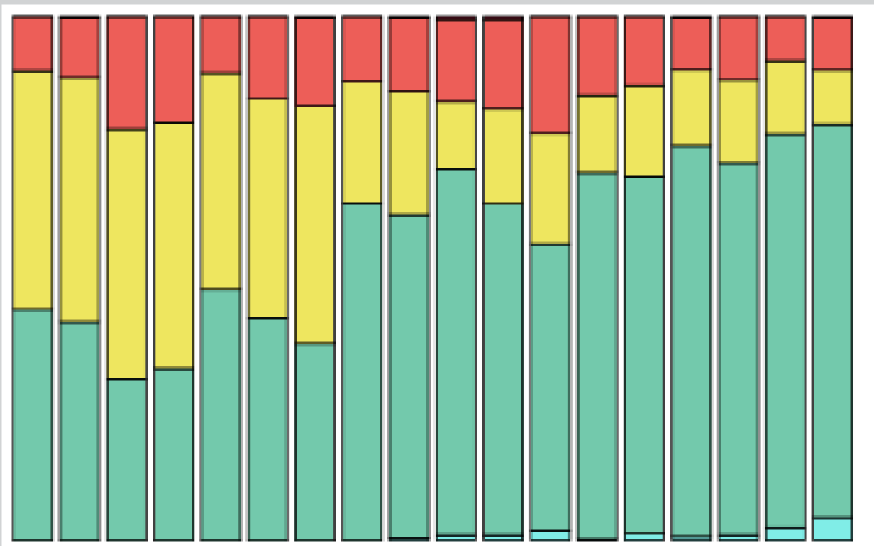
Eionet Report - ETC/ATNI 2019/16

Air Quality Trends in Europe: 2000-2017

Assessment for surface SO_2, NO_2, Ozone, PM_{10} and $PM_{2.5}$

September 2020



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The withdrawal of the United Kingdom from the European Union did not affect the production of the report.
Data reported by the United Kingdom are included in all analyses and assessments contained herein, unless otherwise indicated.

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Summary

We present an assessment of observed air quality trends in Europe aimed at making the most of the regulatory monitoring network to document and explain the effectiveness of air pollution mitigation policies. The focus is on the 2000-2017 time period and surface SO_2, NO_2, ozone, PM_{10} and $PM_{2.5}$, for which we can rely on more than 10,000 stations. Data related to 3,500 stations complied with the requirements, in terms of completeness and representativeness for long-term trend assessments. Such long-term records are only available for countries of the European Union, with one exception for Norway.

Substantial improvements are found for all air pollutants. We assess in detail the absolute and relative trends for a wide range of air pollutant indicators and also discuss the spatial variability of the trends as well as changes in monthly, weekly and hourly variability. These changes are put in perspective with emission reductions in Europe in order to point out the pollutants where a potential mismatch may occur between expected and observed improvements in air pollutant concentration.

The relative change in SO_2 concentrations lies in the 70 to 85% range. This reduction is lower but still in line with the reported emission decrease in Europe (-89%). There is however a slight mismatch between emissions and concentrations in the aftermath of the 2008 economic crisis. Such a mismatch after 2008 also appears for NO_2. But on the contrary to SO_2, this mismatch has a more substantial impact on the overall trend as reduction in concentrations is 30% which is lower than expected given the 53% reduction in emission over the same time period.

The magnitude of ozone peaks (as the fourth highest annual daily maximum of 8hr running mean) decreases by 10% and the number of days exceeding the long-term objective of daily maximum hourly ozone above 120µg/m^3 is reduced by 30 to 50%. Annual ozone mean however increases, especially at urban sites. The increase is less pronounced at rural sites, suggesting that it is mainly related to lower NOx titration effect rather than hemispheric changes. Annual mean ozone increase also contributes to higher health exposure, with median SOMO35 and SOMO10 increasing by 1.3% and 13.4%, respectively at urban stations. This needs however to be considered with respect to the NO_2 reduction in order to understand the net impact on health, for instance by looking at O_x (sum of NO_2 and O_3) which decreases.

Particulate matter annual mean concentrations decrease by 25 to 45%, depending on station typology. The reductions are similar for PM_{10} and $PM_{2.5}$ when comparing collocated measurements. The highest peaks of particulate matter exhibit less relative reduction than the average, showing that episodes of high PM would deserve more focus. PM concentrations decrease faster than primary PM emissions (-30% for primary PM_{10} and -18% for primary $PM_{2.5}$), thanks to the additional impact of the reduction of precursors of secondary PM, such as SOx, NOx and NH_3.

The air quality index over Europe was computed for the whole time period. It gradually improves over the 2000-2017 time period. But most of the improvement concern days in the “moderate” air quality category, whereas the number of days classified as “poor” for air quality remain quite constant. This observation raises specific concern for future improvement of high air pollution episodes.

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1 Introduction

It is well documented that air pollution poses a serious threat for human health and ecosystems. It has been mitigated since the end of the 20th century, in particular through international policy instruments such as the Geneva Convention on the Long Range Transboundary Air Pollution (CLRTAP, 1979) and, as far as Europe is concerned, the National Emission Ceiling Directives (EC, 2001, 2016), which set objectives to be achieved by the implementation of national and local regulations. In order to assess the magnitude of the threat, and the efficiency of mitigation strategies and policies, scientific assessments based on tools to monitor and predict atmospheric composition changes were developed. The CLRTAP launched the European Monitoring and Evaluation Programme (EMEP, www.emep.int), with a dedicated in situ monitoring network, and the European Commission released a number of air quality directives (EC, 1996, 2008, 2004) defining common monitoring principles for countries of the European Union as well as maximum air pollution levels not to be exceeded to ensure a clean air for European citizen.

Decades after having initiated emission reduction strategies and dedicated monitoring networks, several studies taking stock of long-term air quality monitoring have been performed by the EC and the CLRTAP to assess the efficiency of air pollution mitigation strategies (Maas and Grennfelt 2016;EEA, 2009;Colette et al., 2016). The topic has also been of interest for the scientific community with a number of articles devoted to the assessment of air quality trends and relating them to the efforts achieved in terms of emission reductions. The majority of such studies were focused on ozone (Vautard et al., 2006;Sicard et al., 2013;Derwent et al., 2003;Derwent et al., 2010;Jonson et al., 2006;Wilson et al., 2012;Fleming et al., 2018;Simpson et al., 2014) to name just a few, and excluding all the scientific body devoted to tropospheric ozone at a larger scale. But there has also been studies investigating nitrogen and particulate matter trends: (Colette et al., 2011;Guerreiro et al., 2014;Barmpadimos et al., 2012;Turnock et al., 2015;Banzhaf et al., 2015;Turnock et al., 2016;Tørseth et al., 2012).

Several of those investigations relied on both observations and models to discuss policy effectiveness, in general by feeding one or several chemistry-transport models with reported air pollutant emissions before comparing the results with observations to conclude on the effectiveness of policy implementation (Colette et al., 2017). Here we choose a different perspective, by deliberately limiting the scope to the analysis of observations to update the knowledge of the current status of trends in the European air quality. We also intend to relate observed air pollution trends to reported emission changes, to the extent possible.

In this study, the period of interest is 18 years long: 2000-2017, the latest year being constrained by the availability of validated observation released for the year 2017 in 2019 by the European Environment Agency. Such a temporal extent has two positive outcomes. The duration of the record allows concluding on statistical significance of the trends. In addition, larger geographical areas become available for the analysis as the completeness criteria has left in the past wide regions out of the analysis because of the too short monitoring records available.

The input data and statistical methods are presented in Section 2 and the results are discussed in Section 3 where the trend of the various air pollutants of interest are discussed (Sulphur dioxide – SO_2, nitrogen dioxide – NO_2, ozone – O_3, Particulate matter finer than 10µm and 2.5µm – PM_{10} and $PM_{2.5}$) as well as the trends of the Air Quality index in order to provide a synthetic overview of air quality evolution that captures the change for all individual compounds.

2 Methods

2.1 Air quality observations

For this study, we rely on the air quality monitoring databases hosted by the European Environment Agency (EEA). Up to 2012, these datasets were gathered in the AIRBASE database, for which we used the v8 release[1]. After 2013, the EEA database moved to the Air Quality e-reporting system[2]. A technical difficulty lied in matching these two databases because many stations changed names and codes over time. Instead of station names, the matching is performed using the Sampling Point Identification, which is the most reliable meta-data about the consistency of a given record.

The EEA databases differentiate station area (urban, suburban and rural) and typology (background, traffic, industrial). For synthesis, we differentiate background types at urban, suburban and rural areas and considered traffic stations and industrial stations as a whole, irrespectively of their areas.

2.2 Statistical processing

2.2.1 Data completeness

All the surface data available included in the database is used in the present study. We did not apply any outlier detection or filtering considering that the impact of spurious data will be minimised in the aggregation of statistic over a large dataset. We did however perform a completeness check so that too short records were not included in the trend analysis. First the completeness in any given year is assessed so that all datasets (days or hours) within a year where less than 75% of the record are available are discarded. In a second step, we also removed a given station if less than 75% of the years in the 18 year time period (i.e. 5 years or more) were not available.

Regarding temporal resolution, most observations for NO_2, SO_2 and O_3 are available as hourly data so that we used only those records, which allow to investigate daily maximum behaviour and diurnal variations. For PM_{10} and $PM_{2.5}$ there is however a mix of hourly and daily values according to the measurement method used, but most relevant indicators are defined on the basis of daily means. As a consequence, we averaged all hourly records and checked for redundancy before aggregating them in the raw data available as daily means.

A specific work was performed to identify collocated measurements of O_3 and NO_2 in order to discuss Ox (as O_3 + NO_2) trends, but also to compare the relative trends of O_3 and NO_2 at a consistent set of stations. Ideally NO should also be added to O_3 and NO_2 to derive total Ox, but that would have lead to a selection of too few stations because of the scarce collocation of O_3, NO_2 and NO measurements. Likewise, we identified collocated measurements of PM_{10} and $PM_{2.5}$ to compare the trends of fine and coarse PM.

Because until 2007, French authorities reported PM hourly concentrations from automatic devices (TEOM, Beta gaujes) without applying any correction factor to account for the volatilisation of some PM compound during the measurement phase, daily values could not be directly used before that date in that country. Nevertheless, as it was done by the other countries at that time, a correction of annual mean values was applied (factor 1.3, (Malherbe et al., 2017)) so that only annual mean statistics of PM_{10} can be used for the purpose of this study for France up to 2006.

The total number of air quality stations by station type and pollutant available during the period 2000-2017 in the European Union (28 countries) is given in Figure 1. In 2000 only about 2000 records (combinations of stations and pollutants) were available, but in 2017 this number reaches 10000 records. A steady increase of the number of stations is found for all station types and pollutants. Since

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[1] https://www.eea.europa.eu/data-and-maps/data/airbase-the-european-air-quality-database-8, accessed 2/8/2019.

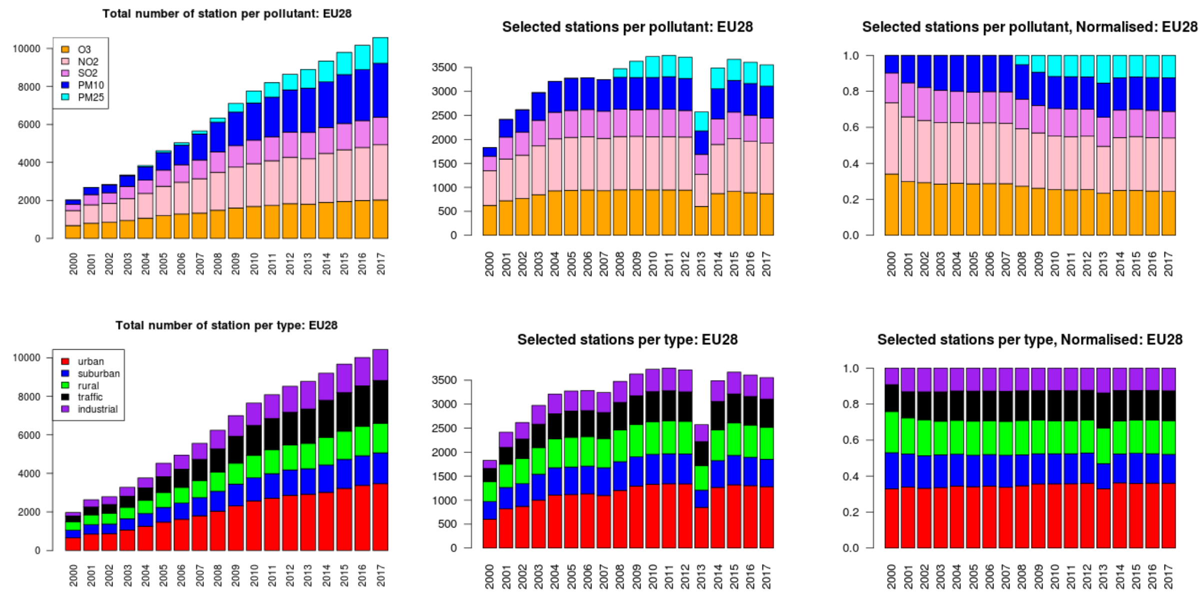
[2] https://www.eea.europa.eu/data-and-maps/data/aqereporting-8, accessed 2/8/2019.

the early 2000s, the PM monitoring network has developed drastically, so that the proportion of gaseous monitoring devices is reduced. Before 2007, $PM_{2.5}$ stations locations were scarce, this is why the trends for $PM_{2.5}$ will be limited to the 2008-2017 time period.

After having applied the completeness checks for trend assessment described above, we kept about 3500 records. If only stations covering the whole period had been selected, the number of station would have been constant in time, but we see here an increase in the number of sites over the first few years because of the relaxed completeness criteria that selects records with only 75% of valid years (i.e. over 2000-2017 for all pollutants except for $PM_{2.5}$ where the time period is 2008-2017). A clear issue occurred in 2013, the year when the EEA system changed from Airbase to AQ e-reporting. As can be seen in the total number of available records, there is no anomaly in the data reported overall. But because of the change in system, some countries used different sampling point identifiers, so that several records cannot be matched with the reminder of the period, making them irrelevant for trend assessment. The vast majority of selected stations passing completeness criteria are located in the 28 countries of the European Union. The only exception being Norway with 6, 15, 1 and 1 stations for O_3, $PM_{2.5}$, PM_{10} and NO_2, respectively.

Apart from this anomaly of 2013, there is no systematic trend in the distribution of station type after 2001. The number of PM monitoring sites increased gradually, so that ozone and NO_2 monitoring became relatively less important.

Figure 1: Number of air quality monitoring station by pollutant (top) and station type (bottom), in the 28 countries of the European Union available over the 2000-2017 time period (left) and passing the completeness criteria for trend assessment in absolute (middle) and relative (right) numbers.



2.2.2 Air pollutant indicators, metrics and indices

We intended to be as comprehensive as possible in terms of statistical indicators, computing for each year: annual, seasonal, monthly, weekly (per day of the week), daily information and corresponding quantiles on the basis of daily means for all compounds. For NO_2 and O_3 we could also compute those aggregated on the basis of hourly observations to derive daily maxima and include diurnal profiles.

We also included a few additional metrics because of their relevance with regards to the European Directive on Air quality (EC, 2008), or health and ecosystem impacts. For ozone, the hourly daily maximum was used to compute the number of days above 120µg/m^3 (long-term objective), 180 µg/m^3 (information threshold) and 240 µg/m^3 (alert threshold). The daily maximum 8-hr average was also used to derive 4DMA8: the annual fourth highest peak, which is considered to be the most representative of ozone peaks given that lower quantiles, or summer average of the peaks are largely influenced by low ozone days (Colette et al., 2016). We also computed health-related metrics: SOMO35 and SOMO10 (sum of ozone daily maxima in excess of 35 ppbv and 10 ppbv, (Malley et al., 2015)) and ecosystem-related metrics : AOT40c and AOT40f (accumulated ozone over 40 ppbv between May and July – included – for crops and between April and September – included – for forests). For NO_2 we considered but eventually excluded the number of hours above 200 µg/m^3 because of the low number of occurrences at most stations. Similarly, the number of days above 125 µg/m^3 and hours above 350 µg/m^3 were excluded for SO_2. For PM_{10} we computed the number of days above 50 µg/m^3 daily limit value.

We also computed air quality indices by country for all air pollutants, using the definition of EEA recalled in Table 1 consist in defining intervals for each air pollutants, the index being subsequently defined as the worst level across available air pollutant observations at a given station. Computing the index therefore requires availability of all pollutants at a given station, which is far from being the case so that modelling is used by EEA as gap filling. In order to avoid such gap filling, we rather compute the index level for all pollutants and take the median by pollutant for all stations in a given country. The country air quality index is then defined here as the worst category for all pollutants.

Table 1: Definition of the EEA Air Quality Index (Source: airindex.eea.europa.eu)

Pollutant	Index level (based on pollutant concentrations in µg/m3)				
	Good	Fair	Moderate	Poor	Very poor
Particles less than 2.5 µm ($PM_{2.5}$)	0-10	10-20	20-25	25-50	50-800
Particles less than 10 µm (PM_{10})	0-20	20-35	35-50	50-100	100-1200
Nitrogen dioxide (NO_2)	0-40	40-100	100-200	200-400	400-1000
Ozone (O_3)	0-80	80-120	120-180	180-240	240-600
Sulphur dioxide (SO_2)	0-100	100-200	200-350	350-500	500-1250

2.2.3 Statistical tests

The statistical method applied for the trend detection is Mann-Kendall (with a p-value of 0.05) and we compute the actual slope using the Sen-Theil approach. Both techniques differ from the more classical least square regression in the fact that they focus on the distribution of pairs of changes, aggregating their sign for Mann-Kendall, or using the median of differences for Sen-Theil. They are thus less sensitive to outliers, but also to autocorrelation and non-normality in the distribution.

The trends presented here are given in unit change per year (µg/m^3/yr in most cases). But we also provide the relative change which is useful to provide order of magnitudes over various pollutants/indicators. The relative change is computed from the Sen-Theil slope, multiplied by the overall duration, and normalised by the estimated level at the beginning of the period. The estimated level at the beginning of the period is the linear fit over the whole time series taken for the year 2000, which minimises the effect of interannual variability compared to using directly the value for the year

2000. Those estimated 2000 levels are used for normalisation of both observed concentrations and emissions in the timeseries. A similar approach is used when comparing distribution or monthly/weekly/daily cycle for the beginning and end of the period, where we use the linear fit for 2000 and 2017 instead of the actual cycle for those years.

2.3 Air pollutant emissions

We used the National air pollutant emission (Primary PM_{10} and $PM_{2.5}$, nitrogen oxides - NO_x, ammonia - NH_3, volatile organic compounds - VOC and sulfur oxides - SO_x), reported to the Convention on Long Range Transboundary Air Pollutant and European National Emission Ceiling Directive. Those were obtained for the EU28 from the EMEP Centre for Emission Inventories and Projections[3] (Emission as used in EMEP models), in the version of July 2019.

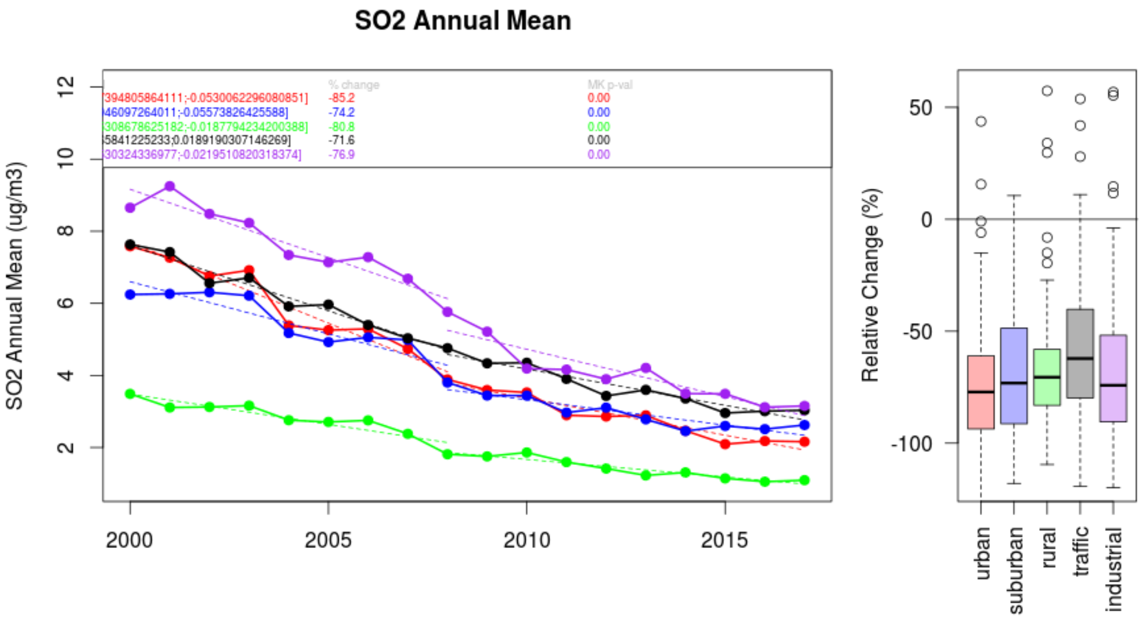
[3] https://www.ceip.at/ms/ceip_home1/ceip_home/webdab_emepdatabase/emissions_emepmodels/, accessed 2/8/2019.

3 Results

3.1 Sulfur dioxide

The trends of sulfur dioxide present the largest decrease of all pollutants. The time series presented in Figure 2 displays the median of annual mean values at all available European stations by site typology as well as several statistical indicators of the trend. It shows that even at industrial sites, the levels in 2017 are comparable to that of rural sites at the beginning of the period. The trends are all significant, and relative change range from 70% (traffic) to 85% (urban background and industrial) depending on station types.

Figure 2: Time series of the European-wide composite (median) of annual mean SO_2 (µg/m^3) per station type and area (red: urban background, blue suburban background, green: rural background, black: traffic, violet: industrial) between 2000 and 2017. The dashed lines show the linear fit between 2000 & 2008 and between 2008 & 2017. The table provides the total number of station (Nsta), the Sen Theil slope of the European-wide composite (ST, µg/m^3/yr), the 95th confidence interval of Sen Theil slopes at all European stations (95th CI), the percentage change between 2000 and 2017 for the European-wide composite (% change), as well as its Mann-Kendall p-value (MK p-val). The boxplots on the right-hand side show the distribution of percentage change between 2000 and 2017 for all stations of each typology.



The distribution of relative change at individual stations also presented in Figure 2 as boxplots that provide the inner 25th to 75th percentiles (filled boxes), median (horizontal line) as well as the 95% confidence intervals (whiskers) and individual values out of that confidence interval (dots). It shows that there are a few sites outside of the 95% confidence interval where the trend is -25% or higher (even positive). The trends and relative changes at individual sites are plotted in the maps provided in supplementary material (Figure S.7). Those smaller decrease, or even increases are really scattered, and would need to be investigated at the site level to check for suspicious records. Such a level of investigation is beyond the scope of a European-wide assessment and would not change our overall conclusions.

Comparing the linear fits over 2000-2008 and 2008-2017, one can notice a flattening out in more recent years. Focusing on EMEP rural sites, it should be noted that most of the decrease for SO_2 had been actually observed in the 1990s, with up to 90% decreases (Colette et al., 2016;Tørseth et al., 2012;Aas et al., 2019).

Figure 3 presents a comparison between reported SOx emission changes between 2000 and 2017 and observed relative change of SO_2 annual means. Here we present the median of relative change over all background (urban, suburban and rural together), traffic and industrial stations. The figure also presents the median over background sites for individual countries in order to provide an indication on the robustness of the trend, even if individual countries are not discussed. Only countries where more than 5 stations of given typology are available are included and the median over those countries is also plotted as a European indicator (which may differ from the time series over all available stations discussed in Figure 2). The annual total emissions by each selected country and their median is also plotted, but without distinction of activity sectors, and therefore to be compared to "background" stations.

The consistency between the rate of change in emissions and observed concentrations is very good until 2007, where a drop was reported in emissions but not matched in observations. Looking in more detail into the emission trends shows that this drop is mainly due to reductions in emissions from the industrial sector and the energy production and distribution sector (EEA, 2018). Between 2007 and 2008, the mismatch is slightly lower when comparing the time series at industrial than for traffic and background sites, but after a few years, the inconsistency is similar at all monitoring sites.

The relative changes are provided for the 8 European countries with dense enough monitoring in Table 2. The agreement between SOx emissions and SO_2 observations is good (-89%, and -81% change, respectively). But the mismatch can be important for a few countries, in particular Germany, Spain, and Italy.

The map of trends in supplementary material (Figure S.7) show that the networks are very scattered in the United Kingdom and Italy. In Spain also the station density is not large, but there seems to be a systematic lower relative decrease of SO_2 in southern Spain for urban background and traffic sites that would deserve further investigation. The lower relative decrease is also pronounced in Germany, which benefits from an excellent coverage of the network.

Figure 3: Time series of country median SO_2 observed at background sites (thin solid lines), and corresponding country SOx emissions (thin dashed lines) normalised to estimated 2000 levels. The thick solid lines are for the median of selected countries of observed over traffic (black), industrial (violet) and background (cyan) sites. The thick dashed red line is for the median of emissions in selected countries. The number of stations is provided in brackets.

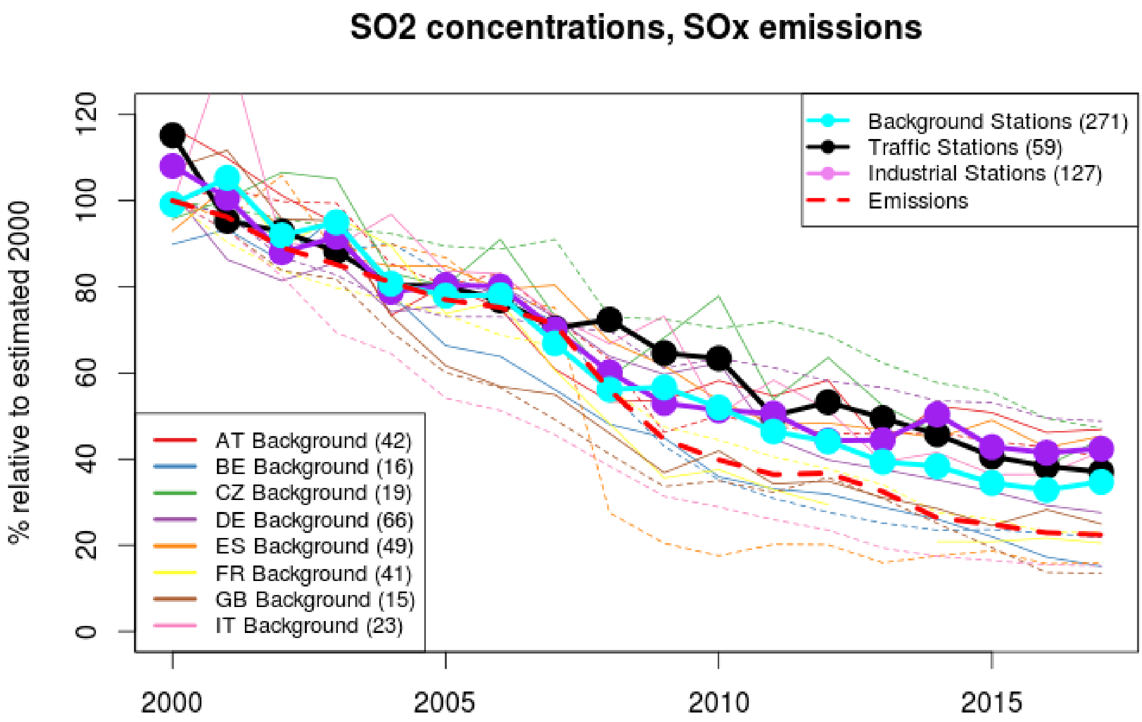


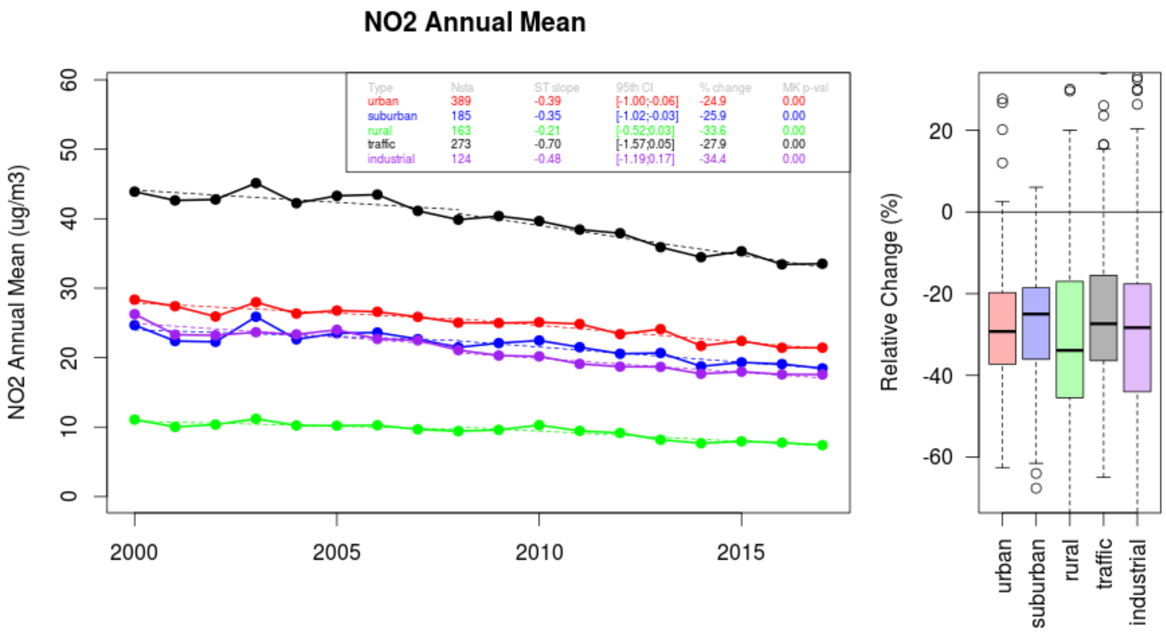
Table 2: Change, relative to 2000 (in %), for emissions and concentrations, as median over countries with enough observations

	SOx,SO_2		NOx,NO_2		PM_{10}		$PM_{2.5}$	
	Emis	Conc	Emis	Conc	Emis	Conc	Emis	Conc
AT	-72	-67	-43	-18	-32	-45		
BE	-94	-98	-53	-34	-44	-43		
CZ	-55	-62	-50	-29	-22	-36		
DE	-49	-74	-41	-25	-30	-40	-46	-32
ES	-102	-60	-53	-31	-34	-43	-29	-23
FI					-35	-32		
FR	-87	-97	-56	-35	-49	-36	-56	-41
GB	-101	-88	-61	-32			-25	-29
IT	-101	-54	-60	-34	-21	-41	-14	-20
NL			-51	-33	-43	-46		
PL					-17	-14	-14	-30
EU28	-89	-82	-53	-30	-30	-44	-18	-33

3.2 Nitrogen dioxide

The median trend of annual mean nitrogen dioxide over Europe is displayed in Figure 4 as the time series of the median across all European sites. It shows a clear downward trend for all station types (Figure 4). The interannual variability is low, except for the year 2003. The relative changes are within -20% to -40% for the central interquartile part of the distribution across stations (25 to 75 percentiles), they are similar for all station types, except for rural station where the median relative change is larger. The comparison of the linear fits over the beginning (2000-2008) and end (2008-2017) of the period indicated as dashed straight lines show that there is no real flattening of the trend except at traffic sites where the decrease is more pronounced over the later period. On the contrary, a slowdown of the decrease has been reported over the United States between (2005-2009) and (2011-2015) (Jiang et al., 2018). Over Europe this slowdown appears to have occurred earlier as the median change was -41% over (1990-2001) and -28% over (1990-2001) (Colette et al., 2016).

Figure 4: Same as Figure 2 for NO_2.

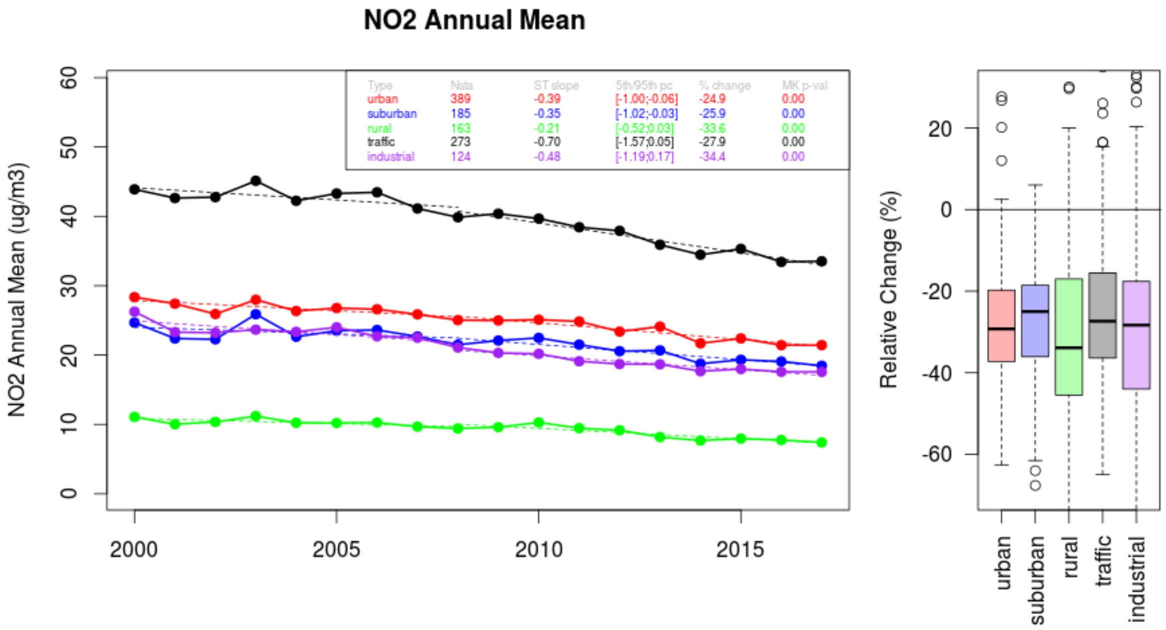


In order to discuss the relative evolution of low and high NO_2 values, the absolute and relative trends per quantiles between 0 and 100 is given in Figure 5. At each monitoring sites, the percentiles distribution of daily mean NO_2 is computed every year to derive the absolute trend and relative change of each corresponding percentiles. Figure 5 provides the median trend and change for each percentile by typology of station. It appears that the absolute largest declines are found for highest percentiles. This is reflected by the fact that the downward trend is larger for the annual mean than for the annual median (-29.8; -28.9, -36.1, -30.8 and -40.1% versus -24.9, -25.9, -33.6, -27.9 and -34.4 for urban, suburban, rural, traffic and industrial sites, respectively), see Table 3. On the contrary, the relative changes are much larger for the lower percentiles (up to 40%), whereas the peaks have only declined by 20%.

Table 3: Summary of observed SO_2 and NO_2 trends for various indicators and station typology: total number of station (Nsta), Sen Theil slope of the European-wide composite (ST, µg/m^3/yr), 5th and 95th quantiles of Sen Theil slopes at all European stations, percentage change between 2000 and 2017 for the European-wide composite (% change), as well as its Mann-Kendall p-value (MK p-val).

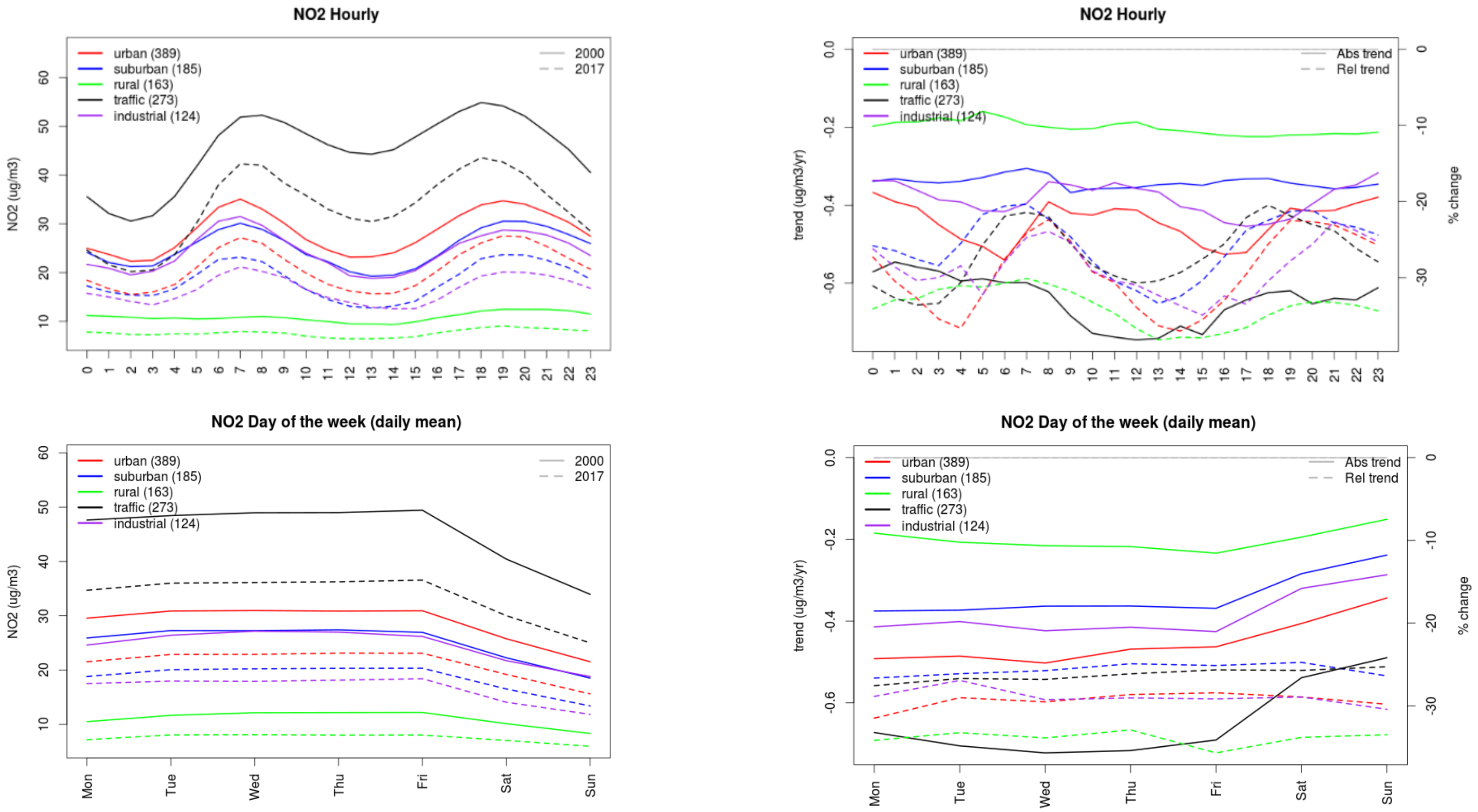
Pollutant	Metric	Type	Nsta	ST Slope	5th and 95th quantiles of ST slope	% change	MK p-val
SO2	Annual Mean	urban	168	-0.34	[-0.94;-0.05]	-85.2	0.00
SO2	Annual Mean	suburban	83	-0.26	[-1.11;-0.06]	-74.2	0.00
SO2	Annual Mean	rural	104	-0.15	[-0.33;-0.02]	-80.8	0.00
SO2	Annual Mean	traffic	75	-0.29	[-1.07;0.02]	-71.6	0.00
SO2	Annual Mean	industrial	155	-0.39	[-1.34;-0.02]	-76.9	0.00
SO2	Annual Median	urban	168	-0.26	[-0.73;0.00]	-83.7	0.00
SO2	Annual Median	suburban	83	-0.20	[-0.68;0.00]	-74.0	0.00
SO2	Annual Median	rural	104	-0.11	[-0.26;0.01]	-75.7	0.00
SO2	Annual Median	traffic	75	-0.20	[-0.82;0.05]	-62.0	0.00
SO2	Annual Median	industrial	155	-0.28	[-0.87;0.05]	-77.5	0.00
NO2	Annual Mean	urban	389	-0.39	[-1.00;-0.06]	-24.9	0.00
NO2	Annual Mean	suburban	185	-0.35	[-1.02;-0.03]	-25.9	0.00
NO2	Annual Mean	rural	163	-0.21	[-0.52;0.03]	-33.6	0.00
NO2	Annual Mean	traffic	273	-0.70	[-1.57;0.05]	-27.9	0.00
NO2	Annual Mean	industrial	124	-0.48	[-1.19;0.17]	-34.4	0.00
NO2	Annual Median	urban	389	-0.44	[-1.01;-0.06]	-29.8	0.00
NO2	Annual Median	suburban	185	-0.36	[-0.98;-0.04]	-28.9	0.00
NO2	Annual Median	rural	163	-0.19	[-0.50;0.03]	-36.1	0.00
NO2	Annual Median	traffic	273	-0.76	[-1.55;0.08]	-30.8	0.00
NO2	Annual Median	industrial	124	-0.52	[-1.20;0.14]	-40.1	0.00

Figure 5: For NO_2 and each typology of station, absolute trend (solid lines) and relative change (dashed lines) of the percentiles of daily means.



This larger decline of high NO_2 levels in absolute terms is also seen in diurnal cycles (Figure 6). The diurnal cycle displays a usual two-peak (morning/evening) profile. What is noticeable is the relative change per hour of the day, where it appears clearly that those peaks were not reduced as efficiently as lower values (see dashed lines in the upper right panel of Figure 6). The same figures also show the median by day of the week, which displays a marked decrease over weekends compared to week days. This cycle also illustrates that NO_2 levels observed in 2017 in working days are similar to those of weekends in 2000, even at traffic sites. But here the relative change is very consistent between week days and weekends, which is contrary with the relative change in the diurnal profile. A possible explanation for the lower relative decrease during the morning/evening rush hour could be the lower efficiency of end-of-pipe technologies in high traffic conditions.

Figure 6: Left column: diurnal cycle (top) and weekly cycle (bottom) of NO_2 at various station type estimated from the whole time series in 2000 (solid lines) and 2017 (dashed lines). Right column: corresponding absolute (solid lines) and relative (dashed lines) trends.

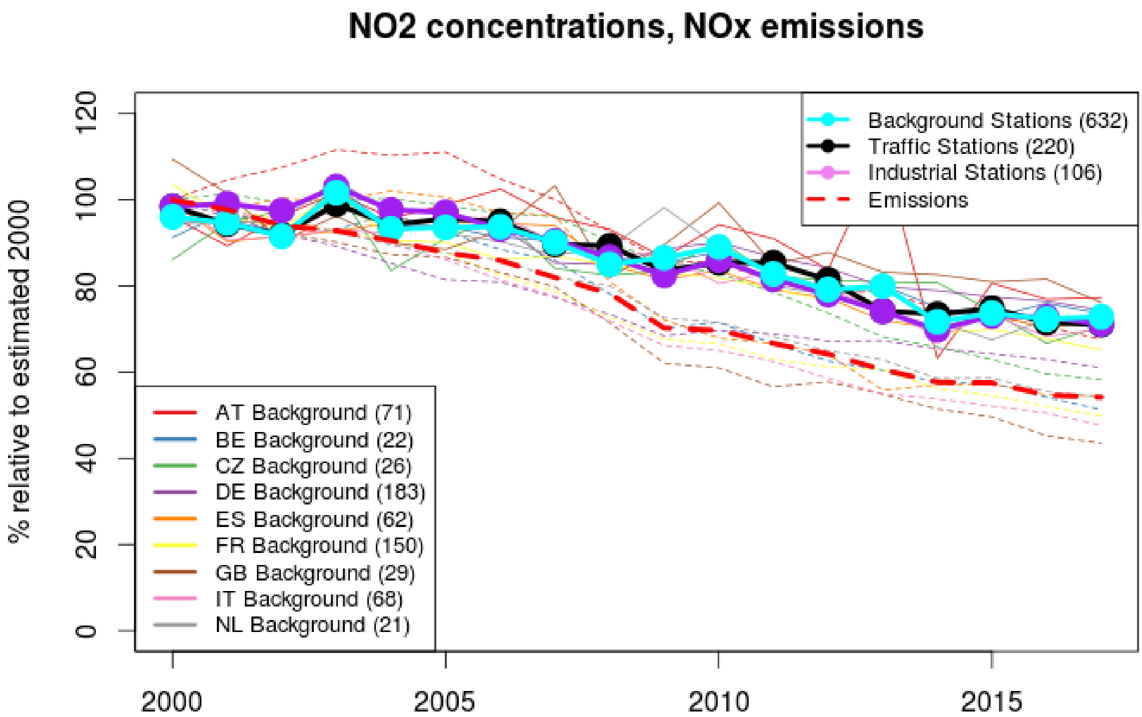


There is some geographical variability in NO_2 annual mean trends but the differences appear more clearly on relative changes (see maps in supplementary material Figure S.16). In particular, a lower relative decline is found over Germany, Austria and the Czech Republic compared to other countries.

The comparison between the trend in emissions and observation is presented in Figure 7 and the corresponding numbers are in Table 2. As mentioned in Section 3.1, this comparison must be handled with care because of network representativeness limitations.

Again, the agreement was quite good up to 2008, but after 2009, the mismatch becomes clear for all station types: background but also traffic and industrial. The mismatch is quite systematic over European countries with enough measurement sites, so that the comparison over EU28 points out a disagreement: -53% change in emissions, whereas NO_2 concentrations only decreased by 32% (see Table 2). As for SOx, the sharp decrease in emission between 2008 and 2009 (except in Austria and Czech Republic) is due to industry and energy sectors (EEA, 2018). The fact that it does not lead to air pollutant concentration reductions would deserve further investigation.

Figure 7: Same as Figure 3 for NOx emissions and NO_2 concentrations.

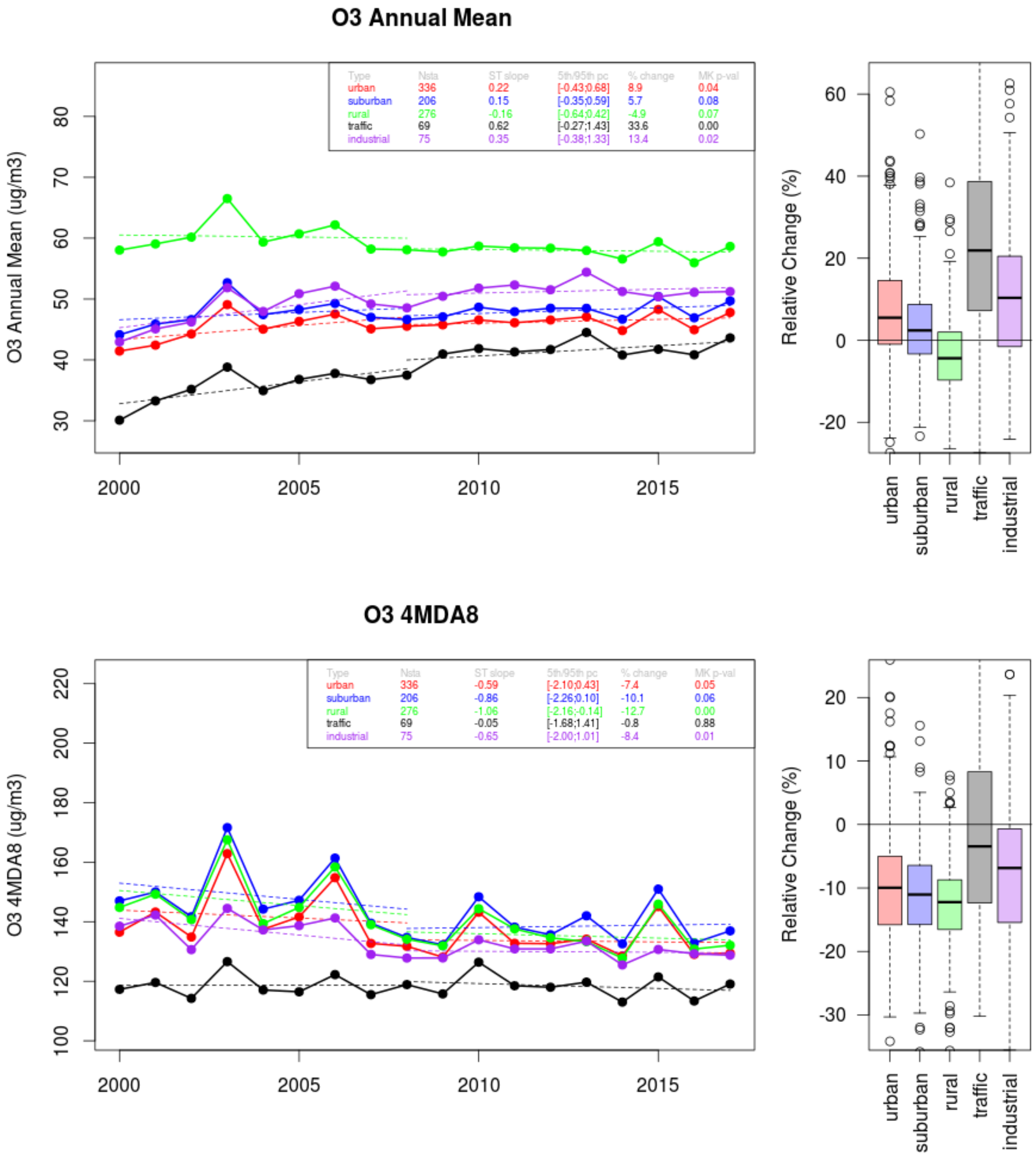


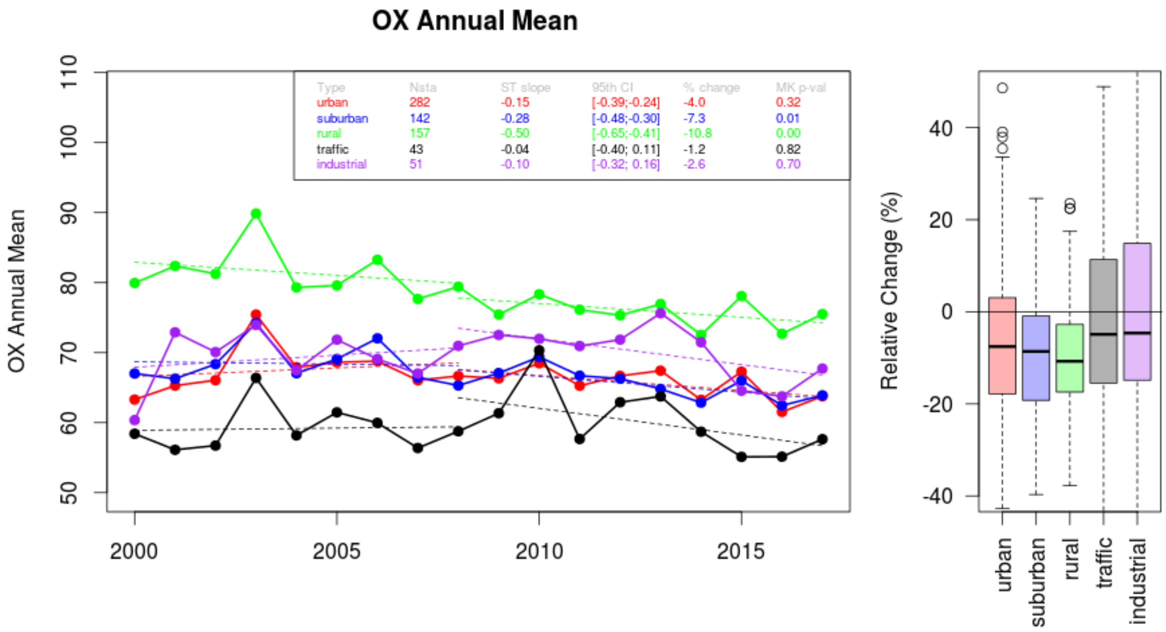
3.3 Ozone

For ozone, opposite trends have been reported before with decreases of high ozone peaks, whereas annual mean ozone increase or display no significant trends (Fleming et al., 2018;Simpson et al., 2014). We confirm this finding and establish that annual mean ozone increases while peaks decrease (Figure 8). The increase of annual mean can be substantial, especially at traffic sites with 25% of the sites showing relative increases of 40% or more. There is however a clear flattening of this upward trend, with more modest increases found since 2008. The only exception is for rural sites where the trend was already flat in the earlier part of the period. Amongst all the factors that bear upon surface ozone, the recent increase of annual mean ozone is generally attributed to hemispheric transport (Cooper et al., 2014) or reduced titration as a result of NOx emission decreases (Monks et al., 2015). The clear difference between rural sites and other typologies indicates that the decreased titration has more impact on the recent trend in Europe than hemispheric transport (Jonson et al., 2006).

The ozone peaks are assessed from the 4MDA8 trend. The fourth highest value is taken instead of the summertime average because when only a handful of significant ozone air pollution episode occur in a year for a given station, the summertime average of daily maxima is not really representative of high ozone episodes. Ozone peaks decrease clearly over the period, of about 10%, except at traffic sites where the decrease is smaller. There is a flattening of the trend over recent year, but interannual variability is high for ozone, so that the apparent flattening is largely influence by the two outstanding years of 2003 and 2006.

Figure 8: Same as Figure 2 for ozone annual mean (top), fourth highest daily peak (4MDA8, middle), and Ox (as O_3+NO_2, bottom).





The trends of daily maxima ozone percentiles illustrate well the difference between ozone trends for high and low concentrations (Figure 9). We show here only percentiles of the maximum daily hour value, but the percentiles of the daily means are provided in Supplementary Material (Figure S.27), they display a similar pattern slightly shifted so that there is no decrease at all at traffic sites.

High quantiles decrease by about 10% at all sites, while at urban background, traffic and industrial sites low quantiles increase by more than 10% below the 20th percentile. As a result of the decrease in ozone peaks, the number of days above the long-term air quality objective of 120µg/m^3 is also reduced by 28%, 31% and 42% at urban, suburban and rural sites, respectively (Table 4).

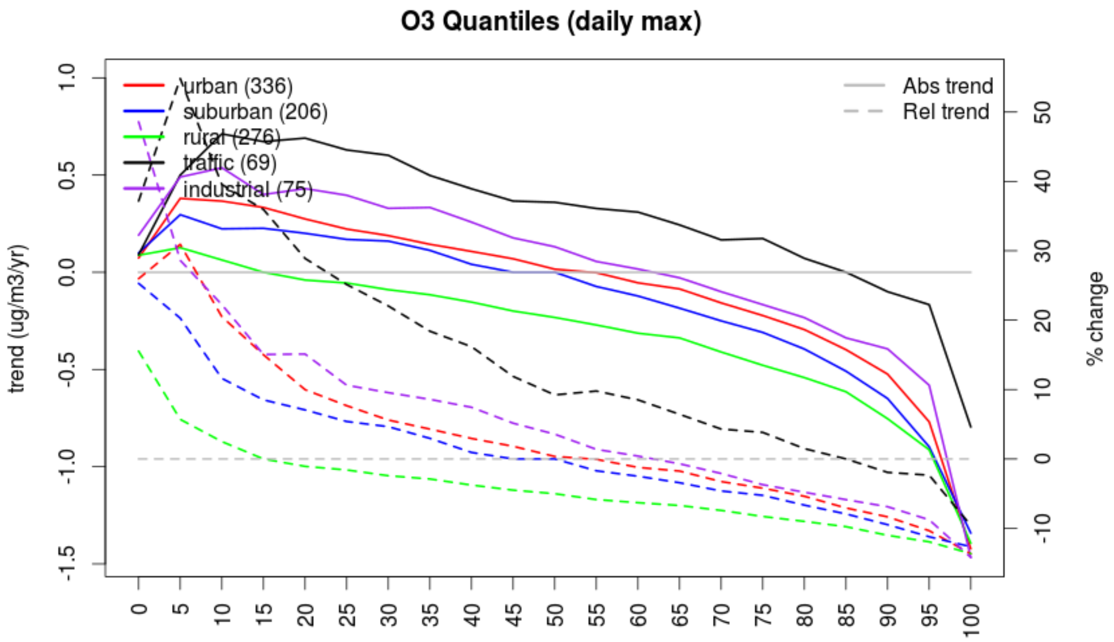
Table 4: Same as Table 3 for ozone indicators

Metric	Type	Nsta	ST Slope	5th and 95th quantiles of ST slope	% change	MK p-val
Annual Mean	urban	336	0.22	[-0.43;0.68]	8.9	0.04
Annual Mean	suburban	206	0.15	[-0.35;0.59]	5.7	0.08
Annual Mean	rural	276	-0.16	[-0.64;0.42]	-4.9	0.07
Annual Mean	traffic	69	0.62	[-0.27;1.43]	33.6	0.00
Annual Mean	industrial	75	0.35	[-0.38;1.33]	13.4	0.02
Annual Median	urban	336	0.20	[-0.44;0.79]	8.2	0.01
Annual Median	suburban	206	0.18	[-0.37;0.70]	6.9	0.03
Annual Median	rural	276	-0.15	[-0.62;0.45]	-4.4	0.04
Annual Median	traffic	69	0.78	[-0.15;1.77]	43.8	0.00
Annual Median	industrial	75	0.38	[-0.42;1.42]	14.7	0.00

Metric	Type	Nsta	ST Slope	5th and 95th quantiles of ST slope	% change	MK p-val
4MDA8	urban	336	-0.59	[-2.10;0.43]	-7.4	0.05
4MDA8	suburban	206	-0.86	[-2.26;0.10]	-10.1	0.06
4MDA8	rural	276	-1.06	[-2.16;-0.14]	-12.7	0.00
4MDA8	traffic	69	-0.05	[-1.68;1.41]	-0.8	0.88
4MDA8	industrial	75	-0.65	[-2.00;1.01]	-8.4	0.01
Nday max > 120ug/m3	urban	336	-0.53	[-2.63;0.36]	-28.2	0.02
Nday max > 120ug/m3	suburban	206	-0.78	[-2.77;0.03]	-31.1	0.01
Nday max > 120ug/m3	rural	276	-1.00	[-3.28;0.00]	-41.9	0.00
Nday max > 120ug/m3	traffic	69	0.00	[-2.32;1.29]	0.0	0.91
Nday max > 120ug/m3	industrial	75	-0.43	[-3.83;1.05]	-25.6	0.04
SOMO35	urban	336	27.90	[-1859.77;1232.45]	1.3	0.82
SOMO35	suburban	205	-170.62	[-2019.53;703.47]	-6.2	0.36
SOMO35	rural	275	-790.42	[-3479.48;766.83]	-23.0	0.01
SOMO35	traffic	69	646.86	[-1398.62;2324.69]	71.4	0.01
SOMO35	industrial	75	459.55	[-2385.27;2339.74]	22.2	0.13
SOMO10	urban	336	1727.67	[-3418.61;5617.34]	13.4	0.02
SOMO10	suburban	206	976.67	[-3011.74;4610.24]	7.0	0.13
SOMO10	rural	276	-759.03	[-4802.58;3331.87]	-4.0	0.23
SOMO10	traffic	69	4146.50	[-1894.72;9685.98]	49.1	0.00
SOMO10	industrial	75	3185.00	[-3181.19;10864.18]	23.5	0.00
AOTcrops	urban	335	-71.02	[-759.43;265.42]	-9.5	0.54
AOTcrops	suburban	205	-206.32	[-828.78;127.56]	-20.7	0.17
AOTcrops	rural	275	-200.73	[-911.08;71.24]	-21.0	0.08
AOTcrops	traffic	65	128.36	[-483.88;580.85]	43.9	0.11
AOTcrops	industrial	74	-85.12	[-772.25;408.13]	-13.1	0.60
AOTforest	urban	335	-183.50	[-1248.70;558.84]	-15.0	0.17
AOTforest	suburban	205	-273.81	[-1293.80;175.36]	-17.1	0.01
AOTforest	rural	275	-530.38	[-1639.38;159.27]	-32.1	0.00
AOTforest	traffic	67	273.40	[-773.85;1289.65]	59.1	0.05
AOTforest	industrial	74	-77.33	[-1413.75;639.11]	-7.1	0.65

The trends in ozone health and ecosystem exposure are influenced by both high and low percentiles of ozone distributions. The trend of SOMO35 at urban and suburban sites is not significant and the relative change is +1.6% and -6.2%, respectively. The decrease is significant at rural sites and reaches -23%. SOMO10 is more influence by the background, so that the increase at urban sites is significant and reaches 13.4%, whereas changes are not significant at suburban and rural sites. Regarding ecosystem, AOT40 for crops is reduced by 21% but the interannual variability is so large that the trend is not significant. For forests, however the -32% change is indeed significant.

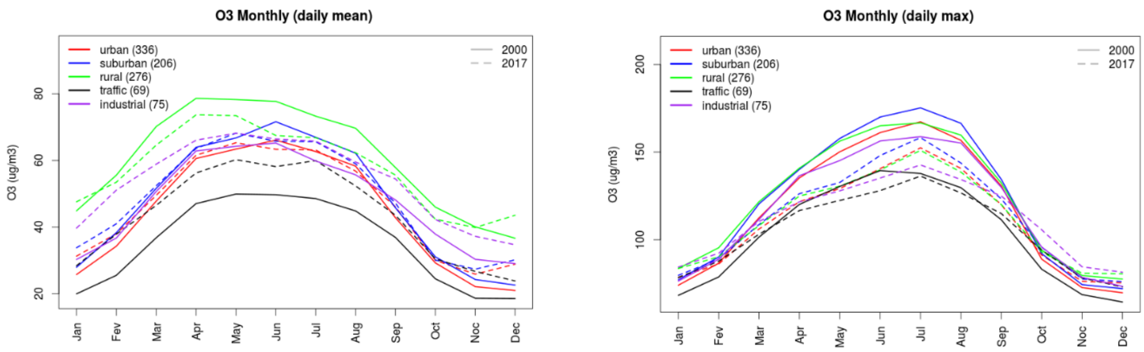
Figure 9: *For ozone and each typology of station, absolute trend (solid lines) and relative change (dashed lines) of the percentiles of daily maxima.*



Ozone displays a strong seasonal cycle illustrated in Figure 10 for both ozone daily means and daily maxima. Comparing the monthly cycle at the beginning (2000) and end (2017) of the period show that the summer peak of daily mean ozone vanished at rural sites, where a clear spring maximum now occurs. This springtime is generally attributed to tropospheric ozone burden increase either in relation to enhance stratosphere-troposphere exchange or long range transport (Butler et al., 2018). The change in summer peak at urban and suburban site is really marginal regarding daily means.

For daily peaks, the change largest from spring to summer compared to fall and winter, but unlike daily mean, there is no real modification in the pattern of the seasonal cycle so that the peaks still occur in June, July and August.

Figure 10: *Monthly cycle of daily mean (left) and daily maxima (right) ozone at various station type estimated from the whole time series in 2000 (solid lines) and 2017 (dashed lines).*

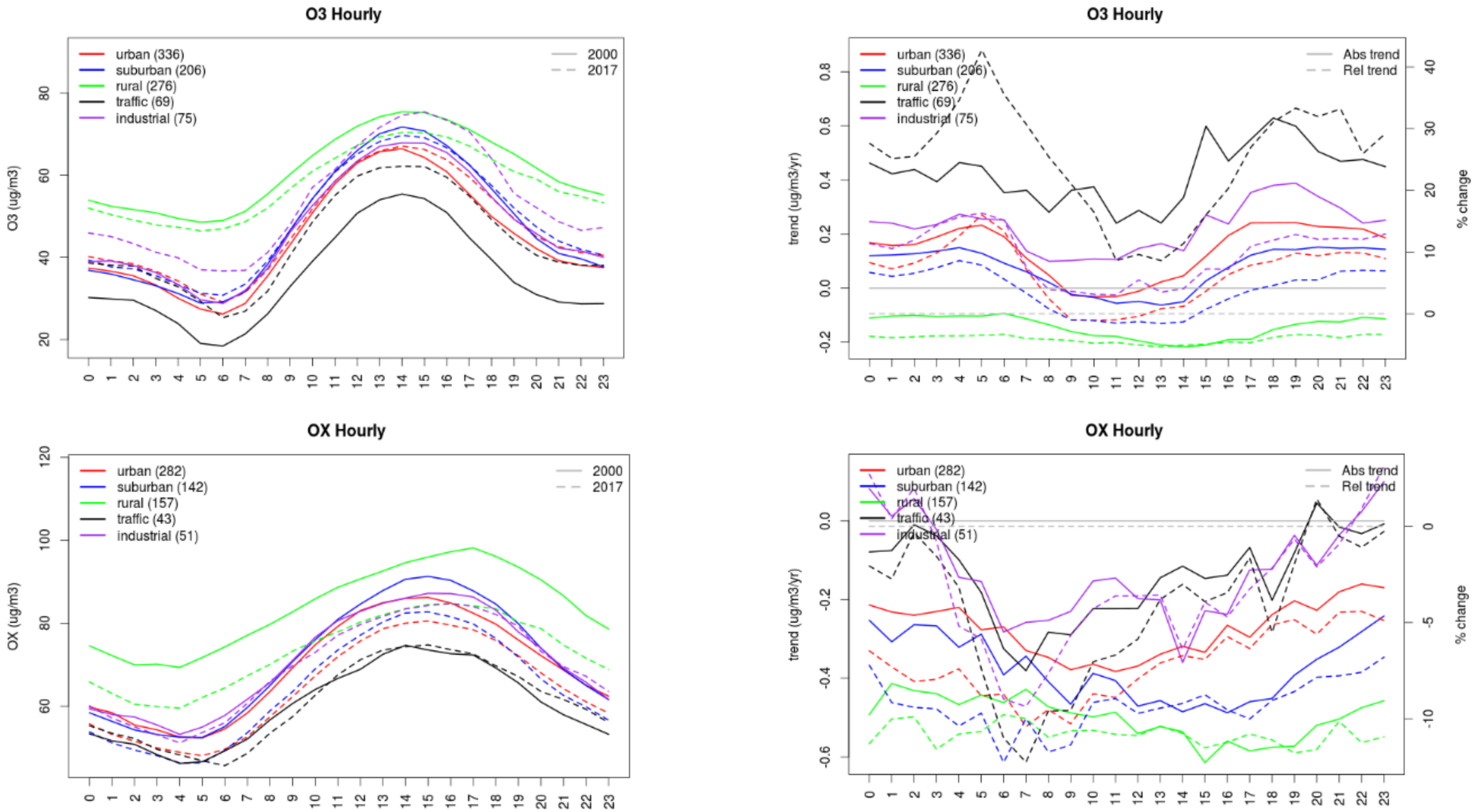


The weekly cycles of ozone are provided in Supplementary Material (Figure S.30). It displays an opposite signal as NO_2 with a weekend increase, especially pronounced for daily mean, but also to some extent for daily maxima, especially at traffic sites. There are differences between the trends by day of the week, with less increase of daily means at industrial sites on Tuesdays, and on Fridays: less decline at rural sites and more increase at urban sites. These features are somehow anecdotic but would be worth investigating in relation to changes in practices (e.g. industrial maintenance or longer week end breaks in summer).

The ozone diurnal cycles are shown in Figure 11. Both the cycle estimated for 2000 and 2017, and the absolute trend and relative change are available on the Figure. For traffic sites, the increase occurs all day long and the relative increase is largest during morning and evening rush hours. For rural sites, the decrease occurs also all day long. But for urban and suburban sites, a night-time increase is compensated by a daytime decrease. Note that those cycles are averaged over a full year and would be shifted if considered only over summer.

Figure 11 also show the diurnal cycle of Ox defined as the sum between O_3 and NO_2. Because of the fast O_3/NO_2 reaction, it is chemically relevant to consider the trend of their sum. They also both have adverse health effects. And as we saw earlier, their trends are opposite (at least in terms of daily means). So that it is difficult to conclude whether, for instance, ozone increases at urban sites should really be a concern for human exposure if it is associated with nitrogen dioxide decreases. Ox trend (Figure 8) are significantly decreasing at suburban and rural sites (-7% and -11% relative change, respectively). At traffic and urban sites, the trends are not significant (-1.2% and -4%, relative change, respectively). In addition, the diurnal cycle of Ox shows that even if the amplitude of the decrease is smallest during the night, there is no actual increase in Ox. The largest decrease at traffic stations is found during the morning rush hour, which demonstrate that even if NO_2 reduction measures led to some increases in ozone, there is no such increase when considering Ox instead of ozone alone.

Figure 11: Left column: diurnal cycle of ozone (top) and Ox (as O_3+NO_2, bottom) at various station types estimated from the whole time series in 2000 (solid lines) and 2017 (dashed lines). Right column: corresponding absolute trend (solid lines) and relative changes (dashed lines



The spatial variability of the trends of ozone peaks (in terms of 4MDA8) is relatively limited. There is no strong latitudinal gradient that would be expected from different chemical regimes and photolysis rates. The amplitudes of the peaks are slightly less reduced in Germany (to be related to the lower NO_2 decreases discussed in Section 3.2) and a couple of coastal stations in Spain. The trends in SOMO35 are much more variable in space, which is consistent with the lack of significant trends in urban and suburban areas.

3.4 Particulate Matter

Strong significant downward trends of annual mean PM_{10} were observed in Europe since 2000 for all station types (Figure 12). The decrease of the median over all European stations of annual mean PM_{10} is higher than 40%. The comparison of trends in the earlier and later part of the period indicate no flattening out, except at rural sites. For annual mean $PM_{2.5}$, we can only consider the decrease after 2008 because of the scarcity of the network before that date. All trends are significant, except at rural sites, but the decrease of annual mean $PM_{2.5}$ is slightly lower, about 30%.

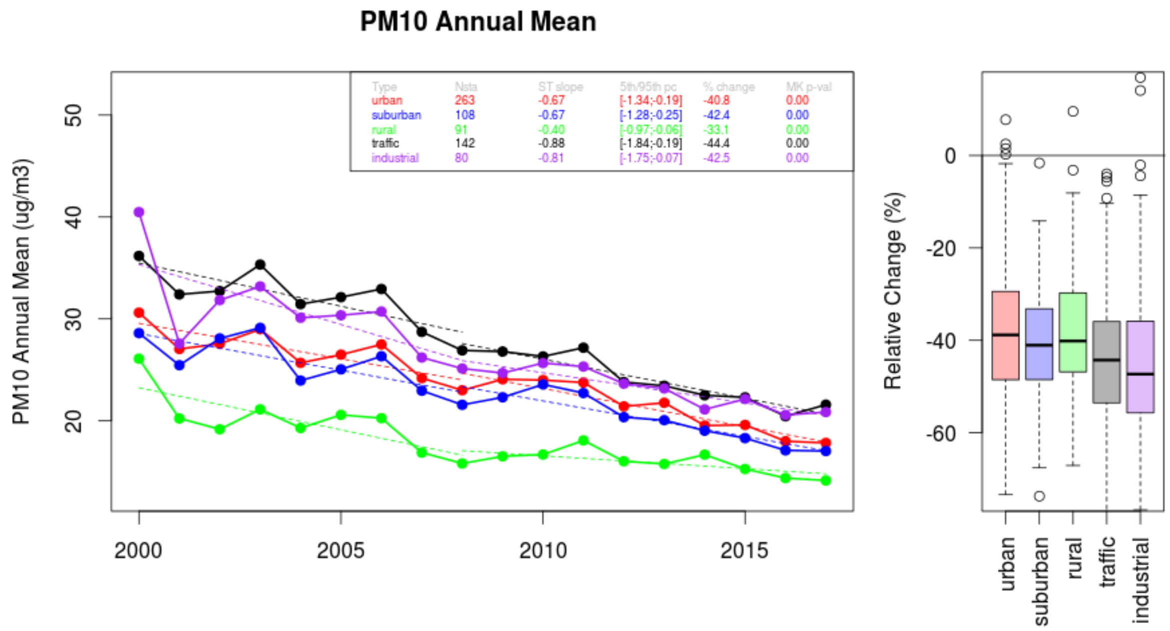
This lower decrease of $PM_{2.5}$ compared to PM_{10} could raise specific question on the efficiency of emission reductions. Those number are however based on different monitoring networks and different time periods for $PM_{2.5}$ and PM_{10}. We should therefore limit the comparison to the sites where both $PM_{2.5}$ and PM_{10} are measured and using a consistent time period (2008-2017). Then we find that annual mean $PM_{2.5}$ and PM_{10} decreases are very similar, yet slightly larger for $PM_{2.5}$, except at rural sites: $PM_{2.5}$ decrease by 15%, 22%, 8%, 32%, and 35% while PM_{10} decrease by 14%, 14%, 10%, 32%, 26% at urban, suburban, rural, traffic and industrial sites, respectively.

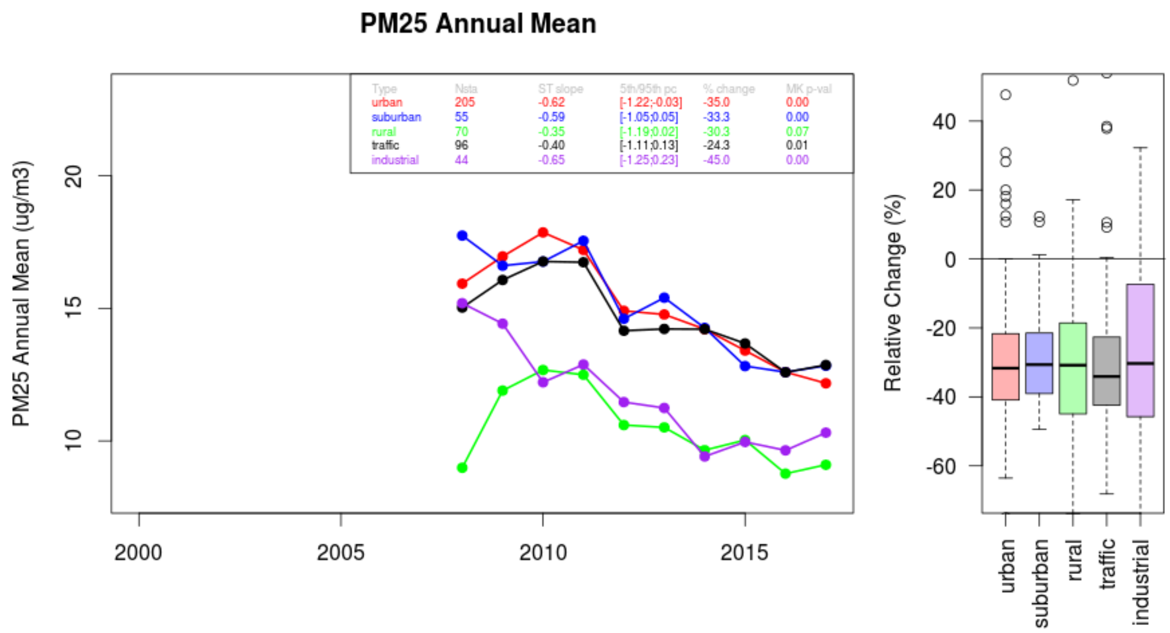
Table 5: Same as Table 3 for PM_{10} and $PM_{2.5}$ indicators

Pollutant	Metric	Type	Nsta	ST Slope	5th and 95th quantiles of ST slope	% change	MK p-val
PM10	Annual Mean	urban	263	-0.67	[-1.34;-0.19]	-40.8	0.00
PM10	Annual Mean	suburban	108	-0.67	[-1.28;-0.25]	-42.4	0.00
PM10	Annual Mean	rural	91	-0.40	[-0.97;-0.06]	-33.1	0.00
PM10	Annual Mean	traffic	142	-0.88	[-1.84;-0.19]	-44.4	0.00
PM10	Annual Mean	industrial	80	-0.81	[-1.75;-0.07]	-42.5	0.00
PM10	Annual Median	urban	186	-0.63	[-1.30;-0.12]	-42.5	0.00
PM10	Annual Median	suburban	90	-0.63	[-1.17;-0.21]	-45.8	0.00
PM10	Annual Median	rural	89	-0.35	[-0.86;-0.05]	-33.7	0.00
PM10	Annual Median	traffic	132	-0.82	[-1.72;-0.15]	-47.0	0.00
PM10	Annual Median	industrial	68	-0.70	[-1.60;-0.12]	-41.6	0.00
PM10	Nday > 50ug/m3	urban	186	-1.64	[-5.07;-0.17]	-87.1	0.00
PM10	Nday > 50ug/m3	suburban	90	-1.36	[-4.71;-0.13]	-80.0	0.00
PM10	Nday > 50ug/m3	rural	89	-0.50	[-2.14;0.19]	-78.0	0.00
PM10	Nday > 50ug/m3	traffic	132	-2.75	[-7.83;-0.03]	-90.3	0.00
PM10	Nday > 50ug/m3	industrial	68	-2.33	[-7.28;0.00]	-85.8	0.00
PM25	Annual Mean	urban	205	-0.62	[-1.22;-0.03]	-35.0	0.00
PM25	Annual Mean	suburban	55	-0.59	[-1.05;0.05]	-33.3	0.00
PM25	Annual Mean	rural	70	-0.35	[-1.19;0.02]	-30.3	0.07
PM25	Annual Mean	traffic	96	-0.40	[-1.11;0.13]	-24.3	0.01
PM25	Annual Mean	industrial	44	-0.65	[-1.25;0.23]	-45.0	0.00

Pollutant	Metric	Type	Nsta	ST Slope	5th and 95th quantiles of ST slope	% change	MK p-val
PM25	Annual Median	urban	205	-0.51	[-1.04;0.03]	-35.7	0.00
PM25	Annual Median	suburban	55	-0.58	[-0.90;-0.08]	-39.6	0.00
PM25	Annual Median	rural	70	-0.31	[-0.98;0.02]	-32.7	0.01
PM25	Annual Median	traffic	96	-0.41	[-1.02;0.11]	-29.1	0.00
PM25	Annual Median	industrial	44	-0.70	[-1.13;0.20]	-54.6	0.00

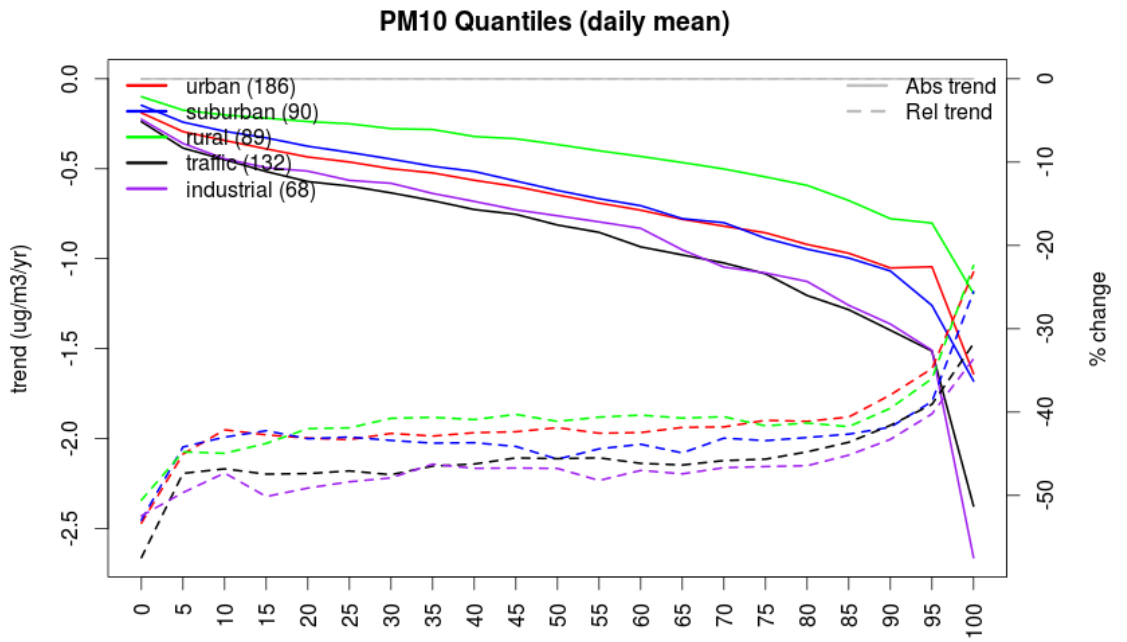
Figure 12: Same as Figure 2 for PM10 (top) and PM2.5 (bottom)





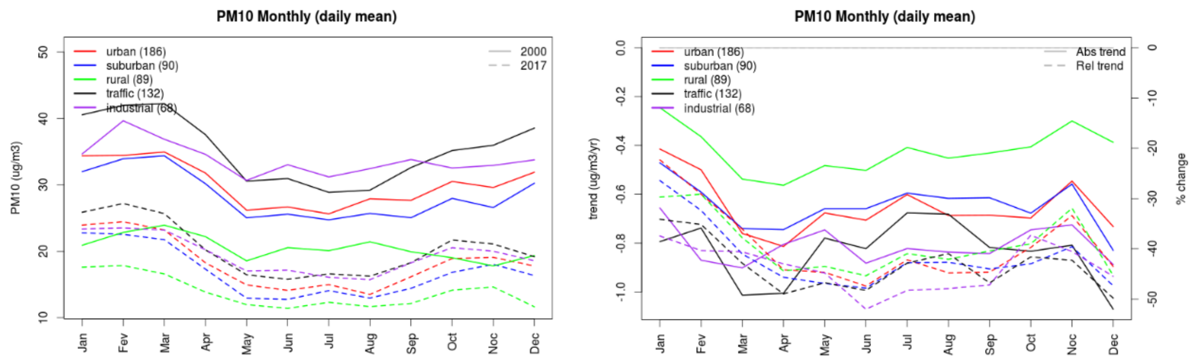
Following the reduction in annual mean PM_{10}, the number of exceedance day is dramatically reduced: 78 to 90% for the 50µg/m^3 limit value (Table 5). Yet, in relative terms, the highest PM_{10} peaks decrease much less than the rest of the distribution (Figure 13), a consequence is that the annual median of PM_{10} decreases less than the annual mean. This shows that the strategy to target long-term air pollution is bearing fruits, but now alternative approaches should also be considered for the highest peaks, unless one considers that such outstanding events are too difficult to mitigate for instance during exceptional stagnation events, or desert dust outbreaks.

Figure 13: For PM_{10} and each typology of station, absolute trends (solid lines) and relative changes (dashed lines) of the percentiles of daily means.



The monthly PM_{10} cycle show a winter maximum at urban, suburban and traffic sites both for the 2000 and 2017 estimates (Figure 14). At rural sites, the monthly cycle was marginal in 2000 but it is becoming more important in 2017, which could be due to enhanced use of wood burning. The importance of residential heating can also be seen at urban and suburban sites, where it is clearly in winter that the decreases are more limited.

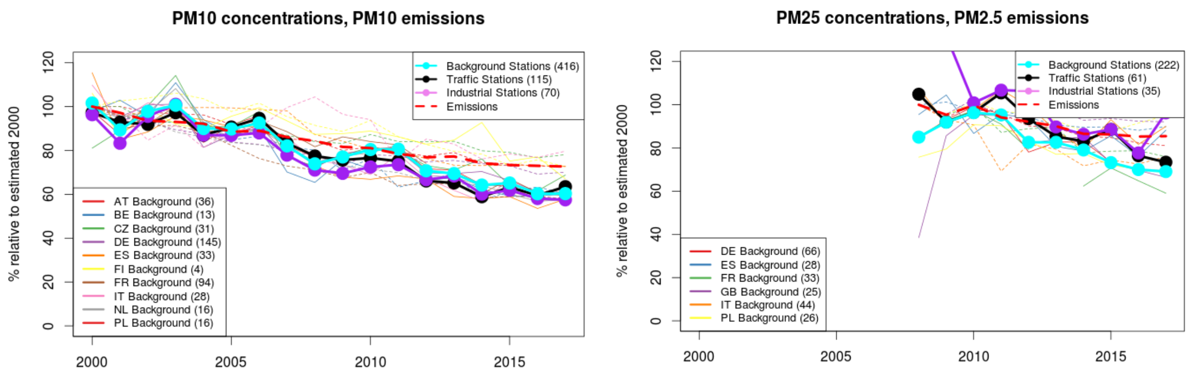
Figure 14: Left: Monthly cycle of PM_{10} at various station types estimated from the whole time series in 2000 (solid lines) and 2017 (dashed lines). Right: corresponding absolute trends (solid lines) and relative changes (dashed lines).



It is difficult to find systematic geographic patterns in the maps of PM_{10} trends in Supplementary Material (Figure S.48). The only specificity which is worth highlighting is a larger decline of PM_{10} at rural sites in the Benelux compared to Germany and Spain.

The comparison between emissions and observation by country displays a good correlation (Figure 15 and Table 2). Over countries where the network is dense enough, PM10 and PM2.5 decrease respectively by 44% and 33%, whereas the corresponding change in primary emissions is 30% and 18%. Note that we are not comparing collocated measurements here. The decreases are larger in observations, which was expected considering the importance of secondary aerosols, which are mitigated by decreasing other precursors than primary PM.

Figure 15: Same as Figure 3 for PM_{10} (left) and $PM_{2.5}$ (right) observations and primary emission. Normalised levels are not compared to the actual value in 2000 but rather the value of the linear fit for 2000 to minimise the impact of interannual variability. Those normalised levels are outlying and beyond the y-range for $PM_{2.5}$ median EU28 concentrations at industrial sites in 2008 and 2009 (131 and 132%, respectively).



3.5 Air Quality Index

The evolution of air quality in Europe is synthetized in Figure 16 using the EEA air quality index that bins air pollutant ($PM_{2.5}$, PM_{10}, NO_2, O_3 and SO_2) concentrations in 5 categories: good, fair, moderate, poor and very poor. For a given time and location, the category is computed for each pollutant, and the index takes worst category of all 5 pollutants. Because of the non-colocalization of various pollutants in the monitoring network, and its substantial development since 2000, it is difficult to compute the index over a long time period for each station. To cope with this limitation, we proposed to compute a median index by country (see Supplementary Figures S.65 to S.87) and for the 28 countries of the European Union (Figure 16), which also allow to use all the monitoring stations and not only those that comply with the completeness criteria.

A clear improvement is found for the overall index. In the early 2000, less than half of the days were "fair" and no "good" days could be detected. On the contrary in 2017 a few days are classified as "good", and about 80% of the days are either "good" or "fair". It is the "moderate" category that was reduced substantially, while in fact the days with "poor" air quality remained quite constant.

Figure 16 also show the long-term evolution of the good/fair/moderate/poor/very poor categories by pollutant over Europe. The number of "good" days for NO_2 and PM_{10} has increased clearly. For PM_{10}, the number of "poor" days have been reduced. But for ozone, the distribution has not changed significantly, and the daily categories for SO_2 were already good at the beginning of the period. Unfortunately, due to thresholding effect and continent-wide aggregation, we cannot attribute the reduction of "moderate" days for the integrated index to a given pollutant.

We should recall that the long-term evolution of the network presented in Figure 1 does not suggest that there could be significant sampling biases related to a change in station typology. The number of stations has increased dramatically but the relative proportion of urban/rural sites remained quite constant. An important effect could be due to the development of PM monitoring that would rather be unfavourable to the air quality trend. So that the improvement we notice in Figure 16 would rather be on the conservative side. The main artefact that could bear upon this time series lie in the number of countries reporting to EEA that has evolved in time. The evolution of AQ index by country is provided in the Supplementary Material but the only way to make a more robust synthesis at the European scale would be to involve models and data fusion to fill the gaps of the monitoring network.

Figure 16: For the whole Europe Union: overall air quality index (percentage of days in a given year) and distribution of daily categories per pollutant (light blue: good, light green: fair, yellow: moderate, orange: poor, red: very poor, see Table 1).

4 Conclusions

With the widespread development of air quality monitoring since 2000, observed records are now becoming long enough to assess trends in a robust statistical manner. This development also allows to cover large areas of Europe, whereas previous assessment relying on the limited records available since the 1990s, where heavily biased towards the few areas where early monitoring was undertaken.

We rely on measurements extracted from two air quality databases hosted by the European Environment Agency: Airbase (2000-2012) and Air Quality e-reporting (2013-2017). For SO_2, NO_2, ozone, PM_{10} and $PM_{2.5}$, we reach more than 10,000 stations, amongst which 3,500 comply with our requirements in terms of completeness and representativeness for long-term assessment.

Important reductions of air pollution are found for all compounds for which we assess the overall trend over Europe, but also the geographical variability of the trend, and the various indicators of their statistical distribution (percentiles, seasonal cycles, exposure indicators etc.). In the case where a given air pollutant can be directly related to a primary emission, we also compare the observed trend to the reported emission change.

The reduction of ambient SO_2 can reach 70 to 85%, which is in-line with the reported emission decrease, except for a clear mismatch in the aftermath of the 2008 economic crisis. A similar mismatch is also found for NO_2, where an important discrepancy is found between a 53% reduction in reported emission changes, whereas the actual observed NO_2 change is only 30%.

For ozone peaks, a 10% decrease is found so that the number of days of exceedance of the European long-term objective is reduced by 30 to 50%. Annual mean ozone increases however, in line with the lower NOx titration effect. But when considering the sum of NO_2 and O_3 (as Ox), decreases are found.

Particulate matter reductions are similar for $PM_{2.5}$ and PM_{10} (when identical monitoring networks are considered) and of the order of 25% to 45%, depending on station typology. This reduction is slightly larger than the reduction in primary PM emissions, thanks to the additional effect of reducing other PM precursors.

The long-term evolution of air quality index over Europe displays a clear improvement, with most days earlier classified as "moderate" are now "good", but it is worth noting that the relative proportion of days classified as "poor" is quite constant, which can also be related to the lower relative reduction of high PM episodes compared to moderate levels.

With this analysis, we tried to extract as much information as we could from the investigation of in-situ air pollution observations, only put in perspective with reported emission changes. We face however important limitations in terms of spatial coverage and representativeness because of the un-even coverage of the in-situ network. This limitation could be addressed by involving modelled results, possibly corrected with observations by means of data fusion. It would in turn allow the quantification of long-term evolution of air quality impacts on human health or ecosystems. The other limitation of the study is related to the comparison of observed air concentration with reported emissions, that ignores the transport and transformation of atmospheric trace species, which would also be covered by involving chemistry-transport models.

5 References

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