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# Urban population exposure to air pollution in Europe over the last decades

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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 (European Council 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 2.5 μm and 10 μm: ~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 and the implementation of fresh air corridors can help meet air quality standards, but also answer to social needs, as recently highlighted by the COVID-19 lockdowns.

**Keywords:** Air pollution, EU-28, Mann-kendall test, Population exposure, Risk assessment, Trend

## Background

Outdoor air pollution is a major global public health issue [48], leading to 4.2 million premature deaths worldwide [74] and half a million in the European Union (EU) in 2016 [24]. The EU identifies seven main air pollutants [45]: 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 [33], 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 [9, 55, 75]. 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 [24]. Air pollution also damages plant ecosystems [35, 49, 63], and surface O<sub>3</sub> is considered as the most detrimental air pollutant in terms of effects on vegetation and biodiversity [1, 52, 63].

The legislated ambient air quality standards and the emission control policies (e.g., [10, 18, 77]) control

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

emissions of harmful substances into the atmosphere, and regulate the concentrations of air pollutants such as  $PM_{2.5}$ ,  $PM_{10}$ ,  $NO_2$  and  $O_3$ , by setting limit and target values for the protection of human health Table 1 and requirements to ensure that Member States adequately monitor air quality in a harmonised manner. Therefore, the number of air quality monitoring stations grew rapidly in Europe, by an order of magnitude in 1996, with databases gathering air quality data such as the AirBase system of the European Environment Agency. The number of urban and suburban monitoring stations in Europe ranged from 1300 in 1990 to 3600 in 2000 and about 5000 stations in 2020. 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. 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 [16].

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  $PM_{2.5}$  and  $O_3$  levels over time.

## Materials and methods

### Data collection

The official national emissions of main air pollutants ( $SO_x$ ,  $NH_3$ ,  $PM_{2.5}$ ,  $PM_{10}$ ) and main  $O_3$  precursors ( $NO_x$ , NMVOCs, CO), submitted by the Parties to the LRTAP Convention, were obtained through the Centre on Emission Inventories and Projections (CEIP) under the European Monitoring and Evaluation (EMEP) Program.<sup>2</sup> The EU-28 urban population exposure was estimated by the European Environmental Agency (EEA) from data reported in Airbase, and the number of premature deaths attributed to exposure to ambient  $PM_{2.5}$  and  $O_3$  (per  $10^6$  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.

**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 $\mu g m^{-3}$ )	WHO AQG (threshold in $\mu g m^{-3}$ )
$PM_{10}$ <sup>a</sup>	Annual mean (40)	Annual mean (20)
$PM_{10}$ <sup>a</sup>	Number of exceedance of 24-h mean (50)	Number of exceedance of 24-h mean (50)
$PM_{2.5}$ <sup>b</sup>	Annual mean (25)	Annual mean (10)
$PM_{2.5}$ <sup>b</sup>	–	Number of exceedance of 24-h mean (25)
$O_3$ <sup>c</sup>	Number of exceedance of maximum daily 8-h mean (120)	Number of exceedance of maximum daily 8-h mean (100)
$NO_2$ <sup>d</sup>	Annual mean (40)	Annual mean (40)
$NO_2$ <sup>d</sup>	Number of exceedance of 1-h mean (200)	Number of exceedance of 1-h mean (200)
$SO_2$ <sup>e</sup>	Number of exceedance of 24-h mean (125)	Number of exceedance of 24-h mean (20)
$CO$ <sup>f</sup>	Maximum daily 8-h (10,000)	Maximum daily 8-h (10,000)

<sup>a</sup> Annual mean  $PM_{10}$  concentration and number of days with 24-h  $PM_{10}$  concentration over  $50 \mu g m^{-3}$  for the protection of human health. The annual mean  $PM_{10}$  concentration does not to exceed  $40 \mu g m^{-3}$  (Directive 2008/50/EC) or  $20 \mu g m^{-3}$  (WHO AQG). The 24-h  $PM_{10}$  mean concentration does not to exceed  $50 \mu g m^{-3}$  (WHO AQG) or more than 35 times a year (EC)

<sup>b</sup> Annual mean  $PM_{2.5}$  concentration and numbers of days with 24-h  $PM_{2.5}$  mean concentration over  $25 \mu g m^{-3}$  (WHO AQG). The annual mean  $PM_{2.5}$  concentration does not to exceed  $25 \mu g m^{-3}$  (EC) or  $10 \mu g m^{-3}$  (WHO AQG)

<sup>c</sup> For the protection of human health, the Directive 2008/50/EC has introduced a threshold of  $120 \mu g m^{-3}$  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  $O_3$  concentrations over  $100 \mu g m^{-3}$  as limit value for the protection of human health (WHO AQG)

<sup>d</sup> Annual mean  $NO_2$  concentration and number of hours with  $NO_2$  concentrations above  $200 \mu g m^{-3}$ . The annual mean  $NO_2$  concentration does not to exceed  $40 \mu g m^{-3}$  (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  $SO_2$  mean concentration does not to exceed  $125 \mu g m^{-3}$  more than 3 times a year (EC) and does not to exceed  $20 \mu g m^{-3}$  (WHO AQG). <sup>f</sup> The Directives have introduced a threshold of  $10 mg m^{-3}$  for the maximum daily 8-h mean concentration

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

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

### Estimation of urban population exposure

For each city included in the Urban Audit,<sup>4</sup> the EU-28 urban population exposure to air pollutants above the EU limit values and WHO AQG was estimated by combining the concentration maps, from measured concentrations at urban and suburban background monitoring stations with more than 75% of validated hourly data per year, with the population density, and considering that the entire population is potentially exposed to the averaged concentrations, i.e., excluding human mobility [22–32]. The estimation of population exposure was based on data from about 1300 stations in 2000 to 3100 stations in 2017 in EU-28 countries.

### 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 [36] and widely used for the health risk assessment of air pollution [2, 3, 12, 37, 42–44, 61].

WHO set daily maximum 8-h concentrations for O<sub>3</sub> and 24-h average concentration for PM<sub>2.5</sub> as metrics to represent the mean daily exposure of population [76]. The daily population exposure to O<sub>3</sub> and PM<sub>2.5</sub> is estimated by combining concentrations maps from satellite and modeled data, and calibrated by ground measurements, with epidemiological data including relative risk values and baseline incidence rates [36]. 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  $\mu\text{g m}^{-3}$  of the air pollutant  $c$  concentration [73].

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

The demographic data were taken from Eurostat [34], and the mortality data and BI were obtained by WHO [72]. The RR values were obtained from exposure–response functions, based on epidemiological studies,

following recommendations from the Health Risks of Air Pollution in Europe project, and published by WHO [75]. For the non-accidental mortality (all ages), RR = 1.0123 and RR = 1.0029 are reported for PM<sub>2.5</sub> and O<sub>3</sub>, respectively, i.e., for instance, a 10  $\mu\text{g m}^{-3}$  increase in the 24-h average PM<sub>2.5</sub> concentration is associated with a 1.2% increase in the risk for mortality attributed to non-accidental causes. However, the use of RR values and BI data from local (or national) epidemiological studies is recommended to obtain robust results.

### Statistical estimation of annual trends

A 10-year time-series is considered long enough to assess short-term changes [66]. 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 [38, 65]. 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. In this study, we used MAKESENS program version 1.0 [56]. Results were considered significant at  $p < 0.05$ .

### Literature review

To report robust short-term air pollutants changes over the last 2 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  $< 0.05$ .

## Results and discussion

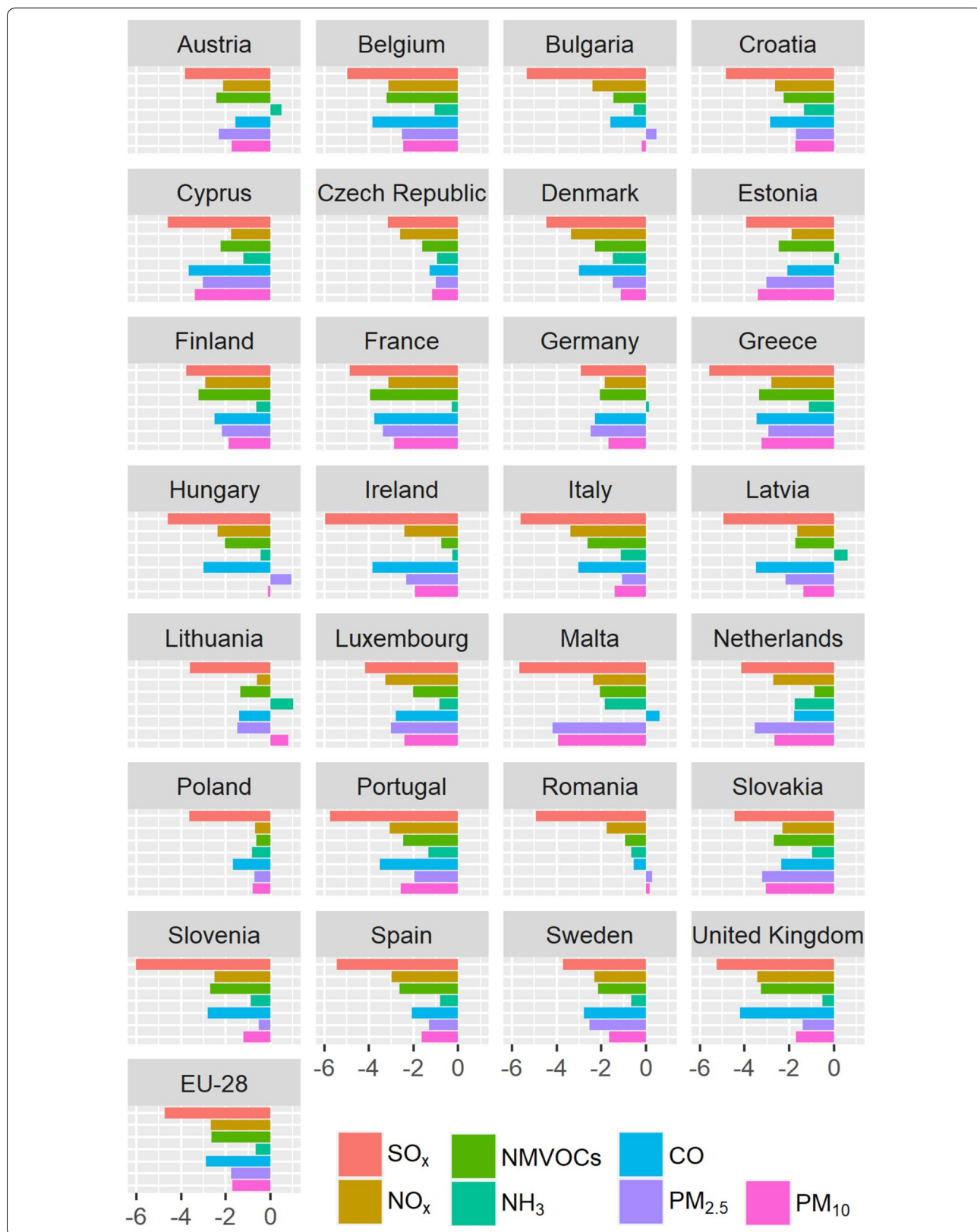
### Trends in national emissions

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,

(See figure on next page.)

**Fig. 1** 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  $\mu\text{m}$  and 10  $\mu\text{m}$  (PM<sub>2.5</sub> and PM<sub>10</sub>) over the time period 2000–2017 (see Additional file 1: Table S1 for raw data). All trends are significant at  $p < 0.05$  (Mann–Kendall)

<sup>4</sup> <https://ec.europa.eu/eurostat/web/cities/data/database>.



over the time period 2000–2017 in the EU-28 countries (Fig. 1). The  $\text{SO}_x$  emissions decreased in all EU-28 countries, from  $-2.9\%$  year<sup>-1</sup> (Germany) to  $-6.0\%$  year<sup>-1</sup> (Slovenia). For  $\text{NO}_x$ , the highest decrease was observed in the United Kingdom ( $-3.4\%$  year<sup>-1</sup>), while the lowest reduction was found in Lithuania ( $-0.6\%$  year<sup>-1</sup>) and Poland ( $-0.7\%$  year<sup>-1</sup>). For NMVOCs, the decrease ranged from  $-0.6\%$  year<sup>-1</sup> (Poland) to  $-4.0\%$  year<sup>-1</sup> (France). In general, small reductions were exhibited in the agricultural sector, contributing to 92% of  $\text{NH}_3$  emissions [22], but an increase could be determined in Austria, Estonia, Germany, Latvia and Lithuania, ranging from 0.1 to 1.0% per year. The domestic heating represents 48% of CO emissions [22]. Also, the CO emissions usually decreased, except in Malta ( $+0.6\%$  year<sup>-1</sup>). A decrease of  $\text{PM}_{2.5}$  emissions was observed in all EU-28 countries, except Bulgaria ( $+0.5\%$  year<sup>-1</sup>), Hungary ( $+0.9\%$  year<sup>-1</sup>) and Romania ( $+0.3\%$  year<sup>-1</sup>), associating with a slighter reduction in  $\text{PM}_{10}$  emissions ( $-0.2\%$  year<sup>-1</sup> in Bulgaria,  $-0.1\%$  year<sup>-1</sup> in Hungary). An increase of  $\text{PM}_{10}$  emissions was noted in Lithuania ( $+0.8\%$  year<sup>-1</sup>) and Romania ( $+0.1\%$  year<sup>-1</sup>). The highest decrease for  $\text{PM}_{2.5}$  ( $-4.2\%$  year<sup>-1</sup>) and  $\text{PM}_{10}$  ( $4.0\%$  year<sup>-1</sup>) emissions occurred in Malta (Fig. 1).

The emissions of all primary air pollutants contributing to ambient levels of PM,  $\text{O}_3$ , and  $\text{NO}_2$  decreased between 2000 and 2017 in the EU-28 (observed reductions  $\text{SO}_x$ :  $-80\%$ ;  $\text{NO}_x$ :  $-46\%$ ; NMVOCs:  $-44\%$ ;  $\text{NH}_3$ :  $-10\%$ ; CO:  $-49\%$ ;  $\text{PM}_{2.5}$ :  $-31\%$ ;  $\text{PM}_{10}$ :  $-29\%$ ), in line with stringent EC Directives, e.g. Air Quality Framework Directive [21], Large Combustion Plant Directive [19], and National Emission Ceilings Directives [17, 20], setting emission reduction commitments by 2030 compared to 2005 (expected reductions  $\text{SO}_2$ :  $-79\%$ ,  $\text{NO}_x$ :  $-63\%$ , NMVOCs:  $-40\%$ ,  $\text{NH}_3$ :  $-19\%$ ;  $\text{PM}_{2.5}$ :  $-49\%$ ). 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 [28, 71], and vehicle technologies related to legislative “Euro” standards [59].

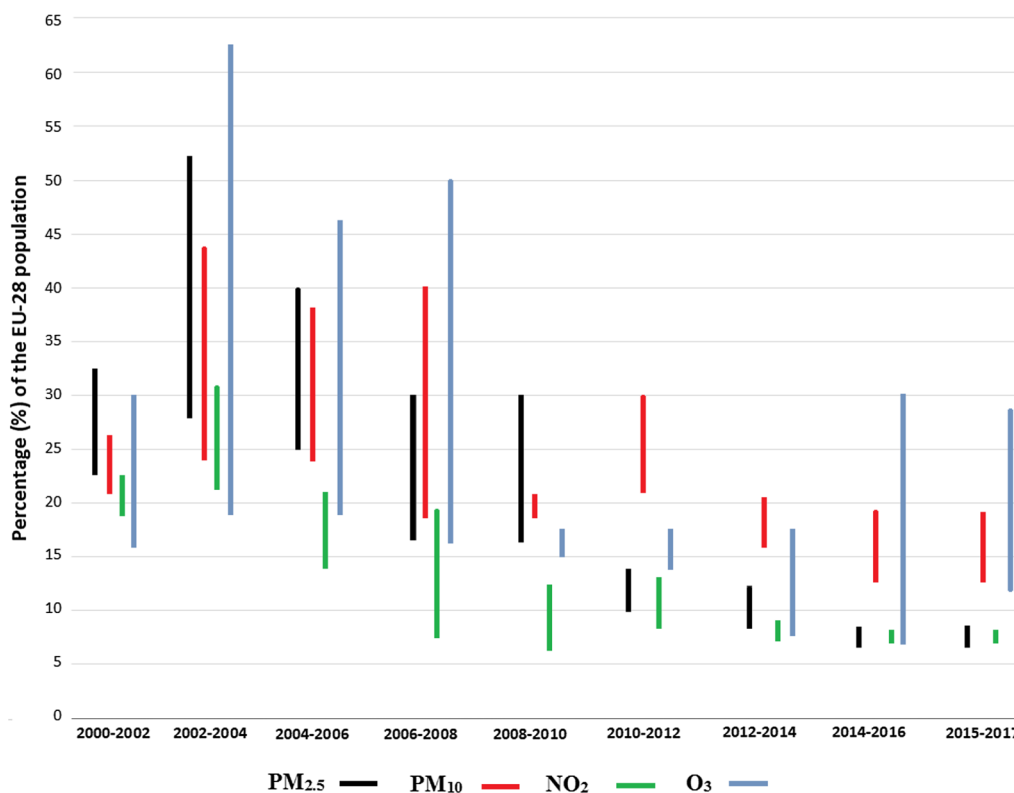
In EU-28 countries, the “on-road transport” sector is the largest contributor to total  $\text{NO}_x$  emissions (road transport: 40–55%), and represents 8–15% of VOCs emissions [22]. Diesel-powered motor vehicles account for about 91% of the fleet (from 81% in Czech Republic to 99% in Portugal) in all EU countries except for Greece (37%), and gasoline-powered motor vehicles account for about 7% of the fleet [41]. The Euro-2 to Euro-6 standards for light-duty vehicles were enforced from 1997 to 2015. For diesel cars, the average  $\text{NO}_x + \text{VOCs}$  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

$\text{NO}_x + \text{VOCs}$  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  $\text{NO}_x + \text{VOCs}$  (76%) emission intensity in the EU compared to early 2000s. An investigation by Breuer et al. [7] in Germany showed that 91% of road transport  $\text{NO}_x$  emissions are produced by diesel-powered motor vehicles. At national level, emissions of  $\text{NO}_x$  from on-road transport decreased in all EU countries (from  $-0.81\%$  year<sup>-1</sup> in Lithuania to  $-4.29\%$  year<sup>-1</sup> in Finland) except in Poland ( $+1.51\%$  year<sup>-1</sup>) and Romania ( $+1.17\%$  year<sup>-1</sup>) between 2000 and 2017 (Additional file 1: Table S1). Investigations on  $\text{NO}_x$  emissions by diesel cars showed that, on average, their real-world  $\text{NO}_x$  emissions are seven times the limit of 0.08 g/km mandated by the Euro 6 standard [41]. Therefore, the reported reduction of  $\text{NO}_x$  emissions ( $-46\%$ ) can be overestimated compared to the real-world  $\text{NO}_x$  emissions.

#### Trends in urban population exposure

Despite the reduction of  $\text{PM}_{10}$  emissions over the time period 2000–2017, the minimum and maximum percentage of the EU-28 urban population exposed to  $\text{PM}_{10}$  concentrations above the EU daily limit value ranged from 18 to 44% in 2000–2010 to 13–30% in 2010–2017 (Fig. 2), with the highest extent of exposure observed in 2003 (44%). Between 2000 and 2017, the EU daily limit value for  $\text{PM}_{10}$  was widely exceeded in Europe, mostly in Eastern Europe [38], e.g., Bulgaria, Cyprus, Czech Republic, Hungary, Poland, Slovakia, Greece, and Italy. In 2017, the EU daily limit value was exceeded in Bulgaria, Croatia, Czech Republic, Poland and Italy [22, 31]. Before 2006, more than 80% of the EU-28 population was exposed to  $\text{PM}_{10}$  levels exceeding the WHO AQG, reaching 42–52% in 2014–2017 (Additional file 1: Table S2). From 2000 to 2017, the annual averaged  $\text{PM}_{10}$  concentrations decreased by  $0.65 \mu\text{g m}^{-3}$  year<sup>-1</sup> on average at urban stations in the EU-28 [22]. In 2010–2017, 6–14% of the EU-28 population was exposed to  $\text{PM}_{2.5}$  levels above the EU annual target value, while the range was 16–52% in 2000–2010 (Fig. 2). 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 (Additional file 1: Table S2). 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 [22].

The percentage of the EU-28 population exposed to  $\text{NO}_2$  concentrations above the EU annual limit value and the WHO AQG decreased from 14 to 31% before 2006,



**Fig. 2** Minimum and maximum percentage of EU-28 population (in %) exposed to air pollutants concentrations (particulate matter PM<sub>2.5</sub> and PM<sub>10</sub>, nitrogen dioxides NO<sub>2</sub> and tropospheric ozone O<sub>3</sub>) exceeding the European Union limit or target values between 2000 and 2017 (see Additional file 1: Table S2; data source: [22–32])

with the maximum recorded in 2003, to less than 10% since 2012 (Fig. 2). The annual limit value was mostly exceeded in Italy, Greece, and in the United Kingdom in 2000–2005, and in Germany in 2010–2016 [22–31]. The NO<sub>2</sub> 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 [38]. The percentage of the EU-28 urban population exposed to SO<sub>2</sub> levels above the EU daily limit value ranged from 1 to 2% in 2000–2005 to lower than 0.5% since 2007 (data not shown). The percentage of the EU-28 urban population exposed to SO<sub>2</sub> levels exceeding the WHO AQG decreased from more than 70% before 2006 to less than 40% since 2013 [22–31]. Less than 2% of the EU-28 urban population was exposed to maximum CO daily 8-h mean concentrations above the EU and the WHO AQG (data not shown). Only a few traffic stations in Bulgaria, Poland and Romania have reported exceedances of the SO<sub>2</sub> and CO EU limit values over the time period 2000–2017 [22, 38].

The EU-28 urban population exposed to O<sub>3</sub> levels above the EU target value for human health protection ranged from 7 to 62% since 2000 (Fig. 2), with the

highest extent of exposure observed in 2003. As for NO<sub>2</sub> and PM<sub>10</sub>, the maximum O<sub>3</sub> concentrations were observed in 2003, due to extremely warm summer in Europe, with a heatwave occurred in August, and stagnant weather conditions leading to accumulation of air pollutants [70]. The EU target value was mostly exceeded in Southern Europe, where higher background O<sub>3</sub> levels (annual mean > 30 ppb) are observed [65], 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 O<sub>3</sub> levels exceeding the WHO AQG since 2000 (Additional file 1: Table S2). In the EU, the annual mean of daily O<sub>3</sub> concentrations increased by on average 0.05 ppb year<sup>-1</sup> at 260 urban stations over the time period 2000–2014 (Table 2). The annual O<sub>3</sub> mean concentrations increased by on average 0.34 ppb year<sup>-1</sup> at more than 80% of urban stations between 2005 and 2014, except in the United Kingdom where a decrease (−0.18 ppb year<sup>-1</sup>) was observed at 65% of urban stations [59]. In Germany, an increase of 0.18 ppb year<sup>-1</sup> was reported at 79 urban stations over the time period 2005–2018 [59]. A significant increase in the annual O<sub>3</sub>

**Table 2** National-averaged trends magnitude (ppb per year  $\pm$  standard deviation) of annual ozone mean concentrations at urban and rural background monitoring stations in Europe

Countries	Time period	References	<i>n</i>	Urban stations
Europe	1995–2012	[78]	289	+0.27 $\pm$ 0.10
Austria	1995–2014	[62]	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	[64]	179	+0.14 $\pm$ 0.19
France	2000–2010	[46, 65]	29	+0.10 $\pm$ 0.30
Greece			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	[8]	260	+0.05 $\pm$ 0.13
Belgium	2005–2014	[59]	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
Spain			77	+0.54 $\pm$ 0.73
United Kingdom			29	-0.18 $\pm$ 0.34
Germany	2005–2018	[59]	79	+0.18 $\pm$ 0.15

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)

mean (on average, +0.29 ppb year<sup>-1</sup>) was found at urban stations in Southern Europe between 2000 and 2010 [46, 65]. In France, an increase of +0.14 ppb year<sup>-1</sup> at 76% of urban stations was reported between 1999 and 2012 [64]. Despite an increasing fleet size, the reduction in NO<sub>x</sub> and VOCs emissions since the early 1990s, due to the vehicle emission regulations, allowed a reduction in O<sub>3</sub> peaks and high percentiles [11, 26, 62]. At EU-28 urban

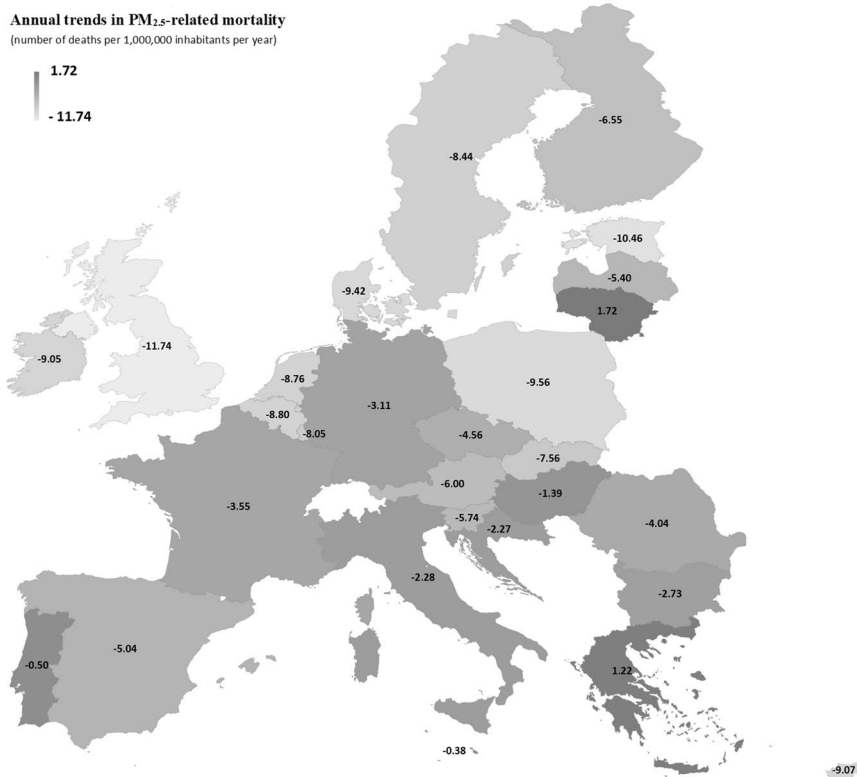
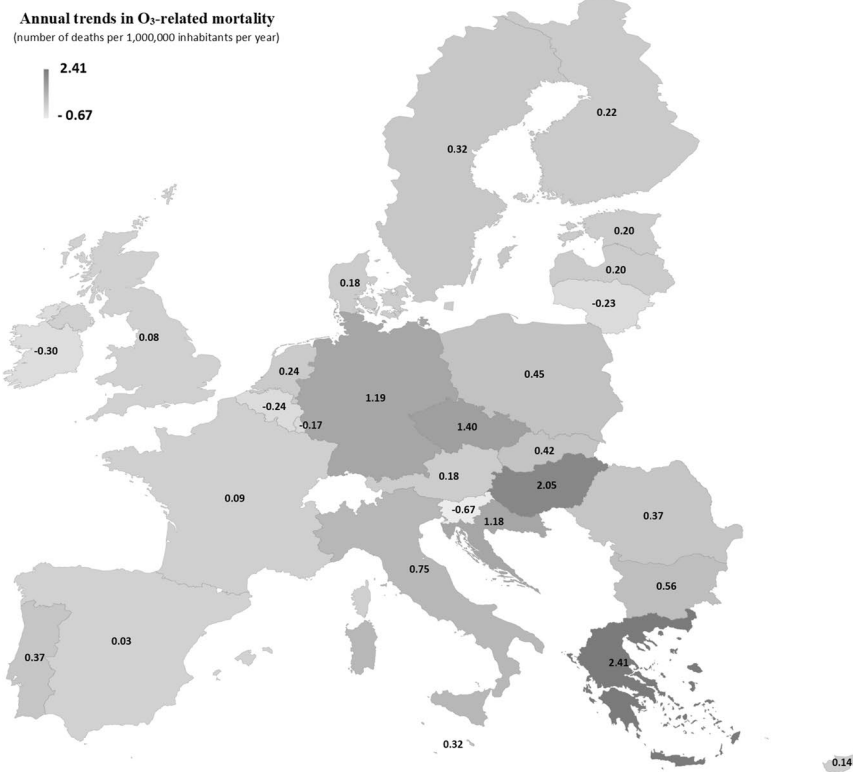
stations, a reduction in O<sub>3</sub> annual mean of the maximum daily 8-h mean values (-0.75 ppb year<sup>-1</sup>) was found over the time period 2000–2014 [26]. In Southern Europe, significant reductions in 98th percentile (-0.51 ppb year<sup>-1</sup>) and hourly maximum (-1.81 ppb year<sup>-1</sup>) values were found at urban stations between 2000 and 2010 [65]. Simpson et al. [68] found an increase of O<sub>3</sub> concentrations of 0.1–0.4 ppb year<sup>-1</sup> up to the 95th O<sub>3</sub> percentile over the time period 1990–2009. The surface O<sub>3</sub> levels are rising in cities in Europe from 2000 (e.g., [8, 47, 59, 64, 67, 78]), mainly due to a reduced titration of O<sub>3</sub> by NO [40, 59].

### 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> [9, 55]. 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 (Fig. 3). 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 [59, 62], 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 [4, 59]. In this study, only the outdoor exposure to air pollution was considered while people spend about 80–90% of time in indoor environments [54]. As the spatio-temporal

(See figure on next page.)

**Fig. 3** 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  $\mu$ m (PM<sub>2.5</sub>) and tropospheric ozone (O<sub>3</sub>) over the time period 2000–2017 in the 28 European Union countries (EU-28). Points below the thick line show a decrease in O<sub>3</sub>- and PM<sub>2.5</sub>-related mortality, while points above the thick line show an increase (see Additional file 1: Table S3 for raw data)





variability of air pollutants levels and human mobility were ignored, the individual exposure estimates are slightly biased.

### Conclusions

Between 2000 and 2017, the EU-28 emissions fell for  $\text{SO}_x$  by about 80%,  $\text{NO}_x$ : 46%, NMVOCs: 44%,  $\text{NH}_3$ : 10%, CO: 49%,  $\text{PM}_{2.5}$ : 31%, and  $\text{PM}_{10}$ : 29%. This confirms 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 the secondary air pollutant  $\text{O}_3$  [12, 23, 38, 61]. In 2015–2017, the percentages of EU-28 urban population exposed to concentrations exceeding the WHO limit values were 74–81% for  $\text{PM}_{2.5}$ , 42–52% for  $\text{PM}_{10}$ , 95–98% for  $\text{O}_3$ , 21–31% for  $\text{SO}_2$  and 7–8% for  $\text{NO}_2$  [22]. In agreement with a reduction of ambient  $\text{PM}_{2.5}$  levels in cities, the annual  $\text{PM}_{2.5}$ -related number of deaths decreased (– 4.85 per  $10^6$  inhabitants) between 2000 and 2017. The control strategies of  $\text{O}_3$  precursor emissions were effective in rural areas [53, 65]. However, the rising  $\text{O}_3$  levels have become a major public health issue in the EU-28 cities [47, 59, 62], where the annual  $\text{O}_3$ -related number of premature deaths increased (+ 0.55 deaths per  $10^6$  inhabitants) over the time period 2000–2017.

Barmadimos et al. [5] have reported a positive correlation between  $\text{PM}_{10}$  and air temperature in summer (e.g., higher emissions from agriculture), and negative in winter (e.g., lower emissions by tertiary sector for heating). In Europe, the average annual air temperature increased by 0.22–0.40 °C per decade since 1965 [24]. The highest air temperature increase was observed over Eastern and Northern Europe in winter, and over Southern Europe in summer (EEA, 2018b). Climate change is projected to reduce the benefits of PM and  $\text{O}_3$  precursor emissions controls leading to higher PM and  $\text{O}_3$  levels.

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 [15]. 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 and an implementation of fresh air corridors can help improve air quality and meet air quality standards in cities [6, 13, 51], but also answer to social needs, e.g., recreation, cultural, aesthetic [57, 58]. The cold air corridors are needed to reduce the climatic extreme events in large cities, which can lead to air pollution peaks.

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  $\text{NO}_2$  (~ 52%) concentrations in Southern European cities due to the reduction of road and non-road transport [60, 69]. However, these measures did not significantly reduce the  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  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  $\text{O}_3$  levels increased by ~ 17% due to a lower titration of  $\text{O}_3$  by NO [60]. 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>5</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 [14]. 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. 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 [39], which hopefully may have relevant and lasting impacts on the quality of urban air. 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 [62]. For improving air quality and thermal comfort in cities, tree planting programs need to: (a) plant and sustain healthy trees by selecting a diversity species well adapted to local conditions, (b) avoid species sensitive to air pollution, (c) use low VOCs and pollen emitting trees, (d) supply ample water to vegetation; (e) use long-lived and low maintenance species; and (f) implement cold air corridor in large cities to minimize the health risk of air pollutants [50, 62].

<sup>5</sup> [http://www.xinhuanet.com/english/2020-05/19/c\\_139070452.htm](http://www.xinhuanet.com/english/2020-05/19/c_139070452.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; ICCT: International Council on Clean Transportation;  $NC_c$ : Number of cases attributed to the exposure to the air pollutant  $c$ ;  $NH_3$ : Ammonia; NMVOCs: Non-methane volatile organic compounds;  $NO_x$ : Nitrogen oxides;  $O_3$ : Tropospheric ozone; OECD: Organization for Economic Co-operation and Development;  $PM_{10}$ : Particulate matter with an aerodynamic diameter lower than 10  $\mu m$ ;  $PM_{2.5}$ : Particulate matter with an aerodynamic diameter lower than 2.5  $\mu m$ ; RR: Relative risk;  $SO_x$ : Sulfur oxides; WHO: World Health Organization.

## Supplementary Information

The online version contains supplementary material available at <https://doi.org/10.1186/s12302-020-00450-2>.

**Additional file 1: Table S1.** 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>), on-road transport NO<sub>x</sub> (NO<sub>x\_road</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  $\mu m$  and 10  $\mu m$  (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-Kendal). The increasing trends are in bold. **Table S2.** Minimum and maximum percentage of EU-28 population (in %) exposed to air pollutants concentrations (tropospheric ozone O<sub>3</sub>, nitrogen dioxides NO<sub>2</sub>, particulate matter PM<sub>2.5</sub> and PM<sub>10</sub>) exceeding the European Union (EU) and World Health Organization Air Quality Guidelines (WHO AQG) limit or target values between 2000 and 2017. **Table S3.** 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  $\mu m$  (PM<sub>2.5</sub>) and tropospheric ozone (O<sub>3</sub>) over the time period 2000–2017 in the 28 European Union countries (EU-28) with associated significance level  $p$  (Mann-Kendal \*\*\*  $p < 0.001$ ; \*\*  $p < 0.01$ ; \*  $p < 0.05$ ; +  $p < 0.1$  and  $p < 0.1$ ).

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## Authors' contributions

PS, VC, and EA conceived the project. PS, VC, and EA analyzed the data. All authors participated in writing and revising the manuscript.

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## Availability of data and materials

Not applicable.

## Ethics approval and consent to participate

Not applicable.

## Consent for publication

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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