Science of the Total Environment 735 (2020) 139542

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Amplified ozone pollution in cities during the COVID-19 lockdown

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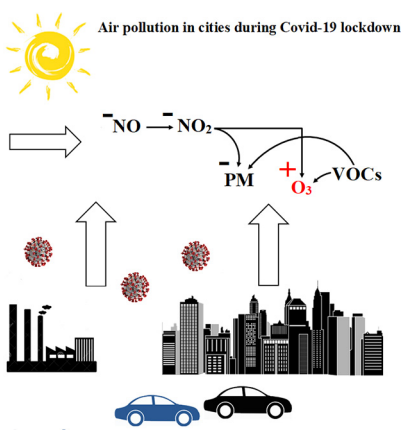
HIGHLIGHTS

- Air quality during the COVID-19 lockdown in 4 European and 1 Chinese cities
- The lockdown caused a substantial reduction in NO_x in all cities (~ 56%)
- Reductions in PM were much higher in Wuhan (~ 42%) than in Europe (~ 8%)
- The lockdown caused an ozone increase in all cities (17% in Europe, 36% in Wuhan)
- The lockdown effect on O_3 production was higher than the weekend effect

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 11 May 2020
Received in revised form 17 May 2020
Accepted 17 May 2020
Available online 20 May 2020

Keywords:
Air quality
Ozone
Coronavirus
Lockdown
Risk assessment

ABSTRACT

The effect of lockdown due to coronavirus disease (COVID-19) pandemic on air pollution in four Southern European cities (Nice, Rome, Valencia and Turin) and Wuhan (China) was quantified, with a focus on ozone (O_3). Compared to the same period in 2017–2019, the daily O_3 mean concentrations increased at urban stations by 24% in Nice, 14% in Rome, 27% in Turin, 2.4% in Valencia and 36% in Wuhan during the lockdown in 2020. This increase in O_3 concentrations is mainly explained by an unprecedented reduction in NO_x emissions leading to a lower O_3 titration by NO. Strong reductions in NO_2 mean concentrations were observed in all European cities, ~53% at urban stations, comparable to Wuhan (57%), and ~65% at traffic stations. NO declined even further, ~63% at urban stations and ~78% at traffic stations in Europe. Reductions in $PM_{2.5}$ and PM_{10} at urban stations were overall much smaller both in magnitude and relative change in Europe (~8%) than in Wuhan (~42%). The PM reductions due to limiting transportation and fuel combustion in institutional and commercial buildings were partly offset by increases of PM emissions from the activities at home in some of the cities. The NO_x concentrations during the lockdown were on average 49% lower than those at weekends of the previous years in all cities. The lockdown effect on O_3 production was ~10% higher than the weekend effect in Southern Europe and 38% higher in Wuhan, while for PM the lockdown had the same effect as weekends in Southern Europe (~6% of

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https://doi.org/10.1016/j.scitotenv.2020.139542
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difference). This study highlights the challenge of reducing the formation of secondary pollutants such as O_3 even with strict measures to control primary pollutant emissions. These results are relevant for designing abatement policies of urban pollution.

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1. Introduction

Nowadays, 55% of the world population lives in urban areas (United Nations, 2019). Outdoor air pollution is a major global public health issue (Lelieveld et al., 2015), leading to 4.2 million premature deaths worldwide in 2016 (WHO, 2019). In cities, particulate matter with an aerodynamic diameter lower than 2.5 μm and 10 μm ($PM_{2.5}$ and PM_{10}), nitrogen dioxide (NO_2) and tropospheric ozone (O_3) are among the most threatening air pollutants in terms of harmful effects on human health associated with respiratory and cardiovascular diseases and mortality (Weinmayr et al., 2010; Pascal et al., 2013; Stafoggia et al., 2013; Cohen et al., 2017; Nuvolone et al., 2018). As a result, the legislated ambient air quality standards and the emission control policies around the world (e.g. World Health Organization Air Quality Guidelines, 2006; European Council Directive 2008/50/EC, Ministry of Environmental Protection, 2012; National Emission Ceilings Directive (2016/2284/EU); Convention on Long-range Transboundary Air Pollution, 2017) regulate air pollutants concentrations such as $PM_{2.5}$, PM_{10}, NO_2 and O_3, by setting limit and target values, long-term objectives, information thresholds, and alert threshold values for the protection of human health (Table 1).

In Europe, despite successful legislation implemented over several decades, and success in reducing air pollutants emissions, the current air pollution levels continue to cause important impacts on human health by exceeding the EU standards and the WHO Air Quality Guidelines for the protection of health (Guerreiro et al., 2014; De Marco et al., 2018; Sicard et al., 2019). In 2015, 47,300, 84,300 and 38,600 non-accidental premature deaths were attributed to air pollution ($PM_{2.5}$, NO_2, O_3) in France, Italy and Spain, respectively (EEA, 2018). In 2016, the total number of premature deaths for non-accidental causes attributed to $PM_{2.5}$ and PM_{10} (above 10 $\mu g.m^{-3}$) and O_3 (above 20 $\mu g.m^{-3}$) was 1800 in Rome and 144 in Nice (Sicard et al., 2019). Moreover, in a study of nine European cities including Rome and Valencia, the percentage increase in all deaths from natural causes per °C increase in air temperature tended to be greater during high O_3 days (Analitis et al., 2018), suggesting interactions with climate change. Air pollution in China has been a rising threat to human health (Liu et al., 2016; Feng et al., 2019) with annually about 2.5 million premature deaths attributed to air pollution (Lelieveld et al., 2015). Based on WHO metrics for human health protection, the O_3 levels led to 59,844 additional deaths in 2015 across China (Feng et al., 2019). In Wuhan, a total of 19,948 deaths were attributed to PM_{10}, NO_2 and SO_2 over the time period 2007–2009 (Ren et al., 2017).

Table 1
Air quality standards and limit values (in $\mu g.m^{-3}$) established by the European Commission in Europe (Directive 2008/50/EC) and the Ministry of Environmental Protection in China (2012), as well as the World Health Organization Air Quality Guidelines (2006) for the human health protection in urban areas.

	Averaging time	EC (2008)	MEP (2012)	WHO (2006)
$PM_{2.5}$	Annual mean	25	35	10
	24-hour mean	–	75	25
PM_{10}	Annual mean	40	70	20
	24-hour mean	50 (×35*)	150	50
NO_2	Annual mean	40	40	40
	1-hour mean	200 (×18)	200	200
O_3	Daily 8-hour maximum	120 (×25)	160	100

* Not to be exceeded >35 times a year.

Tropospheric O_3 formation occurs when NO_x and volatile organic compounds (VOCs) react in the atmosphere in the presence of sunlight (Seinfeld and Pandis, 1998). Despite control efforts, the rising O_3 levels become a major public health concern in cities worldwide (Paoletti et al., 2014; Sicard et al., 2018; Lefohn et al., 2018). The O_3 background levels are significantly rising at urban stations in Nice (+ 0.30 ppb per year), Rome (+ 0.49 ppb per year) and Valencia (+ 1.21 ppb per year) since 2005 (Sicard et al., 2013, 2018). In China, following the implementation of Air Quality Standards in 2012 (Ministry of Environmental Protection, 2012), a network of ground stations became operational in 2013. Some studies showed an O_3 increase in cities, e.g. maximum daily 8-hour average O_3 concentrations increased by on average 0.46 ppb per year in 74 cities across China from 2013 to 2017 (Liu et al., 2018; Liu and Wang, 2020). The ground-level O_3 is considered one of the most harmful air pollutants in terms of effects on human health (e.g. respiratory and cardiovascular systems), vegetation and materials (Mills et al., 2011; WHO, 2013; Sicard et al., 2016a, 2016b; Nuvolone et al., 2018; Sicard et al., 2019).

The surface O_3 concentrations tend to be higher on the weekends (Saturday and Sunday) compared to the weekdays (Monday to Friday) at urban sites, despite lower emissions of NO_x, VOCs and PM. This "weekend effect" has been widely studied worldwide (e.g. Qin et al., 2004; Jiménez et al., 2005; Blanchard et al., 2008; Schipa et al., 2009; Wolff et al., 2013; Adame et al., 2014; Shen et al., 2014; Diéguez et al., 2014; Xie et al., 2016; Zou et al., 2019; Sicard et al., 2020). Observations of how O_3 formation responds to emissions reduction provide an insight into the effectiveness of policies to plan future suitable strategies for reducing emissions of O_3 precursors (Karl et al., 2017).

Due to the coronavirus disease (COVID-19) pandemic, lockdown measures were implemented in China, Italy, Spain and France to limit social contacts and flatten the epidemic curve. Such measures were implemented from 23rd January 2020 in Wuhan, China, while in Italy, the measures were first applied in Northern regions (from 8th March 2020), where first COVID-19 cases were reported, before to be extended to the whole country from 10th March 2020. Tightened restrictive measures (i.e., close of schools and non-essential businesses, limitation of motorized transports) were fully implemented nationwide from 10th March 2020 in Italy, 14th March 2020 in Spain and 18th March 2020 in France, where schools were closed from 16th March 2020.

Plenty of newspapers and other mass media promoted that air pollution levels significantly dropped in major global cities since the lockdown measures, for instance NO_2 levels fell by up to 60% in cities relative to the same period in 2019 in the United Kingdom (BBC news, 8 April 2020) and $PM_{2.5}$ decreased by 60% in New Delhi from 23rd March to 13th April compared to the same period in 2019 (CNN, 23 April 2020). The NO_2 pollution over New York (USA) was 30% lower in March 2020, compared to the monthly average from 2015 to 2019 (The Conversation, 15 April 2020). Measurements performed by the European Space Agency (Sentinel-5P satellite) showed that between late January and early February 2020, NO_2 levels over cities in Asia and Europe were reduced by 40–50% compared to the same period in 2019 (ESA, 16th April 2020). In parallel with a reduction of the levels of some pollutants (e.g. black carbon, NO_2, PM), in Barcelona (Spain) it was also observed that O_3 increased (Tobías et al., 2020). Due to the complex chemistry of the atmosphere, there are still open questions regarding air pollution formation and spatiotemporal patterns during the lockdown.

Most of studies are based on the mean biases from the lockdown period (March–April 2020) compared to the same period in 2019, which are subject to the fluctuations in emissions and meteorological conditions. Here, we analyze short-term changes at 36 urban stations in Nice (France), Rome and Turin (Italy), Valencia (Spain) and Wuhan (China) from 1st January 2017 until 18th April 2020. Our aim was to detect and quantify the lockdown effect on the levels of the most health-threatening air pollutants in cities (NO_x, $PM_{2.5}$, PM_{10}, and O_3 in particular). We hypothesized that air pollution in the cities is mainly due to local urban emissions and that a 3-year time-series of data is long enough to more reliably detect short-term changes within time series, likely due to short-term emissions changes (e.g. PM, NO_x). We also expected an O_3 increase in cities due to a lower NO titration.

2. Materials and methods

2.1. Description of study areas

Five cities were selected to have a spatial representativeness and larger air pollution gradients for NO_2, $PM_{2.5}$, PM_{10} and O_3: Rome and Turin in Italy, Nice in France, Valencia in Spain, and Wuhan in China (Fig. 1).

The COVID-19 outbreak was firstly identified in Wuhan (Central China) in December 2019 (WHO, 2020). Wuhan, capital city of Hubei province, has a total area of 1530 km^2 and a population of 8.8 million; Wuhan is the ninth most populous city in China. The climate is humid subtropical with abundant rainfall in summer. The annual mean air temperature is 17.1 °C (from 4.0 °C in January to 29.1 °C in July, on average) with an annual mean rainfall of 1320 mm (China Meteorological Administration). The population in Nice (Southeastern France) is estimated at 345,000 inhabitants (INSEE, 2019) over 72 km^2 with an annual mean air temperature of 14.8 °C (8.0 °C in January −22.3 °C in July) and an annual mean rainfall of 811 mm. Rome, capital city of Italy, has a total area of 1285 km^2. The population in Rome is estimated at 2.9 million inhabitants (ISTAT, 2019). The Italian capital is characterized by a Mediterranean climate with an annual mean air temperature of 15.7 °C (from 7.7 °C in January to 24.4 °C in August) and the annual mean rainfall is 798 mm. The city of Turin, with 876,000 inhabitants over 130 km^2, is located in North-western Italy and experiences a warm temperate climate. The annual mean air temperature is 12.0 °C (from 2.0 °C in January to 22.4 °C in July) with an annual mean rainfall of 980 mm. Valencia is the third largest city in Spain, with around 800,000 inhabitants over 135 km^2. Valencia has a Mediterranean climate with short, very mild winters and long, hot and dry summers: with daily mean from 11.9 °C in January to 26.1 °C in August, and the annual mean rainfall is 475 mm (AEMET, 2020).

Nice, Rome, Turin and Valencia are located in the European region that is the most affected by air pollution, in particular by PM_{10} (Stafoggia et al., 2013) and O_3 (Sicard et al., 2013), due to high temperature, strong insolation, anticyclonic subsidence combined with high road traffic and industrial emissions (Millán et al., 2000). The highest hourly O_3 maxima (exceeding 120 ppb) are found in Southeastern France (Nice) and North-western Italy (Turin), in particular during summer (Sicard et al., 2013). In Italy, the highest PM_{10} concentrations are observed in Turin, where the air quality is among the worst in Europe, exceeding the limit values for PM_{10} and O_3 e.g. 86 and 61 days, respectively in 2019 (Forni et al., 2019). Wuhan is a major transport hub with dozens of railways, roads, and expressways passing through the city and connecting to major cities in China (Wang et al., 2017). In Wuhan, the NO_2 concentrations are increasing (+ 0.67 μg.m^{-3} per year) over the time period 2001–2014, while the PM_{10} concentrations showed a significant downward trend (− 2.0 μg.m^{-3} per year) since 2008 (Song et al., 2016). Nevertheless, the current annual PM_{10} (100–120 μg.m^{-3}) and NO_2 (50–60 μg.m^{-3}) mean concentrations are still quite high, exceeding the air quality standards (Qian et al., 2007; Song et al., 2016).

2.2. Data selection and methodology

The hourly NO, NO_2, $PM_{2.5}$, PM_{10} and O_3 concentrations were provided by the local and regional agencies in charge of air monitoring stations, i.e. the Certified Associations of Air Quality Monitoring in France (*Atmo Sud*), Regional Environmental Protection Agency in Italy (ARPA), Regional Ministry of the Environment of the *Generalitat Valenciana* in Spain, China's National Environmental Monitoring Center, and obtained from 1st January 2017 to 18th April 2020. A total of 36 monitoring stations with >75% of validated hourly data in a year were selected to calculate a valid aggregated value (24-h average concentration) and subsequent calculations. In Nice, data from 3 stations were used (3 with NO_2, NO, PM_{10} and $PM_{2.5}$, 2 with O_3). In Rome, 15 stations

Fig. 1. Location of the air quality monitoring stations in China (Wuhan), France (Nice), Italy (Turin and Rome) and Spain (Valencia).

were analyzed (15 with NO_2 and NO, 9 with O_3, 13 with PM_{10}, and 6 with $PM_{2.5}$). In Turin, 4 stations were selected (4 with NO_2 and NO, 3 with O_3 and PM_{10}, and 2 with $PM_{2.5}$). For Valencia, 6 stations were considered (6 with NO_2, NO and O_3, 4 with PM_{10} and $PM_{2.5}$), while 8 stations were used for Wuhan (with all pollutants except NO). Among the stations, 1 station in Nice, 5 in Rome, and 3 in Valencia were identified as being closer to roads with a higher traffic load than in the other stations. Besides the analysis per city of all stations, this subset of stations was also analyzed separately and referred to as "traffic".

To detect and estimate the changes within the time series, and quantify the lockdown effect on air pollutants levels, the deviations of 24-h mean concentrations (expressed in %) were computed for each day of the year (DOY) and station, by calculating the mean bias between the period before the lockdown (from 1st January 2020 until the start date of the lockdown) and during the lockdown in 2020 (from start date of the lockdown until 8th April in Wuhan i.e. the end date of the lockdown, and until 18th April in Nice, Turin, Rome and Valencia where the lockdown was still running at the time of data collection) and the same time period averaged over the 3 previous years (2017–2019), representing the baseline conditions. For each city, the mean NO, NO_2, $PM_{2.5}$, PM_{10} and O_3 concentrations during the lockdown period in 2020 were calculated and compared with the mean concentrations during weekdays and weekends of the equivalent time period averaged over the 3 previous years (2017–2019). The objective was to estimate how different was a long and substantial reduction in activity in comparison with the reduced activity typical of the weekends. The non-parametric Kruskal-Wallis test followed by a post-hoc test using the criterion Fisher's Least Significant Difference and p adjusted with the Holm correction was used to test for statistical significances between groups. A p-value <0.05 was considered statistically significant.

3. Results

By averaging all stations, the seasonality of daily O_3, NO_2, NO, $PM_{2.5}$, PM_{10} mean concentrations differed among cities from 1st January to 31st December averaged over the time period 2017–2019 (Figs. S1–S5). The highest O_3 mean concentrations, reaching 90 μg.m^{-3} in summer, were observed in Turin and Wuhan following a bell-shaped function, while lower mean concentrations were recorded in Nice, Rome and Valencia (Fig. S1). The seasonal variations observed for NO and NO_2 consisted of a winter peak and summer minima (Figs. S2, S3) with a marked seasonality and higher concentrations in both highly industrialized cities (Turin and Wuhan) and lower levels in Nice and Valencia. This enhanced seasonality of NO_x levels in winter may be partly attributed to increased fossil fuels for domestic heating and driving. The PM levels were higher in Wuhan (PM_{10} up to 100 μg.m^{-3} in winter) than in the European cities, although values were also high in Turin, up to 50 μg.m^{-3} in winter (Figs. S4, S5). No remarkable seasonality of PM levels was observed in Nice, Rome and Valencia.

From 1st January to 18th April 2020, the highest O_3 mean concentrations were observed in Wuhan (54.1 μg.m^{-3}), followed by Nice (50.4 μg.m^{-3}), while the lowest mean concentrations (37.3 μg.m^{-3}) were recorded in Turin (Fig. 2). The highest NO_2 concentrations were recorded in Turin (40.8 μg.m^{-3}) and the lowest in Valencia (21.5 μg.m^{-3}). The NO ranged from 10.6 μg.m^{-3} in Nice to 25.6 μg.m^{-3} in Turin (Fig. S6), while the NO data were not available from Wuhan. The average values of $PM_{2.5}$ (Fig. 2) and PM_{10} (Fig. S7) were 43.1 μg.m^{-3} and 56.1 μg.m^{-3} in Wuhan, respectively, and 31.1 μg.m^{-3} and 42.9 μg.m^{-3} in Turin, respectively. The lowest mean values were recorded in Valencia, with concentrations of 11.3 μg.m^{-3} and 21.1 μg.m^{-3} of $PM_{2.5}$ and PM_{10}, respectively. When traffic stations were analyzed separately, they showed higher NO_2 and NO concentrations and somewhat lower O_3 concentrations. For example, in Valencia, NO_2 was 59% higher in traffic stations, NO 28% and O_3 5% (data not shown).

In 2020, the daily O_3 mean concentrations from 1st January 2020 until the start date of the lockdown were similar to the same period averaged over 2017–2019 (Table S1a) in Nice (+ 2.0%), Rome (−9.0%) and Wuhan (+4.2%), while the O_3 concentrations were much lower in Valencia (−18.7%) due to cloudy and rainy conditions (AEMET, 2020), and much higher in Turin (+53.3%) due to high air temperature (5–10 °C above the 1971–2000 baseline) and no rainfall in February (ARPA, 2020). In Valencia, the O_3 levels at traffic stations before the lockdown were lower (−13.1%) than the baseline conditions (Table S1b). Compared to 2017–2019, the daily O_3 mean concentrations clearly increased at all stations during the lockdown (Fig. 3): +24.0% in Nice, +13.6% in Rome, +27.0% in Turin, +2.4% in Valencia and +36.4% in Wuhan. In a context of cloudy and rainy conditions before and during the lockdown in Valencia (AEMET, 2020), the response to lockdown measures was amplified at traffic stations (+ 11.4%).

The mean bias of daily NO_2 concentrations before the lockdown at all stations were: - 14.2% in Nice, +1.6% in Rome, +2.6% in Turin, −20.0% in Valencia and −21.6% in Wuhan (Table S1a). During the lockdown, the changes in daily O_3 mean concentrations at all stations were associated with a strong decline in NO_2 mean concentrations compared to baseline conditions (Fig. 3): −62.8%, −45.6%, −30.4%, −69.0% and −57.2% in Nice, Rome, Turin, Valencia and Wuhan, while NO declined by 70.7%, 68.5%, 52.6% and by 61.9% in Nice, Rome, Turin and Valencia, respectively. For both NO and NO_2, stronger reductions were observed during lockdown at traffic stations: −88.1% and −68.9% in Nice, −70.5% and −55.1% in Rome, −75.5% and −70.6% in Valencia, respectively (Table S1b).

At all stations, compared to baseline conditions 2017–2019, PM_{10} concentrations decreased during the lockdown (Fig. 3) by 5.9% in Nice (−7.8% before lockdown), 8.9% in Turin (+9.9%), 32.1% in Valencia (+14.5%) and 48.7% in Wuhan (−31.7%) while PM_{10} slightly increased by 1.8% in Rome (+18.5% before lockdown). Looking at $PM_{2.5}$ we found: −2.9% (−19.0%), +10.6% (+22.6%), −12.6% (+8.1%), −12.6% (+24.3%) and −36.3% (−34.2%) in Nice, Rome, Turin, Valencia and Wuhan, respectively (Table S1a). The lockdown measures had a greater effect at traffic stations: PM_{10} and $PM_{2.5}$ levels decreased by 7.6% and 8.0% in Nice, by −3.0% and −1.5% in Rome, and by 51.3% and 29.3% in Valencia (Table S1b).

By comparing mean concentrations during the 2020 lockdown with mean concentrations during weekdays and weekends of the equivalent time period over 2017–2019, we showed that higher O_3 concentrations occurred during the lockdown, except in Valencia by combining all stations (Fig. 4). At all stations, the relative effect of lockdown relative to weekdays was 29.1% in Nice, i.e. the O_3 mean concentration during the lockdown was 29.1% higher than on weekdays, 14.6% in Rome, 26.5% in Turin, 4.4% in Valencia and 35.1% in Wuhan (Table S2a). During the lockdown, the O_3 levels were 9.8% higher than during the weekends in Nice, 7.1% in Rome, 24.8% in Turin, 37.7% in Wuhan, and 4.2% lower in Valencia. However, at traffic stations in Valencia, the O_3 levels during the lockdown were 14.1% higher than on weekdays and 3.8% higher than on weekends (Table S2b). In all cities, the lowest mean NO_2 concentrations were observed during the lockdown (Fig. 4). At all stations, the lockdown NO_2 levels were 65.7% lower than on weekdays in Nice, 50.3% in Rome, 32.5% in Turin, 71.6% in Valencia and 56.8% in Wuhan. During the lockdown, the NO_2 concentrations were 54.7% lower than on weekends in Nice, 42.2% in Rome, 20.6% in Turin, 62.9% in Valencia and 57.0% in Wuhan (Table S2a). At traffic stations, the NO_2 levels during the lockdown were 70.9% lower than on weekdays in Nice, 57.5% in Rome and 72.8% in Valencia and 61.9% lower than on weekends in Nice, 51.3% in Rome and 64.6% in Valencia. Similar observations were found for NO (Table S2). The highest levels of PM_{10} were observed during the weekdays in Nice, Rome, Turin, and Valencia and during the weekends in Wuhan (Fig. 4). For PM_{10}, the differences in daily concentrations between weekends and lockdown were <1 μg.m^{-3} in Nice, Rome and Turin (Table S2a). At all stations, the lockdown-weekday PM_{10} difference was - 7.3% in Nice, −0.4% in Rome, −13.0% in Turin, −41.1% in Valencia and −47.5% in Wuhan. The lockdown-weekend PM_{10} difference was of - 3.6% in Nice, +3.7% in Rome, −0.4% in Turin, −35.6% in

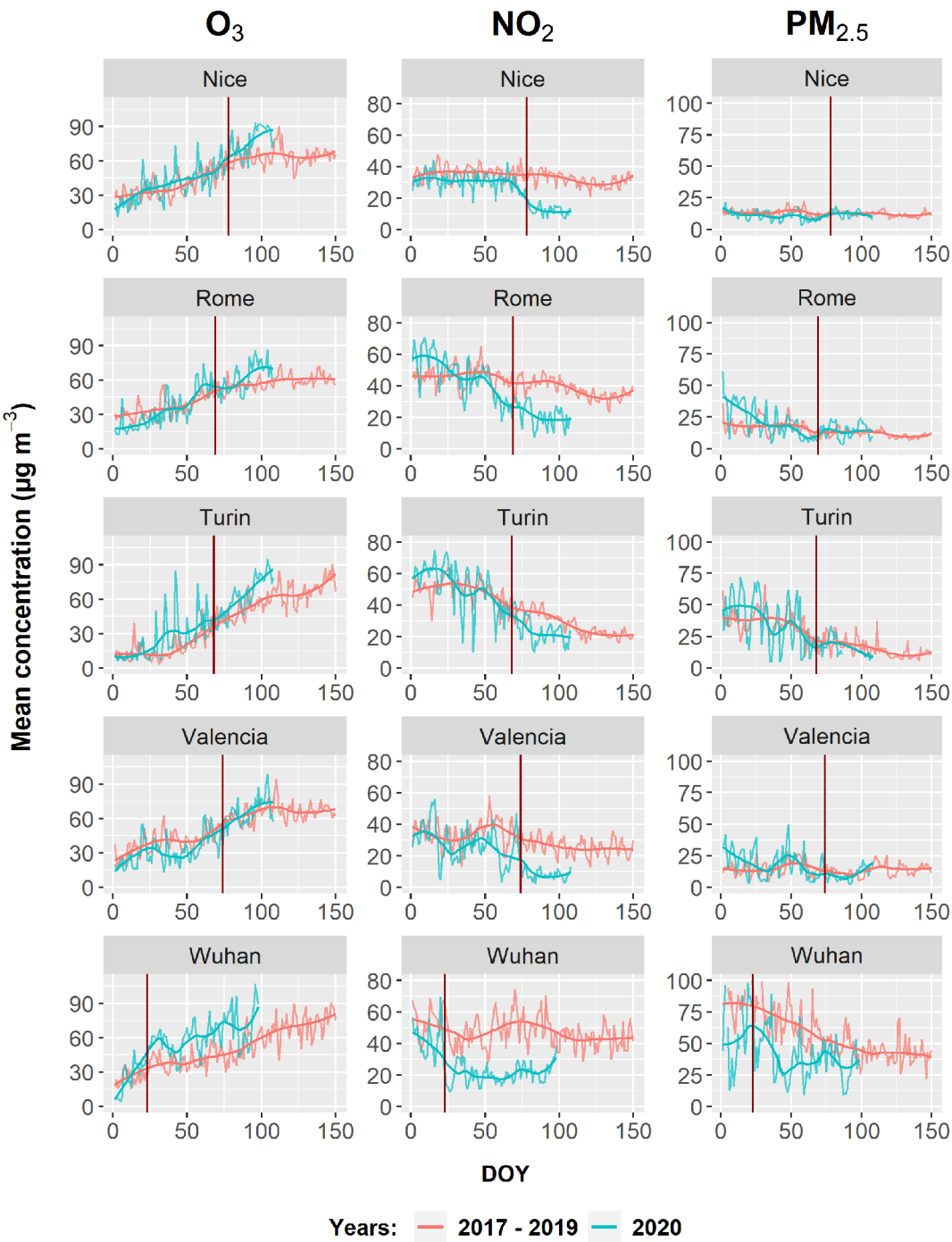

Fig. 2. Daily ozone (O_3), nitrogen dioxide (NO_2) and particulate matter with an aerodynamic diameter lower than 2.5 µm ($PM_{2.5}$) mean concentrations (µg.m^{-3}) by joining all stations in Nice, Rome, Turin, Valencia and Wuhan from 1st January (Day of the Year, DOY = 1) to DOY = 150, averaged over the 3 previous years (2017–2019), and from DOY = 1 to the end date of the analyzed period in Europe (DOY = 109) and the lockdown in Wuhan (DOY = 98) in 2020. Vertical line: start date of the lockdown in 2020. Smoothing line: locally weighted smoother (LOESS).

Valencia and −49.5% in Wuhan. At traffic stations, the PM_{10} levels during the lockdown were 7.2% lower than on weekdays in Nice, 4.3% in Rome and 53.3% in Valencia, and 6.9% lower than on weekends in Nice, 0.7% in Rome and 49.5% in Valencia (Table S2b). Similar observations were found for $PM_{2.5}$ (Table S2). Despite the overall consistency in the observed changes in all cities for the different air pollutants, at city level, some differences were statistically significant and others not due to the variability between stations, with the differences being more pronounced at traffic stations (Fig. 4).

4. Discussion

Urban air pollution levels are mainly influenced by local emissions and chemical mechanisms (Kent et al., 2007; Huszar et al., 2015; Monks et al., 2015). For instance, the local photochemical formation accounted for 75% of the daytime O_3 in Wuhan in summer 2016 (Zeng et al., 2018). In this study, we considered that 3-year baseline conditions were long enough to reduce inter-annual variability in air pollution levels, here more influenced by local short-term emissions reduction

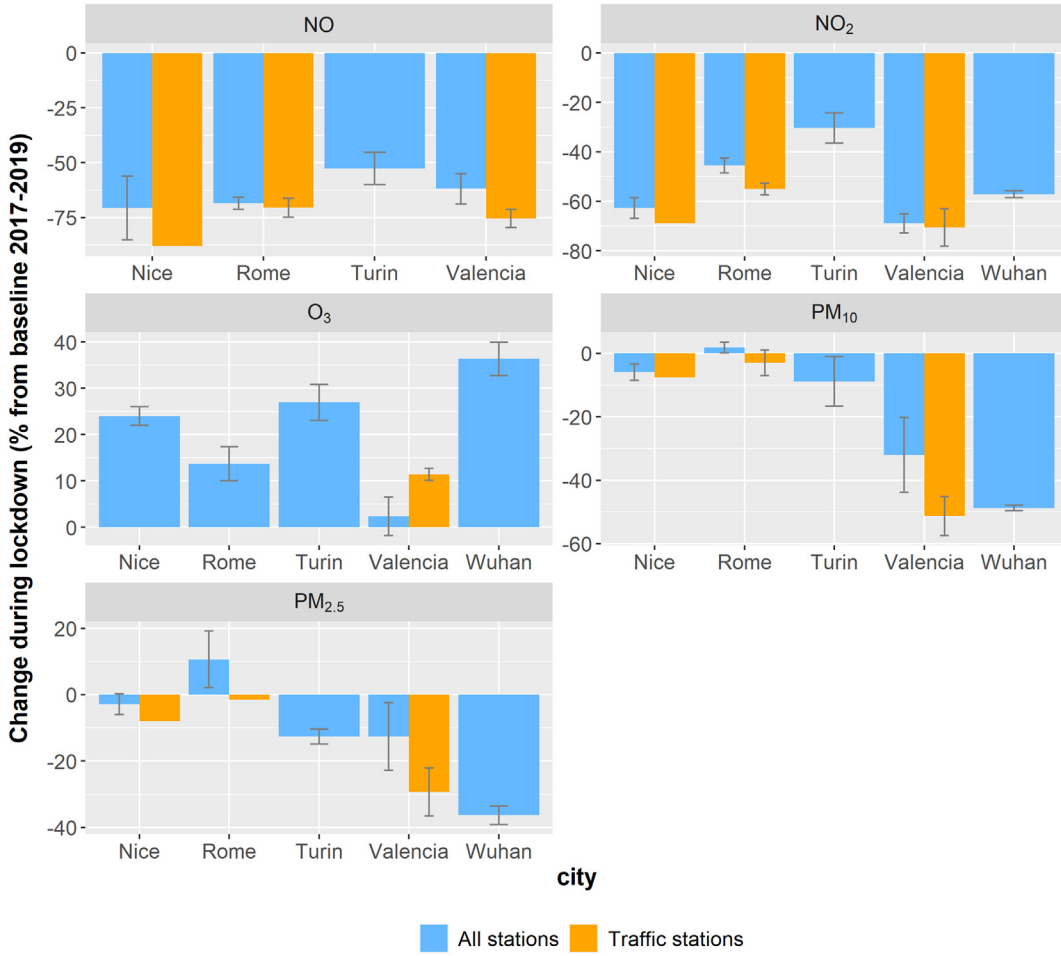

Fig. 3. Mean bias (±standard error, in %) at city-scale of 24-hour mean concentrations ($PM_{2.5}$, PM_{10}, NO, NO_2 and O_3) at all stations and traffic stations in Nice, Rome, Turin, Valencia and Wuhan between the lockdown period in 2020 and the same time period averaged over the 3 previous years (2017–2019).

rather than meteorological variations. By investigating the lockdown effects on air quality in cities, we have a better understanding of contributions from sectoral emissions to air pollution, in particular the O_3 production, in order to formulate effective emission control policies. Among the dominant sectors contributing to air pollution in urban areas, the sectors "transport", "industrial processes" and "commercial, institutional and households" were strongly impacted by the lockdown measures in China, France, Italy and Spain.

In Europe, the "transport" sector is the largest contributor to NO_x emissions (road transport: 39%, non-road transport: 8%) and represents 13% of $PM_{2.5}$ and PM_{10} emissions (EEA, 2019). The fuel and biomass combustion in the "commercial, institutional and households" (domestic heating) sector is the largest contributor to $PM_{2.5}$ and PM_{10} emissions (56% and 39%, respectively) and represents 14% of NO_x emissions, while 46% of non-methane VOCs and 8% of NO_x are emitted by "industrial processes" sector. In Wuhan, the main source of air pollution recently shifted from coal combustion to a mixture of coal combustion and road traffic emissions (Wang et al., 2017) where the combustion of fossil fuel and road traffic are the largest contributors to $PM_{2.5}$ and NO_2 emissions, respectively (Wang et al., 2017).

The lockdown measures led to a decrease of NO (~63%) and NO_2 (~53%) concentrations in Nice, Rome, Valencia and Wuhan, while NO and NO_2 declined by 53% and 30% in the highly industrialized city of Turin. The lower reduction in Turin can be explained by a higher contribution of the "industrial" sector, where essential industrial activities (e.g. food, pharmaceutical) continued during the lockdown. Stronger reductions were observed at traffic stations for NO (~78%) and NO_2 (~65%), where the "road transport" sector is the largest contributor to NO_x emissions. The magnitude of changes in NO_2 levels was similar between European cities (~52%) and Wuhan in China (57%), where automobile exhausts are the major source of NO_x (Wang et al., 2017). As road and non-road transport were drastically reduced, the lockdown effect on NO_x reduction was much higher than the weekend effect, with NO_x concentrations during the lockdown on average 49% lower than on weekends at all stations, and 60% lower at traffic stations.

In Southern Europe, the lockdown measures did not significantly impact the $PM_{2.5}$ and PM_{10} levels at all stations. Indeed, $PM_{2.5}$ and PM_{10} concentrations decreased in Nice (3% and 6%) and Turin (13% and 9%) and increased in Rome (11% and 2%). Stronger reductions were observed in Valencia (13% and 32%). At traffic stations, the lockdown measures strongly decreased both $PM_{2.5}$ and PM_{10} levels in Nice (8%), Rome (1–3%) and Valencia (29–51%). The observed decreases were due to the reduction of road and non-road transport, representing up to 15% of wintertime PM levels (Karamchandani et al., 2017), and the reduction of fuel combustion in closed institutional and commercial buildings. However, this decrease was counter-balanced by a PM increase from domestic heating ("requiring people to stay at home") and garden activities (e.g. biomass burning). In Nice, and surrounding cities, the local authorities issued a regulation to drastically reduce the green waste burning. By comparing both the lockdown and weekend effects on PM levels at all stations, we showed that the lockdown had the same effect as weekends in Nice, Rome and Turin (~6% of difference). Such slight difference suggests that road traffic was not a large contributor to PM emissions but derived more from residential and tertiary sector in these cities. The citizens behave similarly during lockdown and weekends. The strong reductions in particles at traffic stations observed in

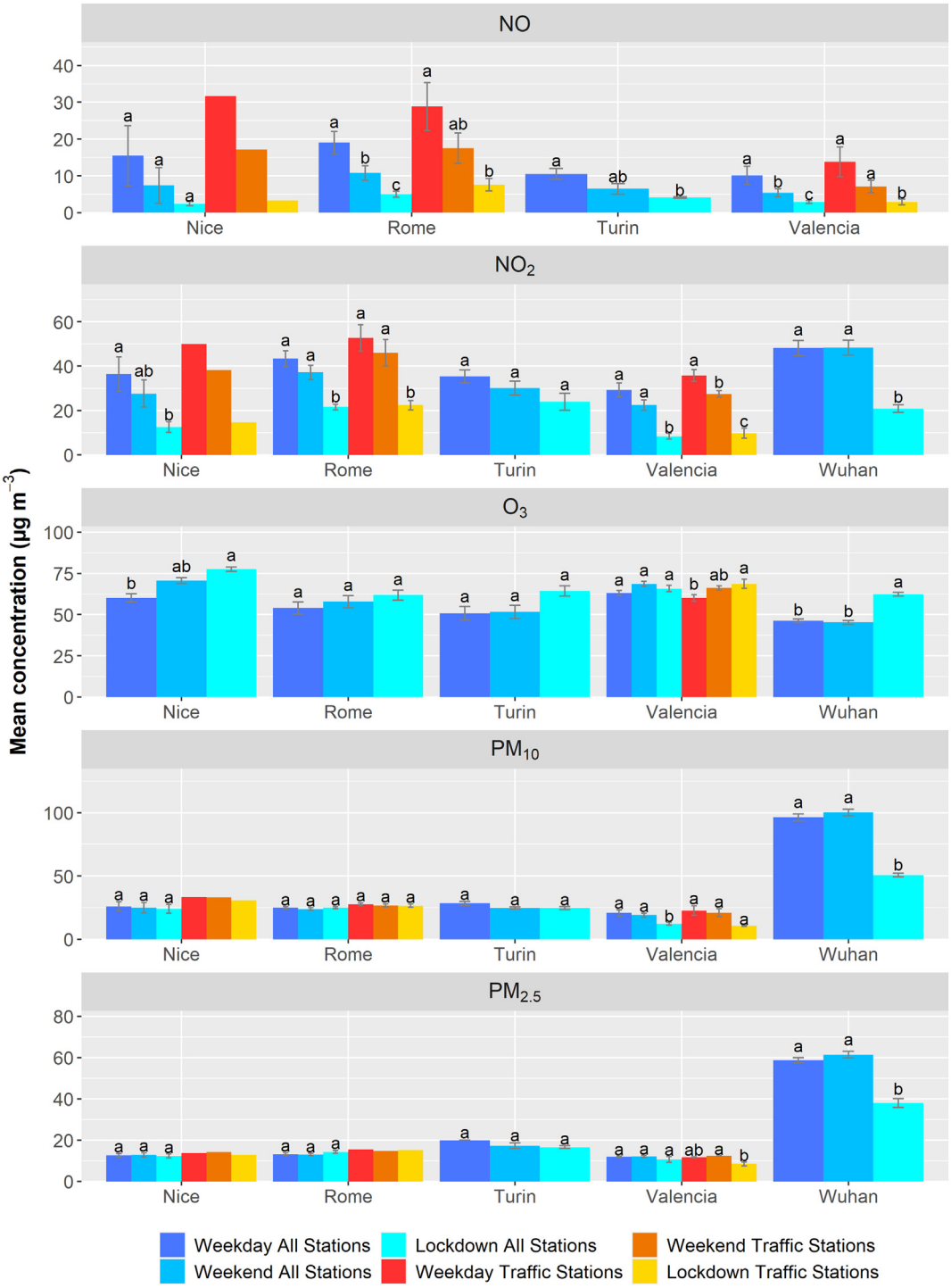

Fig. 4. Mean concentrations (±standard error, in µg.m^{-3}) by joining all stations at city-scale of 24-hour mean concentrations ($PM_{2.5}$, PM_{10}, NO, NO_2 and O_3) in Nice, Rome, Turin, Valencia and Wuhan between the lockdown period in 2020 and the weekday and weekend of the equivalent time period averaged over the 3 previous years (2017–2019). Different letters represent significant differences between groups per city (Kruskal-Wallis test, *p*-value <0.05 significant).

Valencia during the lockdown resulted from a combination of factors: i) a higher baseline due to landscaping works in one of the traffic stations in previous years and ii) several rainy days (wash-out effect) during the lockdown period. A higher contribution of "transport" (road transport: 18%; non-road transport: 21%) to PM emissions, as reported in Barcelona (Karamchandani et al., 2017), could also partly explain the largest reduction observed in Valencia. In Wuhan, the $PM_{2.5}$ and PM_{10} decreases by 36% and 49% were higher than those observed in Southern Europe (~4% and ~ 11%, respectively). In Wuhan, the emissions from household heating and cooking activities (ab. 32%), coal consumption and heavy industries harboring iron and steel smelts (accounted for 34% of secondary PM and 57% of primary dust) were the largest contributors to $PM_{2.5}$ and PM_{10} emissions compared with ab. 5% from the "transport" sector (Qian et al., 2007; Wang et al., 2017). The lockdown had a higher effect than the weekends on PM reduction in Wuhan (~44% less), thus, the strong reduction can be attributed to the reduction of coal combustion in the tertiary sector and cessation of industrial activities.

During the lockdown 2020, the surface O_3 levels increased by 24–27% in Nice and Turin, by 14% in Rome and by 36% in Wuhan. The

slight increase of O_3 levels in Valencia (2.4%) was mainly due to rainy and cloudy conditions (AEMET, 2020). In a recent paper, Tobías et al. (2020) reported an increase in O_3 of 29% at urban stations of Barcelona between February 16th to March 13th and March 14th to 30th, 2020. At city-scale, the O_3 formation depends on the VOC-NOx ratio (Pusede and Cohen, 2012). The urban areas are characterized by a low ratio due to high NO_x concentrations (Beekmann and Vautard, 2010). The local O_3 formation is generally limited by VOCs in Wuhan (Zeng et al., 2018) and in Southern Europe (Anav et al., 2019; Sicard et al., 2020). In this case, i.e. with "VOC-limited" conditions, a reduction in VOCs emission reduces the O_3 formation, but a reduction in NO_x emission increases the O_3 formation. To effectively control O_3 pollution in Wuhan, the reduction ratio of VOCs to NO_x concentrations should not be lower than 0.73 (Zeng et al., 2018). The implementation of stringent lockdown measures produced more reduced NO_x emissions than VOCs emissions in the investigated cities, leading to higher VOC-NO_x ratio, which enhanced the O_3 production. During the lockdown, an increase in O_3 precursors emissions such as carbon monoxide (CO) and VOCs from home (e.g. cleaning, fireplaces) and garden activities (e.g. barbeques, biomass burning) may also have contributed to the O_3 increase (Coe et al., 2003; Su et al., 2003; Murphy et al., 2007; Wolff et al., 2013).

In cities, the freshly emitted NO, in particular from road traffic, depletes O_3 locally (Solberg et al., 2005; Molina et al., 2009). The O_3 titration occurs particularly in winter (less photolysis reactions of NO_2) under high NO_x levels (Sillman, 1999). Following the lockdown measures, the clear upward trend observed at all stations, resulted primarily from a lower titration of O_3 by NO due to the reduction in local NO_x emissions by road transport (e.g. Huszar et al., 2015; Sicard et al., 2016a). In different European cities, the relative contribution of road traffic emissions to O_3 levels was 12–35% and 20–24% in three Mediterranean cities: Lisbon, Barcelona and Athens (Valverde et al., 2016; Karamchandani et al., 2017; Mertens et al., 2019). In Wuhan, the vehicle exhausts made the largest contribution to O_3 production, with 30% during non-high O_3 days (Zeng et al., 2018). These findings are in agreement with the rate of O_3 increase observed due to the lockdown measures.

Furthermore, a reduction in $PM_{2.5}$ and PM_{10} could also lead to an increase in surface O_3 concentrations (Liu et al., 2013; Li et al., 2017). As PM emissions were lower during the lockdown, the higher solar radiation favored O_3 formation (Heuss et al., 2003; Murphy et al., 2007; Wolff et al., 2013). In addition to photochemical reactions, the heterogeneous chemical processes occurring on the surface of $PM_{2.5}$ and aerosols in the atmosphere are also an important way for the interaction between O_3 and $PM_{2.5}$ (Meng et al., 1997; Jacob, 2000; Deng et al., 2010; Li et al., 2011). In Nanjing, high concentrations of $PM_{2.5}$ (rising from 100 to 250 $\mu g.m^{-3}$) resulted in a reduction of 130 $W.m^{-2}$ of the irradiance and a 12% reduction of near-surface O_3 (Li et al., 2017).

In urban stations in France, Italy and Spain, the mean O_3 concentration on the weekend was 12% higher than on weekdays over the time period 2005–2014 (Sicard et al., 2020). The O_3 weekend effect is more pronounced in winter (Sicard et al., 2020). In this study, we showed that the mean O_3 concentrations during the lockdown were on average 10% and 38% higher than on weekend in Southern Europe and Wuhan, respectively. Generally, the lockdown effect on O_3 production was higher than O_3 weekend effect, mainly due to the longer period of NO_x reduction. Furthermore, the average lifetime of O_3 in the troposphere is estimated at 20–24 days (Stevenson et al., 2006).

5. Conclusions

Following the implementation of stringent lockdown measures in the framework of the COVID-19 pandemic, the reduction in road and non-road transport, non-essential businesses and industrial activities led to significant declines in NO_x and PM concentrations, especially in Wuhan. The lockdown measures led to a reduction of NO_x concentrations of ~56% due to the large reduction of the "transport" sector (e.g. 70% in France). During the lockdown, the PM levels slightly changed (<10% of change). Indeed, the restrictive measures reduced emissions of $PM_{2.5}$ and PM_{10} by road and non-road transport and by fuel combustion in institutional and commercial buildings, but these decreases were counter-balanced by an increase of PM emissions from the activities at home (e.g. domestic heating, biomass burning). Due to stringent lockdown measures, generally the near-surface O_3 increased by ~17% in Southern European cities and by 36% in Wuhan, i.e. similarly to the relative contribution of road traffic emissions to O_3 levels. The O_3 increase is due to a lower titration of O_3 by NO due to the strong reduction in local NO_x emissions by road transport. Overall, the largest effect of the lockdown measures on concentrations of NO_x, PM and O_3 came from the large reduction in road transport, as observed at traffic stations. The NO_x reduction during the lockdown was higher than the VOCs reduction. Similarly to the O_3 weekend effect, the main causes of the higher O_3 concentrations in cities during the lockdown, under VOC-limited conditions, are: i) a reduction in NO_x emissions from road traffic leading to a lower O_3 titration by NO (dominant cause); ii) as PM emissions were lower, the higher solar radiation favored O_3 formation; and iii) an increase of O_3 precursors emissions from home and garden activities.

In Southern Europe and Wuhan, NO_x concentrations during the lockdown were on average 49% lower than on weekends. The lockdown effect on O_3 production was 10% higher than the O_3 weekend effect in Southern Europe and 38% higher in Wuhan. The lockdown did not lead to lower PM pollution than a « routine » weekend effect in Southern Europe (~6% of change). The unprecedented reduction in mobility and economic activity caused by the COVID-19 lockdown represents an exceptional opportunity for studying the contribution of different sources of primary pollutant and for understanding the changes in the atmospheric chemistry under conditions of reduced primary pollutant emissions in the cities. For secondary pollutants like O_3, the lockdown has also shown that its reduction will remain challenging even with effective policies for reducing primary pollutants.

CRediT authorship contribution statement

Pierre Sicard: Conceptualization, Investigation, Writing - original draft. **Alessandra De Marco:** Investigation, Writing - review & editing. **Evgenios Agathokleous:** Writing - review & editing. **Zhaozhong Feng:** Investigation, Writing - review & editing. **Xiaobin Xu:** Investigation, Writing - review & editing. **Elena Paoletti:** Writing - review & editing. **José Jaime Diéguez Rodriguez:** Investigation, Writing - review & editing. **Vicent Calatayud:** Conceptualization, Investigation, Data curation, Writing - original draft.

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.

Acknowledgements

In the framework of the Research Group 8.04.00 "*Air Pollution and Climate Change*" and the Working Party 8.04.05 "Ground-level ozone" of the International Union of Forest Research Organizations (IUFRO), this work was made possible with the access to the air quality database of the Certified Associations of Air Quality Monitoring in France (*Atmo Sud*), Regional Environmental Protection Agency in Italy (ARPA), Regional Ministry of the Environment of the *Generalitat Valenciana* in Spain, China's National Environmental Monitoring Center. E.P. acknowledges Sofia Martini (CNR) and Roberta De Maria (ARPA) for data provision, and the INTERREG ALCOTRA project MITIMPACT for support. E.A. and Z.F. acknowledge multi-year support from *The Start-up Foundation for Introducing Talent of Nanjing University of Information Science &*

Technology (NUIST), Nanjing, China (No. 003080 to E.A. and No. 002992 to Z.F.). V.C. thanks Project IMAGINA (PROMETEU 2019, Generalitat Valenciana).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.139542.

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