Temporal Air Quality (NO_2, O_3 and PM_{10}) Changes in Urban and Rural Stations in Catalonia during COVID-19 Lockdown: An Association with Human Mobility and Satellite Data

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Abstract

In this study, changes in air quality by NO_2, O_3 and PM_{10} in Barcelona metropolitan area and other parts of Catalonia during the COVID-19 lockdown with respect to pre-lockdown and to previous years (2018 and 2019) were evaluated. Selected air monitoring stations included 3 urban (Gràcia, Vall d'Hebron and Granollers), 1 control site (Fabra Observatory), 1 semi-urban (Manlleu), and 3 rural (Begur, Bellver de Cerdanya, and Juneda). NO_2 lockdown levels showed a diminution, which in relative terms was maximum in two rural stations (Bellver de Cerdanya, -63% and Begur, -61%), presumably due to lower emissions from the ceasing hotel and ski resort activities during eastern holidays. In absolute terms and from an epidemiologic perspective, decrease in NO_2, also reinforced by the high amount of rainfall registered in April 2020, was more relevant in the urban stations around Barcelona. O_3 levels increased in the transited urban stations (Gràcia, +42%, and Granollers, +64%) due to the lower titration effect by NO_x. PM_{10} lockdown levels decreased, mostly in Gràcia, Vall d'Hebron and Granollers (-35, -39% and -39%, respectively) due to traffic depletion (-90% in Barcelona's transport). Correlation among mobility index in Barcelona (-100% in retail & recreation) and contamination was positive for NO_2 and PM_{10} and negative for O_3 (P<0.001). Satellite images evidenced two hotspots of NO_2 in Spain (Madrid and Barcelona) in April 2018 and 2019 that disappeared in 2020. Overall, the benefits of lockdown on air quality in Catalonia were evidenced with NO_2, O_3 and PM_{10} levels below WHOAQG values in most of stations opposed to the excess registered in previous years.

Highlights

- Air quality changes during COVID-lockdown compared to past years were studied in urban and rural areas.
- NO_2 and PM_{10} levels decreased while O_3 levels increased (lockdown *vs* pre-lockdown).
- Correlation among mobility index and contaminant levels was demonstrated.
- NO_2, O_3 and PM_{10} lockdown levels were below WHOAQC values in most stations.
- Satellite images of NO_2 showed two hotspots in Spain in 2019 that vanished in 2020.

Introduction

Monitoring studies of environmental pollution have always been necessary in order to evaluate the impact of air contaminants on human health and the environment. In the last decade, the fast-growing population around the world, especially localized in metropolitan areas, resulted in increments of industrialization, transport demand and transport flow. Large anthropogenic emissions from these sectors led to various environmental concerns regarding poor quality outdoor air, altered climate (Ramanathan and Feng 2009; Shakun et al. 2012) and harmful effects on human health (Kim et al. 2015; Anenberg et al. 2019). In the major metropolitan cities, nitrogen dioxide (NO_2), particulate matter ($PM_{2.5}$ and PM_{10}), and in minor concentrations, sulfur dioxide (SO_2), carbon monoxide (CO) and ozone (O_3) are among the most hazardous air pollutants (Cohen et al. 2017; Nuvolone et al. 2018).

However, in 2020, the outbreak of COVID-19, which started in China but quickly spread to many countries over the world and eventually turned into a global pandemic, caused a repercussion on the environmental panorama. On the 30th of January 2020, the World Health Organization declared a global health emergency (WHO 2020a; Sohrabi et al. 2020), and shortly after, many major human activities, in the field of transportation, industrial manufacturing, culture, and education, were globally constrained to prevent further spreading of SARS-CoV-2 virus. Pandemic lockdowns started in all parts of the world, generating reduced industrial production and energy consumption, lower road traffic and therefore, lower emissions of pollutants in the atmosphere (Isaifan 2020; Tobías et al. 2020; Mahato et al. 2020). This situation gives a unique opportunity to evaluate the impact of these restricted anthropogenic emissions on air quality. Various studies that evaluate the effects of COVID-19 lockdown on air contamination have already been performed in different parts of the world (Delhi (Srivastava et al. 2020), London (Kumari and Toshniwal 2020), Milan (Altuwayjiri et al. 2020), Lima (Kumari and Toshniwal 2020), Ghaziabad (Lokhandwala and Gautam 2020), Nigeria (Zabbey et al. 2020), Tunisia (Chekir and Ben Salem 2021), Baghdad (Hashim et al. 2021), and Spain (Tobías et al. 2020; Briz-Redón et al. 2021)). All these studies agree on the association between contingency measures and improvement in air quality, but also in clean beaches and environmental noise reduction (Zambrano-Monserrate et al. 2020). Nevertheless, for the Iberian Peninsula, most of the studies are concentrated on the two cities of Madrid and Barcelona (Baldasano 2020; Tobías et al. 2020), and the few ones covering a larger region (*e.g.*, Catalonia (Saez et al. 2020; Marquès et al. 2021)) are focused on the lockdown period. Thus, more in-depth evaluation on the effects of the lockdown on air contamination covering a largest period of time (*i.e.*, pre, during and post-lockdown), a wider region (*i.e.*, the metropolitan region of Barcelona extended to Catalonia too) and different types of geographical locations (*i.e.*, urban, semi-urban and rural) will provide new insights on the reasons and social aspects related to such reductions.

Within this context, the objective of this study is to assess the changes in three of the most emitted air pollutants (*i.e.*, NO_2, O_3 and PM_{10}) linked to COVID-19 lockdown restrictions in one of the most populated regions of Spain: Catalonia, including Barcelona and its metropolitan area. Given that the lockdown in Catalonia started on March 14th and lasted until June 21st (Mitjà et al. 2020), with April being the month with the strictest confinement restrictions, in this study two distinct data analysis approaches were followed. First, a study covering the pre-lockdown, lockdown and post-lockdown period was performed. Secondly, an exhaustive evaluation of the month of April (the period when the strictest lockdown was imposed) was performed. In both approaches, the data analysis compared the same time periods with the two immediately preceding years 2018 and 2019. Moreover, the analysis of air contamination has been performed in three types of air quality monitoring stations (urban, semi-urban and rural) and a control site, in order to evaluate the influence of geographical location on the monitored air pollution. In addition, the Satellite observations of Spain in April 2018, 2019 and 2020 showed the total tropospheric column of NO_2 and O_3 to add more evidences of air pollution changes linked to the COVID-19 lockdown. This study is structured to provide the following information (i) of the overall situation regarding air contamination and COVID-19 lockdown in Barcelona and 5 other parts of Catalonia; (ii) of the air quality stations, the acquired data and the methods used for their analysis; and (iii) discussion on the causes of air pollution reduction due to lockdowns, offering daily and hourly contaminant profiles and percentages of change of air contamination during lockdown compared to pre-lockdown and to previous years, associated to traffic and social aspects measured with mobility index.

Materials And Methods

2.1. Lockdown scenario in Barcelona and Catalonia

The first case of COVID-19 disease in Catalonia (NE Spain) was registered on February 25th, 2020 and the first death caused by the SARS-CoV-2 virus happened on March 6th, 2020 (see Fig. 1). Soon after, the Spanish government declared a state of alarm due to COVID-19 health crisis, which started with the publication of the Royal Decree 463/2020 (Gobierno de España 2020a) on March 14th and imposed the lockdown of all non-essential industries and activities. Stores, hotels and restaurants were ordered to close, together with shopping and administrative centers. Restrictions on mobility became obligatory, and remote working was imposed whenever possible. These measures became stricter on March 27th, time when only essential services were allowed to remain open (Gobierno de España 2020b).

Transition towards a new normality began on May 4th with the start of the de-escalation. In Catalonia, such de-escalation was gradual and was organized in four different phases: 0 or preparatory phase, 1 or initial phase, 2 or intermediate phase and 3 or advanced phase. The progress from one phase to another was specific for each region and was determined according to the capacities of the primary healthcare and hospital system, the epidemical situation and the implementation of collective protective measures. For this reason, there was a delay in the start of the different phases in Barcelona and metropolitan area respect to other parts of Catalonia (see Fig. 1 to know the exact dates for these phases). The end of the four-phase of de-escalation was produced on June 21st with the end of the state of alarm and the beginning of the "new normality".

2.2. NO_2, O_3 and PM_{10} air pollution

In this study, air pollution by NO_2, O_3 and PM_{10} was evaluated in Barcelona, its metropolitan area and other parts of Catalonia. NO_2 is part of the group of nitrogen oxides (NO_x) that includes nitrogen monoxide, nitrous acid and nitric acid. NO_2 primarily is emitted into the air from the burning of fuel of vehicles (cars, trucks and buses), power plants and combustion facilities and off-road equipment. The chemistry of NO_2 is complex (see Fig. 2), since it is subject to extensive further atmospheric transformations to form both ozone and particulate matter; for the latter, NO_2 is the precursor of other organic, NO_3^- and SO_4^{2-} particles currently measured as $PM_{2.5}$ or PM_{10}. Nitrogen dioxide exerts a range of health effects including effects on lung metabolism, structure, function, inflammation and host defense against pulmonary infections (WHO 2020b). The current World Health Organization Air Quality Guideline (WHOAQG) (WHO 2020b) reference values for nitrogen dioxide are 40 and 200 µg/m^3 for annual and 1-hour mean NO_2 concentration, respectively.

Ozone is the primary ingredient of photochemical smog and can produce harmful effects on human health, most of them associated with the respiratory system (*i.e.*, asthma, chest pain, throat irritation, coughing, reduced lung function and damaged lung tissue (WHO 2020b)). The formation processes of ozone are interdependent and complex and their reaction and production rates are not linear. The major part of ozone is created by chemical reactions between oxides of nitrogen (NO_X) and volatile organic compounds (VOC) emitted by cars, power plants, industries and natural (biogenic) sources in the presence of sunlight (*hv*) ($NO_x + VOC + hv \rightarrow O_3$, see reaction 1 in Fig. 2). Elevated ozone in polluted regions is usually due to the ozone production with VOC and NO_x during daytime. However, ozone concentrations are depressed through the process of NO_x titration. Such process consists on the removal of O_3 through reaction with NO_x ($NO + O_3 \rightarrow NO_2 + O_2$ or $NO_2 + O_3 \rightarrow NO_3 + O_2$; see respective ractions 2 and 3 in Fig. 2). The titration process can happen both at daytime and at nighttime, in the immediate vicinity of very large emissions of NO_x. The WHOAQG reference value of ozone is 100 µg/m^3 for 8-hour mean O_3 concentration.

PM refers to a complex mixture of substances: organic plus elemental carbon, mineral dust (Al and Si oxides, CO_3^{2-}, Ca, Mg, P, Fe, K and some other trace elements), marine aerosols ($SO_{4marine}^{2-}$, Cl^- and Na) and secondary inorganic phases ($SO_{4non-marine}^{2-}$, NH_4^+ and NO_3) (Querol et al. 2004). These substances can be in solid or liquid states and exhibit different properties (size distribution, gas–solid/liquid partitioning, toxicity, etc.). Different PM sources exist including building sector (construction, demolition and domestic heating), traffic (motor emissions and tire, pavement and brakes abrasion products), industry (high levels of sulfate, nitrate and other burning products), and natural (*i.e.*, marine aerosols and air masses, especially African dust) (Querol et al. 2004, 2008). In this study only the fraction of particulate matter that passes through a size selective impactor inlet with a 50% efficiency cut-off at 10 µm aerodynamic diameter (*i.e.*, PM_{10}) has been studied. The current WHOAQG reference values for this pollutant are 20 and 50 µg/m^3 for annual and daily PM_{10} concentration, respectively.

2.3. Air monitoring stations

Catalonia is located in the northeast corner of Spain and its capital city is Barcelona (see Fig. 3). Holding more than seven million inhabitants, Catalonia is the most populated urban area in the Mediterranean coast and Barcelona and its metropolitan area, with 5.5 million people is the second-most populated city in Spain and the fifth-most populous urban area in the European Union. The metropolitan area of Barcelona is composed of multiple urban centers that are closely connected to each other but also to other external surrounding areas. Other Catalan provinces studied are Lleida (0.14 million inhabitants) and Girona (0.1 million inhabitants). The main pillars of the Catalan economy are tourism and hospitality industry. Tourism is enhanced by the excellent connections within the Catalan capital, such as the port, a high speed train station, an international airport and the second largest trade fair of Europe (Ajuntament de Barcelona 2020a). All of them, but especially the port and airport involve a high flow of people, goods and vehicles. The average traffic volume in the main accesses of Barcelona is approximately 1.08 million vehicles per day, and constitutes one of the biggest emission sources of air pollutants (Ajuntament de Barcelona 2020a).

The air quality monitoring network in Barcelona and Catalonia is managed by the Generalitat de Catalunya (Spain) and is made up of 129 automatic remote stations distributed across 15 air quality zones (Gencat). In this study, we analyzed 8 of these stations, the ones containing full information of the pollution (NO_2, O_3 and PM_{10}) and meteorological parameters in the period of time studied, which were located in six distinct air quality zones (see Table 1). Among these eight air quality stations, three were urban (Gràcia, Vall d'Hebron and Granollers), one semi-urban (Manlleu) and one control site (Fabra Observatory), all of them located in the province of Barcelona, and the remaining three were rural: Juneda and Bellver de Cerdanya in the province of Lleida, and Begur (Costa Brava, NE Catalonia), in the province of Girona (Fig. 3). Information regarding the air quality monitoring stations, that includes the number of inhabitants and density, emission source, type of background, geocoordinates, altitude, province they belong to and the air quality zone (AQZ) to which they correspond are displayed in Table 1. As can be observed, the source of emission corresponds in all cases to background contamination except for Gràcia station, where traffic is responsible. Furthermore, it is important to highlight the particular features of Fabra Observatory, which is a control site located in the AQZ of Barcelona that shows especial characteristics since it is placed in Collserola Mountain at 415 m of altitude and receives less impact from the city. Such emplacement is useful to provide information of vertical contamination near the city of Barcelona, which complements superficial information registered in Gràcia and Vall d'Hebron.

Table 1
Information of the air quality monitoring stations.

Air quality monitoring station	Type of station	Emission source	Inhabitants	Density (inhabit/ km^2)	Area (km^2)	Latitude	Longitude	Altitude (m)	Province	Air quality zone (AQZ)
Gràcia	Urban	Traffic	120907	60300	4.,19	41º 23' 55" N	2º 9' 12" W	57	Barcelona	Area of Barcelona
Vall d'Hebron	Urban	Background	5687	7700	0.74	41º 25' 33" N	2º 8' 52" W	136	Barcelona	Area of Barcelona
Granollers	Urban	Background	62419	4073	14.9	41º 35' 55" N	2º 17' 13" W	133	Barcelona	Vallès Oriental
Observatori Fabra	Control site	Background	-	-	-	41º 25' 6" N	2º 7' 26" W	415	Barcelona	Area of Barcelona
Manlleu	Semi-urban	Background	20573	1194	17.2	42º 0' 11" N	2º 17' 14" W	460	Barcelona	Plana de Vic
Begur	Rural	Background	3925	190	20.7	41º 57' 31" N	3º 12' 46" W	200	Girona	Empordà
Bellver de Cerdanya	Rural	Background	2138	22	98.14	42º 22' 5" N	1º 46' 36" W	1060	Lleida	Pirineu Oriental
Juneda	Rural	Background	3475	73	47.3	41º 32' 38" N	0º 49' 47" W	255	Lleida	Terres de Ponent

2.4. Air quality monitoring data

Hourly concentrations of NO_2, O_3 and PM_{10} were measured from February 15th to August 31st of 2018, 2019 and 2020 in the above mentioned eight permanent air quality monitoring stations. NO_2 concentrations were measured by means of chemiluminescence according to the UNE method 77212:1993, using automatically operated MCV 30QL analyzers. Ozone concentrations were measured by means of UV photometry according to ISO FDIS 139464:1998, automatically operated with MCV 48 AUV analyzers. Finally, PM_{10} concentrations were measured by means of gravimetric determination, using manually operated high volume samplers MCV CAV-A/MS. The generated databases with all the concentrations measured were compiled by the Department of Air Monitoring and Control Service of the Generalitat de Catalunya. In this study, two periods of time were comparatively evaluated for the three years (2018, 2019 and 2020). First, the period compressed between 15th of February and the 31st of August was chosen, as a representation of a period of time including pre-lockdown, lockdown and post-lockdown and was used to evaluate the correspondence between contamination profiles and anthropogenic activity. Secondly, the month of April was studied in more detail, being this month a representation of a period of time when the strictest lockdown was applied, since the de-escalation process started at the beginning of May (see Fig. 1). The month of April was used to perform an exhaustive analysis of the effects of COVID-19 lockdown on air contamination.

For the analysis of the period of 15th February to 31st August, an initial database containing the experimental data measured every hour during 198 days was processed. Such database gave a vector of size (1 x 4752). A total number of 72 data vectors of this size were obtained, considering three years, eight air quality monitoring stations and the levels of three air pollutants. Folding these 72 long data vectors produce 72 data tables sized (198 x 24), one per year and monitoring station, where the 198 rows correspond to the 198 days monitored and the 24 columns correspond to the hour times when the contaminants were measured every day. To make all these data tables have the same size, the extra day of February 2020 was removed. All these data tables were used to evaluate the time trends of the three air pollutants during the pre, lockdown and post-lockdown periods, to determine the differences among periods by calculating the percentages of change, to define the specific profiles in each location associated to the activities carried out in each area and to study their correlation with the mobility index by calculating the Pearson's correlation coefficients.

For the analysis of the month of April, an initial database containing the experimental data measured every hour during 30 days gave a vector of size (1 x 720). A total number of 72 data vectors of this size were obtained, considering three years, eight air quality monitoring stations and three contaminants (3 x 8 x 3). Folding these 72 long data vectors produce 72 data tables sized (30 x 24), one per year and monitoring station, where the 30 rows correspond to the 30 days of April and the 24 columns correspond to the hour times when the contaminants were measured every day. These data tables were used to perform a comparative analysis of the levels of the three pollutants in April 2020 respect to the levels registered in April of the two immediately preceding years (*i.e.*, 2018 and 2019). Only these two years were selected since they include the time when the implementation of important measures to improve air quality in the city of Barcelona started; introduction of shared transport systems and a cycling infrastructure, reduction of space for vehicles and implementation of the low emission zones (LEZs (AMB)), which was first implemented on December 31st 2017 and finally put into permanent effect on January 1st, 2020. Thus, the composition of the vehicle fleet and the mobility structure of Barcelona in 2018 and 2019 significantly differs to that of previous years and makes those past years less comparable in the present study.

Raw data vectors containing all meteorological data are given in excel data files as Supplementary material to this manuscript. In all cases, occasional day missing data were replaced by the mean (average) of all the values in the same column of the data table.

2.5. Space Observations Data

Satellites in space provide global observation data for air quality monitoring over the Earth. For this study, data from Satellite measurements of background tropospheric NO_2 and O_3 concentrations for the region of the Iberian Peninsula supplied by S-5P/TROPOMI-ESA were used. The Sentinel-5P mission, launched by the European Space Agency in 2017, is a low-orbit polar Satellite used to monitor Earth's atmosphere with a high spatio-temporal resolution using the TROPOMI. Concretely, it is a multispectral sensor that registers reflectance values at ultraviolet-visible (250–500 nm), near-infrared (675–775 nm) and short-wave infrared (2305–2385 nm) wavelengths which measures concentrations of key atmospheric constituents such as O_3, CH_4, CO, SO_2, CH_2O, NO_2 and aerosol properties (Veefkind et al. 2012). In this study, the measurements of Sentinel-5P were used to collect and plot data of NO_2 and O_3 from April 2018 and 2019 (under no pandemic) versus April 2020 (during the pandemic, under the strictest lockdown restrictions). To do that, Satellite data were downloaded from https://scihub.copernicus.eu/ and further analyzed using Panoply software (NASA GISS).

2.6. Anthropogenic activity: mobility index

The Mobility index is a parameter calculated by Google and provided in COVID-19 Community Mobility Reports (Google reports) that indicates the change in daily human mobility (including number of visits and length of stay at different places) from the start of the lockdown restrictions (February 15th) until present, respect to a referential value. Such referential value corresponds to the mean value calculated for the five weeks (January 3rd to February 6th of 2020) previous to the lockdown. The MI is provided for six categories, the ones that are useful to indicate social distancing efforts together with access to essential services. These categories comprehend grocery and pharmacy (including grocery markets, food warehouses, farmer markets, specialty food shops, drug stores, and pharmacies), parks (including local and national parks, public beaches, marinas, dog parks, and public gardens), transit stations (including subway, bus, and trains stations), retail and recreation (containing restaurants, cafes, shopping centers, theme parks, museums, libraries, and movie theaters), residential (*i.e.*, places of residence) and workplaces (*i.e.*, places of work). The Community Mobility Datasets were created to be helpful to control the lockdown restrictions imposed by the governments and were constructed with anonymized sets of data from users who had turned on the Location History for their Google Accounts. Therefore, the overall MI data represents a sample of Google users at each part of the world and may not represent the exact behavior of all the population.

In this study, data of MI were tracked from Google (Google reports) for the region of Catalonia (Spain), covering the period of time compressed among February 15th and August 31st 2020, with the aim of better understanding the anthropogenic changes caused by the pandemic lockdown as well as the lockdown scenario in Catalonia.

Results And Discussion

3.1. Global air contamination from Satellite Observations

As observed in Fig. 4a and 4b, in April 2018 and 2019, under normal human activity and no lockdown restrictions, recorded NO_2 levels were higher in the two most populated, highly transited and industrialized cities of Spain: Madrid (5390 inhabitants/km^2) and Barcelona (16499 inhabitants/km^2), revealing two hotspots with levels of NO_2 up to 100 µmol/m^2. Contrarily, in the same period of time in 2020, under the strictest lockdown, transportation restrictions and industry emission shutdown led to a clear decrease in NO_2 emission. According to S-5P/TROMPOMI Satellite records and as observed in Fig. 4c, levels of NO_2 in the cities of Madrid and Barcelona were in the range of 0–40 µmol/m^2 on April 2020. Therefore, the two hotspots observable on April 2018 and 2019 (Figs. 4a and 4b) completely receded during the lockdown in April 2020, giving a map with homogeneous contamination distribution over the whole country (Fig. 4c). Similarly, Tobías et al. compared tropospheric NO_2 concentrations supplied by TROPOMI-ESA in the lockdown period respect to the pre-lockdown. The authors reported a -57% of decrease in 2020 (lockdown versus pre-lockdown) compared with a -22% of decrease for the same period of time in 2019. In addition, the Satellite images provided in their study also evidenced two hotspots of contamination in Spain, corresponding to Madrid and Barcelona, with NO_2 levels in the same order as ours (up to 200 µmol/m^2 in 2020).

Different concentration distribution changes were observed for O_3 (Fig. 4d-4e). According to S-5P TROPOMI data, total O_3 tropospheric column showed an increase in the Iberian Peninsula in April 2020 with respect to April 2019. As observed in Fig. 4d, levels of O_3 were high in the upper north-east part of Spain, but low in the rest of the Peninsula. In contrast, on April 2020, the amount of O_3 increased over all the country, achieving levels up to ~ 0.16 mol/m^2. Such increase of O_3 linked to the lockdown has been also reported in the literature (Tobías et al. 2020). No georeferenced Satellite images of O_3 on April 2018 neither of PM_{10} are provided in this study since they are not available by the S-5P/TROPOMI-ESA.

3.2. Traffic reduction linked to lockdown

The restrictions in mobility imposed during the lockdown in Spain resulted in serious limitations on traffic all over the country, producing dramatic decreases in vehicular mobility, as reported by the General Directorate of Traffic (DGT).

Data of traffic density in Spain provided by the DGT is divided in two periods of time considering the traffic ratios registered on an equivalent period of time in 2019. Period 1 of time (March 9th to June 7th) (Dirección General del Tráfico 2020a) corresponds to the period of time when the strictest lockdown was imposed, and period 2 of time (July 1st to August 31st) (Dirección General del Tráfico 2020b) responds to the end of the state of alarm and the beginning of the "new normality". The main recorded results for the two periods of time include: 1) long-distance movements all over the country fell 72.48 and 13.26% (periods 1 and 2, respectively) for light vehicles, and 33.77 and 2.25% (periods 1 and 2, respectively) for heavy vehicles; 2) traffic across the borders of Spain decreased by 81.85 and 28.77% (periods 1 and 2, respectively) with Portugal and by 76.68 and 20.09% (periods 1 and 2, respectively) with France; 3) entries to and exits from the city of Barcelona decreased by 55 and 17% in periods of time 1 and 2, respectively; and 4) traffic across the distinct Spanish cities decreased by 37 and 12.11% in periods of time 1 and 2, respectively. As observed, traffic depletion was larger in period 1, when the strictest lockdown was imposed by the Spanish government, and showed a recovery in period 2, after the end of the state of alarm.

Not only the travel across cities suffered a decrease but also traffic inside cities, especially in the city of Barcelona. Traffic data inside Barcelona was provided by Barcelona City Council's (Ajuntament de Barcelona 2020b, c), at different moments of the pandemic. During the first two weeks of lockdown, the maximum diminution was registered: public transport conducted only 10% of the usual trips, taxis performed only 5% of their services and travel by bicycles and other personal mobility vehicles decreased by 87% (Ajuntament de Barcelona 2020c). In phase 1 of de-escalation (see Fig. 1), the traffic inside the city of Barcelona was a bit higher compared to the two first weeks of lockdown: public transported conducted 19,9% of the usual trips, private vehicles performed only 47.3% of the movements, and travel by bicycles and other personal mobility devices only decreased by 23% (Ajuntament de Barcelona 2020b).

3.3. Meteorological characteristics during lockdown respect to previous years

Meteorological factors have a significant effect on atmospheric pollution. As stated by Gkatzelis et al. 2021, wind velocity, stability, and turbulence have an impact on the dilution, transport, and dispersion of chemicals. Sunshine activates the photochemical production of oxidants that constitute smog, while rainfall has an effect that eliminates from the atmosphere some particles and gases.

In this study, meteorological data were obtained from Barcelona open datasets (Opendata), supplied by the Meteorological Service of Catalonia (Meteocat). As observed in Table 2, some meteorological differences between April 2018, 2019 and 2020 were reported in the control site of Barcelona: Fabra Observatory. During the lockdown period, higher average temperatures and higher humidity were registered (+ 1.2 and + 0.3 ºC and + 3 and + 4.5 %RH, respect to 2019 and 2018, respectively). However, the most important meteorological variation was the total amount of rain registered in the catalan city. As observed in Table 2, the total rainfall registered in Fabra Observatory in April 2020 was 254.6 mm, a value + 6.75 and + 4.3 times higher than the rainfall recorded in 2019 and 2018, respectively. In fact, April 2020 achieved an historical record and stood as the more rainy April month registered in Fabra Observatory for the past 107 years, constituting a 454% of the meteorological average of this month (Betevé; Meteocat).

Table 2
Meteorological parameters registered in Fabra Observatory in April 2018, 2019 and 2020. Data supplied by Barcelona open data sets (Opendata).

Meteorological parameters registered in Fabra Observatory			
	April 2018	April 2019	April 2020
Mean daily temperature (ºC)	13.6	12.7	13.9
Maximum daily temperature (ºC)	24.9	21.1	21.2
Minimum daily temperature (ºC)	5.1	3.1	6.8
Relatively daily humidity (%)	69	70,5	73.5
Rainfall (mm)	58.6	37.7	254.6
Mean daily insolation (MJ/m^2)	22.5	18.3	21.2
Mean wind speed (m/s)	4.2	4.2	3.7
Average wind direction (º)	223	228	218
Maximum wind speed	18.2	21.1	20.2
Maximum wind direction	355	334	292

Finally, some other minor variations were detected regarding the insolation ratio (-0.5 MJ/m^2 lower in 2020 respect to the previous two years), in wind speed, which was a bit lower in April 2020 respect to April 2019 – 2018 (3.7 m/s versus 4.2 m/s), and in the wind direction.

Overall, the meteorological differences registered in April 2020 under pandemia respect to the same time in the previous two years (under no pandemia) were fundamentally due to the amount of rain recorded during the lockdown, which was much higher than in the previous two years.

3.4. Time trend profiles and percentage changes of NO_2, O_3 and PM_{10} for pre-, during and post-lockdown and its relation with anthropogenic mobility

Mobility index (MI) of different human activities along with mean daily profiles of contaminants before (February 15th to March 13th), during (March 14th to June 21st) and after lockdown (June 22nd to August 31st) were studied to evaluate the impact of the anthropogenic mobility on air contamination (Fig. 5). In addition, the percentage changes of average concentrations of contaminants during lockdown and after lockdown respect to the period of time before lockdown were calculated and are shown in Table 3. It can be observed that the levels vary according to urban, semi-urban and rural sampling stations, with the highest NO_2 and PM_{10} values in urban areas linked to a heavier traffic and mobility indices.

Table 3
Average concentrations (μgm^{-3}) and standard deviations of NO_2, O_3 and PM_{10} for time periods March 14th-June 21st (during lockdown) and June 22nd - August 31st (post-lockdown) together with the percentages of change (lockdown versus pre-lockdown and post-lockdown versus lockdown).

	February 15th- March 13th 2020 (pre- lockdown) **lockdown)**	**March 14th- June 21st 2020 (lockdown)**	**% of change** **respect to** **pre-lockdown**	**June 22nd- August 31st 2020 (post- lockdown)**	**% of change respect to** **pre-lockdown**
NO_2					
Gràcia	40.7 ± 5	21.5 ± 4	-47%	26.0 ± 3	-36%
Vall d'Hebron	27.7 ± 5	16.0 ± 4	-42%	18.8 ± 2	-32%
Granollers	38.0 ± 5	16.4 ± 4	-57%	20.2 ± 3	-47%
Fabra Observatory	10.3 ± 2	5.9 ± 2	-43%	8.5 ± 1	-17%
Manlleu	24.2 ± 2	10.5 ± 3	-57%	10.4 ± 2	-57%
Begur	4.9 ± 1	1.9 ± 0.4	-61%	2.2 ± 0.4	-55%
Bellver de Cerdanya	7.9 ± 2	2.9 ± 1	-63%	4.8 ± 2	-39%
Juneda	7.5 ± 1	5.5 ± 1	-27%	5.2 ± 1	-31%
O_3					
Gràcia	43.4 ± 5	61.6 ± 7	+ 42%	51.9 ± 4	+ 20%
Vall d'Hebron	54.1 ± 5	67.3 ± 6	+ 24%	62.5 ± 5	+ 16%
Granollers	35.0 ± 5	57.4 ± 7	+ 64%	56.4 ± 4	+ 61%
Fabra Observatory	73.3 ± 3	86.6 ± 9	+ 18%	85.3 ± 7	+ 16%
Manlleu	31.1 ± 7	46.8 ± 6	+ 50%	55.1 ± 6	+ 77%
Begur	67.5 ± 2	77.8 ± 9	+ 15%	78.1 ± 5	+ 16%
Bellver de Cerdanya	46.4 ± 7	51.4 ± 3	+ 11%	58.1 ± 7	+ 25%
Juneda	46.9 ± 12	58.0 ± 6	+ 24%	63.1 ± 6	+ 35%
PM_{10}					
Gràcia	28.3 ± 5	18.5 ± 3	-35%	22.8 ± 3	-19%
Vall d'Hebron	21.4 ± 3	13.0 ± 3	-39%	17.8 ± 2	-17%
Granollers	32.1 ± 4	19.5 ± 3	-39%	24.2 ± 2	-25%
Fabra Observatory	17.2 ± 3	12.5 ± 2	-27%	16.7 ± 2	-3%
Manlleu	32.1 ± 4	18.9 ± 4	-41%	23.0 ± 2	-28%
Begur	n.d.	n.d.	n.d.	n.d.	n.d.
Bellver de Cerdanya	13.2 ± 3	9.9 ± 3	-25%	15.1 ± 3	-14%
Juneda	21.3 ± 4	15.3 ± 2	-28%	20.8 ± 3	-2%
n.d.: no data available					

Figure 5a illustrates the changes in MI in terms of different human activities for the periods before, during and after lockdown in the three different regional areas where the air quality stations of this study are located (Barcelona, Girona and Lleida). It can be clearly observed that all the activities including transport, industries, social places, and educational sectors were running normally before lockdown (see curves in light-grey-shaded areas of Fig. 5a). However, after the beginning of the state of alarm and lockdown, the mobility index of all the human activities except for the residential (the latter showing an increment during lockdown) notably decreased (see curves in yellow-shaded areas of Fig. 5a). The decline of MI of human activities during the COVID lockdown reported in this study is in agreement with the findings of Zhang et al. 2020. In that study the authors evidenced the same MI trend not only in Spain but in other countries (*i.e.*, United States, France, Italy, Germany, United Kingdom, India, Bangladesh and Pakistan), in which the MI of all human activities decreased a large extent (up to -90% of drop) while the MI for residential activities significantly increased (up to + 30% of increment) since the start of the pandemic.

In Catalonia, the decline of MI was maximum in April (up to 100% decrease in retail and recreation), when the strictest lockdown was produced, and started to recover right up to 21st of June, when the state of alarm finished and the "new normality" started (see curves in dark-grey-shaded areas of Fig. 5a). Interestingly, in Girona and Lleida, the activity in parks after the lockdown suffered a substantial increase (up to 400% in Girona and up to 250% in Lleida) whereas in Barcelona, all the different type of human activities returned to normal levels.

In Fig. 5b, daily averages (24 h means) of NO_2, O_3 and PM_{10} concentration (μg/m^3) for the equivalent period of time (*i.e.*, before, during and after lockdown) are represented.

Concerning NO_2, averaged concentrations of this contaminant substantially decreased during lockdown period in 2020 in contrast to the same period of time in 2018 and 2019. Moreover, the differences in the amount of NO_2 were evident when comparing the periods of time pre-lockdown, lockdown and post-lockdown during 2020. The percentages of decrease in the eight monitoring stations during the lockdown period respect to the pre-lockdown are shown Table 3 and followed this order: Bellver de Cerdanya (-63%) > Begur (-61%) > Manlleu and Granollers (-57%) > Gràcia (-47%) > Fabra Observatory (-43%) ~ Vall d'Hebron (-42%) and Juneda (-27%). The highest decrease in the two rural stations (Bellver the Cerdanya and Begur) is explained by the fact that they are widely populated in winter as the former is a ski resort and the latter a famous holiday and second residence emplacement and most houses are heated by gasoil or wood burning, which are emission sources of NO_2 (Michael Alberts 1994; Saud et al. 2011). However, in both sites people were asked to return to their main residence during lockdown, which was reflected in a high decrease in NO_2 levels. This did not happen in the third rural station (Juneda), since this is not a holiday spot. The urban and semi-urban areas had similar NO_2 decrease during lockdown and reflect the decrease in mobility observed in all urban areas. As expected, in the period of time right after the end of the state of alarm (June 22nd to August 31st), the levels of NO_2 incremented in all stations, but in no case they returned to the pre-lockdown levels (see last column of Table 3). Moreover, during the lockdown in 2020, the WHOAQG (EUR-Lex) daily reference value of 40 μg/m^3 was not exceeded in any site, although this standard value was exceeded in the three urban stations (Gràcia, Vall d'Hebron and Granollers) during the same period in 2018 and 2019. These results are in agreement with those reported by Baldasano, J.M. et al., who reported NO_2 levels below the WHOAQG reference value during the second half of March 2020 in 24 stations located in Madrid and 9 stations placed in Barcelona.

Concentrations of O_3 showed a substantial increase during lockdown, in the highly populated urban stations of Gràcia (+ 42%) and Granollers (+ 64%), and in the semi-urban station of Manlleu (+ 50%). In the rest of rural stations and in the control station, the registered percentages of change were lower, but still showing an increment of O_3 respect to the pre-lockdown period: Juneda and Vall d'Hebron (+ 24%), Fabra Observatory (+ 18%), Begur (+ 15%) and Bellver de Cerdanya (+ 11%) (see Table 3). Increment in O_3 levels during the lockdown is related to the reported diminution of NO_2 levels and the suppression of the titration effect and was more evident in the most transited and populated stations: Gràcia (60300 inhabit/km^2), Granollers (4121 inhabit/km^2) and Manlleu (1194 inhabit /km^2), see Table 1. Vall d'hebron, despite also being a populated station (7700 inhabit/km^2), showed a lower increment of O_3 since this station does not receive the direct impact of traffic. After the end of the state of alarm and the return to the "new normality", with increased traffic and NO_2 emissions, concentration of O_3 started to decrease again, still showing percentages of increase respect to pre-lockdown levels but in a lesser extend (see last column of Table 3). During the lockdown, the WHOAQG reference value of 100 μg/m^3 for O_3 was slightly exceeded in one time in Observatory Fabra, and more times in previous years in Fabra Observatory and Begur.

Concerning PM_{10}, concentrations of this contaminant during lockdown decreased in all stations, but in a minor extent in comparison to NO_2. The percentages of decrease during the lockdown respect to the pre-lockdown were as follows: Manlleu (-41%) > Granollers and Vall d'Hebron (-39%) > Gràcia (-35%) > Juneda (-28%) ~ Fabra Observatory (-27%) and Bellver de Cerdanya (-25%) (no data for Begur, see Table 3). Thus, highest decrease was observed for the semi-urban station of Manlleu and the urban stations of Granollers, Vall d'Hebron and Gràcia. In the stations of Fabra Observatory, Vall d'Hebron, Bellver and Juneda, levels of PM_{10} during lockdown in 2020 were lower than the WHOAQG annual reference value of 20 μg/m^3, in contrast to 2018 and 2019, when the reference value was exceeded. However, in Gràcia, Manlleu and Granollers, such reference value was slightly exceeded at some moments during the lockdown. The percentages of change and the levels of PM_{10} obtained in our study are in agreement with those reported by Tobías, A et al., in a study performed in two air quality stations in Barcelona. In that study the authors also reported PM_{10} levels slightly over the WHOAQG limit value in the station placed in the urban center of Barcelona, which suggests a location very similar to our Gràcia station.

Pearson's correlation coefficients of percentages of change of contaminants in the stations located in Barcelona (*i.e.*, Gràcia and Vall d'Hebron) respect to the mobility index, MI, in the same city were calculated for the period of time February 15th to August 31st and are summarized in Table 4. As it can be observed in the table, NO_2 showed a positive correlation with MI both in Gràcia (+ 0.51) and Vall d'Hebron (+ 0.38), which may suggest that the diminution of NO_2 levels was in a significant part caused by the MI reduction. In contrast, O_3 showed a negative correlation both in Gràcia (-0.56) and Vall d'Hebron (-0.41), indicating that the diminution of human mobility and traffic depletion contributed in an increase of O_3 levels in these two neighborhoods of Barcelona due to the lower titration effect. Finally, the correlation of PM_{10} with MI was, as which occurred with NO_2, positive in the two locations, despite a bit lower (+ 0.41, in Gràcia and + 0.33, in Vall d'Hebron), also indicating that part of the diminution of PM_{10} contamination can be attributed to traffic restrictions during lockdown.

Table 4
Pearson's correlation coefficient index among the mobility index (MI) and the percentage of change of contaminants for the period of time (February 15th - August 31st).

	NO_2	O_3	PM_{10}
	Pearson's correlation coefficient	Pearson's correlation coefficient	Pearson's correlation coefficient
Gràcia	+ 0.51	-0.56	+ 0.41
Vall d'Hebron	+ 0.38	-0.41	+ 0.33

3.5. Hourly profiles and percentage changes of NO_2, O_3 and PM_{10} during strictest lockdown: April 2020 versus April 2019 and 2018

Detailed evaluation of air contamination changes during the strictest lockdown was performed, focusing the study on data from April 2020 and comparing them to data acquired in April 2019 and April 2018, the latter used as basal concentrations. With that purpose, for these periods of time, hourly profiles of the contaminants and their percentages of change were calculated and evaluated to determine the sources of pollution and human impacts.

Hourly average profiles are represented in the plots of Fig. 6, each plot containing information of one contaminant, one station and three years simultaneously (in blue data from 2018, in green data from 2019 and in red data from 2020). Average values and the associated standard deviations were calculated for each hour as the mean ± SD of all the month of April (n = 30) and are represented in Fig. 6 with continuous lines and shaded areas, respectively.

In order to obtain the percentages of change, the mean concentrations of each contaminant in each station were first calculated as averages of the whole month of April for each year (see Table 5). Then, the percentages of change were calculated as the % of variation among April 2019 versus April 2018 and April 2020 versus April 2019. The reason why the percentages of change were calculated in that way and not considering April 2020 versus April 2018 is the fact that, in most locations, air quality was higher in 2019 respect to 2018. The improvement in air quality observed in 2019 can be attributed to a combination of factors: on the one hand, the implementation of LEZ and on the other hand, the weather patterns of 2019.

Daily profiles calculated from hourly averages shown in Fig. 6 evidence that NO_2 had similar hourly profiles in all stations, showing two maxima. The first maximum appeared between 8:00 and 10:00 am (two-hours delayed in Fabra Observatory), coincident with the rush traffic hour and, thus, can be assigned to the increasing fuel combustion by vehicles. It is interesting to stand out that despite the lockdown and the traffic depletion, this maximum was still observed. The second maximum, a bit lower and wider, appeared around 22:00 pm in all stations. However, mean ranges of NO_2 differed among stations. Higher levels were registered in highly populated urban stations of Gràcia, Vall d'Hebron and Granollers (~ 10–60 μgm^{-3}), followed by semi urban station of Manlleu and control site Fabra Observatory (~ 5–30 μgm^{-3}), ending with rural stations of Begur, Bellver de Cerdanya and Juneda (~ 2–15 μgm^{-3}). Thus, we observed a correlation between the NO_2 levels and the density of population of the air quality stations, the latter provided in Table 1. When comparing amounts of NO_2 among 2018 and 2019, in three stations (*i.e.*, Gràcia, Vall d'Hebron and Fabra Observatory) amount of NO_2 decreased, in Granollers' station there were no differences (*i.e.*, 0% of change) among these two years and in four stations (Manlleu, Begur, Bellver de Cerdanya and Juneda) there was an increment in NO_2 levels. In contrast, there was a uniform tendency in % of change of NO_2 levels among April 2020 versus April 2019 (see Table 5): in all stations the amount of this contaminant was lower in 2020 (under pandemic lockdown) respect to the same period of time in 2019 (under no pandemic). In particular, the percentages of change from higher to lower were as follows: Bellver de Cerdanya (-63%) ~ Fabra Observatory (-62%) > Granollers (-52%) > Gràcia (-45%) ~ Begur (-44%) > Manlleu (-38%) > Vall d'Hebron (-33%) > Juneda (-23%). As previously observed in Sect. 3.4., the rural station of Bellver de Cerdanya showed the highest relative decrease of NO_2, again presumably due to the ceasing hotel and ski resort activities during eastern holidays. These changes in the concentrations of NO_2 in the Bellver de la Cerdanya rural station are however only in relative terms and local. Absolute changes show that the NO_2 concentrations depletion was much more important in the Barcelona urban area than in the rural areas due to the lockdown situation. Moreover, the weather conditions of April 2020 in Barcelona urban area favored the cleansing of the atmosphere, including NO_2 gases, as they were especially rainy. The decrease of NO_2 reported in this study due to meteorological and lockdown restrictions is in accordance to previous air quality monitoring studies performed in the cities of Barcelona and Madrid (Baldasano 2020).

Moreover, in all stations NO_2 depletion was more evident in the second maximum and in general for the second part of the day (12-24h). In addition, it is worthy to stand out that in all stations, NO_2 concentration levels showed significantly lower standard deviation in 2020 respect to the previous years (narrowed red-shaded areas in Fig. 6). In the lockdown period, the WHOAQG annual reference value of 40 $\mu g/m^3$ was not exceeded in any site, whereas in previous years it was exceeded in Gràcia, Vall d'Hebron and Granollers stations (see Fig. 6), as previously observed when comparing the whole pre-, lockdown and post-lockdown period for the three years (Sect. 3.3).

Hourly average profiles of O_3 showed a marked minimum between 8:00 and 10:00 am (again two-hours delayed in Fabra Observatory), coincident with the maximum NO_2 concentration, a maximum around 16:00 pm, coincident with the increase of solar radiation and simultaneous to the NO_2 minimum between the two maxima, and a minimum around 22:00 pm, coincident with the second NO_2 maximum. The latter increase of O_3 at night can be attributed to the suppression of the titration effect (see Fig. 2).

Mean ranges of O_3 also varied among stations. Contrarily to NO_2, higher levels of O_3 were registered in Fabra Observatory and Begur stations (~ 60–120 μgm^{-3}), followed by Gràcia, Vall d'Hebron, Bellver de Cerdanya and Juneda (~ 20–100 μgm^{-3}), and finally with Granollers and Manlleu (~ 0-100 μgm^{-3}). When comparing O_3 levels among 2018 and 2019 we observed that there was already an increment in ozone in April 2019 respect to April 2018. Such increment was registered in all stations except for Begur, where a slightly decrease was detected. Differences in O_3 in April 2020 versus 2019 showed two different tendencies. In two stations the amount of O_3 was higher in April 2020 and the percentages of change, from higher to lower, were as follows: Gràcia (+ 12%) > Granollers (+ 3%). However, for the remaining six stations, the amount of O_3 was lower in April 2020 respect to the previous year, in the following order: Bellver de Cerdanya (-27%) > Manlleu (-18%) > Juneda (-16%) > Begur (-14%) > Vall d'Hebron (-9%) and Fabra Observatory (-5%). It is important to highlight that the increase produced in Gràcia (and in a lesser extend in Granollers) occurred in the second part of the day (12-24h, see Fig. 6), coincident with the previously reported decrease of NO_2. In the lockdown period, the WHOAQG 8-hour reference value of 100 µg/m^3 was not exceeded in any site, whereas in previous years it exceeded in Fabra Observatory and Begur stations (see Fig. 6).

PM_{10} hourly profiles showed a clear increase between 8:00 and 10:00 am in most of stations (Gràcia, Granollers, Manlleu, Bellver de Cerdanya and Juneda). Mean ranges of PM_{10} were similar for all the stations (~ 10–50 µgm^{-3}), except for the control site (Fabra Observatory), which registered the lowest levels (~ 5–30 µgm^{-3}). The comparison among years evidenced a decrease of PM_{10} in 2020 respect to 2019 in all stations, despite the diminution in Bellver de Cerdanya was little. Larger diminution was registered in the three more transited urban stations: Vall d'Hebron, showing a percentage of diminution of -60%, Gràcia, with a diminution of -25% and Granollers of -26%. This is due to the fact that PM levels are very dependent on the traffic influence (dust resuspension, erosion of road pavements and brakes), and during the lockdown, the density of vehicles in the city of Barcelona decreased: -90% in public transport, -95% in taxis and − 87% in bicycles and other personal mobility vehicles (see Sect. 3.2 of this manuscript). Lower percentages of decrease of PM_{10} in April 2020 respect to April 2019 were registered in the other stations of Juneda (-23%), Fabra Observatory (-21%) Manlleu (-18%) and Bellver de Cerdanya (-4%). No data of concentration of PM_{10} in Begur station in April 2020 were available and thus, their percentage of change could not be calculated. Moreover, in the lockdown period, the WHOAQG daily reference value of 50 µg/m^3 was not exceeded in any site, and the annual reference value of 20 µg/m^3 was only slightly exceeded among 10–12 hours in Gràcia, Granollers, Manlleu and Juneda, while highly exceeded in the same stations and also in Vall d'Hebron and Bellver de Cerdanya in previous years (see Fig. 6).

There are quite a few other studies concerning changes in air quality during the COVID lockdown in many areas throughout the globe (Menut et al. 2020; Ropkins and Tate 2021; Filonchyk et al. 2021), and most of them report NO_2 diminution, small increase in O_3 and PM_{10} diminution; the latter being generally a modest depletion in comparison to that of NO_2. For instance Menut et al. 2020, reported a large reduction in NO_2 concentrations, a lower reduction in particulate matter and a mitigated effect on ozone concentrations over western Europe. Filonchyk et al. 2021, reported reductions of tropospheric NO_2 approximately by -10 to 19%, and reductions of PM_{10} from − 8.5 to -33.9% in 2020 respect to 2019, in Poland, eastern Europe. Also Ropkins and Tate 2021, reported NO_2 decreased from − 32 to -50% and O_3 increase by + 20% across the United Kingdom. These findings are in accordance to the ones obtained in the present study. Therefore, the results hereby presented confirm the improvement of air quality due to the lockdown that has been observed worldwide, in the region of Catalonia (Spain).

However, a more exhaustive analysis of the acquired data using multivariate statistical and chemometric methods is pursued with the goal of the apportionment of the different sources of the three investigated air quality parameters (NO_2, O_3 and PM_{10}) and to describe how their temporal and geographical profiles changed during COVID-19. The results of this more exhaustive analysis will be hopefully reported when the COVID-19 pandemic situation finishes.

Table 5

Average concentrations (µgm^{-3}) and standard deviations (n = 30) of NO_2, O_3 and PM_{10} for April 2018, April 2019 and April 2020 together with the percentages of change (April 2019 versus 2018 and April 2020 and 2019).

	Average April 2018	Average April 2019	Average April 2020	% of change 2019 vs 2018	% of change 2020 vs 2019
NO_2					
Gràcia	49.2 ± 13	36.1 ± 10	19.7 ± 5	-27%	-45%
Vall d'Hebron	32.1 ± 10	25.7 ± 9	17.3 ± 6	-20%	-33%
Granollers	29.7 ± 7	29.7 ± 9	14.3 ± 5	0%	-52%
Fabra Observatory	13.7 ± 3	13.2 ± 2	5.0 ± 1	-4%	-62%
Manlleu	15.2 ± 3	16.5 ± 4	10.2 ± 2	+ 9%	-38%
Begur	3.0 ± 1	3.4 ± 1	1.9 ± 0.5	+ 13%	-44%
Bellver de Cerdanya	7.0 ± 1	7.8 ± 1	2.9 ± 1	+ 10%	-63%
Juneda	6.3 ± 2	8.2 ± 2	6.3 ± 1	+ 30%	-23%
O_3					
Gràcia	50.9 ± 23	61.1 ± 20	68.2 ± 20	+ 20%	+ 12%
Vall d'Hebron	62.1 ± 23	77.2 ± 25	70.6 ± 20	+ 24%	-9%
Granollers	55.5 ± 25	63.2 ± 31	64.9 ± 21	+ 14%	+ 3%
Fabra Observatory	88.8 ± 7	97.3 ± 8	92.2 ± 9	+ 10%	-5%
Manlleu	52.8 ± 19	60.1 ± 26	49.2 ± 18	+ 14%	-18%
Begur	101.0 ± 6	96.6 ± 7	83.2 ± 7	-4%	-14%
Bellver de Cerdanya	67.9 ± 18	70.2 ± 19	51.3 ± 13	+ 4%	-27%
Juneda	60.0 ± 23	68.9 ± 26	58.0 ± 23	+ 15%	-16%
PM_{10}					
Gràcia	31.7 ± 19	22.5 ± 10	16.8 ± 8	-29%	-25%
Vall d'Hebron	23.6 ± 13	27.2 ± 18	11.0 ± 6	+ 15%	-60%
Granollers	27.3 ± 15	23.8 ± 9	17.7 ± 6	-13%	-26%
Fabra Observatory	n.d.	15.4 ± 9	12.1 ± 7	n.d.	-21%
Manlleu	23.1 ± 13	18.4 ± 9	15.1 ± 7	-20%	-18%
Begur	n.d.	10.2 ± 5	n.d.	n.d.	n.d.
Bellver de Cerdanya	17.3 ± 13	10.4 ± 8	10 ± 6	-40%	-4%
Juneda	23.5 ± 17	18.2 ± 12	14.0 ± 7	-23%	-23%

n.d.: no data available

Conclusions

This study shows the impact of social movements on air quality by integrating chemical analysis, Satellite observation data and mobility indexes. It also demonstrates the importance to accurately evaluate the time frame, from days to years, to determine the changes and evolution of NO_2, O_3 and PM_{10} during the pandemic. In the 8 areas studied, considering urban, semi-urban and rural, the concentration of NO_2, O_3 and PM_{10} varied during the COVID-19 lockdown with respect to the pre-lockdown period and with respect to previous years. In the major part of air quality stations, the levels of these air contaminants were lower than the WHOAQG reference values. Only for O_3 the reference value was slightly exceeded one time during lockdown in the control station (Observatory Fabra) and for PM_{10} in four stations (Gràcia, Granollers, Manlleu and Juneda). However, these standards were exceeded multiple times in most stations (especially urban areas) during the same period of time in the two previous years.

In this study we observed a significant correlation among the levels of contaminants in the two stations located in Barcelona and the diminution in anthropogenic mobility registered in the same city (up to 100% decrease in retail and recreation). Pearson's correlation coefficients were positive for NO_2 and PM_{10}, since lower mobility and traffic depletion resulted in lower levels of these contaminants. Conversely, a negative correlation was observed for O_3, as the reduction of NO_2 emissions went together with the lower titration effect.

The overall consequences of the COVID-19 lockdown regarding NO_2, O_3 and PM_{10} air pollution extracted in the comparison of lockdown respect to pre-lockdown periods in 2020 were analogous to those extracted in the comparison of April 2020 (time for the strictest lockdown) respect to April 2019 (under no pandemic). For NO_2, the lockdown restrictions, together with the higher rainfall registered in 2020, especially in the Barcelona urban area, produced a decrease in the levels of this pollutant, especially in the urban stations in absolute terms, although also significant in relative terms because of the ceasing activities during eastern holidays in some rural stations close to ski hotels and resorts.

The effects of the lockdown regarding O_3 levels were opposed to those of NO_2. The traffic depletion originated by the lockdown resulted in a decrease of NO_2 levels, and thus, a suppression of the titration effect, resulting in higher amounts of ozone. Larger increments of O_3 were registered in the urban and most transited stations of Gràcia and Granollers. Levels of PM_{10} also suffered a depletion due to the lockdown, as occurred with NO_2, but in a lesser extent. Largest decrease in PM_{10} levels due to lockdown was registered in the three urban stations of Gràcia, Vall d'Hebron and Granollers. Satellite S-5P/TROMPOMI images confirmed the results for NO_2, by showing two hotspots of contamination in Spain (Madrid and Barcelona) in April 2018 and 2019 that disappeared in April 2020, and for O_3 by showing higher levels of O_3 all over the country in April 2020.

Overall, the scenario originated by the COVID-19 pandemic evidences that the reduction in traffic emissions in cities has clear effects on decreasing air contamination, which represents a significant improvement in public health and quality of life. The experimental evidences about the improvement of air quality during the pandemic have made possible to see cities with clean and healthier skies, which were not observed for decades and confirmed the potential of applying traffic restriction policies in the near future.

Declarations

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Supplemental Materials

Supplemental Materials are not available with this version

Figures

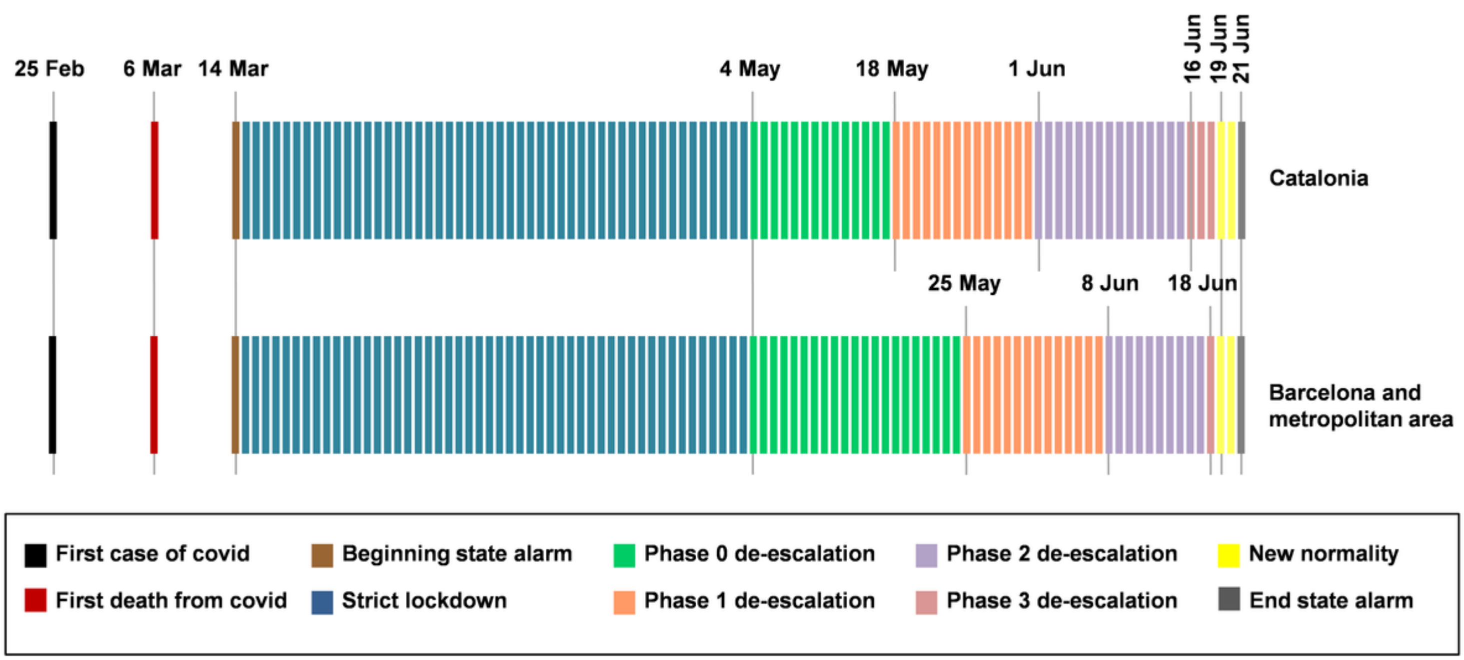

Figure 1

Dates of the scenario of COVID-19 health crisis (including start of state of alarm, beginning of lockdown and de-escalation phases) of Catalonia and Barcelona and its metropolitan area.

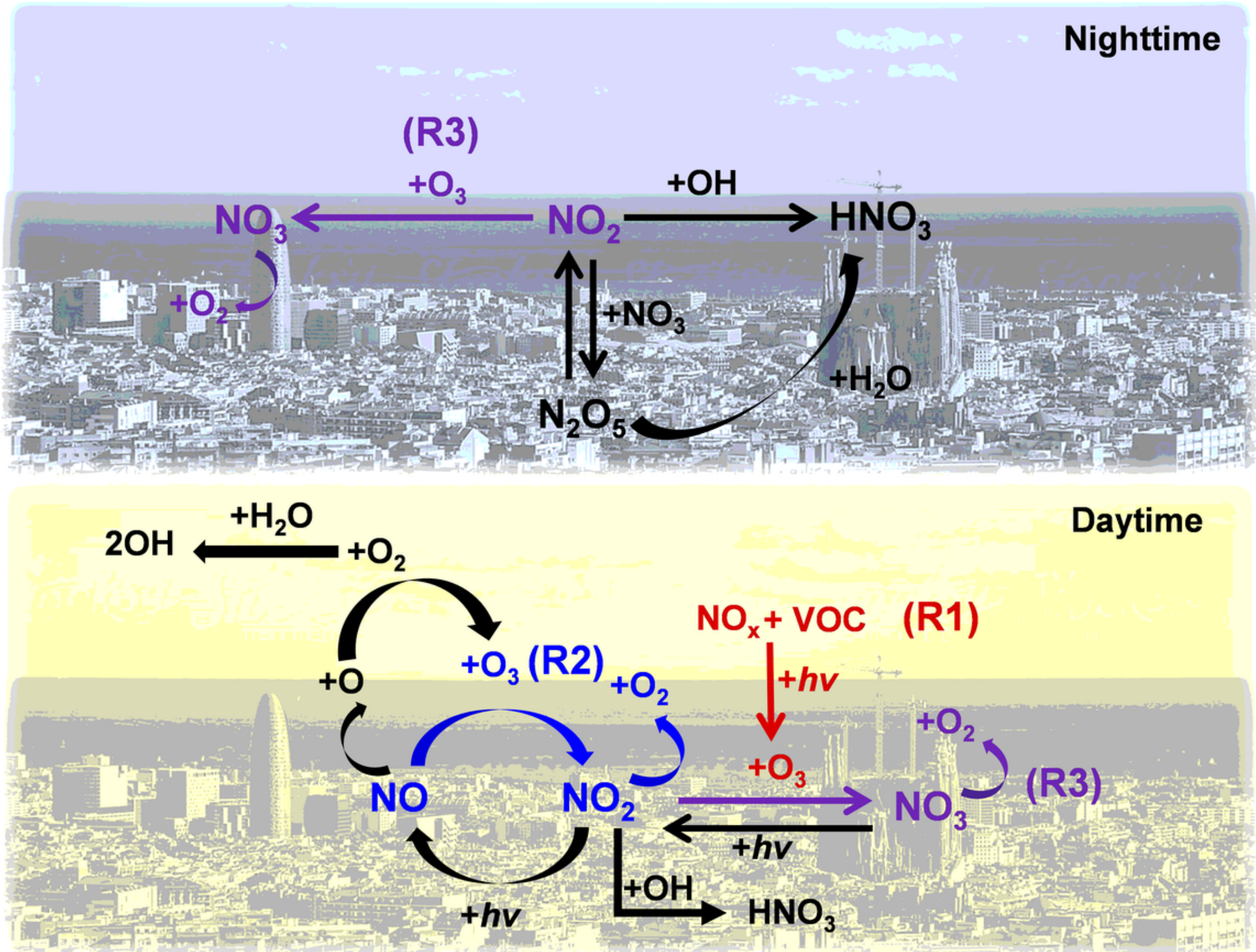

Figure 2

Reactions among pollutants (including NO2 and O3) in the troposphere; daytime versus nighttime. R1, R2 and R3 indicate the main reactions: ozone formation from NOx and VOCs (R1) and ozone suppression through NOx titration at daytime (R2) and at nighttime (R3).

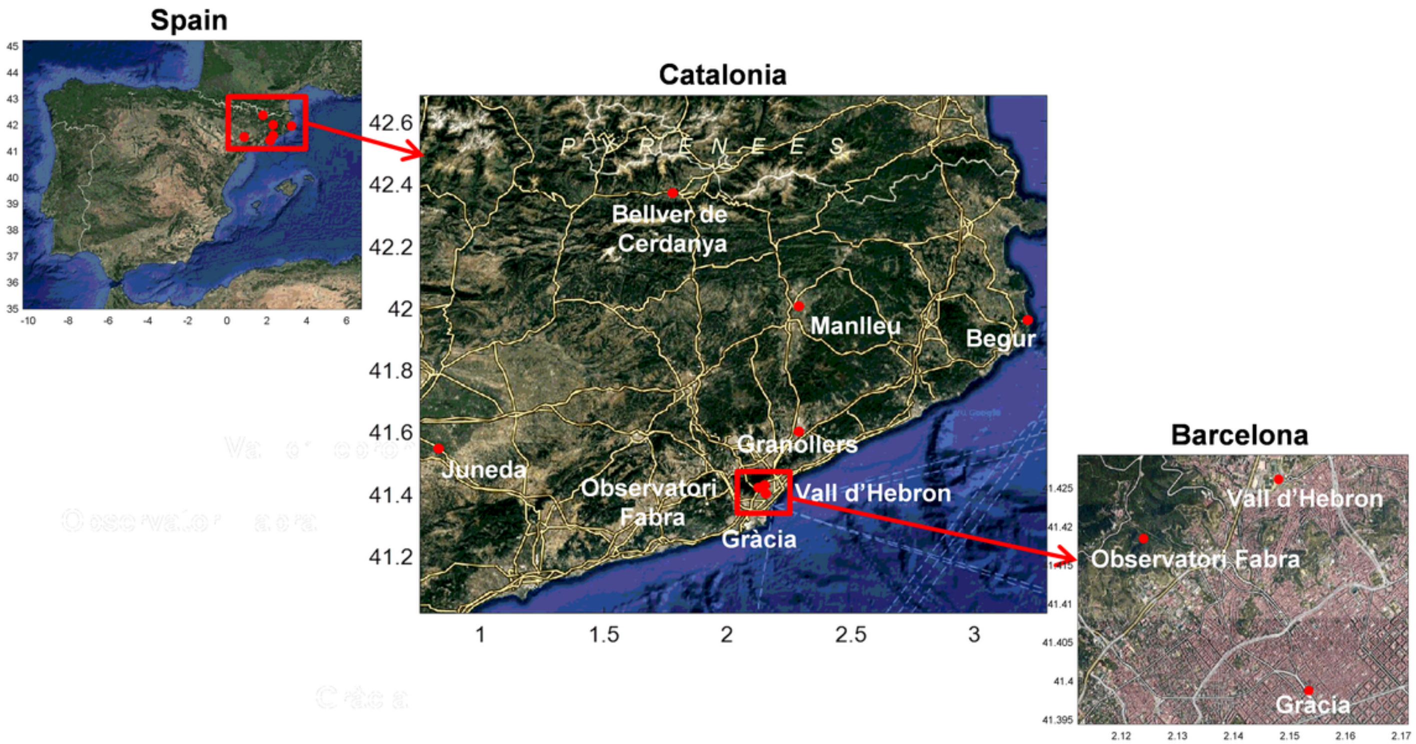

Figure 3

Map of Spain with a zoom in the region of Catalonia showing the eight air quality monitoring stations used in this study, also with a second zoom in the area of Barcelona.

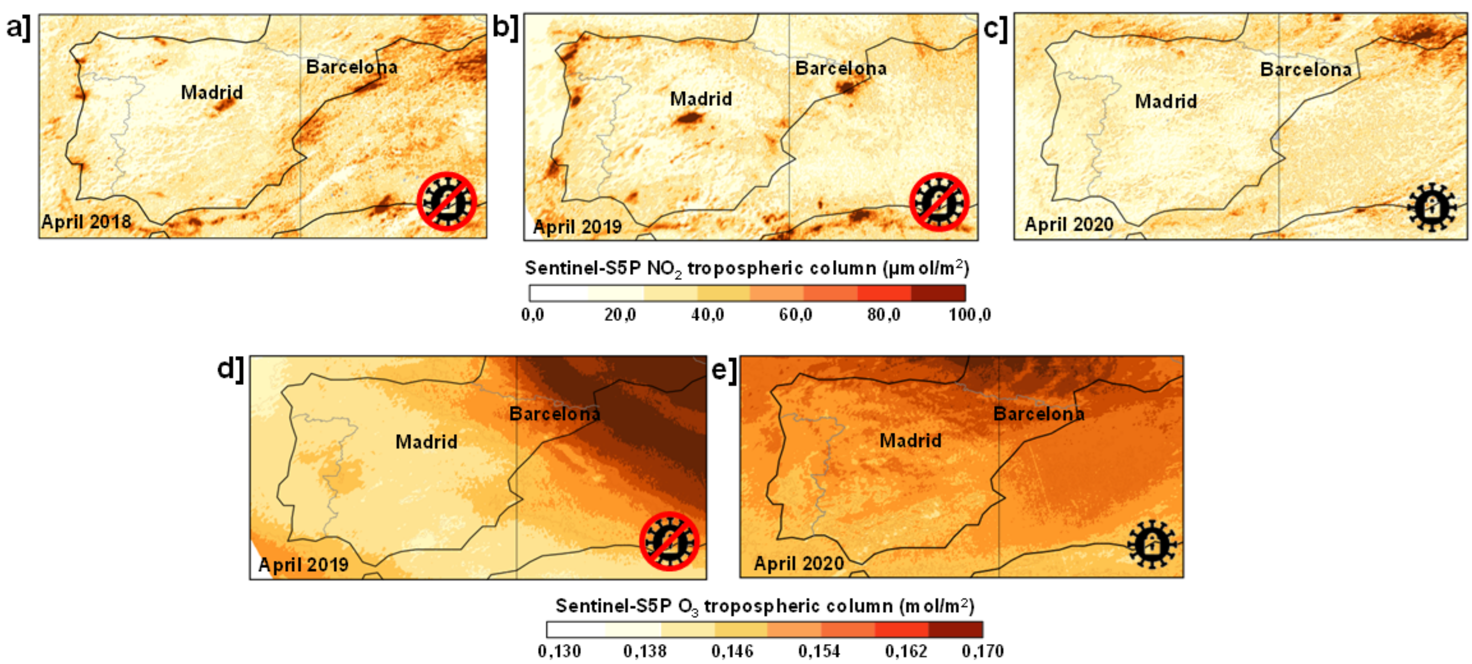

Figure 4

Levels of background tropospheric NO2 and O3 measured by TROPOMI-ESA (Veefkind et al. 2012) in the Iberian Peninsula. Top panels: NO2 tropospheric column measured on a] April 2018, b] April 2019, c] April 2020 (during the strictest lockdown). Bottom panels: O3 tropospheric column measured on d] April 2019, e] April 2020 (during the strictest lockdown).

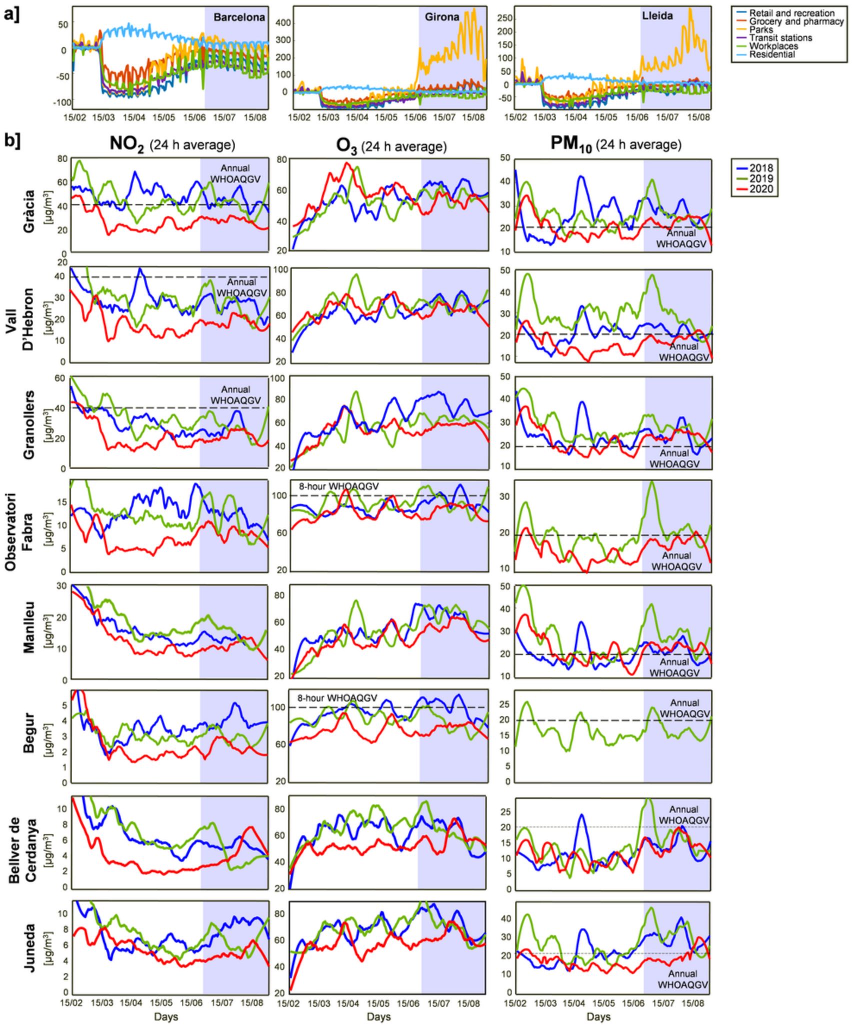

Figure 5

a) Mobility index (MI) of different human activities before (light grey-shaded area), during (yellow-shaded area) and after (dark grey-shaded area) the lockdown being put into effect. b) daily average (24 h) concentrations of the three contaminants from 15th February to 31st August 2018 (blue), 2019 (green) and 2020 (red). Observe that concentration values in y-axes are different depending on contaminant and station.

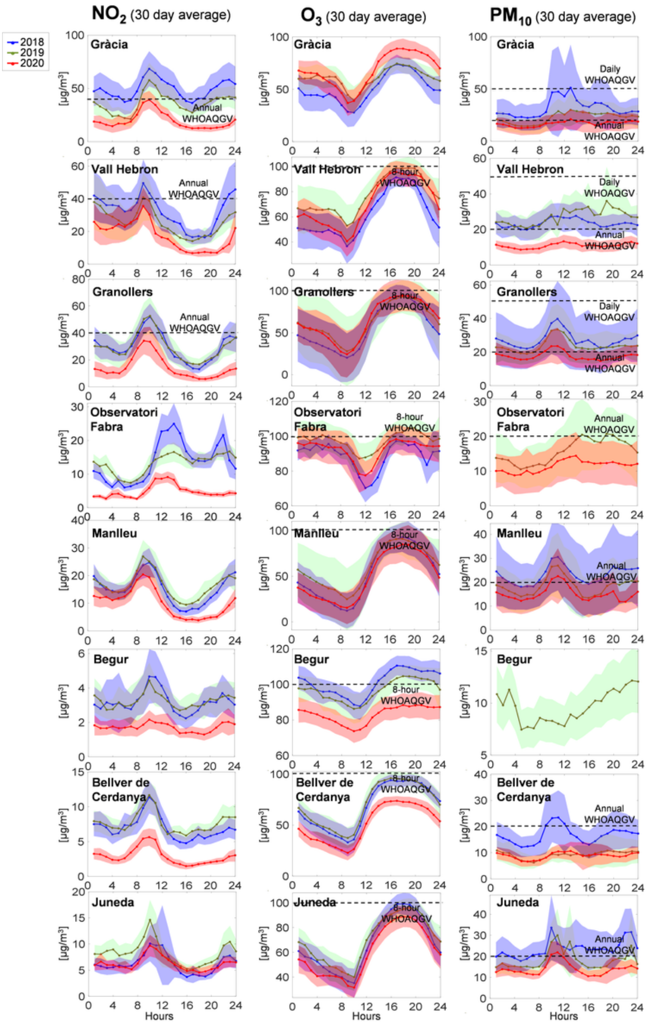

Figure 6

Hourly average (30 days) concentrations of NO2, O3 and PM10 together with their standard deviation (shaded areas) calculated during April 2018 (blue), 2019 (green) and 2020 (red) for the different air quality monitoring stations. Observe that concentration values in y-axes are different depending on contaminant and station.