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ORIGINAL PAPER

Trends in tropospheric ozone concentrations and forest impact metrics in Europe over the time period 2000–2014

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Abstract In Europe, tropospheric ozone pollution appears as a major air quality issue, and ozone concentrations remain potentially harmful to vegetation. In this study we compared the trends of two ozone metrics widely used for forests protection in Europe, the AOT40 (Accumulated Ozone over Threshold of 40 ppb) which only depends on surface air ozone concentrations, and the Phytotoxic Ozone Dose which is the accumulated ozone uptake through stomata over the growing season, and above a threshold Y of uptake (PODY). By using a chemistry transport model, we found that European-averaged ground-level ozone concentrations (−2%) and AOT40 metric (−26.5%) significantly declined from 2000 to 2014, due to successful control strategies to reduce the emission of ozone precursors in Europe since the early 1990s. In contrast, the stomatal ozone uptake by forests increased from 17.5 to 26.6 mmol O_3 m^{-2} despite the reduction in ozone concentrations, leading to an increase of potential ozone damage on plants in Europe. In a climate change context, a biologically-sound stomatal flux-based standard (PODY) as new European legislative standard is needed.

Keywords Tropospheric ozone · AOT40 · POD · Trendm · Mann–Kendall test

Introduction

Tropospheric ozone (O_3) is a secondary short-lived climate pollutant (Shindell et al. 2012), formed by the photochemical oxidation of NO_x in the presence of carbon monoxide (CO), methane (CH_4) and volatile organic compounds (VOCs) (Chameides et al. 1988). It is also the third most important greenhouse gas in terms of radiative forcing (Mickley et al. 2001). Despite the implementation of legislative standards to control the emission of O_3 precursors worldwide (Cooper et al. 2014; Monks et al. 2015; Simon et al. 2015; Sicard et al. 2016a), O_3 concentrations remain potentially harmful to vegetation over some regions around the world (Sicard et al. 2016a, 2017; Cailleret et al. 2018; Mills et al. 2018). In Europe, surface O_3 pollution appears as a major air quality issue (Sicard et al. 2013, 2018, 2020a,b; EEA 2018), particularly in Southern Europe where road traffic and industrial emissions, combined with higher solar radiation, enhance O_3 formation (Millán et al. 2000), and causes threat to vegetation (e.g. Sanz et al. 2000; Paoletti 2006; Wittig et al. 2009; Anav et al. 2011; Mills et al. 2011; Sicard et al. 2016b). Currently, the European standard used to protect vegetation against negative impacts of O_3 is the Accumulated Ozone over a Threshold of 40 ppb (AOT40), i.e. the cumulative exposure to hourly O_3 concentrations above 40 ppb over the daylight hours of the growing season (Directive 2008/50/EC). In Europe, a target value of 9,000 ppb h,

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averaged over 5 years, is recommended by the Directive 2008/50/CE for the vegetation protection whilst a critical level of 5,000 ppb h is recommended by UNECE (2017) for forest protection. Although AOT40 metric is widely used, the O_3 uptake through stomata is a better metric to assess plant damage because it estimates the quantity of O_3 entering in the leaf tissues (Musselman et al. 2006; De Marco et al. 2015; Sicard et al. 2016c). The Phytotoxic Ozone Dose above a threshold Y of uptake (PODY) is the accumulated stomatal O_3 flux over the growing season and can be modelled using the Deposition of Ozone and Stomatal Exchange (DO3SE) model (UNECE 2017). The threshold Y represents a detoxification threshold below which any O_3 molecule absorbed by the plant is detoxified (CLRTAP 2017). High ambient O_3 levels may not damage plants when stomata are closed (Ronan et al. 2020). Conversely, high PODY and resulting damages can occur at low O_3 levels when stomata are open under favourable environmental conditions such as optimal air temperature and soil moisture (Ronan et al. 2020). For these reasons, the stomatal flux-based approach is recommended as more realistic compared to the exposure-based approach (Paoletti and Manning 2007; Sicard et al. 2016c; Agathokleous et al. 2018).

The evaluation of temporal trends in air pollutant levels in European Union (EU) countries is an essential tool to assess the improvement of air quality due to emissions control strategies (Guerreiro et al. 2014). To date, many studies have investigated O_3 trends for a small number of monitoring stations, in particular at rural sites representative of background O_3 conditions (De Leeuw 2000). In 2016, a report was published by the co-operative programme for monitoring and evaluation of the long-range transport of air pollutants in Europe (EMEP) focusing on background sites showing the evolution of ground-level O_3 over the time period 1990–2012 (Colette et al. 2017). The report highlighted a relatively flat trend for annual O_3 mean concentrations at EMEP background stations whilst a reduction of 37% for AOT40 was found between 2002 and 2012. Sicard et al. (2016a) found a decline (−27%) for O_3 vegetation impact metrics at 332 background stations in France between 1999 and 2012. Similarly, Araminienė et al. (2019) found a decreasing trend over the time period 2001–2014 for O_3 annual mean (−1.3%) and AOT40 (−16%) in Lithuania whereas they found an increase for POD0 (+2.9%). Anav et al. (2019) found a decreasing trend of AOT40 (−22%) and O_3 concentrations (−1.6%) and a slight increase of POD0 (+5.9%) in Europe over the time period 2000−2014. In this study, we performed a spatio-temporal analysis of short-term annual trends in O_3 exposure-based and flux-based metrics for the protection of forests for all European countries over the time period 2000–2014.

Materials and methods

Environmental data: the WRF-CHIMERE modelling system

Hourly air temperature data and O_3 concentrations were obtained, respectively, from the Weather Research and Forecasting (WRF), a mesoscale meteorological model (Skamarock and Klemp 2008) and CHIMERE, an Eulerian offline chemistry-transport model developed to analyse the gas-phase chemistry, aerosol formation, transport and deposition at regional scale. Data were provided at 1-h temporal resolution and 12 km × 12 km of spatial resolution over the time period 2000–2014. The O_3 concentrations at 20–25 m of height from the ground (top of the canopy) provided by the CHIMERE model were used to calculate AOT40 and PODY. Further information about the validation of data obtained by the WRF-CHIMERE modelling system can be found in Menut et al. (2013), Martin et al. (2014) and Anav et al. (2016).

Calculation of O_3 metrics

AOT40 calculation

The O_3 exposure index AOT40 (in ppb hours, abbreviated to ppb h) was calculated as sum of the hourly exceedances above 40 ppb, for daylight hours (8 am–8 pm) during the growing season, i.e. 1st April-30th September for the protection of forest trees (UNECE 2010), according to the methodology for O_3 risk assessment in Europe.

$$\mathrm{AOT40} = \int_{i=1}^{n} \max\left(\left([O3] - 40\right), 0\right).dt \tag{1}$$

where $[O_3]$ is hourly O_3 concentration (ppb), n denotes the number of hours to be included in the calculation period and *dt* is time step (1-h). The function "maximum" ensures that only values exceeding 40 ppb are taken into account. In Europe, the critical values for the protection of forests is 5000 ppb h as recommended by UNECE (2010).

However, Klingberg et al. (2014) showed that AOT40 does not consider the influence of climate change on the growing season duration. By consequence, we used in this study the AOT40 formula proposed by Anav et al. (2016) that is more plausible from a physiological point of view as the revised AOT40 was calculated from 1st January to 31st December for hours with stomatal conductance (g_{sto}) higher than 0:

$$\mathrm{AOT40} = \int_{t=1Jan}^{31Dec} \max\left(\left([O3] - 40\right), 0\right).dt; gsto > 0 \tag{2}$$

where $[O_3]$ is the hourly O_3 concentration (ppb), *dt* is the time step (1-h) and g_{sto} is the stomatal conductance computed according to Eq. (3). However, AOT40 does not provide any information on the O_3 uptake by leaves (Anav et al. 2016).

Phytotoxic ozone dose calculation

The PODY was calculated using the DO_3SE model, based on the multiplicative Jarvis' algorithm (Jarvis 1976) for estimation of g_{sto} (mmol O_3 m^{-2} s^{-1}). The g_{sto} is calculated as a species-specific function where the maximum value of stomatal conductance (g_{max}) is reduced by limiting functions, scaled from 0 to 1 as described in Eq. 3.

$$g_{sto} = g_{max} \times f_{phen} \times f_{light} \times max\{f_{min}, (f_{temp} \times f_{VPD} \times f_{SWC})\} \tag{3}$$

where g_{max} is the maximum stomatal conductance of a plant species to O_3 (mmol O_3 m^{-2} s^{-1} per leaf area). The functions f_{phen}, f_{light}, f_{temp}, f_{VPD} and f_{SWC} stand for the g_{max} variation with leaf age, photosynthetically flux density at the leaf surface (PPFD, µmol photons m^{-2} s^{-1}), surface air temperature, (T, °C), vapor pressure deficit (VPD, kPa) estimated through the surface air humidity, and volumetric soil water content (SWC, m^3 m^{-3}), respectively. The function $f_{\min}$ is the minimum g_{sto} expressed as a fraction of g_{max}. We assumed that f_{phen} was 1 throughout the growing season. The following formulas were applied:

$$f_{light} = 1 - \exp(-light_a * PPFD) \tag{4}$$

$$f_{temp} = \left(\frac{T - T_{min}}{T_{opt} - T_{min}}\right)\left\{\frac{T_{max} - T}{T_{max} - T_{opt}}^{\left(\frac{T_{max}-T_{opt}}{T_{opt}-T_{min}}\right)}\right\} \tag{5}$$

$$f_{VPD} = \min\left[1, \max\left\{f_{\min}, \left(\frac{(1 - f_{\min}) * (VPD_{\min} - VPD)}{(VPD_{\min} - VPD_{\max})}\right) + f_{\min}\right\}\right] \tag{6}$$

$$f_{SWC} = \min\left[1, \left(f_{\min}, \left((1 - f_{\min}) * \left(\frac{SWC - WP}{FC - WP}\right) + f_{\min}\right)\right)\right] \tag{7}$$

where $light_a$ is an adimensional constant; PPFD is hourly photosynthetic photon flux density estimated through the solar radiation; T_{opt}, T_{min}, and T_{max}, represent the optimum, minimum, and maximum temperature for stomatal conductance, respectively; $VPD_{\min}$ and $VPD_{\max}$ are minimum and maximum vapor pressure deficit for stomatal conductance, respectively; WP is SWC at wilting point and FC is SWC at field capacity. These two parameters are constant and depend on the soil type obtained from a module included into the WRF-CHIMERE model.

The dominant vegetation data, required to estimate g_{sto}, were retrieved from the spatial tree distribution, based on the European Forest Institute database (Brus et al. 2011). Species-specific values of DO3SE parameters were derived from UNECE (2017) for each dominant plant species.

Once the stomatal conductance was computed, the stomatal O_3 flux was calculated over the growing season and expressed as PODY (nmol O_3 m^{-2} s^{-1} per leaf area). PODY (mmol m^{-2}) was calculated from hourly data as:

$$PODY = \int_{i=1}^{n} \left[\left(g_{sto} \times [O_3] - Y\right), 0\right].dt \tag{8}$$

where PODY is the accumulated stomatal O_3 flux above a threshold Y over the growing, g_{sto} represents the hourly stomatal conductance values, $[O_3]$ is the hourly O_3 concentrations (ppb) and *dt* is the time step (1-h). PODY was calculated with Y = 0 nmol O_3 m^{-2} s^{-1} per leaf area, by considering that any O_3 molecule is harmful for plants (Musselman et al. 2006), and Y = 1 nmol O_3 m^{-2} s^{-1} per leaf area, as recommended by CLRTAP (2017).

Estimation of annual trends

A 10-year time-series of O_3 data is considered long enough to assess short-term changes as reported in Sicard et al. (2016a). The non-parametric tests are robust and suitable for non-normally distributed data with missing and extreme values (Sicard et al. 2009). The non-parametric Mann–Kendall test was used to assess whether there is a monotonic upward or downward trend of O_3 over time (Sicard et al. 2013; Guerreiro et al. 2014). To quantify linear trends, the non-parametric Sen's slope estimator was used (Sicard et al. 2013, 2016a; Araminienė et al. 2019). Annual trends were calculated for O_3 metrics over the time period 2000–2014 for each European country as well as for four European regions as classified in UNECE (2010). Table 1 shows the regional classification of countries. The results are considered statistically significant at $p < 0.05$.

Results

European distribution of air temperature and ozone metrics

Over the time period 2000–2014, the minimum annual air temperatures (7.0 ± 0.3 °C) occurred in Northern Europe while the maximum values (12.7 ± 0.5 °C) were found in Mediterranean Europe (Fig. 1). Atlantic and Continental central Europe showed mean annual temperature ranging from 8.2 to 10.2 °C (Fig. 1). Taking into account mean O_3 concentrations, Mediterranean Europe showed values

Table 1 Regional classification of countries adopted in this study based on Manual on methodologies and criteria for modelling and mapping critical loads and levels of air pollution effects, risks and trends (UNECE 2017)

Region	Countries
Atlantic central Europe	Belgium, Ireland, Luxembourg, Netherlands, Unite Kingdom
Continental central Europe	Austria, Belarus, Czech Republic, Finland, Faroe Is., France, Germany, Hungary, Slovakia, Liechtenstein, Moldova, Poland, Romania, Russia, Switzerland, Ukraine
Mediterranean Europe	Albania, Bosnia & Herzegovina, Bulgaria, Greece, Croatia, Italy, Macedonia, Malta, Montenegro, Portugal, Slovenia, Spain, Serbia
Northern Europe	Denmark, Estonia, Finland, Iceland, Latvia, Lithuania, Norway, Sweden

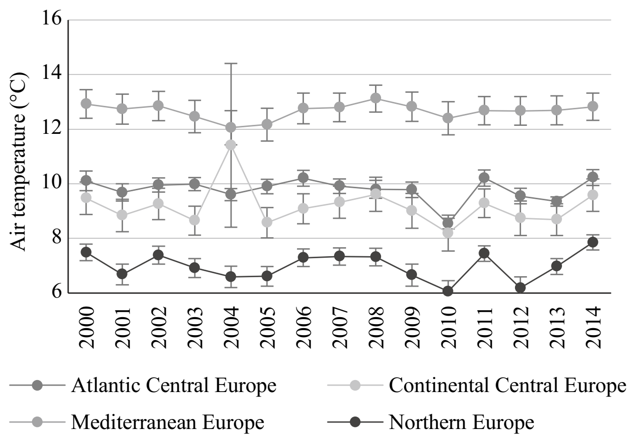

Fig. 1 Mean annual air temperature (in °C ± Standard Error) in Europe over the time period 2000–2014

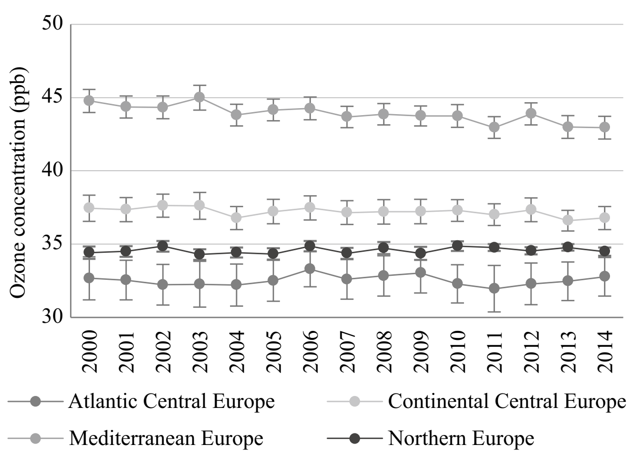

Fig. 2 Mean ozone concentration (in ppb ± Standard Error) in Europe over the time period 2000–2014

ranging from 42.9 ppb in 2011 to 45.0 ppb in 2003 whilst the other parts of Europe showed annual average values well below 40 ppb for all considered years (Fig. 2). Similarly, the highest AOT40 values (38,359 ppb h) were observed in Mediterranean Europe whilst the lowest AOT40 values (5094 ppb h) were found in Northern Europe (Fig. 3). In Continental central Europe, AOT40 ranged from 13,636 to

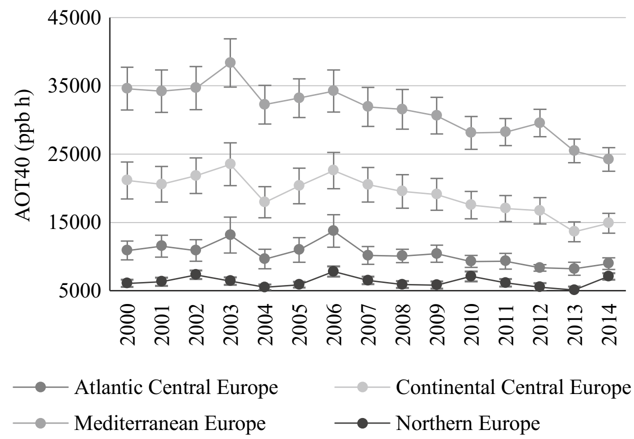

Fig. 3 Mean AOT40, i.e. accumulated exposure over a threshold of 40 ppb (in ppb h ± Standard Error) in Europe over the time period 2000–2014

23,515 ppb h while in Atlantic central Europe AOT40 varied from 8207 to 13,751 ppb h (Fig. 3). Taking into account POD0 values, the minimum value (14.0 mmol O_3 m^{-2}) was found in 2006 in Northern Europe while the maximum values, 29.7 and 32.1 mmol O_3 m^{-2} were observed in Atlantic and Mediterranean Europe, respectively (Fig. 4). Similar results were found for POD1 (Fig. 5).

Trends in ground-level ozone metrics at European level

The annual trend magnitudes for O_3 concentrations, AOT40, POD0 and POD1 over the time period 2000–2014 are shown in Tables 2, 3, 4 and 5. The O_3 mean concentrations decreased significantly ($p < 0.05$) by 0.4 ppb per decade in Continental Central Europe and by 1.1 ppb per decade in Mediterranean Europe (Table 2). In Atlantic Central Europe and Northern Europe, the trends for O_3 mean concentration were not statistically significant (Table 2). The exposure index AOT40 significantly declined in Atlantic, Continental and Mediterranean Europe with a magnitude of 2124, 5532 and 7161 ppb h per decade, respectively (Table 3). POD0 and POD1 increased significantly over the time period 2000–2014 all over Europe, except in Northern Europe showing a positive but not significant ($p > 0.5$)

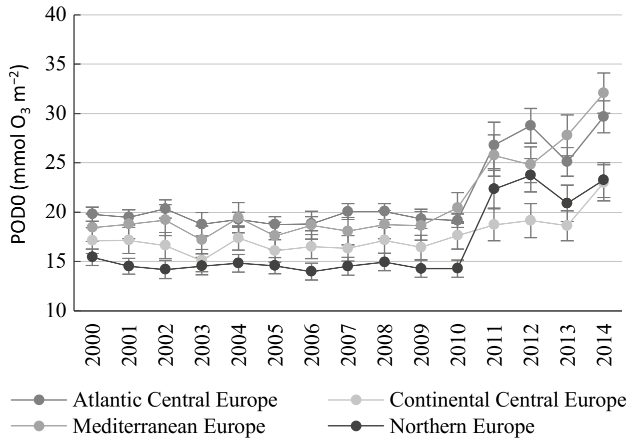

Fig. 4 Mean Phytotoxic Ozone Dose above a threshold Y of 0 nmol m^{-2} s^{-1} (POD0 in mmol O_3 m^{-2} ± Standard Error) in Europe over the time period 2000–2014

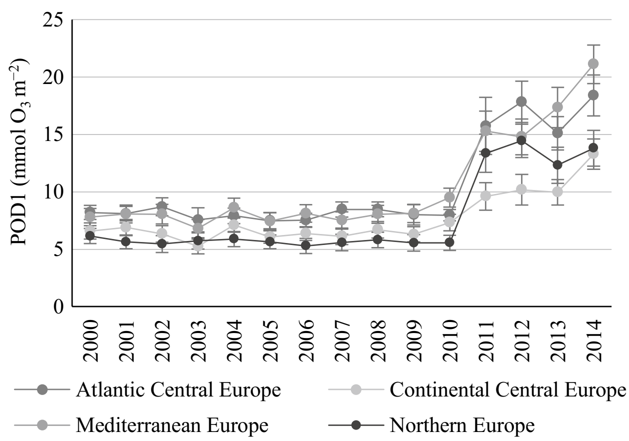

Fig. 5 Mean Phytotoxic Ozone Dose above a threshold Y of 1 nmol m^{-2} s^{-1} (POD1 in mmol. O_3 m^{-2} ± Standard Error) in Europe over the time period 2000–2014

Table 2 Regional average ± standard deviation (SD) and annual trend, obtained by the Mann Kendall test for ozone concentrations over the time period 2000–2014 (not significant, ns)

Region	Ozone concentrations (2000–2014)			
	Mean	SD	Trend (ppb/year)	*p* value
Atlantic central Europe	32.53	0.35	0.00	ns
Continental central Europe	37.21	0.30	−0.04	< 0.05
Mediterranean Europe	43.90	0.61	−0.11	< 0.05
Northern Europe	34.58	0.21	0.01	Ns

trend (Tables 4, 5). POD0 and POD1 increased by 4.3 and 3.3 mmol O_3 m^{-2} per decade in Atlantic Central Europe respectively, and by 2.4 and 3.0 mmol O_3 m^{-2} per decade in Continental Central Europe while higher trend magnitudes were found in Mediterranean Europe: POD0 and POD1 significantly increased by 5.7 and 6.1 mmol O_3 m^{-2} per decade, respectively. It is important to underline that POD0 and POD1 values are increasing since 2010.

Table 3 Regional average ± standard deviation (SD) and annual trend, obtained by the Mann Kendall test for AOT40 over the time period 2000–2014 (not significant, ns)

Region	AOT40 (2000–2014)			
	Mean	SD	Trend (ppb h/year)	*p* value
Atlantic central Europe	10,371.4	1581.5	−212.5	< 0.05
Continental central Europe	19,134.3	2812.6	−553.2	< 0.05
Mediterranean Europe	31,415.7	3790.2	−716.2	< 0.05
Northern Europe	6302.3	755.2	−34.7	Ns

Table 4 Regional average ± standard deviation (SD) and annual trend, obtained by the Mann Kendall test for POD0 over the time period 2000–2014 (not significant, ns)

Region	POD0 (2000–2014)			
	Mean	SD	Trend (mmol m^{-2}/year)	*p* value
Atlantic central Europe	21.6	3.9	0.43	< 0.05
Continental central Europe	17.5	1.8	0.24	< 0.05
Mediterranean Europe	21.0	4.4	0.57	< 0.05
Northern Europe	16.7	3.7	0.29	Ns

Table 5 Regional average ± standard deviation (SD) and annual trend, obtained by the Mann Kendall test for POD1 over the time period 2000–2014 (not significant, ns)

Region	POD1 (2000–2014)			
	Mean	SD	Trend (mmol m^{-2}/year)	*p* value
Atlantic central Europe	10.4	4.1	0.33	< 0.05
Continental central Europe	7.6	2.2	0.30	< 0.05
Mediterranean Europe	10.4	4.4	0.61	< 0.05
Northern Europe	7.7	3.6	0.15	Ns

Trends in ground-level ozone metrics at country-level

Over the time period 2000–2014, the exposure AOT40 index significantly declined in most of European countries (Fig. 6a). The lowest value was found in Switzerland (− 1376 ppb h $year^{-1}$) and the highest values were observed in the Isle of Man (self-governing British Island) and Malta

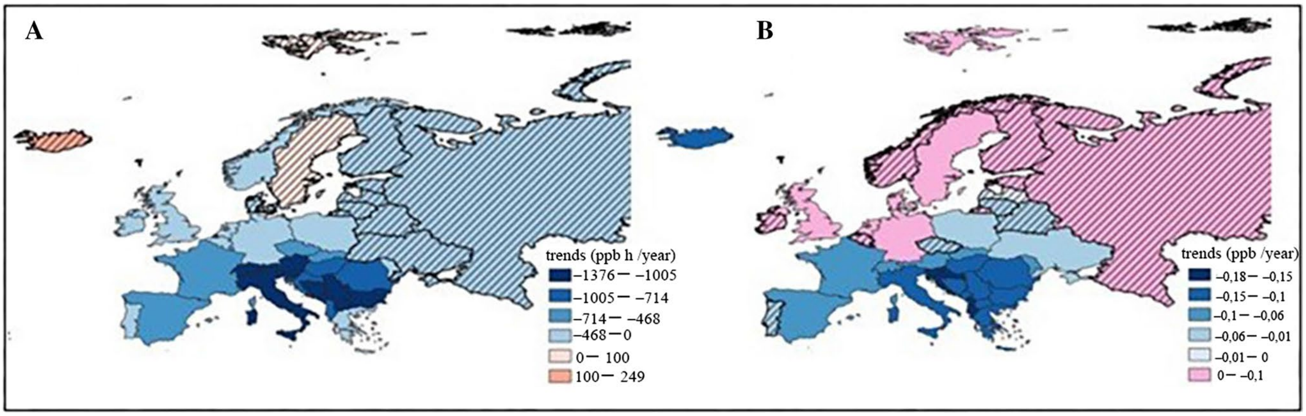

Fig. 6 Annual trends for AOT40 (in ppb h per year) **a** and ozone concentrations (in ppb) **b** in European countries. Blue color is for negative trends, pink for positive ones. Countries with not significant trends ($p > 0.05$) are striped

(+ 200 ppb h year^{-1} and + 249 ppb h year^{-1} respectively). Similar results were found for trends in O_3 concentrations (Fig. 6b). In particular, only six countries showed positive trends (Denmark, United Kingdom, the Netherlands, Germany, Sweden and Svalbard). On the contrary, trends of POD0 and POD1 were positive in all countries (Fig. 7a and b) with minimum values of + 0.03 and + 0.04 mmol m^{-2} per year and maximum values of + 1.06 and + 0.93 mmol m^{-2} per year for POD0 and POD1, respectively.

Discussion and conclusions

Our results showed a general decrease of O_3 concentrations and exposure-based index, namely AOT40, all over Europe and a general increase of flux-based metrics (POD0 and POD1) over the time-period 2000–2014. These results are in line with other studies conducted in different European countries. Anav et al. (2019) found that O_3 concentrations and AOT40 declined all over Europe from 2000 to 2014, while PODY did not. Karlsson et al. (2017) found a significantly decreasing trends of AOT40 in Fennoscandia and United Kingdom over the time period 1990–2015 highlighting that AOT40 is projected to no longer exceed the critical level established for forest protection for most of Northern Europe by 2050, while POD1 does not fall below its critical level. Similarly, the analysis conducted by Mills et al. (2018) at global level by using the international Tropospheric Ozone Assessment Report (TOAR) database showed a statistically significant decreasing trends of 45% of AOT40 (for perennial vegetation) and highlighted the reduction of AOT40 at many European sites. At country level, Araminienė et al. (2019) found negative trends of O_3 concentrations (− 0.28 ppb decade^{-1}) and AOT40 (− 2,540 ppb h decade^{-1}) but a positive trend for POD0 (+ 0.39 mmol O_3 m^{-2} decade^{-1}) in Lithuania. Our results demonstrate the large-scale success of European control strategies, such as the Air Quality Framework

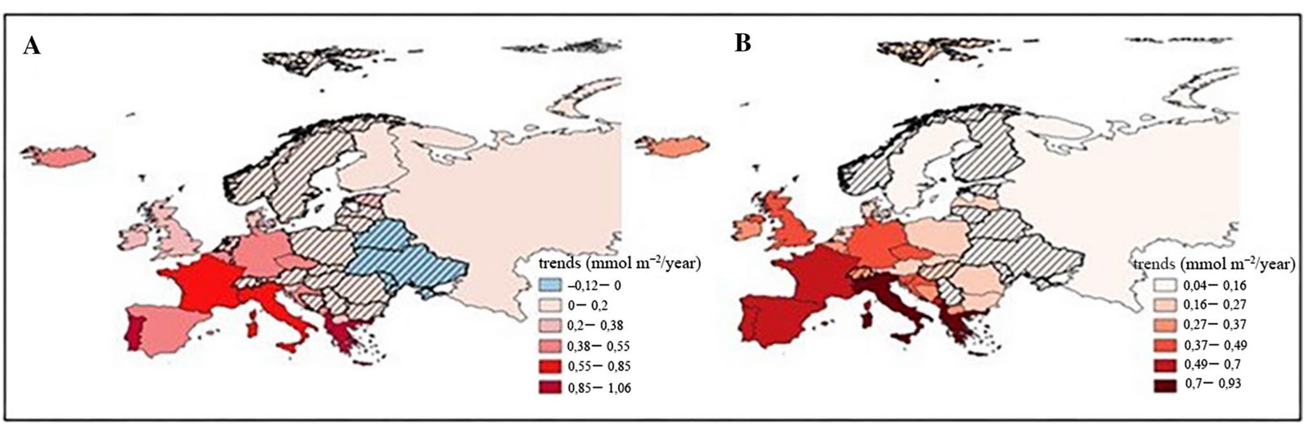

Fig. 7 Annual trends for POD0 **a** and POD1 **b** (in mmol O_3 m^{-2} per year) in European countries. Blue color is for negative trends, red for positive ones. Countries with not significant trends ($p > 0.05$) are striped

Directive (96/62/EC), Large Combustion Plant Directive (2001/80/EC), National Emission Ceilings Directives (2001/81/EC) and the Gothenburg Protocol (1999) under the United Nations Convention on Long-Range Transboundary Air Pollution (LRTAP), targeted at decreasing peak O_3 levels and reducing the risk of O_3 impacts on vegetation and human health. The emission reductions have primarily been achieved as a result of the progress in vehicle technologies, the stringent inspection systems legislation related to the "Euro" standards, the use of flue gas abatement techniques, the progress in the storage and distribution of solvents (Vestreng et al. 2008; EEA 2018; Sicard et al. 2020b). However, our results showed a general increase of PODY all over Europe between 2000 and 2014 highlighting the relative insensitivity of PODY to O_3 precursors control strategies. Climate change is identified as the responsible of this insensitivity as postulated in previous studies (Liu et al. 2016a, b; Fu et al. 2017; Anav et al. 2019). They reported earlier green-up dates and delayed dormancy dates then a longer growing season due to changing climate. Moreover, climate change increases the stomatal conductance thanks to the positive effects of higher air temperature and solar radiation on stomata opening (Hoshika et al. 2015). Even if the O_3 mean concentrations decreased, higher PODY levels were observed over time leading to higher O_3 risk to European forests (Proietti et al. 2016; Anav et al. 2019). Anav et al. (2019) hypothesized that the positive feedback between climate change and PODY will increase in the near future and the efforts in controlling emissions of O_3 precursors could be significantly offset by climate change, thus increasing the O_3 risk for forests. A primary goal is to define a metric for O_3-risk assessment, which can identify ecosystems at O_3 risk to protect them using standards and policies. In Europe, AOT40 has been widely used, under the assumption that plant injury and exposure to O_3 concentrations are correlated (EPA 2007; UNECE 2011; Fares et al. 2013). To date, several studies report a general growing consensus for moving toward a biologically-sound stomatal flux-based standard (PODY) as new European legislative standard (Mills et al. 2011; Fares et al. 2013; Sicard et al. 2016b,c; Anav et al. 2016, 2019) although critical levels for vegetation protection still need to be validated (Sicard et al. 2016c). Epidemiological observation of O_3-induced injury and environmental variables including O_3 can be used to derive consistent stomatal flux-based critical levels for different type of vegetation under natural field conditions (De Marco and Sicard 2019; Paoletti et al. 2019). The question about deriving new critical levels is still a challenge for the scientific community (De Marco and Sicard 2019), even because some advantage of AOT40 are still present. Indeed the simplicity and fast applicability of AOT40 could be an advantage for the use of the concentration-based metric (Anav et al. 2016). But on the other side AOT40 is not taking into consideration more biological processes linked to the stomatal aperture and can have spatially and temporally different patterns (De Marco et al. 2015). These consideration highlights the role of climate into determination of the impacts of ozone on forests. Consequently, strategies integrating both climate and air quality policies are urgently needed for forest protection against the negative impacts of O_3 (Ainsworth et al. 2012).

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