

# Urban Population Exposure to Air Pollution in Europe Over the Last Decades

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## Research

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# Abstract

**Background** - The paper presents an overview of air quality in the 27 member countries of the European Union (EU) and the United Kingdom (previous EU-28), from 2000 to 2017. We reviewed the progress made towards meeting the air quality standards established by the EU Ambient Air Quality Directives (Directive 2008/50/EC) and the World Health Organization (WHO) Air Quality Guidelines by estimating the trends (Mann-Kendal test) in national emissions of main air pollutants, urban population exposure to air pollution, and in mortality related to exposure to ambient fine particles (PM<sub>2.5</sub>) and tropospheric ozone (O<sub>3</sub>).

**Results** - Despite significant reductions of emissions (e.g. sulfur oxides: ~80%, nitrogen oxides: ~46%, non-methane volatile organic compounds: ~44%, particulate matters with a diameter lower than 2.5µm and 10µm: ~30%), the EU-28 urban population was exposed to PM<sub>2.5</sub> and O<sub>3</sub> levels widely exceeding the WHO limit values for the protection of human health. Between 2000 and 2017, the annual PM<sub>2.5</sub>-related number of deaths decreased (- 4.85 per 10<sup>6</sup> inhabitants) in line with a reduction of PM<sub>2.5</sub> levels observed at urban air quality monitoring stations. The rising O<sub>3</sub> levels became a major public health issue in the EU-28 cities where the annual O<sub>3</sub>-related number of premature deaths increased (+ 0.55 deaths per 10<sup>6</sup> inhabitants).

**Conclusions** - To achieve the objectives of the Ambient Air Quality Directives and mitigate air pollution impacts, actions need to be urgently taken at all governance levels. In this context, greening and re-naturing cities can help meet air quality standards, but also answer to social needs, as recently highlighted by the COVID-19 lockdowns.

## Background

Outdoor air pollution is a major global public health issue (Lelieveld et al., 2015), leading to 4.2 million premature deaths worldwide (WHO, 2016) and half a million in the European Union (EU) in 2016 (EEA, 2018). The EU identifies seven main air pollutants (Koolen and Rothenberg, 2019): ammonia (NH<sub>3</sub>), nitrogen oxides (NO<sub>x</sub>), carbon monoxide (CO), particulate matter with an aerodynamic diameter lower than 2.5 µm and 10 µm (PM<sub>2.5</sub> and PM<sub>10</sub>), sulfur oxides (SO<sub>x</sub>), tropospheric ozone (O<sub>3</sub>), and non-methane volatile organic compounds (NMVOCs). In cities, where 74% of the EU population lives (EU, 2016), PM<sub>2.5</sub> and ground-level O<sub>3</sub> have potentially the most significant effects on human health associated with respiratory and cardiovascular diseases and mortality, compared to other air pollutants (Pascal et al., 2013; WHO, 2013; Cohen et al., 2017). In 2016, 374,000 and 14,600 non-accidental premature deaths were attributed to air pollution (PM<sub>2.5</sub> and O<sub>3</sub>, respectively) in the EU-28<sup>1</sup> countries (EEA, 2018). Air pollution also damages plant ecosystems (Mills et al., 2011; Sicard et al., 2016a; Feng et al., 2019), and surface O<sub>3</sub> is considered as the most detrimental air pollutant in terms of effects on vegetation and biodiversity (Paoletti, 2007; Sicard et al., 2016a; Agathokleous et al., 2020).

The legislated ambient air quality standards and the emission control policies (e.g. European Council Directive 1996/62/EC; World Health Organization Air Quality Guidelines, WHO-AQG 2006; European Council Directive 2008/50/EC; National Emission Ceilings Directive, 2016; Convention on Long-range Transboundary Air Pollution, CLRTAP 2017) control emissions of harmful substances into the atmosphere, and regulate the concentrations of air pollutants such as PM<sub>2.5</sub>, PM<sub>10</sub>, NO<sub>2</sub> and O<sub>3</sub>, by setting limit and target values for the protection of human health (Table 1). The Clean Air Programme for Europe (CAPE), published by the European Commission in 2013, aims to improve air quality in Europe by 2030 and to reduce the number of premature deaths by half compared with 2005 (EC, 2013). Following the above directives, the number of air quality monitoring stations grew rapidly in Europe with databases gathering air quality data such as the European Environment Agency Airbase system. Due to the spatial representativeness of monitoring stations and the duration of time series, the above database offers an unprecedented way for trends analysis, and peer-reviewed articles.

Table 1

– Examples of air quality standards for common air pollutants as given in the European Ambient Air Quality Directive (Directive 2008/50/EC) and World Health Organization Air Quality Guidelines (WHO AQG) for the protection of human health.

Air pollutant	EU limit and target value (threshold in $\mu\text{g m}^{-3}$ )	WHO AQG (threshold in $\mu\text{g m}^{-3}$ )
<sup>a</sup> PM <sub>10</sub>	Annual mean (40)	Annual mean (20)
<sup>a</sup> PM <sub>10</sub>	Nb of exceedance of 24-h mean (50)	Nb of exceedance of 24-h mean (50)
<sup>b</sup> PM <sub>2.5</sub>	Annual mean (25)	Annual mean (10)
<sup>b</sup> PM <sub>2.5</sub>	-	Nb of exceedance of 24-h mean (25)
<sup>c</sup> O <sub>3</sub>	Nb of exceedance of maximum daily 8-h mean (120)	Nb of exceedance of maximum daily 8-h mean (100)
<sup>d</sup> NO <sub>2</sub>	Annual mean (40)	Annual mean (40)
<sup>d</sup> NO <sub>2</sub>	Nb of exceedance of 1-h mean (200)	Nb of exceedance of 1-h mean (200)
<sup>e</sup> SO <sub>2</sub>	Nb of exceedance of 24-h mean (125)	Nb of exceedance of 24-h mean (20)
<sup>f</sup> CO	Maximum daily 8-h (10,000)	Maximum daily 8-h (10,000)
<p><sup>a</sup> Annual mean PM<sub>10</sub> concentration and number of days with 24-hour PM<sub>10</sub> concentration over <math>50 \mu\text{g m}^{-3}</math> for the protection of human health. The annual mean PM<sub>10</sub> concentration does not to exceed <math>40 \mu\text{g m}^{-3}</math> (Directive 2008/50/EC) or <math>20 \mu\text{g m}^{-3}</math> (WHO AQG). The 24-h PM<sub>10</sub> mean concentration does not to exceed <math>50 \mu\text{g m}^{-3}</math> (WHO AQG) or more than 35 times a year (EC). <sup>b</sup> Annual mean PM<sub>2.5</sub> concentration and numbers of days with 24-h PM<sub>2.5</sub> mean concentration over <math>25 \mu\text{g m}^{-3}</math> (WHO AQG). The annual mean PM<sub>2.5</sub> concentration does not to exceed <math>25 \mu\text{g m}^{-3}</math> (EC) or <math>10 \mu\text{g m}^{-3}</math> (WHO AQG). <sup>c</sup> For the protection of human health, the Directive 2008/50/EC has introduced a threshold of <math>120 \mu\text{g m}^{-3}</math> for the daily maximum 8-h average. The threshold level should not be exceeded on more than 25 times a year. Number of days with daily maximum 8-h O<sub>3</sub> concentrations over <math>100 \mu\text{g m}^{-3}</math> as limit value for the protection of human health (WHO AQG). <sup>d</sup> Annual mean NO<sub>2</sub> concentration and number of hours with NO<sub>2</sub> concentrations above <math>200 \mu\text{g m}^{-3}</math>. The annual mean NO<sub>2</sub> concentration does not to exceed <math>40 \mu\text{g m}^{-3}</math> (EC and WHO AQG) while the hourly threshold should not be exceeded more than 18 times a year (EC). <sup>e</sup> The 24-h SO<sub>2</sub> mean concentration does not to exceed <math>125 \mu\text{g m}^{-3}</math> more than 3 times a year (EC) and does not to exceed <math>20 \mu\text{g m}^{-3}</math> (WHO AQG). <sup>f</sup> The Directives have introduced a threshold of <math>10 \text{mg m}^{-3}</math> for the maximum daily 8-hour mean concentration.</p>		

For the first time, through an extensive literature review and trends analysis, this study aims to i) quantify the annual trends in national emissions of main air pollutants in the EU-28 countries over the time period 2000–2017, ii) analyze the trends in real-world air pollutants concentrations over the last two decades; iii) assess the effectiveness of emissions control policies for reducing the exposure of EU-28 population to ambient air pollution, and iv) evaluate the impact of control policies on the number of premature deaths attributed to exposure to ambient PM<sub>2.5</sub> and O<sub>3</sub> levels over time.

<sup>1</sup>Including the United Kingdom, which withdrew from the European Union on 31<sup>st</sup> January 2020.

## Materials And Methods

### Data collection

The national emissions of main air pollutants (SO<sub>x</sub>, NH<sub>3</sub>, PM<sub>2.5</sub>, PM<sub>10</sub>) and main O<sub>3</sub> precursors (NO<sub>x</sub>, NMVOCs, CO), officially submitted by the Parties to the LRTAP Convention, were obtained by the European Monitoring and Evaluation (EMEP) Program<sup>2</sup>. The population exposure was estimated by the European Environmental Agency (EEA) from measured concentrations at urban and suburban background (non-traffic) monitoring stations across the EU-28 countries, with more than 75% of validated hourly data per year and station, and considering that the entire population is potentially exposed to the averaged concentrations (EEA, 2007; 2011–2019). The number of premature deaths attributed to exposure to ambient PM<sub>2.5</sub> and O<sub>3</sub> (per 10<sup>6</sup> inhabitants) were obtained by the Organization for Economic Co-operation and Development<sup>3</sup> (OECD). The above datasets were obtained over the time period 2000–2017.

### Estimation of the national number of premature deaths

The number of non-accidental premature deaths attributable to ambient PM<sub>2.5</sub> and O<sub>3</sub> were estimated for each EU member country and year by the method described in detail in Global Burden of Diseases (GDB, 2018) and widely used for the health risk assessment of air pollution (Goudarzi et al. 2017; Amoatey et al., 2019a,b; Khaniabadi et al., 2018a,b, 2019; De Marco et al. 2018; Sicard et al., 2019).

WHO set daily maximum 8-hour concentrations for O<sub>3</sub> and 24-hour average concentration for PM<sub>2.5</sub> as metrics to represent the mean daily exposure of population (WHO, 2008). The population exposure to O<sub>3</sub> and PM<sub>2.5</sub> is estimated by combining satellite data, a chemical transport model, land use information, and finally calibrated by using ground measurements (GBD, 2018). For a health endpoint, the number of cases  $NC_c$  attributed to the exposure to the air pollutant  $c$  is calculated as  $NC_c = BI \times AP$  where BI is the baseline incidence rates and AP the attributable proportion, i.e. the fraction of a health endpoint that can be related to the exposure to  $c$  in a population  $P_c$  where RR is the relative risk value, i.e. the probability of developing a disease associated to an increase of 10 µg m<sup>-3</sup> of the air pollutant  $c$  concentration (WHO, 2018).

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

These RR values are obtained from exposure-response functions, based on epidemiological studies, and published by WHO (2013). For the mortality “non-accidental causes”, RR = 1.015 and RR = 1.003 are reported for PM<sub>2.5</sub> and O<sub>3</sub>, respectively, i.e. for instance, a 10 µg m<sup>-3</sup> increase in the 24-hour average PM<sub>2.5</sub> concentration is associated with a 1.5% increase in the risk for mortality attributed to non-accidental causes.

## Statistical estimation of annual trends

A 10-year time-series is considered long enough to assess short-term changes (Sicard et al., 2009). The non-parametric Mann-Kendall test and the non-parametric Sen’s slope estimator were used to detect changes within time-series and estimate the magnitude of trends (Sicard et al., 2013; Guerreiro et al., 2014). Both tests were applied for annual national emissions of main air pollutants and the number of premature deaths attributed to exposure to ambient PM<sub>2.5</sub> and O<sub>3</sub> levels in EU-28 countries over the time period 2000–2017. Results were considered significant at p < 0.05.

## Literature review

To report robust short-term air pollutants changes over the last two decades, approximately 50 peer-reviewed articles and technical report spanning over the time period 2000–2017 were retrieved from literature databases (Science Direct, Web of Science, and Google scholar). We selected the studies with: i) in-situ observations from air quality monitoring networks (excluding modeled data); ii) annual mean concentrations; iii) at least 10-year time-series of data; iv) more than 75% of data coverage annually; and v) significant trend, i.e. with a p-value < 0.05.

<sup>2</sup><https://www.ceip.at>

<sup>3</sup><https://stats.oecd.org>

## Results And Discussion

### Trends in national emissions

Significant reductions were observed for the emission of all primary pollutants, i.e. – 4.7% year<sup>-1</sup> for SO<sub>x</sub>, – 2.7% year<sup>-1</sup> for NO<sub>x</sub>, – 2.6% year<sup>-1</sup> for NMVOCs, – 0.6% year<sup>-1</sup> for NH<sub>3</sub>, – 2.9% year<sup>-1</sup> for CO and – 1.8% year<sup>-1</sup> and – 1.7% year<sup>-1</sup> for PM<sub>2.5</sub> and PM<sub>10</sub>, respectively, over the time period 2000–2017 in the EU-28 (Table 2). The SO<sub>x</sub> emissions decreased in all EU-28 countries, from – 2.9% year<sup>-1</sup> (Germany) to – 6.0% year<sup>-1</sup> (Slovenia). For NO<sub>x</sub>, the highest decrease was observed in the United Kingdom (– 3.4% year<sup>-1</sup>),

while the lowest reduction was found in Lithuania ( $-0.6\% \text{ year}^{-1}$ ) and Poland ( $-0.7\% \text{ year}^{-1}$ ). For NMVOCs, the decrease ranged from  $-0.6\% \text{ year}^{-1}$  (Poland) to  $-4.0\% \text{ year}^{-1}$  (France). A slight decrease was also observed in Ireland ( $-0.7\% \text{ year}^{-1}$ ), Netherlands and Romania ( $-0.9\% \text{ year}^{-1}$ ). The sector “agriculture” contributes to 92% of  $\text{NH}_3$  emissions (EEA, 2019), and their emissions usually exhibited small reductions, with an increase in Austria, Estonia, Germany, Latvia, and Lithuania, ranging from 0.1 to  $1.0\% \text{ year}^{-1}$ . The domestic heating represents 48% of CO emissions (EEA, 2019). Also, the CO emissions usually decreased, except in Romania ( $-0.5\% \text{ year}^{-1}$ ) and Malta ( $+0.6\% \text{ year}^{-1}$ ). A decrease of  $\text{PM}_{2.5}$  emissions was observed in all EU-28 countries, except Bulgaria ( $+0.5\% \text{ year}^{-1}$ ), Hungary ( $+0.9\% \text{ year}^{-1}$ ) and Romania ( $+0.3\% \text{ year}^{-1}$ ), associating with a slighter reduction in  $\text{PM}_{10}$  emissions ( $-0.2\% \text{ year}^{-1}$  in Bulgaria;  $-0.1\% \text{ year}^{-1}$  in Hungary). An increase of  $\text{PM}_{10}$  emissions was noted in Lithuania ( $+0.8\% \text{ year}^{-1}$ ) and Romania ( $+0.1\% \text{ year}^{-1}$ ). The highest decrease for  $\text{PM}_{2.5}$  ( $-4.2\% \text{ year}^{-1}$ ) and  $\text{PM}_{10}$  ( $4.0\% \text{ year}^{-1}$ ) emissions occurred in Malta (Table 2).

Table 2

Annual trends of national emissions (% year<sup>-1</sup>) in the 28 European Union countries (EU-28) for sulfur oxides (SO<sub>x</sub>), nitrogen oxides (NO<sub>x</sub>), non-methane volatile organic compounds (NMVOCs), ammonia (NH<sub>3</sub>), carbon monoxide (CO), particulate matter with an aerodynamic diameter lower than 2.5 µm and 10 µm (PM<sub>2.5</sub> and PM<sub>10</sub>) over the time period 2000–2017. All trends are significant at  $p < 0.05$  (Mann-Kendall). The increasing trends are in bold.

EU-28 Countries	SO <sub>x</sub>	NO <sub>x</sub>	NMVOCs	NH <sub>3</sub>	CO	PM <sub>2.5</sub>	PM <sub>10</sub>
Austria	- 3.83	- 2.11	- 2.41	<b>+ 0.51</b>	- 1.57	- 2.31	- 1.74
Belgium	- 4.98	- 3.12	- 3.20	- 1.07	- 3.85	- 2.53	- 2.45
Bulgaria	- 5.35	- 2.41	- 1.47	- 0.54	- 1.59	<b>+ 0.47</b>	- 0.19
Croatia	- 4.84	- 2.63	- 2.25	- 1.34	- 2.85	- 1.71	- 1.72
Cyprus	- 4.58	- 1.77	- 2.23	- 1.22	- 3.65	- 3.03	- 3.39
Czech Republic	- 3.15	- 2.60	- 1.60	- 0.96	- 1.27	- 0.99	- 1.16
Denmark	- 4.45	- 3.35	- 2.30	- 1.48	- 3.00	- 1.49	- 1.13
Estonia	- 3.92	- 1.88	- 2.48	<b>+ 0.24</b>	- 2.07	- 3.01	- 3.41
Finland	- 3.77	- 2.92	- 3.23	- 0.64	- 2.51	- 2.17	- 1.88
France	- 4.85	- 3.12	- 3.96	- 0.30	- 3.75	- 3.37	- 2.87
Germany	- 2.93	- 1.86	- 2.07	<b>+ 0.13</b>	- 2.29	- 2.48	- 1.67
Greece	- 5.59	- 2.79	- 3.36	- 1.11	- 3.46	- 2.94	- 3.25
Hungary	- 4.59	- 2.35	- 2.04	- 0.44	- 3.00	<b>+ 0.93</b>	- 0.10
Ireland	- 5.97	- 2.40	- 0.75	- 0.26	- 3.84	- 2.33	- 1.94
Italy	- 5.63	- 3.40	- 2.63	- 1.12	- 3.03	- 1.07	- 1.40
Latvia	- 4.94	- 1.63	- 1.73	<b>+ 0.61</b>	- 3.49	- 2.16	- 1.36
Lithuania	- 3.60	- 0.61	- 1.35	<b>+ 1.01</b>	- 1.39	- 1.49	<b>+ 0.79</b>
Luxembourg	- 4.18	- 3.25	- 2.02	- 0.84	- 2.80	- 3.00	- 2.42
Malta	- 5.66	- 2.37	- 2.06	- 1.86	<b>+ 0.60</b>	- 4.20	- 3.95
Netherlands	- 4.14	- 2.72	- 0.88	- 1.76	- 1.78	- 3.53	- 2.65
Poland	- 3.62	- 0.69	- 0.62	- 0.81	- 1.67	- 0.72	- 0.79
Portugal	- 5.73	- 3.06	- 2.46	- 1.34	- 3.50	- 1.96	- 2.56
Romania	- 4.94	- 1.77	- 0.95	- 0.67	- 0.55	<b>+ 0.28</b>	<b>+ 0.15</b>

EU-28 Countries	SO <sub>x</sub>	NO <sub>x</sub>	NMVOCs	NH <sub>3</sub>	CO	PM <sub>2.5</sub>	PM <sub>10</sub>
Slovakia	- 4.46	- 2.31	- 2.69	- 0.98	- 2.36	- 3.22	- 3.06
Slovenia	- 6.03	- 2.50	- 2.70	- 0.89	- 2.79	- 0.52	- 1.22
Spain	- 5.44	- 2.99	- 2.62	- 0.82	- 2.08	- 1.30	- 1.63
Sweden	- 3.71	- 2.32	- 2.15	- 0.66	- 2.77	- 2.54	- 1.64
United Kingdom	- 5.26	- 3.42	- 3.28	- 0.52	- 4.21	- 1.40	- 1.70
EU-28	- 4.74	- 2.67	- 2.63	- 0.65	- 2.89	- 1.76	- 1.70

The emissions of all primary air pollutants contributing to ambient levels of PM, O<sub>3</sub>, and NO<sub>2</sub> decreased between 2000 and 2017 in the EU-28 (observed reductions SO<sub>x</sub>: - 80%; NO<sub>x</sub>: - 46%; NMVOCs: - 44%; NH<sub>3</sub>: - 10%; CO: - 49%; PM<sub>2.5</sub>: - 31%; PM<sub>10</sub>: - 29%), in line with stringent EC Directives, e.g. Air Quality Framework Directive (1996/62/EC), Large Combustion Plant Directive (2001/80/EC), and National Emission Ceilings Directives (2001/81/EC; 2016/2284/EC), setting emission reduction commitments by 2030 compared to 2005 (expected reductions SO<sub>2</sub>: - 79%; NO<sub>x</sub>: - 63%; NMVOCs: - 40%; NH<sub>3</sub>: - 19%; PM<sub>2.5</sub>: - 49%). The emission reductions were mainly achieved as a result of the progress in e.g. the use of flue-gas abatement techniques, energy production and distribution, storage and distribution of solvents (Vestreng et al., 2008; EEA, 2014), and vehicle technologies related to legislative “Euro” standards (Sicard et al., 2020a). In EU-28 countries, the “transport” sector is the largest contributor (road transport: 39%) to total NO<sub>x</sub> emissions (EEA, 2019). The Euro-2 to Euro-6 standards for light-duty vehicles were enforced from 1997 to 2015. For diesel cars, the average NO<sub>x</sub> + VOCs limit ranged from 0.70 g/km (Euro-2) to 0.17 g/km (Euro-6), from 1.00 g/km to 0.50 g/km for CO and from 0.08 g/km to 0.0045 g/km for PM. For gasoline cars, the average NO<sub>x</sub> + VOCs limit ranged from 0.500 g/km (Euro-2) and 0.128 g/km (Euro-6) and from 2.2 g/km to 1.0 g/km for CO. In 2017, the successive Euro standards have lowered the PM (94%), CO (50%) and NO<sub>x</sub> + VOCs (76%) emission intensity in the EU compared to early 2000s.

## Trends in urban population exposure

Despite the reduction of PM<sub>10</sub> emissions over the time period 2000–2017, the minimum and maximum percentage of the EU-28 urban population exposed to PM<sub>10</sub> concentrations above the EU daily limit value ranged from 18–44% in 2000–2010 to 13–30% in 2010–2017 (Fig. 1), with the highest extent of exposure observed in 2003 (44%). Between 2000 and 2017, the EU daily limit value for PM<sub>10</sub> was widely exceeded in Europe, mostly in Eastern Europe (Guerreiro et al., 2014), e.g. Bulgaria, Cyprus, Czech Republic, Hungary, Poland, Slovakia, Greece, and Italy. In 2005, Estonia, Finland, Ireland, Luxembourg, and the United Kingdom did not record exceedances of this limit value. In 2017, the limit value was exceeded in Bulgaria, Croatia, Czech Republic, Poland and Italy (EEA, 2011–2019). Before 2006, more than 80% of the EU-28 population was exposed to levels exceeding the WHO AQG value for the protection of human

health, decreasing to 42–52% in 2014–2017 (EEA, 2007; 2011–2019). From 2000 to 2017, the annual averaged  $\text{PM}_{10}$  concentrations decreased by  $0.65 \mu\text{g m}^{-3} \text{ year}^{-1}$  on average at urban stations in the EU-28 (EEA, 2019). In 2010–2017, 6–14% of the EU28 population was exposed to  $\text{PM}_{2.5}$  levels above the EU annual target value, while the range was 16–52% in 2000–2010. The target value was exceeded mostly in Bulgaria, Czech Republic, Poland, and Slovakia between 2000 and 2013. The population exposure to  $\text{PM}_{2.5}$  levels above the WHO AQG ranged from more than 90% before 2006 to 74–80% in 2014–2017. Between 2000 and 2017, the annual averaged concentrations of  $\text{PM}_{2.5}$  decreased by on average  $0.42 \mu\text{g m}^{-3}$  per year at urban background stations in the EU-28 (EEA, 2019).

The percentage of the EU-28 population exposed to  $\text{NO}_2$  concentrations above the EU annual limit value and the WHO AQG value decreased from 14–31% before 2006, with the maximum recorded in 2003, to less than 10% since 2012 (Fig. 1). The annual limit value was mostly exceeded in Italy, Greece, and in the United Kingdom in 2000–2005, and in Germany in 2010, 2011, 2012, 2014, and 2016 (EEA, 2011–2019). The  $\text{NO}_2$  annual mean concentrations decreased by on average  $0.39 \mu\text{g m}^{-3} \text{ year}^{-1}$  over the time period 2002–2011 by joining 708 urban stations in the EU-28 (Guerreiro et al., 2014). The percentage of the EU-28 urban population exposed to  $\text{SO}_2$  levels above the EU daily limit value ranged from 1–2% in 2000–2005 to lower than 0.5% since 2007 (data not shown). The percentage of the EU-28 urban population exposed to  $\text{SO}_2$  levels exceeding the WHO AQG decreased from more than 70% before 2006 to less than 40% since 2013 (EEA, 2011–2019). Less than 2% of the EU-28 urban population was exposed to maximum CO daily 8-hour mean concentrations above the EU and the WHO AQG limit values (data not shown). Only a few traffic stations in Bulgaria, Poland and Romania have reported exceedances of the  $\text{SO}_2$  and CO EU limit values over the time period 2000–2017 (Guerreiro et al., 2014; EEA, 2019).

The EU-28 urban population exposed to  $\text{O}_3$  levels above the EU target value for human health protection ranged from 7–62% since 2000 (Fig. 1), with the highest extent of exposure observed in 2003. Higher background  $\text{O}_3$  levels (annual mean > 30 ppb) were observed in Southern Europe (Sicard et al., 2013). The EU target value was mostly exceeded in Southern Europe, such as Croatia, Cyprus, France, Greece, Italy, Slovenia, Spain, Malta, Portugal, but also in Austria, Hungary, Luxembourg, and Poland recently. More than 95% of the total EU-28 urban population was exposed to  $\text{O}_3$  levels exceeding the WHO AQG since 2000 (data not shown). In the EU, the annual mean of daily  $\text{O}_3$  concentrations increased by on average  $0.05 \text{ ppb year}^{-1}$  at 260 urban stations over the time period 2000–2014 (Table 3). The annual  $\text{O}_3$  mean concentrations increased by on average  $0.34 \text{ ppb year}^{-1}$  at more than 80% of urban stations between 2005 and 2014, except in the United Kingdom where a decrease ( $-0.18 \text{ ppb year}^{-1}$ ) was observed at 65% of urban stations (Sicard et al., 2020a). In Germany, an increase of  $0.18 \text{ ppb year}^{-1}$  was reported at 79 urban stations over the time period 2005–2018 (Sicard et al., 2020a). A significant increase in the annual  $\text{O}_3$  mean (on average,  $+0.29 \text{ ppb year}^{-1}$ ) was found at urban stations in Southern Europe between 2000 and 2010 (Sicard et al., 2013; Kulkarni et al. 2015). In France, an increase of  $+0.14 \text{ ppb year}^{-1}$  at 76% of urban stations was reported between 1999 and 2012 (Sicard et al., 2016b). Despite an increasing fleet

size, the reduction in  $\text{NO}_x$  and VOCs emissions since the early 1990s, due to the vehicle emission regulations, allowed a reduction in  $\text{O}_3$  peaks and high percentiles (EEA, 2016; Sicard et al., 2018; de Foy et al., 2020). At EU-28 urban stations, a reduction in  $\text{O}_3$  annual mean of the maximum daily 8-hour mean values ( $-0.75 \text{ ppb year}^{-1}$ ) was found over the time period 2000–2014 (EEA, 2016). In Southern Europe, significant reductions in 98th percentile ( $-0.51 \text{ ppb year}^{-1}$ ) and hourly maximum ( $-1.81 \text{ ppb year}^{-1}$ ) values were found at urban stations between 2000 and 2010 (Sicard et al., 2013). Simpson et al. (2014) found an increase of  $\text{O}_3$  concentrations of  $0.1\text{--}0.4 \text{ ppb year}^{-1}$  up to the 95th  $\text{O}_3$  percentile over the time period 1990–2009. The surface  $\text{O}_3$  levels are rising in cities in Europe from 2000 (e.g. Simon et al., 2015; Sicard et al., 2016b; Chang et al., 2017; Lefohn et al., 2018; Yan et al., 2019; Sicard et al., 2020a), mainly due to a reduced titration of  $\text{O}_3$  by NO (Huszar et al., 2015; Sicard et al., 2020a).

Table 3

National-averaged trends magnitude (ppb per year  $\pm$  standard deviation) of annual ozone mean concentrations at urban and rural background monitoring stations worldwide. The studies were selected for more than 10-year time-series of ozone data, for stations with at least 75% of validated hourly data over the time period, and with a significant trend, i.e. with a *p-value*  $< 0.05$ . Number of stations (*n*, with  $n \geq 2$ ).

Countries	Time period	References	n	Urban stations	
Europe	1995–2012	Yan et al., 2019	289	+ 0.27 $\pm$ 0.10	
Austria	1995–2014	Sicard et al., 2018	6	+ 0.17 $\pm$ 0.12	
Belgium			2	+ 0.08 $\pm$ 0.15	
Germany			60	+ 0.19 $\pm$ 0.06	
Greece			3	+ 0.18 $\pm$ 0.50	
Netherlands			5	+ 0.19 $\pm$ 0.11	
Slovenia			2	+ 0.14 $\pm$ 0.08	
Spain			12	+ 0.36 $\pm$ 0.24	
Sweden			3	+ 0.37 $\pm$ 0.10	
Switzerland			11	+ 0.28 $\pm$ 0.11	
United Kingdom			12	+ 0.21 $\pm$ 0.12	
France	1999–2012	Sicard et al., 2016b	179	+ 0.14 $\pm$ 0.19	
France	2000–2010	Kulkarni et al., 2015	29	+ 0.10 $\pm$ 0.30	
Greece			Sicard et al., 2013	3	+ 0.41 $\pm$ 0.15
Italy				20	+ 0.04 $\pm$ 0.30
Portugal				8	+ 0.40 $\pm$ 0.33
Spain				14	+ 0.48 $\pm$ 0.53
Europe	2000–2014	Chang et al., 2017		260	+ 0.05 $\pm$ 0.13
Belgium	2005–2014	Sicard et al., 2020a	2	+ 0.42 $\pm$ 0.05	
France			136	+ 0.31 $\pm$ 0.42	
Germany			79	+ 0.09 $\pm$ 0.17	
Greece			4	+ 0.85 $\pm$ 0.43	
Italy			50	+ 0.43 $\pm$ 0.84	
Portugal			2	+ 0.48 $\pm$ 0.12	

Countries	Time period	References	n	Urban stations
Spain			77	+ 0.54 ± 0.73
United Kingdom			29	- 0.18 ± 0.34
Germany	2005–2018	Sicard et al., 2020a	79	+ 0.18 ± 0.15

## Trends in national mortality from exposure to ambient PM<sub>2.5</sub> and O<sub>3</sub> levels

At present compared to other air pollutants, PM<sub>2.5</sub> poses the most serious health risk in the EU-28 cities, associated with premature deaths and increased morbidity, followed by ground-level O<sub>3</sub> (Pascal et al., 2013; Cohen et al., 2017). In the EU-28, the number of deaths due to ambient PM<sub>2.5</sub> levels decreased by on average 4.85 per 1,000,000 inhabitants annually between 2000 and 2017 (Table 4). The highest annual decreases were observed in the United Kingdom and Estonia (- 11.74 and - 10.46 deaths per 10<sup>6</sup> inhabitants, respectively) while a slighter reduction was found in Portugal (- 0.50 deaths per 10<sup>6</sup> inhabitants). In Greece and Lithuania, an increase of annual mortality due to ambient PM<sub>2.5</sub> levels was observed (+ 1.22 and + 1.72 deaths per 10<sup>6</sup> inhabitants, respectively). In line with rising O<sub>3</sub> levels in cities (Sicard et al., 2018, 2020a), the annual O<sub>3</sub>-related number of premature deaths increased in the EU-28 (on average + 0.55 deaths per 10<sup>6</sup> inhabitants). The highest annual decrease of mortality was observed in Greece (+ 2.41 deaths per 10<sup>6</sup> inhabitants), Hungary (+ 2.05 deaths per 10<sup>6</sup> inhabitants) and Czech Republic (+ 1.40 deaths per 10<sup>6</sup> inhabitants), while a non-significant increase was found in Spain (+ 0.03 deaths per 10<sup>6</sup> inhabitants). Between 2000 and 2017, the annual number of deaths attributed to O<sub>3</sub> declined mostly in Northern Europe (e.g. Belgium: - 0.24; Ireland: - 0.30; Lithuania: - 0.23 deaths per 10<sup>6</sup> inhabitants per year) where lower background O<sub>3</sub> levels (annual mean < 20 ppb) were observed (Araminiené et al., 2019; Sicard et al., 2020a).

Table 4

Annual trends of mortality (number of deaths per 1,000,000 inhabitants per year) due to ambient particulate matter with an aerodynamic diameter lower than 2.5  $\mu\text{m}$  ( $\text{PM}_{2.5}$ ) and tropospheric ozone ( $\text{O}_3$ ) over the time period 2000–2017 in the 28 European Union countries (EU-28) with associated significance level  $p$  (Mann-Kendall \*\*\*  $p < 0.001$ ; \*\*  $p < 0.01$ ; \*  $p < 0.05$ ; +  $p < 0.1$  and  $p > 0.1$ ).

EU-28 Countries	$\text{PM}_{2.5}$	<i>p level</i>	$\text{O}_3$	<i>p level</i>
Austria	- 6.00	***	0.18	**
Belgium	- 8.80	***	- 0.24	*
Bulgaria	- 2.73	*	0.56	***
Croatia	- 2.27	+	1.18	***
Cyprus	- 9.07	***	0.14	
Czech Republic	- 4.56	***	1.40	***
Denmark	- 9.42	***	0.18	
Estonia	- 10.46	***	0.20	***
Finland	- 6.55	***	0.22	***
France	- 3.55	***	0.09	
Germany	- 3.11	***	1.19	***
Greece	1.22	*	2.41	***
Hungary	- 1.39		2.05	***
Ireland	- 9.05	***	- 0.30	+
Italy	- 2.28	**	0.75	***
Latvia	- 5.40	*	0.20	***
Lithuania	1.72	+	- 0.23	***
Luxembourg	- 8.05	***	- 0.17	*
Malta	- 0.38		0.32	***
Netherlands	- 8.76	***	0.24	*
Poland	- 9.56	***	0.45	***
Portugal	- 0.50		0.37	***
Romania	- 4.04	***	0.37	***

EU-28 Countries	PM <sub>2.5</sub>	<i>p level</i>	O <sub>3</sub>	<i>p level</i>
<b>Slovakia</b>	- 7.56	***	0.42	***
<b>Slovenia</b>	- 5.74	***	- 0.67	***
<b>Spain</b>	- 5.04	***	0.03	
<b>Sweden</b>	- 8.44	***	0.32	***
<b>United Kingdom</b>	- 11.74	***	0.08	
<b>EU-28</b>	<b>- 4.85</b>	<b>***</b>	<b>0.55</b>	<b>***</b>

## Conclusions

In the EU-28, SO<sub>x</sub>, NO<sub>x</sub>, NMVOCs, NH<sub>3</sub>, CO, PM<sub>2.5</sub> and PM<sub>10</sub> emissions fell by about 80%, 46%, 44%, 10%, 49%, 31% and 29%, respectively, over the time period 2000–2017, thus confirming successful control strategies of air pollutants emissions. However, the current levels of air pollutants in cities continue to exceed the EU standards and WHO AQG for the protection of human health in Europe, especially for secondary air pollutants such as PM<sub>2.5</sub> and O<sub>3</sub> (Guerreiro et al., 2014; De Marco et al., 2018; EEA, 2018; Sicard et al., 2019). In 2015–2017, the percentages of EU-28 urban population exposed to concentrations exceeding the WHO limit values were 74–81% for PM<sub>2.5</sub>, 42–52% for PM<sub>10</sub>, 95–98% for O<sub>3</sub>, 21–31% for SO<sub>2</sub> and 7–8% for NO<sub>2</sub> (EEA, 2019). In agreement with a reduction of ambient PM<sub>2.5</sub> levels in cities, the annual PM<sub>2.5</sub>-related number of deaths decreased (- 4.85 per 10<sup>6</sup> inhabitants) between 2000 and 2017. The control strategies of O<sub>3</sub> precursor emissions were effective in rural areas (Sicard et al., 2013; Paoletti et al., 2014). However, the rising O<sub>3</sub> levels have become a major public health issue in the EU-28 cities (Lefohn et al., 2018; Sicard et al., 2018, 2020a), where the annual O<sub>3</sub>-related number of premature deaths increased (+ 0.55 deaths per 10<sup>6</sup> inhabitants).

There is an urgent need to take decisive actions at all governance levels to achieve the objectives of the Ambient Air Quality Directives as reported by the EC (COM(2018)330 “A Europe that protects: Clean air for all”). These actions span from improving air quality monitoring network, control of emission sources, improved mobility plans and raising awareness to citizens on the problem of air pollution, among others. In this context, urban and peri-urban reforestation can help improve air quality and meet air quality standards in cities (Nowak, 2006; Escobedo et al., 2011; Baró et al., 2014), but also answer to social needs, e.g. recreation, cultural, aesthetic (Selmi et al., 2016; Samson et al., 2019).

Although outside the period of analysis, it is relevant to note that the recent COVID-19 pandemic could represent an opportunity for adopting measures that contribute to improve air quality in European cities in the future. Compared to the same period in 2017–2019, the lockdown measures in 2020 led to a decrease of NO (~ 63%) and NO<sub>2</sub> (~ 52%) concentrations in Southern European cities due to the reduction of road and non-road transport (Sicard et al. 2020b; Tobías et al., 2020). However, these measures did not

significantly reduce the PM<sub>2.5</sub> and PM<sub>10</sub> levels (~ 8%) attributed to an increase of PM emissions from the activities at home (e.g. domestic heating, biomass burning), and during the lockdown, the ground-level O<sub>3</sub> levels increased by ~ 17% due to a lower titration of O<sub>3</sub> by NO (Sicard et al. 2020b). While it is true that “*Air pollution rebounds in Europe’s cities as lockdowns ease*” (Financial Times, 24 June 2020) and that COVID discourages the use of public transport, there are some positive changes that, if sustained over time, might result in improvements of air quality in the cities in the future. Partial or total telework has been implemented in many companies and public offices, a change that will last to certain extent after the COVID pandemic reducing private car mobility. Cities like Barcelona and Paris have widened sidewalks to ensure social distancing on pedestrians, created more bicycle lanes and separated traffic and bus lanes for each direction<sup>4</sup>.

The COVID-19 lockdowns showed us the value of green urban spaces for our physical and mental wellbeing. Greening and re-naturing cities are keywords of the EU Biodiversity Strategy for 2030 (EC COM(2020)380). European Commission calls on European cities of at least 20,000 inhabitants to develop “*ambitious Urban Greening Plans*” by including the promotion of green infrastructure, nature-based solutions, and by planting at least 3 billion additional trees in the EU by 2030. However, to efficiently reduce air pollution in cities, municipalities and city planners urgently need to base the selection of tree species upon quantitative and concrete assessments of the role of urban trees in affecting air quality either positively or negatively (Sicard et al., 2018). Then, the COVID pandemic can be taken as an opportunity for the cities to foster changes in organization of the urban public space and re-think mobility (Honey-Rosés et al., 2020), which hopefully may have relevant and lasting impacts on the quality of urban air.

<sup>4</sup>[http://www.xinhuanet.com/english/2020-05/19/c\\_1139070452.htm](http://www.xinhuanet.com/english/2020-05/19/c_1139070452.htm);

<https://www.rfi.fr/en/france/20200520-france-bicycle-use-jumps-44-percent-since-end-coronavirus-confinement-paris-anne-hidalgo>

## Abbreviations

AP: attributable proportion, AQG: Air Quality Guidelines, BI: baseline incidence, CAPE: The Clean Air Programme for Europe, CLRTAP: Convention on Long-range Transboundary Air Pollution, CO: carbon monoxide, EEA: European Environmental Agency, EMEP: European Monitoring and Evaluation Program, EU: European Union, GDB: Global Burden of Diseases,  $NC_c$ : number of cases attributed to the exposure to the air pollutant  $c$ , NH<sub>3</sub>: ammonia, NMVOCs: non-methane volatile organic compounds, NO<sub>x</sub>: nitrogen oxides, O<sub>3</sub>: tropospheric ozone, OECD: Organization for Economic Co-operation and Development, PM<sub>10</sub>: particulate matter with an aerodynamic diameter lower than 10 µm, PM<sub>2.5</sub>: particulate matter with an aerodynamic diameter lower than 2.5 µm, RR: relative risk, SO<sub>x</sub>: sulfur oxides, WHO: World Health Organization.

## Declarations

### **Ethics approval and consent to participate -**

Not applicable

### **Consent for publication -**

Not applicable

### **Availability of data and materials -**

Not applicable

### **Competing interests -**

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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### **Authors' contributions -**

P.S., V.C., and E.A. conceived the project. P.S., V.C., and E.A. analyzed the data. All authors participated in writing and revising the manuscript.

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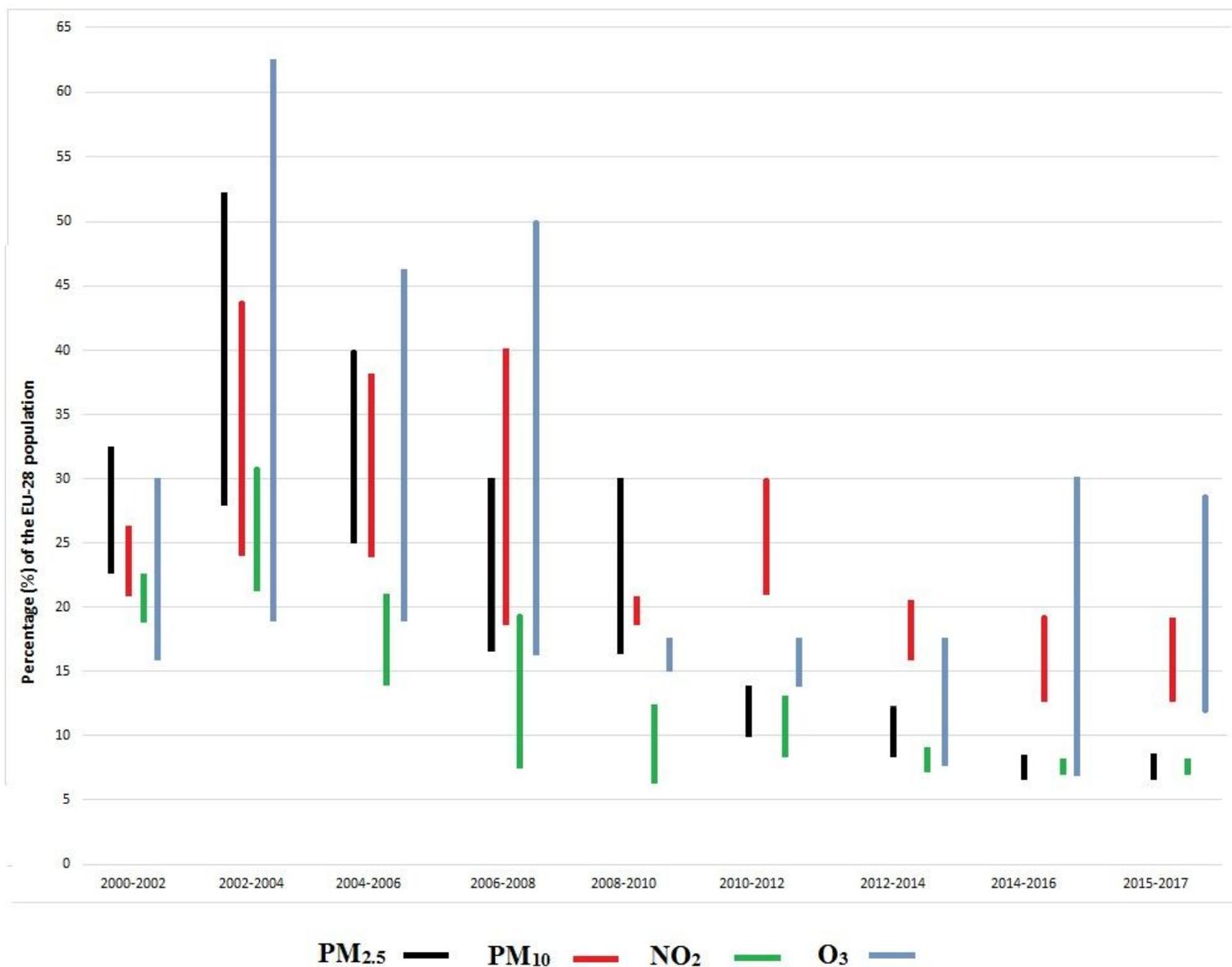
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## Figures



**Figure 1**

Minimum and maximum percentage of EU-28 population (in %) exposed to air pollutants concentrations exceeding the European Union limit or target values between 2000 and 2017.