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Impact of the SARS-CoV-2 lockdown measures in Southern Spain on PM10 trace element and gaseous pollutant concentrations

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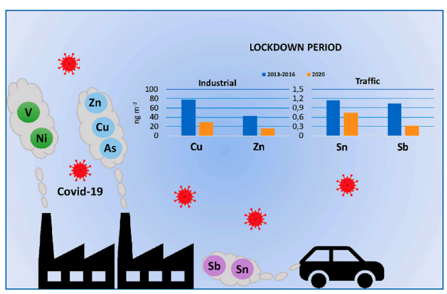
HIGHLIGHTS

- The effect of Covid-19 lockdown over air pollutants has been studied at Andalusia.
- PM10 trace elements and NO_2 levels, related to traffic, were highly reduced.
- General conclusions for industrial sites cannot be drawn.

GRAPHICAL ABSTRACT



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ABSTRACT

Trace element concentrations within PM10, gaseous pollutants (NO_2 and SO_2), and PM10 levels were studied during the Covid-19 lockdown at a regional level in Southern Spain (Andalusia). Pollutant concentrations were compared considering different mobility periods (pre-lockdown, lockdown, and relaxation) in 2020 and previous years (2013–2016). An acute decrease in NO_2 levels (<50%) was observed as a consequence of traffic diminution during the confinement period. Moreover, a lower reduction in PM10 levels and a non-clear pattern for SO_2 levels were observed.

During the lockdown period, PM10 elements released from traffic emissions (Sn and Sb) showed the highest concentration diminution in the study area. Regarding the primary industrial sites, there were no significant differences in V, Ni, La, and Cr concentration reduction during 2020 associated with industrial activity (stainless steel and oil refinery) in Algeciras Bay. Similarly, concentrations of Zn showed the same behaviour at Cordoba, indicating that the Zn-smelter activity was not affected by the lockdown. Nevertheless, stronger reductions of Cu, Zn, and As in Huelva during the confinement period indicated a decrease in the nearby Cu-smelter emissions. Brick factories in Bailen were also influenced by the confinement measures, as corroborated by the marked decrease in concentrations of Ni, V, Cu, and Zn during the lockdown compared to that from previous years.

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This work has shown the baseline concentrations of trace elements of PM10, which is of great value to air quality managers in order to minimise pollution levels by applying the confinement of the population, affecting both traffic and industrial anthropogenic activities.

1. Introduction

By the end of 2019, the first case of Covid-19 (derived from the coronavirus SARS-CoV-2) was reported in Wuhan (China). The virus rapidly spread into Europe throughout Italy, and on 11th March a global pandemic was declared by the World Health Organization (WHO, 2020a). Since then, several countries have established lockdown measures to minimise the number of people infected. With the aim of diminishing human mobility, most economic activities, except those for essential purposes, were reduced. Consequently, flights, trains, and vehicular and public transportation were stopped. This exceptional situation supposed an enormous change from a social, economic, and environmental point of view.

The scientific community took this opportunity to study air quality with less human influence on the environment. TROPOMI and OMI satellite image analysis showed that NO_2 levels decreased in China (40%), South Korea (30%), the United States (42–48%), and Western Europe (20–38%) (Bauwens et al., 2020) during the corresponding confinement periods. Other studies in China have also reported the reduction of gaseous pollutants and PM10 and PM2.5, as a result of lower emissions from road traffic and coal combustion (Xu et al., 2020; Silver et al., 2020). Furthermore, a marked decrease in PM2.5 concentrations was observed in India and the USA, especially in major urban cities with traffic influence (Chen et al., 2020; Mahato and Ghosh, 2020). However, these observations did not always apply to SO_2 levels, which experienced only a slight decrease (Shi and Brasseur, 2020; Kandari and Kumar, 2021). SO_2 emissions were associated with coal burning for residential heating and energy production, and these activities were not suspended during the confinement periods. Apart from evaluating the drop in air pollutant concentrations, other studies have emphasised the relationship between exposure to atmospheric pollution and Covid-19 deaths (Wu et al., 2020; Shakoor et al., 2020). It has been demonstrated that the presence of certain air pollutants (i.e. NO_2, PM10, and PM2.5) increases respiratory and cardiovascular health problems. Therefore, polluted air can involve an extra risk, considering that the virus spreads by droplet and aerosol transmission (Wang and Du, 2020; Zhu et al., 2020; Naqvi et al., 2021).

The Covid-19 virus quickly reached Europe at the end of March 2020; therefore, restriction measures were taken across the territory. Filonchyk et al. (2021) showed the greatest reduction in PM10 and PM2.5 concentrations in April and March 2020 compared to the same period in 2019 in large cities of Poland. Other European cities from the UK and Italy found a clear decrease in NO_x levels, while SO_2 did not have the same pattern at all monitoring sites (Collivignarelli et al., 2020; Donzelli et al., 2020; Higham et al., 2021; Wyche et al., 2021). In spite of the numerous works mentioned above, all of them focus on the study of gaseous and particulate matter (PM) concentration changes, neglecting its chemical composition.

Spain is one of the countries most affected by Covid-19, becoming the fifth state with the highest number of cases (WHO, 2020b). On 14th March, Spain's President declared a state of emergency, imposing a lockdown across the country and limiting all unnecessary displacements of the people. The confinement period in Spain was approximately two months followed by a progressive relaxation period. Subsequently, a "new normal" was created. In addition to the rest of the world, many research groups have studied the changes in air quality in Spain considering these three periods. The authors concluded in an important reduction in NO_x, NO_2, and NO emissions (~40–60%) due to the diminution of road traffic, notably in large cities such as Madrid, Barcelona, and Valencia (Baldasano, 2020; Tobías et al., 2020; Donzelli et al., 2021). The PM concentrations also decreased in these cities (30% PM10), although variations in PM2.5 values were less pronounced because this secondary pollutant has other potential emission sources such as industry, farming, or agricultural biomass burning, but also because of the contribution of secondary component to its total mass concentration. Another important finding was the relative change in SO_2 and CO levels: a smaller decrease or increase was observed because of their origin from specific industrial activities, cargo shipping, or domestic coal heating, which were less affected by government restrictions (Martorell-Marugán et al., 2021; Querol et al., 2021). All of the above works concurred that the confinement imposed in 2020 has offered to the scientific community ideal conditions to study the factors most implied in air quality improvement. Nevertheless, there is no background information about the influence of mobility measures on the PM chemical composition. Until now, most of the authors have focused on gaseous pollutants and changes in PM10 levels. They coincided with a lower decrease in PM10 concentrations compared to NO_2 levels. In this sense, it is interesting to emphasise the complex origin of PM given the physicochemical variety of its components and sources (Moreno et al., 2006). Thus, understanding the evolution of the PM chemical composition during the lockdown should allow a better comprehension of the PM diminution origin and its respective sources.

In this study, we aimed to assess the change in PM10 trace elements during the year 2020 at a regional level (Andalusia, Southern Spain) as a consequence of the adopted lockdown to avoid the spread of Covid-19. For this purpose, the concentrations of PM10 trace elements related to traffic and industrial emissions obtained during 2020 were compared to values recorded in earlier periods (2013–2016). Furthermore, we provide gaseous pollutants (SO_2 and NO_2) and PM10 levels during this year with mobility restrictions. These results can be used in the future to improve air quality in areas with a significant contribution of traffic and industry.

2. Methodology

2.1. Study area

Andalusia is the southernmost region of the Iberian Peninsula and the largest community in Spain (approximately 8.5 million inhabitants, INE, 2021). This region is separated from North Africa by the Strait of Gibraltar, where the Mediterranean Sea (East) and the Atlantic Ocean (West) meet (Fig. 1). The topography of Andalusia encompasses three different areas: the Sierra Morena Range in the north, the Baetic Mountains in the south, and the basin of the Guadalquivir River lies between these two mountainous areas. The climate in Andalusia is generally Mediterranean (hot summers and mild winters), with great variation due to its complex topography: the weather is dryer in the east, and rainfall increases in the coastal locations. Furthermore, a more continental climate is found in areas adjacent to the main mountainous ranges. North African dust outbreaks affect the air quality of Andalusia, which normally occurs during February, March, and summer months. (Rodríguez et al., 2001; Cachorro et al., 2008; Fernández-Camacho et al., 2010; Millán-Martínez et al., 2021b).

The economy of Andalusia is primarily based on service and agricultural sectors, although there are also important industrial estates located across the territory. In this sense, several studies highlight emissions related to different industrial activities. High concentrations of SO_2, and certain toxic metals released from metallurgic processes, ceramic industries, and oil refineries are some examples (Querol et al., 2004, 2008; de la Rosa et al., 2010; Pandolfi et al., 2011; Sánchez de la

Campa et al., 2018; Millán-Martínez et al., 2021a, 2021b). Another important source of pollution in Andalusia is road traffic. The levels of NO_2 and PM10 are higher in populated urban areas because public transportation is, in most cases, insufficient (de la Rosa et al., 2010; Amato et al., 2014).

For PM10 geochemical composition characterisation purposes, 12 monitoring sites belonging to the Air Quality Monitoring Network of the Regional Government of Andalusia were considered in this study. They are distributed in the eight provinces of Andalusia (Fig. 1) and were selected considering their environments and nearby emission sources.

1. Four traffic monitoring stations: Principes (Seville), Palacio Congresos (Granada), Carranque (Malaga), and Mediterraneo (Almeria). These sites are located in metropolitan areas of some of the most populated cities of Andalusia (Seville, 700,000 inhabitants; Malaga 578,000 inhabitants, INE, 2021) and, hence, are affected by dense urban traffic emissions. The air quality in these locations has been previously studied (Fernández-Camacho et al., 2016; Sánchez-Rodas et al., 2017).
2. Five urban-industrial monitoring stations: Campus (Huelva), La Rabida (Huelva), Nerva (Huelva), Puente Mayorga (Cadiz), and Bailen (Jaen). Campus and La Rabida sites have been broadly studied because of the industrial estates established in this area. Thus, the emission of sulphide-related trace elements in PM10 (As, Se, Cd, Sb, Bi, or Pb) from a Cu-smelter has been the principal geochemical anomaly found in many studies (Chen et al., 2016; Sánchez de la Campa et al., 2018; Millán-Martínez et al., 2021a). The Nerva monitoring station is located in the vicinity of a mining district (Riotinto) characterised by sulphide geochemical anomalies derived from fugitive emissions (Sánchez de la Campa et al., 2020). Very close to the Strait of Gibraltar (Algeciras Bay, Cadiz) (Fig. 1) are located a petrochemical plant and oil refinery, a power plant, and a stainless-steel industry. Considering this industrial activity and the high maritime transport, high concentrations of Cr, V, and Ni have been reported in the nearby selected monitoring sites (La Linea, Fig. 1) (de la Rosa et al., 2010; Pandolfi et al., 2011). Bailen is another industrial site characterised by the presence of one of the largest ceramic industries in Spain (Sánchez de la Campa et al., 2010; Sánchez de la Campa and de la Rosa, 2014).
3. Two urban monitoring stations, San Fernando (Cadiz) and Lepanto (Cordoba). The latter is affected by emissions from not-too-far metallurgy factories (Sánchez-Rodas et al., 2017).
4. One rural monitoring station was Matalascañas (Huelva). This monitoring site is situated on the eastern coastline of Huelva, 30 km from the industrial estate already mentioned for the Campus site. Furthermore, Matalascañas is within Doñana National Park, declared a UNESCO "World Reserve of the Biosphere".

To evaluate the Covid-19 restriction measures over the concentration of pollutants, we compared the following subperiods:

1. The pre-lockdown period: 1st February to 15th March when the state of emergency was declared in Spain.
2. Lockdown period: 16th March to 10th May when a relaxation period was established.
3. Relaxation period: 11th May to 31st July. In Spain, different relaxation measures were taken depending on the region (Querol et al., 2021), but in Andalusia, this relaxation applied to nearly the entire territory.

These three sub-periods were studied for PM10 trace element concentrations comparing the year 2020 to the average concentrations obtained during 2013–2016, since no data were available during 2017–2019.

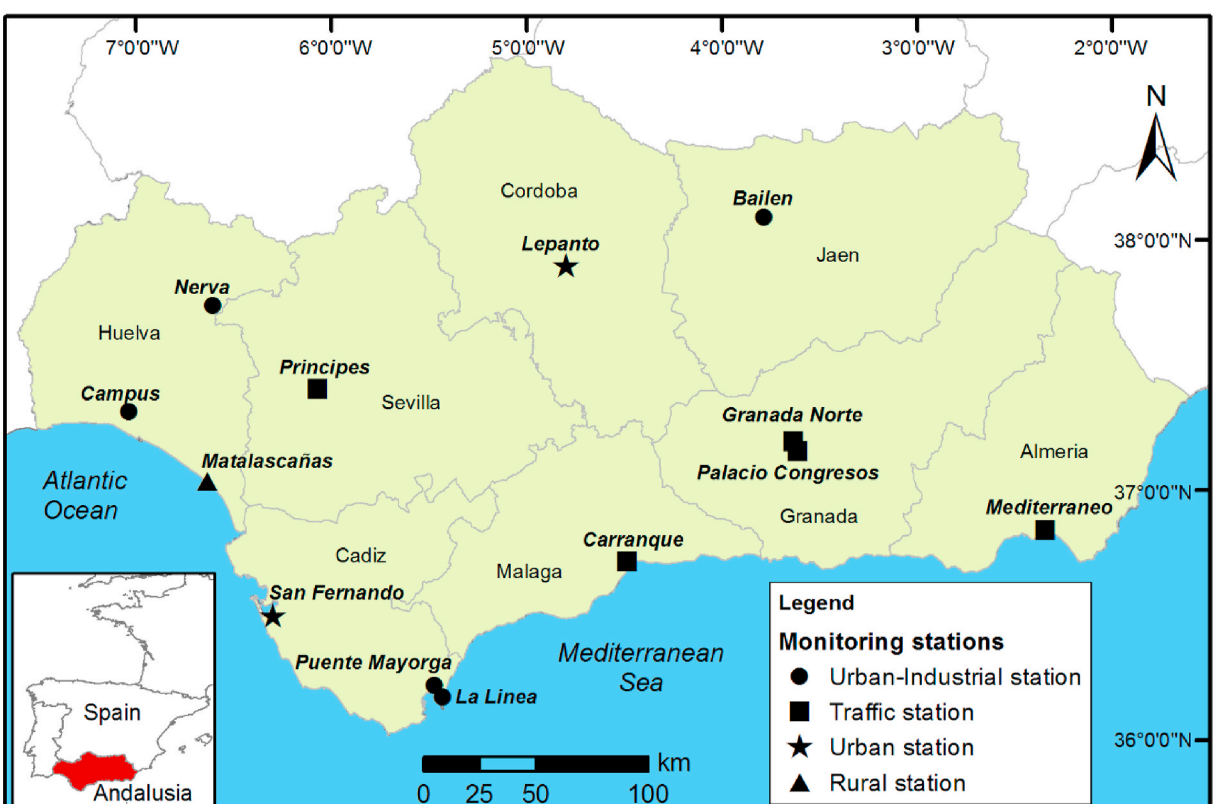

Fig. 1. Map of the study area indicating the provinces and the monitoring stations.

2.2. Industrial activity

The confinement imposed in 2020 supposed strict mobility restrictions and, thus, a direct impact on transport and traffic-related atmospheric emissions. Nevertheless, essential services were not wholly suppressed; therefore, the influence of the lockdown on the industrial sector remains unclear. It has already been mentioned that some monitoring stations are influenced by different industrial activity emissions in Andalusia. Consequently, it is very important to study the main air pollutant emissions derived from these activities during 2020 and previous years using the European Pollutant Emission Register (EPER, 2021). Furthermore, Emissions Inventory of Andalusia Government (2003–2019) has been consulted in order to select the main pollutant contributions in the different Andalusian provinces (https://bit.ly/38QfZxE). From this inventory, it can be inferred that the most representative emissions correspond to several activities located at specific industrial estates (Huelva, Algeciras Bay and Bailen).

In the province of Huelva, several industrial estates affect the monitoring sites of Campus and La Rabida (Millan-Martínez et al., 2021a). The predominant activities contributing to SO_2 emissions are an oil refinery (La Rabida) and a Cu-smelter, and to a much lesser extent, the TiO_2 pigment manufacturing industry. Comparing SO_2 emissions to those obtained in previous years (Fig. S1), a no relevant variation can be observed in 2020. The production of Cu is also responsible for the release of Cu and other sulphide-related elements such as Zn, As, Sb, and Pb, which have significantly decreased over the years (Fig. S1). Attending to Ni emissions coming from the refinery, a decreasing trend was noticed beginning in 2015, whereas Ni levels from the Cu-smelter were higher in 2020 than in previous years.

The other relevant industrial area of Andalusia corresponds to the Algeciras Bay (province of Cadiz, Fig. 1), where is located the Puente Mayorga monitoring station. In this case, SO_2 levels proceed from an oil refinery and a coal-fired power station, which has been inactive since March 2019. Furthermore, the substitution of coke as fuel by natural gas in the oil refinery has resulted in a progressive decrease in SO_2 emissions since 2015 (Fig. S2). High Cr emissions have also been registered in the area, primarily as a result of the presence of a stainless-steel manufacturing plant (Pandolfi et al., 2011; Millán-Martínez et al., 2021b). The amount of Cr emitted from this industry does not present a high variation in 2020 (Fig. S2), but considering the oil refinery, the emissions of Cr have decreased significantly since 2014.

Finally, the ceramic industry (brick and pottery) is highly developed in the city of Bailen (province of Jaen, Fig. 1). These activities are characterised by the emissions of air pollutants such as SO_2, Ni, Zn, Pb, and Cu derived from coke combustion emission plumes (Sánchez de la Campa et al., 2010). Selecting a representative ceramic industry, a reduction in the emissions of these pollutants has been observed in 2020 (Fig. S3).

2.3. PM sampling and analysis

During 2020, PM10 sampling was performed using low-volume captors (DIGITEL DPA14; 58 m^3 day^{-1}). Sampling frequency was one filter every six days for a duration of 24 h. Furthermore, intensive campaigns were also performed at the industrial monitoring stations of Puente Mayorga and Campus (192 and 184 filters/year, respectively). Quartz glass filters were used to collect PM10.

For PM10 chemical analysis, a half fraction of each filter was acid digested (2.5 mL HNO_3: 5 mL HF: 2.5 mL $HClO_4$) for the analysis of trace elements by ICP-MS (Agilent model, 7900) using a modified method proposed by Querol et al. (2001). For quality control, analysis of the NIST-1633c (fly ash, Standard Reference Material) was carried out during every analytical run of the ICP-MS methodology. The digestion procedure of the PM10 samples and ICP-MS analysis were also validated using NIST-1648a (urban particulate matter, Standard Reference Material). External calibration was performed by ICP-MS using 1, 2, and 4 Agilent® multielemental solutions (1–250 μg L^{-1}, as well as HNO_3 5% blank). To minimise the possible fluctuations in the plasma, ^{103}Rh was used as an internal standard. The average precision and accuracy fall for most of the elements under typical analytical errors (in the range of 5–10%).

PM10 chemical information obtained in 2020 was compared to the mean values from 2013 to 2016. In addition, PM10 and gaseous pollutant levels (NO_2 and SO_2), recorded by the corresponding automatic instrumentation (reference methods, EU 2008), were studied during 2020 and the period 2013–2016. In this respect, the monitoring stations Granada Norte site, next to Palacio Congresos, and La Linea site in Algeciras Bay (Fig. 1), were also considered.

3. Results and discussion

3.1. Pluviometry and air masses origin

It is widely known that dust coming from the Saharan desert is often transported to Southern Europe, causing an increase in PM10 levels (Viana et al., 2002; Querol et al., 2009; Salvador et al., 2013). For this reason, it is important to consider these events during the study period. To this end, air masses in the monitoring stations were studied considering two representative locations: Western Andalusia (37° N, 6° W) and Eastern Andalusia (37° N, 3° W). A five-day back-trajectory analysis starting at three different altitudes (500, 1500, and 2500 m a. s.l.) was carried out using the HYSPLIT model (Stein et al., 2015) of the NOAA Air Resources Laboratory (ARL) (https://www.ready.noaa.gov/HYSPLIT.php). Additionally, North African dust events affecting the monitoring stations were also studied using aerosol, dust maps, and satellite images from NRL (http://www.nrlmry.navy.mil/aerosol), SKIRON (https://forecast.uoa.gr/en/forecast-maps/dust/north-atlantic), BSC DUST (https://ess.bsc.es/bsc-dust-daily-forecast), and Earth Data NASA project (https://worldview.earthdata.nasa.gov/).

The daily atmospheric episodes are classified as North African (NAF) when an African dust outbreak occurs, Atlantic (ATL), including the ones from N, NW, SW, and W Atlantic air masses, and other minor air mass origins (regional, Mediterranean, and European origins >5%).

By analysing the mass origin frequency during 2020 and the period 2013–2016, we found Atlantic to be the most frequent origin for Western (58% and 62%, respectively) and Eastern Andalusia (46% and 52%, respectively). In addition, when comparing air masses during lockdown months, similar NAF events occurred for 2013–2016 and 2020: 30% and 34% for Western Andalusia, and 41% and 44% for Eastern Andalusia (Fig. S4).

Rainfall is another relevant driving parameter for pollutant levels since it is well known that precipitation can “wash” the atmosphere and result in the deposition of air pollutants. Data from the Spanish State Meteorological Agency show that during lockdown months in 2020, cumulative precipitation was 80 L, which is comparable to the average obtained for the period 2013–2016 (74 L) for the same months.

3.2. Gaseous pollutants and PM10 levels

The percentages of change in NO_2, SO_2, and PM10 levels in 2020 are shown in Table 1 for each monitoring site compared to the same subperiods in 2013–2016. A general overview reveals an important decrease in NO_2 emissions and PM10 levels in most of the monitoring sites during confinement. In the case of SO_2, a clear pattern is not observed.

During the lockdown period, NO_2 levels were reduced by half in most of the monitoring sites. This fact is especially important in traffic monitoring stations with high NO_2 levels (i.e. the annual mean in Granada Norte in 2016 was 44 μg NO_2 m^{-3}), where the annual NO_2 concentration standard (40 μg m^{-3}) established by European directives (EU, 2008) is sometimes exceeded. The primary NO_2 anthropogenic sources are vehicles exhaust emissions and fossil fuel combustion

Table 1
Variation of percentages in 2020 pollutants levels at the considered monitoring stations of Andalusia during the pre-lockdown, lockdown and relaxation periods, compared with the respective averaged values for the same three periods in 2013–2016.

MONITORING STATION		PRE-LOCKDOWN	LOCKDOWN	RELAXATION
TRAFFIC				
Granada Norte	SO_2	−30	−20	−9
	NO_2	8	−58	−20
	PM10	11	−35	−11
Principes	SO_2	−17	−31	−22
	NO_2	−21	−61	−41
	PM10	8	−33	−15
Carranque	SO_2	−76	−79	−53
	NO_2	7	−55	−11
	PM10	7	−23	−23
Mediterraneo	SO_2	−55	−64	−72
	NO_2	−6	−66	−29
	PM10	20	−41	−28
URBAN-INDUSTRIAL				
Bailen	SO_2	25	−2	20
	NO_2	−1	−55	−2
	PM10	−4	−41	−12
Campus	SO_2	80	−1	−5
	NO_2	35	−35	−36
	PM10	1	−12	−5
La Linea	SO_2	−77	−84	−82
	NO_2	−19	−58	−37
	PM10	−26	−34	−37
URBAN				
Lepanto	SO_2	−34	−47	−20
	NO_2	−11	−72	−40
	PM10	30	−27	−4
San Fernando	SO_2	−67	−76	−62
	NO_2	−26	−77	−25
	PM10	11	−13	7
RURAL				
Matalascañas	SO_2	−13	33	39
	NO_2	−18		−71
	PM10	15	3	−19

(Hesterberg et al., 2009), and in Spain, more than 75% of NO_2 emissions are due to road traffic (MITECO, 2020). This gaseous pollutant contributes to the formation of PM2.5, and O_3 in the air, representing a human health risk (Santurtún et al., 2017). Fig. 2 shows a pronounced decrease in NO_2 levels corresponding to the beginning of the lockdown period at the traffic monitoring stations. Although levels tended to recover during the relaxation period, they did not reach the values recorded in January 2020. This is due to the mobility restrictions imposed by the Spanish government between certain regions, as well as the enhancement of teleworking with the aim of avoiding human contact. Therefore, the influence of the lockdown on traffic emissions and the corresponding NO_2 level reduction in Andalusia agrees with previously published results in other regions of Spain (Baldasano, 2020; Tobías et al., 2020; Querol et al., 2021; Ceballos-Santos et al., 2021; Donzelli et al., 2021).

In the case of PM10 levels, a less marked decrease occurred in the study area. PM10 concentrations were reduced between 12% and 41%, with the highest variation in the inland cities of Andalusia. There are various sources that contribute to the total mass content of PM10; marine aerosols and mineral dust from natural resuspension are two of the primary sources. This could explain why Campus (urban-industrial) and Matalascañas (rural) PM10 levels remained stable or slightly decreased during the lockdown period. These monitoring stations, located in the western part of Andalusia, have a high contribution of mineral dust and marine aerosols to PM10 (Millán-Martínez et al., 2021) and are strongly affected by NAF episodes. Nevertheless, a PM10 chemical composition study is needed to comprehend the actual origin of the decrease.

SO_2 is a gaseous pollutant generated by high-temperature processes such as industrial activities, energy production or domestic heating. In Andalusia, 34% of SO_2 emissions were derived from oil refineries (EPER, 2019). SO_2 levels are normally low, not higher than 10 μg m^{-3}, and small differences in concentrations could involve high variation percentages. The European normative does not establish any annual SO_2 limit; however, it is recommended that it does not exceed 350 SO_2 μg·m^{-3}·h^{-1} more than 24 times per year. This target value is rarely exceeded; nevertheless, sporadic hourly SO_2 peaks (>20 μg m^{-3} h^{-1}) have often been measured at industrial sites. In this sense, it is more interesting to evaluate the number of days with SO_2 impacts over the previous years to those in 2020.

The monitoring stations of Campus and Puente Mayorga have been previously described because of the high industrial influence of the oil refineries, petrochemical plants, and metallurgic activities developed in the area (Pandolfi et al., 2011; Sánchez de la Campa et al., 2018). In Fig. 3, it can be clearly seen that there has been no decrease in SO_2 impact days in Campus since 2012. In the case of Puente Mayorga, impact days started to be reduced from 2018 to 2020, possibly as a result of the use of natural gas as fuel in several industrial activities (personal communication). This is in concordance with the high reduction percentage of SO_2 levels in La Linea, not only in the lockdown period.

Analysing the year 2020, impact days became lower than in Campus over several months, contrary to what has been observed in previous

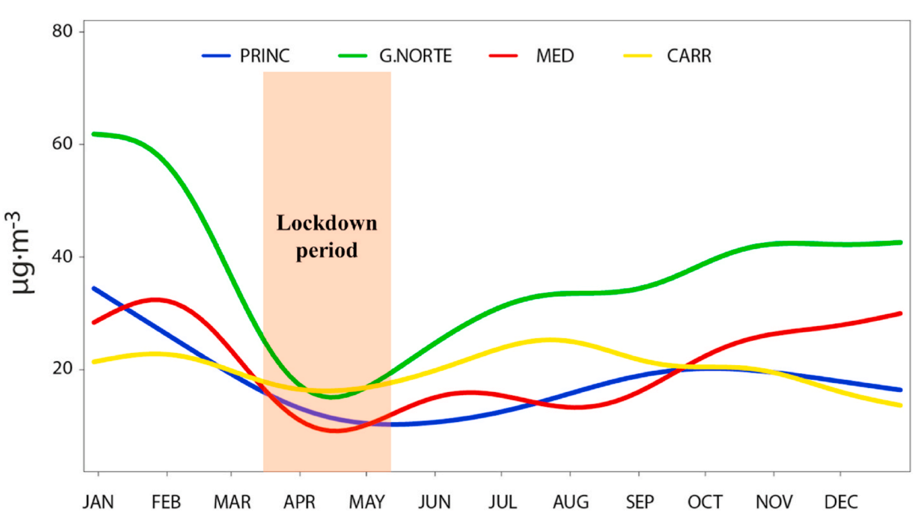

Fig. 2. Time variation of NO_2 concentrations during the year 2020 at the traffic monitoring stations considered in this study. PRINC: Principes; G.NORTE: Granada Norte; MED: Mediterraneo; CARR: Carranque.

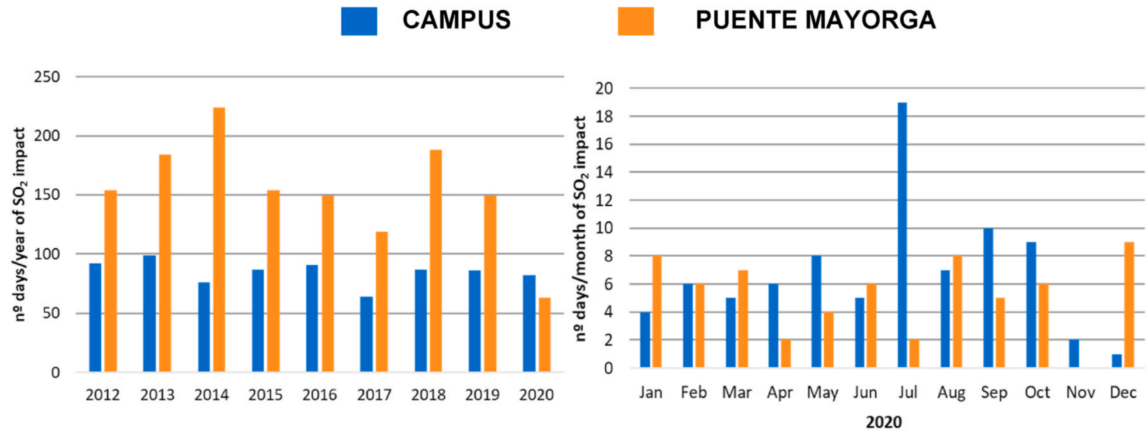

Fig. 3. SO_2 impacts days per year in the period 2012–2020 (left) and per month during 2020 at the urban-industrial monitoring stations of Campus and Puente Mayorga.

years. Another important observation is the slight reduction of the SO_2 impact days in March in Campus, whereas in Puente Mayorga, impacts are higher in March than in February. Nevertheless, SO_2 concentrations are highly dependent on the wind regimes, making it more difficult to evaluate industrial activity during the lockdown period.

In summary, as a consequence of the Covid-19 adopted measures, there was a remarkable reduction in NO_2 emissions at a regional level as a result of the lower traffic during the stay-home period. To a lesser extent, due to the contribution of natural sources, lockdown measures also resulted in a decreasing pattern in PM10 levels in most cases. A different scenario was observed for SO_2 levels, with no clear trends in change percentage and impact days during the lockdown period.

3.3. Geochemical maps of PM10 trace elements (2020 vs 2013–2016)

Annual average concentrations of main PM10 trace elements during 2020 are summarized in Table 2 and represented in more detail in the geochemical maps in Fig. S5. When comparing with the mean concentrations obtained for the period 2013–2016 certain anomalies are still observed in 2020 as a consequence of traffic or specific industrial emissions.

V and Ni are two elements whose emissions are derived from industrial activities such as oil refineries, coal-fired power plants, and maritime transport from fuel-oil combustion. Previous studies have highlighted the high Ni and V concentrations in PM10 in Puente Mayorga as a consequence of industrial estate emissions (Moreno et al.,

Table 2
Mean PM10 trace elements concentrations (in ng m^{-3}) at the monitoring stations of Andalusia during 2020. P.CON: Palacio Congresos; PRINC: Principes; MED: Mediterraneo; CARR: Carranque; PMAY: Puente Mayorga; CAMP: Campus; BAIL: Bailen; RAB: La Rabida; NERV: Nerva; LEP: Lepanto; S.FER: San Fernando; MAT: Matalascañas.

Monitoring station	Traffic				Urban-Industrial					Urban		Rural
	P.CON	PRINC	MED	CARR	PMAY	CAMP	BAIL	RAB	NERV	LEP	S.FER	MAT
N, ng m^{-3}	54	61	60	59	192	184	64	58	63	62	59	62
Li	0.78	0.41	0.39	0.26	0.23	0.29	0.68	0.33	0.59	0.48	0.23	0.31
Be	0.02	0.02	0.01	0.01	0.01	0.01	0.03	0.01	0.02	0.03	0.01	0.06
Sc	0.17	0.12	0.10	0.08	0.05	0.09	0.15	0.10	0.18	0.13	0.07	0.08
V	3.28	1.79	2.75	4.08	2.55	1.60	23.71	1.91	1.71	1.76	1.65	1.39
Cr	3.46	3.34	3.21	2.94	13.1	1.24	2.32	2.36	1.57	2.56	1.75	1.12
Co	0.25	0.20	0.18	0.16	0.33	0.14	0.24	0.28	0.28	0.17	0.10	0.13
Ni	2.98	3.45	3.10	3.14	6.59	1.32	6.90	2.54	1.45	1.62	2.41	1.38
Cu	8.88	13.5	7.74	9.26	7.48	21.2	5.95	276	12.8	20.3	3.30	30.4
Zn	21.5	34.3	16.2	26.1	39.7	16.8	24.2	60.4	14.5	80.7	14.1	19.1
Ga	0.30	0.20	0.15	0.14	0.11	0.13	0.29	0.15	0.27	0.26	0.11	0.11
Ge	0.33	0.31	0.39	0.31	0.12	0.13	0.32	0.16	0.14	0.42	0.33	0.11
As	0.41	0.87	0.51	0.33	0.36	3.29	0.46	6.11	1.61	0.51	0.41	0.94
Se	0.10	0.12	0.10	0.10	0.04	0.12	0.13	0.19	0.13	0.14	0.13	0.10
Rb	1.50	0.98	0.67	0.71	0.32	0.68	2.14	0.76	1.18	1.87	0.47	0.60
Sr	11.0	3.97	4.83	3.24	2.28	2.07	5.33	3.59	3.37	4.19	3.40	3.32
Y	0.69	0.68	0.47	0.55	0.17	0.27	0.62	0.33	0.41	0.67	0.51	0.26
Zr	3.23	3.45	3.36	3.05	0.73	2.46	3.27	1.60	3.11	3.38	2.60	1.44
Mo	11.6	11.7	8.28	11.1	0.60	2.41	9.36	5.01	4.44	8.19	10.5	3.63
Cd	0.07	0.08	0.06	0.07	0.29	0.37	0.08	0.61	0.11	0.30	0.03	0.10
Sn	2.06	2.46	2.14	2.44	1.39	1.28	0.88	0.97	0.65	2.24	0.84	0.60
Sb	1.26	1.58	1.02	1.16	0.55	0.66	0.79	1.23	0.48	1.24	0.33	0.24
Cs	0.07	0.04	0.04	0.02	0.02	0.03	0.11	0.03	0.06	0.10	0.02	0.02
Ba	17.9	19.2	16.2	20.9	3.12	6.92	19.2	8.73	8.77	20.9	13.8	7.11
La	0.63	0.48	0.37	0.32	1.66	0.33	0.56	0.35	0.50	0.67	0.30	0.28
W	0.03	0.03	0.03	0.03	0.08	0.02	0.04	0.08	0.10	0.08	0.03	0.09
Tl	0.01	0.01	0.03	0.01	0.01	0.02	0.25	0.03	0.02	0.07	0.00	0.01
Pb	5.28	4.98	4.06	3.19	6.05	10.8	14.5	26.6	6.37	7.32	3.32	3.63
Bi	0.12	0.34	0.18	0.17	0.12	0.57	0.15	0.85	0.26	0.21	0.16	0.10
U	0.18	0.21	0.15	0.17	0.03	0.07	0.17	0.11	0.09	0.18	0.15	0.06

2008; Pandolfi et al., 2011; Sánchez de la Campa and de la Rosa, 2014). Even though a high concentration of V was still found in Bailen in 2020 (24 ng m^{-3}), the recent use of natural gas as fuel in the nearby industries of Puente Mayorga station (CEPSA, 2020) and the inactivity of the coal-fired power plant (European Policies to reduce CO_2 emissions), resulted in a strong reduction of V concentrations (2.6 ng m^{-3} 2020 vs 22 ng m^{-3} 2013–2016). A similar situation was observed for Ni concentrations, with maximum values in Bailen (6.9 ng m^{-3}) and an important diminution in Puente Mayorga (average concentrations of 6.6 and 17 ng m^{-3} in 2020 and 2013–2016, respectively). The highest concentrations of Cr in 2020 (13 ng m^{-3}) corresponded to Puente Mayorga, derived from a stainless-steel factory located in Algeciras Bay. In addition, the catalysts used in oil refining processes can release La, explaining why this metal has the highest values in Puente Mayorga (1.7 ng m^{-3} 2020).

The concentrations of As were highest in the monitoring stations of Campus and La Rabida in 2020 (3.3 and 6.1 ng m^{-3}, respectively) due to Cu-smelter emissions in the area. As a result of this industrial activity, high concentrations of Cu, Zn, and Pb were also found in La Rabida (276, 60, and 27 ng m^{-3}, respectively in 2020). In the city of Cordoba (Lepanto monitoring station), previous studies have described high Cu and Zn concentrations from the nearest Zn-smelters (de la Rosa et al., 2010; Sánchez-Rodas et al., 2017; Millán-Martínez et al., 2021b). However, an important decrease was observed in 2020 (Cu: 20 ng m^{-3}; Zn: 81 ng m^{-3}).

Sn and Sb are typical elements released from non-exhaust traffic emissions (brake wear) (Amato et al., 2014). The brake-pads of vehicles contain Sb_2S_3 as lubricant for friction material, which is emitted as particles during break wear. The heat generated during this process can oxide considerably Sb_2S_3 (von Uexküll et al., 2005; Lijima et al., 2007; Amato et al., 2009). The related-origin of Sb with traffic emissions have been previously observed in other traffic monitoring stations of Andalusia (Sánchez-Rodas et al., 2017). Accordingly, the highest concentrations of these elements were registered in 2020 in the four traffic-monitoring stations selected for this study (Fig. S4).

In summary, the principal geochemical anomalies already described in previous years were also observed in 2020. However, the decrease or inactivity of certain industrial activities, as well as the implementation of abatement measures, have resulted in mean concentration diminutions for some trace elements. Relevant reduction percentages were registered in Puente Mayorga in V (88%) and Ni (60%) in 2020 with respect to 2013–2016. Similarly, the mean concentrations of Cu and Zn also decreased in 2020 at Campus (71% and 53%, respectively) and La Rabida (32% and 58%, respectively). In addition, the concentrations of these two elements were diminished by approximately 80% at Lepanto. Cr (Puente Mayorga) and As (Campus and La Rabida) did not display relevant variations in the annual concentrations of PM10 in both periods.

3.4. *Evolution of PM10 trace elements in year 2020*

To study the influence of the Covid-19 mobility restrictions on the PM10 trace element concentrations, a more detailed study was performed during the year 2020. To this end, 2020 mean concentrations were compared to those obtained in the earlier period of 2013–2016. In both cases, the above-mentioned subperiods (pre-lockdown, lockdown, and relaxation) have been considered. The concentration variations are shown in Fig. 4.

The mean concentrations of V and Ni at the Puente Mayorga monitoring site showed significant differences during the three subperiods when comparing 2020 and 2013–2016. Even though the percentage reductions of both metals were approximately 80%, this decrease is

Fig. 4. Comparison between mean trace elements concentrations (ng m^{-3}) during 2020 and the period 2013–2016 taking into account the three different mobility periods: pre-lockdown, lockdown and relaxation.

related to the fuel change in the Algeciras Bay refinery and the inactivity of the nearby coal-fired power station since 2019. In addition, regarding La concentrations (cracking process catalysts of oil refinery) and Cr (stainless steel industry), no significant diminutions were observed in any subperiod. From these results, it can be inferred that the industrial activity developed in the area did not decrease during the lockdown.

A different scenario occurred at the industrial site of Bailen. V and Ni concentrations decreased during the lockdown and relaxation periods during 2020 with respect to 2013–2016 (Fig. 4). In this case, the high concentrations of Ni and V have been associated with the use of coke as fuel in brick factories; therefore, a lower demand for construction sector materials during the confinement may have produced a reduction in these emissions.

Concerning Cu and Zn concentrations derived from the Zn metallurgy activity at Lepanto, significant reductions were observed, between 70% and 90%, in the three subperiods. These activities do not seem to be affected by the confinement measures because the diminution concentrations were also maintained during the pre-lockdown and relaxation periods.

In the province of Huelva, Cu, Zn, and As concentrations showed the highest diminution in 2020 with respect to the 2013–2016 mean values in the lockdown and relaxation periods at Campus. This indicates a decrease in the emissions from the Cu-smelter previously defined. However, these toxic trace element concentrations did not show a high diminution during the lockdown at La Rabida, which is also affected by the Cu-smelter. The different scenarios given between La Rabida and Campus can result from fugitive emissions due to the handling of polymetallic sulphides in a port area close to La Rabida. (Millán-Martínez et al., 2021a).

Finally, significant reductions in Sn and Sb concentrations during the lockdown stage were higher than those observed in the pre-lockdown and relaxation periods. This decrease was registered in all the traffic monitoring sites as well as in urban and industrial stations such as San Fernando, Campus, and Puente Mayorga (Fig. 4). Traffic emissions were remarkably reduced by the imposed confinement in 2020.

The comparison of the PM10 trace element mean concentrations obtained in the three different subperiods in 2020 and 2013–2016 revealed the change in anthropogenic activities under mobility restrictions. Road traffic-related elements (mainly Sn and Sb) underwent a significant reduction during this period. In the industrial sector, the results showed that emissions from the stainless-steel factory (Cr) and the oil refinery (V, Ni, and La) in Puente Mayorga were maintained. Zn-smelters located in Lepanto were not affected during lockdown. Conversely, the ceramic industry in Bailen and the Cu-smelter affecting Campus (Huelva) experienced a decrease during confinement months.

4. Conclusions

Mobility restrictions imposed by the Spanish Government during the global pandemic of SARS-CoV-2 (Covid-19) gave rise to unprecedented conditions for evaluating the variation of certain air pollutant concentrations. In the present study, gaseous pollutants (NO_2 and SO_2) PM10 levels and trace element concentrations in PM10 during 2020 were compared to those obtained in 2013–2016 in Andalusia (Southern Spain). The results showed a strong decrease in NO_2 in the study area during the lockdown period resulting from traffic diminution. A smoother reduction was observed in PM10 levels, and a non-defined pattern was found for SO_2 levels.

The trace element results revealed an accused diminution of road traffic emissions during the lockdown period compared to previous years. Considering the elements derived from particular industrial emissions, different conditions can be distinguished. In Algeciras Bay, the concentration variations of Cr, V, and Ni, as well as the lack of reduction of SO_2 impacts during the lockdown period, corroborated the continuity of the industrial activity (oil refinery and stainless-steel factory) in the area. At the Bailen site, a significant decrease in elements derived from the brick production industry (Ni, V, Cu, Zn, and Pb) during the confinement period can be associated with a reduction in construction sector demand. In the city of Huelva, trace elements associated with Cu-smelter (As, Cu, and Zn) showed larger decreases during the lockdown and relaxation periods. This fact seems to be in concordance with the lower number of SO_2 impact days during March 2020, which points to a steady reduction in the Cu-smelter emissions.

The results obtained in this study are of great interest to air quality managers to determine the degree of reduction in the concentration of metals in ambient air during the confinement of the population. The most significant reduction in the metal concentration in PM10 is related to traffic mobility. However, metals derived from industry are dependent on the type of activity present.

Sample CRediT author statement

María Millán Martínez: Investigation, Writing – original draft. Daniel Sánchez-Rodas: Reviewing, Methodology. Ana M. Sánchez de la Campa: Reviewing, Methodology. Jesús D. de la Rosa: Funding acquisition, Reviewing.

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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Appendix A. Supplementary data

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