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Analysis of European ozone trends in the period 1995–2014

Yingying Yan[1, 2], Andrea Pozzer[1], Narendra Ojha[1], Jintai Lin[3], Jos Lelieveld[1]

[1] Atmospheric Chemistry Department, Max Planck Institute for Chemistry, Mainz, Germany

[2] Department of Atmospheric Science, China University of Geosciences, Wuhan, China

[3]Laboratory for Climate and Ocean-Atmosphere Studies, Department of Atmospheric and Oceanic Sciences, School of Physics, Peking University, Beijing 100871, China

Email: andrea.pozzer@mpic.de

Abstract

Surface-based measurements from the EMEP network are used to estimate the changes in surface ozone levels during the 1995-2014 period over Europe. It is shown that a significantly decreasing trend in the 95th percentile ozone concentrations has occurred, especially during noontime (0.9 μg/m^3/y), while the 5th percentile ozone concentrations continued to increase with a trend of 0.3 μg/m^3/y during the study period. With the help of numerical simulations performed with the global chemistry-climate model EMAC, the importance of anthropogenic emissions changes in determining these changes are investigated. The EMAC model is found to successfully capture the observed temporal variability in mean ozone concentrations, as well as the contrast in the trends of 95th and 5th percentile ozone over Europe. Sensitivity simulations and statistical analysis show that a decrease in European anthropogenic emissions had contrasting effects on surface ozone trends between the 95th and 5th percentile levels, and that background ozone levels have been influenced by hemispheric transport, while climate variability generally regulated the inter-annual variations of surface ozone in Europe.

1. Introduction

Tropospheric ozone has detrimental effects on human health, and elevated concentrations at the surface are of concern over most of the European region (Hjellbrekke and Solberg, 2002; WHO, 2013; EEA, 2013; Lelieveld et al., 2015). The European Union (EU) Air Quality Directive sets four standards for surface ozone to reduce its impacts on human health and crop yields (http://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri=CELEX:32008L0050&from=EN). The standards are: information threshold, (1-hour average: 180 μg/m^3), alert threshold (1-hour average: 240 μg/m^3), long-term objective (maximum diurnal 8-hour mean: 120 μg/m^3), and the target value (long-term objective should not be exceeded more than 25 days per year, averaged over 3 years). Exceedances are particularly frequent in regions close to high ozone precursor emissions during summer with stagnant meteorological conditions, associated with persistent high temperatures. Since a substantial decrease in precursor concentrations has been achieved in Europe in recent decades, the number of exceedances has declined (Guerreiro et al., 2014), in line with a long-term downward trend of pollution emissions (Colette et al., 2011; Wilson et al., 2012). Further, a number of studies have shown that European ozone levels are on average decreasing in the last 20 years (as example, Jonson et al, 2010). Nevertheless, background ozone changes over Europe are not so clear (Wilson et al., 2012), being sensitive to climate conditions and intercontinental transport of O_3 and its precursors, and are significant in view of tropospheric chemistry (Lelieveld and Dentener, 2000).

The response of surface ozone to a changing climate, with potentially more frequent heat extremes (Bloomer et al., 2009; Jacob and Winner, 2009; Cooper et al., 2012; Fu et al., 2015; Lin et al., 2015; Simon et al., 2015), and concurrent changes in anthropogenic emissions of precursor gases (Bloomer et al., 2009; Fu et al., 2015; Strode et al., 2015; Yan et al., 2017) may pose a challenge for air quality management. Observation and model-based analyses of ozone trends in responses to climate change (Bloomer et al., 2009), precursor emissions (Bloomer et al., 2009; Lefohn et al., 2010), and long-range transport (Lin et al., 2015) have been conducted for North America (Strode et al., 2015; Lin et al., 2017; Yan et al., 2017), several Asian regions (Brown-Steiner et al., 2015; Lin et al., 2017) and also for Europe (Meleux et al., 2007, Wilson et al., 2012, Jonson et al., 2006). For Europe, the connection between climate and ozone levels has been subject of large number of studies, notably to investigate the effects of climate change on surface ozone levels (Langner et al., 2005; Meleux et al., 2007; Colette et al., 2011; Langner et al., 2012.)

Tropospheric ozone is produced photochemically during daytime, mainly from the photolysis of nitrogen dioxide (NO_2), while NO_2 levels are strongly influenced by radicals and their precursors, including organic compounds. Due to the complex photo-chemistry involved, the amount of ozone formed responds nonlinearly to changes in precursor emissions and is sensitive to variations in air temperature, radiation and other climatic factors (Fu et al., 2015; Monks et al., 2015; Coates et al., 2016). Ozone can be destroyed via reactions with NO_x (i.e., ozone titration) especially during nighttime, and thus a reduction in NO_x emissions could result in more ozone (Jhun et al., 2014; Yan et al., 2017). Previous studies of European ozone have focused on daytime or diurnal mean ozone with little attention paid to the daytime-nighttime contrast in ozone changes (Colette et al., 2011; Wilson et al., 2012; Guerreiro et al., 2014).

Our work contrasts the trends of the monthly 5th and 95th percentile European ozone levels at hourly levels over the period 1995–2014, based on the hourly ozone measurements from the EMEP network. Additionally, numerical simulations from the global chemistry-climate model ECHAM5/MESSy (EMAC) are conducted to evaluate the model's ability in capturing ozone trends over Europe and to investigate the underlying importance of the meteorology and emission changes for the observed ozone trends.

The manuscript is organized as follows: the observational dataset, model simulations and analysis methods are described in Section 2. In Section 3, the average linear trends for the European domain are estimated and analyzed separately for the monthly, seasonal and annual surface 5th, 50th, and 95th percentiles of the observed ozone concentrations. We then compare the observed ozone trends and variability to results of the atmospheric chemistry – general circulation model EMAC. To investigate the effects of anthropogenic emissions and climate variability on observed European ozone changes, we conduct a sensitivity simulation with constant emissions and statistical analysis with the ERA-Interim 2-meter temperature data in Section 4. Followed by the conclusions in Section 5.

2. Methods and Data

2.1 Ozone measurements

The hourly ground-level ozone measurements over 1995–2014 have been obtained from the Chemical Coordination Centre of *European Monitoring and Evaluation Programme (*EMEP) network (http://www.nilu.no/projects/ccc/emepdata.html). Table 1 shows the number of measurement sites (varies from 113 to 137) and the percentage of missing hourly data in each year. Fig. 1 further shows the site distribution. Since many of the stations are not operating continuously during the study period (Fig. 1), we have included only those sites in the analysis which fulfill the criteria defined by Cooper et al. (2012). Such data selection criteria are further applied for the US ozone trends analysis with the EPA-AQS measurements by Yan et al. (2017). First, we discard the observational days with the valid hourly data less than 66.7% in any daytime or nighttime. Then, we discard the particular season with less than 60 days containing valid data in any season. Finally, for any season, we keep the data with valid seasonal mean ozone more than 15 years during 1995–2014; otherwise we discard the data in all years for the particular season. Fig. 1 shows the final selected 93 sites satisfying above criteria for the analysis.

We calculated the linear trends for the European surface ozone at individual hours, and mean values for daytime (local time: 07:00–19:00), nighttime (local time: 19:00–07:00) and full days (24 h). For each daytime or nighttime period, the missing data varies between 6.8 and 34.6% (Table 1). The monthly 5th, 50th and 95th percentile ozone concentrations for each period (per hour, daytime, nighttime and diurnal) are derived from the lowest, middle and highest 5th percentile hourly ozone mixing ratios of the corresponding period in each month.

To calculate the ozone trends per hour, during daytime, nighttime and per day, we then use the following statistical trend model (Weatherhead et al., 1998; Yoon and Pozzer, 2014):

$$Y_t = \mu + S_t + \omega X_t + N_t$$

Where Y_t denotes the monthly time series of ozone, μ is a constant term representing the offset, $X_t = t/12$ (with t as month) the number of years in the timeseries, and ω is the magnitude of the trend per year. S_t is a seasonal component in the trend estimates. N_t is the residual term of the interpolation. As the seasonal component does not have much impact on the statistical properties of the estimates of the other terms in the model, we use the deseasonalized monthly data to perform the trend analysis with a model of the form:

$$Y_t = \mu + \omega X_t + N_t$$

Using this formulation the linear trends are also analyzed separately for the observed monthly, seasonal and annual surface ozone concentration. The 5th, 50th and 95th percentile ozone concentrations are derived from the lowest, middle and highest 5th percentile of the full set of measurements (i.e., hourly ozone mixing ratios) of the corresponding period.

The standard deviation of ozone trends over the European stations is calculated with:

$$\sigma = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(\omega_i - \alpha)^2}$$

where N is the total number of sites, ω_i is ozone trend at individual sites and α represents the average ozone trend.

2.2 ERA-Interim 2-meter temperature data

The 2-meter temperature data is from the reanalysis product ERA-Interim, provided by the European Centre for Medium Range Weather Forecast (ECMWF) Public Datasets web interface (http://apps.ecmwf.int/datasets/), covering the data-rich period from 1979 and continuing in real time (Dee et al., 2011). Compared to the ERA-40, the ERA-Interim has an improved representation of the hydrological cycle, and stratospheric circulation (Dee and Uppala, 2009; Dee et al., 2011). The ERA-Interim atmospheric model and reanalysis system uses cycle 31r2 of ECMWF's Integrated Forecast System (IFS), configured for 60 vertical levels up to 0.1 hPa. The horizontal-spatial resolution is either in a full T255 spectral resolution or in the corresponding N128 reduced Gaussian grid (Dee et al., 2011). ERA-Interim assimilates four analyses per day, at 00, 06, 12 and 18 UTC. ECMWF public website provides a large variety of data in uniform lat/long grids varying from 0.125° to 3°. Out of those, here, we analyze the monthly mean 2-meter temperature data which are archived on the 0.75° latitude by 0.75° longitude grid. Additional information (e.g. on current data availability) is available on the ECMWF website at http://www.ecmwf.int/research/era.

2.3 Atmospheric chemistry modeling

The ECHAM5/MESSy Atmospheric Chemistry (EMAC) model has been used to simulate surface ozone for the 1995–2014 periods. The EMAC model applies the second version of the Modular Earth Submodel System (MESSy2) to link multi-institutional computer codes (Jockel et al., 2016). The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5) (Roeckner et al., 2006). EMAC simulated gas-phase tracers as well as aerosols have been extensively evaluated in previous studies (e. g. Pozzer et al., 2007; Pozzer et al., 2012).

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In this work, we use the archived RC1SD-base-10a simulation results from the EMAC model conducted by the ESCiMo project (Jockel et al., 2016). The model results were simulated with version 5.3.02 for ECHAM5 and version 2.51 for MESSy. The archived data were obtained with a T42L90MA spatial resolution, i.e., with a T42 spherical representation which is corresponding to a quadratic Gaussian grid with approximately 2.8 latitude by 2.8 longitude, and 90 levels in the vertical, with the top level up to 0.01 hPa. To reproduce the observed meteorology, the method of Newtonian relaxation towards ERA-Interim reanalysis data (Dee et al., 2011) is applied to weakly nudge the dynamics of the general circulation model. Differently from the work of Jöckel et al. (2016), the model was re-run to cover the full period of measurements and also with a 1-hourly temporal resolution for ozone, in order to compare model results with hourly observational data. We also conducted a sensitivity simulation in which the anthropogenic emissions were kept constant throughout the years to investigate the effects of emissions on ozone trends.

Anthropogenic emissions in the model are incorporated as prescribed sources following the Chemistry-Climate Model Initiative (CCMI) recommendations (Eyring et al., 2013), using the MACCity (Monitoring Atmospheric Composition & Climate/City Zero Energy) emission inventory, which includes a seasonal cycle (monthly resolved). Additionally, the emissions are vertically distributed as described by Pozzer et al. (2009). Since the total NMVOCs (non-methane volatile organic compounds) values for anthropogenic sectors are not provided by the MACCity raw dataset, they are recalculated from the corresponding species (Jockel et al., 2016).

Emissions from natural sources have been prescribed as well, either as monthly resolved or annually constant climatology. The spatial and temporal distributions of biogenic NMHCs are based on Global Emissions InitiAtive (GEIA). Further details of the model setup on the emissions, physical and chemical processes as well as the model evaluation with observations can be found in Jöckel et al. (2016).

3. Results

3.1 Ozone trends in EMEP measurements

Fig. 2 shows the trends in ozone concentrations (monthly mean, 5th, 50th and 95th percentile) over Europe during the 1995-2014 period, for each hour of the day. While the average ozone concentrations (and 50th percentiles) do not show significant trends, the 5th and 95th percentile ozone show significant trends with a clear diel cycle. The 95th percentile ozone shows a decreasing trend over Europe during the 1995-2014 period with the trend being most pronounced (-0.9 ± 0.5 μg/m^3/y) during midday (1100-1500 h). 95th percentile ozone concentrations also show a decreasing trend during the night, however the trends are observed to be smaller (-0.5 ± 0.35 μg/m^3/y). Here the standard deviation depicts the variability of the trends among the stations, and therefore reflects the almost homogeneous decrease over entire Europe. Interestingly, in contrast with the 95th percentile, the 5th percentile ozone over Europe shows an increasing trend especially during midday (0.3 ± 0.16 μg/m^3/y). Further, the temporal evolutions of ozone anomalies during the 1995–2014 period are shown for 5th and 95th percentile in Fig. S1. The 95th percentile ozone trend indicates a general decline in the photochemical buildup of ozone during noon hours, with the exception of strongly enhanced ozone during 2003. The inter-annual variability is observed to be very large with ozone anomalies in excess of 35 μg/m^3 in 2003 relative to 2014. For 95th percentile ozone, the sharp increase by up to 20 μg/m^3 in the year 2003 occurred during a strong European heat wave (Section 4.2). The analysis of individual year observations here shows that the increasing trend in the 5th percentile ozone is a robust feature with most of the recent years showing stronger noontime build up in ozone as compared to the 1990s. During the study period the variability in noontime ozone anomalies is however lower (~10 μg/m^3) in the 5th percentile ozone compared to the 95th percentile ozone.

Consistently with the results obtained for hourly ozone, when the observational data is reduced to diurnal values, a growth rate of 0.22 ± 0.15 μg/m^3/y is calculated for the 5th percentile ozone, while a stronger decline rate of -0.57 ± 0.34 μg/m^3/y is estimated for the 95th percentile ozone (see Table 2). Hereafter we will mainly focus on trends in the daytime mean, nighttime mean, 5th percentile and 95th percentile ozone concentrations.

The observed long-term reduction in 95th percentile ozone concentrations over Europe concurs with the reduction in anthropogenic emissions of ozone precursors. Anthropogenic emissions of NO_x and CO over Europe declined by 35% and 58%, respectively, as calculated from the MACCity inventory. Slower rates of ozone reduction during nighttime are suggested to be combined effects of reduced titration due to lower NO_x emissions, and an increase in the global background ozone concentrations during this period, probably due to growing precursor emissions in China since 1995, which has been predicted by Lelieveld and Dentener (2000) based on atmospheric chemistry – transport modeling, and corroborated by satellite observations (Richter et al., 2005; Krotov et al., 2016). The effect of anthropogenic emissions is discussed in more detail in the Section 4.1.

Fig. 3 further shows ozone trends for each month of the year. The slight growth rates in the 5th percentile ozone are approximately equally distributed at the level of 0.1 ± 0.12 μg/m^3/y. Conversely, the monthly trends for the 95th percentile ozone are negative with a most rapid decrease rate of -1.67 ± 0.4 μg/m^3/y in August. For the 50th percentiles (mean) the seasonal cycle of ozone trends decline unevenly from January to August, then pick up in the following months. It leads to the fastest ozone growth in December when the ozone production is minor due to the relatively lowest solar UV fluxes and temperatures, and the maximum ozone decline in August, which is the photochemically most active month in Europe. In December, the 50th (mean) percentile ozone increases at a rate of 0.41 ± 0.21 μg/m^3/y (0.32 ± 0.09 μg/m^3/y), while a decline rate of -0.40 ± 0.24 μg/m^3/y (-0.51 ± 0.13 μg/m^3/y) is calculated in August.

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Table 3 shows the trends in seasonal ozone concentrations over Europe analyzed separately for day- and nighttime. The ozone concentrations show pronounced differences in trends over the different seasons. The mean surface ozone in summer, averaged over the selected 93 sites, declines at rates of -0.32 ± 0.24 μg/m^3/y and -0.20 ± 0.27 μg/m^3/y during day- and nighttime, respectively. It is mainly related to the rapid decline in the highest levels (95th percentile) of ozone with rates of -1.10 ± 0.61 μg/m^3/y (daytime) and -0.71 ± 0.52 μg/m^3/y (nighttime). Although the 95th percentile ozone in spring declines almost as fast as during summer, the decrease in spring for the 95th percentile ozone is compensated by the growth in 5th percentile ozone, leading to much lower decrease rates in spring compared to summer for the mean ozone concentrations. Finally, in winter ozone grows at a rate of ~0.10 μg/m^3/y. This increase occurs mostly in the lower level (5th percentile) ozone concentrations, with growth rates of 0.25 ± 0.15 μg/m^3/y (daytime) and 0.14 ± 0.22 μg/m^3/y (nighttime).

For the trends in annual mean ozone mixing ratios, a decline in the 95th percentile ozone (daytime: -0.81 ± 0.46 μg/m^3/y; nighttime: -0.57 ± 0.36 μg/m^3/y) is observed while an increase in the 5th percentile ozone (0.22 ± 0.17 and 0.16 ± 0.17 μg/m^3/y for day- and nighttime, respectively, is calculated, resulting in statistically not-significant decreasing trends (daytime: -0.09 ± 0.24; nighttime: -0.05 ± 0.23 μg/m^3/y) (Table 3).

Fig. 4 further shows the ozone trends distribution site-by-site over the 93 selected stations for daytime mean, 5th and 95th percentile ozone during the four seasons. The 95th percentile ozone trend shows a decline at most of the selected sites, although ozone increases are also visible at several sites, especially in fall-to-winter. The annual ozone trend averaged over all sites during daytime (-0.61 μg/m^3/y) is nearly twice that during nighttime (-0.34 μg/m^3/y, Fig. S2). For the 5th percentile ozone, the annual means have grown over the western and central European sites, in contrast with declines in ozone at other locations over the northern and southern Europe. Averaged across all sites, the 5th percentile ozone has slightly grown during day- as well as nighttime. The regional trend contrast is most significant in summertime with an average decrease rate of -0.03 μg/m^3/y. The ozone trends spatial distribution in the daytime (Fig. 4) much resembles that of the ozone trends in nighttime (Fig. S2) for the mean, 5th percentile as well as 95th percentile ozone.

3.2 Ozone exceedance trends

Based on the European directive for ozone concentrations limits, we calculate the number of exceedances for the information threshold and long-term objective (Fig. 5). Averaged over the selected 93 sites, the exceedances of the information threshold as well as the long-term objective have declined at rates of -3.2% and -2.5% per year relative to 1995. The decrease accelerated after the year 2003, during which a European heat wave raised summer temperatures by 20 to 30% (in degrees Celsius) compared to the seasonal average over a large part of the continent, extending from northern Spain to the Czech Republic and from Germany to Italy. The variations

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in the exceedances are inter-annually consistent with the changes in the annual 95th percentile ozone, with a significant correlation coefficient of 0.93 for information threshold exceedances and 0.90 for long-term objective exceedances.

3.3 Ozone trends from EMAC simulation

The same analysis performed on the observations has been carried out on the EMAC model results, i.e., for the same period covered by the observations. To ensure spatiotemporal consistency with the EMEP data, modeled ozone concentrations are sampled at the times and locations of the measurements. Fig. 6 compares the time series of modeled and observed monthly mean ozone over Europe. Although the model overestimates the measurements with a mean bias of 4.3 μg/m^3 over the 1995–2014 period, the simulation results are highly correlated with observed ozone, with a significant correlation coefficient of 0.91. The high bias may be explained by the coarse grid resolution of 2.8 degrees that was applied, leading to the artificial dispersion of localized NO_x emissions, which optimizes NO_x concentrations over Europe with respect to chemical O_3 formation, also noticed by Joeckel et al (2016). Such overestimation of the observed ozone due to coarse model horizontal resolution has been reported by Lin et al. (2008) and Yan et al. (2014, 2016). The overestimation after 2010 becomes more evident (mean bias 5.4 μg/m^3), mostly due to the emissions used in the model version used, being prescribed up to the year 2005 and predicted in the subsequent period. The modeled ozone biases are slightly higher (mean bias: 5.2 μg/m^3 and 6.7 μg/m^3 for 1995–2014 and 2010–2014, respectively) compared to the observed de-seasonalized time series. Nevertheless, EMAC model can reproduce the observed inter-annual and seasonal variabilities of ozone, with statistically significant correlation coefficients at most observation sites. For the diurnal, daytime as well as nighttime mean ozone averaged across the 93 sites, the model-observation correlation is 0.84–0.92 (0.62–0.70 for de-seasonalized time series).

The EMAC modeled ozone trends per hour are shown in Fig. 6. The agreement with the observationally estimated trends is good, although the model tends to overestimate the trends by 0.12 μg/m^3/y, 0.23 μg/m^3/y, 0.08 μg/m^3/y, and 0.36 μg/m^3/y for the mean, 5th, 50th and 95th percentile ozone, respectively. The measured diurnal cycle of the ozone trends (Fig. 2) is well captured by the EMAC model for the 5th and 95th percentile ozone concentrations. Consistently, the modeled temporal evolutions (Fig. S3) of annual European 5th percentile ozone anomalies are larger compared to the observations (~15 μg/m^3 versus ~10 μg/m^3 enhancements during photochemical buildup of ozone at midday hours during 1990–2014), while being smaller for the 95th percentile (~21 μg/m^3 versus ~30 μg/m^3). Further, the EMAC model reproduces the jump in high level ozone concentrations during the year 2003 that was affected by a major heat wave.

For the diurnal mean values, averaged over Europe, the model produces higher growth rates for the 5th percentile ozone and weaker decrease rates for the 95th percentile ozone compared to the observed trends (Table 2). For the 50th percentile and mean ozone trends averaged over Europe,

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the model shows statistically insignificant changes, similar to the observed trends (Table 2). Fig. S4 further shows the spatial distribution of the simulated diurnal ozone trends. It corroborates that central Europe experiences the highest growth rate for the averaged (also 50th percentile) and 5th percentile ozone concentrations, and the strongest reduction for the 95th percentile ozone during all seasons.

For the trends per month, the EMAC model reproduces the observed variability with statistically significant correlation coefficients of 0.88–0.90 for the mean, 50th and 95th percentile ozone trends (Fig. 3 and Fig. S5). Seasonally, for the 95th percentile ozone the modeled ozone trends are much weaker than from measurements in all seasons except the autumn (Table 3). The decreased higher level ozone is probably driven by the anthropogenic ozone precursor emission decline over these years, which has been studied in previous work of ozone change drivers and corroborated in Sect. 3.4 with a sensitivity simulation. For the 5th percentile ozone, especially for the daytime period, the increasing trends are enhanced in the model results during all seasons (Table 3).

4. Anthropogenic emissions and climate variability

4.1 Effects of anthropogenic emissions

A sensitivity simulation is conducted with constant global anthropogenic emissions to test the sensitivity of observed European ozone to inter-annual variability in climate, by removing the effects of anthropogenic emission changes. Consequently, the decline in European emissions (Fig. S6) is removed from the EMAC model. With constant emissions, the modeled ozone shows a slight increase at the midday hours for the 95th percentile and a slight decrease for the 5th percentile, in contrast to the trends calculated from the control simulation. In the sensitivity simulations no significant trend (less than 0.1 μg/m^3/y) for any hour of the day is found, and also no contrast in ozone trends between the 5th and 95th percentiles (Fig. 7), which was well reproduced by the control simulation. Therefore, it appears that both the decreases in 95th percentile ozone and the enhancements in 5th percentile ozone are associated with the rapid decline in the precursor gases anthropogenic emissions over Europe, notably of NO_X, prescribed by the MACCity inventory (Fig. S6). These results reflect the effectiveness in controlling high-level ozone, but being unsuccessful in controlling the lower level ozone. Evidently, the 35% reduction in NO_x emissions in Europe was not sufficient to achieve substantial reductions in ozone, especially of background levels, which are affected by growing emissions in Asia that are transported hemispherically (Lelieveld and Dentener, 2000).

Averaging over the selected 93 sites, we calculate the number of exceedances for the information threshold both in the control and the sensitivity simulation (Fig. 8). In the control simulation, the exceedances of the information threshold have declined at rates of -2.5% per year relative to 1995, slightly smaller than the observed decrease rate of -3.2%. The variations in exceedances are inter-annually consistent with the observations, with a significant correlation coefficient of 0.61. However, in the sensitivity simulation, the decline rate (-0.6%) in the exceedances is much smaller than the rates in the control simulation and in the observations.

By fixing the anthropogenic emissions, ozone trends in each month for the 95th percentile ozone show no obvious decline but rather a slight enhancement with growth rates of -0.23 – 0.50 μg/m^3/y. For the 5th percentile ozone and compared to the control simulation, there is no increase but a slight decrease at a rate of -0.51 – 0.15 μg/m^3/y in months of the year (Fig. S7). For the trends in annual mean ozone mixing ratios simulated in the sensitivity simulation, an enhancement in the 95th percentile ozone (daytime: 0.16 $\pm$0.18 μg/m^3/y; nighttime: 0.10 $\pm$0.15 μg/m^3/y) is calculated while a decline in the 5th percentile ozone (-0.11 $\pm$0.14 and -0.07 $\pm$0.12 μg/m^3/y for daytime and nighttime, respectively) is estimated, contrasting to but smaller in the absolute value than the trends in the control simulation. This contrast has been also shown in the trends for individual hour of the day between control and sensitivity simulations (Fig. 7). These results show that the effects of decline in anthropogenic emissions on European ozone change are somewhat offset by the impacts of climate variability. This compensation effect is not only for the high level ozone concentrations, which has been reported by previous studies (Lin et al., 2017), but also for the low level ozone concentrations.

4.2 Effects of heat waves

As discussed in number of studies (e.g., Filleul et al, 2006, Vautard et al, 2005, Garcia-Herrera et a 2010, Vieno et al 2010), the 2003 heat waves caused favorable meteorology for ozone buildup, leading to very high ozone concentrations during the summer period (from July to August). Fig. 9 shows the distribution of the difference in the exceedances between 2003 and averaged over 1995-2002 for the information threshold as well as the long-term objective over individual site. Except for some northern sites, the exceedances in 2003 are much more frequent than the average from 1995 to 2002 over most of the observational sites, especially over central Europe. This exceedance anomaly distribution in 2003 relative to the period of 1995-2002 coincides with the 2-meter temperature anomaly distribution, with a statistically significant correlation up to 0.64 (Fig. S8).

The exceedance anomaly of information threshold and long-term objective during the year 2003 with respect to the 1995-2002 period follows the anomaly in ozone concentrations, in turn consistent with the temperature anomaly. Fig. S9 shows the correlations between the monthly mean 2-meter temperature and the monthly mean, 5th and 95th percentile ozone for diurnal, daytime and nighttime concentrations. It corroborates the high correlations over central Europe with statistically significant values up to ~0.82. Ozone concentrations over the northern sites are negative and insignificantly correlated with temperature. This may be related to the influence of the Northern Atlantic Oscillation, which had an opposite impact on temperature over northern

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compared to central Europe (Fig. S10). These results underscore that the large-scale climate variability affects the inter-annual variability of European ozone.

In the simulation with constant emissions, however, the modeled ozone fluctuation of annual European ozone anomalies for individual hours is comparable in magnitude with the results in the control simulation (Fig. S7). In both simulations, the fluctuation dominates around midday for 5th (~15 μg/m^3 in the base simulation versus ~13 μg/m^3 in sensitivity simulation) and 95th (~21 μg/m^3 versus ~20 μg/m^3) percentile ozone (Fig. S7 and Fig. S3). In addition, the variations in the exceedances of the information threshold are inter-annually consistent with the observations and the control simulation, with significant correlation coefficients of 0.54 and 0.56, respectively, comparable to the correlations between observations and control simulation (Fig. 8). Further correlations between the European averaged monthly mean 2-meter temperature and the modeled monthly mean (50th), 5th and 95th percentile ozone in the sensitivity simulation are statistically significant with correlation coefficients of 0.69–0.78 for diurnal, day- and nighttime concentrations, consistent with the correlations (0.70–0.81) between 2-meter temperature and simulated European ozone in the control simulation. These results clearly show that the ozone variations are regulated by climate variations.

5. Conclusions and outlook

Based on EMEP observed ozone in the period 1995-2014, we analyzed the annual and seasonal trends of the mean, the 5th, 50th and 95th percentile of the ozone concentrations at different temporal distributions, i.e., hourly, diurnal, day- and nighttime. Results show that although reductions in anthropogenic emissions have lowered the peak ozone concentrations, especially during daytime in the period 1995-2014, the lower level ozone concentrations have increased continually since 1995 over the 93 sites. This leads to insignificant trends in the 50th percentile and mean ozone. Both the 5th and 95th percentile ozone trends follow a diel cycle with largest trends during periods of strong photochemical activity. These contrasting ozone trends per hour during the day and at different concentration levels are well reproduced by the EMAC chemistry-climate model, although the model slightly overestimates observed ozone at the surface. Furthermore, the numbers of exceedances of the information threshold and long-term objective have continuously declined during the 20-year period considered, and the decrease has accelerated since the year 2003.

Sensitivity simulations with constant emissions in the EMAC model, and correlation analysis between modeled ozone and the ERA-Interim 2-meter temperature help distinguish effects of climate and anthropogenic emissions on ozone variations and trends. Climate variability generally regulates the interannual variations of European surface ozone, while the changes in anthropogenic emissions predominantly contribute to ozone trends. However, it appears that the negative ozone trend due to European emission controls has been counteracted by a climate related tendency as well as hemispheric dispersion of pollutants from other regions, notably China..

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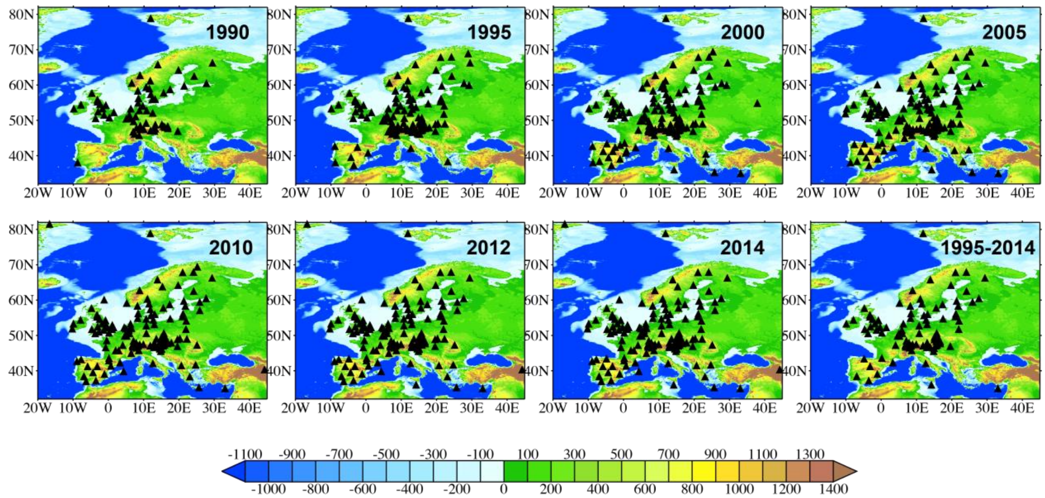

Fig. 1. Site distribution for the EMEP datasets (1990-2014) as well as the selected 93 sites (1995-2014). The overlaid map shows the surface elevation (m) from a 2 min Gridded Global Relief Data (ETOPO2v2) available at NGDC Marine Trackline Geophysical database (http://www.ngdc.noaa.gov/mgg/global/etopo2.html).

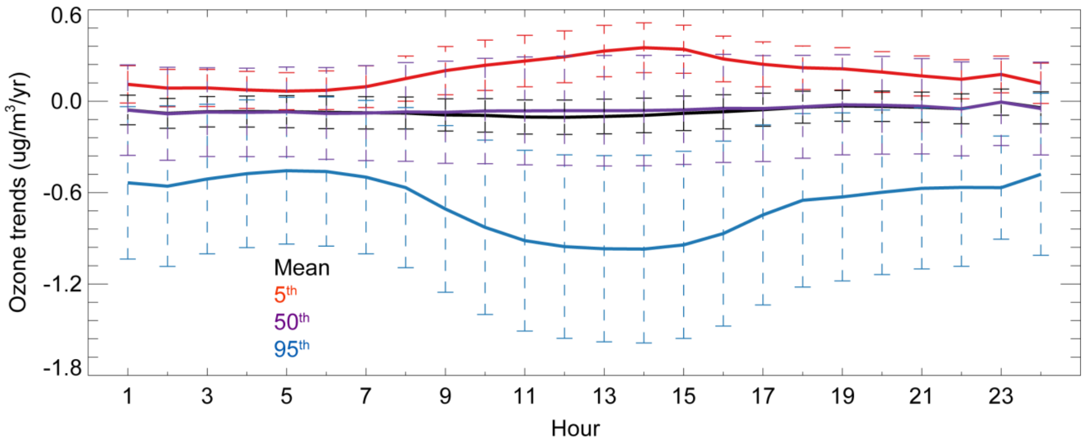

Fig. 2. Trend in the observed surface ozone averaged over Europe, calculated for the selected 93 sites. The black line shows the 1995–2014 linear trends in the deseasonalized European monthly ozone anomalies for each hour of the day (local standard time), the red, purple and blue lines depict the observed trend for 5th, 50th and 95th percentile ozone, respectively, and the dashed bars indicate their standard deviations.

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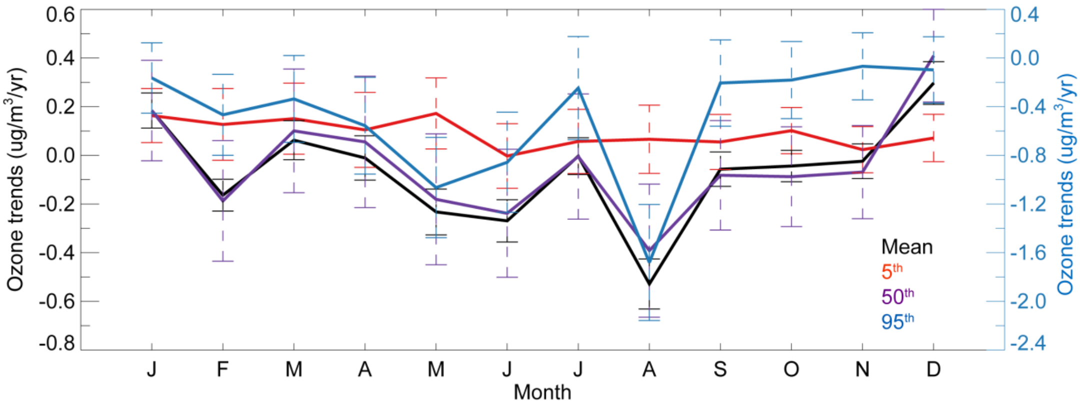

Fig. 3. Monthly trend in the observed surface ozone averaged over Europe for the selected 93 sites. The black line shows the 1995–2014 linear trends in the European mean ozone for each month of the year, the red, purple and blue lines depict the observed trend for 5th, 50th and 95th percentile ozone, respectively, and the dashed bars indicate their standard deviations. The left axis is for the trends of mean, 5th, and 50th percentile ozone, while the right axis for the 95th percentile ozone.

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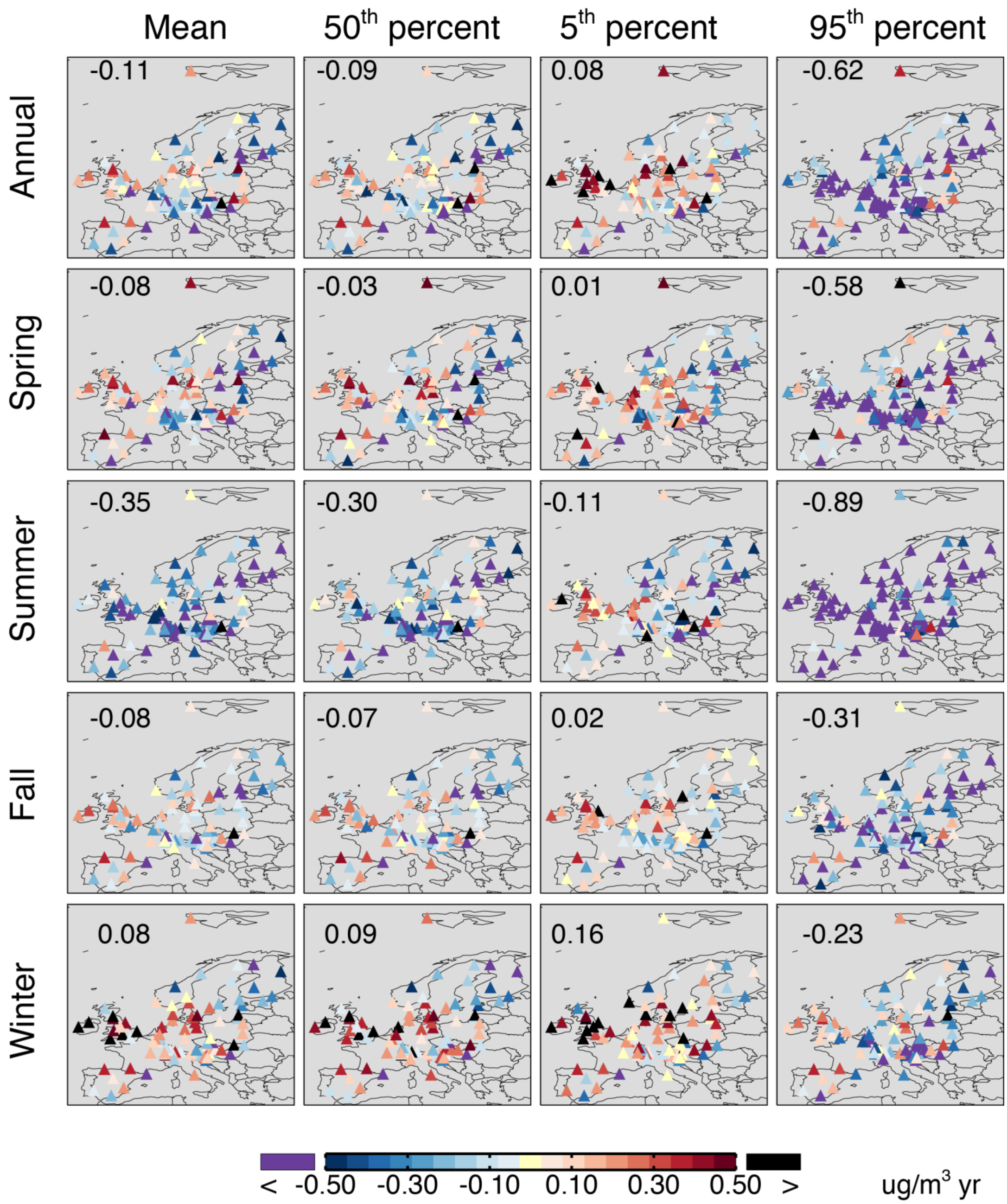

Fig. 4. Spatial distribution of measured daytime ozone trends in μg/m^3 across the selected 93 sites for average, 5th, 50th and 95th percentile ozone in annual mean and four seasons. Also shown in each panel are the average trends over all sites.

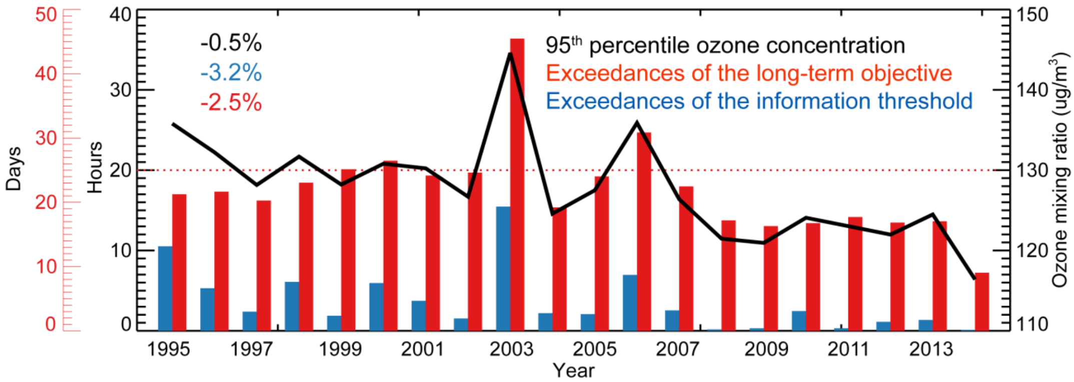

Fig. 5. Annual exceedances of the information threshold (for blue bars, hours should be multiplied by 100, 1-hourly averages: 180 μg/m^3) as well as the long-term objective (red bars, maximum diurnal 8-hourly mean: 120 μg/m^3), compared with the annual 95th percentile ozone concentrations (black line). Red dotted line shows the target value (long-term objective that should not be exceeded more than 25 days per year, averaged over 3 years).

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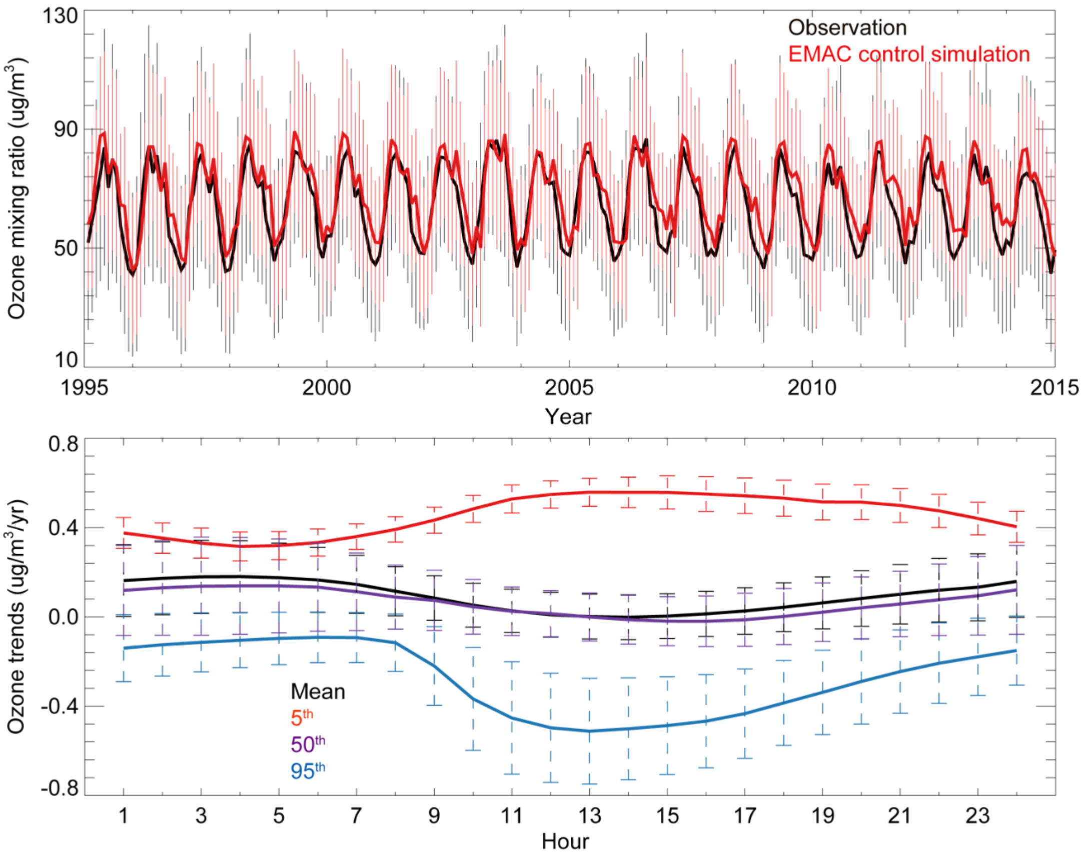

Fig. 6. EMAC modeled ozone in μg/m^3 over Europe during 1995-2014. Time series of measured (black) and modeled (red) monthly mean ozone over the 93 selected sites (top). Trend in the modeled surface ozone averaged over the selected 93 sites for all hours of the day (local time, bottom). The black line shows the 1995-2014 linear trends in the European mean ozone, the red, purple, and blue lines are the modeled trends for 5th, 50th and 95th percentile ozone, respectively. The dashed bars indicate their standard deviations.

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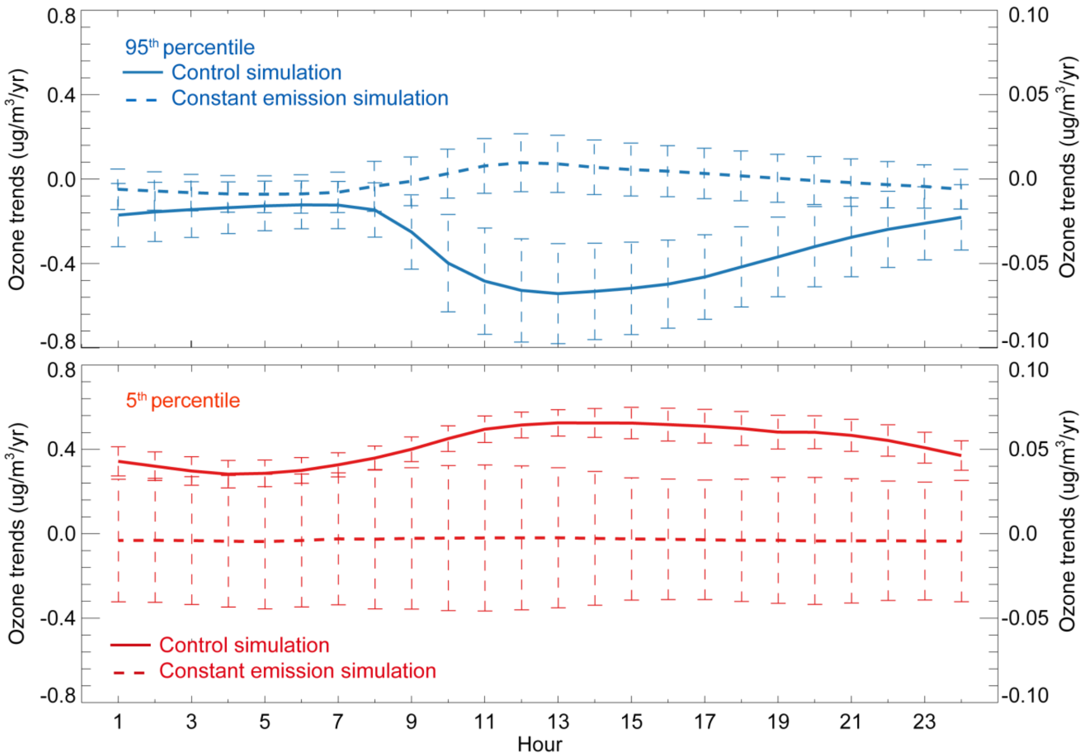

Fig. 7. Modeled trend in the surface ozone averaged over the selected 93 sites for all hours of the day (local time). The solid lines (left legends) show the 1995-2014 linear trends in the control simulation for 95th (top) and 5th percentile (bottom) ozone, respectively. The dashed lines (right legends) represent the modeled trends by the constant emission simulation. The bars indicate their deviations.

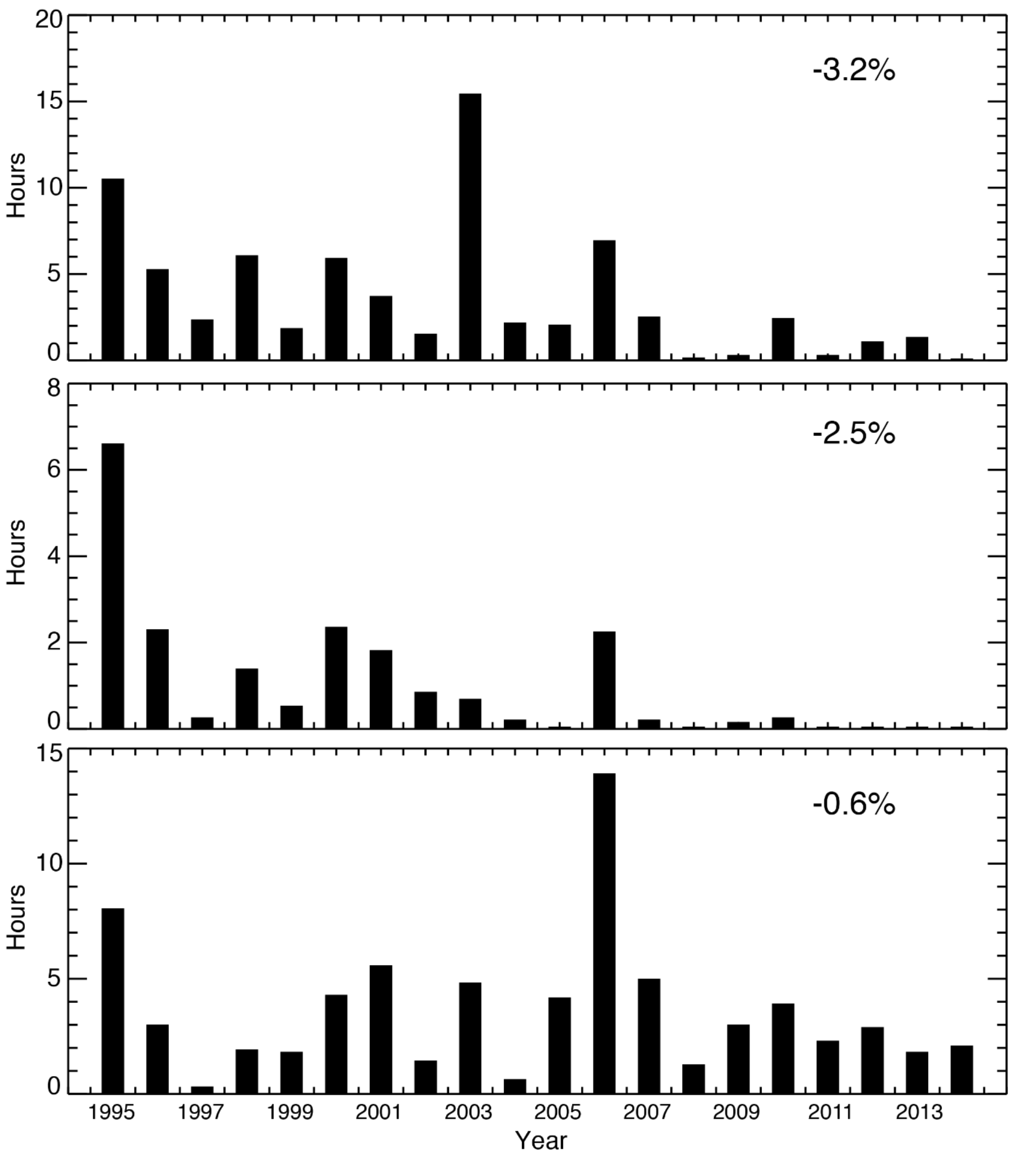

Fig. 8. Annual observed (top) and modeled (middle: control simulation; bottom: constant emission simulation) exceedances of the information threshold (1-hourly averages: 180 $\mu g/m^3$). The hours along the y-axis should be multiplied by 100.

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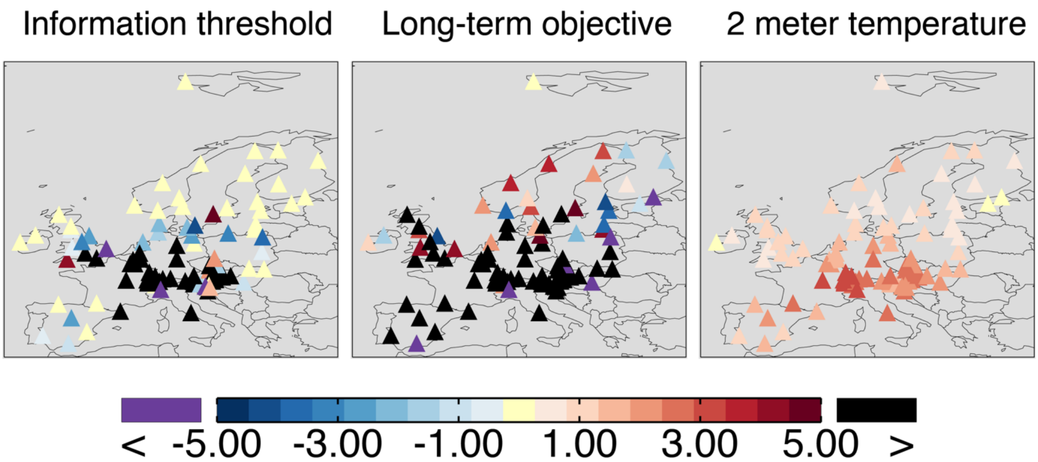

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583 Fig. 9. Spatial distribution of the exceedance anomalies in 2003, relevant to the averages over 1995-2002 and for the
584 information threshold as well as the long-term objective, in comparison with the 2-meter temperature anomalies in
585 each of the sites.
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587 Table 1. Percentage of missing hourly data in each year.

Year	Number of sites	Missing data		
		Whole day	Daytime	Nighttime
1995	113	32.6%	30.6%	34.6%
1996	115	28.8%	26.7%	30.9%
1997	121	23.9%	21.6%	26.2%
1998	120	18.5%	16.0%	21.0%
1999	127	10.4%	7.9%	12.8%
2000	132	9.8%	7.2%	12.3%
2001	134	11.9%	9.4%	14.4%
2002	136	9.3%	6.8%	11.8%
2003	137	12.1%	9.8%	14.4%
2004	135	10.9%	8.5%	13.3%
2005	132	10.5%	8.1%	12.9%
2006	130	10.6%	8.1%	13.1%
2007	132	9.5%	7.0%	12.0%
2008	136	10.8%	8.2%	13.4%
2009	134	10.6%	7.8%	13.3%
2010	136	15.0%	12.6%	17.5%
2011	135	13.8%	11.4%	16.2%
2012	136	14.1%	11.8%	16.4%
2013	136	19.9%	17.8%	22.0%
2014	137	21.0%	19.1%	23.0%

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Table 2. Modeled and observed ozone trends and their standard deviations based on diurnal average ozone concentrations. The mean, 5^{th}, 50^{th}, and 95^{th} percentile represent the monthly statistics of the diurnal averages. The model has been sampled in the same location of the EMEP stations.

	5^{th} percentile	50^{th} percentile	Mean	95^{th} percentile
EMEP (μg/m^3/y)	0.22±0. 15	-0.05±0. 23	-0.07±0. 21	-0.57±0. 34
EMAC (μg/m^3/y)	0.42±0. 14	0.01±0. 10	0.06±0. 09	-0.23±0. 10

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Table 3. Modeled and observed linear trends and their standard deviations of the 1995–2014 European mean annual and seasonal averaged daytime and nighttime mean as well as their 5th, 50th and 95th percentile ozone concentrations.

	Seasons	Mean		5th percentile		50th percentile		95th percentile	
		EMEP	EMAC	EMEP	EMAC	EMMP	EMAC	EMEP	EMAC
Daytime (μg/m³/y)	Annual	-0.09±0.24	0.00±0.06	0.22±0.17	0.45±0.14	-0.06±0.24	-0.01±0.06	-0.81±0.46	-0.48±0.15
	MAM	-0.09±0.27	-0.05±0.08	0.13±0.24	0.52±0.17	-0.02±0.27	-0.02±0.08	-0.93±0.53	-0.49±0.16
	JJA	-0.32±0.24	-0.10±0.07	-0.03±0.26	0.41±0.20	-0.26±0.24	-0.09±0.13	-1.10±0.61	-0.54±0.16
	SON	-0.03±0.19	-0.04±0.05	0.09±0.14	0.36±0.12	-0.04±0.20	-0.02±0.05	-0.24±0.25	-0.44±0.23
	DJF	0.10±0.25	0.18±0.14	0.25±0.15	0.39±0.22	0.05±0.27	0.15±0.20	-0.28±0.31	-0.08±0.05
Nighttime (μg/m³/y)	Annual	-0.05±0.23	0.12±0.11	0.16±0.17	0.38±0.19	-0.05±0.24	0.07±0.12	-0.57±0.36	-0.21±0.10
	MAM	-0.06±0.29	0.08±0.10	0.18±0.23	0.23±0.23	-0.00±0.29	0.04±0.08	-0.64±0.43	-0.20±0.12
	JJA	-0.20±0.27	0.06±0.14	0.07±0.24	0.36±0.22	-0.15±0.28	0.04±0.14	-0.71±0.52	-0.36±0.21
	SON	-0.03±0.21	0.06±0.10	0.05±0.12	0.19±0.16	-0.05±0.23	0.04±0.11	-0.21±0.24	-0.23±0.19
	DJF	0.09±0.24	0.24±0.18	0.14±0.22	0.43±0.27	0.06±0.25	0.20±0.25	-0.24±0.29	-0.05±0.06