginal effects for NO₂ levels

Fig. 3 shows that the estimated reductions of NO₂ 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.

However, due to being comparatively small, Santander did not show significant changes after the COVID-19 lockdown, with lower NO₂ pollution (see no lockdown level in Fig. 3). However, the differences between the reduced NO₂ 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. NO₂ is mainly produced from oxidation of NO by O₃ and peroxy radicals, while NO is mainly from direct emissions from various combustion processes, including vehicles, industrial boilers, power plants and ships (Tobías et al., 2020). Therefore, the significant reduction of NO₂ accompanied with rising O₃ concentration (see Fig. 4) during the lockdown period can be mainly attributed to the reduction of NO emission sources.

3.5.2. City-specific marginal effects for O₃ levels

The result obtained for the NO₂ levels contrasts with those found for O₃ (see Fig. 4).

An increasing trend of O₃ levels during lockdown can thus be seen, although there are only statistically significant differences for Barcelona and Valencia. Most O₃ increases are not significant or are moderate compared to NO₂ 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_x levels in urban areas in VOC-limited environments, which leads to reduced local titra-

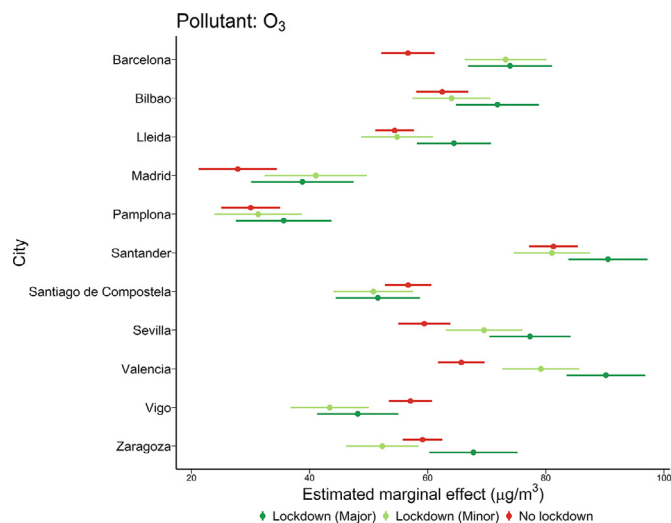


Fig. 4 – City-specific marginal effects estimated by Eq. (1) for O_3 levels in each of the three periods. Differences between marginal effects are statistically significant when the associated confidence intervals do not overlap.

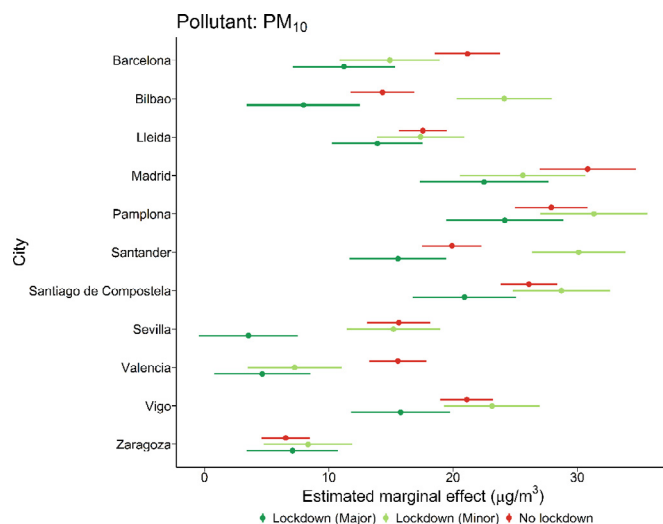


Fig. 5 – City-specific marginal effects estimated by Eq. (1) for PM_{10} levels in the three study periods. Differences between marginal effects are statistically significant when the associated confidence intervals do not overlap.

tion of NO_x and thus to a more moderate increase of O_3 levels (Souza and Ozonur, 2019; Tobías et al., 2020). It is important to mention that NO_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_{10} levels

Lockdown periods also led to a general reduction in particulate matter (PM_{10}) levels (Fig. 5).

Significant reductions of PM_{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_{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 (Celis et al., 2004).

3.5.4. City-specific marginal effects for SO_2 levels

The lockdown periods also led to a reduction in SO_2 levels in some of the cities (Fig. 6).

Significant reductions of SO_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_2 levels or in-

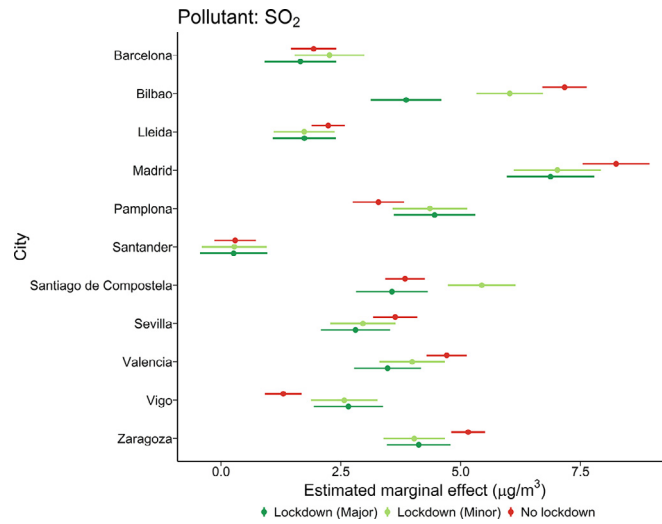


Fig. 6 – City-specific marginal effects estimated by Eq. (1) for SO₂ levels in each of the three periods. Differences between marginal effects are statistically significant when the associated confidence intervals do not overlap.

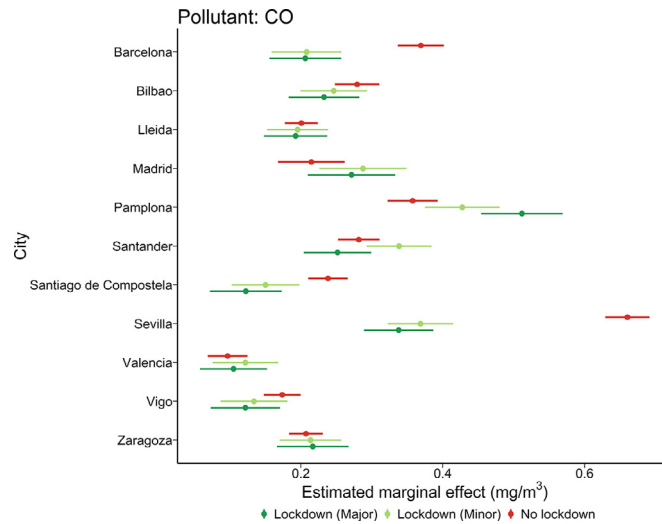


Fig. 7 – City-specific marginal effects estimated by Eq. (1) for CO levels in each of the three periods. Differences between marginal effects are statistically significant when the associated confidence intervals do not overlap.

significant variations, e.g. in Vigo and Pamplona. As in the case of NO₂ and PM₁₀, 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 O₃ like NO and NO₂ (Yousefian et al., 2020) in Barcelona and Sevilla are in agreement with the higher O₃ 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 NO₂ was achieved in most cities, mainly due to the significant reduction in transport and industry. Secondly, SO₂, CO and PM₁₀ were only reduced in some of the cities, which is beneficial because these chemical compounds can be transformed into H₂SO₄ and HNO₃ 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 NO₂ 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.

Acknowledgements

Financial support was received from the Universitat de València for the VLC-CAMPUS “Atracció de Talent” PhD fellowship (awarded to C.B-S) and from the Fundación Universidad Católica de Valencia San Vicente Mártir through Grant 2019–231–003UCV (awarded to Á.S-A).

REFERENCES

- Arroyo, V., Díaz, J., Salvador, P., Linares, C., 2019. Impact of air pollution on low birth weight in Spain: an approach to a National Level Study. *Environ. Res.* 171, 69–79. doi:10.1016/j.envres.2019.01.030.
- Ayuntamiento de Madrid, 2020. Portal De Datos Abiertos Del Ayuntamiento de Madrid. Bicimad [WWW Document] URL <https://datos.madrid.es/portal/site/egob/>.
- Ayuntamiento de Zaragoza, 2020. La Ciudad. Medio Ambiente [WWW Document]. URL <https://www.zaragoza.es/sede/portal/medioambiente/>
- Becker, S., Soukup, J.M., 1999. Exposure to urban air particulates alters the macrophage-mediated inflammatory response to respiratory viral infection. *J. Toxicol. Environ. Heal. - Part A* 57, 445–457. doi:10.1080/009841099157539.
- Bourouiba, L., 2020. Potential implications for reducing transmission of COVID-19. *JAMA Insights* doi:10.1001/jama.2020.4756.
- Briz-Redón, Á., Serrano-Aroca, Á., 2020. A spatio-temporal analysis for exploring the effect of temperature on COVID-19 early evolution in Spain. *Sci. Total Environ.* 728, 138811. doi:10.1016/j.scitotenv.2020.138811.
- Calidad del aire - navarra.es [WWW Document], 2020. URL https://www.navarra.es/home_es/Temas/Medio+Ambiente/Calidad+del+aire/
- Catalunya, G.de, n.d. Dades obertes de Catalunya [WWW Document]. URL http://governobert.gencat.cat/ca/dades_obertes/
- Celis, J.E., Morales, J.R., Zaror, C.A., Inzunza, J.C., 2004. A study of the particulate matter PM10 composition in the atmosphere of Chillán. *Chemosphere* 54, 541–550. doi:10.1016/S0045-6535(03)00711-2.
- Chen, R., Kan, H., Chen, B., Huang, W., Ba, Z., Song, G., et al., 2012. Association of particulate air pollution with daily mortality | *American Journal of Epidemiology* | Oxford Academic. *Am. J. Epidemiol.* 175, 1173–1181. doi:10.1093/aje/kwr425.
- Chiu, A.S.F., Aviso, K.B., Tan, R.R., 2020. On general principles at the sustainability science-policy interface. *Resour. Conserv. Recycl.* 158, 104828. doi:10.1016/j.resconrec.2020.104828.
- CMAOT, 2020. Consejería de Medio Ambiente y Ordenación del Territorio, Junta de Andalucía [WWW Document]. Localización la Red Automática Calid. del Aire. URL <http://www.juntadeandalucia.es/medioambiente/site/portalweb/>
- Czerwinska, J., Wielgosinski, G., 2020. The effect of selected meteorological factors on the process of “Polish smog” formation. *J. Ecol. Eng.* 21, 180–187. doi:10.12911/22998993/112764.
- Dadashi, M., Pages, D., Hernandez, I., 2020. Chemometrics modelling of temporal changes of ozone half hourly concentrations in different monitoring stations. *Chemom. Intell. Lab. Syst.* 201, 104015. doi:10.1016/j.chemolab.2020.104015.
- Dong, Y., Li, J., Guo, J., Jiang, Z., Chu, Y., Chang, L., et al., 2020. The impact of synoptic patterns on summertime ozone pollution in the North China Plain. *Sci. Total Environ.* 735, 139559. doi:10.1016/j.scitotenv.2020.139559.
- European Environment Agency, 2014. *Costs of Air Pollution from European Industrial Facilities 2008-2012*. European Environment Agency (EEA).
- Euskadi.eus, 2020. Euskadi.eus - Toda la información, trámites y servicios del Gobierno Vasco [WWW Document]. URL <https://www.euskadi.eus/inicio/>
- Fenech, S., Aquilina, N.J., 2020. Science of the total environment trends in ambient ozone, nitrogen dioxide, and particulate matter concentrations over the Maltese Islands and the corresponding health impacts. *Sci. Total Environ.* 700, 134527. doi:10.1016/j.scitotenv.2019.134527.
- Fox, J., Weisberg, S., 2018. *An R Companion to Applied Regression*. SAGE Publications.
- Gerhardsson, L., Skerfving, S., Oskarsson, A., 1997. Effects of acid precipitation on the environment and on human health. In: *Advances in Environmental Control Technology: Health and Toxicology*. Gulf Professional Publishing, pp. 355–364. doi:10.1016/b978-088415386-3/50017-5.
- Gobierno de Cantabria. Consejería de Medio Ambiente. Ordenación del Territorio y Urbanismo, 2020. Red de Control y Vigilancia de la Calidad del Aire de Cantabria.
- Grolemund, G., Wickham, H., 2011. Dates and times made easy with lubridate. *J. Stat. Softw.* 40, 1–25.
- GVA, 2020. Calidad del aire - Generalitat Valenciana [WWW Document]. URL <http://www.agroambient.gva.es/es/web/calidad-ambiental/calidad-del-aire>
- Hoque, M.M., Ashraf, Z., Kabir, H., Sarker, E., 2020. Meteorological influences on seasonal variations of air pollutants (SO₂, NO₂, O₃, CO, PM_{2.5} and PM₁₀) in the Dhaka megacity. *Am. J. Pure Appl. Biosci.* 2, 15–23. doi:10.34104/ajpab.020.15023.
- Horne, B.D., Joy, E.A., Hofmann, M.G., Gesteland, P.H., Cannon, J.B., Lefler, J.S., et al., 2018. Short-term elevation of fine particulate matter air pollution and acute lower respiratory infection.

- Am. J. Respir. Crit. Care Med. 198, 759–766. doi:10.1164/rccm.201709-1883OC.
- INE, 2019. Instituto Nacional de Estadística. (National Statistics Institute) [WWW Document]. URL <https://www.ine.es/dynt3/inebase/es/index.htm?padre=517&capsel=522>
- ISCIII, n.d. Situación de COVID-19 o Coronavirus en España. [WWW Document].
- Jiang, S., Du, L., Shi, Z., 2020. An emerging coronavirus causing pneumonia outbreak in Wuhan, China: calling for developing therapeutic and prophylactic strategies. *Emerg. Microbes Infect.* doi:10.1080/22221751.2020.1723441.
- Lai, C.C., Shih, T.P., Ko, W.C., Tang, H.J., Hsueh, P.R., 2020. Severe acute respiratory syndrome coronavirus 2 (SARS-CoV-2) and coronavirus disease-2019 (COVID-19): the epidemic and the challenges. *Int. J. Antimicrob. Agents* doi:10.1016/j.ijantimicag.2020.105924.
- Lang, D.T., 2020. XML: tools for parsing and generating XML within R and S-Plus.
- Lang, D.T.:the CRAN team, 2015. RCurl: general network (HTTP/FTP/...) client interface for R. R package version 1.95-4.7.
- Li, X., Ma, Y., Wang, Y., Liu, N., Hong, Y., 2017. Temporal and spatial analyses of particulate matter (PM10 and PM2.5) and its relationship with meteorological parameters over an urban city in northeast China. *Atmos. Res.* 198, 185–193. doi:10.1016/j.atmosres.2017.08.023.
- Liu, B., Sun, X., Zhang, J., Bi, X., Li, Y., Li, L., et al., 2020. Characterization and spatial source apportionments of ambient PM10 and PM2.5 during the heating period in Tianjin, China. *Aerosol. Air Qual. Res.* 20, 1–13. doi:10.4209/aaqr.2019.06.0281.
- Lu, R., Zhao, X., Li, J., Niu, P., Yang, B., Wu, H., et al., 2020. Genomic characterisation and epidemiology of 2019 novel coronavirus: implications for virus origins and receptor binding. *Lancet* 395, 565–574. doi:10.1016/S0140-6736(20)30251-8.
- Ludecke, D., 2016. sjPlot: data visualization for statistics in social science. R package version 2.0.1. <https://CRAN.R-project.org/package=sjPlot>.
- Manisalidis, I., Stavropoulou, E., Stavropoulos, A., Bezirtzoglou, E., 2020. Environmental and health impacts of air pollution: a review. *Front. Public Heal.* doi:10.3389/fpubh.2020.00014.
- Mao, J., Wang, L., Lu, C., Liu, J., Li, M., Tang, G., et al., 2020. Meteorological mechanism for a large-scale persistent severe ozone pollution event over eastern China in 2017. *J. Environ. Sci.* 92, 187–199. doi:10.1016/j.jes.2020.02.019.
- Ministerio de Medio Ambiente y Medio Rural y Marino. AEMET, 2011. Atlas Climático Ibérico Iberian Climate Atlas Gobierno De España.
- Mitjà, O., Arenas, À., Rodó, X., Tobias, A., Brew, J., Benlloch, J.M., 2020. Experts' request to the Spanish Government: move Spain towards complete lockdown. *Lancet* 395, 1193. doi:10.1016/s0140-6736(20)30753-4.
- Morawska, L., Cao, J., 2020. Airborne transmission of SARS-CoV-2: the world should face the reality. *Environ. Int.* doi:10.1016/j.envint.2020.105730.
- Nuvolone, D., Petri, D., Voller, F., 2018. The effects of ozone on human health. *Environ. Sci. Pollut. Res.* 25, 8074–8088. doi:10.1007/s11356-017-9239-3.
- Radzka, E., 2020. The effect of meteorological conditions on air pollution in Siedlce. *J. Ecol. Eng.* 21, 97–104. doi:10.12911/22998993/113074.
- Ren, L.-L., Wang, Y.-M., Wu, Z.-Q., Xiang, Z.-C., Guo, L., Xu, T., et al., 2020. Identification of a novel coronavirus causing severe pneumonia in human. *Chin. Med. J. (Engl.)* 1. doi:10.1097/cm9.0000000000000722.
- Sarkis, J., Cohen, M.J., Dewick, P., Schröder, P., 2020. A brave new world: lessons from the COVID-19 pandemic for transitioning to sustainable supply and production. *Resour. Conserv. Recycl.* 104894. doi:10.1016/j.resconrec.2020.104894.
- Shi, C., Yuan, R., Wu, B., Meng, Y., Zhang, H., Zhang, H., et al., 2018. Meteorological conditions conducive to PM2.5 pollution in winter 2016/2017 in the Western Yangtze River Delta. *China. Sci. Total Environ.* 642, 1221–1232. doi:10.1016/j.scitotenv.2018.06.137.
- Souza, A.De, Ozonur, D., 2019. Statistical behavior of O₃, OX, NO, NO₂, and NOx in urban environment. *Ozone Sci. Eng.* 42. doi:10.1080/01919512.2019.1602468.
- Team, R.C., 2020. R: a language and environment for statistical computing.
- Tobías, A., 2020. Evaluation of the lockdowns for the SARS-CoV-2 epidemic in Italy and Spain after one month follow up. *Sci. Total Environ.*, 138539 doi:10.1016/j.scitotenv.2020.138539.
- Tobías, A., Carnerero, C., Reche, C., Massagué, J., Via, M., Minguilón, M.C., et al., 2020. Changes in air quality during the lockdown in Barcelona (Spain) one month into the SARS-CoV-2 epidemic. *Sci. Total Environ.* 726, 138540. doi:10.1016/j.scitotenv.2020.138540.
- Trasande, L., Malecha, P., Attina, T.M., 2016. Particulate matter exposure and preterm birth: estimates of U.S. attributable burden and economic costs. *Environ. Health Perspect* 124, 1913–1918. doi:10.1289/ehp.1510810.
- UK Department for Environment Food and Rural Affairs, n.d. Site Environment Types - Defra, UK.
- Uttamang, P., Campbell, P.C., Aneja, V.P., Hanna, A.F., 2020. A multi-scale model analysis of ozone formation in the Bangkok Metropolitan Region. *Atmos. Environ.* 229, 117433. doi:10.1016/j.atmosenv.2020.117433.
- van Doremalen, N., Bushmaker, T., Morris, D.H., Holbrook, M.G., Gamble, A., Williamson, B.N., et al., 2020. Aerosol and surface stability of SARS-CoV-2 as compared with SARS-CoV-1. *N. Engl. J. Med.* doi:10.1056/NEJMc2004973, NEJMc2004973.
- Venter, Z.S., Aunan, K., Chowdhury, S., Lelieveld, J., 2020. COVID-19 lockdowns cause global air pollution declines with implications for public health risk Authors Affiliations : oRCIDs : key words : introduction : medRxiv. 10.1101/2020.04.10.20060673
- Verity, R., Okell, L.C., Dorigatti, I., Winskill, P., Whittaker, C., Imai, N., et al., 2020. Estimates of the severity of coronavirus disease 2019: a model-based analysis. *Lancet Infect. Dis.* doi:10.1016/S1473-3099(20)30243-7.
- Wang, P., Guo, H., Hu, J., Kota, S.H., Ying, Q., Zhang, H., 2019. Responses of PM 2.5 and O 3 concentrations to changes of meteorology and emissions in China. *Sci. Total Environ.* 662, 297–306. doi:10.1016/j.scitotenv.2019.01.227.
- Wang, P., Chen, K., Zhu, S., Wang, P., Zhang, H., 2020. Severe air pollution events not avoided by reduced anthropogenic activities during COVID-19 outbreak. *Resour. Conserv. Recycl.* 158, 104814. doi:10.1016/j.resconrec.2020.104814.
- Wickham, H., 2016. *ggplot2: Elegant Graphics For Data Analysis*. Springer-Verlag, New York.
- Wu, B., Li, T., Baležentis, T., Štreimikienė, D., 2019. Impacts of income growth on air pollution-related health risk: exploiting objective and subjective measures. *Resour. Conserv. Recycl.* 146, 98–105. doi:10.1016/j.resconrec.2019.03.037.
- Xiao, Y., Murray, J., Lenzen, M., 2018. International trade linked with disease burden from airborne particulate pollution. *Resour. Conserv. Recycl.* 129, 1–11. doi:10.1016/j.resconrec.2017.10.002.
- Xie, J., Teng, J., Fan, Y., Xie, R., Shen, A., 2019. The short-term effects of air pollutants on hospitalizations for respiratory disease in Hefei. *China. Int. J. Biometeorol.* 63, 315–326. doi:10.1007/s00484-018-01665-y.
- Xu, Q., Li, X., Wang, S., Wang, C., Huang, F., Gao, Q., et al., 2016. Fine particulate air pollution and hospital emergency room visits for respiratory disease in urban areas in Beijing, China, in 2013. *PLoS ONE* 11, e0153099. doi:10.1371/journal.pone.0153099.

- Xu, Y., Yang, L., Wang, X., Zheng, M., Li, C., Zhang, A., 2020. Risk evaluation of environmentally persistent free radicals in airborne particulate matter and influence of atmospheric factors. *Ecotoxicol. Environ. Saf.* 196, 110571. doi:10.1016/j.ecoenv.2020.110571.
- Xunta de Galicia, 2020. Calidade do Aire de Galicia [WWW Document]. URL <https://www.meteogalicia.gal/Caire/index.action>
- Yousefian, F., Faridi, S., Azimi, F., Aghaei, M., Shamsipour, M., 2020. Temporal variations of ambient air pollutants and meteorological influences on their concentrations in Tehran during 2012 – 2017. *Sci. Rep.* 10, 292. doi:10.1038/s41598-019-56578-6.
- Yu, D.E.C., Razon, L.F., Tan, R.R., 2020. Can global pharmaceutical supply chains scale up sustainably for the COVID-19 crisis? *Resour. Conserv. Recycl.* 104868. doi:10.1016/j.resconrec.2020.104868.
- Zhu, Y., Xie, J., Huang, F., Cao, L., 2020. Association between short-term exposure to air pollution and COVID-19 infection: evidence from China. *Sci. Total Environ.* 727, 138704. doi:10.1016/j.scitotenv.2020.138704.