Impact of COVID-19 Lockdowns on Air Quality and Health. Association between Concentrations of Tropospheric Ozone and Infection Cases and Deaths in Spain

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Research

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Abstract

Background

This work describes the changes of the air quality and the health implications caused by the lockdown of the first-wave provoked by the SARS-CoV-2 pandemic. Air pollutants were studied in 83 locations in Southern Spain. The study covered urban and industrial gases, NO_2, CO, SO_2, H_2S and O_3, and also PM_{10} and $PM_{2.5}$ particles.

Methods

It was evaluated the increase and decrease of concentrations during the state of alarm declared on 14th March. Pearson correlations for air pollutants, meteorological factors, vehicular traffic densities (VTDs) and data of infections and deaths caused by the COVID-19 disease were also assessed.

Results

It was found a clear reduction in carbon monoxide (-25% to -83%), particulate matter (-21% to -42%) and mainly nitrogen dioxide (-55% to -81%) in trafficked areas during the lockdown, reducing cardiovascular and respiratory problems. CO, SO_2 and H_2S increased (+26 to 34%, +68 to +85% and +32 to +84%) at industrial locations. O_3 increased along the lockdown period coinciding with reductions in NO_2 and CO (r = -0.90 and -0.81). This ozone rising constitute the ozone lockdown effect (OLE), increasing the risk of pneumonia hospital admissions. Regarding traffic, Pearson coefficients between ozone and VTDs were higher during lockdown than pre-lockdown period, and in the most trafficked areas a reduction in PM_{10} and $PM_{2.5}$ levels was observed, contributing this also to the OLE.

Conclusions

Effects of ozone on COVID-19 disease was revealed by the graphic associations and correlations found between O_3 levels and infection cases and deaths, which were remarkable, constituting in this case the ozone COVID effect (OCE): when concentrations of O_3 increase, the incidence of the disease is higher; when O_3 falls, infection cases are reduced.

Introduction

The crisis generated by COVID-19 pandemic is consolidating itself as a phenomenon unprecedented with unpredictable consequences. In the hope of finding a definitive vaccine, most of the governments induced their population to lockdown in their homes or cities. Considering the number of deaths and infection cases, Spain has been from the beginning one of the countries most affected worldwide and in Europe by this crisis [1].

In the first wave of the pandemic the Spanish Government declared the 'state of alarm' on Saturday 14th March 2020 [2]. As the days go by, the lockdown limited significantly the activity of the population. Once the trend of infections decreased, on 4th May the Government started the de-escalation process, probably too soon. Consequently, a first outbreak occurred on 17th May with people infected by a flight from abroad. At the end of July, Spain suffered numerous outbreaks that increased the spread exponentially to more than 1000 infections/day, more than 6000 cases in August/September and more than 20,000 (4,000 in Andalusia) in October/November [3].

End of September Spain declared the second wave with the highest rate of daily cases of COVID-19 with 38,273 new cases (data of 16th November), which was significantly higher in comparison with the new cases in Germany, 14,580, Italy, 27,352, United Kingdom, 21,363 or France with 9,072 cases [4]. The Region of Madrid (839 cases/100,000 inhab.) had many cities with incidences over 1000 cases, that is why finally on 5th October it was declared the second lockdown here. As a second "state of Alarm" was declared on 25th October for 6 months [5], the first objective of the present work is the need to provide data on the first lockdown, focusing toward the lockdowns in late 2020 and early 2021, since health experts predict a third wave after Christmas holidays in terms of the COVID-19 disease and other health effects. For instance, some authors demonstrated a 24%-35% reduction before and after lockdown in hospital admissions for other health effects, such as acute myocardial infarctions in France [6]. Besides, according health experts, it is expected.

The number of infections is different for different cities, so in addition to the determinant factors, such as the number of inhabitants and population density, it is necessary to know if other factors can influence the incidence of the disease. So, the second objective is the search of possible relationships between the SARS-CoV-2 and some air pollutant or meteorological variable. For this aim we will quantify by means of graphical and statistical tools the variations produced by the lockdown on atmospheric composition and the relationships of air pollutants and meteorological factors with the COVID infection cases and deaths.

Methodology

The region of study in Spain. Characteristics of the Southern Europe

The Southern Europe studied was the Region of Andalusia (8.4 million inhab., 87,268 Km^2) and with a particular emphasis on the cities of Sevilla, Barcelona and Madrid. Andalusia can be differentiated climatically into two areas by the Strait of Gibraltar. The Western zone of the Atlantic Ocean includes the provinces of Huelva https://goo.gl/maps/ooQL5u1XgTK2nwQ48, Cadiz https://goo.gl/maps/f8q3TrfRR2ApQFrd9, Sevilla https://goo.gl/maps/g9juFtEyEh1ucBZR7 and Cordoba https://goo.gl/maps/5sVSpxdwGfebPJQH7, and the Eastern area of the Mediterranean Sea includes Jaen https://goo.gl/maps/Y12NQsXtXeC5ftTv5, Malaga https://goo.gl/maps/iShggpNQAbMX8tMy5, Granada https://goo.gl/maps/zeCvBPS6Y6jGHbWt5 and Almeria https://goo.gl/maps/Jjwk1wK152TtuGym7.

In the Western zone the Guadalquivir River Valley forms a triangle of about 15,000 Km^2 (Figure 1) through which the Atlantic winds enter from S-SW direction crossing the Valley. Winters are short and mild, so domestic heating systems are not a source of air pollution. There are numerous anticyclone events and frequent thermal inversions. Saharan dust intrusions are also carried toward the region. The Eastern part is characterised by four microclimatic subzones: two adjacent and very different areas, 'Sierra Nevada' (Granada) and the 'Costa del Sol' (Malaga), and two more arid and drier areas: Jaen, widely dedicated to the olive cultivar, and Almeria, with a desert climate.

The results obtained in the Region of Andalusia can be extrapolated to the rest of the Iberian Peninsula, to the Mediterranean countries of Southern Europe and to other European countries or the world.

Characteristics of the Air Monitoring network

The Andalusian Government has an air quality monitoring network with more than eighty stations distributed throughout the eight provinces of the region to measure air pollutants (Figure 1, Table 1). The corresponding measurements can be downloaded from the website of the Department of Environment of the Government of Andalusia [7], however validated concentrations were provided by the same Department through official requests. The monitoring stations are classified in relation to the emission source (background, industrial, traffic) and the type of area (rural, urban, suburban). Table 1 shows the name of each station, the city council and the province where they are located, as well as the type of area and emission source, mentioned above. Data of the region of Madrid and its capital was provided by the Department of Environment of the Community of Madrid from its Air Quality Network [8]. The data of the city of Barcelona was provided by the Department of Territory and Sustainability of the Government of Catalonia from its Air Pollution Monitoring and Forecasting Network [9].

Air pollutants, meteorological factors, traffic densities and COVID data

In this work we have processed concentrations of gas pollutants, NO_2, CO, SO_2, H_2S and O_3 and suspended particles, PM_{10} and $PM_{2.5}$. In order to study whether there was significant variation in the magnitude of these vectors caused by the lockdown period, daily medians were gathered from all stations for four months, from January 1st to April 30th 2020. In parallel, daily records of meteorological parameters for the same period were obtained from the Spanish Meteorological Agency [10]. They were wind speed (WS), rainfall (RF), air temperature (AT), atmospheric pressure (AP) and air humidity (AH).

The first consequence of a lockdown is a drastic reduction in vehicle traffic through the cities and roads. In the first days of the lockdown, some press [11], blogs [12, 13] and preprints [14, 15] reported that the restrictions of economic activities and population movements resulted in a decrease in the level of air pollution. Thus, in order to study the influence of the lockdown on the vehicle transport, the data of traffic densities were available in the largest city in Andalusia, the city of Sevilla (37º23'N, 5º58'W, 141.3 Km^2, 7 m a.s.l.), the most representative city in terms of air pollution by urban traffic. Daily data of thirty entry/exit routes from the city were provided by the Mobility Management Centre [16], the traffic control centre of the City of Sevilla.

The "state of alarm" did not stop the progress of the number of daily virus infections and deaths until the end of April 2020. The mechanism of the virus transmission is not yet fully controlled, but it is also not clear if there are other factors that may contribute to the incidence of the disease. For this reason, our study investigated the relationships between meteorological and pollutants data with the number of daily infections and deaths [17] caused by the SARS-CoV-2.

Positive-negative effects of lockdown. Strategy for the cause-effect patterns recognition

The methodology was divided into three stages: the first consisted of detecting the change of slope from 14th March in each concentration-time plot. Then differences before-after 14th March were estimated and quantified in percentage. Thirdly it was performed a study on correlations including meteorological variables, air pollutants, traffic densities and data of infection cases and deaths. The Pearson high coefficients were validated confirming with its graphic profiles [18].

Alternatively we could evaluate the lockdown effects by comparing the 2020 data with those of previous years, such as the 2019 year; however it is not clear that these comparisons truly reflected the impact of lockdown on environmental concentrations as was declared by Jia et al. [19]. In consequence we have considered more effective to compare two periods: a) A pre-lockdown period, 1st January/13th March and b) a during-lockdown period, 14th March/30th April.

Results And Discussion

Air pollutants in pre-lockdown and during-lockdown periods. Health consequences

We have evaluated the behaviour of NO_2, CO, SO_2, H_2S, O3, PM10 and PM2.5 along the four months of the study at each station of the network to determinate graphically how the lockdown has influenced the air quality. Besides, we have calculated the change in slope and quantified the percentage of variation of the concentrations before and after 14th March according with the type of area and source.

Nitrogen dioxide and carbon monoxide

Nitrogen dioxide is the most clearly reduced its concentration (global mean of -53.7%) in all stations [20]. Only three nearby industrial areas increased the level. NO_2 is the typical pollutant emitted by car exhausts and energy production, in both NO and NO_2 species, i.e. NO_x [21]. Carbon monoxide is originated from similar sources [22] and its concentrations down by -27.0%.

For the whole region, starting from pre-lockdown levels (1st January – 13th March) the highest decreases produced from 14th March were obtained in stations of the province of Granada (-60.4% from 28.5 μg m^{-3}), Sevilla (-62.4% from 18.9 μg m^{-3}), Malaga (-65.0% from 21.4 μg m^{-3}) and Jaen (-67.7% from 15.8 μg m^{-3}), indicating that the highest reductions were observed for the highest pre-lockdown values. These changes are clearly shown by the SENTINEL-5P satellite measurements of tropospheric NO_2 concentrations (μmol m^{-2}) when comparing both periods (Figure 2, top): pre-lockdown values were around 50 μmol m^{-2} and much less than 45 μmol m^{-2} during lockdown. Observing the concentration-time plots for the most populated city, Sevilla, most stations have a decreasing trend of NO_2 (Figure 3a), being reduction rates from -55.4% (Torneo) to -72.0% (Alcala), with rapid trafficked roads. Similar results were found in the Lombardia Region where only in a heavy traffic zone NO_2 showed a significant decrease [23]. The rest of urban and suburban stations decreased over 60.4%. rural stations the percentages were -24.2% in Gerena and -37.9% in Sierra Norte, both located more than 50 km from the city, so the rate of reduction decreases the more distance from the big city. The behaviour of NO_2 in the rest of cities is shown in Figure 3b. Some representative reduction rates were -66.1% (in Pozo Dulce, the centre of Huelva), -68.4% (in Marbella, a city of Malaga Province) and -81.3% (in San Fernando, a city of Cadiz Province), having all of them high traffic densities and nearby industrial activities (Huelva and Cadiz), as it was also reported by other studies [24, 25, 26].

Due to the direct relationship between the concentration of NO_2 in the air and hospital admissions for cardiovascular disease [27], the health consequences of the NO_2 reduction were extremely beneficial, such as the lower number of acute hospital admissions for cardiovascular disease in the short-term (acute ischemic syndromes, atrial fibrillation, and decompensated heart failure [28], and possibly a lower cardiovascular mortality due to environmental causes in the long run.

Regarding carbon monoxide, stations decreased its concentration after lockdown and others increased it, as it was reported in USA [19], Italy [29] and by interdisciplinary studies developed for 40 cities [30]. The highest decreases starting from the pre-lockdown values were observed in stations of Sevilla (-45.0% from 477 μg m^{-3}), Jaen (-48.7% from 432 μg m^{-3}), Cadiz (-52.6% from 522 μg m^{-3}) and Huelva (-58.2% from 705 μg m^{-3}), with values two times higher than in Cordoba (-32.7% from 370 μg m^{-3}) and Granada (-33.0% from 363 μg m^{-3}). Pre-lockdown levels in the period 1st January-13 March 2020 were similar to the 2019 values for the same stations and period. Thus, the highest decreases were also related to high pre-lockdown concentrations, as in the case of NO_2. In conclusion, for both NO_2 and CO the highest reduction tases were in stations where pre-lockdown levels are elevated. These stations were near to the two most important industrial areas, the Chemical Park of Huelva, and the Campo de Gibraltar of Cadiz, but also in cities with high rate of vehicles circulation, the province of Sevilla. These results are observed by the SENTINEL-5P satellite measurements of CO concentrations when comparing both periods (Figure 2, bottom): pre-lockdown values were between 106 and 129 μmol m^{-2}, and between 60 and 83 μmol m^{-2} during lockdown.

In the case of carbon monoxide the health consequences of the CO reduction are not clear because no direct relation between CO at low-medium concentrations and hospital admissions are usually found by researchers [31]. Some authors found direct association with negative effects of short-term CO exposure only over 2,000 μg m^{-3} [32].

The reduction rates of CO in Sevilla were from -24.8% in Bermejales (-63.1% NO_2) to -50.0% in Torneo (-55.4% NO_2)(Figure 3c). Curiously, within the lockdown period we have observed a small and unexpected period of increase inside the decreasing profile. This coincides with the Easter holiday period (6th April), which suggest the departures of private vehicles from the city despite of being forbidden for citizen. When the behaviour of CO in the rest of cities was decreasing (Figure 3d) the stations with the most reduction rates had high traffic densities, such as Pozo Dulce (in

Huelva, -70.8% CO, -66.1% NO_2), Jerez (in Cadiz, -83.4% CO, -71.0% NO_2), Lepanto (in Cordoba, -64.4% CO, -79.9% NO_2), Fuentezuelas (in Jaen, -52.0% CO, -68.6% NO_2) and Congresos (in Granada, -56.1% CO, -48.7% NO_2).

On the other hand, if a lockdown consists of a total braking of all vital activities, an increase in CO should indicate an increase in 'essential' economic activities that cannot be stopped. In the province of Sevilla, the increases corresponded to stations located near major industrial areas (see above google maps links): Gerena (+35.8% CO, -24.2% NO_2) is next to a big copper mine. Similarly, three stations close to each other, Ranilla (+25.0% CO, -69.2% NO_2), Alcala (+16.5% CO, -71.8% NO_2) and Dos Hermanas (+19.4% CO, -71.4% NO_2) receive the influence of a big stainless steel factory, two detergent plants and a cement factory. In addition, the CO profile presented a continuous increase in the form of 'sawtooth cycles' (Figures 3e and 3f). This type of variation was also observed for aerosol particles [33] and for particles and nitrogen dioxide. In our case, observing these cycles we found that they occured coinciding with rainfall episodes as it can see in Figure 3e.

In the rest of cities the increases of CO corresponded to stations Joya, Cartuja, Barrios and Marconi (Figure 3f). Joya (Almeria, +43.2% CO, +16.6% NO_2) and Barrios (Cadiz, +27.7% CO, -37.0% NO_2) are next to coal thermal power plants, and Barrios is near the first major stainless steel factory of Andalusia (see above google maps links). Cartuja (in Jerez de la Frontera, Cadiz, +26.6% CO, -79.6% NO_2) is next to the industrial zone of the city and Marconi (Cadiz, +25.1% CO, -85.0% NO_2) is next to the 'Zona Franca' industrial area and the 'Bay of Cadiz' Port Area. Consequently, differences between decreases and increases of carbon monoxide concentrations were caused by the increasing activity in power generating industries and others essential industries during the lockdown period.

Sulfur dioxide and hydrogen sulfide

Sulfur dioxide emissions come mainly from industrial activities [34], however no clear trend was observed in the evolution of SO_2, as it was reported in Barcelona (Spain) [35]. Regarding H_2S, emissions can come from incomplete combustions and from wastewater [36]. H_2S emissions increase on lockdown in practically all stations where the gas sensor is installed. In the majority of stations no significant variations were observed for SO_2, except in some places with extreme values (Figure 3h), such as in Ejido (Almeria), in Moguer (Huelva), in Obejo and Villaharta (Cordoba) and in Gerena (Sevilla).

Ejido (+190.5% in SO_2, -79.0% in NO_2) has no relevant industries, but it contains the largest extension of plastic greenhouses in Spain ('sea of plastic'), and H_2S also increased +103.2%. Greenhouse plastics are usually degraded by elemental sulfur, which is used to disinfect greenhouses, which is why commercial plastics for greenhouses are manufactured protected against sulfur. So, these extreme values are due to burning of used plastics mixed with organic residues. Also, in Moguer, next to the petrol refinery in Huelva Chemical Park, an increase of +84.7% in SO_2 was observed (-56.2% in NO_2), similar to those reported in UK [37, 38]. Obejo and Villaharta are near a coal thermal power plant, and the values increased too much for SO_2 (+82.6% and +92.3%) and for H_2S (+84.3% and +32.5%) after 14th March. Villaharta also receive NO_2 emissions (+34.7%) probably from the chimney of the power plant (see above google maps link). Regarding Gerena, the mine zone showed an increase of +67.9% in SO_2 in comparison with a decrease of -24.2% in NO_2. All these extreme increases in SO_2 levels must be attributed to the increase in burning of coal [39] or other fossil fuels by thermal power plants [40] or other industrial activities, such as mines or refineries. According Wang et al. [25], concentrations of SO_2 and other pollutants such as CO and NO_2 appeared to have positive effect on hospitalizations, especially at high concentrations, for instance by bronchiectasis. Also other authors [41] reported that only sulfur dioxide was associated with out-patient visits. Associations of SO_2 and CO with pneumonia were reported [42]. Consequently, it would be advisable to observe especially these unusual patterns to alert the population, although they are less frequent than the general decreases observed for the studied air pollutants.

Regarding decreases of sulfur dioxide concentrations Figure 3g shows higher differences between levels before and after 14th March. Stations such as Ranilla (Sevilla, -46.0% SO_2, -69.2% NO_2), Algeciras (Campo de Gibraltar, -51.7% SO_2, -21.3% NO_2) and Rosales (Huelva, -55.4% SO_2, -63% NO_2) were located in residential places of industrial areas, suggesting the stopping of private and economic activities. Similar results were obtained in Milan (Italy) [28].

Ozone and Particulate matter (PM_{10} and $PM_{2.5}$)

In 2018, 1,206 exceedances of threshold limits [43] were reported, which 979 were due to tropospheric ozone, 224 to particulate matter, 3 to nitrogen dioxide and none to sulphur dioxide.

<u>Ozone</u>

The atmospheric conditions most suitable for achieving high levels of this secondary pollutant are the result of the reactions of nitrogen oxides (NO_x) with carbon monoxide, methane and non-methane volatile organic compounds (NMVOCs), i.e. their precursors, under high solar radiation and temperatures [44]. Excessive ozone in the air can have a marked effect on human health. It can cause breathing problems, trigger asthma, reduce lung function and cause lung diseases [45].

In connection with our study, we observed that the concentration of ozone increased during the lockdown in all monitoring stations. In this sense, the increase was related to the fall of NO_2 and CO, because of O_3 concentrations were negatively correlated with NO_2, CO and also $PM_{2.5}$ and PM_{10}. The increase is attributed in almost all studies to the decrease of NO_2 (NO_x or NO) and VOCs [46]. The highest increases occur in traffic stations where the pre-lockdown levels of ozone are low. It is certainly not normal that whilst in most stations nitrogen oxides decreased, they increased in parallel ozone levels. Therefore we propose an explanation: if under certain conditions the reduction of NO_2 emissions can involve an increase in ozone levels, a process well known as 'Ozone Weekend Effect (OWE)' [47, 48], we asked ourselves, Could the weekend effect explains similarly the increase in ozone during lockdown? Our hypothesis affirm: "*despite the decrease in the levels of its main precursors, NO_2 and CO, the increase in ozone levels during lockdown is explained by an effect that we will be calling the 'Ozone Lockdown Effect (OLE)', similar to the 'weekend effect' but longer in time*". Other similar study developed in four cities from Southern Europe, including Valencia (Spain), also reported a similar concept, the 'lockdown effect on O_3 production' as the increase of ozone during lockdown for a longer period [49].

In terms of health effects, some authors declare that no significant relationship between tropospheric ozone and hospital admissions was found for respiratory diseases [50]. However, the majority of studies found associations with respiratory and cardiovascular affections [27, 51, 52], such as pneumonia and COPD, chronic obstructive pulmonary disease [53].

If we make a joint study for CO, NO_2, O_3 before/after 14th March in Sevilla, the levels of NO_2 were reduced by up to -19 μg m^{-3} in Centro (historical Centre of Sevilla), Torneo and Ranilla, while the maximum reduction of CO reached -401 μg m^{-3} in Torneo, Ranilla and also Santa Clara (in the border of Sevilla). As ozone was negatively correlated with NO_2 and CO (Figures 4a and 4b) the increases of O_3 of up to 40 μg m^{-3} (Figure 5a) occurred when pre-lockdown levels of ozone were significantly low, such as in Torneo (+41.2 μg m^{-3}, initially from 10.8 μg m^{-3}), Santa Clara (+40.1 μg m^{-3}, initially from 8.2 μg m^{-3}) and Centro (+36.2 μg m^{-3}, initially from 13.3 μg m^{-3}), being all of areas with usual high vehicle traffic, which emit the ozone precursors NO_x, CO and also VOCs [54]. In areas where pre-lockdown levels of O_3 are significantly high, the Ozone Lockdown Effect is not so marked, and the increases were significantly low (Figure 5a), as it can see in Sierra Norte (+19.1 μg m^{-3}, initially from 35.1 μg m^{-3}), Gerena (+15.1 μg m^{-3}, initially from 26.0 μg m^{-3}) and Alcala (+24.8 μg m^{-3}, initially from 33.3 μg m^{-3}), being all far from the city with low traffic in background or rural areas, some of these near industries (Gerena). Furthermore, it would be interesting to study if the ozone lockdown effect is also visible in provinces with cities many industrialised, that is, in Huelva and Cadiz.

So, in these cases, the highest ozone increases were also related with low pre-lockdown values, being the most representative stations Punta Umbria (a coastal city near Huelva, +32.1 μg m^{-3}, initially from 25.1 μg m^{-3}) and Matalascañas (a coastal city near Huelva, +31.3 μg m^{-3}, initially from 28.4 μg m^{-3}), both in Huelva province, and Marconi (Figure 5b, +43.2 μg m^{-3}, initially from 34.1 μg m^{-3}) and Jerez (+33.0 μg m^{-3}, initially from 29.1 μg m^{-3}) in Cadiz province. These monitoring stations are located in very touristic coastal cities, very transited before the lockdown. On the opposite side, the 'ozone lockdown effect' is not so remarkable in stations with high pre-lockdown levels, such as Rabida (+10.1 μg m^{-3}, initially from 41.2 μg m^{-3}) and Moguer (+18.2 μg m^{-3}, initially from 40.1 μg m^{-3}) near the Chemical Park of Huelva, and Arcos (+13.0 μg m^{-3}, initially from 69.9 μg m^{-3}) and Campamento (Figure 5b, in the 'Campo de Gibraltar', +16.2 μg m^{-3}, initially from 78.9 μg m^{-3}) in Cadiz. Thus, ozone formation is more important in trafficked areas [52] where the reduction of road traffic is more significant than in those areas where the reduction occurs near industrial activities.

These results confirm our hypothesis and thus, despite the decrease of NO_2 and CO, the increase of tropospheric ozone during the lockdown constitutes the 'ozone lockdown effect, OLE'. In addition, in the 4-month period studied, the 'ozone weekend effect, OWE' was observed in most of the weekly periods of all stations.

PM_{10} and $PM_{2.5}$

As mentioned above, the poor air quality was mainly due to particles and ozone. Nowadays, particulate matter is a primary pollutant that has a negative correlation with ozone [45] because of aerosols can have a radiation effect through both absorption and scattering [55, 56]. Furthermore, other authors propose that aerosols reduced the actinic flux photolysis rates under absorption and scattering, so the surface ozone production decreases [57, 58]. Li et al. [59] demonstrated that O_3 was strongly inhibited under the condition of high $PM_{2.5}$ concentration. In addition, as in the case of CO and NO_2, our correlation study shows also high negative correlation coefficient between O_3 and PM_{10} $PM_{2.5}$ (Figures 4c and 4d).

In the province of Sevilla, particle concentrations decreased in all the areas, particularly in stations located inside the city, Torneo and Principes, with higher PM_{10} and $PM_{2.5}$ decreases than stations far from the city (Figure 5c), Gerena and Sierra Norte. The magnitude of the decrease depends on the pre-lockdown values, i.e., the greatest decreases occur in stations with higher pre-lockdown concentrations (23 - 24 μg m^{-3}), all of them trafficked stations. Additionally, if we compare between concentrations of PM_{10} and $PM_{2.5}$, the result shows that the decrease in $PM_{2.5}$ levels (-57%) in lockdown were higher than in PM_{10} (-42.0%), suggesting and corroborating the much more anthropogenic contribution (Figure 5d).

In Cadiz (Figure 5e) and Huelva (Figure 5f), levels of PM decrease and increase also. In Cadiz, the greatest decreases in PM_{10} levels occur in urban stations with a high traffic, such as Jerez (-42.5%) and Marconi (-28.1%). Rural and background areas, such as the Alcornocales National Park, and the 'Arcos de la Frontera' city, show low decreases, -7.3%, while in industrial areas PM_{10} seems to decline very lightly, such as in Rinconcillo (+5%), Linea and Palmones (-5%) or even rising in Zabal (+14%), all them near a big stainless steel factory and a big petrol refinery of the Bay of Algeciras. In Huelva, the same perceptions were observed. The greatest decreases in PM_{10} occur within the city of Huelva, where the most intense urban traffic was stopped at 14th March. Titan (-32%), Rosales (-29%), Pozo Dulce (-25%) and Carmen (the University Campus, -21%) were the representative stations, whereas inside industrial areas, Palos and Moguer (-14%), San Juan and Rabida (-11%, -9%) and the coastal areas, Punta Umbria (-10%), Mazagon (0%) and Matalascañas (+8%) had lower decreases or even increased the particle concentrations.

In conclusion, these results in Sevilla, Cadiz and Huelva reinforce the idea that since ozone and $PM_{10}/PM_{2.5}$ are negatively correlated and traffic zones suffer the greatest reductions, we must consider PMs as further variables involved in the 'ozone lockdown effect OLE' like CO and NO_2. Some authors found that during lockdown the ozone concentrations were higher than in normal situations, being this related with the increase of organic carbon and decrease of elemental carbon in aerosols [60].

Nevertheless, if ozone is the most irritating gas pollutant, it may be assumed that its presence could influence the incidence of the COVID-19 disease. Therefore, can it be assumed that there is an important relationship between airborne ozone levels and the number of COVID-19 infections or deaths?

Correlation study for air pollutants: relationship with the incidence of COVID-19. Meteorological and traffic implications

As mentioned above, correlation studies were done between atmospheric and meteorological variables, traffic densities and also with number of infections and deaths due to *COVID*-19 disease. The study was done for all representative locations in the region. In the case of traffic densities the study was cantered in Sevilla, the most trafficked city.

Ozone and number of infections and deaths by COVID-19

On the other hand, we have detected an interesting relation between daily infections/daily deaths and ozone concentration. The correlation coefficient between ozone and number of infections was r = + 0.71 (Figure 6a): when ozone is high the number of infections is also elevated and when ozone decreases the infection cases also was reduced.

In addition, Figure 6b shows how infections cases in Sevilla increased 1-2 days after the ozone concentration rise. The possibility of a coincidental parallel increase of ozone and infection cases is not plausible because the profile also shows a period from 1st April where decreasing infections is preceded by decreasing ozone. In order to check this behaviour in other important cities outside the Southern Spain, we also studied this relationship in the cities of Madrid and Barcelona (Figures 6c and 6d), corroborating the behaviour observed in Sevilla: in graphics b), c) and d), every peak/valley of the infections black lines (INF) is preceded by a peak/valley of the ozone red lines (O_3) (see grey lines), even in the both rising and decreasing periods. This fact confirms our hypothesis, which states that the presence of high levels of ozone is a determinant factor on the incidence of COVID-19 disease, further reducing the infection rate when the ozone level decreases. This behaviour constitutes an *effect that we will be calling the 'Ozone COVIDEffect (OCE)'.*

The number of deaths was also correlated with the ozone level (r = + 0.59), so that, despite the high rate of person-to-person transmission of the new COVID-19 virus, the high oxidizing character of ozone constitutes the first ambient factor that enhances the incidence of the COVID-19. As a result, we propose a mechanism for infected people, in which the ozone entering into the lungs accelerates the occurrence of the virus symptoms, even in healthy people, increasing the seriousness of the disease.

Air pollutants and meteorological parameters

Firstly, regarding Pearson correlations studied (Table 2), we confirmed the well known relationships along air pollutants, such as mentioned above for ozone, O_3 with NO_2, O_3 with CO, ozone with PM_{10}, ozone with $PM_{2.5}$ and for nitrogen dioxide, NO_2 with CO, NO_2 with PM_{10}, NO_2 with $PM_{2.5}$ or CO with PM_{10}. No significant correlations were found for SO_2 ($r < \pm 0.32$). Other well known correlations were also found between meteorological parameters and air pollutants, for instance those for atmospheric pressure (AP), such as AP with O_3 (Figure 7a), AP with NO_2, AP with CO or AP with PM_{10}, those for wind speed (WS), such as WS with O_3, WS with NO_2 or WS with CO (Figure 7b), or those for air temperature (AT), such as AT with O_3 or AT with NO_2 (Figure 7c).

Correlations with rain should be performed in a different way: a significant value of precipitation (RF > 0.4 mm) implies an air-cleaning effect the next day, which is why concentrations of one day need to be correlated with that of the following. So, correlations found (Table 2) for the rainfall (RF) were for CO, RF with SO_2, RF with NO_2 and RF with PM_{10} (Figure 7d), but no correlation was found for ozone. This way, these correlations indicate a gas/particles-cleaning effect, being possible a dissolving effect in the raindrops and thus obtaining rainwater with nitrate or sulphate in solution [61]. Thus, we have seen above that increases of CO in sawtooth form of Figure 3e were related to the correlation with rain events (Table

2). A particle-cleaning effect is also produced by rain events [62]. Regarding infection cases and deaths, atmospheric pressure showed negative correlations with cases of infections and deaths, AP with INF (r = - 0.88), AP with DEA (r = - 0.71), and positives with temperature, AT with INF (r = + 0.73), AP with DEA (r = + 0.51).

Air pollutants and traffic of vehicles

The correlation study also included the vehicular traffic densities (VTDs) in the city of Sevilla, which should be correlated with air pollutants. Thirty-three street and roads routes were included. Since the main consequence of the lockdown was the reduction of traffic densities, the correlation study compared Pearson coefficients before 14th March and after 13th March, as was studied by Hashim et al. [24].

The results showed that Pearson coefficient for VTD/NO_2 was higher before the lockdown than after it (r = + 0.72 < 14th March, r = + 0.59 > 13th March, NO_2, Figures 7e, 7f) but for ozone it was lower before (r = - 0.47 < 14th March, r = - 0.63 > 13th March, O_3, Figures 7g, 7h). This fact confirms the regular and major transit of cars into pre-lockdown, and during lockdown, the much minor traffic and the ozone lockdown effect. What is more, the result is different in the case of carbon monoxide, where Pearson coefficient for VTD/CO is positive (r = + 0.53 < 14th March) within pre-lockdown, as NO_2, but negative and higher (r = - 0.62 > 13th March) into lockdown, as O_3, confirming the rise of road transport along industrial areas. No correlation was found for SO_2.

Conclusions

According to the results obtained, we have succeeded to describe the effects of COVID-19 lockdown on the change of atmospheric composition. In general, we have demonstrated the clear decrease in carbon monoxide, particulate matter and nitrogen dioxide due to the traffic of vehicles. On the other hand, we detected the cases in which carbon monoxide and sulfur dioxide increased concentrations in stations located near large industrial areas, such as stainless steel or thermal power plants. Besides we have concluded that some extremely high values of sulfur dioxide and hydrogen sulfide observed were caused by the frequent burning activities of greenhouse plastics. Studying the whole of region, it is concluded that there are no differences by geographical factors, only by type of emission. Additionally, the major reduction rates in air pollutants occurred, in general, when the pre-lockdown concentrations were high.

In particular, the most interesting conclusions of this study were related to ozone and airborne particles. Regarding ozone, which is a secondary pollutant negatively correlated with the rest of pollutants, the increase in concentration during the lockdown is attributed to the 'ozone lockdown effect' OLE, similar to the 'ozone weekend effect' OWE, but over the longer term. With regard to airborne particles, PM_{10} and $PM_{2.5}$, which were also negatively correlated with ozone, the reduction of levels in areas strongly influenced by traffic contributes significantly to the 'ozone lockdown effect' as CO and NO_2. Regarding traffic intensities (VTDs), we have confirmed the evidence of the ozone lockdown effect through the fact that the O_3/VTDs correlations were higher during the lockdown than in the pre-lockdown, in contrast to the VTDs/NO_2 correlations. As we studied more than 80 monitoring stations in the Southern Spain, it is expected that the results and conclusions can be extrapolated to the rest of the country and probably to other Mediterranean countries, as we already demonstrated the same ozone OCE effect in Sevilla, Barcelona and Madrid.

Finally, as a result of the high positive correlation found between ozone and the number of COVID-19 cases, which was graphically confirmed, we can affirm that the presence of significant concentrations of tropospheric ozone in the breathing air constitutes a critical factor on the major incidence of the disease. This effect was confirmed on the cities of Madrid and Barcelona with the same results of Sevilla. Consequently, we can stated that ozone is those air pollutant that more health risk present for the population in relation to harmfull respiratory effects and the SARS-CoV-2 disease.

Declarations

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.

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Availability of data and materials

Data will be available if required.

Consent for publication

Consent for publication was obtained from each participant.

Competing interests

There are no competing interests.

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Tables

Table 1 List of the air monitoring stations studied in Southern Spain from the total of 83 stations assessed

Province	Stations	Name	City Council	Emission source	Type or area
Almeria	2/12	Joya	Nijar	Industrial	Rural
		Ejido	El Ejido	Background	Urban
Cadiz	13/23	Campamento	San Roque	Industrial	Suburban
		Barrios, Palmones	Los Barrios	Industrial	Suburban
		Cartuja	Jerez de la Frontera	Industrial	Suburban
		Algeciras, Rinconcillo	Algeciras	Industrial	Urban
		Linea, Zabal	La Linea de la Concepción	Industrial	Urban
		Marconi	Cadiz	Traffic	Urban
		Jerez	Jerez de la Frontera	Traffic	Urban
		San Fernando	San Fernando	Traffic	Suburban
		Arcos	Arcos de la Frontera	Background	Urban
		Alcornocales	Los Barrios	Background	Rural
Cordoba	3/6	Villaharta	Villaharta	Industrial	Suburban
		Lepanto	Cordoba	Traffic	Urban
		Obejo	Obejo	Industrial	Suburban
Granada	1/4	Congresos	Granada	Traffic	Urban
Huelva	11/16	Pozo Dulce, Rosales, Carmen	Huelva	Traffic	Urban
		Moguer, Mazagon	Moguer	Industrial	Suburban
		Rabida, Palos	Palos de la Frontera	Industrial	Suburban
		Punta Umbria	Punta Umbria	Industrial	Urban
		Matalascañas	Matalascañas	Background	Urban
		San Juan	San Juan del Puerto	Industrial	Urban
		Titan	Huelva	Industrial	Urban
Jaen	2/4	Fuentezuelas	Jaen	Traffic	Suburban
		Valle	Jaen	Background	Urban
Malaga	1/6	Marbella	Marbella	Traffic	Suburban
Sevilla	11/12	Torneo, Bermejales, Ranilla, Centro, Santa Clara, Príncipes	Sevilla	Traffic	Urban
		Dos Hermanas	Dos Hermanas	Traffic	Suburban
		Mairena	Mairena del Aljarafe	Background	Suburban
		Alcala	Alcala de Guadaira	Industrial	Suburban
		Gerena	Gerena	Industrial	Rural
		Sierra Norte	San Nicolas del Puerto	Background	Rural
Total	44/83				

Table 2 Matrix of correlation for Pearson coefficients formed by ambient variables, meteorological variables, vehicular traffic densities before (VTDb) and after (VTDa) lockdown, Covid-19 infection cases (INF) and Covid-19 death cases (DEA)

	O_3	NO_2	CO	SO_2	H_2S	PM_{10}	$PM_{2.5}$	AP	WS	AT	RF	AH	VTD_b	VTD_a	INF	DEA
O_3	1.00	-0.90	-0.81	0.11	0.12	-0.62	-0.63	-0.83	0.68	0.58	0.15	-0.51	-0.47	-0.63	0.71	0.59
NO_2	-0.90	1.00	0.74	0.21	0.09	0.62	0.73	0.74	-0.62	-0.59	-0.65	0.54	0.72	0.59	-0.58	-0.46
CO	-0.81	0.74	1.00	0.31	0.21	0.59	0.57	0.71	-0.66	-0.37	-0.72	-0.43	0.53	-0.62	-0.26	-0.19
SO_2	0.11	0.21	0.31	1.00	0.75	0.14	0.09	0.57	-0.48	-0.29	-0.70	-0.39	-0.15	-0.17	-0.30	-0.22
H_2S	0.12	0.09	0.21	0.75	1.00	0.11	0.13	0.47	-0.31	-0.22	-0.66	-0.43	-0.08	-0.05	-0.21	-0.14
PM_{10}	-0.62	0.62	0.59	0.14	0.11	1.00	0.88	0.65	-0.52	-0.53	-0.82	-0.57	0.64	0.48	-0.33	-0.12
$PM_{2.5}$	-0.63	0.73	0.57	0.09	0.13	0.88	1.00	0.48	-0.59	-0.58	-0.89	-0.62	0.67	0.44	-0.27	-0.09
AP	-0.83	0.74	0.71	0.57	0.47	0.65	0.48	1.00	-0.54	-0.71	-0.58	-0.17	—	—	-0.88	-0.71
WS	0.68	-0.62	-0.66	-0.48	-0.31	-0.52	-0.59	-0.54	1.00	0.38	0.36	-0.50	—	—	-0.38	-0.13
AT	0.58	-0.59	-0.37	-0.29	-0.22	-0.53	-0.58	-0.71	0.38	1.00	-0.36	-0.46	—	—	0.73	0.51
RF	0.15	-0.65	-0.72	-0.70	-0.66	-0.82	-0.89	-0.58	0.36	-0.36	1.00	0.86	—	—	0.41	0.28
AH	-0.51	0.54	-0.43	-0.39	-0.43	-0.57	-0.62	-0.17	-0.50	-0.46	0.86	1.00	—	—	0.36	0.21
VTD_b	-0.49	0.72	0.53	-0.15	-0.08	0.64	0.67	—	—	—	—	—	1.00	—	—	—
VTD_a	-0.63	0.59	-0.62	-0.17	-0.05	0.48	0.44	—	—	—	—	—	—	1.00	—	—
INF	0.71	-0.58	-0.26	-0.30	-0.21	-0.33	-0.27	-0.88	-0.38	0.73	0.41	0.36	—	—	1.00	0.63
DEA	0.59	-0.46	-0.19	-0.22	-0.14	-0.12	-0.09	-0.71	-0.13	0.51	0.28	0.21	—	—	0.63	1.00

—: correlation not applicable or not relevant

Figures

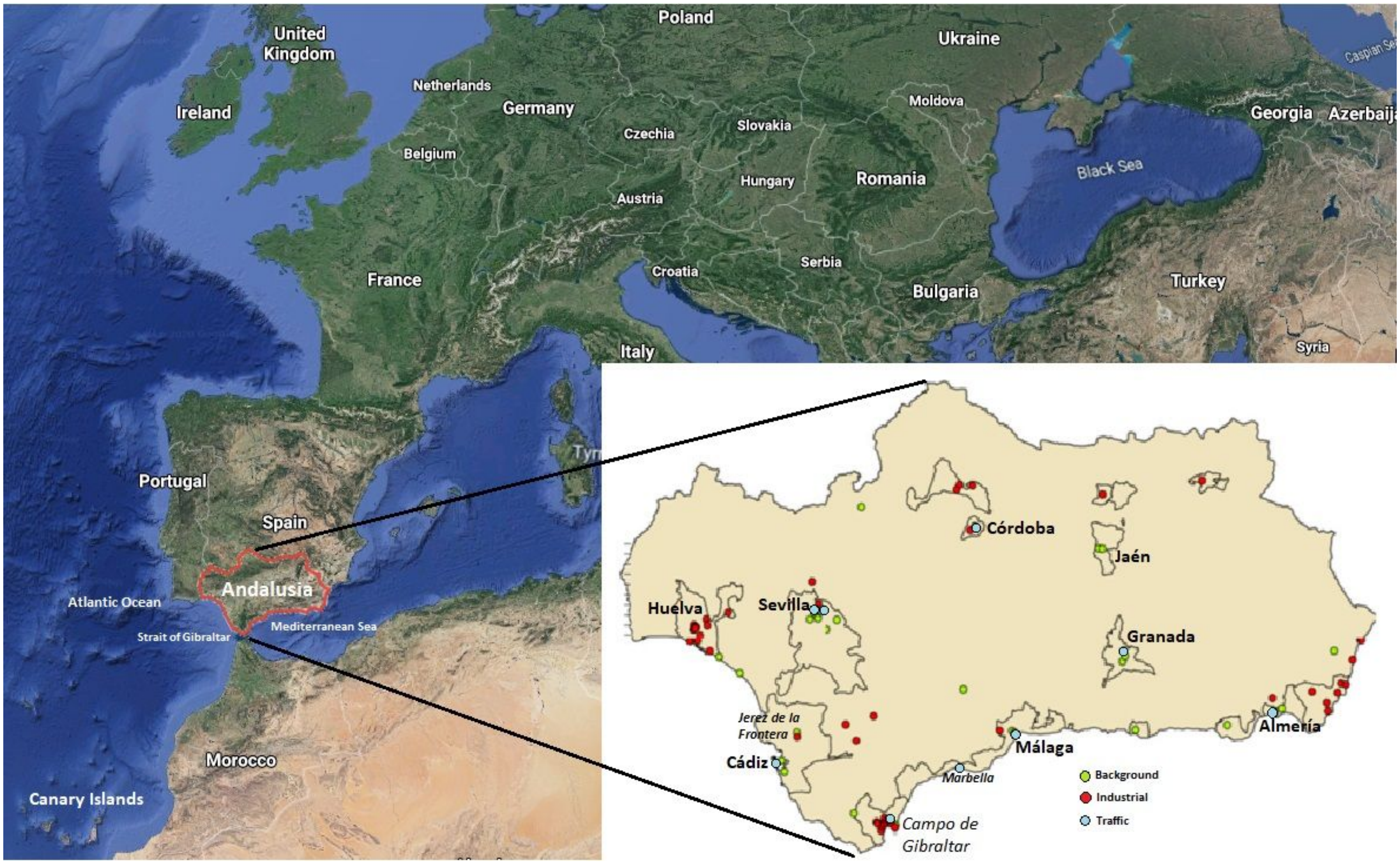
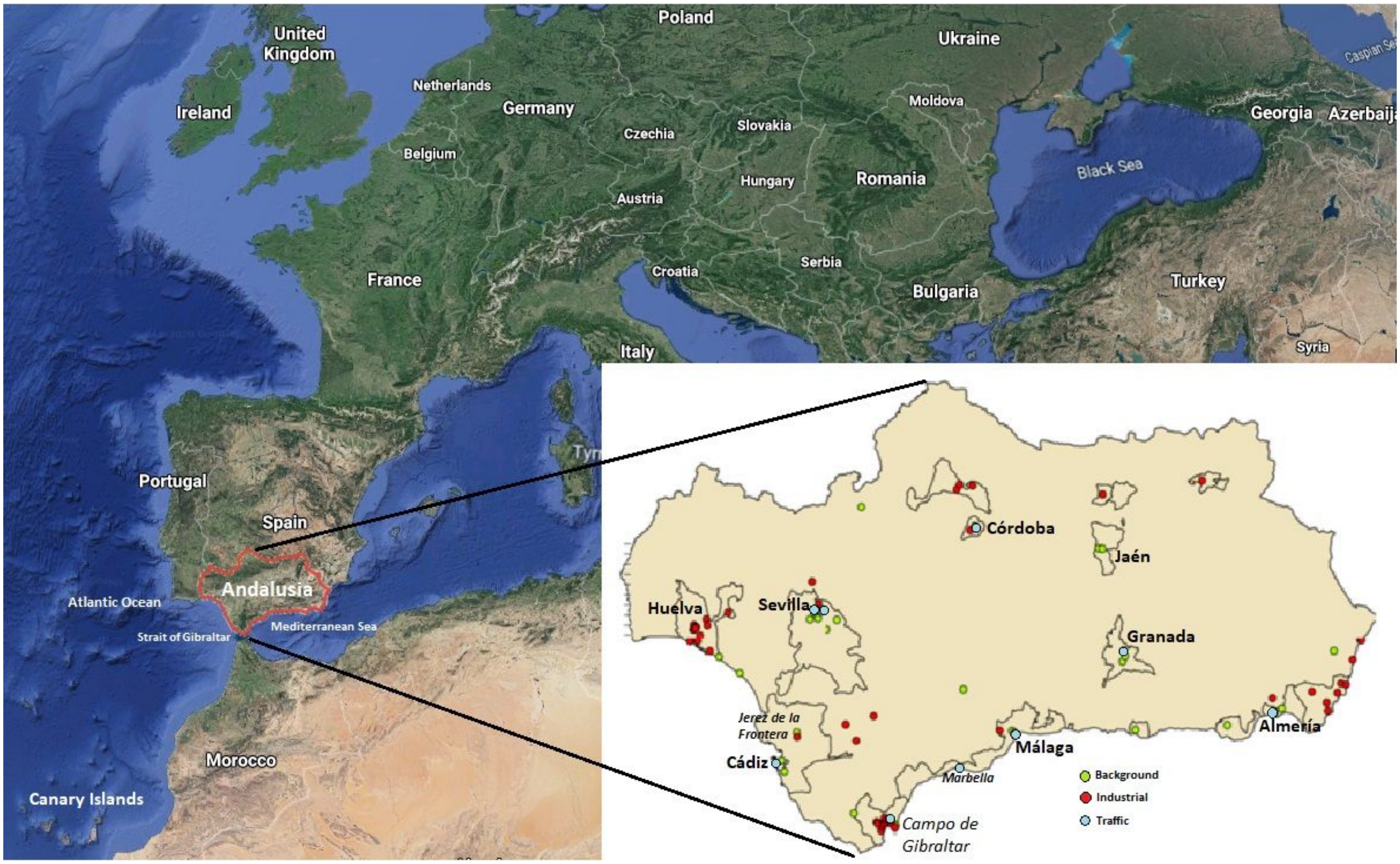


Figure 1

Region of Southern Spain studied with the eight provinces and cities. Spatial distribution of the air monitoring stations. (red contour, https://earth.google.com/web/@37.57746982,-4.79866842,394.48234909a,723551.45849437d,35y,0h,0t,0r) Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.



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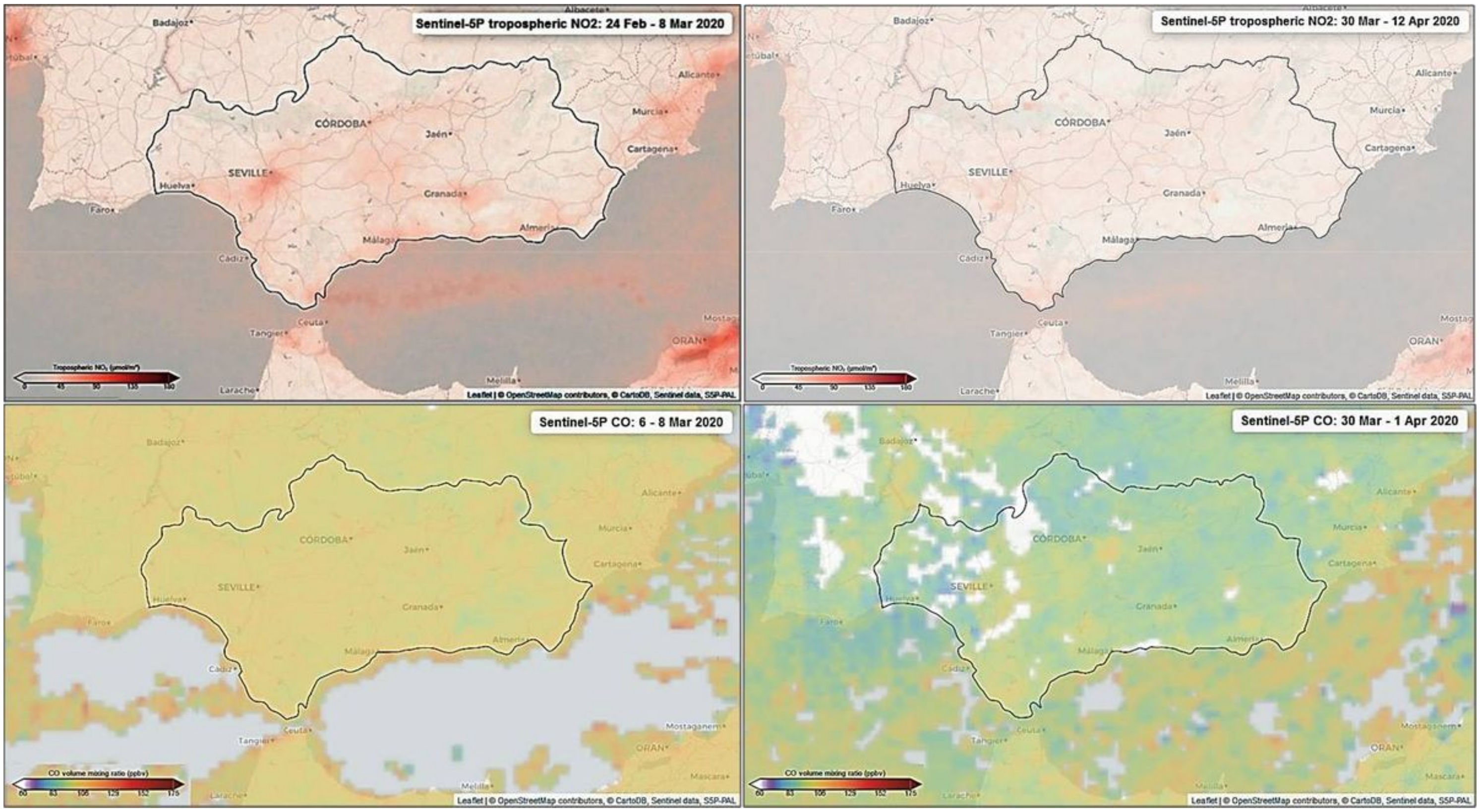
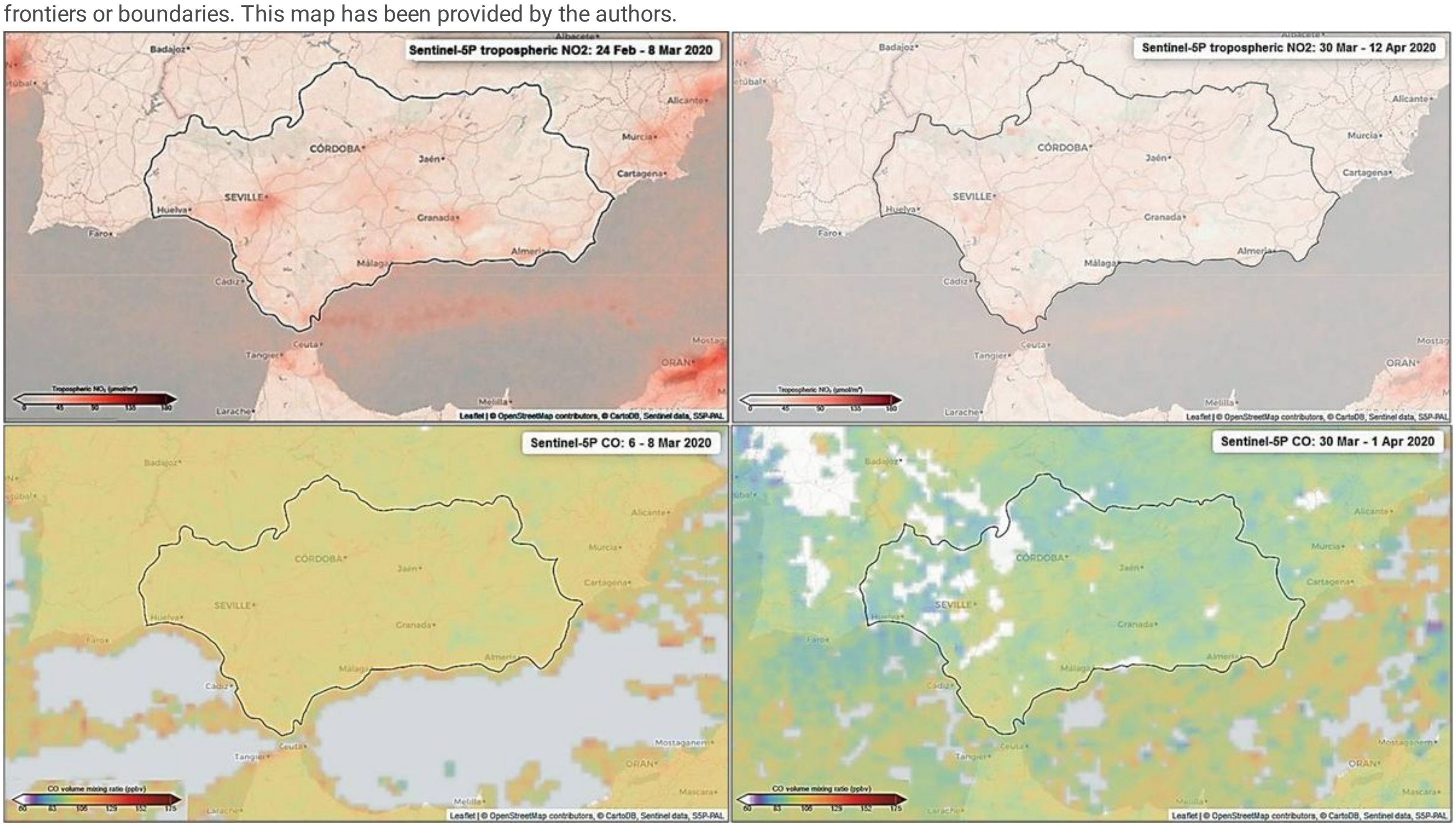


Figure 2

Concentrations in µmol m-2 of tropospheric NO2 averaged for 14 days and CO for 3 days by the Sentinel-5P satellite supplied by COPERNICUS-ESA in the Southern Spain (Andalusia region). Top maps for NO2, bottom maps for CO; left maps before 14th March, right maps after 13th March Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.



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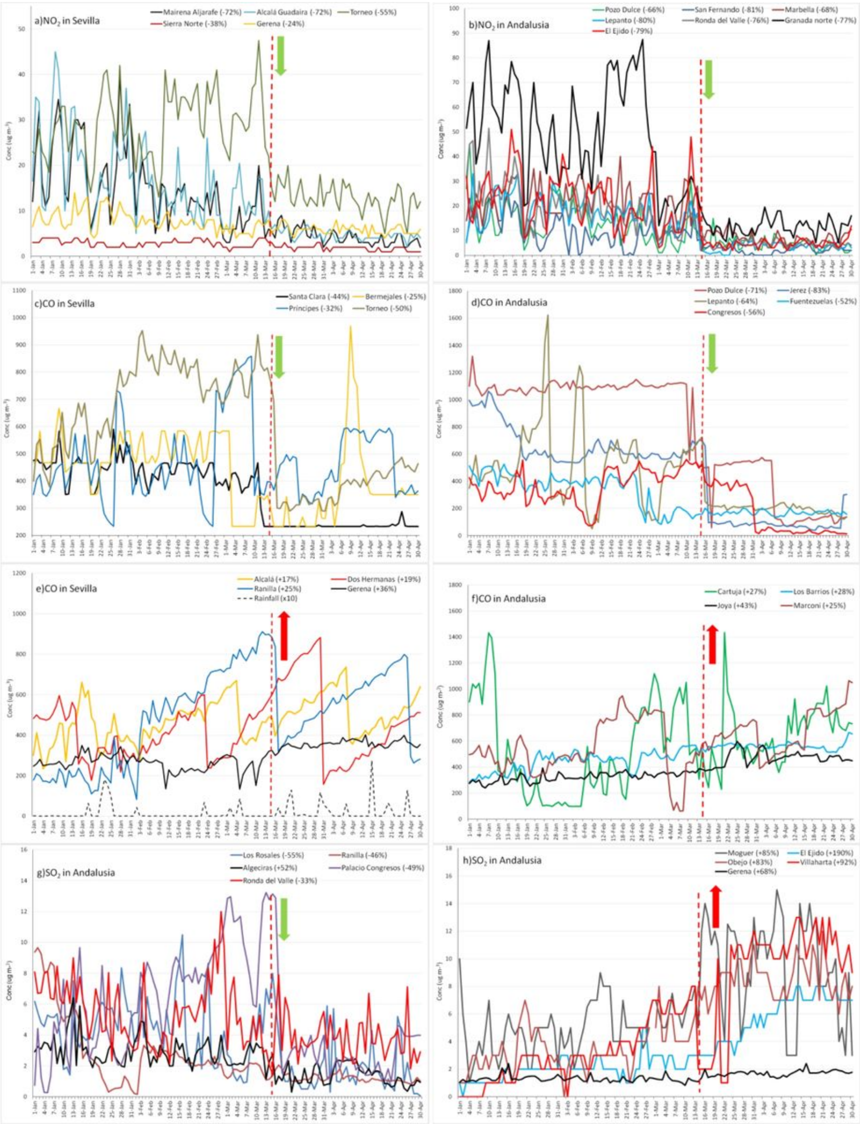
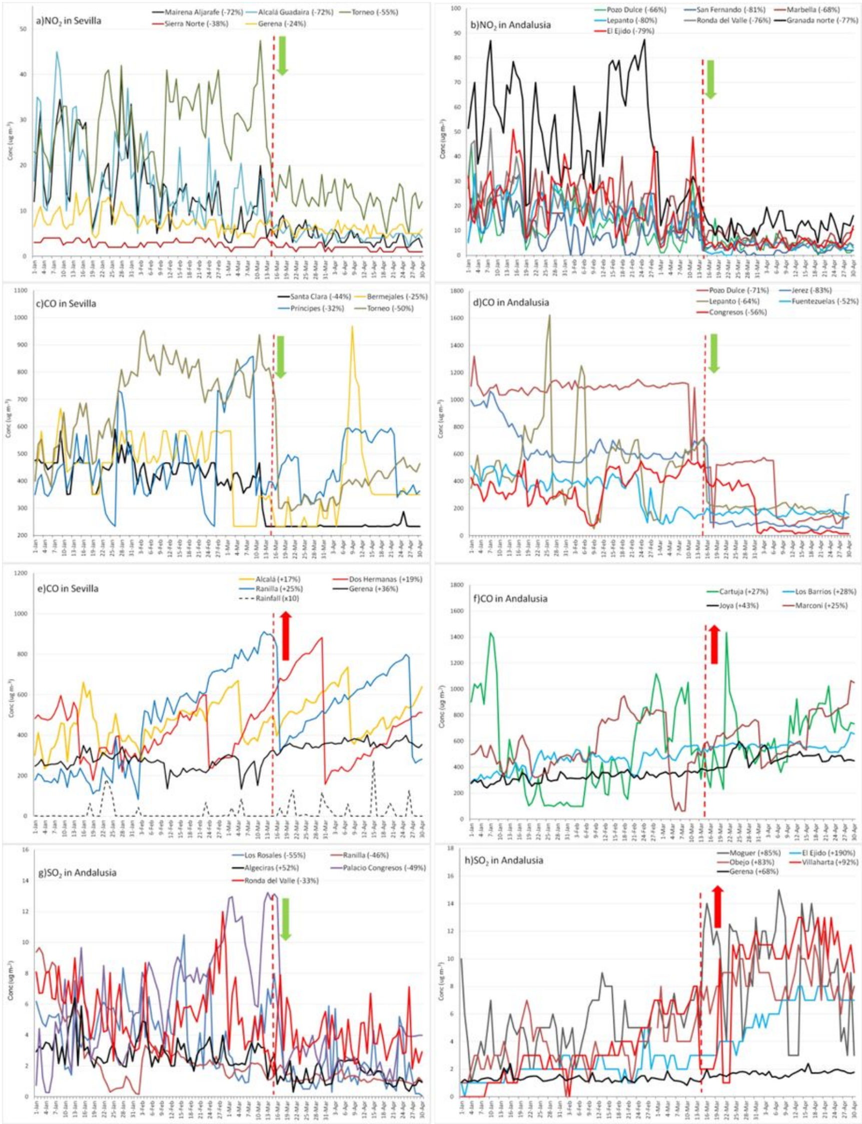


Figure 3

Air pollutants from 1st January - 30th April: NO2 decreasing in Sevilla (a) and in the rest of cities (b); CO decreasing in Sevilla (c) and in the rest of cities (d); CO increasing in Sevilla (e) and in the rest of cities (f); SO2 decreasing in Andalusia (g); SO2 increasing in Andalusia (h)



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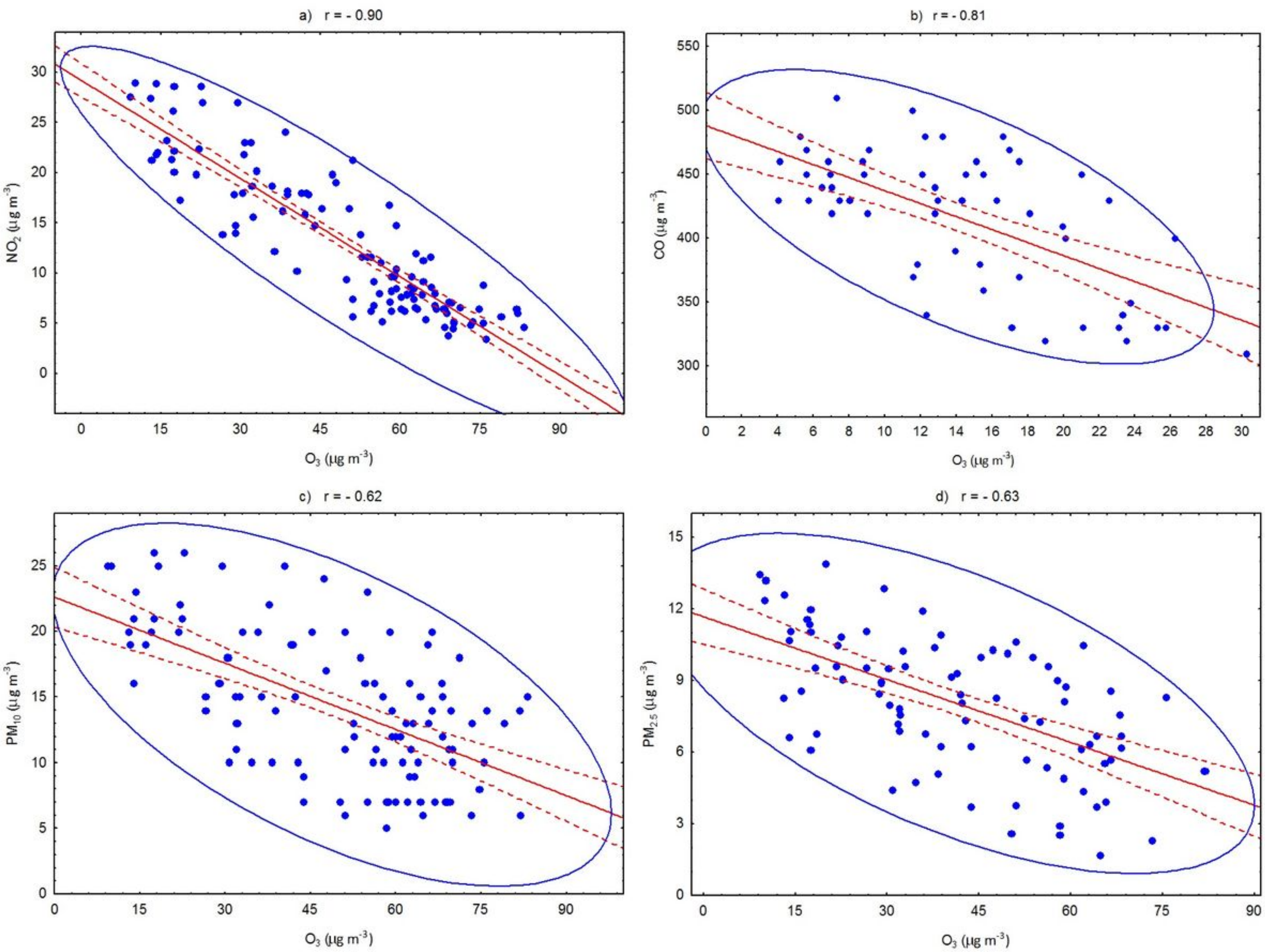
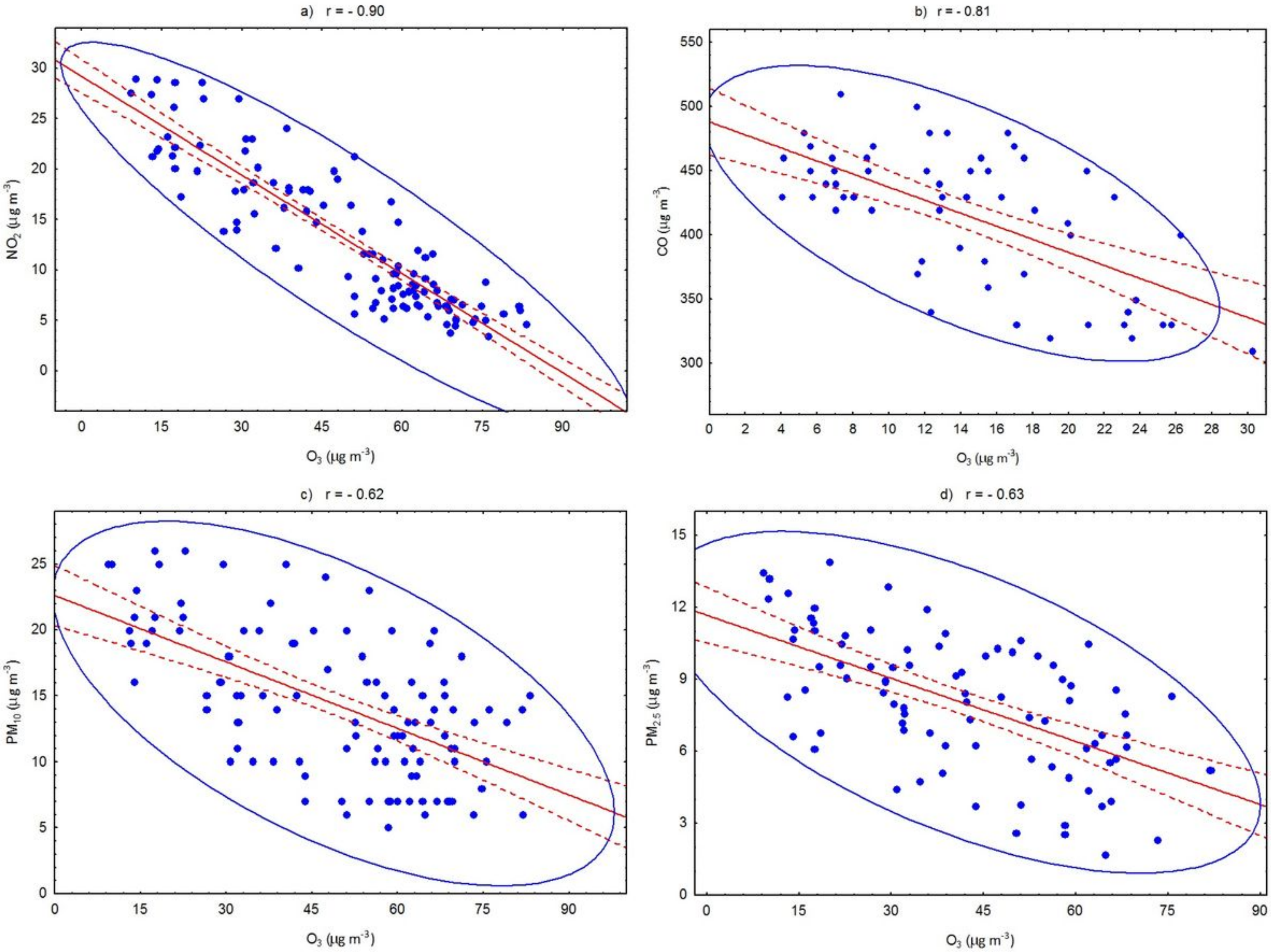


Figure 4

Pearson correlations for ozone: a) ozone vs. nitrogen dioxide; b) ozone vs. carbon monoxide; c) ozone vs. PM10; d) ozone vs. PM2.5



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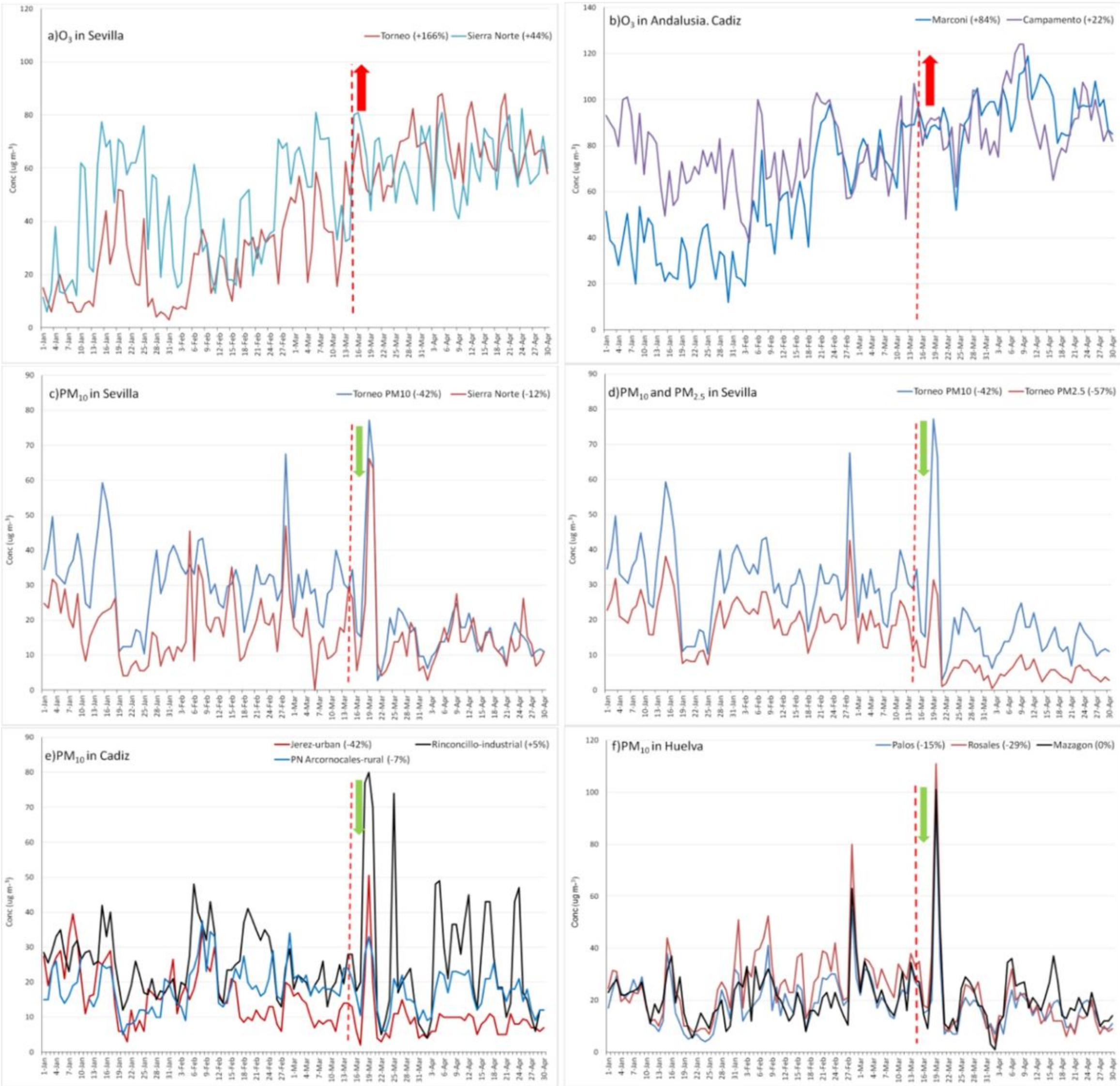
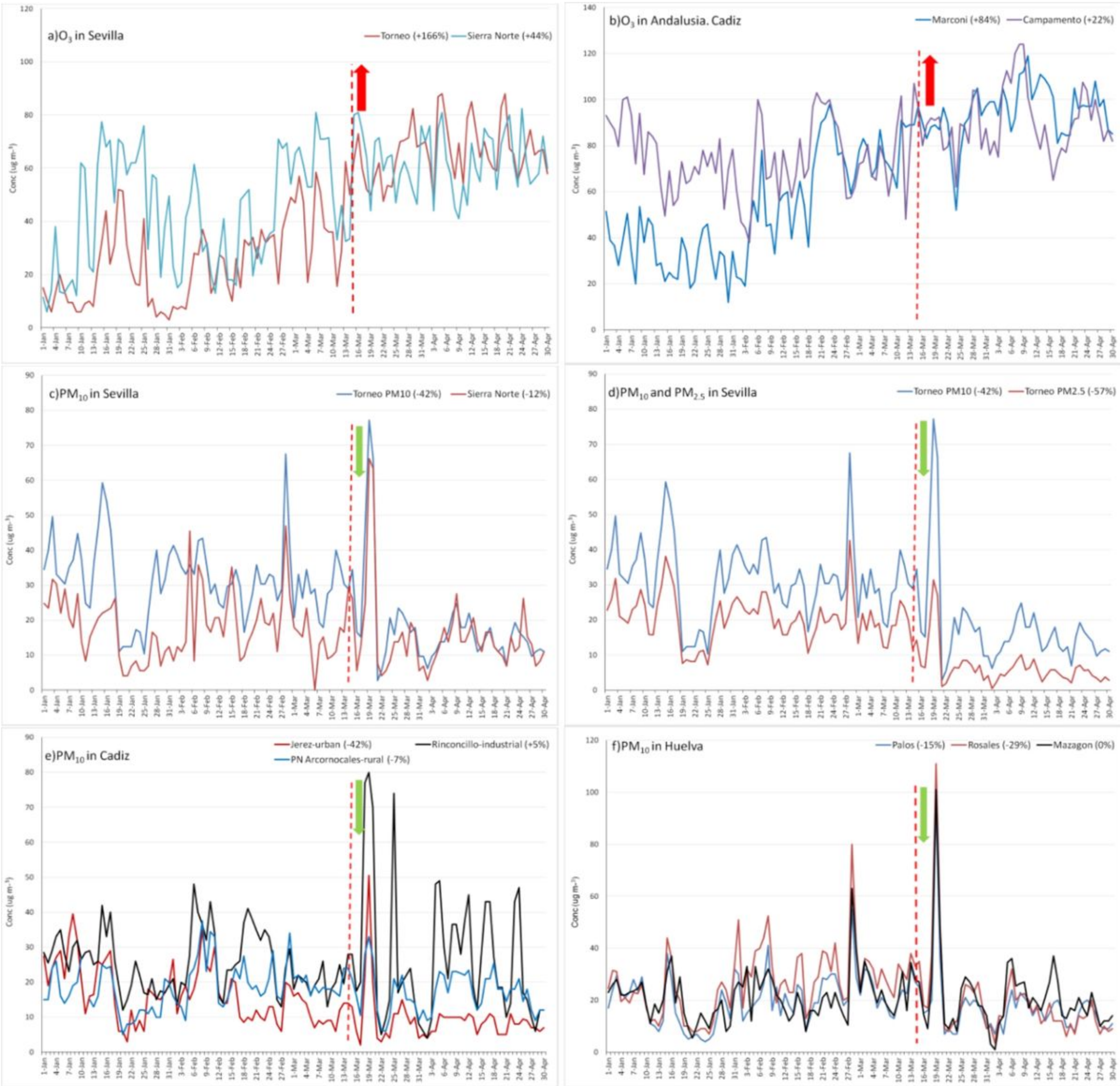


Figure 5

Ozone from 1st January - 30th April: O3 increasing in Sevilla (a) and Cadiz (b); PM10 decreasing in Sevilla (c); PM10 and PM2.5 comparison (d); PM10 decreasing in Cadiz (e) and Huelva (f)



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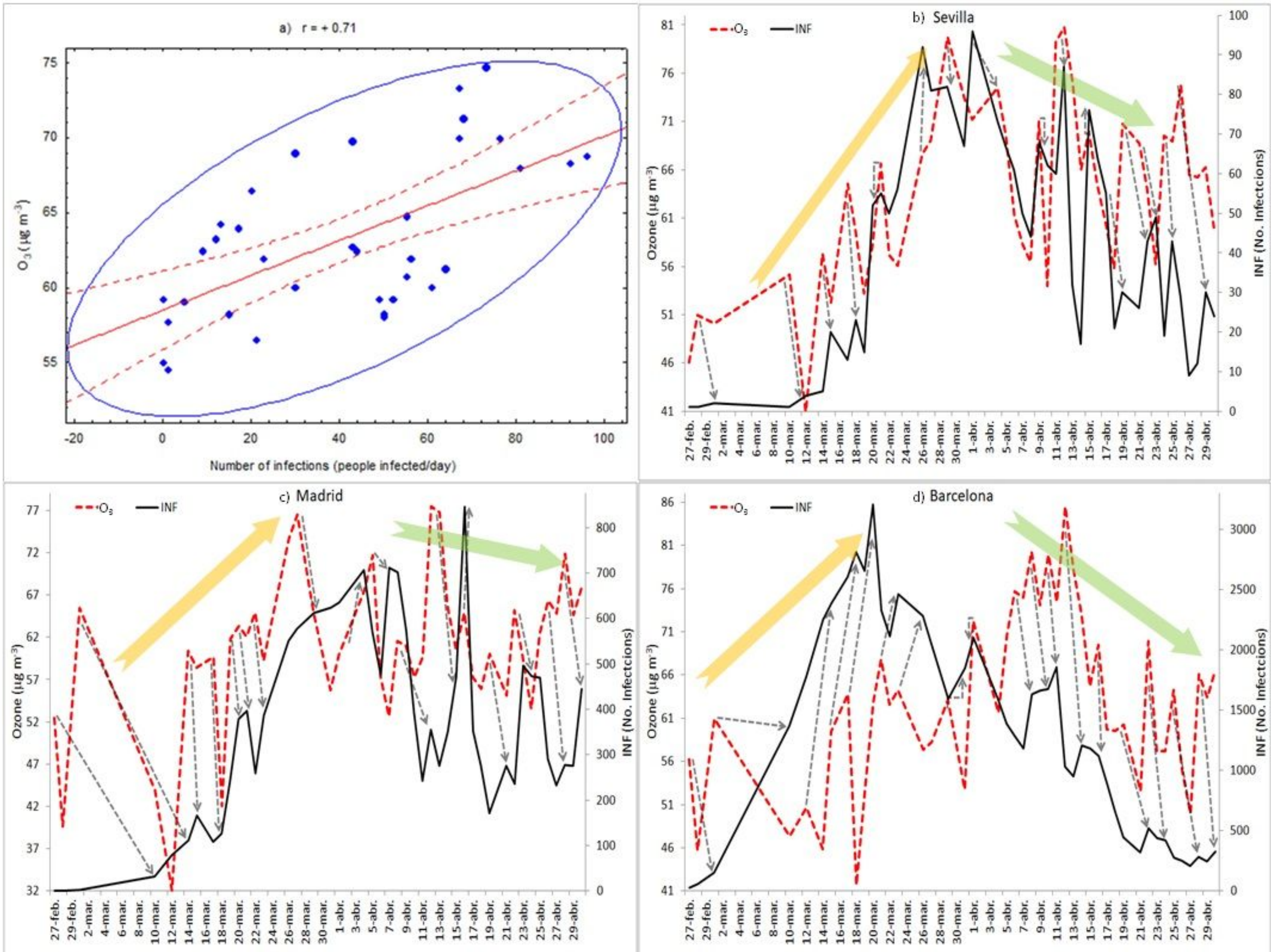
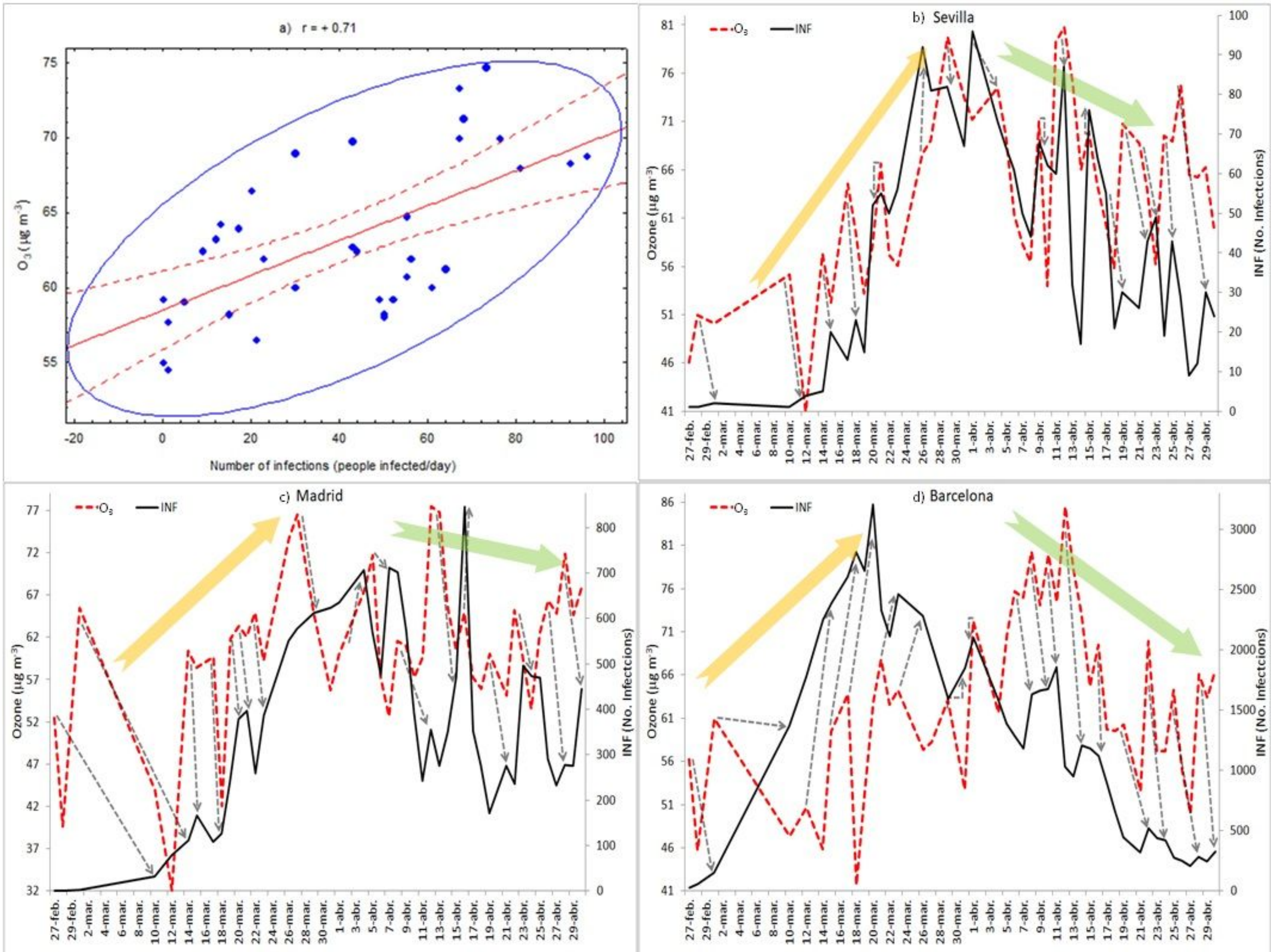


Figure 6

Relationship between Ozone and Infection cases (INF): a) Pearson correlation O3 vs. INF in Sevilla. Profiles of Infection cases vs. Ozone from 27th February to 30th April in Sevilla (b), Madrid (c) and Barcelona (d)



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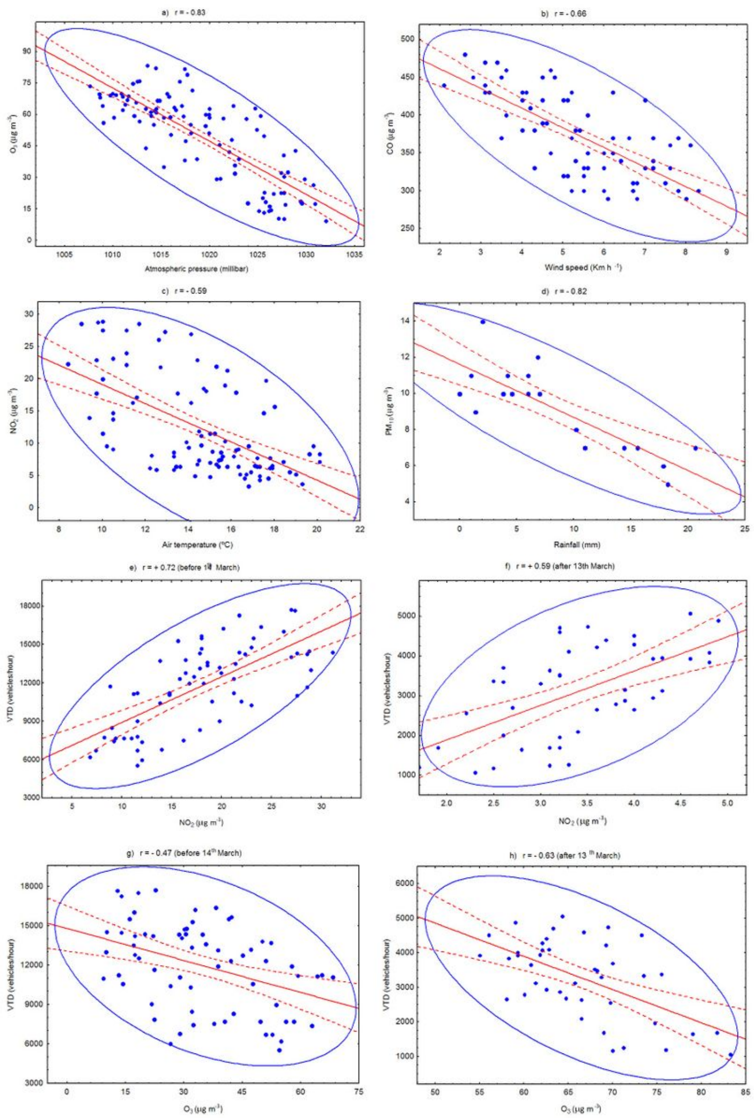
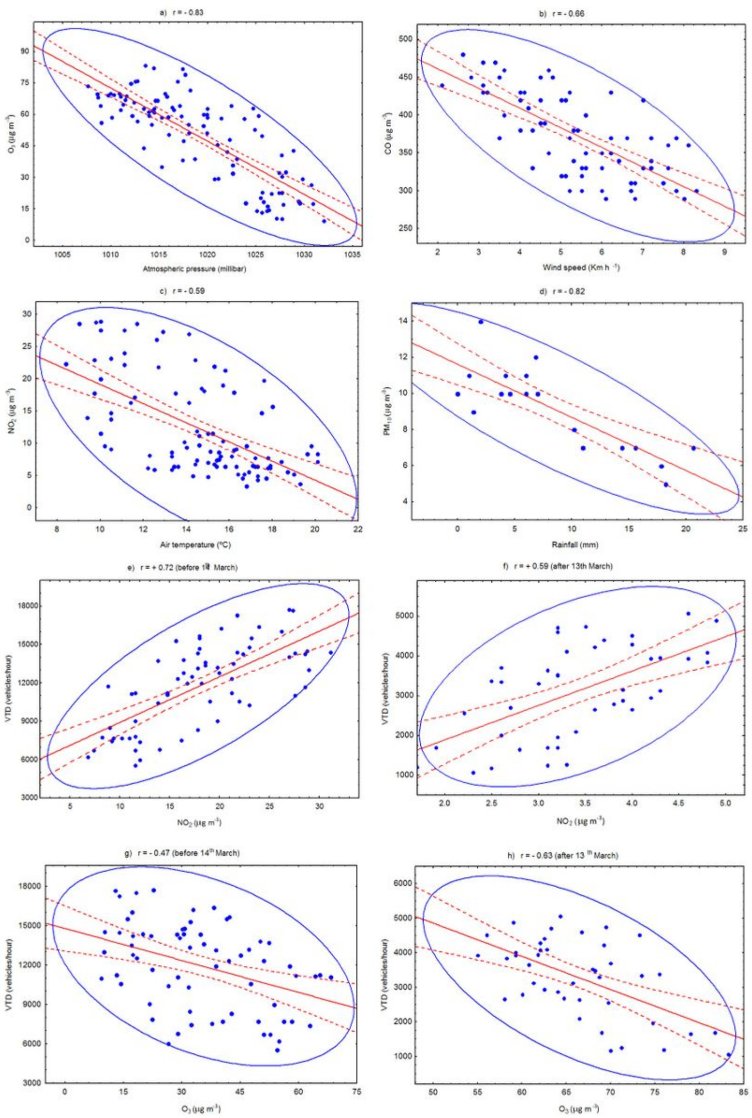


Figure 7

Correlations for air pollutants, meteorological and traffic variables: a) atmospheric pressure vs. O3; b) wind speed vs. CO; c) temperature vs. NO2; d) rainfall vs. PM10; e) VTD vs. NO2 before 14th March; f) VTD vs. NO2 after 13th March; g) VTD vs. O3 before 14th March; h) VTD vs. O3 after 13th March



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