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Impact of COVID-19 restrictions on hourly levels of PM10, PM2.5 and black carbon at an industrial suburban site in northern Spain

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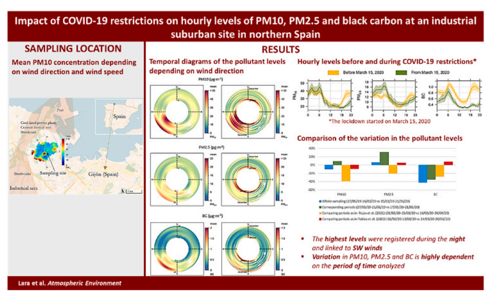
HIGHLIGHTS

- Hourly levels of pollutants were jointly analysed with wind direction and speed.
- The highest levels of PM and black carbon were found during night hours.
- SW winds associated with high levels, pointing to industries in that direction.
- The lockdown period considered affects the conclusions on the level variations.
- PM10 may vary by −39% or 8.7%; PM2.5 by −21% or 31% and BC by −42% or 7.4%.

GRAPHICAL ABSTRACT



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ABSTRACT

Due to the COVID-19 pandemic, lockdown restrictions were established around the world. Many studies have assessed whether these restrictions affected atmospheric pollution. Comparison between them is difficult as the periods of time considered are generally not the same and thus, different conclusions may be reached. Besides, most of them consider mean daily pollutant concentration, despite differences being observed according to the time of day. In this study, the hourly levels of PM10, PM2.5 and black carbon (BC) in an industrial suburban area in the north of Spain were analysed from May 2019 to June 2020 and compared with those from the literature, using the same period in each case. In general, the highest concentrations were reached when the wind direction came from the southwest (where a steelworks, a coal-fired power plant and other industries are located) and during the night-time, both before and during the lockdown. The highest concentrations of PM10, PM2.5 and BC were observed from December to February (on average: 45, 17 and 1.3 μg m^{-3}, respectively). The decrease/increase in those pollutants levels during the lockdown were found to be highly dependent on the period considered. Indeed, PM10 can be found to decrease by up to 39% or increase by 12%; PM2.5 can decrease by 21% or increase by up to 36%; and BC, although it generally decreases (by up to 42%), can increase by 7.4%.

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1. Introduction

Particulate matter is considered one of the most problematic pollutants for human health. PM10 and PM2.5 fractions include inhalable particles that are small enough to penetrate the thoracic region of the respiratory system. PM2.5 can penetrate the lung barrier and enter the circulatory system (WHO, 2021) and hence, spread to the whole body (Feng et al., 2016; Megido et al., 2016). The smaller the particles are, the greater impact they may produce. Li et al. (2018) found that when the PM2.5 concentration increased, so did susceptibility to respiratory diseases, including acute respiratory distress, asthma, chronic obstructive pulmonary disease, and lung cancer.

Based on numerous epidemiological studies and clinical observations, PM2.5 has been considered as the main cause of the adverse cardiovascular effects of air pollution on human health (Du et al., 2016). Moreover, Hayes et al. (2020) reported that after a long-term exposure to fine particulate matter, air pollution is associated with ischaemic heart disease and stroke mortality, with excess risk occurring in the range of and below the present US annual average standard (12 μg m^{-3}) for ambient exposure to PM2.5.

Data on cancer hospitalizations and annual PM2.5 concentrations were collected from 1814 Brazilian cities during 2002–2015 by Yu et al. (2021). They estimated that 34% of total cancer hospitalizations could be attributed to PM2.5 exposure in Brazil during the study time. Long-term exposure to ambient PM2.5 was positively associated with hospitalization for many cancer types in Brazil (Yu et al., 2021).

The World Health Organization (WHO) updated its Global Air Quality Guidelines in September 2021. Table S1 shows the comparison between the values recommended by the WHO (WHO, 2021) and those set as maximums in Spanish legislation. There are several differences between them, for example, the WHO recommendation for annual mean concentrations of PM2.5 is not exceeding 5 μg m^{-3}, whereas in Spain the maximum annual mean allowed is 25 μg m^{-3}.

One of the major contributors to PM2.5 in the air is black carbon (BC), which is small enough to be easily inhaled into the lungs and has been associated with adverse health effects. BC not only has a negative impact on local and regional air quality, but also warms the regional and global climate (von Schneidemesser et al., 2017). Indeed, it is the second-largest contributor to Arctic warming (Shapovalova, 2016).

First cases of COVID-19 were reported in the Wuhan region of Hubei (China) at the end of 2019. Consequently, WHO declared the COVID-19 pandemic on 11/03/2020 (WHO, 2020). From 14/03/2020 to 21/06/2020, the Spanish Government declared a state of alarm, which imposed severe restrictions on industrial activities, traffic, and urban mobility (Querol et al., 2021). Based on local circumstances and the evolution of the pandemic, the restrictions were gradually reduced by phases. In Gijón, phases 0 and 1, where most restrictions continued in force, were established on 04/05/2020 and 11/05/2020, respectively; meanwhile, phases 2 and 3 were established on 25/05/2020 and 08/06/2020, with partial reopening of activities with capacity limitations and reduction of mobility restrictions. Phase 3 ended on 21/06/2020, coinciding with the end of the alarm state and the beginning of new normality (GPA, 2020).

The number of movements decreased substantially in all Spanish autonomous communities, ranging from 40 to 50% compared to the pre-pandemic baseline, being those reduction more than 75% for trips related to workplace or public transport journeys (Radics and Christidis, 2022). Therefore, those reductions were also associated to a large reduction in the industrial activity. The emissions into the atmosphere registered by industrial complexes in the region of Asturias also reflect these variations. PM10 emissions were reduced from 917 t in 2019 to 634 t in 2020 (PRTR, 2023). After the relaxation of most of the restrictions, the movements were increasing to 70–80% compared to the pre-pandemic values in the summer of 2020 (Radics and Christidis, 2022). Some studies have compared situations with and without the lockdown restrictions due to the COVID pandemic by analysing different periods of time. Some of them have compared the levels observed during the lockdown with those in the previous period (Tobías et al., 2020; Rojas et al., 2021), whereas others made comparisons with the same period as the lockdown in previous years (Briz-Redón et al., 2021; Sokhi et al., 2021; Toro et al., 2021; Shakoor et al., 2020; Jephcote et al., 2021). However, most of these studies did not take into account the effect that the meteorology of the area could have had on the increase/decrease observed in the concentrations of PM10, PM2.5 and BC during the periods analysed, an aspect of great relevance.

The objective of this study was to analyse the variations of the hourly levels of PM10, PM2.5 and BC over time before and during restrictions due to the COVID-19 pandemic. To do so, hourly, daily (distinguishing between weekday and weekend) and monthly average concentrations were assessed. Furthermore, to compare them with the different studies found in the literature, the same periods of time were used in each case, finding that the decrease/increase in the levels of these three pollutants were highly dependent on the period of time considered.

The site of study was the western area of the city of Gijón (northern Spain), which is affected by nearby urban and industrial activities (a steelworks, coking plant, coal-fired power station, heavy traffic road …) and is highly contaminated by settleable particulate matter (Lara et al., 2021; Negral et al., 2021).

Most of the studies in the literature use mean daily pollutant concentrations, despite differences being observed in these levels depending on the hour of the day. Besides, the variability in the levels may depend not only on the pandemic situation, but also on the meteorology of the analysed area, the activity of the pollution sources in the surroundings, etc. Thus, in the present study, a joint analysis of wind direction, wind speed and the hourly levels of the three air pollutants was conducted for both periods of time (before and during lockdown). The effect of wind was assessed to better understand its relevance and whether its influence was enough to explain the variations observed in the levels recorded. Likewise, the mobility restrictions and the reduction of industrial activities during the COVID-19 pandemic were considered.

2. Materials and methods

2.1. Studied area

The present study was carried out in an industrialized area in the western neighbourhoods of Gijon, a city on the northern coast of Spain. According to the Köppen-Geiger climate classification (Kottek et al., 2006), Gijón is classified as Cfb (west coast maritime, oceanic, temperate climate with cool summers, with abundant and well-distributed rainfall throughout the year). This region has a mean annual temperature of 14 °C and a relative humidity of 75%.

Fig. 1 shows the location of the sampling site and the meteorological station (43°32′47.9"N 5°42′14.5"W, 13 m AMSL), as well as the main industrial activities therein. The industries with the highest emissions of particulate matter during 2019–2020 (2–3 km around the sampling site) were: a steelworks, a coal-fired power plant and a cement factory. In the vicinity there is an industrial estate where other smaller industries are located that produce aluminium, refractory products, metallic structures, and wires etc., a galvanizing plant, a plant that manufactures iron castings, etc. In addition, there is a Port of $4.2 \cdot 10^6$ m^2. Seaports have been reported to be a major source of PM due to the handling of materials, which causes the resuspension of loose materials (Wirth et al., 2022). Indeed, the Port of Gijón moves a large quantity of bulk commodities (mainly coal, iron ore and cement), which are handled and stored in the open air, and thus, it can give rise to important diffuse emissions of particles due to the action of the wind. During 2019 and 2020, the Port loaded and unloaded $8.6 \cdot 10^5$ and $9.9 \cdot 10^5$ t of dry bulks, respectively (Port of Gijón, 2021).

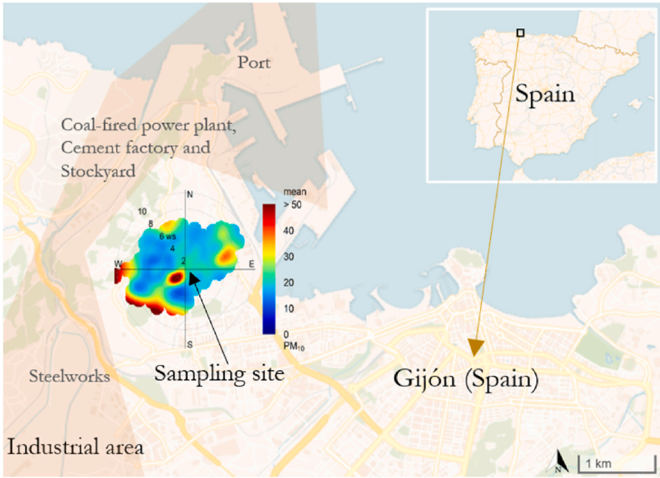

Fig. 1. Location of the sampling site and industrial activities in the area, polar plot representing mean concentration of PM10 observed (in μg m^{-3}) depending on wind direction and wind speed (indicated as concentric circles, in m s^{-1}).

2.2. Sampling of PM10, PM2.5 and BC

PM10, PM2.5 and BC were measured at the air quality mobile control station located in the western area of Gijón, from 17/05/2019 to 21/06/2020 for PM10 and PM2.5 (n = 382 days) and from 28/05/2019 to 21/06/2020 for BC (n = 375 days). Due to maintenance procedures the sampling was stopped from 07/02/2020 to 20/02/2020.

The determination of the level of the pollutant was carried out by recording hourly measurements. The equipment used for PM10 and PM2.5 was a Met One Instruments BAM 1020 continuous particulate monitor, whose operation is based on the absorption of beta radiation. In the case of BC, a Magee Scientific Aethalometer, model AE31, was used. The equipment was installed inside a van and the sampling heads and the weather monitoring station were on the roof of the van.

2.3. Statistical analysis and data treatment

The software used for data analysis was R version 4.1.2: openair package version 2.9–2 (Carslaw and Ropkins, 2012) and summary tools package version 0.6.5. From the mean hourly values, the daily mean value concentration for PM10, PM2.5 and BC were determined, considering only the days when it had been possible to carry out more than 75% of the hourly readings. Season-separated average has been also calculated, subsetted as 3-month data corresponding to: JJA (June-July-August) and SON (September-October-November), DJF (December-January-February), MAM (March-April-May). Data from 01/06/2019 to 31/05/2020 were used in this analysis to compare complete seasons.

The hourly and daily variation in pollutant levels were followed to obtain information about pollutant level patterns. Reductions/increments observed in pollutant levels during the period of the COVID-19 restrictions were calculated as the difference between the mean concentration during restrictions compared to the mean concentration previously registered, using the same periods used by other authors in the literature to determine the variability of the results. The Mann-Whitney *U* test (Fagerland and Sandvik, 2009) was used to determine if there were significant differences ($p < 0.05$) between the means of the samples collected before and during the COVID-19 restrictions.

In order to consider the temporal aspects of pollutant concentration according to wind direction, polar annulus plots were also performed using hourly data. Polar plots have been widely used to support the identification of the origin of the emission sources (Carslaw et al., 2006; Beddows et al., 2015; Grange et al., 2016; Bodor et al., 2021). These plots provide important information with respect to the sources of the particles, showing hourly and monthly variations depending on the wind. Bivariate polar plots are constructed by dividing data into wind speed/hour/month-wind direction bins and the mean concentration for each bin is calculated. Furthermore, the Kruskal-Wallis test (Kruskal and Wallis, 1952; Mann, 1945) was also used to assess significant differences ($p < 0.05$) depending on the wind direction of each sample collected to support these results.

3. Results

3.1. Levels of PM10, PM2.5 and BC

3.1.1. PM10

The mean PM10 concentration throughout the whole sampling period was 31 μg m^{-3}, which was below 40 μg m^{-3} (the legal Spanish limit value of the annual mean per calendar year), although it exceeds the value recommended by the WHO (15 μg m^{-3} annual mean). The maximum and minimum values registered were 138 and 4.1 μg m^{-3}, respectively. Fig. S1 shows the PM10 levels recorded during the period.

The daily Spanish legal limit value for PM10 is 50 μg m^{-3} and must not be exceeded on more than 35 occasions per year. During the sampling period, it was exceeded on 38 occasions: 1 in JJA (2019), 1 in SON (2019), 26 in DJF (2019–2020), with maximum values at the end of February, and 10 in MAM (2020), with maximum values in March. In short, the daily average value was exceeded mainly in DEF and in March. In Chungcheong province, Korea, Cha et al. (2019) obtained mean PM10 values of 37 μg m^{-3} in SON 2015, 48.8 μg m^{-3} in DJF 2015/2016, and 66.4 μg m^{-3} in MA 2016. These authors indicated that these results were affected by the wind direction. They linked the high concentrations found in MA and DJF with W winds, which transported pollutants, whereas the low concentrations in SON were probably observed because of increased wind variations, which drove turbulent mixing. In the present study, the highest PM10 concentrations were also found in DJF and MAM (mean values of 45 and 31 μg m^{-3}, respectively). Season-averaged mean values for PM10, PM2.5 and BC can be consulted in Table 1.

In a previous study carried out in this area to assess the impact of restrictions on the dry atmospheric deposition fraction, Lara et al. (2022) reported a high influence of wind speed and wind direction on the atmospheric pollutant levels registered. In the present study, the highest values of PM10 were generally associated with SW winds, blowing from where the integral steel industry is located (Figs. 1 and 2). Nonetheless, high values were also found under the influence of winds from other directions. The largest contribution of SW winds to PM10 compared to the other directions is also reflected in Fig. 3, which shows that both mean and maximum concentration values occurred under winds from this direction, finding significant differences ($p < 0.05$) with respect to the levels registered under winds from different directions. The assessment of the significance of the differences in PM10, PM2.5 and BC concentrations with respect to the wind direction is provided in the Supplementary Material.

The highest concentrations occurred at night, with maximum mean values being reached between 22:00 and 08:00, this contrast between day and night being even higher in DEF (consult Figs. S2 and S3). This may be explained by thermal inversion, which mainly occurs during the night and prevents the dispersion of PM10 particles (Czarnecka and

Table 1
Season-averaged concentration levels of PM10, PM2.5 and BC.

	Mean concentration (μg m^{-3})		
Season	PM10	PM2.5	BC
JJA 2019	25 ± 24	8.3 ± 6.4	0.56 ± 0.57
SON 2019	26 ± 22	9.0 ± 5.7	0.67 ± 0.70
DJF 2019–2020	45 ± 41	17 ± 11	1.3 ± 1.1
MAM 2020	31 ± 29	13 ± 7.9	0.53 ± 0.53

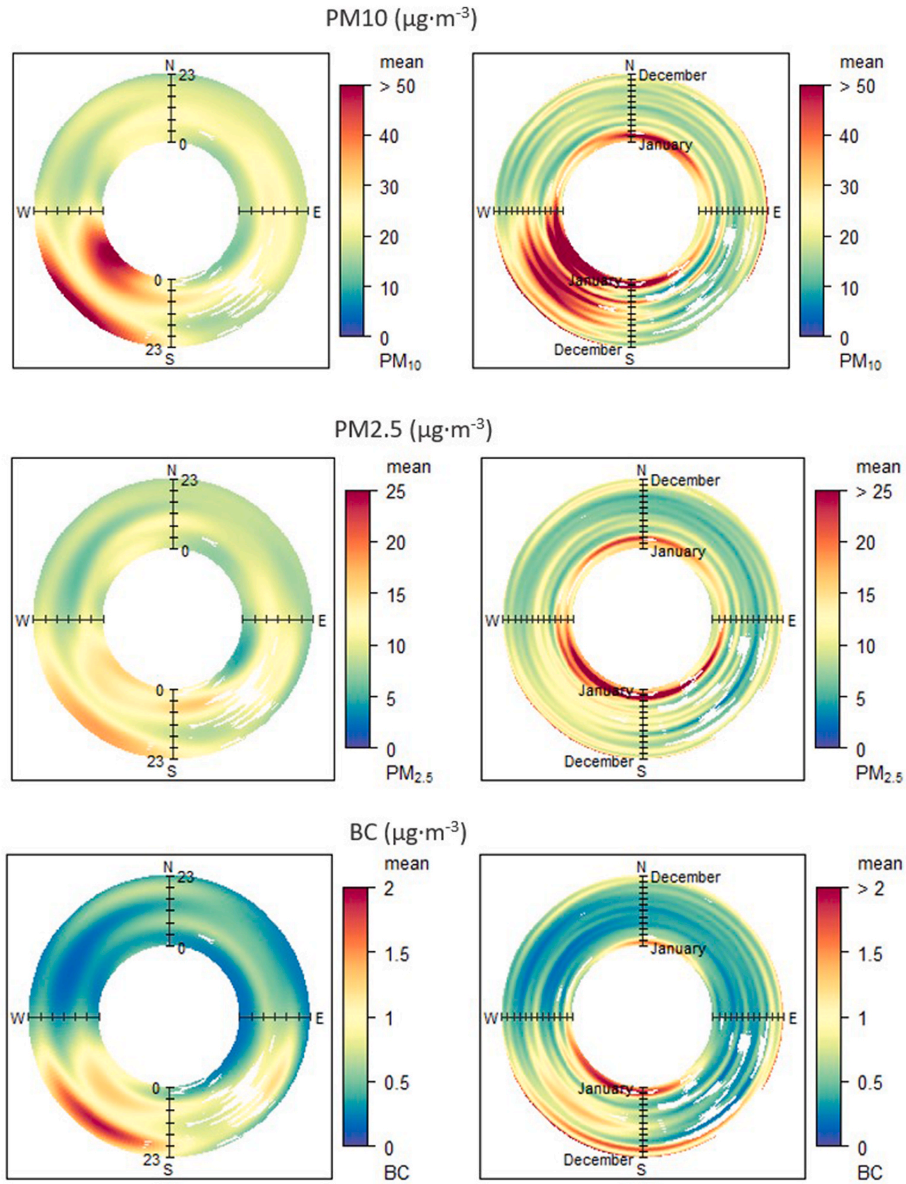

Fig. 2. Temporal diagram of the variation of PM10, PM2.5 and BC (μg m^{-3}) during the whole sampling period depending on the wind direction: hourly average (left) and monthly average (right).

Nidzgorska-Lencewicz, 2017).

In a previous work carried out in the eastern area of Gijón to assess the human health hazard of potentially toxic elements in PM10, the results showed that potential risks of As and Pb should not be overlooked (Megido et al., 2017b). Moreover, in the western area of Gijón, the results of studies related to settleable particulate matter showed that the highest levels of risk seemed to be associated with the presence of As, Pb, Sb and Fe (Lara et al., 2021; Negral et al., 2021).

3.1.2. *PM2.5*

The mean concentration of PM2.5 was 11 μg m^{-3}, lower than the Spanish legal annual mean value, which is 25 μg m^{-3}. The temporal variation of the levels of PM2.5 can be consulted in Fig. S1. The maximum level registered was 38 μg m^{-3} (coinciding with the day on which the maximum value for PM10 occurs), while the minimum value was 2.9 μg m^{-3}. In Korea, Cha et al. (2019) obtained mean PM2.5 values of 31 μg m^{-3} in SON 2015, 38 μg m^{-3} in DEF 2015/2016, and 42 μg m^{-3} in MA 2016. As in PM10 (Table 1), PM2.5 concentrations were higher in DEF (17 μg m^{-3}) and MAM (13 μg m^{-3}) than in SON (9.0 μg m^{-3}) and JJA (8.3 μg m^{-3}).

Higher concentrations of PM2.5 occurred under SW winds, as observed with PM10, although in this case there was not such a marked contrast with respect to the rest of the directions (Figs. 2 and 3). In the same way, the mean and maximum concentrations were higher in this direction, presenting a significant difference with respect to the values associated to other wind directions. The highest concentrations were observed between December and February and during the night-time (22:00–08:00).

PM10 and PM2.5 levels correlated to some extent ($R^2 = 0.67$). In the study carried out by Gong et al. (2015) at a station located in an urban area in Wuhan affected by industrial activity (steel production), the correlation between PM10 and PM2.5 was higher, R^2 of 0.89. In Chungcheong province, Korea, the studies carried out by Cha et al. (2019) obtained R^2 values of 0.45 and 0.47 in SON (2015) and DEF (2015–2016), respectively. Correlations between PM10 and PM2.5 were higher in our study, reaching values of 0.70 and 0.44 in DEF and SON, respectively.

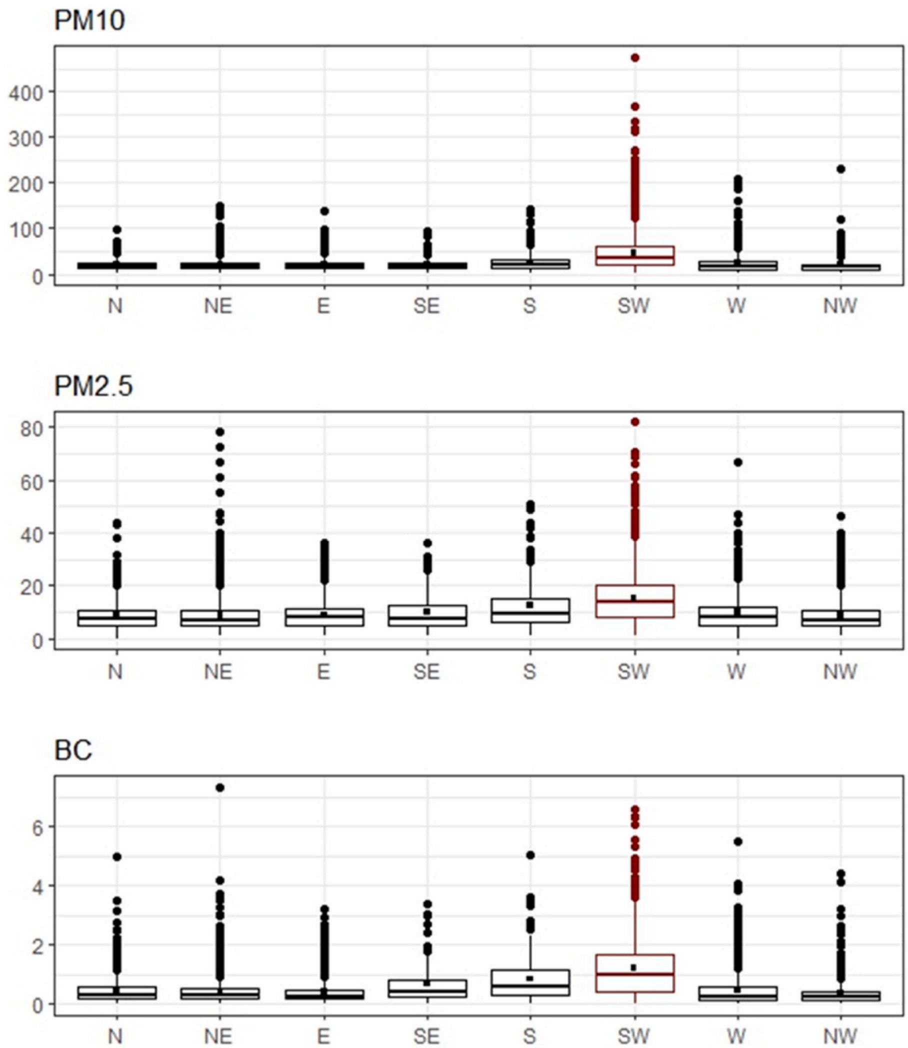

Fig. 3. Boxplot of concentrations measured (in μg m^{-3}) group by wind direction. The top, middle, and bottom lines of the box represent the 75th, median, and 25th percentiles, respectively. The square represents the arithmetic mean. Error bars outside the box represent 1.5-times the interquartile range and outliers are depicted by circles.

3.1.3. BC

BC is formed during the incomplete combustion of carbon-based fuels (Seita, 2018), is composed of solid particles, and is emitted by diesel engines, biomass burning, domestic heating, coal-fired power plants, gas flaring and maritime shipping, among other sources. According to Viidanoja et al. (2002), who studied BC in PM2.5 and PM10 in Helsinki, more than 90% of BC is part of PM2.5. In this study, BC values were relatively low (Fig. S1), the mean value being 0.70 μg m^{-3}, a value lower than that found by Viidanoja et al. (2002) in Helsinki, which was 1.2 μg C·m^{-3} in PM2.5 and 1.3 μg C·m^{-3} in PM10. It was also lower than values recorded in Jakarta by Santoso et al. (2020), with a mean BC value of 3.8 μg m^{-3}. However, Milinković et al. (2021) found at a site on the Central Adriatic coast, near the town of Šibenik (~34000 inhabitants) and its port (~2 km from the site) during February–July 2019, a mean value for BC of 0.60 ± 0.60 μg m^{-3}, slightly lower than those found in Gijón.

In line with the PM10 and PM2.5 results, the highest concentrations of BC appeared under SW winds (Fig. 2). In that direction the integrated iron and steel industry was located (Fig. 1). Although the BC concentrations associated to SW winds were significantly higher than those observed in the rest of directions (Supplementary Material), the maximum values were found linked to NE winds (Fig. 3) and greater variability in the levels was found for other wind directions.

BC concentration was much higher in DEF (1.3 μg m^{-3}) than in the

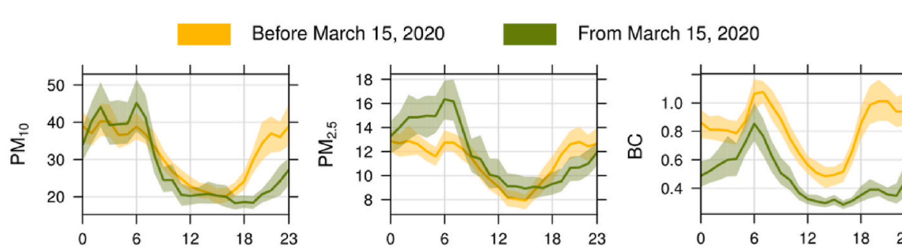

Fig. 4. Hourly averaged concentration of PM10, PM2.5 and BC (in μg m^{-3}) before and after March 15, 2020, shading showing 95% confidence intervals of the mean.

Table 2
Mean and standard deviation of daily levels (μg m^{-3}) of PM10, PM2.5 and BC in different periods of time.

Pollutant	Whole sampling	Whole sampling		Corresponding periods		Comparing the same period as in Rojas et al. (2021)		Comparing the same period as in Tobías et al. (2020)	
Date	17/05/19–21/06/20	17/05/19-14/03/20	15/03/20–21/06/20	17/05/19–21/06/19	17/05/20–21/06/20	01/02/20-15/03/20	16/03/20–30/04/20	16/02/20-13/03/20	14/03/20–30/03/20
	N = 382	N = 284	N = 98	N = 35	N = 35	N = 30	N = 46	N = 22	N = 17
PM10	31 ± 17	31 ± 18	28 ± 14	25 ± 12	28 ± 10	48 ± 31	29 ± 17	46 ± 31	42 ± 17
PM2.5	11 ± 5.8	11 ± 6.0	12 ± 5.2	8.8 ± 4.1	12 ± 4.5	18 ± 7.5	13 ± 5.9	17 ± 7.6	18 ± 5.8
Date	28/05/19–21/06/20	28/05/19-14/03/20	15/03/20-21/06/20	28/05/19–21/06/19	28/05/20–21/06/20	01/02/20-15/03/20	16/03/20–30/04/20	16/02/20-13/03/20	14/03/20–30/03/20
	N = 375	N = 276	N = 98	N = 25	N = 25	N = 30	N = 46	N = 22	N = 17
BC	0.72 ± 0.55	0.81 ± 0.58	0.47 ± 0.31	0.54 ± 0.32	0.35 ± 0.25	0.76 ± 0.47	0.55 ± 0.35	0.68 ± 0.41	0.73 ± 0.39

other periods studied, when mean values ranged from 0.53 to 0.67 μg m^{-3}. The variation of the concentration with respect to the time of day is also very clear. In contrast with the patterns observed in PM10 (Fig. S3) and PM2.5 (Fig. S4), the highest concentrations in BC appeared from 04:00 to 08:00 and from 18:00 to 00:00 and (Fig. 4 and Fig. S5).

The correlations found between BC and PM2.5 ($R^2 = 0.52$ for the whole period and 0.64, 0.53, 0.51 and 0.25 in SON, DEF, MAM and JJA, respectively) were similar to those found in Chungcheong province, Korea, by Cha et al. (2019): SON 2015 ($R^2 = 0.47$), DEF 2015–2016 ($R^2 = 0.45$), and MA 2016 ($R^2 = 0.58$). Those values indicated the importance of other pollutants in PM2.5, such as secondary pollutants (sulphates, nitrates and ammonium) or metals/metalloids, generated from primary emissions from industrial plants or as long-distance pollution. Megido et al. (2017a) analysed the influence of secondary inorganic aerosols (sulphates, nitrates and ammonium) on PM10 in the eastern area of Gijón, finding that these accounted for an average of 17% of the total. The correlation between BC and PM10 was $R^2 = 0.48$, slightly lower than that found for BC and PM2.5.

Major contribution of the pollutants studied occurred under SW, highlighting especially the PM10 levels registered. The source apportionment studies carried out in this area (Lara, 2023) and in other zones of the conurbation (Megido et al., 2017a) suggest an important industrial contribution.

3.2. *Influence of the lockdown – comparison with literature*

To analyse the influence of the lockdown due to the COVID-19 pandemic, mean values for different periods of time before and during lockdown for the three pollutants are shown in Table 2. Different methods have been used to assess the effect of COVID-19 restrictions on pollution levels. Some of them compared data obtained during restrictions with the period before restrictions while others compared it with similar periods from previous years (Ródenas et al., 2022). Air quality data from the sampling site, available only from 17/05/2019 because sampling was carried out in a mobile control station, were very different from other areas studied in Gijón. Therefore, the data from other stations would not be helpful for this analysis. Given the available data, variation in pollutant levels during the restrictions were calculated by first determining the mean values for the whole sampling period, and secondly comparing these with the corresponding period in 2019. Rigueira et al. (2022) studied PM10 variation during COVID-19 restrictions comparing with data collected from 2015 in a station located in the western area of Gijón. They concluded that reductions observed in PM10 concentration during the lockdown were nothing extraordinary.

Over the whole sampling period, PM10 and BC underwent reductions of 9.7% (non-significant reduction, $p > 0.05$) and 42% (significant reduction, $p < 0.05$), respectively, while PM2.5 experimented an increase of 9% (non-significant increase, $p > 0.05$). Fig. 5 represents the hourly variation during these periods. As stated before, the highest concentrations of PM10, PM2.5 and BC were produced during the nighttime. In the case of PM10 and PM2.5, in the morning the concentrations were slightly higher during the lockdown than before the lockdown, and just the opposite occurs in the afternoon. With respect to BC, the concentrations were higher before lockdown regardless of the time of day.

However, variations observed when comparing corresponding periods differ drastically from those found when a comparison was made between the COVID restriction period and the period immediately before it. On examining increases during the same period sampled in different years, values of 12% (non-significant increase, $p > 0.05$) and 36% (significant increase, $p > 0.05$) were found for PM10 and PM2.5, respectively, while BC underwent a reduction of 35% (significant reduction, $p < 0.05$). Although in the 2020 period studied there were still restrictions, these were no longer so limiting and therefore some mobility and industrial activity had already been resumed. Results of the daily, hourly and monthly averages comparing those periods can be consulted in Fig. S3, S4 and S5.

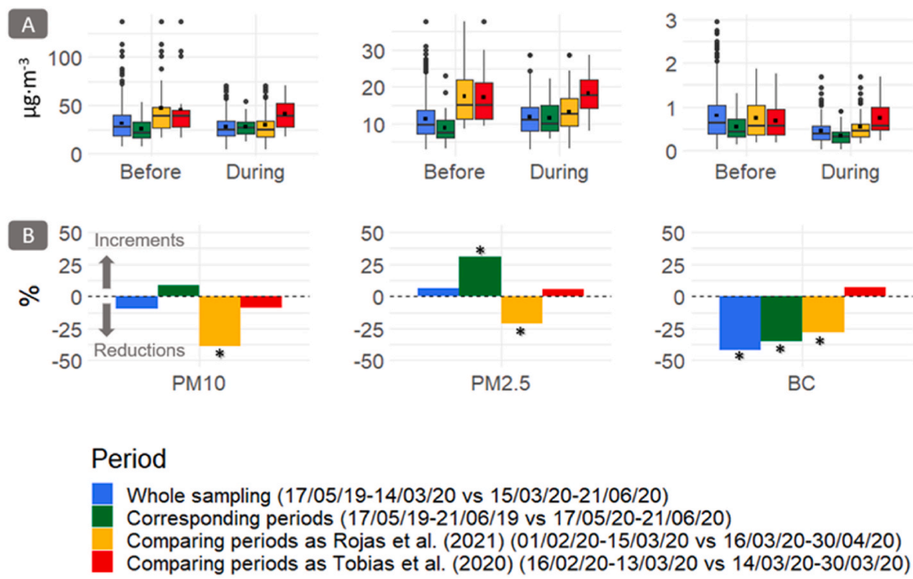

Fig. 5. Comparison of PM 10, PM2.5 and BC concentrations measured before and during COVID-19 restrictions depending on the period considered (colour code is explained in the legend). Fig. 5A: Boxplot of concentrations measured before and during COVID-19 restrictions. The top, middle, and bottom lines of the box represent the 75th, median, and 25th percentiles, respectively. The black square represents the arithmetic mean. Error bars outside the box represent 1.5-times the interquartile range and outliers are depicted by circles. Fig. 5B: Increments (positive values) and reductions (negative values) observed during COVID-19. Significant differences ($p < 0.05$) between periods are indicated by an asterisk.

In a global and regional overview of the changes in ambient concentrations of air-quality-related species, Sokhi et al. (2021) observed reductions of between 30% and 40% in mean PM2.5 concentrations during the 2020 full lockdown as compared to the same period in 2015–2019. However, they found that PM2.5 exhibited increases in some Spanish cities, which they attributed mainly to the long-range transport of African dust and/or biomass burning. Some Chinese cities showed similar increases in PM2.5 during the lockdown periods, but in this case, it was likely due to secondary PM formation (Sokhi et al., 2021).

Other authors studied this influence, comparing different time periods; for example, Toro et al. (2021) analysed the effects of the measures taken during the COVID-19 pandemic (school closures, confinement, etc.) on PM10 and PM2.5 in Santiago de Chile in MAM of 2017, 2018, 2019 (without confinement) and 2020 (during confinement), at various sampling stations. The results obtained showed that PM10 concentrations decreased by 5.2% during lockdown in 2020, while PM2.5 decreased by 11%. Although PM2.5 decreases during confinement, the decrease is much less than that obtained by Sokhi et al. (2021).

Eleven of Spain's largest cities were considered in the study of Briz-Redón et al. (2021): Barcelona, Bilbao, Lleida, Madrid, Pamplona, Santander, Santiago de Compostela, Seville, Valencia, Vigo and Zaragoza. One traffic station was selected by them for data collection and analysis in each of the cities studied. The period analysed, for each station and day, was from 17/03/2019 to 31/03/2019 and from 15/03/2019 to 29/03/2020. During the lockdown, PM10 decreased in all the stations studied except in Bilbao, Santander and Vigo. These three stations are in the north of Spain. Gijón, the city studied in this work, has similar weather conditions to those three and is on the coast, as are Santander and Vigo.

In the city of Milan, Italy, similar results to those obtained by Tobías et al. (2020) have been reported by Collivignarelli et al. (2020). Significant reductions were observed for PM10 and PM2.5 (up to 47%) and BC (up to 71%). It was probably attributable to the significant reduction in vehicular traffic caused by the lockdown, considering that means of transport represents the main source of PM10 in Milan.

In states in the USA significantly affected by COVID (California, Florida, Louisiana and North Carolina), Shakoor et al. (2020) studied the effect of lockdown on PM10 and PM2.5 levels by comparing the results obtained in the first quarter of 2019 and the first quarter of 2020. PM10 and PM2.5 decreased in 2020, with the exception of Louisiana, where PM10 increased in 2020 (a 25% increase in comparison with the previous year). The increase in the concentration of PM10 might be due to the contribution of diverse anthropogenic sources (Shakoor et al., 2020).

An increase in PM2.5 concentrations during lockdown was observed by Benchrif et al. (2021) in Addis Ababa and Jakarta. Patel et al. (2020) analysed the influence of the lockdown on PM10 and PM2.5 in three fixed-site air quality monitoring sites within the Auckland urban area (New Zealand), which is largely unaffected by long-range pollution transport or industrial sources of air pollution. The time periods studied were from 27/03/2020 to 17/04/2020 (lockdown) and comparable periods in the historical air pollution record. The reductions observed in PM2.5 and PM10 were 8–17% for PM2.5 and 7–20% for PM10.

Using the WRF-CHIMERE modelling system, Menut et al. (2020) determined the impact of the measures to contain the Covid-19 pandemic on air quality during the month of March 2020 in the west of Europe, for 799 stations for PM10 and 399 stations for PM2.5. Reductions in PM2.5 concentrations ranged between 5 and 10%. The sources of primary particle emissions from residential heating and emissions from the agricultural sector contributed to the formation of secondary fine particles in the atmosphere.

At the same time, other periods of time were considered to compare the results of the present study with other studies from the literature. In the seven sampling stations located in the metropolitan area of Lima (Peru), the concentrations of PM10 and PM2.5 decreased during the six weeks of lockdown compared to the previous six weeks. PM10 and PM2.5 levels were reduced by 24–45% and 14–45%, respectively (Rojas et al., 2021). In the present study, for the same period of time, PM10 decreased by 39% and PM2.5 by 21% (significant reductions, $p < 0.05$), falling between the values obtained by Rojas et al. (2021).

Tobías et al. (2020) collected PM10 and BC data from 16/02/2020 to 13/03/2020 (before lockdown) and during lockdown (from 14/03/2020 to 30/03/2020), at the urban background and traffic air quality monitoring stations in the city of Barcelona, Spain. Comparing the two periods, they found that the mean concentrations of PM10 decreased by 28%–31%, during the lockdown period compared to previous weeks without lockdown; for BC, the reduction oscillated between 45% and 51%. In the present study, analysing the same period, the decrease in PM10 was 9.2% (non-significant reduction, $p > 0.05$), much lower than that found by Tobías et al. (2020). And for BC there was an increase of 7.4% (non-significant reduction, $p > 0.05$).

These abovementioned results indicate that the decrease/increase in PM10, PM2.5 and BC will largely depend on the period of time analysed before and during the lockdown (Fig. 5). The variations (increases or decreases) found between both periods, in PM10, PM2.5 and BC, also depend on the emission sources and the weather conditions in the area where the station is located. The area studied here is strongly influenced by industrial activity, which did not stop during the lockdown, while in other places analysed emissions depend mainly on traffic, which decreased considerably during the lockdown (Collivignarelli et al., 2020; Tobías et al., 2020; Rojas et al., 2021, etc).

4. Conclusions

In the present study, PM10, PM2.5 and BC were analysed in a sub-urban area in the North of Spain affected by nearby industrial activities. The influence of the lockdown restrictions due to the COVID-19 pandemic (from May 2019 to June 2020) and wind speed and wind direction were considered in the assessment.

- The highest concentrations of PM10, PM2.5 and BC occurred in DEF (2019–2020). The highest diurnal concentrations occurred at night.
- The highest concentrations of PM10, PM2.5 and BC were associated with SW winds, blowing from where the integrated steel industry is located. Indeed, the statistical analysis has confirmed that those levels were significantly higher than the ones recorded under different wind directions.
- Correlations between the pairs PM10-PM2.5, PM10-BC and PM2.5-BC were $R^2 = 0.67$, $R^2 = 0.48$ and $R^2 = 0.52$, respectively. The relatively low values between PM2.5 and BC compared with those in the literature might indicate the presence of secondary pollutants that contributed to PM2.5 such as sulphates, nitrates, and ammonium.
- The variations in the levels of PM10, PM2.5 and BC during the lockdown are highly dependent on the period of time analysed (i.e., the dates stablished as the period "before restrictions" and the period "during restrictions"). Some authors considered the period "before restrictions" as the previous year(s), whereas others used data from the previous months of the same year, when restrictions had still not been stablished. In the present study, PM10 was found to decrease by 39% or increase by up to 12% depending on the period considered. Likewise, PM2.5 decreased by 21% or increased by up to 36% and BC decreased by 42% or increased by up to 7.4%. These results highlight the degree of complexity when comparing with other studies of the literature.

CRediT authorship contribution statement

Rosa Lara: Investigation, Data curation, Formal analysis, Writing, Writing – review & editing. **Laura Megido:** Formal analysis, Writing – review & editing, Supervision. **Beatriz Suárez-Peña:** Investigation, Writing – review & editing. **Luis Negral:** Investigation. **Yolanda Fernández-Nava:** Writing – review & editing. **Jesús Rodríguez-Iglesias:** Writing – review & editing. **Elena Marañón:** Writing – review & editing. **Leonor Castrillón:** Writing, Funding acquisition, Project administration, Supervision.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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References

Beddows, D.C.S., Harrison, R.M., Green, D.C., Fuller, G.W., 2015. Receptor modelling of both particle composition and size distribution from a background site in London. UK. Atmospheric Chemistry and Physics 15, 10107–10125. https://doi:10.5194/acp-15-10107-2015.

Benchrif, A., Wheida, A., Tahri, M., Shubbar, R.M., Biswas, B., 2021. Air quality during three covid-19 lockdown phases: AQI, PM2.5 and NO2 assessment in cities with more than 1 million inhabitants. Sustain. Cities Soc. 74, 103170 https://doi.org/10.1016/j.scs.2021.103170.

Bodor, Z., Bodor, K., Keresztesi, A., Szép, R., 2020. Major air pollutants seasonal variation analysis and long-range transport of PM10 in an urban environment with specific climate condition in Transylvania (Romania). Environ. Sci. Pollut. Control Ser. 27 (30), 38181–38199. https://10.1007/s11356-020-09838-2.

Briz-Redón, A., Belenguer-Sapiña, C., Serrano-Aroca, A., 2021. Changes in air pollution during COVID-19 lockdown in Spain: a multi-city study. Journal of Environmental Sciences 101, 16-26. https://doi.org/10.1016/j.jes.2020.07.029.

Carslaw, D.C., Beevers, S.D., Ropkins, K., Bell, M.C., 2006. Detecting and quantifying aircraft and other on-airport contributions to ambient nitrogen oxides in the vicinity of a large international airport. Atmos. Environ. 40 (28), 5424–5434. https://doi.org/10.1016/j.atmosenv.2006.04.062.

Carslaw, D.C., Ropkins, K., 2012. "openair — An R package for air quality data analysis." Environmental Modelling & Software 27–28, 52–61. https://doi.org/10.1016/j.envsoft.2011.09.008, 0.

Cha, Y., Lee, S., Lee, J., 2019. Measurement of black carbon concentration and comparison with PM10 and PM2.5 concentrations monitored in Chungcheong province, Korea. Aerosol Air Qual. Res. 19 (3), 541–547. https://doi.org/10.4209/aaqr.2018.08.0325.

Collivignarelli, M.C., Abbà, A., Bertanza, G., Pedrazzani, R., Ricciardi, P., Carnevale Miino, M., 2020. Lockdown for CoViD-2019 in Milan: what are the effects on air quality? Sci. Total Environ. 732 (139280) https://doi.org/10.1016/j.scitotenv.2020.139280.

Czarnecka, M., Nidzgorska-Lencewicz, J., 2017. The impact of thermal inversion on the variability of PM10 concentration in winter seasons in Tricity. Environ. Protect. Eng. 43 (2), 157–172. https://doi.org/10.5277/epe170213.

Du, Y., Xu, X., Chu, M., Guo, Y., Wang, J., 2016. Air particulate matter and cardiovascular disease: the epidemiological, biomedical and clinical evidence. Thorac Dis 8 (1), 8–19. https://doi.org/10.3978/j.issn.2072-1439.2015.11.37.

Fagerland, M.W., Sandvik, L., 2009. The wilcoxon–mann–whitney test under scrutiny. Stat. Med. 28, 1487–1497. https://doi.org/10.1002/SIM.3561.

Feng, S., Gao, D., Liao, F., Zhou, F., Wang, X., 2016. The health effects of ambient PM2.5 and potential mechanisms. Ecotoxicol. Environ. Saf. 128, 67–74. https://doi.org/10.1016/j.ecoenv.2016.01.030.

Grange, S.K., Lewis, A.C., Carslaw, D.C., 2016. Source apportionment advances using polar plots of bivariate correlation and regression statistics. Atmos. Environ. 145, 128–134. https://doi.org/10.1016/j.atmosenv.2016.09.016.

Gong, W., Zhang, T., Zhu, Z., Ma, Y., Ma, X., Wang, W., 2015. Characteristics of PM1.0, PM2.5, and PM10, and their relation to black carbon in wuhan, Central China. Atmosphere 6, 1377–1387. https://doi.org/10.3390/atmos6091377.

GPA, 2020. BOPA provisions related to COVID-19. Government of the Principality of Asturias, 24.01.23. https://coronavirus.asturias.es/bopa1.

Hayes, R.B., Lim, C., Zhang, Y., Cromar, K., Shao, Y., Reynolds, H.R., Silverman, D.T., Jones, R.R., Park, Y., Jerrett, M., Ahn, J., Thurston, G.D., 2020. PM2.5 air pollution and cause-specific cardiovascular disease mortality. Int J Epidemiol 49 (1), 25–35. https://doi.org/10.1093/ije/dyz114.

Jephcote, C., Hansell, A.L., Adams, K., Gulliver, J., 2021. Changes in air quality during COVID-19 'lockdown'in the United Kingdom. Environ. Pollut. 272, 116011 https://doi.org/10.1016/j.envpol.2020.116011.

Kottek, M., Grieser, J., Beck, C., Rudolf, B., Rubel, F., 2006. World map of the Köppen-Geiger climate classification updated. Meteorol. Z. 15, 259–263. https://doi.org/10.1127/0941-2948/2006/0130.

Kruskal, W.H., Wallis, W.A., 1952. Use of ranks in one-criterion variance analysis. J. Am. Stat. Assoc. 47, 583. https://doi.org/10.2307/2280779.

Lara, R., 2023. Suspended and settleable particulate matter in the western area of the conurbation of Gijón. In: Doctoral Thesis. University of Oviedo.

Lara, R., Megido, L., Negral, L., Suárez-Peña, B., Castrillón, L., 2022. Impact of COVID-19 restrictions on the dry deposition fraction of settleable particulate matter at three industrial urban/suburban locations in northern Spain. Atmos. Environ. 284, 119216. https://doi.org/10.1016/j.atmosenv.2022.119216.

Lara, R., Suárez-Peña, B., Megido, L., Negral, L., Rodríguez-Iglesias, J., Fernández-Nava, Y., Castrillón, L., 2021. Health risk assessment of potentially toxic elements in the dry deposition fraction of settleable particulate matter in urban and suburban locations in the city of Gijón. Spain. J. Environ. Chem. Eng 9, 106794. https://doi.org/10.1016/j.jece.2021.106794.

Li, T., Hu, R., Chen, Z., Li, Q., Huang, S., Zhu, Z., Zhou, L.F., Li, T., Hu, R., Chen, Z., Li, Q., Huang, S., Zhu, Z., Zhou, L.-F., 2018. Fine particulate matter (PM2.5): the culprit for chronic lung diseases in China. Chronic Diseases and Translational Medicine 4, 176–186. https://doi.org/10.1016/j.cdtm.2018.07.002.

Mann, H.B., 1945. Nonparametric tests against trend. The Econometric Society Stable 13, 245–259. https://doi.org/10.2307/1907187.

Megido, L., Suárez-Peña, B., Negral, L., Castrillón, L., Suárez, S., Fernández-Nava, Y., Marañón, E., 2016. Relationship between physico-chemical characteristics and potential toxicity of PM10. Chemosphere 162, 73–79. https://doi.org/10.1016/j.chemosphere.2016.07.067.

Megido, L., Negral, L., Castrillon, L., Fernandez-Nava, Y., Suarez-Peña, B., Marañón, E., 2017a. Impact of secondary inorganic aerosol and road traffic at a suburban air quality monitoring station. J. Environ. Manag. 189, 36–45. https://doi.org/10.1016/j.jenvman.2016.12.032.

Megido, L., Suárez-Peña, B., Negral, L., Castrillón, L., Fernández-Nava, Y., 2017b. Suburban air quality: human health hazard assessment of potentially toxic elements in PM10. Chemosphere 177, 284–291. https://doi.org/10.1016/j.chemosphere.2017.03.009.

Menut, L., Bessagnet, B., Siour, G., Mailler, S., Pennel, R., Cholakian, A., 2020. Impact of lockdown measures to combat Covid-19 on air quality over western Europe. Sci. Total Environ. 741, 140426 https://doi.org/10.1016/j.scitotenv.2020.140426.

Milinković, A., Gregorič, A., Grgičin, V.D., Vidič, S., Penezić, A., Cvitešić Kušan, A., Alempijević, S.B., Kasper-Giebl, A., Frka, S., 2021. Variability of black carbon aerosol concentrations and sources at a Mediterranean coastal region. Atmos. Pollut. Res. 12 (11), 101221 https://doi.org/10.1016/j.apr.2021.101221.

Negral, L., Suárez-Peña, B., Amado, Á., Megido, L., Lara, R., Marañón, E., Castrillón, L., 2021. Settleable matter in a highly industrialized area: chemistry and health risk assessment. Chemosphere 274, 129751. https://doi.org/10.1016/j.chemosphere.2021.129751.

Patel, H., Nick Talbot, N., Salmond, J., Dirks, K., Xie, S., Davy, P., 2020. Implications for air quality management of changes in air quality during lockdown in Auckland (New Zealand) in response to the 2020 SARS-CoV-2 epidemic. Sci. Total Environ. 746, 141129 https://doi.org/10.1016/j.scitotenv.2020.141129.

Port of Gijón, 2021. Annual reports. Port Authority of Gijón, 04.02.23. https://www.puertogijon.es/en/port-authority/traffic-statistic-annual-reports-2/.

PRTR, 2023. The European pollutant release and transfer register, 29.01.23. http://prtr.ec.europa.eu/.

Querol, X., Massagué, J., Alastuey, A., Moreno, T., Gangoiti, G., Mantilla, E., Duéguez, J. J., Escudero, M., Monfort, E., Pérez García-Pando, C., Petetin, H., Jorba, O., Vázquez, V., de la Rosa, J., Campos, A., Muñóz, M., Monge, S., Hervás, M., Javato, R., Cornide, M.J., 2021. Lessons from the COVID-19 air pollution decrease in Spain: now what? Sci. Total Environ. 779, 146380 https://doi.org/10.1016/j.scitotenv.2021.146380.

Radics, M., Christidis, P., 2022. Impact of the COVID-19 Pandemic on Mobility in Spain, EUR 31033 EN. Publications Office of the European Union, Luxembourg, JRC128451. https://doi.org/10.2760/040554. ISBN 978-92-76-49855-1.

Rigueira, X., Araújo, M., Martínez, J., García-Nieto, P.J., Ocarranza, I., 2022. Functional data analysis for the detection of outliers and study of the effects of the COVID-19 pandemic on air quality:A case study in Gijón. Spain. Mathematics 10 (2374). https://doi.org/10.3390/math10142374.

Ródenas, M., Soler, R., Borrás, E., Vera, T., Diéguez, J.J., Muñoz, A., 2022. Assessment of COVID-19 lockdown impact on the air quality in eastern Spain: PM and BTX in urban, suburban and rural sites exposed to different emissions. Atmosphere 13 (97). https://doi.org/10.3390/atmos13010097.

Rojas, J.P., Francesco, R., Urdanivia, F.R., Roy, A., Garay, R.A., Alan, J., García, A.J., Enciso, C., Medina, E.A., Toro, R.A., Manzano, C., Leiva Guzmán, M.A., 2021. Effects of COVID 19 pandemic control measures on air pollution in Lima metropolitan area, Peru in South America. Air Quality. Atmosphere & Health 764, 142874. https://doi.org/10.1007/s11869-021-00990.

Santoso, M., Lestiani, D.D., Damastuti, E., Kurniawati, S., Kusmartinia, I., Atmodjo, D.P. D., Sari, D.K., Muhtarom, T., Permadi, D.A., Philip, K., Hopke, P.K., 2020. Long term characteristics of atmospheric particulate matter and compositions in Jakarta, Indonesia. Atmos. Pollut. Res. 11, 2215–2225. https://doi.org/10.1016/j.apr.2020.09.006.

Sokhi, R.S., Vikas Singh, V., Querol, X., Finardi, S., Targino, A., Andrade, M.F., Pavlovic, R., Garland, R.M., Massagué, J., Kong, S., Baklanov, A.S., Ren, L., Tarasova, O., Carmichael, G., Peuch, V., Anand, V., Arbilla, G., Badali, K., Zavala, M., 2021. A global observational analysis to understand changes in air quality during exceptionally low anthropogenic emission conditions. Environ. Int. 157, 106818 https://doi.org/10.1016/j.envint.2021.106818.

Seita, R., 2018. Arctic climate governance via EU law on black carbon? John Wiley & Sons Ltd. Downloaded from University of Eastern Finland's eRepository. https://doi.org/10.1111/reel.12241. https://erepo.uef.fi/handle/123456789/6748.

Shakoor, A., Chen, X., Farooq, T.H., Shahzad, U., Ashraf, F., Rehman, A., Sahar, N., Yan, W., 2020. Fluctuations in environmental pollutants and air quality during the lockdown in the USA and China: two sides of COVID-19 pandemic. Air Quality. Atmosphere & Health 13, 1335–1342. https://doi.org/10.1007/s11869-020-00888-6.

Shapovalova, D., 2016. The effectiveness of the regulatory regime for black carbon mitigation in the arctic. Arctic Review on Law and Politics 7 (2), 136–151.

Tobías, A., Carnerero, C., Reche, C., Massagué, J., Via, M., Minguillón, M.C., Alastuey, A., Querol, X., 2020. Changes in air quality during the lockdown in Barcelona (Spain) one month into the SARS-CoV-2 epidemic. Sci. Total Environ. 726, 138540 https://doi.org/10.1016/j.scitotenv.2020.138540.

Toro, R., Catalán, F., Leiva-Guzman, M., 2021. Air pollution and COVID-19 lockdown in a large south American city: Santiago metropolitan area, Chile. Urban Clim. 36, 100803 https://doi.org/10.1016/j.uclim.2021.100803.

Viidanoja, J., Sillanpää, M., Laakiaa, J., Kerminen, V.M., Hillamo, R., Aarnio, P., Koskentalob, T., 2002. Organic and black carbon in PM2.5 and PM10: 1 year of data from an urban site in Helsinki, Finland. Atmos. Environ. 36 (19), 3183–3193. https://doi.org/10.1016/S1352-2310(02)00205-4.

von Schneidemesser, E., Mar, K.A., Saar, D., 2017. Black carbon in Europe – targeting an air pollutant and climate forcer, IASS policy brief. https://doi.org/10.2312/iass.2017.010.

WHO, 2020. COVID-19: Archived: WHO Timeline - COVID-19. World Health Organization, 25.01.23. https://www.who.int/es/news/item/27-04-2020-who-timeline–-covid-19.

WHO, 2021. What Are the WHO Air Quality Guidelines? World Health Organization, 16.06.22. https://www.who.int/news-room/feature-stories/detail/what-are-the-who-air-quality-guidelines.

Wirth, E., Betancourt-Odio, M.A., Cabeza-García, M., Zapatero-González, A., 2022. Footprints of COVID-19 on pollution in southern Spain. Atmosphere 13 (1928). https://doi.org/10.3390/atmos13111928.

Yu, P., Xu, R., Coelho, M.P., Saldiva, P., Shanshan, L., Zhao, Q., Mahal, A., Sim, M., Abramson, M., Guo, Y., 2021. The impacts of long-term exposure to PM2.5 on cancer hospitalizations in Brazil. Environ. Int. 154, 106671 https://doi.org/10.1016/j.envint.2021.106671.