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A 10-year assessment of ambient fine particles and related health endpoints in a large Mediterranean city

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HIGHLIGHTS

- A 10-year assessment of $PM_{2.5}$-related mortality and morbidity in Marseille.
- Significant reduction of $PM_{2.5}$: 0.80 μg m^{-3} $year^{-1}$.
- Exposure to $PM_{2.5}$ led to 871 deaths and 1868 hospital admissions in 2010–2019.
- Number of $PM_{2.5}$-related deaths decreased by 1.15 per 10^5 people annually.
- COVID-19 pandemic led to 2.6 $PM_{2.5}$-related deaths averted in 2020.

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ABSTRACT

Fine particles i.e., with an aerodynamic diameter lower than 2.5 μm ($PM_{2.5}$) have potentially the most significant effects on human health compared to other air pollutants. The main objectives of this study were to i) investigate the temporal variations of ambient $PM_{2.5}$ in Marseille (Southern France), where air pollution is again a major public health issue, and ii) estimate their short-term health effects and annual trend (Mann-Kendall test) over a 10-year period from 2010 to 2019. In Marseille, the main sources of $PM_{2.5}$ could be related to road traffic, industrial complexes, and oil refineries surrounded the city. The number of premature deaths and hospital admissions attributable to ambient $PM_{2.5}$ exposure for non-accidental causes, cardiovascular and respiratory diseases were estimated by using in-situ air quality data, city-specific relative risk values and baseline incidence. Despite significant reduction of $PM_{2.5}$ (-0.80 μg m^{-3} $year^{-1}$), Marseille citizens were exposed to $PM_{2.5}$ levels exceeding the World Health Organization (WHO) Air Quality Guideline for human health protection (10 μg m^{-3}) during entire study period. Exposure to ambient $PM_{2.5}$ substantially contributed to mortality and hospital admissions: 871 deaths for non-accidental causes, 515 deaths for cardiovascular diseases, 47 deaths for respiratory diseases, as well as 1034 hospital admissions for cardiovascular diseases and 834 for respiratory diseases were reported between 2010 and 2019. Compliance with WHO annual limit values can result in substantial socio-economic benefits by preventing premature deaths and hospital admissions. For instance, based on the value of a statistical life and average cost of a hospital admission, the associated benefit for healthcare would have been €131 million in 2019. Between 2010 and 2019, the number of $PM_{2.5}$-related non-accidental deaths decreased by 1.15 per 10^5 inhabitants annually. Compared to 2010–2019, the restrictive measures associated to COVID-19 pandemic led to a reduction in $PM_{2.5}$ of 11% in Marseille, with 2.6 $PM_{2.5}$-related deaths averted in 2020.

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1. Introduction

Air pollution is a major public health issue in the world (e.g., De Marco et al., 2018; Sicard et al., 2019; Rovira et al., 2020; Zahro and Sulistyorini, 2020), leading to 4.2 million premature deaths worldwide (WHO, 2016) and half a million in the European Union (EU) in 2015 (EEA, 2018). Industrial activities, motor vehicle emissions, transport, domestic heating, and dust are different sources of air pollution in urban environment (EEA, 2019; Sicard et al., 2021).

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These sources release a wide range of air pollutants such as nitrogen oxides (NO_x), volatile organic compounds (VOCs), sulfur dioxide (SO_2), black carbon, polycyclic aromatic hydrocarbon (PAH) and particulate matter (PM) in the atmosphere (EEA, 2019). PM can be released by biological and non-biological sources, or from chemical reactions in the air (Martins et al., 2016; Amoatey et al., 2019a). Atmospheric PM can be classified into different size ranges: particles with an aerodynamic diameter lower than 10 μm (PM_{10} or coarse particles), lower than 2.5 μm ($PM_{2.5}$ or fine particles), lower than 0.1 μm ($PM_{0.1}$ or ultrafine particles), and lower than 0.05 μm or nanoparticles (Clifford et al., 2018). Like most European cities, fuel and biomass combustion in "commercial, institutional and households" sector (e.g., domestic heating) in Marseille was the largest contributor to $PM_{2.5}$ and PM_{10} emissions (56% and 39%, respectively), while maritime transport contributes to 4–5% of PM emissions (Air PACA, 2017). In addition, high PM_{10} and $PM_{2.5}$ concentrations can be recorded in Mediterranean cities during Saharan dust storm (Querol et al., 2019).

Epidemiological studies have reported an association between ambient PM exposure and mortality and hospital admissions (morbidity) for respiratory and cardiovascular diseases (e.g., Analitis et al., 2006; Bell et al., 2008; Sun et al., 2010; Langrish et al., 2012; Mannucci et al., 2019; Yang et al., 2020). In European Union cities, where 74% of the population lives (EU, 2016), $PM_{2.5}$ has potentially the most significant health effects, in terms of mortality and morbidity, compared to other common air pollutants (Pascal et al., 2014; Cohen et al., 2017) as they deeply penetrate the airways system (Pascal et al., 2014; Clifford et al., 2018). The European Union (EU) and the World Health Organization (WHO) regulate the concentrations of air pollutants such as $PM_{2.5}$ by setting limit and target values for the protection of human health (e.g., Directive, 2008/50/EC, WHO Air Quality Guidelines 2006). The annual mean $PM_{2.5}$ concentration does not to exceed 25 μg m^{-3} (EU limit value) or 10 μg m^{-3} (WHO, 2006 Air Quality Guidelines), and the 24-h $PM_{2.5}$ mean concentration does not to exceed 25 μg m^{-3} (WHO, 2006 Air Quality Guidelines).

Despite reductions of PM emissions in EU over the time period 2000–2017 (~30%), 74–81% of the urban population was exposed to $PM_{2.5}$ levels exceeding the WHO limit values for the protection of human health in 2015–2017 (Sicard et al., 2021). In 2016, 374,000 non-accidental premature deaths were attributed to $PM_{2.5}$ in the EU (EEA, 2018). The health risk assessment of air pollution is widely investigated worldwide e.g., in Asia (Liu et al., 2019; Qi et al., 2020; Yang et al., 2020; Zhang et al., 2020), Iran (Hadei et al., 2017; Daryanoosh et al., 2017; 2018; Khaniabadi et al., 2017a; b; c; d, e, 2018, 2019a,b; Karimi et al., 2019; Manojkumar et al., 2020; Zallaghi et al., 2020), and Europe (Fattore et al., 2011; De Marco et al., 2018; Sicard et al., 2019; Todorović et al., 2019; Amoatey et al. 2019b, 2020). However, most of published studies focused on the health impacts of exposure to ambient PM_{10} over short time period (e.g., Pascal et al., 2014; De Marco et al., 2018).

For the first time, a 10-year assessment focused on a Mediterranean large city in Southern France (Marseille as study case) where air pollution is a major public health issue (Salameh et al., 2018) to: i) investigate the temporal variability of $PM_{2.5}$ and its relationships with most common air pollutants in cities (i.e., PM_{10}, NO_2 and ozone); ii) estimate the number of premature deaths and hospital admissions for cardiovascular and respiratory diseases attributed to ambient $PM_{2.5}$ exposure; iii) quantify the annual trends in $PM_{2.5}$-related mortality and morbidity over the time period 2010–2019; and iv) analyze the effect of COVID-19 pandemic on $PM_{2.5}$ levels and $PM_{2.5}$-related deaths and hospital admissions in 2020.

2. Materials and methods

2.1. Study area

The Southern France, experiencing a Mediterranean climate, is among the most affected regions by air pollution in Europe (Sicard et al., 2019). Marseille (43°17′N, 5°22′E) is an urban-industrial city in Southern France (Fig. 1) close to the biggest industrial area in Southern Europe, namely Berre Pond, surrounded by major industrial complexes and oil refineries (Salameh et al., 2018). Marseille is the second largest city in France (after Paris), with a population ranging from 850,700 inhabitants in 2010 to 868,300 in 2020. Marseille is the second most polluted city with regard to $PM_{2.5}$ in France. Air pollution is a major public health issue, mainly due to industrial development, road traffic increment, high insolation and sea/land breeze recirculation (Sicard et al., 2013; Salameh et al., 2018), including tropospheric ozone (O_3) pollution with significant increases in cities in Southern France (on average + 0.31 ppb $year^{-1}$) since the early 2000s (Sicard, 2021). Furthermore, the Mediterranean Basin, in particular Southeastern France, is expected to be more strongly affected by climate change than most other regions in the world (Sicard and Dalstein-Richier, 2015). In Marseille, an annual mean rainfall and air temperature of 588 mm and 14.2 °C, respectively (6.3 °C in January - 23.1 °C in July, on average) are observed (Sicard et al., 2019).

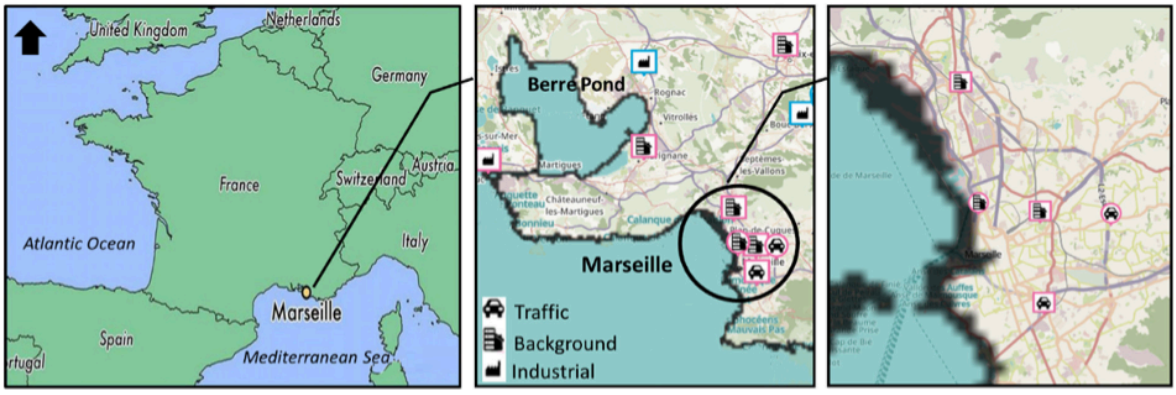

Fig. 1. Location of the study area and air quality monitoring stations (blue/pink: suburban/urban station; square/circle: fixed/mobile station). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

2.2. Air pollution data and population exposure

In Marseille, there are five urban air quality monitoring stations (3 background and 2 traffic). Only urban background stations, with more than 75% of validated hourly data in a year, were selected to better represent average exposure and to calculate valid aggregated values (24-h average concentration). The hourly $PM_{2.5}$ concentrations were provided by the Certified Associations of Air Quality Monitoring in Southeastern France (Atmo Sud) and obtained from January 1, 2010 to December 31, 2020. In this study, we detected outliers within time-series by using Z-scores (Al-Hemoud et al., 2020). Z-scores are the number of standard deviations above and below the average. A standard cut-off value for finding outliers are Z-scores of ± 3. The $PM_{2.5}/PM_{10}$, $PM_{2.5}/NO_2$ and $PM_{2.5}/O_3$ ratios were calculated to fully understand the relationship between $PM_{2.5}$ and other major air pollutants (Xu et al., 2017). These ratios can be used as indicators of air pollution over time to characterize the underlying atmospheric processes and to provide further insights into the spatio-temporal variability of air pollutants in Marseille. In risk assessment, exposure is considered equal to air pollutants concentrations at a specific point in space and time (Bogaert et al., 2009). In this study, the urban population exposure to ambient $PM_{2.5}$ was estimated by considering that the entire population is potentially exposed to the averaged concentrations i.e., excluding human mortality. The non-parametric Spearman rank correlation test was applied to this dataset to measure statistical dependence among variables.

2.3. Evaluation of the health impacts

To estimate short-term mortality and morbidity attributed to $PM_{2.5}$ exposure, the requested inputs are city-specific baseline incidence (BI) rates, relative risk (RR) values and the population at risk (N) for a given health endpoint (WHO, 2020). For each health endpoint, the RR represents the probability of developing the disease when $PM_{2.5}$ concentration increases by 10 μg m^{-3} (Eq. (1)). The city-specific RR values were obtained from exposure-response functions, based on epidemiological studies in Mediterranean cities such as Marseille, following recommendations from the Health Risks of Air Pollution in Europe project, and published by Samoli et al. (2013), Stafoggia et al. (2013) and WHO (2013a).

$$RR = exp[\beta(X - X_0] \quad \text{(eq. 1)}$$

where β is a parameter that regulates the rate of RR increment, X (μg m^{-3}) is the measured $PM_{2.5}$ concentration in air quality monitoring stations, and X_0 (μg m^{-3}) denotes the cut-off value corresponding to a lower level of pollution, i.e., the background concentration where no health effect is reported (Amoatey et al., 2020). The WHO recommends a cut-off value of 10 μg m^{-3} for $PM_{2.5}$ (WHO, 2013a). For the non-accidental mortality in Marseille, RR = 1.015 for $PM_{2.5}$, i.e., a 10 μg m^{-3} increase in the 24-h average $PM_{2.5}$ concentration is associated with a 1.5% increase in the risk for mortality attributed to non-accidental causes.

The attributable proportion (AP, in %) is the fraction of a health endpoint that can be related to the exposure to *c* in a population $P_{(c)}$ where $RR_{(c)}$ is the relative risk value of the air pollutant *c* concentration (WHO, 2020).

$$AP = \frac{\sum [(RR_c - 1) \times P_c]}{\sum [RR_c \times P_c]} \quad \text{(eq. 3)}$$

For a selected health endpoint, the number of cases NC_c per 10^5 people at risk attributed to the air pollutant *c* is calculated as NC_c = BI × AP where BI is the baseline incidence rates and AP the attributable proportion. The number of excess cases NE_c attributed to the air pollutant *c* is calculated as $NE_c = 10^{-5} \times [NC_c \times N]$ where *N* is the number of people at risk exposed to the air pollutant *c* (WHO, 2020).

Table 1
The city-specific baseline incidence rates (BI per 10^5 inhabitants) and Relative Risk (RR) values for health outcomes for people at risk (all ages), for 10 μg m^{-3} increase in particulate matters ($PM_{2.5}$, 24 h average) in Marseille ([1] WHO, 2013a, b;[2] Héroux et al., 2015;[3] Samoli et al., 2013;[4] Stafoggia et al., 2013) with 95% confidence intervals (lag 0–1 days).

Health endpoints	BI	RR (95% CI)
Mortality		
Non-accidental causes	1007	1.015[1,2] [1.007; 1.022]
Cardiovascular diseases	320	1.028[1,2] [1.009; 1.047]
Respiratory diseases	91	1.009[1,2] [1.003; 1.020]
Hospital admissions		
Cardiovascular diseases	1960	1.0091[3] [1.002; 1.015]
Respiratory diseases	760	1.0190[3] [0.998; 1.040]

The number of premature deaths for non-accidental causes, cardiovascular diseases and respiratory diseases, and the number of hospital admissions for cardiovascular and respiratory diseases, due to ambient $PM_{2.5}$ exposure were evaluated based on city-specific RR values and BI rates (Table 1).

2.4. Statistical estimation of annual trends

To detect changes within time-series and estimate the magnitude of trends, we applied the non-parametric Mann-Kendall test and the non-parametric Sen's slope estimator, by using the MAKESENS program developed by the Finnish Meteorological Institute. A 10-year time-series is considered long enough to assess short-term changes (Sicard et al., 2016). Both tests were applied for the annual mean $PM_{2.5}$ concentrations, the number of premature deaths and hospital admissions attributed to ambient $PM_{2.5}$ exposure in Marseille over the time period 2010–2019. Results were considered significant at $p < 0.05$.

2.5. Economic valuation of $PM_{2.5}$ pollution

Valuation of health effects is performed by multiplying the number of premature deaths and hospital admissions by respectively the "value of a statistical life" (VSL in France: €3.15 million in 2010–3.63 million in 2019), and the average cost of a hospital admission (2220€ in 2010–2520€ in 2019) considering the costs of healthcare, lost productivity and welfare losses provided by the Organization for Economic Co-operation and Development OECD (https://stats.oecd.org).

3. Results

3.1. Temporal variability in $PM_{2.5}$ levels

Over the time period 2010–2019, the lowest city-averaged annual $PM_{2.5}$ concentrations (Table 2) were recorded in 2019 (11.1 μg m^{-3}) while the highest concentrations were observed in 2011 (20.7 μg m^{-3}). The annual $PM_{2.5}$ concentrations exceeded the annual limit value established by the WHO for the protection of human health (10 μg m^{-3}) while they did not exceed the EU limit value (25 μg m^{-3}). Between 2010 and 2019, the daily limit value established by the WHO Air Quality Guidelines (25 μg m^{-3}) was exceeded 95 times in 2011 and 10 times in 2019 (Table 2). From 2010 to 2019, the annual averaged $PM_{2.5}$ concentrations decreased by 0.80 μg m^{-3} $year^{-1}$ on average in Marseille (Table 3). By selecting

Table 2
City-averaged annual mean $PM_{2.5}$ concentration (±standard deviation) and numbers of days with 24-h $PM_{2.5}$ mean concentration over 25 μg m^{-3} (WHO Air Quality Guidelines). Number of cases per 10^5 people at risk attributed to $PM_{2.5}$ exposure above the cut-off value (10 μg m^{-3}) for the following health endpoints: mortality for non-accidental causes (M-all causes), cardiovascular diseases (M-CVD), and respiratory diseases (M-RD), and for hospital admissions due to cardiovascular diseases (HA-CVD) and respiratory diseases (HA-RD), with lower and upper limits of the relative risk 95% confident interval [95% CI].

	2010	2011	2012	2013	2014	2015
$PM_{2.5}$ conc. [a]	18.4 ± 8.9	20.7 ± 9.6	16.8 ± 11.2	15.2 ± 8.9	16.2 ± 6.2	15.9 ± 7.7
WHO AQG [b]	65	95	74	46	28	41
M-all causes	13.7 [6.4; 20.1]	16.7 [7.9; 24.4]	12.5 [5.9; 18.3]	9.7 [4.5; 14.2]	10.2 [4.8; 14.9]	10.0 [47.7; 14.6]
M-CVD	8.1 [2.6; 13.4]	9.8 [3.2; 16.3]	7.4 [2.4; 12.3]	5.7 [1.8; 9.5]	6.0 [1.9; 9.9]	5.9 [1.9; 9.8]
M-RD	0.75 [0.25; 1.65]	0.91 [0.31; 2.01]	0.68 [0.23; 1.51]	0.53 [0.18; 1.16]	0.55 [0.19; 1.22]	0.54 [0.18; 1.20]
HA-CVD	16.3 [3.0; 29.6]	19.9 [3.7; 36.0]	14.8 [2.8; 27.0]	11.4 [2.1; 20.8]	12.1 [2.3; 21.9]	11.9 [2.2; 21.6]
HA-RD	13.1 [- 1.3; 27.4]	16.0 [- 1.5; 33.3]	12.0 [- 1.1; 25.1]	9.2 [- 0.9; 19.4]	9.7 [- 0.9; 20.3]	9.6 [- 0.9; 20.0]
	2016	2017	2018	2019	2020	
$PM_{2.5}$ conc. [a]	16.0 ± 7.9	14.6 ± 7.1	12.5 ± 5.9	11.1 ± 5.8	10.0 ± 5.1	
WHO AQG [b]	42	27	11	10	6	
M-all causes	10.2 [4.8; 15.0]	8.4 [3.9; 12.3]	5.5 [2.6; 8.1]	4.2 [1.9; 6.1]	3.0 [1.4; 4.3]	
M-CVD	6.0 [2.0; 10.1]	5.0 [1.6; 8.3]	3.3 [1.0; 5.4]	2.5 [0.8; 4.1]	1.7 [0.6; 2.9]	
M-RD	0.56 [0.19; 1.23]	0.46 [0.15; 1.01]	0.30 [0.10; 0.66]	0.23 [0.08; 0.50]	0.16 [0.05; 0.36]	
HA-CVD	12.1 [2.3; 22.1]	9.9 [1.9; 18.1]	6.5 [1.2; 11.9]	4.9 [0.9; 9.0]	3.5 [0.6; 6.4]	
HA-RD	9.8 [- 0.9; 20.5]	8.0 [- 0.8; 16.8]	5.3 [- 0.5; 11.1]	4.0 [- 0.4; 8.4]	2.8 [- 0.3; 5.9]	

[a] The annual mean $PM_{2.5}$ concentration does not to exceed 25 μg m^{-3} (EU limit value) or 10 μg m^{-3} (WHO Air Quality Guidelines).
[b] Numbers of days with 24-h $PM_{2.5}$ mean concentration over 25 μg m^{-3} (WHO Air Quality Guidelines).

Table 3
Annual trends (Sen's slope estimator) for mean $PM_{2.5}$ concentrations (in μg m^{-3} $year^{-1}$), and for mortality (M) and hospital admissions (HA-) due to ambient $PM_{2.5}$ (expressed as number of cases per 10^5 inhabitants per year) for non-accidental causes (all), and for cardiovascular (CVD) and respiratory diseases (M-RD) over the time period 2000–2019 in Marseille (Mann-Kendall $p < 0.05$). Estimated mean $PM_{2.5}$ concentration and number of cases per 10^5 people at risk attributed to $PM_{2.5}$ exposure above the cut-off value in 2020 by applying the Sen's slope estimator.

	Trends 2010–2019	Estimated 2020
$PM_{2.5}$ conc.	- 0.80	11.1
M-all	- 1.15	3.3
M-CVD	- 0.68	1.9
M-RD	- 0.06	0.18
HA-CVD	- 1.35	4.0
HA-RD	- 1.10	3.1

the year 2015 as an example, and representative of the whole study period, the highest hourly $PM_{2.5}$ concentrations (93.1 μg m^{-3}) was recorded on 1st January and during the week-end (Fig. 2a). The daily limit value established by the WHO Air Quality Guidelines (25 μg m^{-3}) was exceeded 41 times (Fig. 2b), and higher concentrations were recorded in winter, e.g. on average 22.2 μg m^3 in February (Fig. 2c).

3.2. Relationship between $PM_{2.5}$ and other air pollutants

In 2015, the hourly $PM_{2.5}$ concentrations were well correlated ($r^2 = 0.63$, $p < 0.05$) with hourly PM_{10} concentrations (Fig. 3a), with an averaged $PM_{2.5}/PM_{10}$ ratio of 0.52 (Fig. 3a'). The highest hourly (153.0 μg m^{-3}) and daily PM_{10} concentration (57.3 μg m^{-3}) were recorded on 5th June and March 11, 2015, with a $PM_{2.5}/PM_{10}$ ratio of 0.17 and 0.68, respectively. A significant correlation (r^2=0.51, $p < 0.05$) was found between hourly $PM_{2.5}$ and NO_2 concentrations (Fig. 3b), with a $PM_{2.5}/NO_2$ ratio of 0.35 (Fig. 3b'). A negative correlation was measured between ambient hourly $PM_{2.5}$ and O_3 concentrations (Fig. 3c). The highest hourly O_3 concentration (107.6 μg m^{-3}) was recorded on April 16, 2015, with a $PM_{2.5}/O_3$ ratio of 0.10. The averaged $PM_{2.5}/O_3$ ratio was 0.42 (Fig. 3c').

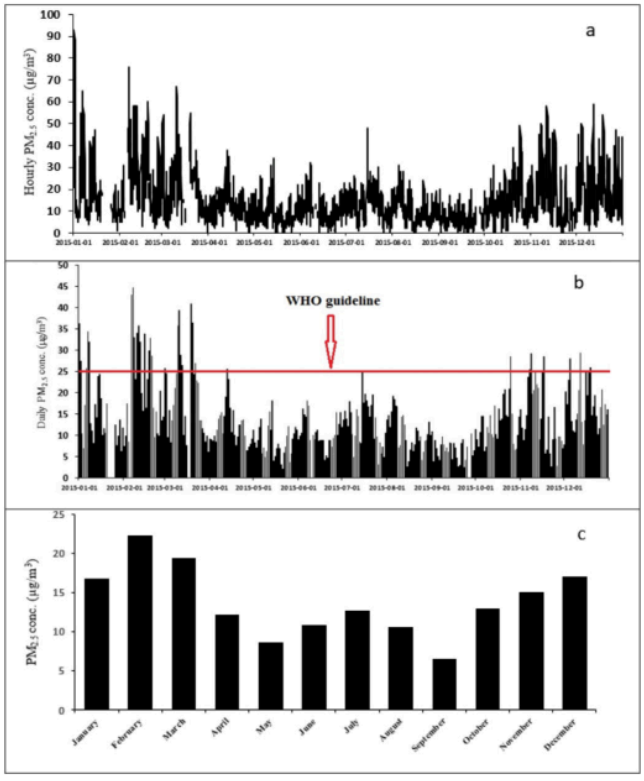

Fig. 2. Time series plots; (a) Hourly $PM_{2.5}$ variations in Marseille in 2015, (b) daily variations with the daily limit value for $PM_{2.5}$ established by the WHO Air Quality Guidelines (25 μg m^{-3}); (c) Monthly $PM_{2.5}$ mean concentrations in 2015.

3.3. Trends in mortality and morbidity from ambient $PM_{2.5}$ exposure

Over the time period 2010–2019, the number of premature deaths for non-accidental causes attributed to $PM_{2.5}$ above 10 μg m^{-3} ranged from 16.7 per 10^5 inhabitants (incl. 9.8 M-CVD) in 2011 to 4.2 per 10^5 inhabitants (incl. 2.5 M-CVD) in 2019 (Table 2).

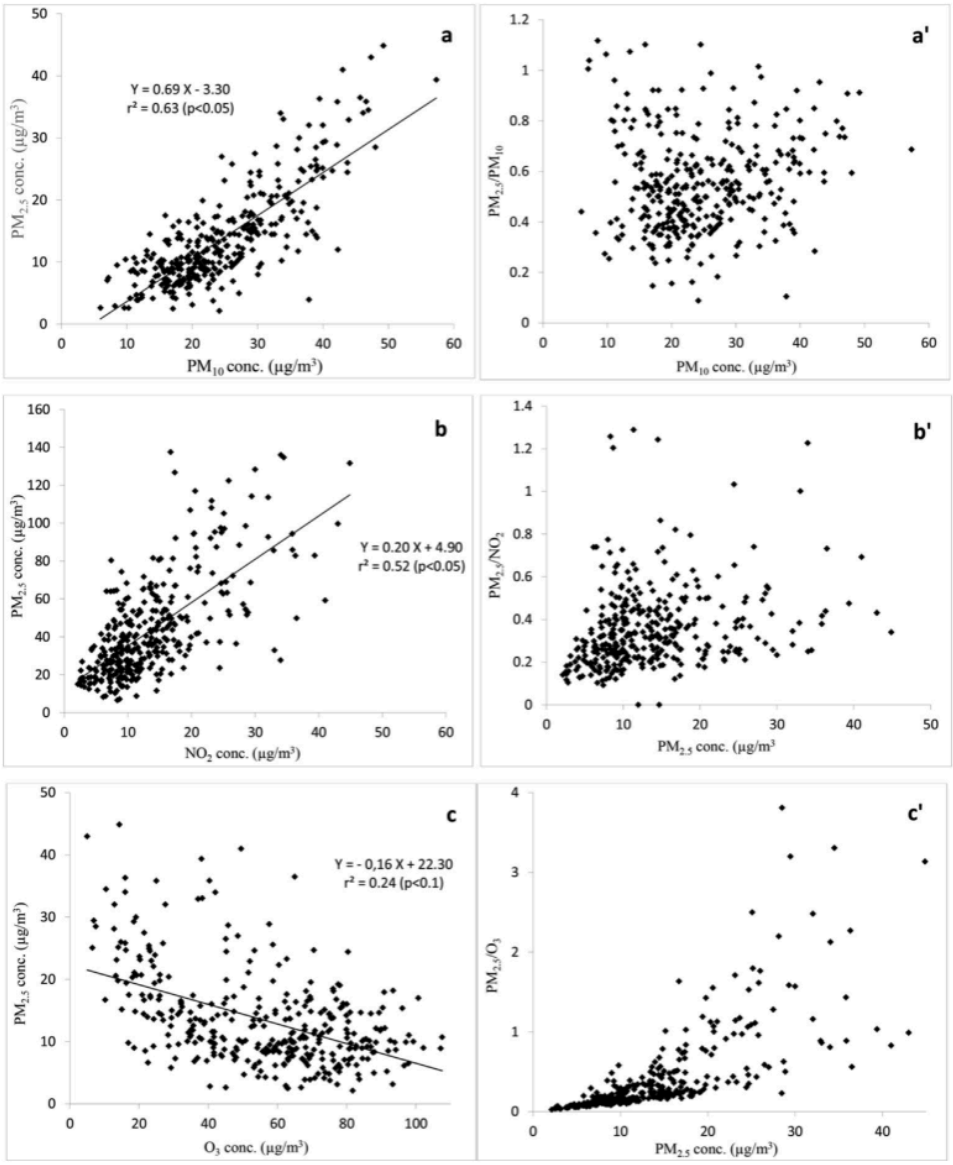

Fig. 3. Correlation between hourly $PM_{2.5}$ concentrations and (a) PM_{10}; (a') $PM_{2.5}/PM_{10}$ ratio; (b) NO_2 concentrations; (b') $PM_{2.5}/NO_2$ ratio; (c) O_3 concentrations; and (c') $PM_{2.5}/O_3$ ratio in 2015 in Marseille.

The number of HA-CVD and HA-RD per 10^5 people was 19.9 and 16.0 in 2011, and 4.9 and 4.0 in 2019, respectively (Table 2). The CVD are the main cause of non-accidental mortality due to $PM_{2.5}$ (about 60%). The number of non-accidental deaths due to ambient $PM_{2.5}$ levels decreased by on average 1.15 per 10^5 inhabitants annually between 2010 and 2019 (Table 3). The highest annual decreases were observed for M-CVD (- 0.68 deaths per 10^5 inhabitants). The number of hospital admissions declined by 1.35 per 10^5 inhabitants per year for CVD and by 1.10 per 10^5 inhabitants per year for RD (Table 3). For all health endpoints, the trend magnitude was higher in 2015–2019 than 2010–2014.

3.4. Economic valuation of $PM_{2.5}$ pollution

By multiplying the number of cases per 10^5 people by the population at risk, the number of $PM_{2.5}$-related deaths ranged from 142 in 2011 to 36 in 2019 (Table 2). The number of $PM_{2.5}$-related hospital admissions for respiratory and cardiovascular diseases ranged from 305 in 2011 to 77 in 2019. The healthcare cost of $PM_{2.5}$ pollution is estimated between 457.9 million in 2011 and 130.8 million in 2019. The $PM_{2.5}$ pollution can result in substantial socio-economic benefits, for instance 36 premature non-accidental deaths and 77 hospital admissions attributable to $PM_{2.5}$ in 2019 could have been "avoided" by respecting the WHO annual limit value of 10 µg m^{-3}, i.e., a benefit for healthcare of about €131 million.

3.5. $PM_{2.5}$ levels and related health effects during COVID-19 pandemic

In 2020, the annual $PM_{2.5}$ mean concentration was 10.0 µg m^{-3} (Table 2), meeting the air quality limit value established by the WHO for human health protection. The daily WHO limit value was exceeded 6 times. During the full year, 3.0 premature deaths per 10^5

inhabitants for non-accidental causes (incl. 1.7 M-CVD), and 4.9 and 4.0 admissions per 10^5 persons for CVD and RD, respectively, were attributed to $PM_{2.5}$ above 10 μg m^{-3}. By considering a population of 868,300 inhabitants in 2020, the number of premature deaths for non-accidental causes attributed to $PM_{2.5}$ was 26 (incl. 15 M-CVD). Following the 2010–2019 trend, the annual $PM_{2.5}$ mean concentration would have been 11.1 μg m^{-3} in Marseille, without COVID-19 pandemic (Table 3). By applying the Sen's slope estimator for estimating the number of cases in 2020 without COVID-19 impact, we obtained 3.3 premature deaths per 10^5 people for all causes (incl. 1.9 M-CVD) attributed to $PM_{2.5}$ exposure, and 4.0 HA-CVD and 3.1 HA-RD per 10^5 persons at risk (Table 3). The "actual gain" (estimated - observed) due to the $PM_{2.5}$ levels reduction (- 11%), mainly related to lockdown measures during COVID-19 pandemic, could be about 0.3 premature deaths, 0.5 HA-CVD and 0.3 HA-RD per 10^5 people (Table 2, Table 3), i.e. 2.6 deaths were averted in Marseille in 2020.

4. Discussion

Due to urban, industrial, shipping and natural fine particles emissions (Salameh et al., 2018), Marseille is the second most polluted city in France, after Paris, and can be considered as a case study to assess the $PM_{2.5}$ effects on human health in a Mediterranean urban environment. The annual $PM_{2.5}$ mean concentrations recorded in Marseille (ab. 16 μg m^{-3} in 2015) were lower than the $PM_{2.5}$ levels observed in Rome (ab. 18 μg m^{-3} in 2015), mainly due to higher population, and road traffic in Rome (De Marco et al., 2018), and the annual mean is ab. 40 μg m^{-3} in Khorramabad (Iran), mainly due to Middle Eastern dust storms (Khaniabadi et al., 2019b). Dust from Sahara Desert can reach Southern Europe leading to high levels of PM concentrations in cities (Querol et al., 2019). In Western Europe, the highest hourly $PM_{2.5}$ concentrations originating from Sahara reached ab. 145 μg m^{-3} in March 2007 in Brussels, Belgium (Vanderstraeten et al., 2008). During desert dust outbreaks, the maximum hourly $PM_{2.5}$ concentrations ranged from 43 to 86 μg m^{-3} over Southern Europe, representing 43–47% of the PM_{10} mass (Goudie, 2014; Querol et al., 2019). In Marseille, the on-road transport and industrial emissions contribute to on average 17% and 7% of $PM_{2.5}$ concentrations, respectively (El Haddad et al., 2011; Salameh et al., 2018). We observed higher $PM_{2.5}$ levels in winter, in particular during weekend and bank holidays, due to activities at home e.g., domestic heating and biomass burning (e.g., hourly peak of ab. 93 μg m^{-3} on January 1, 2015). Biomass burning from residential heating in winter represents on average 14% of $PM_{2.5}$ (Salameh et al., 2018). During the higher $PM_{2.5}$ pollution episodes in 2011–2012, open green waste burning was the largest source of $PM_{2.5}$ (ab. 33%), in particular in fall during land clearing period (Salameh et al., 2018).

In Marseille, we found a coefficient $r^2 = 0.63$ between hourly $PM_{2.5}$ and PM_{10} concentrations. PM measurements were made at 20 sites in 20 European cities across Europe over the time period 2008–2011 (Eeftens et al., 2012), and the correlation between $PM_{2.5}$ and PM_{10} was highly variable across Europe, with a r^2 ranging from 0.44 (Manchester, United Kingdom) to 0.95 (Paris, France). In Marseille, a mean $PM_{2.5}/PM_{10}$ ratio of 0.52 was calculated, i.e., lower than the ratio found in the literature (Querol et al., 2008; Henschel et al., 2013; Contini et al., 2014) and recommended by the WHO (2013b) for the Mediterranean European region ($PM_{2.5}/PM_{10} = 0.65$). In Europe, the $PM_{2.5}/PM_{10}$ ratio ranged from 0.39 (Heraklion, Greece) to 0.74 (Gyor, Hungary), with the lowest values in Southern Europe and higher values in Eastern Europe between 2008 and 2011 (Eeftens et al., 2012). In Catalonia (Spain), the annual $PM_{2.5}/PM_{10}$ ratio ranged from 0.58 to 0.64 over the time period 2008–2017, with a mean ratio of 0.63 in 2015 (Rovira et al., 2020). In Switzerland, due to lower road traffic and industrial activities, the $PM_{2.5}/PM_{10}$ ratio ranged from 0.72 (rural stations) to 0.75 (urban stations) over the time period 1998–2001, with the exception of the traffic station (ratio = 0.59) in Bern (Gehrig and Buchmann, 2003). In the Middle Eastern cities, the ratio ranged from 0.21 to 0.59, due to high levels of PM during or after dust storms (Goudie, 2014; Khaniabadi et al., 2017d, 2019a; Daryanoosh et al., 2018). Some studies used NO_2 as indicator of traffic-related air pollution (e.g., Lewné et al., 2004; Eeftens et al., 2012). Strong correlations ($r^2 = 0.60–0.80$) were obtained between $PM_{2.5}$ and NO_2 in 40–42 urban and suburban stations each in Germany (Munich), the Netherlands and Sweden (Stockholm County) in 1999–2000 (Lewné et al., 2004). In Europe, the correlations between $PM_{2.5}$ and NO_2 concentrations ranged from 0.02 (Gyor, Hungary) to 0.90 (Barcelona, Spain) with no clear spatial pattern in 2008–2011, and a mean $PM_{2.5}/NO_2$ ratio varied from 0.33 to 1.49 for both cities, respectively (Eeftens et al., 2012). In Marseille, the regression of $PM_{2.5}$ on NO_2 showed a significant intercept ($r^2 = 0.52$), with a mean $PM_{2.5}/NO_2$ ratio of 0.35, representing sources other than local combustion (Lewné et al., 2004). The traffic-induced PM emissions causes a reduction of the $PM_{2.5}/PM_{10}$ ratio, and the correlations between $PM_{2.5}$ and NO_2 and PM_{10} were likely due to common sources of the air pollutants, such as road traffic, heating systems, industries, and other combustion processes (Núñez Alonso et al., 2019). As expected, a negative correlation was found between $PM_{2.5}$ and O_3 in this study. As PM absorb solar radiation (Liu et al., 2013; Li et al., 2017), a reduction in $PM_{2.5}$ could lead to an increase in surface O_3 concentrations (Sicard et al., 2020a). In addition to photochemical reactions, the heterogeneous chemical processes occurring on $PM_{2.5}$ surface in the atmosphere are an important way for the interaction between O_3 and $PM_{2.5}$ (Deng et al., 2010; Li et al., 2011).

The exposure to ambient $PM_{2.5}$ is associated to mortality for cardiovascular and respiratory diseases e.g., chronic obstructive pulmonary disease (COPD), asthma exacerbation, stroke and acute heart failure, lung cancer (Bell et al., 2007, 2008; Polichetti et al., 2009; Pascal et al., 2014; Zhang et al., 2020). A 10 μg m^{-3} increase in daily $PM_{2.5}$ concentrations was associated with 1.04% increase in mortality for non-accidental causes worldwide (Atkinson et al., 2014). The risk in premature mortality has been reported even when $PM_{2.5}$ concentrations are below European standards (Stafoggia et al., 2014; Bourdrel et al., 2017). For better comparison, we reported here the number of cases per 10^5 people at risk from literature. In Marseille, non-accidental mortality attributable to $PM_{2.5}$ above 10 μg m^{-3} was 10.0–10.2 cases per 10^5 people at risk in 2015–2016, respectively. In other Mediterranean cities with similar annual $PM_{2.5}$ concentrations in 2015–2016, we found lower attributable rates in Nice in Southeastern France (8.1–9.7 cases/10^5 incl. 4.9–5.8 M-CVD) and Livorno in Italy (7.3–10.7 cases/10^5 incl. 2.9–4.2 M-CVD), and much lower in Rome in Italy (24.6–34.7 cases/10^5 incl. 10.5–14.8 M-CVD) (De Marco et al., 2018; Sicard et al., 2019). In Catalonia (Spain) all-natural causes mortality due to ambient $PM_{2.5}$ ranged from 6.9 to 41.7 cases per 10^5 individuals between 2005 and 2017 (Rovira et al., 2020). In Tehran (Iran), 87.0 non-accidental deaths per 10^5 people at risk were attributable to $PM_{2.5}$ exposure in 2015, with an annual $PM_{2.5}$ average of 51 μg m^{-3} (Hajizadeh et al., 2020). In Tehran, the number of premature deaths due to $PM_{2.5}$ exposure ranged from 54.8 to 133.0 per 10^5 people at risk over the time period 2014–2019 (Hajizadeh et al., 2020). In Khorramabad (Iran), respectively 98 M-all causes, 28 M-CVD and 4 M-RD per 10^5 individuals were attributed to $PM_{2.5}$ exposure (annual mean: 40 μg m^{-3}) in 2016 (Khaniabadi et al., 2019b). Health effects associated with long-term fine PM exposure are mortality for all-causes (+6%) and M-CVD (+11%) in relation to a 10 μg m^{-3}

increase in annual $PM_{2.5}$ concentration worldwide (Anenberg et al., 2010; Hoek et al., 2013). The cardio-vascular diseases are the main cause of non-accidental mortality (France: 50–60%, Italy: 40–55%) and hospital admissions due to $PM_{2.5}$ exposure (De Marco et al., 2018; Sicard et al., 2019) while M-CVD represents 5–15% of the non-accidental mortality due to $PM_{2.5}$ in Iran (Khaniabadi et al., 2019b; Sicard et al., 2019). Xie et al. (2015) conducted a study in Beijing in 2010–2012 and showed that the dose-response relationships between $PM_{2.5}$ and mortality for cardiac diseases were non-linear, with a higher response at lower concentrations and lower at higher concentrations. In 2015–2016, 6.5–4.7 HA-CVD and 6.1–4.4 HA-RD $\times$ 10^5 people were reported in Rome (Italy) and 3.4–2.8 HA-CVD and 7.4–6.2 HA-RD $\times$ 10^5 people in Nice (France) when $PM_{2.5}$ concentrations are beyond 10 $\mu g\ m^{-3}$ (De Marco et al., 2018; Sicard et al., 2019), while in this study we found 11.9–12.1 HA-CVD and 9.6–9.8 HA-RD $\times$ 10^5 people over the same time period. The difference in the attributable cases of premature deaths and hospital admissions is mainly related to population characteristics and methodological differences (e.g., RR and BI values) in exposure assessment (De Marco et al., 2018).

In Marseille, despite the reduction of $PM_{2.5}$ concentrations (-0.80 $\mu g\ m^{-3}\ year^{-1}$), about 12% of daily $PM_{2.5}$ concentrations exceeded the WHO limit value for human health protection over the time period 2010–2019. The number of non-accidental premature deaths due to $PM_{2.5}$ exposure decreased by 1.15 per 10^5 inhabitants annually. In France, the number of deaths due to ambient $PM_{2.5}$ levels decreased by on average 0.35 per 10^5 people per year between 2000 and 2017 (Sicard et al., 2021). In the EU-28 countries, the highest annual decrease was observed in the United Kingdom (- 1.17 deaths per 10^5 people) while a slight increase was found in Greece (+0.12 deaths per 10^5 people) and Lithuania (+0.17 deaths per 10^5 people) (Sicard et al., 2021).

Compared to the same period in 2017–2019, the lockdown measures in 2020 led to a slight decrease of $PM_{2.5}$ concentrations (about 8%) in Southern European cities due to the reduction of road and non-road transport counterbalanced by an increase of PM emissions from the activities at home e.g., domestic heating and biomass burning (Sicard et al., 2020b). Based on 2010–2019 trends analysis, the restrictive measures associated to COVID-19 pandemic led to a reduction in $PM_{2.5}$ of 11% in Marseille, with 2.6 $PM_{2.5}$-related deaths averted. Compared to the time period 2016–2019, the lockdown measures led to a $PM_{2.5}$ reduction of 17.1% across Europe, and 2190 deaths were averted between 21st February and May 17, 2020 in Europe (Giani et al., 2020).

5. Conclusion

In Marseille, the main sources of $PM_{2.5}$ are road traffic, industries, and occasionally desert dust transported to Europe from Africa and forest fires (Salameh et al., 2018; Querol et al., 2019). At present compared to other air pollutants, $PM_{2.5}$ poses the most serious health risk in European cities (Pascal et al., 2014; Cohen et al., 2017; Sicard et al., 2021). Over the time period 2010–2019, the $PM_{2.5}$ concentration levels regularly exceeded the daily limit value and the annual limit value (on average 1.5 times) set by the WHO, while the levels were in agreement with EU-target values.

Between 2010 and 2019, the urban population exposure to ambient $PM_{2.5}$ substantially contributed to mortality and morbidity. In total, 871 non-accidental deaths, 515 deaths for CVD, 47 deaths for RD as well as 1034 HA-CVD and 834 HA-RD can be attributed to $PM_{2.5}$ exposure above 10 $\mu g\ m^{-3}$. Over the last decade, a significant decreasing trend of $PM_{2.5}$ levels, and $PM_{2.5}$-related mortality and morbidity for CVD and RD, is observed in Marseille. Compliance with WHO annual limit values can result in substantial socio-economic benefits, for instance 36 premature deaths and 77 hospital admissions attributable to $PM_{2.5}$ in 2019 could have been "avoided" in Marseille, with an associated benefit for healthcare of about €131 million. In the 28 countries of the EU, the healthcare cost of $PM_{2.5}$-related mortality and morbidity was estimated at €506 billion (Aman et al., 2017). In 2020, the $PM_{2.5}$ levels and related health endpoints were reduced in Marseille mainly due to COVID pandemic lockdowns, compared to previous years (Sicard et al., 2020b).

In this study, we hypothesized that all individuals were exposed to the same levels of $PM_{2.5}$ exposure in Marseille, averaged from 5 urban air quality monitoring stations, and human mobility and exposure to indoor air pollutants, where people spend more than 80% of time, were ignored. Therefore, estimates of non-accidental mortality and morbidity due to $PM_{2.5}$ exposure were biased (Laumbach et al., 2015; Cincinelli and Martellini, 2017; Park and Kwan, 2017). To reduce population's exposure to PM levels, adequate mitigation measures should be implemented to reduce, not only primary PM emissions, but also the emission of gaseous pollutants (precursors of secondary PM) such as an introduction of electric/hybrid cars, speed reduction, green public transportation, regulation of fuel quality, street cleaning to abate resuspension of deposited dust by road traffic, and green infrastructure (Sofia et al., 2020). Previous studies provided quantitative estimates of the ability of green infrastructure to ameliorate urban air quality (e.g., PM, O_3) at city scale worldwide (Sicard et al., 2018).

Credit author statement

Y.O.K and P.S. have conceived the project. Y.O.K and P.S. analyzed the data, and Y.O.K and P.S. have participated in writing and revising the manuscript.

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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