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Long-term trends of air pollutants at European and national level 2005-2021

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Summary

Context and methods

In this report we investigate the development of European air pollutant concentrations during a 17-years period (2005-2021). Surface concentrations of SO_2, NO_2, PM_{10}, $PM_{2.5}$ and O_3 at different typologies of air quality monitoring stations (rural, suburban, and urban background sites, as well as traffic and industrial sites) are analysed.

We first perform various descriptive statistical analyses to infer the main features of the evolution of air quality. Then we compare the trends in air pollutant concentrations to the trends in reported emissions at country level. This allows putting in perspective the efforts in mitigating air pollution with the resulting benefit for ambient air quality. We also conduct an unsupervised machine learning classification to identify the group of stations at European or country level that exhibit a specific temporal evolution. This last part of the analysis opens the way for further interpretation regarding the efficiency of mitigation measures, which will be conducted in a follow-up study, in which those group of stations displaying outstanding trends will be related to local air quality plans.

Between 2005 and 2021, the air quality monitoring has been extended significantly over Europe. In 2021 about 9000 records (combining all pollutants and stations) were available. After applying temporal completeness criteria in order to ensure the consistency of the long-term trends, about 5000 time series are available for the trend analysis between 2005 and 2021 for the following air pollutants: SO_2, NO_2, PM_{10}, $PM_{2.5}$ and O_3. The body of the report is devoted to the time period 2005-2021. But all the results are also provided in for the 2000-2021 period in the supplementary material.

Descriptive analysis of the observed trends

Significant downward trends are observed for SO_2, NO_2, PM_{10}, $PM_{2.5}$.

The largest decrease is seen for sulphur dioxide for which the levels at all other type of sites (background urban and suburban, and industrial and traffic) in 2021 are comparable to the concentrations which were observed at rural sites at the beginning of the period (2005). A reduction of the order of 62 % to 68 % is found.

For NO_2 the surface concentrations have dropped between 34 % and 47 % on average depending on the station typologies. The decreases are widespread in Europe, but slightly lower in magnitude in Germany, Poland and Austria.

The analysis of the evolution of NO_2 concentrations by day of the week, hour of the day, and also the distribution of all days in a given year allows inferring further important features in the change that occurred for NO_2 air pollution in Europe. We find that the concentrations observed during weekend in 2021 are comparable to those that were classically seen during weekdays in 2005. The sharp diurnal cycle (with two peaks during morning and evening rush hours) is still marked in 2021 as the low levels were more efficiently reduced that the rush hour peaks. The distribution of trends for all days in a given year shows that the low and medium levels were also more efficiently reduced that the highest peaks (except at rural sites).

O_3 is not directly emitted into the atmosphere, but is formed in the atmosphere from precursors as NO_x, VOC, CO and methane which, added to a hemispheric baseline level results in the final concentrations. During the period 2005-2021, at background sites the annual mean ozone concentration has slightly increased, between 2 % and 11 %, while the high peaks have been reduced (by 6 % to 9 %). The increase in annual means, especially noticeable at urban and suburban sites is explained by a reduced titration by NO, because of reduced NO_x levels in the atmosphere. As a result

of these opposite trends between annual average NO_2 and O_3, the sum of the two (Ox), is reduced by 1 % to 23 % for background sites, but increased by 17 % at traffic sites.

The increase of annual average ozone concentrations is markedly more pronounced over the North-Western part of Europe, most densely covered by observations (Northern France, Benelux, Germany), while the trends are often not significant elsewhere. Inversely, the downward trends for ozone peaks are more pronounced in Spain, Portugal, South France Italy and Austria.

PM_{10} concentrations decreased between 39 % and 44 % between 2005 and 2021. The density of the network for $PM_{2.5}$ only allows assessing the trends between 2008 and 2021 and we find declines ranging from 37 % to 52 %. Decreases are observed throughout Europe, but they are lower in magnitude in Germany, Austria and Finland (for both PM_{10} and $PM_{2.5}$) and in Spain (only for $PM_{2.5}$), while some non-significant increases are seen for PM_{10} in Poland.

When comparing only the stations where both $PM_{2.5}$ and PM_{10} are measured, and the same period (2008 to 2021), we find decreasing trends of 37 % and 32 %, respectively. The lower trends for PM_{10} could be induced by a compensation due to natural sources of coarse particulate matter, or to a more efficient reduction of secondary PM, which dominate in the fine fraction.

The number of days with the highest peaks was mitigated less efficiently than the average values, highlighting the importance of high air pollution episodes to the total concentrations. The PM_{10} concentrations were more reduced in winter compared to summer, also because of the higher natural and biogenic contributions in summer months.

A composite index adapted from the air pollutant categories in the EEA Air Quality Index was designed. The number of days categorised as “good” for NO_2, PM_{10} and $PM_{2.5}$ has clearly increased, but for ozone, the distribution has not changed substantially. Considering all pollutants together, and Europe as a whole, it appears that in 2005, only half of the days were considered as having a “fair” air quality, while this proportion has increased to about 70 % in 2021.

Drivers of air quality trends

To put in perspective the efforts devoted to mitigate air pollution with the resulting benefit for ambient air concentrations, we compare the reported emissions at country level with the evolution of concentrations at background sites. This is only done for the countries with a sufficient number of monitoring stations, so that the average trends reported in this part may differ slightly from the descriptive statistics sections. We also note that this comparison is limited as most pollutants are also subject to transport and transformation of related precursors species and a more detailed comparison would benefit from the involvement of Chemistry-Transport model calculations.

For SO_2, reported emissions declined by 76 %, while measured concentrations were reduced by 61 %. There was a relatively good agreement between those trends up to the year 2008, after which a larger mismatch is seen. There are also some differences depending on the countries: whereas the observed rate of decline in SO_2 observations is lower than the decline of emissions in Czechia, Bulgaria, Italy, and Hungary, the opposite is found for Austria, Belgium, Germany, and Poland.

For NO_2 we also find that a larger mismatch occurs after the year 2008. Overall, if reported emissions declined by 55 %, the ambient air concentration decreased only by 38 %. There are notable differences between countries such as Germany, Hungary, Sweden and Poland where the decreasing trend in emission was reported to range from 30 % to 42 %, and the other countries where trends in emissions are rather decreasing between 48 and 60 %. It would be worthwhile to find out if such different trends are due to actual changes in the vehicle fleets, or rather to artifact related to the way in which the reported emissions are computed.

The decreasing trends in the emissions of the main ozone precursors are 33 % and 56 % for NMVOC and NOx, respectively. At the same time, annual mean ozone increased by 3 % and ozone peaks decreased by 8 %. The increase of annual mean ozone is due to the titration effect in relation with the NO_2 decline. The limited decrease in ozone peaks is due to the fact that the reduction in anthropogenic precursors only reduces the ozone peak increment above a non-mitigable natural and hemispheric burden of ozone. It would be insightful to quantify more precisely this burden in order to assess the efficiency of ozone mitigation strategies.

The surface PM concentrations show stronger downward trends than the reported emissions. This is likely an effect of the importance of secondary aerosols which are mitigated on top of the direct PM emissions, thus leading to additional reductions compared to the abatement of primary emissions alone. For PM_{10} and $PM_{2.5}$ we find overall reductions in surface concentrations of the order of 42 % and 45 %, respectively. The reduction of the direct emissions of primary PM_{10} is 36 %, and for PM2.5 it is 41 %. The concentration reduction for $PM_{2.5}$ is also slightly larger than the rate of emission reduction of primary $PM_{2.5}$.

An unsupervised machine learning clustering is applied to produce objective clusters of stations with analogous features with regards to air pollutants concentrations in 2005, relative trends, and relative changes in emissions. When applied to the whole of Europe, this analysis confirms the above statements regarding the consistency between concentrations and emissions.

Applying the clustering on a country basis allows identifying group of stations, within a given country, exhibiting specific features. In the report we present a few examples of this analysis for a few selected countries and pollutants as well as station typologies. The results of the clustering applied systematically for all countries/pollutants/station typologies are presented in Annexes.

For background NO_2 stations in Spain, we identify two groups of stations distributed evenly over the country, and with similar concentrations in 2005, but in one of the groups the concentrations decreased by 60 % while in the second group the decrease is only 20 %. Such sharp differences in the temporal trends will be further investigated in a follow-up study to infer whether some of those stations are located in air quality zones undergoing specific action plans.

Similar outstanding behaviour are provided for NO_2 traffic stations in Spain, PM_{10} background stations in Poland, PM_{10} traffic sites in Germany, and PM_{10} industrial sites in France. Note that in this main text, the later specific countries or station typologies were chosen to be as illustrative as possible of the various possible situations to propose a simple method for identifying outstanding stations. As already said, the result of the clustering for all pollutants and all countries are also delivered in supplementary material to support the future analysis of air quality plans.

1 Introduction

It is well documented that air pollution poses a serious threat for human health and ecosystems. It has been mitigated since the 1970s, in particular through international policy instruments such as the Geneva Convention on the Long Range Transboundary Air Pollution or Air Convention (CLRTAP 1979) and, as far as regulations within the Europe Union are concerned, the National Emission reduction Commitments Directive (EC 2016), which set objectives to be achieved by the implementation of national and local regulations. In order to assess the magnitude of the threat of air pollution, 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 set a number of air quality directives (EC 1996, EC 2004, EC 2008) defining common monitoring principles for countries, as well as maximum air pollution levels not to be exceeded to ensure a clean air for European population and environment.

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 EEA/ETC and the CLRTAP to assess the efficiency of air pollution mitigation strategies (Colette et al. 2016a, Maas and Grennfelt 2016, EEA 2020, Colette et al. 2016b). The topic has also been of interest for the scientific community with several articles devoted to the assessment of air quality trends and their relationships with efforts achieved in terms of emission reductions. The majority of such studies were focused on ozone (Cooper et al. 2020, Lefohn et al. 2016, 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 have also been studies investigating nitrogen and particulate matter trends: (Colette et al. 2011, Guerreiro, Foltescu and de Leeuw 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, Colette et al. 2011, Wilson et al. 2012).

In this report the trends are studied by analysing the observations through linear statistics to update the knowledge of the current status of trends in the European air quality. The results are presented based on aggregated European data in the main report, and for each individual country in the annexes. The method relates the trends in air pollutant concentrations to the reported emission changes over the 2005-2021 period to the extent possible. Additionally, a clustering method was applied to identify stations, urban areas, regions and/or countries with outstanding trends in concentrations and emissions. The later analysis will be a support to identify good practices in improving air quality.

In this study, a 17-year period (2005-2021) is studied and presented in the main text, although the 2000-2021 period is also investigated and the results presented as annexes only. Compared to earlier studies looking at shorter periods a 17-year period makes it easier to conclude on statistical significance of the trends. In addition, thanks to the development of the monitoring in time, there are more stations available in recent years and therefore a better spatial coverage for the analysis.

The methods and their respective input data are described in Chapter 2, and the results in Chapters 3 and 4 where the trend and stations' clustering 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. A summary is given in Section 5 5.

2 Models and methods: Analyses of 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. After 2013, the EEA database moved to the Air Quality e-reporting system. Only validated data are used. 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 by the EEA using the Sampling Point Identification, which is the most reliable meta-data about the consistency of a given record.

The EEA databases differentiate sampling point area (urban, suburban and rural) and station type (background, traffic, industrial). For synthesis, we differentiate background types at urban, suburban and rural areas and also considered separately traffic stations and industrial stations (yet without distinction of whether traffic or industrial stations are located in urban, suburban or rural areas). In other words, in the present report, only background types are considered for urban, rural and suburban areas (for some figures we explicitly stated background-urban (BG-UR), background-suburban (BG-SUB) and background-rural (BG-RU)). Those categories are referred to as “typologies” in the present report.

2.1 Data completeness

All the surface data available included in the EEA databases are 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 statistics 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 17-year time period (i.e., 5 years or more) were not available.

Reducing data after a data completeness test can improve data quality but may also come with trade-offs such as reduced sample size, as can be seen when comparing Total and Selected Stations in Figure 2.1. However, the data selection criteria adopted above is meant to preserve a substantial sample size and minimize potential biases regarding the trend analysis.

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 based on daily means. Consequently, we checked for redundancy if the corresponding record was already provided as daily mean before aggregating the available raw hourly records into daily means This aggregation did not include a verification of missing data over a given day, so that the re-computed daily means were computed irrespective of the number of available records for that day. We note that this is slightly different than in the official statistics usually produced by EEA, where 75 % completion criteria are applied before computing a daily mean value. Given the amount of data used in the present assessment, we considered that this discrepancy would have minor impacts. 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 led 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.

Until 2007, French authorities reported PM hourly concentrations from automatic devices (TEOM, Beta gages) without applying any correction factor to account for the volatilisation of some PM compound during the measurement phase. For that reason, 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 2005-2021 in the European Union (27 countries) is given in Figure 2.1. In 2005, about 5000 records (combinations of stations and pollutants) were available, but in 2021 this number exceeds 9000 records (in 2019 the number has even exceeded 10000 records). A steady increase of the number of stations is found for many station types and pollutants. An exception is noted for PM_{10} and SO_2 where the number of stations has decreased slightly in the years 2020/2021. The same exception applies to urban, suburban and rural station types where the number of stations decline slightly in 2020/2021. Over the early 2000's, the PM monitoring network has developed drastically, so that the proportion of gaseous monitoring devices is reduced. For example, there was a noticeable increase in $PM_{2.5}$ monitoring up to 2011.

Before 2007, $PM_{2.5}$ stations locations were so scarce that after applying the completion criteria they can barely be seen in the histogram in the middle of the first row of Figure 2.1. This is why, only for $PM_{2.5}$ the time window for the trend assessment will be reduced to 2008-2021.

After having applied the completeness checks for trend assessment described above, we kept about 5000 records. If only stations covering the whole period had been selected, the number of stations would have been constant in time, instead we see an increase in the number of sites over the first few years due to the improvement of temporal coverage and the presence of more sampling points. There is also the effect of the relaxed completeness criteria that selects records with only 75 % of valid years (i.e., over 2005-2021 for all pollutants except for $PM_{2.5}$ where the time period is 2008-2021 due to the completeness check presented in Table 2.1). In the previous Eionet report 2021/9 (Solberg 2022) a clear issue appeared regarding the number of stations selected in 2013, the year when the EEA system changed from Airbase to AQ e-reporting. In this report, this anomaly has been partly corrected, mostly with the update of French submissions to AQ e-reporting. The anomaly remains slightly visible, especially in the selected stations per pollutant (Figure 2.1). This lower number of available sites in 2013 can be due either to a lower number of reported data, or a change in sampling point identifiers (which would impair the matching of several records with the reminder of the period, making them irrelevant for trend assessment). Nevertheless, the number of reported stations has improved since the previous assessment (Solberg 2022). The vast majority of selected stations passing completeness criteria are located in the 27 countries of the European Union (Table 2.1). But we also captured a few additional stations located mainly in Norway and Switzerland, as well as Iceland, and North Macedonia. These other countries are not discussed in the main body of the report, but they are included in the supplemental material on trends analysis by country.

Apart from this anomaly of 2013 there is no systematic trend in the distribution of station type after 2005. The number of PM monitoring sites increased gradually, so that the proportion of ozone and NO_2 monitoring stations is lower.

Figure 2.1: Number of air quality monitoring station by pollutant (top) and station type (bottom), in the 27 countries of the European Union (EU27) available over the 2005-2021 time period (left) and passing the completeness criteria for trend assessment in absolute (middle, please be aware of the different Y-axes values) and relative (right) numbers

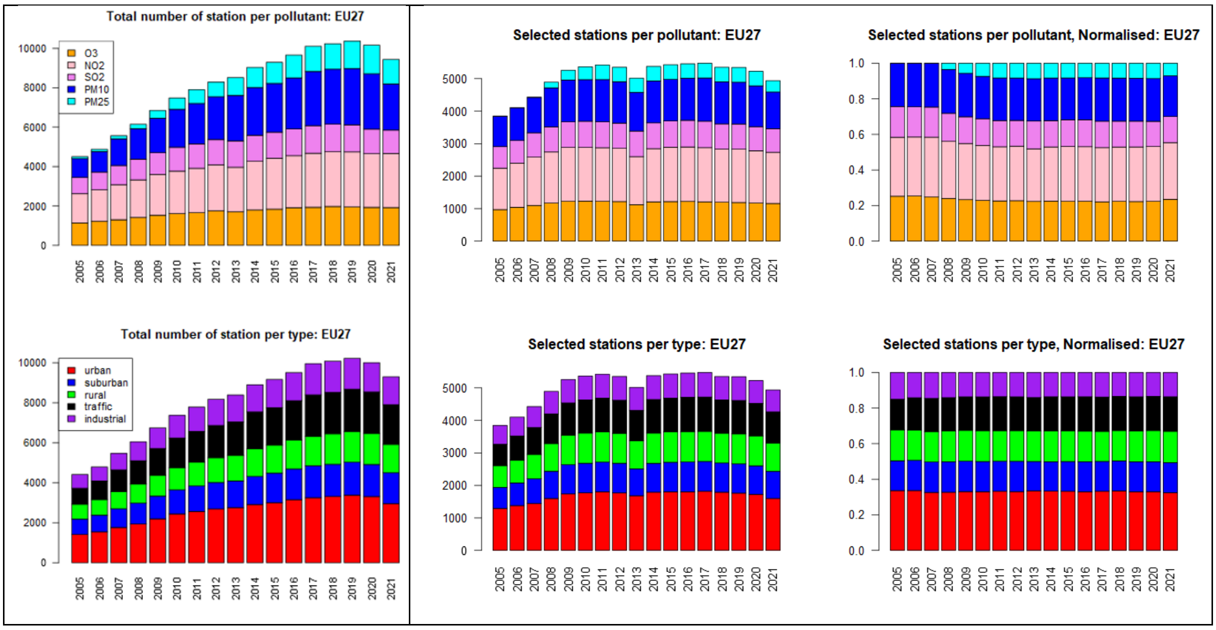


Table 2.1: Number of stations per country and typology passing completeness criteria and therefore included in the present trend assessment for each pollutant. The horizontal line separates the EU27 countries (above the line) from few other European countries (below the line)

Pollutant	SO2	SO2	SO2	SO2	SO2	NO2	NO2	NO2	NO2	NO2	O3	O3	O3	O3	O3	PM10	PM10	PM10	PM10	PM10	PM25	PM25	PM25	PM25	PM25
Metric	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean	Annual Mean
Station Type	urban	suburban	rural	traffic	industrial	urban	suburban	rural	traffic	industrial	urban	suburban	rural	traffic	industrial	urban	suburban	rural	traffic	industrial	urban	suburban	rural	traffic	industrial
AT	9	15	21	0	19	19	41	26	35	7	11	33	41	4	1	19	41	20	16	7	6	4	2	2	0
BE	3	5	9	2	13	6	11	17	2	19	4	8	18	1	5	6	11	13	1	15	7	7	8	0	6
BG	12	0	1	2	2	9	0	1	2	0	10	0	1	2	1	20	2	1	2	2	3	0	1	0	0
CY	0	0	1	1	0	0	0	1	1	0	0	0	1	1	0	0	0	0	0	0	1	2	1	0	0
CZ	12	7	6	2	1	14	11	6	14	1	15	9	16	3	0	27	19	5	12	1	12	7	2	3	1
DE	37	15	31	9	11	90	59	52	98	19	79	59	58	5	12	88	59	49	92	21	45	15	14	31	7
DK	0	0	0	1	0	3	0	2	3	0	3	0	2	1	0	0	0	0	1	0	0	0	0	1	0
EE	3	0	2	1	2	3	0	2	1	2	3	0	3	1	2	0	0	1	1	2	3	0	3	0	1
ES	44	25	32	61	104	54	42	36	89	112	43	51	52	50	74	24	17	20	44	54	6	12	13	14	23
FI	0	0	4	0	4	3	1	4	10	2	2	0	7	0	0	2	2	0	14	0	4	2	1	5	0
FR	28	1	3	1	61	148	46	8	44	20	119	77	31	1	0	126	27	5	31	31	45	2	6	5	0
GR	0	0	0	1	0	0	1	0	1	1	0	1	0	1	1	0	0	0	0	0	0	0	0	0	0
HR	1	0	0	1	0	2	0	0	2	0	1	0	0	0	0	0	0	0	2	0	0	0	0	0	0
HU	5	7	1	7	1	6	7	1	7	1	6	7	2	1	1	6	6	2	7	1	0	0	0	1	0
IE	2	0	1	0	0	1	0	1	2	0	1	1	3	0	0	1	3	1	1	0	0	1	0	0	0
IT	27	10	5	21	20	89	39	43	100	24	68	30	41	3	3	58	28	20	61	14	32	7	15	13	1
LT	3	0	0	2	2	3	0	0	4	4	2	0	4	4	2	5	0	0	5	4	0	0	0	3	0
LU	2	0	1	0	0	1	0	2	0	0	2	0	3	0	0	0	1	0	0	0	1	0	0	0	0
LV	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
MT	0	0	1	1	0	0	0	1	0	0	1	0	1	0	0	0	0	0	0	0	0	0	0	0	0
NL	0	1	4	0	0	10	3	18	15	2	3	3	17	4	0	2	3	15	11	0	0	0	0	0	0
PL	42	2	10	1	3	39	3	11	5	2	23	2	15	0	0	48	3	4	4	2	25	0	1	4	1
PT	3	2	1	2	2	10	1	4	5	4	14	4	8	0	2	9	2	5	6	2	3	0	4	1	0
RO	0	0	0	1	0	2	0	0	0	1	1	0	1	0	1	0	0	0	0	1	0	0	0	0	0
SE	2	0	0	0	0	5	0	1	6	0	5	0	8	1	0	3	0	2	9	0	2	0	3	5	0
SI	3	1	0	1	0	3	1	1	1	0	5	1	3	1	0	4	2	1	1	0	1	0	1	0	0
SK	4	1	0	2	0	2	1	0	4	0	4	2	5	0	0	11	2	1	5	1	1	0	0	1	0
NO	0	0	0	0	1	1	2	0	10	0	0	0	6	0	0	4	2	0	11	0	2	2	0	10	0
CH	2	2	2	1	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
IS	0	0	0	1	0	0	0	0	1	0	0	0	0	0	0	0	0	0	1	0	0	0	0	0	0
MK	0	0	0	3	1	0	0	0	0	0	0	0	0	2	2	0	0	0	0	0	0	0	0	0	0

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 also computed indicators 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 maximum daily eight-hour mean was used to compute the number of days above 120µg/m^3 (long-term objective). The 180 µg/m^3 (information threshold) and 240 µg/m^3 (alert threshold) are computed based on hourly values. 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. The summertime mean of daily maximum ozone is sometimes used as an indicator of ozone peaks, but it should be noted that in many locations in Europe most summer days are not characterized by high ozone levels. In turn, the summertime average is also largely influenced by moderate ozone levels. That is why we use the 4th highest value observed in the summer to provide an indication of the highest peaks (Colette et al. 2016a). We also computed health-related metrics: SOMO35 and SOMO10 (sum of ozone daily maxima in excess of 35 ppbv([a]) and 10 ppbv, (Malley et al. 2015)) and ecosystem-related metrics: AOT40c and AOT40f (accumulated ozone over 40 ppbv between May and July for crops and between April and September 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.

The trends can become complicated to synthetise, with various degrees of reductions for the different pollutants, and even opposite trends depending on the metrics. At the same time, the reader can legitimately call for a synthetic diagnostic illustrating whether the Air Quality situation improves overall (irrespective of the pollutants). This is why we also provide some trend diagnoses inspired from the concentration intervals used in the EEA Air Quality Index (EEA 2021). We computed air quality indices by country for all air pollutants, using the categories defined by EEA and recalled in Table 2.2. The index defines concentration intervals for each air pollutant, and is subsequently categorized for every station as the worst level across available air pollutant observations at that 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 make the calculation easier, we rather compute the index level in a country for all pollutants and at each station, and then we take the median by pollutant for all stations. The country air quality index is then defined here as the worst category for all pollutants.

([a]) Equivalent to 70µg/m^3, using the equivalence 1 ppbv O_3 = 2µg/m^3, applicable to all the other O_3 concentrations in the report.

Table 2.2: Categories of the EEA Air Quality Index (Source: EEA, airindex.eea.europa.eu)

	Good	Fair	Moderate	Poor	Very poor	Extremely poor
Particles less than 2.5 µm ($PM_{2.5}$)	0-10	10-20	20-25	25-50	50-75	75-800
Particles less than 10 µm (PM_{10})	0-20	20-40	40-50	50-100	100-150	150-1200
Nitrogen dioxide (NO_2)	0-40	40-90	90-120	120-230	230-340	340-1000
Ozone (O_3)	0-50	50-100	100-130	130-240	240-380	380-800
Sulphur dioxide (SO_2)	0-100	100-200	200-350	350-500	500-750	750-1250

2.3 Statistical tests

The statistical method applied for the trend detection is Mann-Kendall (with a p-value of 0.05) (Mann 1945, Kendall 1975) and we compute the actual slope using the Sen-Theil approach (Sen 1968). 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 infer 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 2005, which minimises the effect of interannual variability compared to directly using the value for the year 2005. Those estimated 2005 levels are used for normalisation of both observed concentrations and emissions in the timeseries. The same approach is used for comparing the distribution or monthly/weekly/daily cycles at the beginning and end of the period. Instead of using the actual values for the year 2005 and 2021, linear fits are used to estimate the baseline cycles for those years, reducing the impact of interannual variability.

In several figures, the distribution of relative changes is presented as boxplot that provide the inner 25th- to 75th percentiles (filled boxes), median (horizontal line) as well as the 5th and 95th percentiles (whiskers) and individual values outside of that later interval (dots). The term "relative change" refers to a measure of how much a specific value (e.g., concentration or emission) has changed over time relative to a reference value, and in this precise case the reference is the estimated value in 2005 as explained in the previous paragraph.

2.4 Air pollutant emissions

We used the National air pollutant emission inventory (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 Pollution (CLRTAP). Those were obtained (for both EU27 and non-EU27 countries) from the EMEP Centre for Emission Inventories and Projections (Emission as used in EMEP models), in the version updated in July 2023.

2.5 Methodology for the analysis of clustering

An unsupervised machine learning approach is used to produce group stations within clusters with similar trend characteristics. The classification is based on the k-means technique (Jain, Murty and Flynn 1999), applied to observed air concentrations of NO_2, PM_{10}, and $PM_{2.5}$ and related precursor emission trends over the period 2005-2021 (or 2000-2021 presented as supplementary material).

To capture the essential characteristics of each station, we selected three distinct features (i.e., predictors) when the clustering is applied on the whole Europe and two for individual countries. At the European scale, we used the following features: the estimated concentration value in 2005, the relative concentration change trend, and the relative emission change trend. The emission is reported at national scale, so that it is not relevant to include that feature for a country-based clustering. At the country level, we use therefore only the concentration features: the estimated concentration value in 2005 and the relative concentration change trend.

Our clustering methodology encompassed the following station typologies: background-urban (BG-UR), background-suburban (BG-SUB), and background-rural (BG-RU), all background (BG-UR, BG-SUB, and BG-RU), traffic and industrial. To facilitate the interpretation of the clustering, we arbitrarily set a limit to three clusters for this analysis.

Once the clustering is completed on an objective basis with the k-means approach, we rely on probability density functions (PDFs) distribution of the main features of each cluster to interpret what differentiates each cluster. The Map of stations distributed by cluster, and the distribution of station typologies in each cluster is also used in the interpretation.

3 Descriptive analysis of the trends of air pollutants of concentrations

In this Chapter, we perform various descriptive statistical analysis to assess the main features of air pollution trends in Europe. Some of these diagnostics allow pointing out possible causes for the changes in air quality, but the analysis of the main drivers will be further discussed in Section 4. Additional results for individual countries and in EU27, spanning the periods 2005-2021 and 2000-2021, are provided as supplementary material (refer to Annex 1 and 2).

3.1 Sulphur dioxide

The time series presented in Figure 3.1 display, for each typology of stations, the median of annual mean values at all available European stations, their linear fits for 2005-2013 and 2013-2021, as well as the distribution for all sites of the relative changes between the beginning and the end of the period (as boxplots). The corresponding quantitative SO_2 trends are provided in Table 3.1.

The trends of sulphur dioxide present the largest decrease of all pollutants, and they are always significant as indicated by the very small p-values. The decrease is such that at industrial sites, the levels in 2021 are comparable to that of rural sites at the beginning of the period. The relative changes in the Europe-wide composite, computed as the median of all annual mean values between 2005 and 2021, depends on station types: -62.1 % (traffic), -63.5 % (industrial), and -67.7 % (rural background).

In order to provide an indication whether the trend is steady, slowing down, or increasing in time, we also computed the same trends over the first and second halves of the period (indicated by the dashed lines in Figure 3.1). Comparing the linear fits over the first and second halves of the period (2005-2013 and 2013-2021), one can notice a slight flattening of absolute decrease in more recent years, except at suburban background stations (see Table 3.1). The slowdown in the pace of the downward trend becomes even more pronounced when tracing it back to the 1990s, although such estimates are only available for EMEP rural sites in the literature. According to the literature, the significant decrease in SO_2 at those EMEP sites mainly occurred during the 1990s, with reductions of up to 90 % (Colette et al. 2016a, Tørseth et al. 2012, Aas et al. 2019).

Figure 3.1: Time series of the European-wide composite (median) of annual mean SO_2 (µg/m³) per station typology (red: urban background, blue: suburban background, green: rural background, black: traffic, violet: industrial) between 2005 and 2021. The dashed lines show the linear fit between 2005 and 2013 and between 2013 and 2021. The boxplots on the right-hand side show the distribution of relative changes (%) between 2005 and 2021 for all stations of each typology

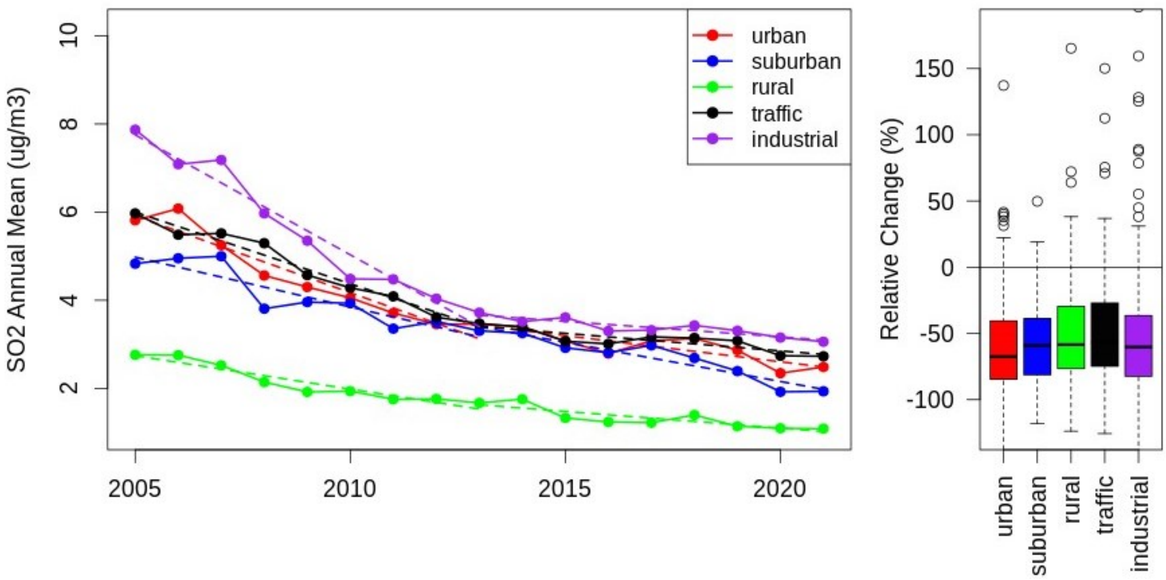
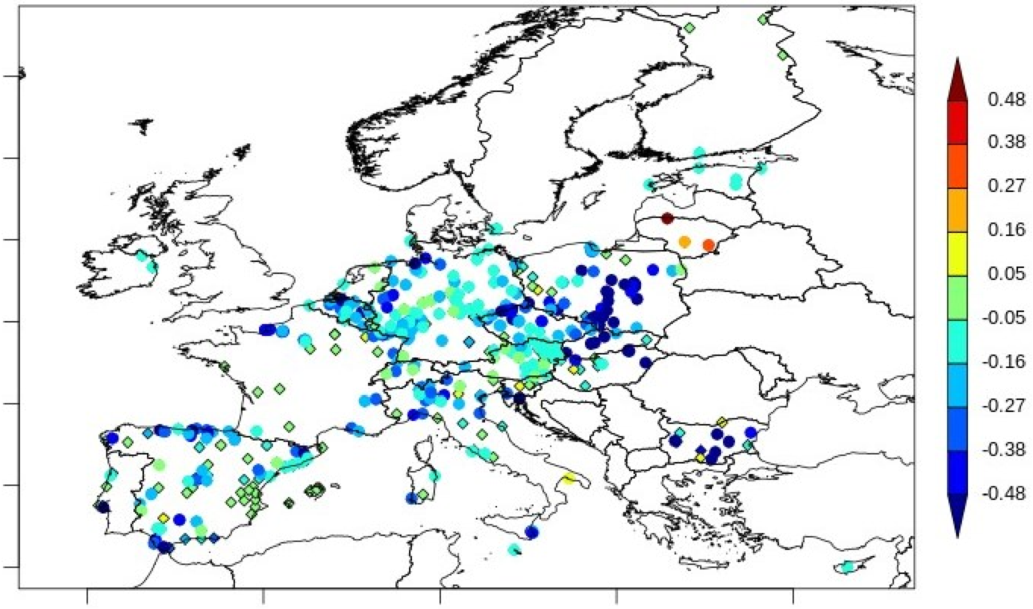


Table 3.1: Summary of observed SO_2 annual mean trends per station typology: total number of stations, trend of the median of the European-wide composite (Sen-Theil slope, µg/m³/yr), 5th and 95th quantiles of Sen Theil slopes at all European stations, significance of the European-wide (EU27) composite (Mann-Kendall p-value, MK p-val), and relative change between: 2021 and 2005, 2013 and 2005, 2021 and 2013 for the European-wide composite (% change)

Metric	Station Typology	Number of Stations	Trend of Median	5th and 95th quantile of trends	MK p-val	Relative Change (2021 vs 2005 %)	Relative Change (2013 vs 2005 %)	Relative Change (2021 vs 2013 %)
Annual Mean	urban	244	[-0.85;0.01]	-0.20	<0.01	-62.4	-47.0	-28.0
Annual Mean	suburban	94	[-0.57;0.00]	-0.18	<0.01	-62.0	-36.0	-42.0
Annual Mean	rural	136	[-0.38;0.04]	-0.10	<0.01	-67.7	-44.0	-37.0
Annual Mean	traffic	125	[-0.68;0.06]	-0.20	<0.01	-62.1	-43.0	-19.0
Annual Mean	industrial	247	[-1.02;0.07]	-0.25	<0.01	-63.5	-56.0	-15.0

The trends of the European-wide composites (median over all stations of the annual means), displayed in Figure 3.1, are all significant as indicated with the Mann-Kendall statistics (MK p-value) of Table 3.1 that are well below 0.05. There are however a few sites where the relative trend is smaller (or even positive) in the boxplots of Figure 3.1. Trends at all urban, suburban and rural background sites are plotted in Map 3.1. Those small decreases or even increases are scattered across Europe and would need to be investigated at site level. Such a level of investigation is beyond the scope of our European-wide assessment and would not change our overall conclusions. Besides these localized increases, it is difficult to point out significant spatial patterns in Map 3.1.

Map 3.1: Map of SO_2 trends (in µg/m3/yr) at all urban, suburban and rural background sites in Europe between 2005 and 2021. Significant trends are displayed as dots, and un-significant trends as diamonds



3.2 Nitrogen dioxide

The median trend of annual mean of nitrogen dioxide over Europe is displayed in Figure 3.2 as the time series of the median across all European sites. It shows a clear downward trend for all station types. The interannual variability is low. The relative changes are within -25 % to -50 % for the central interquartile part of the distribution across all stations. They are similar for all station types, except for rural and industrial stations where the median relative change is larger. For all site typologies except traffic stations, the linear fits over the beginning (2005-2013) and end (2013-2021) of the period indicated as dashed straight lines suggests that there is no strong change in the rate of decline. However, in the quantitative estimates provided in (Table 3.2), it appears that the rate of decline is stronger over the recent years (2013-2021) compared to the earliest years (2005-2013) except for the industrial stations.

Figure 3.2: Time series of the European-wide (EU27) composite (median) of annual mean NO_2 (µg/m³) per station typology (red: urban background, blue: suburban background, green: rural background, black: traffic, violet: industrial) between 2005 and 2021. The dashed lines show the linear fit between 2005 and 2013 and between 2013 and 2021. The boxplots on the right-hand side show the distribution of relative changes (%) between 2005 and 2021 for all stations of each typology

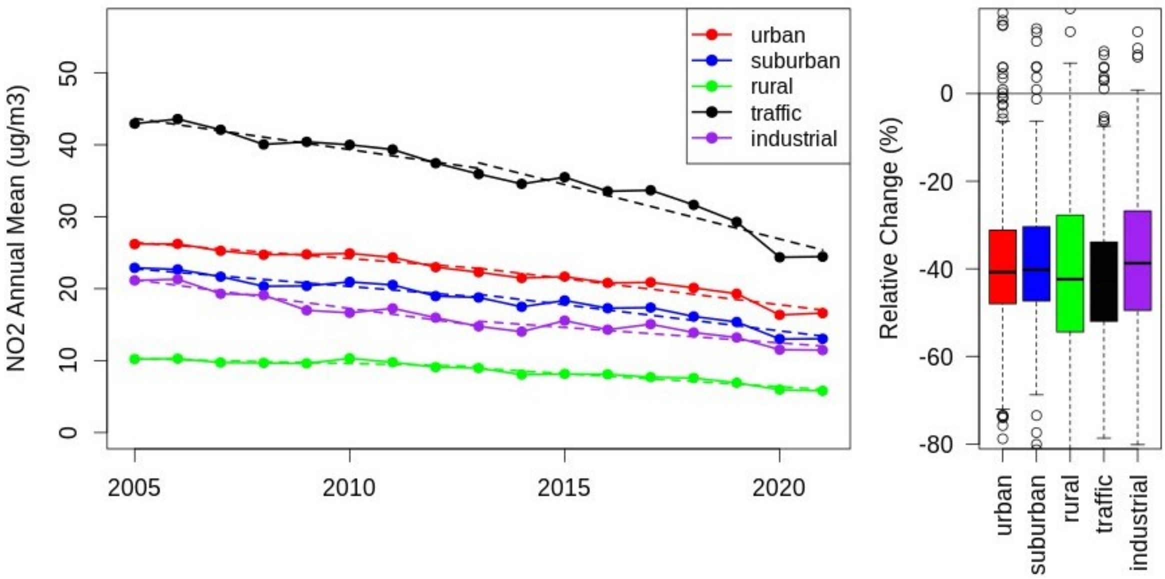
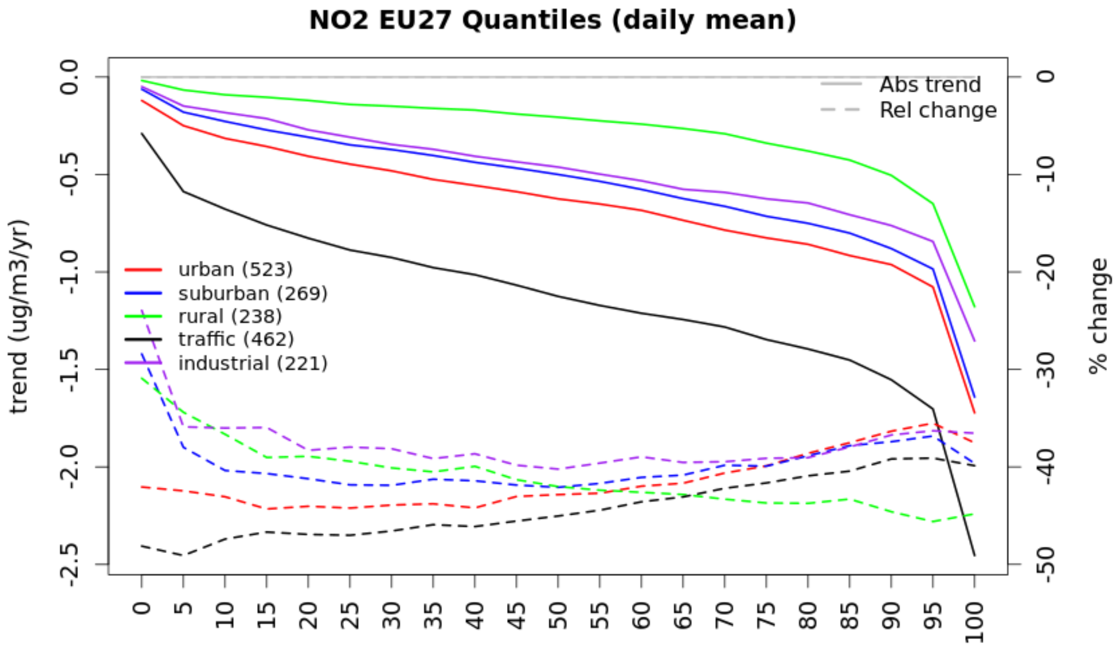


Table 3.2: Summary of observed NO_2 annual mean trends per station typology: total number of stations, 5th and 95th quantiles of Sen Theil slopes at all European stations, trend of the median of the European-wide composite (Sen-Theil slope, µg/m³/yr), significance of the European-wide composite (Mann-Kendall p-value, MK p-val) and percentage change between: 2005 and 2021, 2005 and 2013, 2013 and 2021 for the European-wide composite (% change)

Metric	Station Type	Number of Stations	5th and 95th quantile of trends	Trend of Median	MK p-val	Relative Change (2021 vs 2005 %)	Relative Change (2013 vs 2005 %)	Relative Change (2021 vs 2013 %)
Annual Mean	urban	523	[-1.33;-0.12]	-0.54	0.00	-33.9	-14.0	-25.0
Annual Mean	suburban	269	[-1.06;-0.13]	-0.56	0.00	-41.1	-17.0	-30.0
Annual Mean	rural	238	[-0.78;-0.02]	-0.26	0.00	-41.7	-10.0	-33.0
Annual Mean	traffic	462	[-2.42;-0.27]	-1.02	0.00	-38.8	-16.0	-32.0
Annual Mean	industrial	221	[-1.11;0.01]	-0.57	0.00	-47.1	-30.0	-22.0

In order to discuss the relative evolution of low and high NO_2 concentration, the absolute and relative trends of percentiles 0 to 100 are given in Figure 3.3. At each monitoring site, the percentiles distribution of daily mean NO_2 is computed every year to derive the absolute trend and relative change of each corresponding percentiles. The Figure provides the median absolute trend and relative change for each percentile by typology of station. It appears that the absolute largest declines are found for highest percentiles. On the contrary, the relative changes are larger for the lower/medium percentiles (below 40 %) for urban and traffic sites, whereas there are declines larger than 40 % for highest percentiles at rural sites. Similar patterns were observed in previous assessments, with a few differences. For instance, the earlier assessment of the 2000-2017 period (Colette and Rouïl 2020) revealed a more pronounced trend where high values were less efficiently reduced than medium levels in relative terms.

Figure 3.3: For NO_2 and each typology of station, absolute trend (solid lines, left y-axis) and relative change (dashed lines, right y-axis) of the percentiles of daily means



This larger decline of high NO_2 levels in absolute terms is also seen in diurnal cycles Figure 3.4. 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 lower panel of Figure 3.4). Likewise, Figure 3.5 shows the trends by day of the week. This weekly cycle illustrates that NO_2 levels observed in 2021 in working days are similar to those of weekends in 2005, even at traffic sites. But here, in relative terms, it is the (decreasing) weekday trends that are larger than during weekends.

There is some geographical variability in NO_2 annual mean trends (Map 3.2). In particular, a lower relative decline is found over Germany, Austria and Poland compared to other countries. If we ignore the stations where the trend is not significant (diamond sign), there are only very few and isolated stations which display an increase (one in Italy and one in Romania).

Figure 3.4: Top: diurnal cycle of NO_2 per station type estimated from the whole time series in 2005 (solid lines) and 2021 (dashed lines). Bottom: corresponding absolute (solid lines, left y-axis) and relative (dashed lines, right y-axis) trends

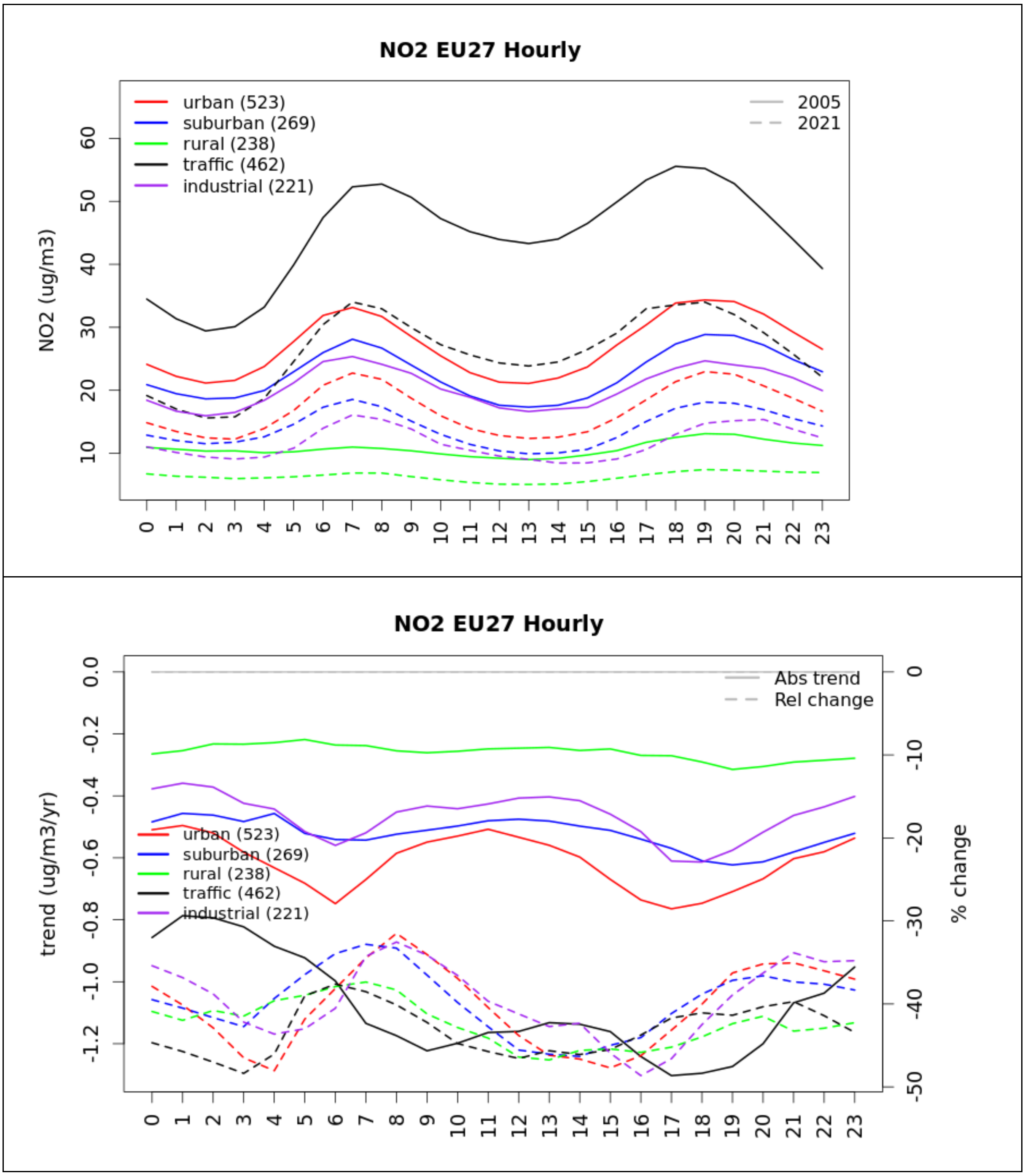
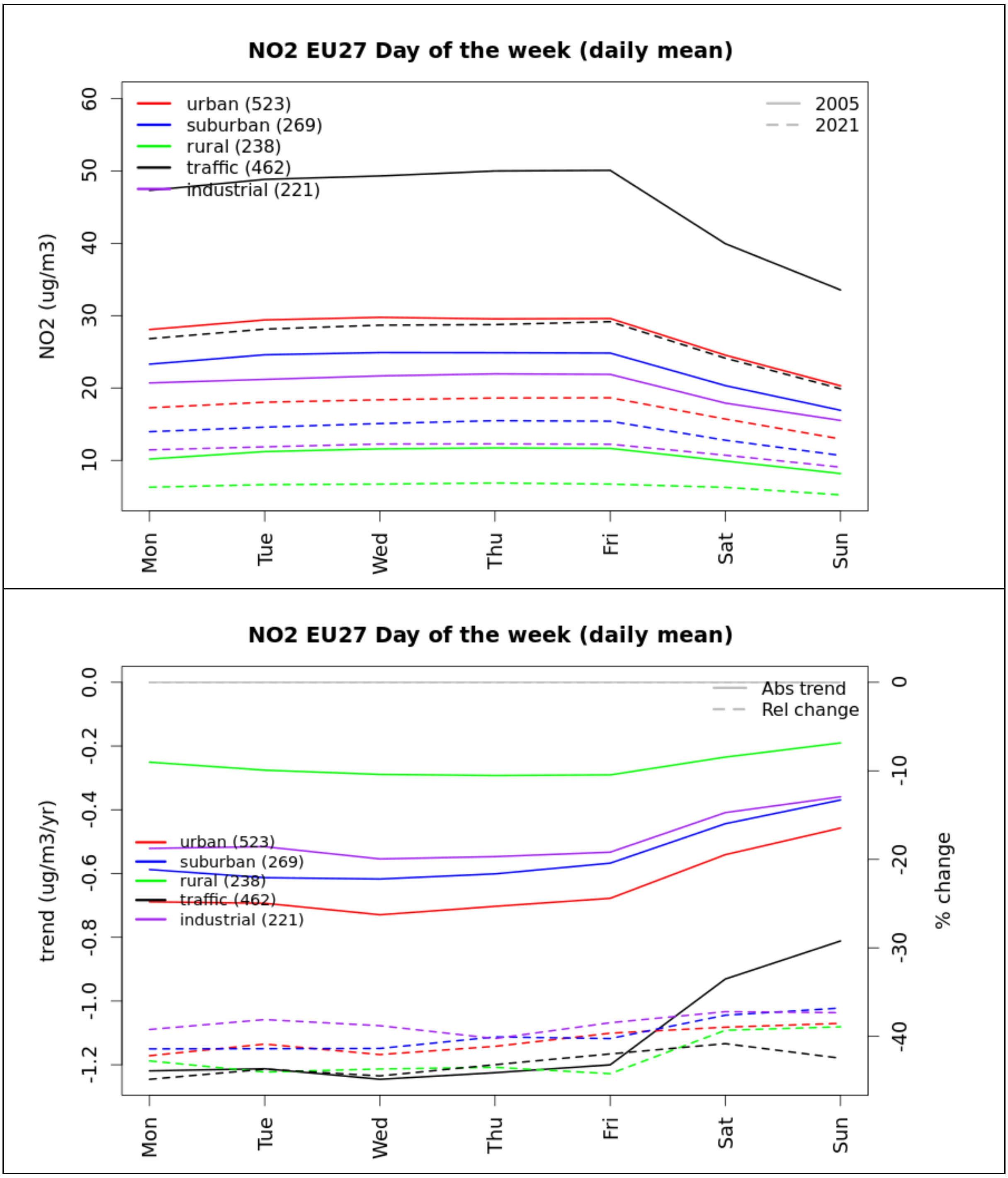
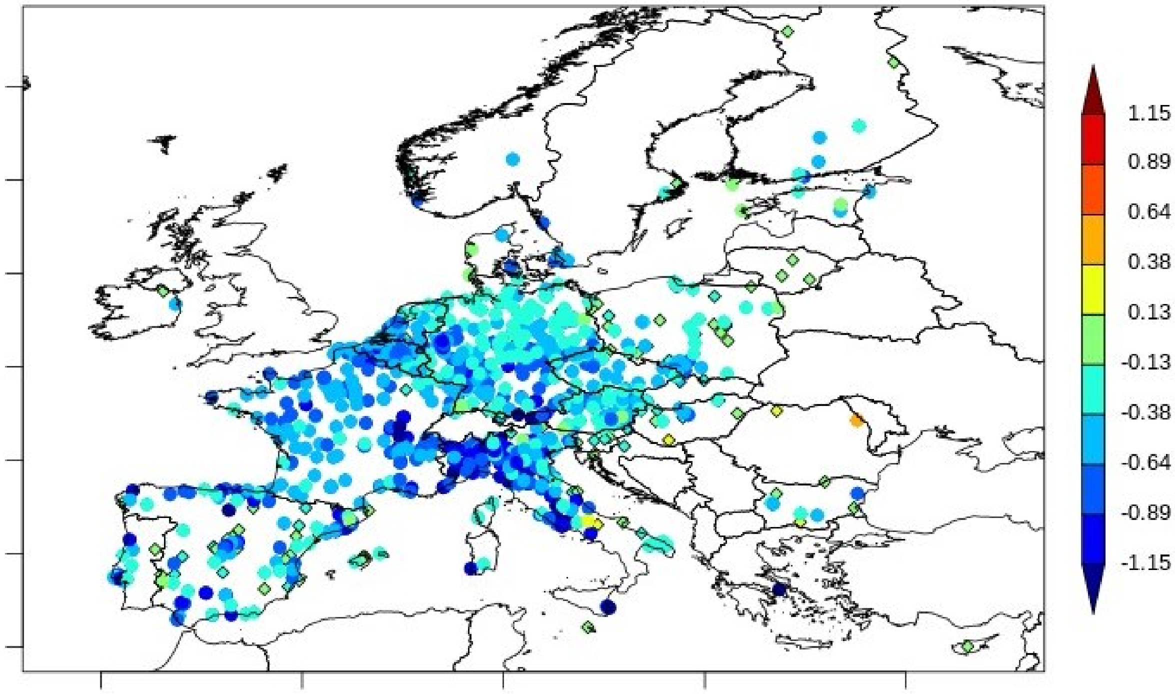


Figure 3.5: Top: weekly cycle of NO_2 per station type estimated from the whole time series in 2005 (solid lines) and 2021 (dashed lines). Bottom: corresponding absolute (solid lines, left y-axis) and relative (dashed lines, right y-axis) trends



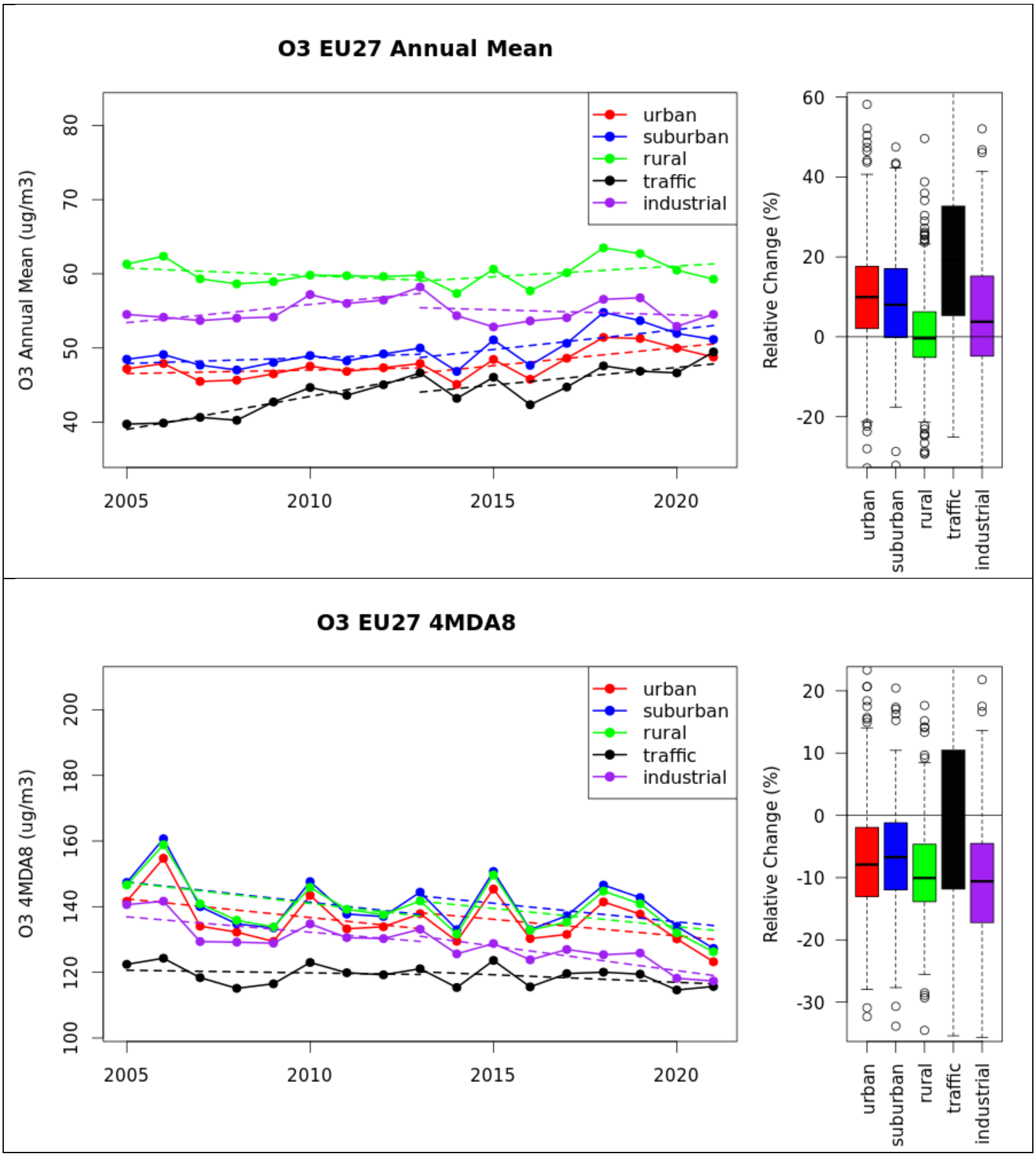
Map 3.2: Map of NO_2 trends (in µg/m³/yr) at all urban, suburban and rural background sites in Europe between 2005 and 2021. Significant trends are displayed as dots, and un-significant trends as diamonds

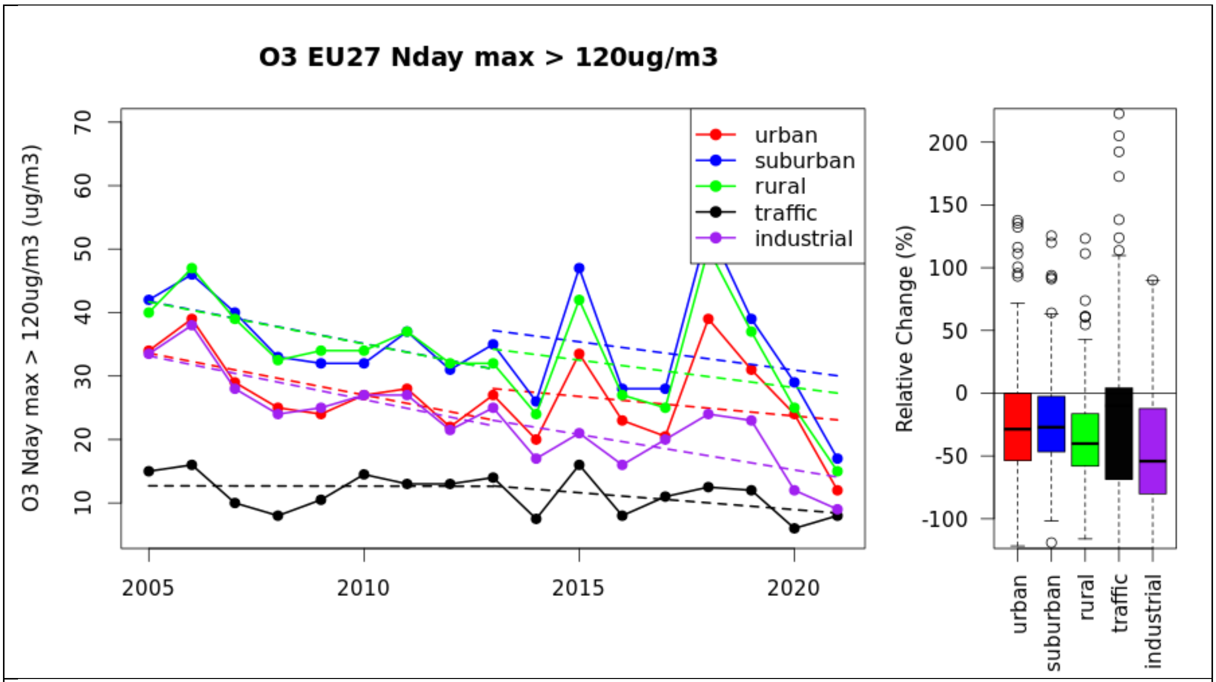


3.3 Ozone

For ozone, opposite trends have been identified in the past depending on the metrics with decreases of high ozone peaks, whereas annual mean ozone increased or displayed no significant trends (Fleming et al. 2018, Simpson et al. 2014). This finding is confirmed here: annual mean ozone increases while peaks decrease (Figure 3.6). The increase of annual mean can be substantial, especially at traffic sites where it reaches almost 22 % over Europe (Table 3.3).

Figure 3.6: For ozone annual mean (top), fourth highest daily peak (4MDA8, second row), number of days of exceedance of maximum daily eight-hour mean over the target value 120µg/m³ (third row), and Ox (as O_3+NO_2) annual mean (bottom): The continuous lines show the time series of the European-wide composite (median) per station typology (red: urban background, blue: suburban background, green: rural background, black: traffic, violet: industrial) between 2005 and 2021. The dashed lines show the linear fit between 2005 and 2013 and between 2013 and 2021. The boxplots on the right-hand side show the distribution of relative changes (%) between 2005 and 2021 for all stations of each typology




O3 EU27 Nday max > 120ug/m3
O3 Nday max > 120ug/m3 (ug/m3)
urban
suburban
rural
traffic
industrial
Relative Change (%)

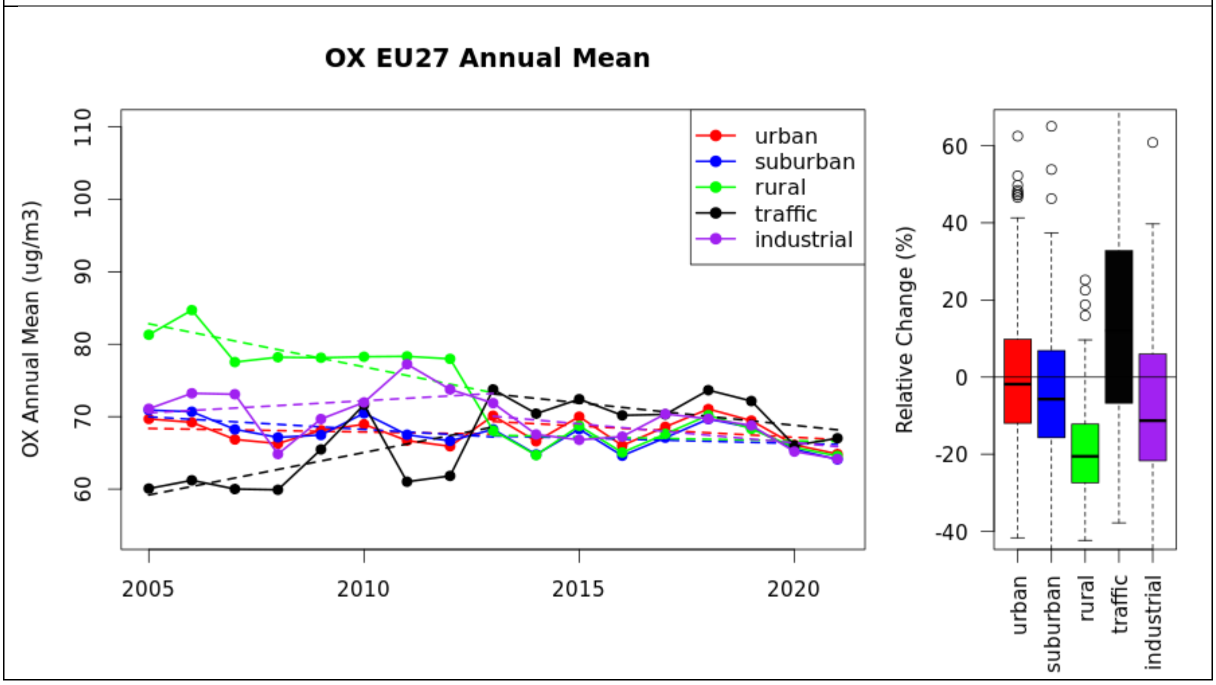

OX EU27 Annual Mean
OX Annual Mean (ug/m3)
urban
suburban
rural
traffic
industrial
Relative Change (%)

Table 3.3: Summary of observed ozone indicator trends per station typology: total number of stations, 5th and 95th quantiles of Sen Theil slopes at all European stations, trend of the median of the European-wide (EU27) composite (Sen-Theil slope, µg/m3/yr or µg/m3.hours/yr for AOT40 or µg/m3.days/yr for SOMO35), significance of the European-wide composite (Mann-Kendall p-value, MK p-val) and percentage change between: 2005 and 2021, 2005 and 2013, 2013 and 2021 for the European-wide composite (% change)

Metric	Station Type	Number of Stations	5th and 95th quantile of trends	Trend of Median	MK p-val	Relative Change (2021 vs 2005 %)	Relative Change (2013 vs 2005 %)	Relative Change (2021 vs 2013 %)
Annual Mean	urban	425	[-0.41;0.75]	0.26	0.01	9.7	2.0	8.0
Annual Mean	suburban	288	[-0.38;0.75]	0.30	0.01	10.7	3.0	9.0
Annual Mean	rural	347	[-0.68;0.61]	0.06	0.65	1.8	-3.0	4.0
Annual Mean	traffic	86	[-0.47;1.27]	0.51	0.00	21.8	18.0	9.0
Annual Mean	industrial	107	[-0.71;0.81]	0.01	0.84	0.4	7.0	-2.0
Seasonal Mean JJA	urban	425	[-0.66;0.78]	0.10	0.65	2.6	1.0	-2.0
Seasonal Mean JJA	suburban	287	[-0.62;0.75]	0.16	0.43	4.2	1.0	-3.0
Seasonal Mean JJA	rural	346	[-0.96;0.47]	-0.10	0.48	-2.4	0.0	-6.0
Seasonal Mean JJA	traffic	86	[-1.08;1.27]	0.40	0.01	13.0	11.0	2.0
Seasonal Mean JJA	industrial	107	[-1.29;0.72]	-0.12	0.48	-3.1	2.0	-7.0
Seasonal Max JJA	urban	425	[-2.28;1.22]	-0.71	0.27	-7.8	-4.0	-6.0
Seasonal Max JJA	suburban	287	[-2.12;0.83]	-0.58	0.27	-6.2	-5.0	-8.0
Seasonal Max JJA	rural	346	[-2.20;0.87]	-0.71	0.20	-7.8	-4.0	-9.0
Seasonal Max JJA	traffic	86	[-2.47;1.61]	-0.34	0.13	-4.4	0.0	-7.0
Seasonal Max JJA	industrial	107	[-3.87;0.94]	-1.29	0.00	-14.6	-8.0	-7.0
4MDA8	urban	425	[-1.88;0.70]	-0.47	0.13	-5.6	-6.0	-6.0
4MDA8	suburban	288	[-1.87;0.57]	-0.59	0.11	-6.8	-7.0	-6.0
4MDA8	rural	347	[-1.93;0.38]	-0.75	0.05	-8.8	-7.0	-6.0
4MDA8	traffic	86	[-2.08;1.34]	-0.23	0.23	-3.3	-1.0	-3.0
4MDA8	industrial	107	[-2.49;0.91]	-1.08	0.00	-13.4	-5.0	-9.0
Nday max > 120ug/m3	urban	425	[-2.99;0.65]	-0.67	0.10	-36.4	-31.0	-18.0
Nday max > 120ug/m3	suburban	288	[-2.86;0.69]	-0.83	0.08	-35.7	-26.0	-19.0
Nday max > 120ug/m3	rural	347	[-3.36;0.32]	-1.10	0.03	-46.5	-25.0	-20.0
Nday max > 120ug/m3	traffic	86	[-2.98;1.38]	-0.28	0.12	-35.2	-1.0	-34.0
Nday max > 120ug/m3	industrial	107	[-3.72;0.66]	-1.17	0.00	-61.3	-33.0	-39.0
SOMO35	urban	422	[-198.63;101.28]	-11.06	0.84	-4.6	-10.0	0.0

Metric	Station Type	Number of Stations	5th and 95th quantile of trends	Trend of Median	MK p-val	Relative Change (2021 vs 2005 %)	Relative Change (2013 vs 2005 %)	Relative Change (2021 vs 2013 %)
SOMO35	suburban	286	[-175.10;100.07]	-18.86	0.43	-6.5	-10.0	-2.0
SOMO35	rural	347	[-239.22;85.54]	-39.56	0.30	-11.9	-7.0	-11.0
SOMO35	traffic	84	[-191.53;178.39]	43.75	0.01	30.4	30.0	5.0
SOMO35	industrial	107	[-273.40;121.36]	-50.61	0.06	-17.4	-4.0	-15.0
SOMO10	urban	425	[-248.63;241.01]	58.90	0.27	5.7	-1.0	6.0
SOMO10	suburban	288	[-212.51;248.24]	50.16	0.34	4.6	-1.0	5.0
SOMO10	rural	347	[-266.34;193.46]	-20.18	0.65	-1.6	-3.0	2.0
SOMO10	traffic	86	[-328.48;526.01]	172.34	0.00	20.0	21.0	7.0
SOMO10	industrial	107	[-367.37;335.68]	-12.75	0.77	-1.1	7.0	-3.0
AOTcrops	urban	422	[-916.10;320.79]	-224.30	0.30	-25.6	-34.0	-17.0
AOTcrops	suburban	285	[-909.61;269.73]	-249.61	0.39	-22.9	-33.0	-18.0
AOTcrops	rural	347	[-1076.09;145.09]	-377.67	0.13	-32.9	-34.0	-21.0
AOTcrops	traffic	84	[-820.05;493.18]	-62.73	0.43	-14.3	-13.0	-11.0
AOTcrops	industrial	104	[-996.68;261.67]	-288.74	0.04	-33.2	-23.0	-37.0
AOTforest	urban	422	[-1380.08;561.52]	-210.33	0.27	-16.2	-18.0	-4.0
AOTforest	suburban	286	[-1372.57;499.56]	-253.62	0.30	-15.6	-17.0	-7.0
AOTforest	rural	347	[-1723.46;375.16]	-449.98	0.09	-25.1	-16.0	-14.0
AOTforest	traffic	84	[-1322.11;949.69]	60.65	0.30	9.1	12.0	6.0
AOTforest	industrial	107	[-1741.29;523.44]	-445.84	0.02	-31.4	-14.0	-20.0
OX	urban	379	[-1.14;1.07]	-0.06	0.43	-1.6	-1.0	-4.0
OX	suburban	206	[-1.26;0.86]	-0.27	0.04	-6.5	-4.0	-2.0
OX	rural	225	[-2.00;0.03]	-1.12	0.00	-23.3	-11.0	-1.0
OX	traffic	79	[-1.09;2.39]	0.60	0.03	16.7	16.0	-7.0
OX	industrial	80	[-2.13;1.05]	-0.39	0.02	-9.2	4.0	-6.0

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 NO_x emission decreases (Monks et al. 2015). The clear difference between rural sites compared to urban and suburban background sites (also seen in the boxplots of Figure 3.6) indicates that the decreased titration has more impact on the trends in Europe than hemispheric transport. Indeed, while hemispheric transport of ozone contributes to the overall tropospheric load (also referred to as burden) of ozone and might have increased since the start of the 20th century, there is no reason that it would have a larger effect in urban and traffic sites. On the contrary, the reduction in NOx emissions is expected to yield an increase of ozone annual mean values. This is why we attribute this sharper increase at urban and traffic sites to the titration effect rather than hemispheric transport.

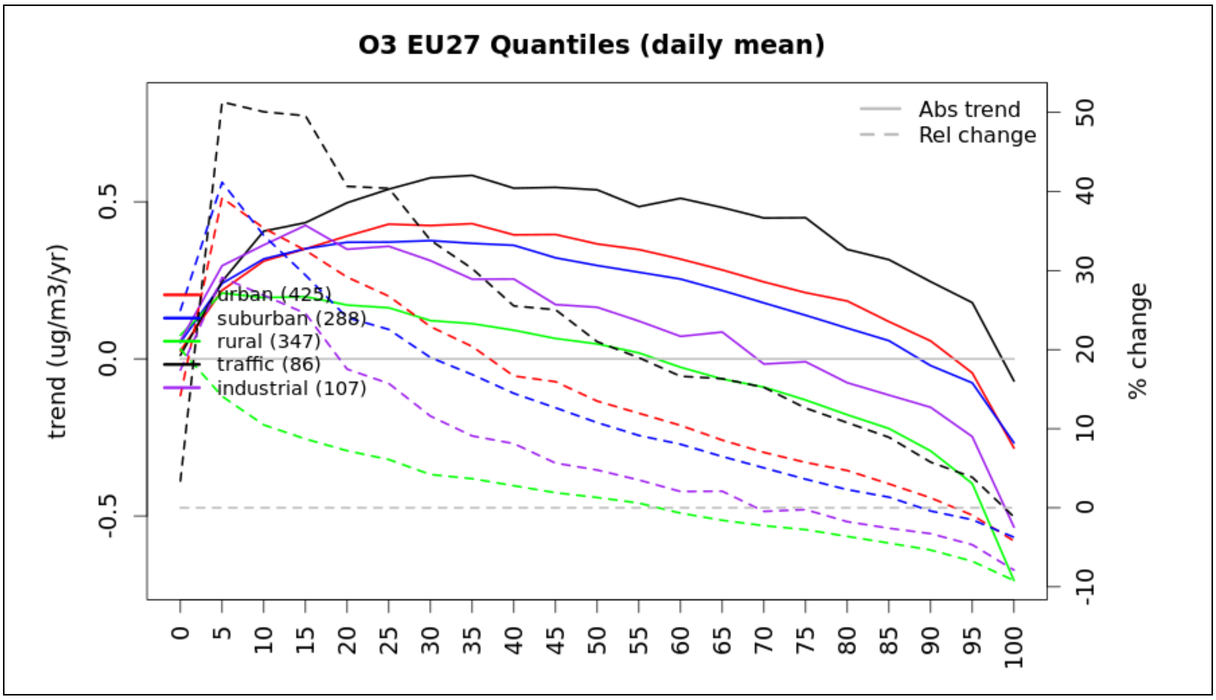
The ozone peaks are assessed from the fourth highest annual daily maximum of 8hr running mean called “4MDA8” trend (see Figure 3.6 and Table 3.3). This metric is taken instead of the summertime average because when only a handful of significant ozone air pollution episode occur in a year for a given station, the summertime average of daily maxima is not representative of high ozone episodes. Ozone peaks decreased over the period, about 6 to 9 % at background sites, but the trend of the European composite is not significant (Table 3.3). There is a flattening of the downward trend over recent years, with several increases reported over the period 2013-2021 even for ozone peaks.

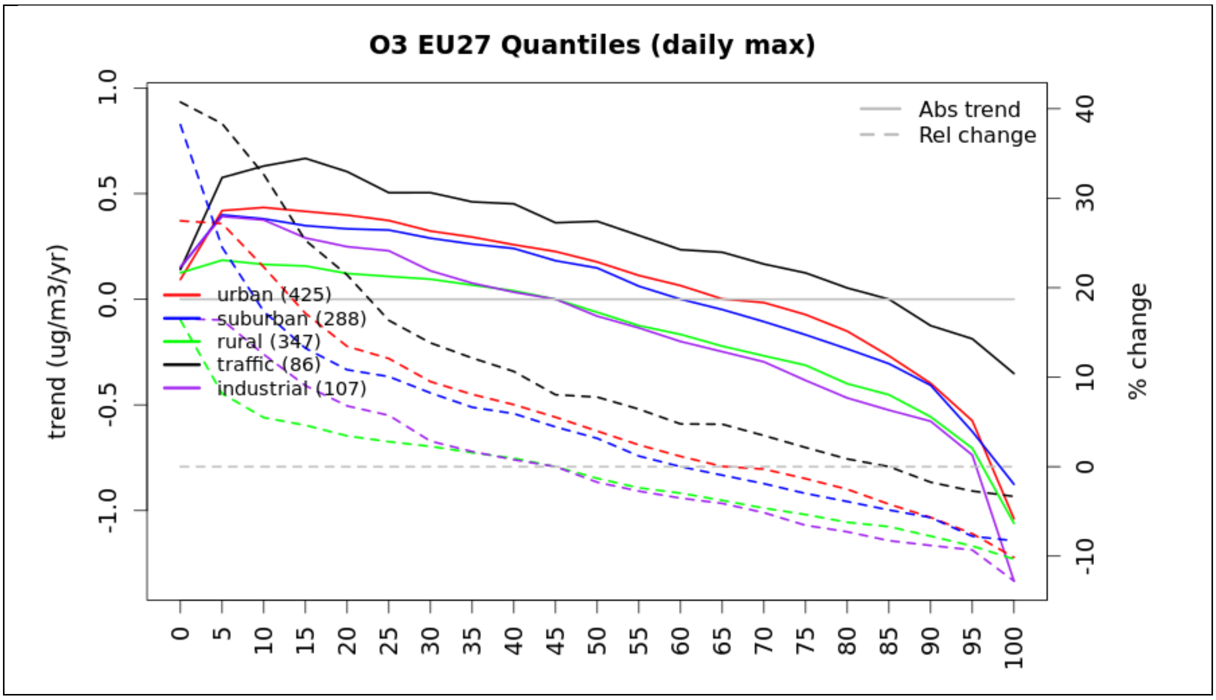
The number of days in exceedance of the target value 120µg/m^3 for the daily maximum eight-hour mean ozone is also given in Figure 3.6. The interannual variability is even stronger than for 4MDA8, with outstanding years in 2006, 2015, 2018 and 2019. The higher number of exceedances is found at background sites, where they declined by 36 %-47 % between 2005 and 2021, but slowly declined by 18 %-20 % between 2013 and 2021 (Table 3.3).

The trends of daily maxima and daily mean ozone percentiles illustrate well the difference between ozone trends for high and low concentrations (Figure 3.7). High quantiles of the daily maxima decrease by about 10 % at all sites with the exception of traffic sites, while at urban background sites low quantiles increase by more than 10 % below the 20th percentile.

It is mainly at rural and industrial sites that some decreases of the highest quantiles in daily means are seen.

Figure 3.7: For ozone and each typology of station, absolute trend (solid lines, left y-axis) and relative change (dashed lines, right y-axis) of the percentiles of daily mean (top) and daily maxima (bottom) over the period 2005-2021

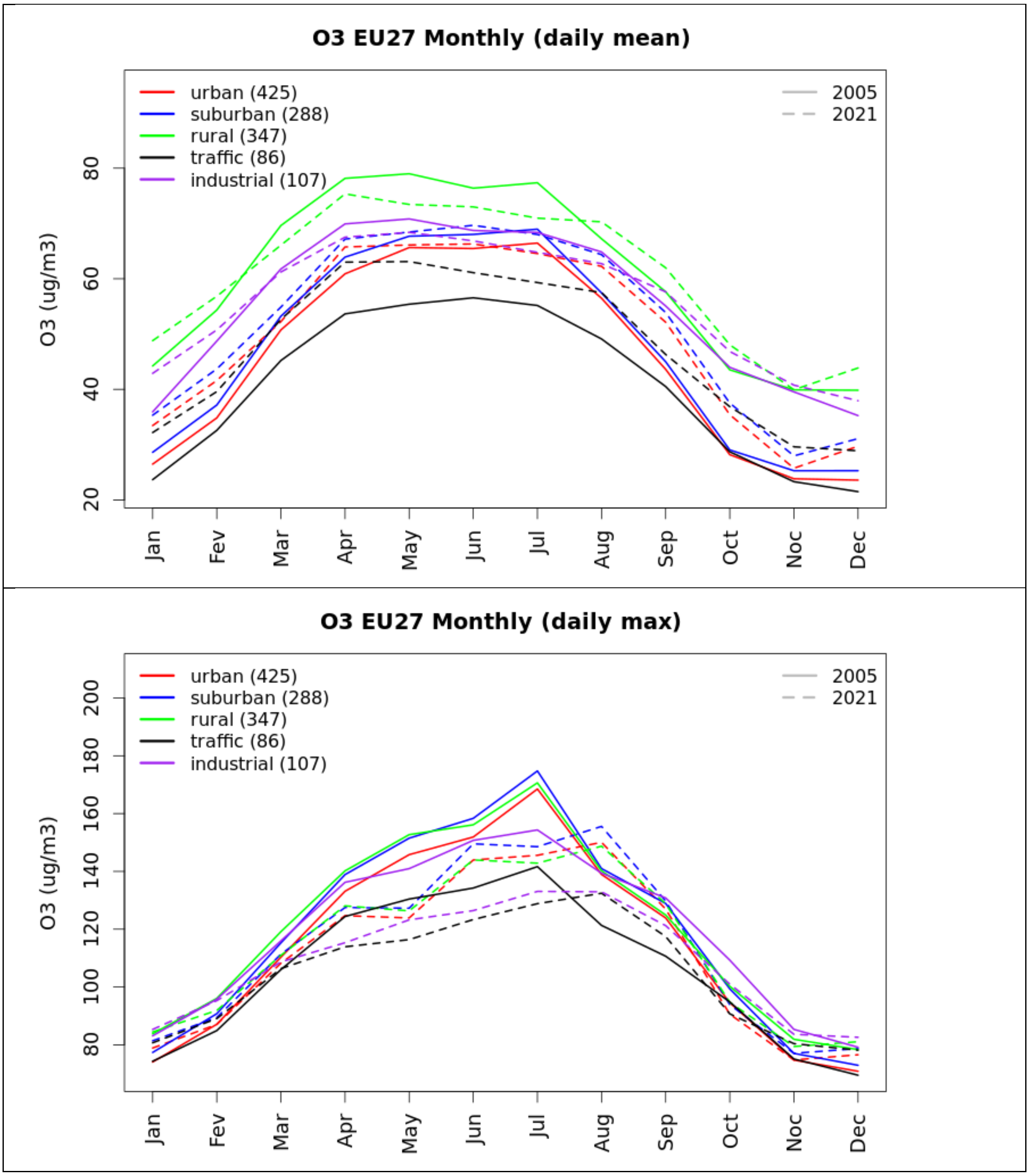




The trends in ozone health and ecosystem exposure are influenced by both high and low percentiles of ozone distributions. As an indicator of human health impact, SOMO35 (sum of ozone daily maxima in excess of 35 ppbv) is mainly relevant at urban and suburban sites where its trend is not significant and the relative change is -4.6 % and -6.5 %, respectively, over the whole period (Table 3.3). By also taking into account lower values of ozone daily maxima (from 10ppbv upwards), SOMO10 is more influenced by the titration effect than SOMO35, so that the increase at urban sites is still un-significant but reaches 5.7 % (Table 3.3). Regarding ecosystem exposure, AOT40 for crops is reduced by 32.9 % at rural sites, but the interannual variability is so large that the trend is not significant. Likewise, for forests, a non-significant 25.1 % reduction is observed.

Ozone displays a strong seasonal cycle illustrated in Figure 3.8 for both ozone daily means and daily maxima. Here the monthly cycles at the beginning (2005) and end (2021) of the period are compared by using estimated values for the years 2005 and 2021 based on a linear fit rather than the actual monthly cycle for those years to minimize the impact of interannual variability. To compute these estimated values for 2005 and 2021, we take the monthly cycle for either daily max or daily mean, for each year over the period. Then a linear regression is done for each month taking the 17 values between 2005 and 2021. And the fitted value for the years 2005 and 2021 are used rather than the actual monthly cycle of 2005 or 2021, which would be very sensitive to the actual choice of the starting/ending years because of interannual variability. If we discard the winter months and traffic sites, when ozone levels are lower, consistent decreases are still found for both ozone max and means, with a notable exception for the month of August and September, where the recent trend contributed to increase ozone levels. This feature was not visible in earlier assessment (Colette and Rouïl 2020) focusing on 2000-2017, highlighting the strong impact of the years 2018 and 2021 on the trend.

Figure 3.8: Monthly cycle of daily mean (top) and daily maxima (bottom) ozone per station typology estimated from the whole time series in 2005 (solid lines) and 2021 (dashed lines)



The ozone diurnal cycles are shown in Figure 3.9. The cycles are estimated for 2005 and 2021, and absolute and relative trends are also available on the figure. For all sites typologies, there is an increase in 2021 for all the hours in the day, although this increase is relatively small during the morning hours, and there is a slight decrease observed between 7UTC and 14UTC, which varied depending on the station type. Note that those cycles are averaged over a full year and would be shifted if considered only over summer.

Figure 3.9: Top: diurnal cycle of ozone per station typology estimated from the whole time series in 2005 (solid lines) and 2021 (dashed lines). Bottom: corresponding absolute trend (solid lines) and relative changes (dashed lines)

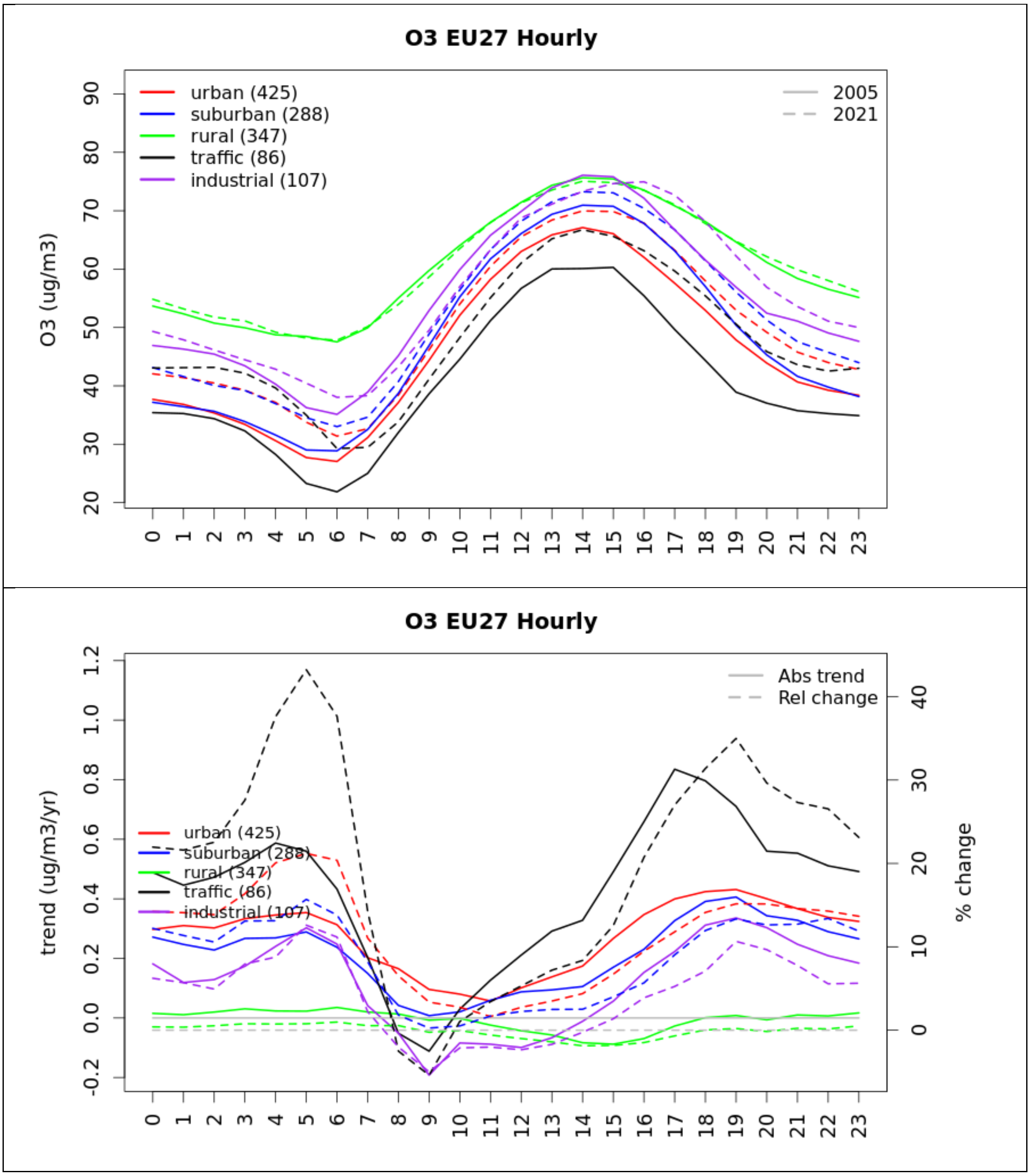


Figure 3.10: Top: diurnal cycle of Ox (as NO_2+O_3) per station typology estimated from the whole time series in 2005 (solid lines) and 2021 (dashed lines). Bottom: corresponding absolute trend (solid lines) and relative changes (dashed lines)

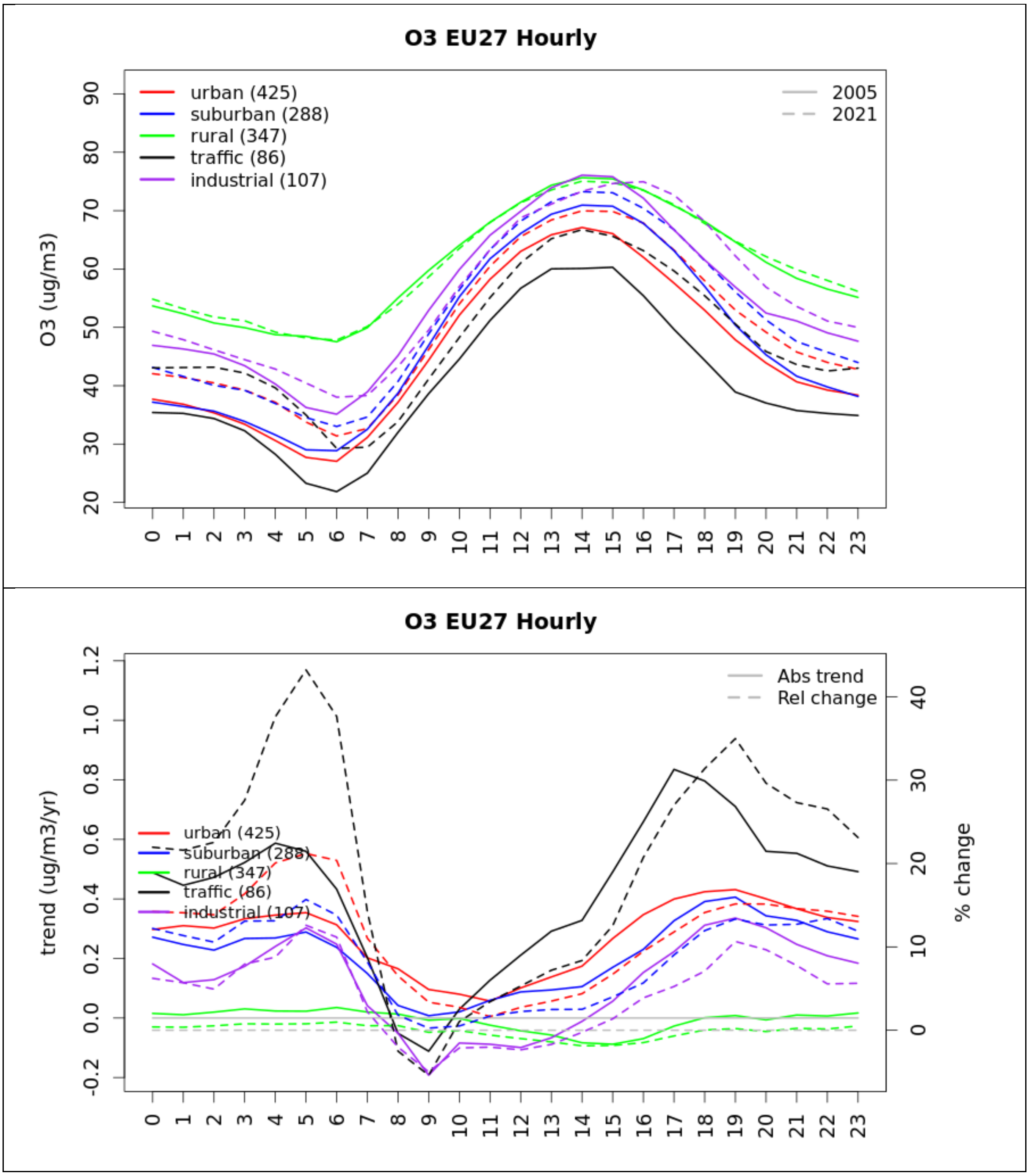
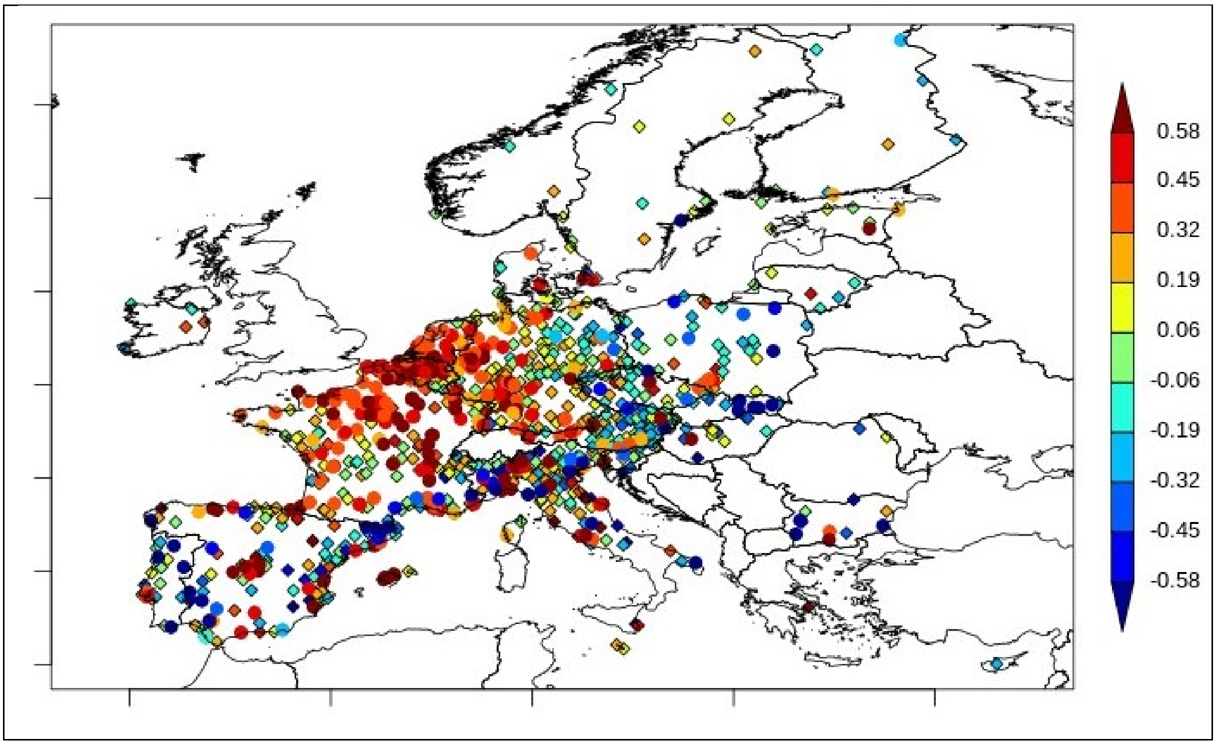
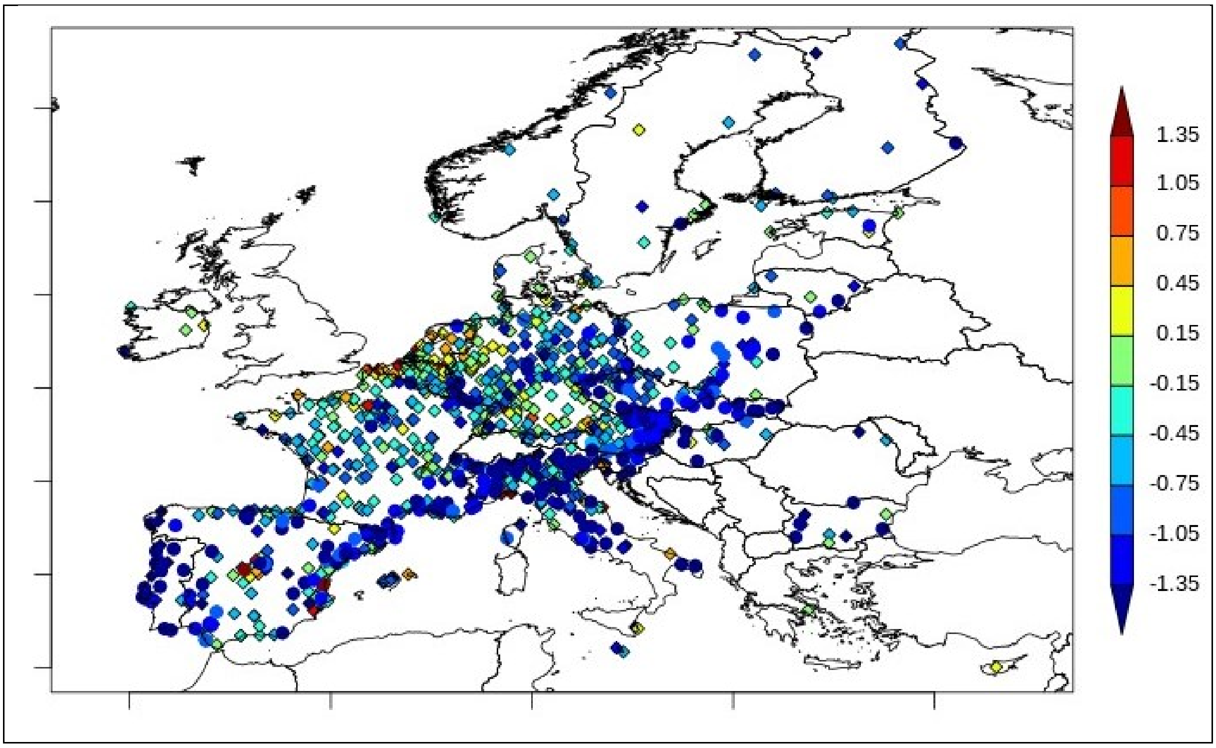


Figure 3.10 shows the diurnal cycle of O_x defined as the sum of O_3 and NO_2 concentrations. Because of the fast O_3/NO_2 reaction, it is chemically relevant to consider the trend of their sum. Both of them have also adverse health effects. However, as mentioned above, their trends are opposite (at 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. O_x trends are significantly decreasing at rural sites and industrial sites (-23.3 % and -9.2 %, respectively), and significantly increasing at traffic sites (+16.7 %) (Figure 3.6 and Table 3.3). At urban and suburban, un-significant reductions in trend of 1.6 and 6.5 % are found, respectively, Table 3.3.

The maps of the trends observed over Europe at background sites are given in Map 3.3 for ozone annual mean and 4MDA8. Decreases in ozone annual mean are recorded over Central/Eastern Europe (Austria, Czech Republic, and Poland) while there are more increases in Western Europe, in particular in Spain (although mixed with decreases), South-eastern France, and in the Benelux. There is however no strong latitudinal gradient that would be expected from different chemical regimes and photolysis rates. The amplitude of ozone peaks (as 4MDA8, which shows a general reduction) is slightly less reduced in Germany (to be related to the lower NO_2 decreases discussed in Section 3.2) and a couple of individual stations in Spain.

Map 3.3: Map of O_3 annual mean (top) and 4MDA8 (bottom) trends, in µg/m³/yr at all urban, suburban and rural background sites in Europe over 2005-2021. Significant trends are displayed as dots, and un-significant trends as diamonds





3.4 Particulate Matter

Strong significant downward trends of annual mean PM_{10} concentrations were observed in Europe since 2005 for all station types (Figure 3.11). The decrease over all European stations of annual mean PM_{10} is 42 % at urban and suburban sites (Table 3.4). The comparison of trends in the earlier and later part of the period indicates no flattening out in any type of sites (on the contrary there was a faster rate of change than in the first period), except for industrial sites, with a slower rate of change in recent years. For annual mean $PM_{2.5}$ concentrations, only changes after 2008 can be considered because of the scarcity of the network before that date. All (decreasing) trends are significant and the decrease of annual mean $PM_{2.5}$ is greater in comparison to that of PM_{10}: for instance, 51 % at urban sites.

Figure 3.11: For PM_{10} (top) and $PM_{2.5}$ (bottom) annual means: The continuous line represents time series of the European-wide composite (median) per station typology (red: urban background, blue: suburban background, green: rural background, black: traffic, violet: industrial) between 2005 and 2021 (top) and between 2008 and 2021 (bottom). The dashed lines show the linear fit between 2005 and 2013 (only top) and between 2013 and 2021. The boxplots on the right-hand side show the distribution of relative changes (%) between 2005 and 2021 (top) and between 2008 and 2021 (bottom) for all stations of each typology

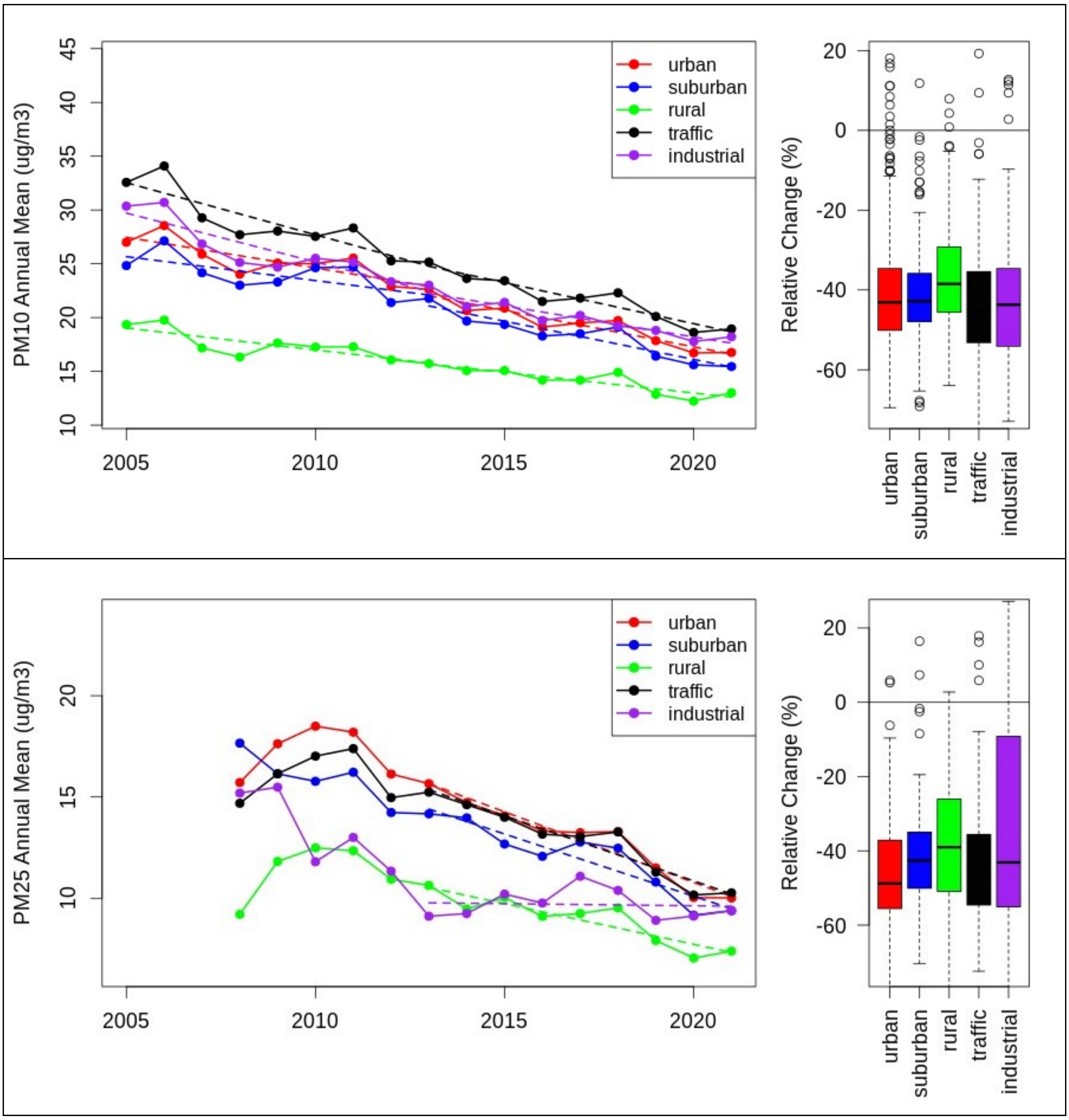


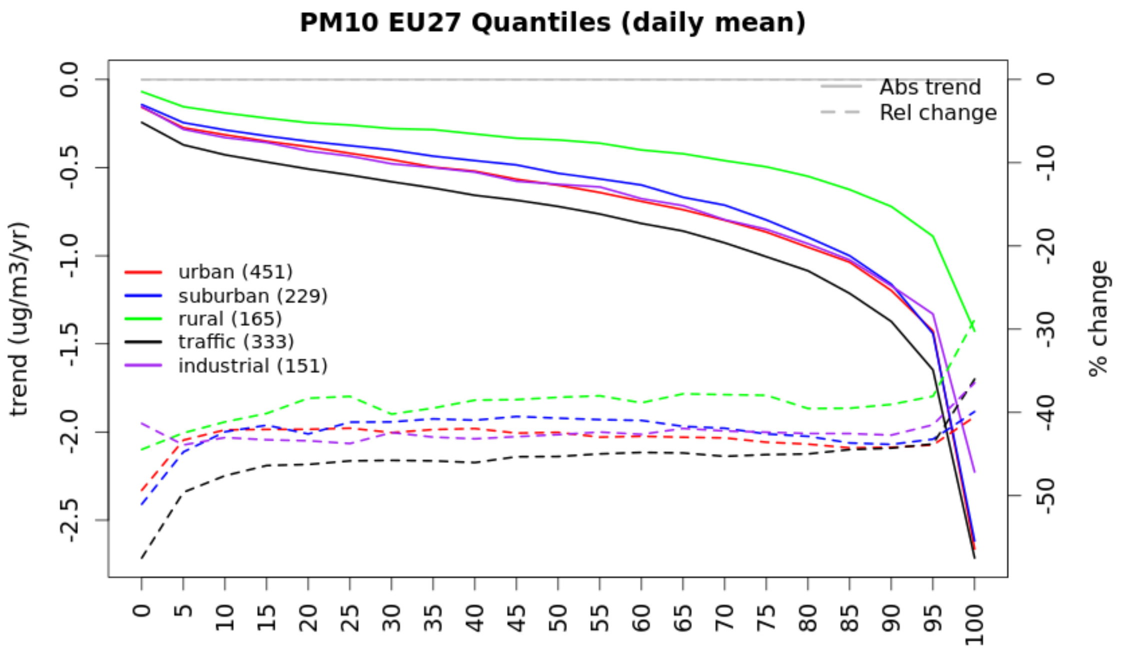
Table 3.4: Summary of observed PM indicator trends per station typology: total number of stations, 5th and 95th quantiles of Sen Theil slopes at all European stations, trend of the median of the European-wide composite (Sen-Theil slope, µg/m³/yr), significance of the European-wide composite (Mann-Kendall p-value, MK p-val) and percentage change between: 2005 (2008 for PM2.5) and 2021, 2005 and 2013 (for PM10 only), 2013 and 2021 for the European-wide (EU27) composite (% change)

Pollutant	Metric	Station Type	Number of Stations	5th and 95th quantile of trends	Trend of Median	MK p-val	Relative Change (2021 vs 2005 %) (PM25 : 2021 vs 2008, %)	Relative Change (2013 vs 2005 %)	Relative Change (2021 vs 2013 %)
PM10	Annual Mean	urban	463	[-1.49;-0.15]	-0.69	<0.01	-42.4	-17.0	-25.0
PM10	Annual Mean	suburban	230	[-1.17;-0.25]	-0.65	<0.01	-42.1	-14.0	-27.0
PM10	Annual Mean	rural	165	[-0.88;-0.05]	-0.43	<0.01	-38.9	-17.0	-20.0
PM10	Annual Mean	traffic	338	[-1.63;-0.31]	-0.82	<0.01	-43.6	-24.0	-25.0
PM10	Annual Mean	industrial	158	[-1.59;-0.12]	-0.71	<0.01	-41.3	-24.0	-21.0
PM10	Nday > 50ug/m3	urban	451	[-5.27;-0.17]	-1.61	<0.01	-92.2	-54.0	-83.0
PM10	Nday > 50ug/m3	suburban	229	[-4.48;0.00]	-1.17	<0.01	-84.6	-29.0	-79.0
PM10	Nday > 50ug/m3	rural	165	[-2.05;0.00]	-0.40	<0.01	-88.4	-24.0	-67.0
PM10	Nday > 50ug/m3	traffic	333	[-6.17;-0.17]	-2.21	<0.01	-90.6	-59.0	-76.0
PM10	Nday > 50ug/m3	industrial	151	[-4.73;0.00]	-1.63	<0.01	-94.7	-73.0	-54.0
PM25	Annual Mean	urban	199	[-1.33;-0.23]	-0.67	<0.01	-51.2	N/A	-36.0
PM25	Annual Mean	suburban	61	[-0.93;-0.01]	-0.58	<0.01	-47.2	N/A	-34.0
PM25	Annual Mean	rural	75	[-0.89;-0.05]	-0.39	<0.01	-46.1	N/A	-30.0
PM25	Annual Mean	traffic	99	[-1.32;0.05]	-0.55	<0.01	-44.9	N/A	-33.0
PM25	Annual Mean	industrial	40	[-0.96;0.14]	-0.36	<0.01	-37.0	N/A	-2.0

The trends of PM_{10} and $PM_{2.5}$ in Table 3.4 cannot be directly compared as they rely on different monitoring networks (about half as many $PM_{2.5}$ stations compared to PM_{10}). We must therefore limit the comparison to those sites with co-located PM_{10} and $PM_{2.5}$ measurements . Then we only include 54 and 29 urban and rural sites, respectively. For urban sites, PM_{10} and $PM_{2.5}$ relative changes between 2008 and 2021 are broadly consistent: 32 and 37 % reduction, respectively. But for rural sites, PM_{10} decrease faster (-19 %) than $PM_{2.5}$ (-14 %). Map 3.5 shows the location of these urban and rural sites where the measurements of both PM_{10} and $PM_{2.5}$ sites are co-located.

In agreement with the reduction in annual mean PM_{10}, the frequency of days exceeding the regulatory limit value (50 µg/m³) decreased significantly, by 85 to 95 % depending on the station typology (Table 3.4). However, when comparing rural sites to other locations (Figure 3.12), the decline in the daily mean PM_{10} above the median (i.e. for quantiles higher than 50) is relatively smaller. This behaviour is illustrative of the fact that anthropogenic sources (affecting primarily non-rural sites) declined faster than natural and biogenic sources. Furthermore, the highest peaks (quantiles higher than 95) declined less at all types of stations, which shows that the most intense episodes affected by adverse stagnating meteorological conditions are less efficiently reduced than average values.

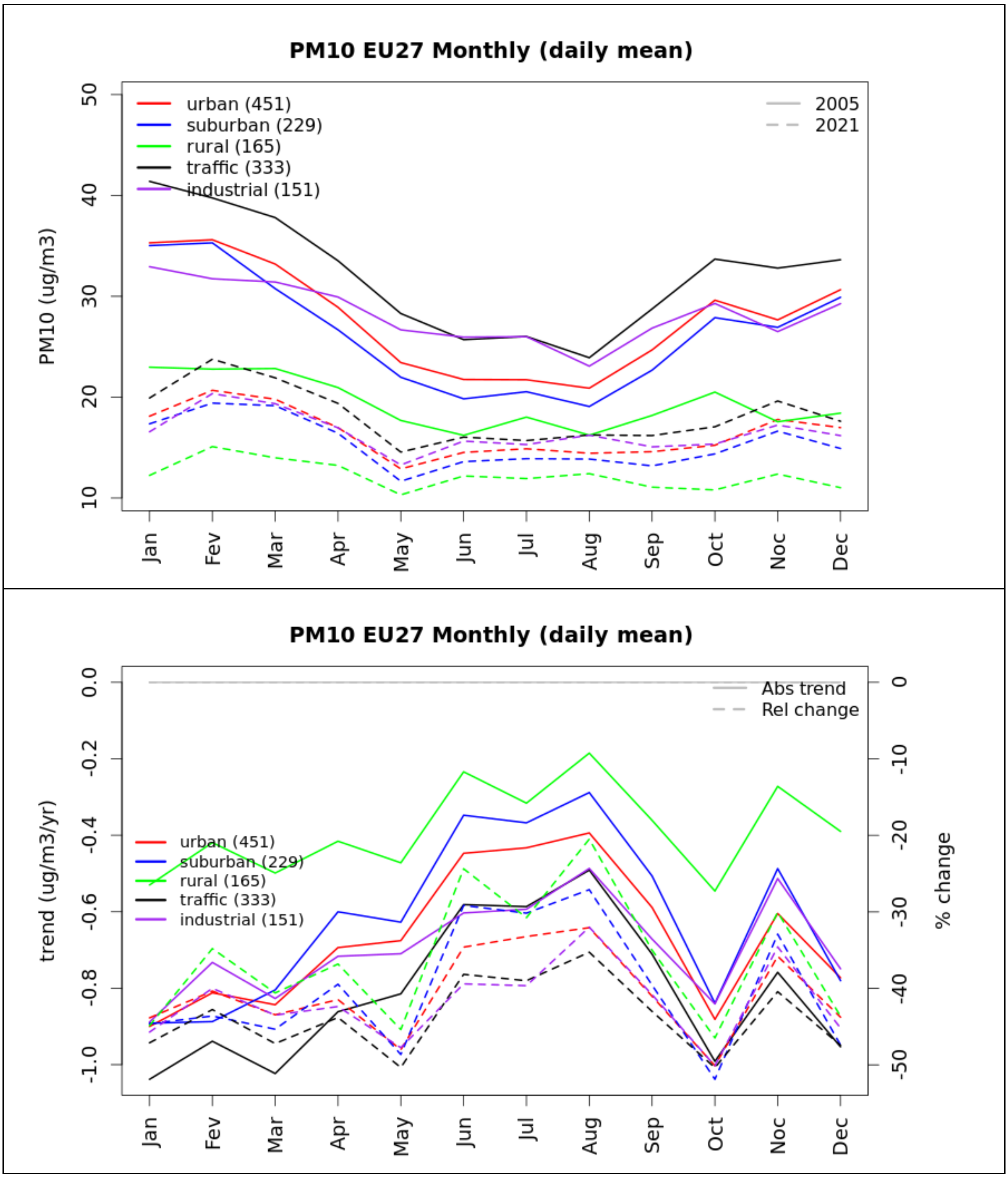
Figure 3.12: For PM_{10} and each typology of station: absolute trend (solid lines, left y-axis) and relative change (dashed lines, right y-axis) of the percentiles of daily mean over the period 2005-2021



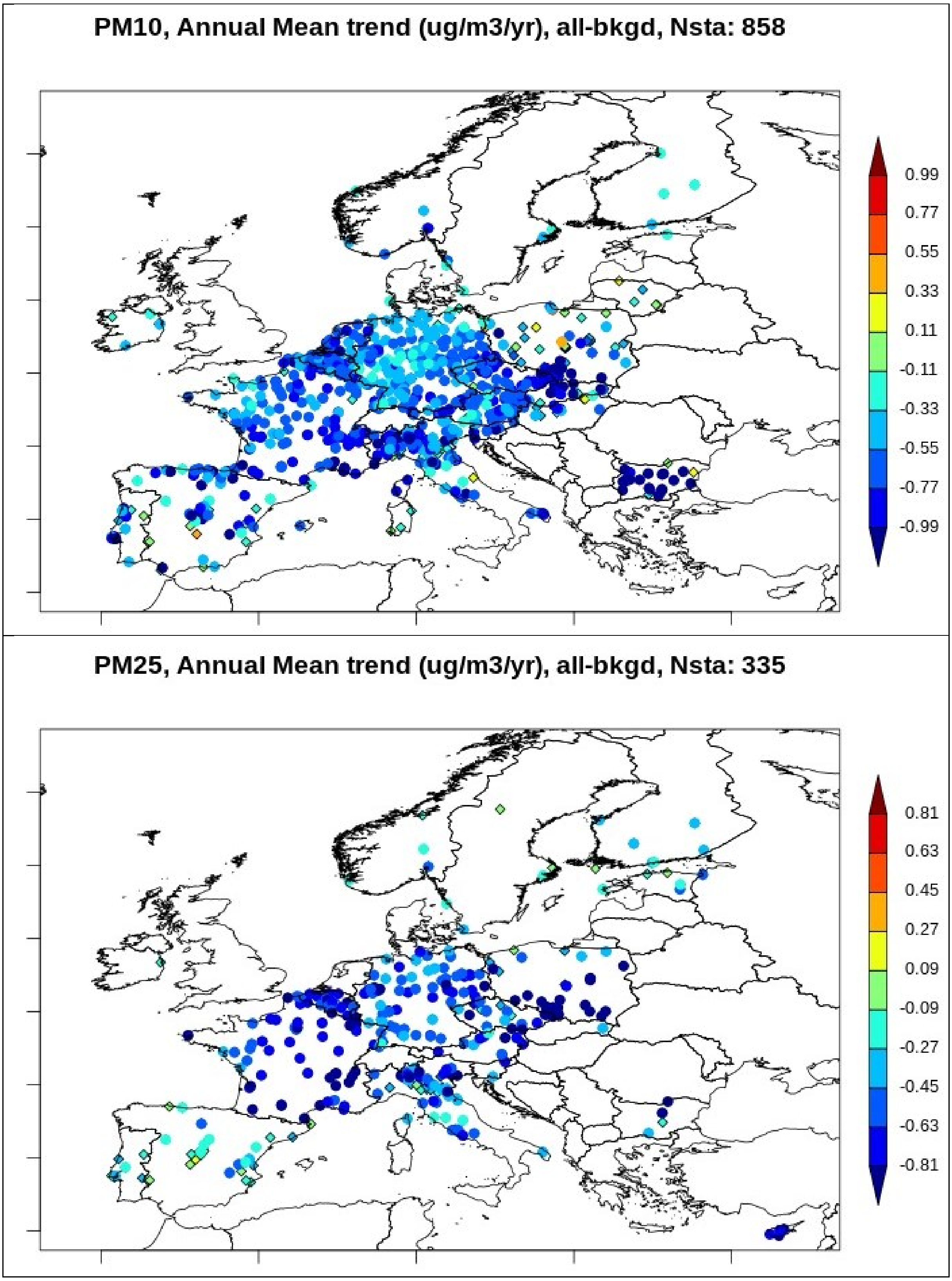
The monthly PM_{10} cycle shows a clear winter maximum at urban, suburban and traffic sites both for the 2005 and 2021 estimates (Figure 3.13). It can also be seen in industrial sites, yet with a smaller amplitude. The amplitude of the monthly cycle has been reduced between both years at all station types. It is in summer (when PM_{10} levels are lower) that the relative reduction has been more limited, in particular because of the contribution of natural sources (including secondary organic aerosols formed from biogenic VOCs).

Decreasing trend in PM_{10} and $PM_{2.5}$ concentrations are recorded in all European countries. Lower rate of declines is seen in Germany, Austria, Lithuania and Finland for both PM_{10} and $PM_{2.5}$, and in Spain (only for $PM_{2.5}$) (Map 3.4). In Poland, some increases in PM_{10} are found, although they remain non-significant from a statistical standpoint.

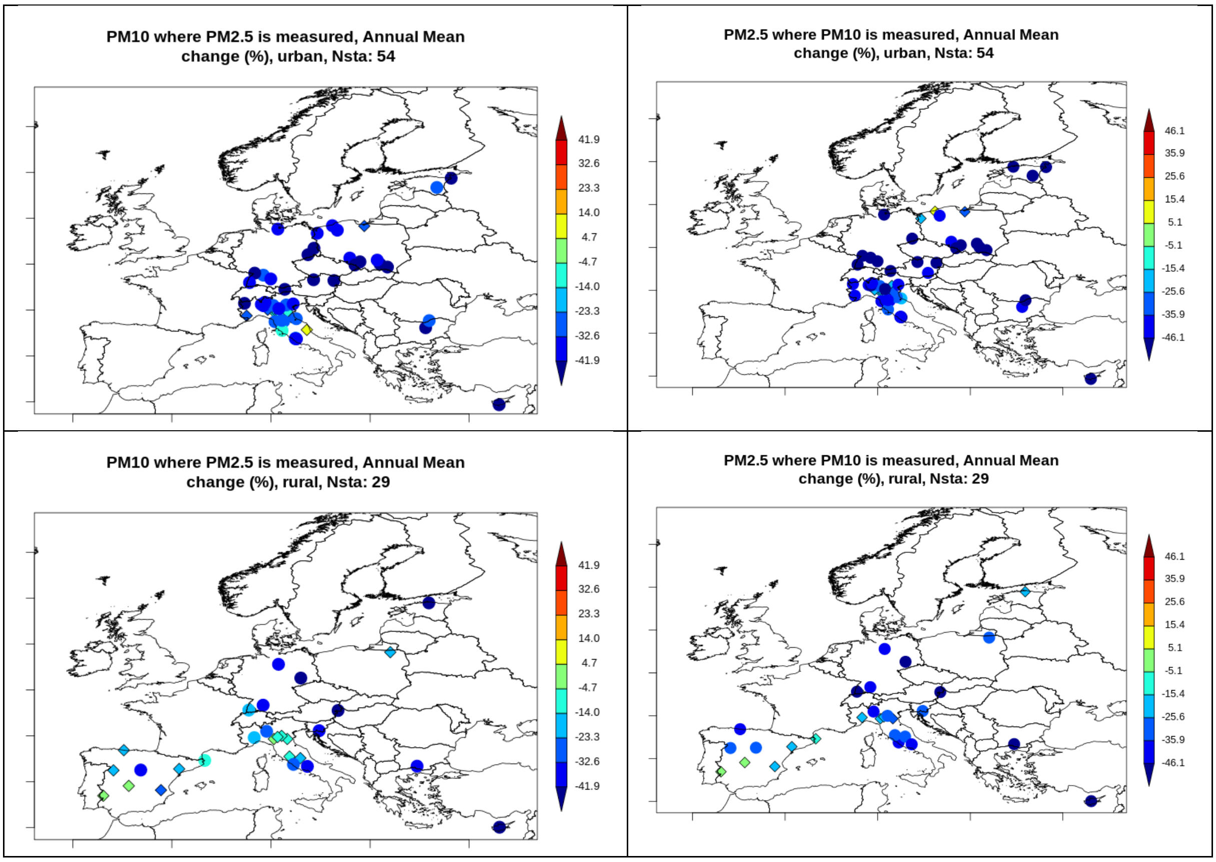
Figure 3.13: Top: Annual cycle of monthly mean of daily PM_{10} per station type estimated from the whole time series in 2005 (solid lines) and 2021 (dashed lines). Bottom: absolute trend (solid lines, left y-axis) and relative change (dashed lines, right y-axis) of the percentiles of daily mean over the period 2005-2021



Map 3.4: Map of PM_{10} annual mean (top) and $PM_{2.5}$ annual mean (bottom) trends, in µg/m^3/yr at all urban, suburban and rural background sites in Europe over 2005-2021 (2008-2021 for $PM_{2.5}$). Significant trends are displayed as dots, and un-significant trends as diamonds



Map 3.5: Left columns: Maps of PM_{10} annual mean change, in %, at urban (top) and rural (bottom) background sites in Europe over 2008-2021, only where $PM_{2.5}$ measurements are also present. Right: same but for $PM_{2.5}$ trends at sites where PM_{10} are also measured. Significant changes are displayed as dots, and un-significant changes as diamonds



3.5 Air Quality Index

The evolution of air quality in Europe is synthetized in Figure 3.14 using an adaptation of the EEA air quality index (AQI) that categorizes air pollutants ($PM_{2.5}$, PM_{10}, NO_2, O_3 and SO_2) concentrations in 6 ranges: good, fair, moderate, poor, very poor and extremely poor. As in the EEA AQI, for a given location, the category is computed for each pollutant, and the index accounts for the worst category of all 5 pollutants. For some pollutants, the EEA AQI uses hourly thresholds, in that case we refer to the daily maxima instead, and for the other pollutants we use the daily means, as in the EEA AQI. While the EEA AQI is designed to apply at a given monitoring station, we have very few stations covering all pollutants for a long time period in the present report. To cope with this limitation, once the categories are estimated for each pollutant for a given day, we compute the index as the median category over each country and over the whole EU27. Ultimately, for each year between 2005 and 2021, we display in Figure 3.14 the distribution of the days falling in each AQI category in order to provide a synthetic view to what extent the overall air quality situation is improving, irrespective of the individual air pollutants.

Figure 3.14: 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, violet: extremely poor, see Table 2.2)

A slight improvement is found for the overall index. In 2005, less than half of the days were “fair”. This proportion reaches about 70 % by 2021 with fewer days classified as “moderate” or “poor”. The use of a median value over the whole Europe (as EU27) evens out the variability so that no “good”, “very poor”, or “extremely poor” days appear in the overall index.

Figure 3.14 also shows the long-term evolution of the good/fair/moderate/poor/very poor/extremely poor categories by pollutant over Europe. The number of “good” days for NO_2, PM_{10} and $PM_{2.5}$ has increased clearly. But for ozone, the distribution has not changed substantially, and the daily categories for SO_2 were almost all already good at the beginning of the period.

Regarding the network presented in Figure 2.1, its long-term evolution does not indicate significant sampling biases related to a change in station typology. Although the number of stations has significantly increased, the relative proportion of urban/rural sites has remained relatively constant. One crucial factor affecting the air quality trend could be the development of PM monitoring, so that the overall index could have been biased towards more favourable categories in the earlier part of the period, what would reinforce the overall improvement of the air quality along the years.

4 Drivers of changes in air pollutant trends in Europe

After the descriptive statistics analysis presented in Chapter 3, we aim here to investigate the main drivers of air pollution trends in Europe. In a first subsection, the observed concentrations changes are put in perspective with the reported emission trends for the various counties. Then we introduce an innovative unsupervised machine learning approach to identify the outstanding group of stations within each country which display specific features in terms of evolution of air quality. The underlying idea of this work is to pave the way for further investigation with the corresponding member states to find insight in relation to the implementation of specific air quality plans, which could explain the larger improvement observed for some air quality stations. It would then be possible to draw some lessons to generalise the most efficient mitigation strategies. Additional results related to drivers for individual countries and for EU27 as a whole, spanning the periods 2005-2021 and 2000-2021, are also presented in Annex 1 and 2.

4.1 Consistency between concentration and emission trends

4.1.1 Sulphur dioxide

Figure 4.1 presents a comparison between reported SOx emission between 2005 and 2021 and observed annual mean SO_2 concentrations, both normalized at their 2005 levels (the intercept of the linear fit is used to estimate 2005 levels and minimize the effect of interannual variability). In order to avoid spurious time series for countries where too few stations are available, we select only the countries where we have more than 5 stations for at least one station type. This selection is performed independently for traffic, industrial and background (either rural, suburban and urban) sites. This means that for each country with more than 5 stations of typology “traffic”, “industrial”, or any “background” type, the median observation of the corresponding type is computed, and subsequently the European median over the countries where more than 5 stations were available is shown in Figure 4.1. In Table 4.1, we provide the corresponding numerical values for the relative trends in emissions and concentrations. Here we only include background stations in the estimate of relative concentration trends. We consider that this is the most sensible comparison with trends in total national emissions, even if the observed trends at traffic or industrial stations offer complementary insight for the analysis in Figure 4.1. The total number of background stations and corresponding relative changes in emissions and concentrations at background sites are also provided in Figure 4.1. As in the boxplot on the right-hand side of Figure 3.1, we can note a case where the relative decrease exceeds 100 % (for SOx emissions in Bulgaria) which would result in unrealistic negative levels in 2021, this is an artifact due to the linear approximation used to fit the trends.

The relation between emission and concentration is not straightforward, and Chemistry-Transport Models (CTM) should be involved for a more detailed analysis, but it remains insightful to compare their qualitative evolution. Emissions of SOx decreased by 76 %, and the measured concentrations showed a reduction of 61 % over the whole period, based on the median, for all background sites in Europe EU27. Note that because we compare to country total reported emissions, in Table 4.1 we take the median observed trend over all stations of background typology in any given country where more than 5 background stations are available for the whole period. As can be seen in Figure 4.1, the agreement is relatively good until 2008, and after that year a substantial gap starts to grow until the end of the period. This mismatch was even more clear in the corresponding figure covering the 2000-2017 period in (Colette and Rouïl 2020).

Figure 4.1: Time series of country median SO_2 observed at traffic (black), industrial (violet) and background (cyan) sites (solid lines), and corresponding national SOx emissions (dashed line) normalised to estimated 2005 levels (%). The median is taken over countries where more than 5 stations of a least one typology is available. The total number of stations is provided in brackets. Straight red and cyan lines are the linear fits of, respectively, emissions and concentrations at background stations over the whole period

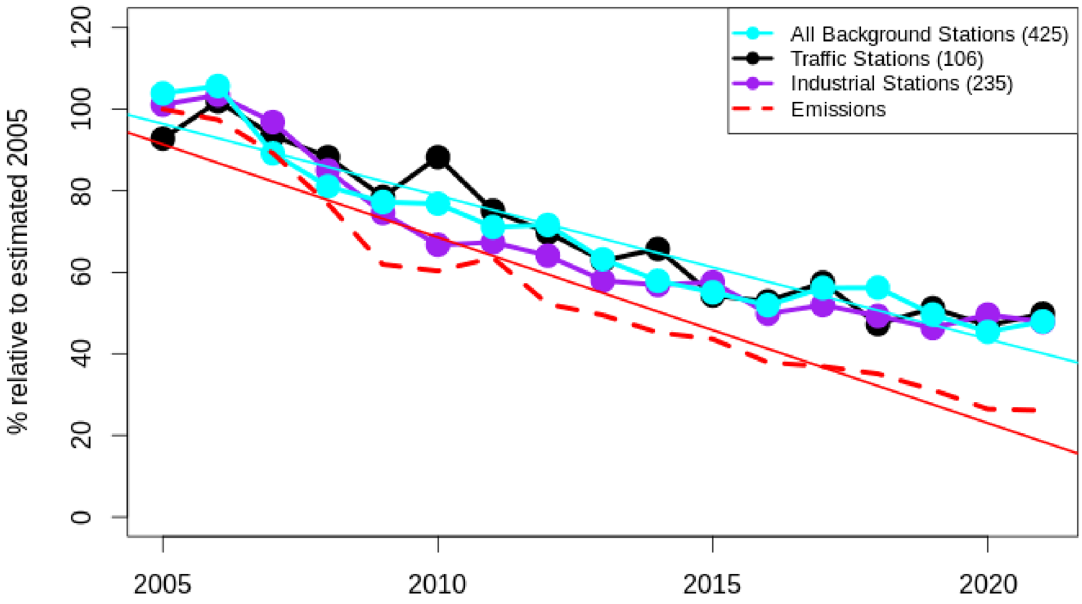


Table 4.1: Change (in %), relative to 2005 for all pollutants except $PM_{2.5}$ which is relative to 2008, for emissions and concentrations, as median over countries with more than 5 background stations of at least one typology (urban, suburban or rural) (Nsta, number of stations, otherwise indicated as "-")

	SOx/SO_2			NOx/NO_2			PPM_{10}/PM_{10} ([a])			$PPM_{2.5}/PM_{2.5}$ ([b])		
Country	Nsta	Emis	Conc	Nsta	Emis	Conc	Nsta	Emis	Conc	Nsta	Emis	Conc
AT	45	-52,8	-55,1	86	-48,3	-38,7	80	-31,9	-46,9	12	-46,9	-49,0
BE	17	-83,1	-91,4	34	-60,4	-48,0	30	-50,3	-45,3	22	-57,8	-54,5
BG	13	-112,7	-37,5	10	-56,7	-18,9	23	-38,1	-52,1	-	-	-
CY	-	-	-	-	-	-	-	-	-	-	-	-
CZ	25	-73,6	-59,3	31	-51,9	-40,7	51	-42,8	-42,3	21	-45,8	-39,7
DE	83	-54,4	-69,1	201	-36,1	-37,3	196	-25,2	-40,9	74	-41,3	-47,8
DK	-	-	-	-	-	-	-	-	-	-	-	-
EE	-	-	-	-	-	-	-	-	-	-	-	-
ES	101	-44,4	-42,7	132	-53,0	-43,7	61	-29,5	-43,3	31	-24,6	-21,3
FI	-	-	-	8	-55,0	-45,0	-	-	-	-	-	-
FR	32	-95,0	-89,1	202	-54,6	-45,0	158	-40,2	-46,1	53	-47,2	-57,0
GR	-	-	-	-	-	-	-	-	-	-	-	-
HR	-	-	-	-	-	-	-	-	-	-	-	-
HU	13	-67,6	-41,0	14	-42,3	-22,3	14	-20,1	-31,0	-	-	-
IE	-	-	-	-	-	-	-	-	-	-	-	-

	SOx/SO_2			NOx/NO_2			PPM_{10}/PM_{10} ([a])			$PPM_{2.5}/PM_{2.5}$ ([b])		
Country	Nsta	Emis	Conc	Nsta	Emis	Conc	Nsta	Emis	Conc	Nsta	Emis	Conc
IT	42	-92,5	-57,3	171	-57,9	-44,3	106	-37,1	-34,9	54	-37,4	-34,7
LT	-	-	-	-	-	-	-	-	-	-	-	-
LU	-	-	-	-	-	-	-	-	-	-	-	-
LV	-	-	-	-	-	-	-	-	-	-	-	-
MT	-	-	-	-	-	-	-	-	-	-	-	-
NL	-	-	-	31	-57,5	-39,4	20	-38,6	-46,7	-	-	-
PL	54	-70,4	-66,8	53	-33,6	-20,3	55	-14,0	-26,0	26	-10,0	-47,8
PT	-	-	-	15	-54,3	-45,8	16	-28,6	-39,9	-	-	-
RO	-	-	-	-	-	-	-	-	-	-	-	-
SE	-	-	-	6	-42,4	-44,3	5	-34,0	-41,9	-	-	-
SI	-	-	-	-	-	-	-	-	-	-	-	-
SK	-	-	-	-	-	-	14	-43,8	-42,6	-	-	-
EU27	425	-75,6	-60,5	994	-55,6	-38,4	833	-35,6	-42,2	293	-40,8	-44,8

Footnotes ([a]) and ([b]): PPM_{10} and $PPM_{2.5}$ refer to the primary emissions in the PM_{10} and $PM_{2.5}$ fraction, respectively.

The sharp decrease in emissions after 2007 is mainly due to reductions in emissions from the industrial sector and the energy production and distribution sector in the lee of the economic downturn of 2008 (EEA 2018). No such decrease is observed at traffic sites. At stations of background type (urban, suburban and rural), a decrease is noted in 2007 but followed by punctual and small concentration increases in 2010, 2012, and 2017 (Figure 4.1). The decrease in 2007 was observed at industrial sites but it was still above the corresponding level in reported emissions inventories.

There are also some noteworthy differences depending on the countries (Table 4.1). Whereas the observed rate of decline in SO_2 observations is slower than the decline of emissions in Czechia, Bulgaria, Italy, and Hungary, the opposite is found for Austria, Belgium and Germany. As previously mentioned, it's important to note that in Table 4.1, Nsta is presented exclusively for the background typology. However, in Figure 4.1, Nsta values are shown for all typologies, with countries selected based on the availability of more than 5 stations for any typology. This requirement similarly applies to the subsequent related figures and tables.

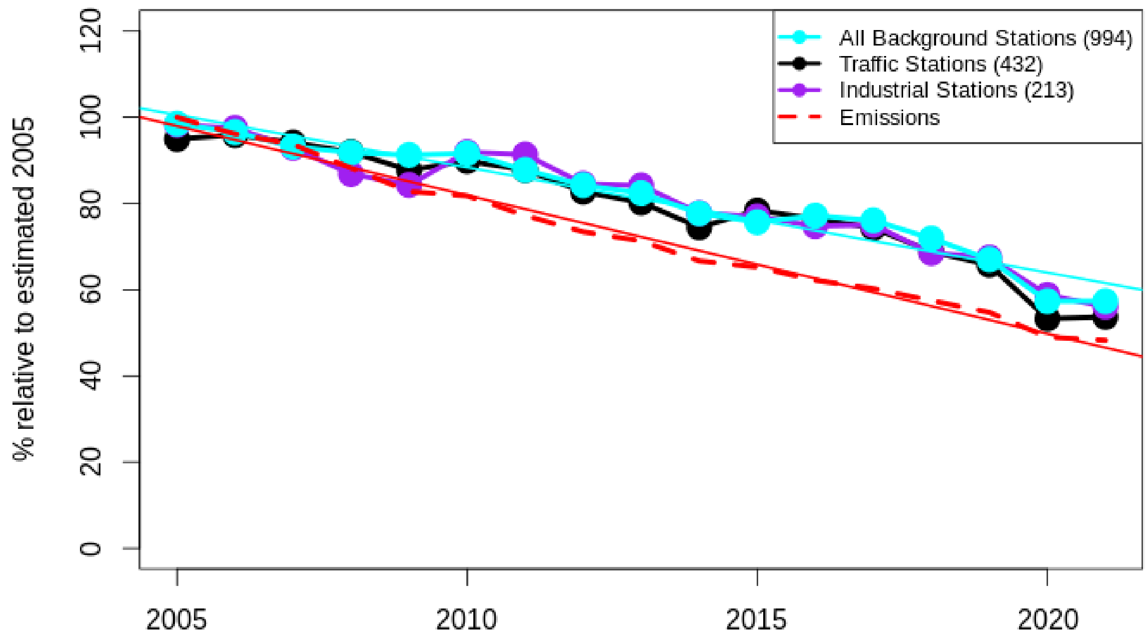
It is not possible to investigate further this issue with the material available here because we ignore the effect of the transport and transformation of SO_2 in the atmosphere and comparing nation-wide emission and point observations raise spatial representativeness issues. Nevertheless, this discrepancy should trigger follow-up works to understand why the 2008 economic downturn seemed to have had a smaller impact on observed SO_2 concentration trends compared to reported emissions.

4.1.2 Nitrogen dioxide

The comparison between the trends in NOx emissions and NO_2 observation is presented in Figure 4.2 and the corresponding numbers are in Table 4.1 above. As mentioned in Section 4.1.1, this comparison must be handled with care, and Chemistry-Transport Model results should be included to be more conclusive.

Again, the agreement between emission and concentration trends was quite good up to 2008, but from 2009 the mismatch becomes clear for all station types. A strong consistency between NO_2 concentration trends monitored at background, traffic and industrial sites can be noted. The mismatch between NOx emissions and NO_2 concentrations is quite systematic over European countries with enough measurement sites, so that the comparison over EU27 points out a disagreement: 56 % reduction in emissions, whereas NO_2 concentrations only decreased by 38 % (see Table 4.1). Significant variations exist among countries like Germany, Hungary, Sweden, and Poland, which reported emission trends ranging from 30 % to 42 %, and other nations where emission trends fall more consistently within the 48 % to 60 % range. This finding raises some concerns regarding the level of ambition that policy measures should achieve to deal with high NO_2 levels still observed in major cities.

Figure 4.2: Time series of country median NO_2 observed at traffic (black), industrial (violet) and background (cyan) sites (solid lines), and corresponding national NOx emissions (dashed line) normalised to estimated 2005 levels (%). The median is taken over countries where more than 5 stations of at least one typology is available. The total number of stations is provided in brackets. Straight lines (red for emissions and cyan for all background stations concentrations) are the linear fits over the whole period



4.1.3 Ozone

The relative evolution of ozone concentration in Europe is put in perspective with emissions of its precursor in Table 4.2. A more in-depth analysis of the effectiveness of emission mitigation policies in reducing ozone levels in Europe would require the implementation of regional and global chemistry-transport models to take into account the effect of meteorology (including its effect on biogenic emissions), intercontinental transport, but also long-lived precursors such as methane. Nevertheless, this table allows pointing out the important gap between the rate of change of the emission of ozone precursors, and the observed ozone evolution. Considering only the countries with more than 5 ozone stations of any typology, the emissions of anthropogenic NMVOC and NO_x decreased in EU-27 over 2005-2021 by 33 % and 56 %, respectively. At the same time, annual mean ozone (ConcAVG) increased by 3 % and the peaks (ConcMDA8) decreased, but only by 8 % (these changes are given for background stations for synthesis purposes in Table 4.2, but the details per typology is given in Table 3.3. This gap is systematic for the European countries included in Table 4.2 and raises legitimate questions in relation to the effectiveness of air quality policies to deal with ozone issues in the European countries. This is essentially because European anthropogenic emissions only contribute to ozone formation in excess to a baseline constituted of natural ozone (from soil and biogenic emissions) and hemispheric background. This baseline is substantial and can be about 80µg/m^3 in summer. Assessing policy effectiveness in mitigating ozone would require focusing on the ozone concentration in excess of that baseline, which would therefore need to be computed more precisely.

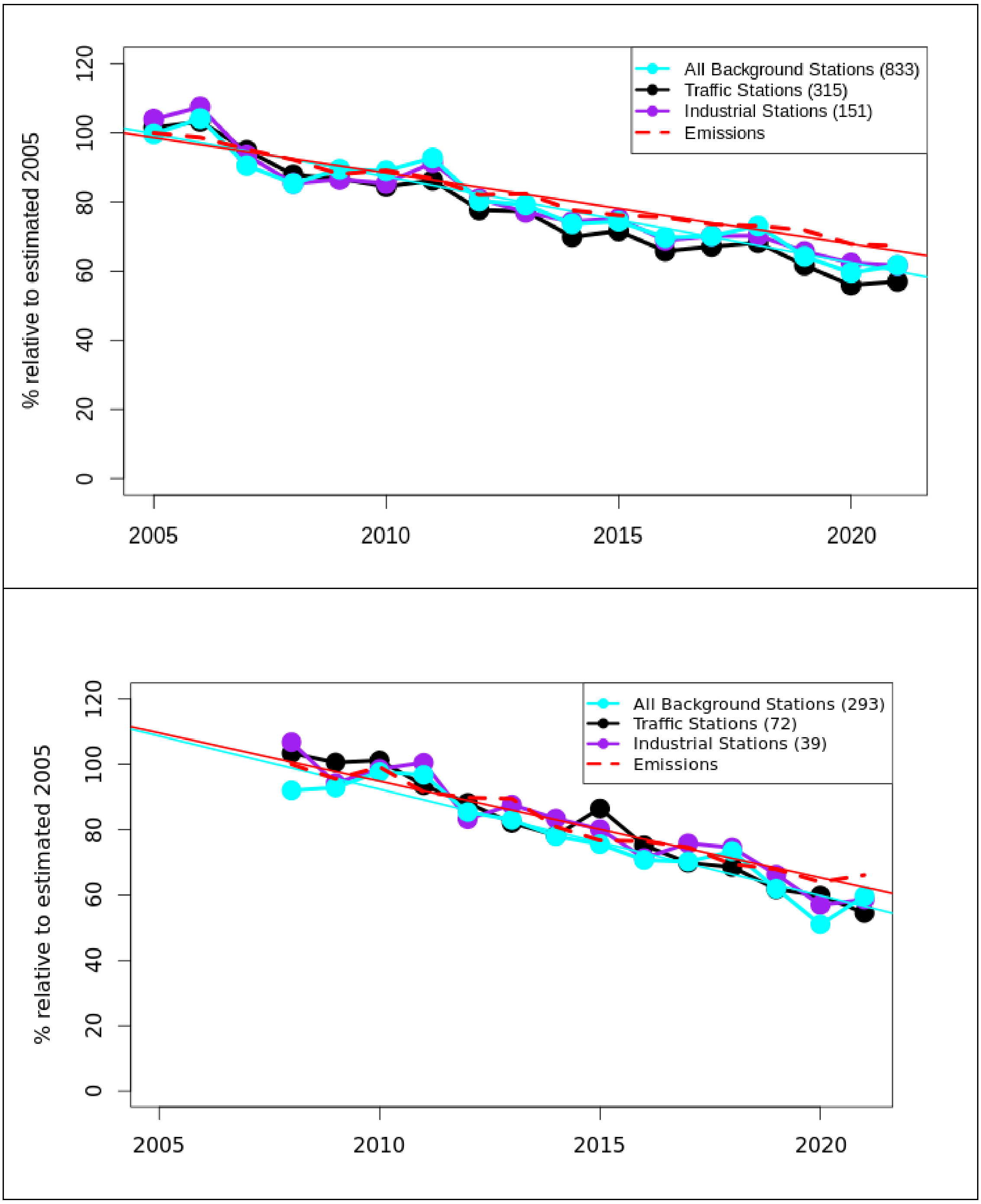
Table 4.2: Change, relative to 2005 (in %), for emissions of anthropogenic NOx and NMVOC and concentrations of ozone annual mean and peaks, as median over countries with more than 5 stations background of either urban, suburban or rural (Nsta, otherwise indicated as "-")

Country	Nsta O3	Emis NOx	Emis NMVOC	Conc AVG	Conc 4MD8
AT	85	-48,3	-39,5	2,0	-10,8
BE	30	-60,4	-39,1	23,5	-1,6
BG	11	-56,7	-24,1	-2,5	-20,1
CY	-	-	-	-	-
CZ	40	-51,9	-33,9	1,3	-10,1
DE	196	-36,1	-33,7	7,8	-4,5
DK	-	-	-	-	-
EE	-	-	-	-	-
ES	146	-53,0	-23,5	3,6	-9,4
FI	9	-55,0	-46,2	-3,9	-13,6
FR	227	-54,6	-37,0	12,3	-6,2
GR	-	-	-	-	-
HR	-	-	-	-	-
HU	15	-42,3	-26,0	-4,7	-12,4
IE	-	-	-	-	-
IT	139	-57,9	-42,6	4,8	-10,3
LT	-	-	-	-	-
LU	-	-	-	-	-
LV	-	-	-	-	-
MT	-	-	-	-	-
NL	23	-57,5	-12,3	18,0	3,0
PL	40	-33,6	-14,8	-3,5	-11,8
PT	26	-54,3	-16,5	0,9	-17,4
RO	-	-	-	-	-
SE	13	-42,4	-39,0	3,8	-8,0
SI	-	-	-	-	-
SK	-	-	-	-	-
EU27	1000	-55,6	-33,4	3,0	-8,1

4.1.4 Particulate Matter

The comparison between emissions and observation, aggregated over EU27 level, displays a good correlation (Figure 4.3). As for SO_2 and NO_2, this comparison remains very qualitative until chemistry-transport models are involved for a more detailed assessment. Over countries where the network is dense enough (more than 5 background stations of either urban, suburban, or rural typology), PM_{10} and $PM_{2.5}$ concentrations decreased respectively by 42 % and 45 % (Table 4.1), whereas the corresponding changes in primary emissions were 36 % and 41 %. The decreases in observed concentrations, particularly for PM_{10}, are more pronounced, as expected due to the significance of secondary aerosols, which are mitigated by reducing precursors other than primary PM. This gap between emission and concentration trends is the only conclusion to be drawn from this Table. The difference between PM_{10} and $PM_{2.5}$ trends shall not be further interpreted on the basis of these figures as this comparison is not for the same time period (2005-2021 is used for PM_{10}, and 2008-2021 is used for $PM_{2.5}$) for collocated measurements for which the reader is referred to Map 3.5.

Figure 4.3: Aggregated time series over EU27 of country median PM_{10} (top) and $PM_{2.5}$ (bottom) observed at traffic (black), industrial (violet) and background (cyan) sites (solid lines), and corresponding country PPM_{10} and $PPM_{2.5}$ emissions (dashed line) normalised to estimated 2005 levels. The total number of stations is provided in brackets. Straight lines are the linear fit over the whole period



4.2 Clustering air quality trends at stations

Given the multitude of clustering scenarios involving different pollutants, countries, and typologies, we have opted to present only a limited number of cases. Those cases were selected to be as illustrative as possible of the various possible situations. The remaining clustering scenarios are visually represented and briefly described in the accompanying supplementary materials (see. Annex 3 and 4). The rationale behind showcasing only a limited number of cases in the main text is to offer a straightforward approach for analyzing clustering to identify exceptional stations. We anticipate that a similar analysis will be carried out for the clustering scenarios presented in the supporting documents, covering other pollutants, countries, and typologies. Also note that if certain clustering scenarios are absent from the annexes, it signifies that the criteria for forming clusters (involving more than 10 stations) were not met.

4.2.1 Europe-wide clustering

When applying the clustering methodology presented in Section 2.5 on the whole of European urban, suburban, and rural background stations of NO_2, we include in the clustering features the relative trend in emission reported for the corresponding country in addition to the concentrations in 2005 and the relative decrease in concentrations. The results are displayed in Figure 4.4.

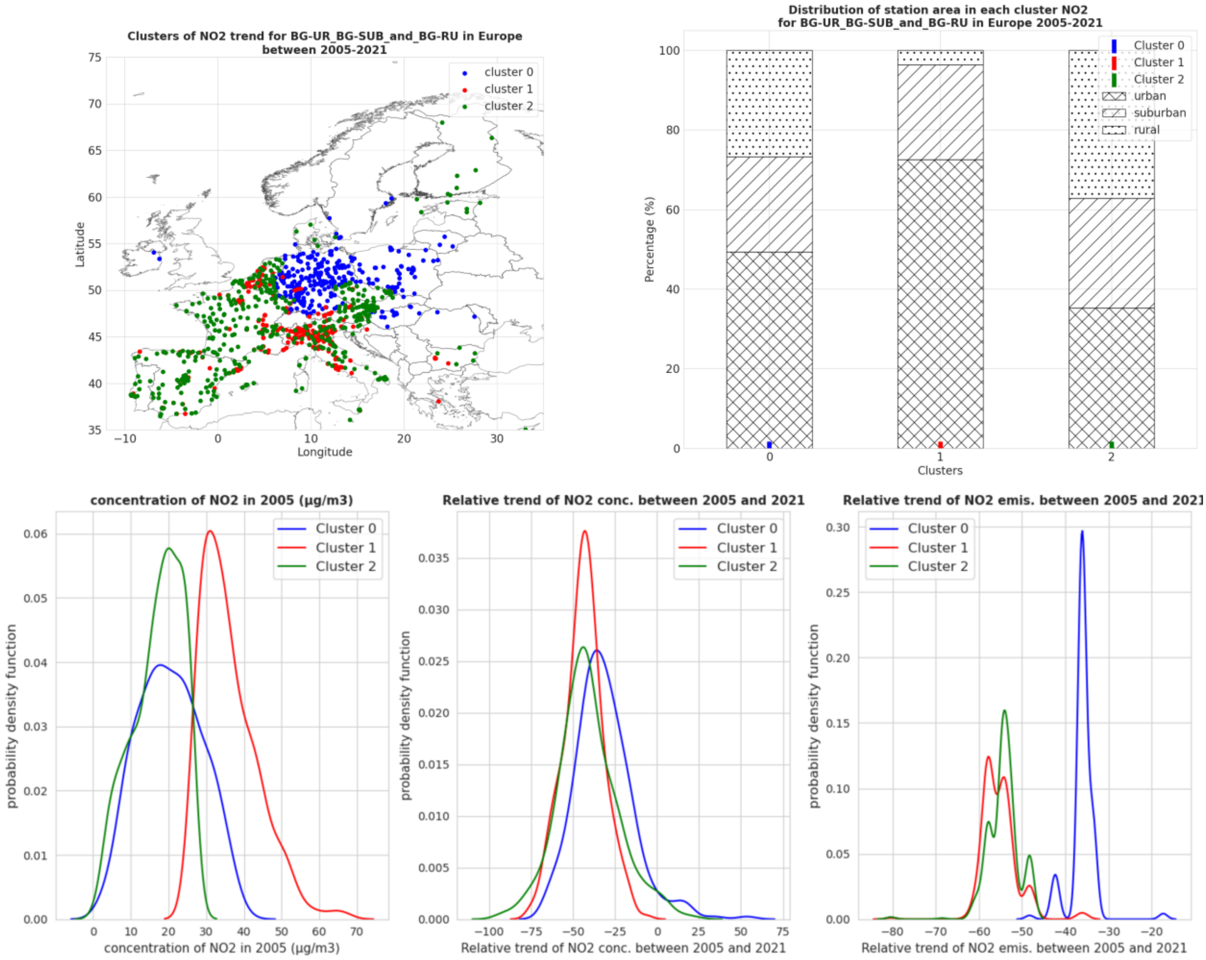
The relative decline in concentrations is quite similar for all clusters (middle plot in the second row of Figure 4.4), it does not appear in this case to be the main feature driving the clustering.

We find two clusters (1 and 2) dominating countries such as Spain, France, Benelux, Italy, Austria and Czechia. Cluster 1 is dominated by urban stations, it had therefore higher concentrations in 2005, but the relative declines in concentrations and emissions are in line with cluster 2. There is therefore not much ground for further interpretation in analyzing the difference between clusters 1 and 2 as we merely disentangled urban and other typologies of stations.

A more interesting feature appears by considering cluster 0. It had very similar levels in 2005 as cluster 2, and again somewhat similar decreases in NO_2 concentrations. But the distribution of relative decline in emissions is notably different. We find again here the discussion of Table 4.1 where a few countries reported declines in emissions of about 30-42 % (DE, HU, PL, SE), while others reported much larger declines between 48-60 %. And at the same time, all these countries witnessed a similar rate of decline in ambient concentrations of NO_2.

To further understand these discrepancies (e.g. within cluster 0, 1 and 2), further analysis of the national reporting methodologies used in countries belonging to either cluster 0 or to one of the other two clusters (1 and 2) would be required to understand why the reported NO_2 emissions trends are so different.

Figure 4.4: Clustering of NO_2 trends applied to urban, suburban, and rural background stations in Europe. Top left: map of stations falling in classes 0,1,2. Top right: distribution (%) of stations of either urban, suburban, and rural background typology in each of the three classes. Bottom, from left to right: distribution of NO_2 concentrations estimated in 2005 at stations belonging to each of the three classes, distribution of the relative trends of NO_2 concentrations between 2005 and 2021 at stations belonging to each of the three classes, distribution of the relative trends of NOx emissions between 2005 and 2021 in the country corresponding to stations of the three classes



4.2.2 Clustering applied for each country

The clustering methodology developed for the whole of Europe is replicated for individual countries. Only countries with more than 10 stations of background rural, suburban, or urban types are included. Here we present a discussion on how the analysis can be performed for selected countries, pollutants, and station typologies, but a similar reasoning can be applied for all other countries. In all cases, the number of classes imposed in the clustering is 3, to facilitate the comparison between countries. While setting a varying number of clusters might capture more nuanced variations in pollutant distributions, it could complicate cross-country comparisons and the interpretation of pollutants patterns for stakeholders.

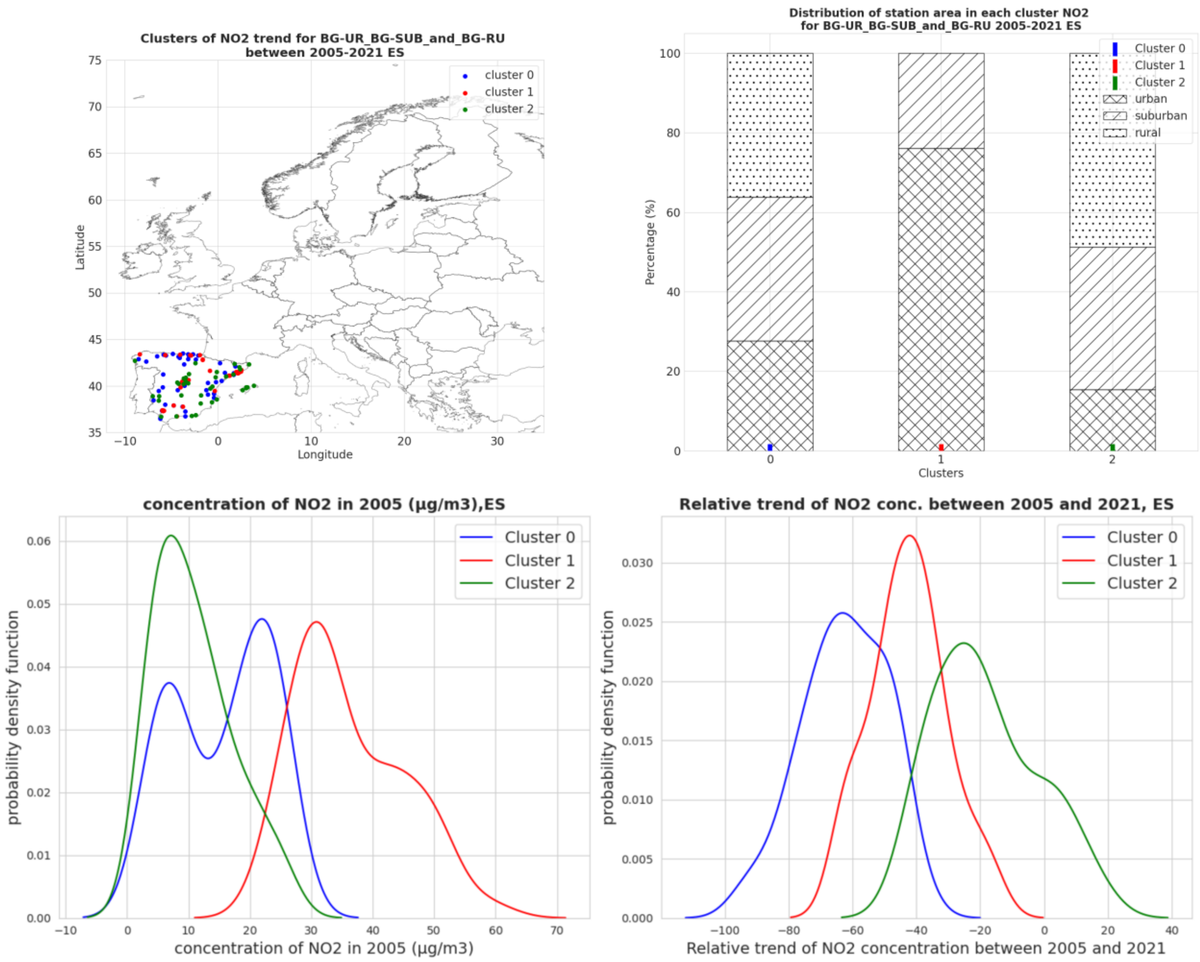
Clustering NO_2 at Background stations in Spain

The results for background NO_2 in Spain are presented in Figure 4.5. In the map in the top right panel, we can notice that the three classes are evenly distributed over the Spanish territory, meaning that each of those classes are represented in all regions of Spain. There is thus no specific geographical feature identified. In the distribution of station typology in the top right, the number of urban, suburban and rural background stations in classes 0 and 2 are similar. On the contrary class 1 is clearly dominated by urban stations, and not a single rural station is included in that class.

The most important distinction is assessed from the probability density functions displayed in the lower two panels. Cluster 1 (dominated by urban sites) is the one where NO_2 concentrations are the highest at the beginning of the period (bottom left), which is precisely because it is dominated by urban sites. The relative NO_2 trends within that class are in the middle of the distribution compared to the other two clusters. On the contrary, the values of NO_2 concentrations estimated for 2005 are within the same range in clusters 0 and cluster 2 (although a 2-peak distribution occurs for cluster 0). But what differentiates cluster 0 and cluster 2 is the relative trends which are notably larger in cluster 0 (centered about -60 %), than in cluster 2 (centered around -20 %).

For that discussion, the specificity which should be further investigated in the case of NO_2 background stations in Spain is: what differentiates the stations in cluster 0 and 2? They appear to have a similar proportion of urban, suburban and rural sites, and they are also evenly distributed geographically. Would it be possible to relate this strong discrepancy in NO_2 trends over the period to any specific plans that would have been introduced in the stations belonging to cluster 0 to explain that the relative trends in concentrations are so much larger than in cluster 2?

Figure 4.5: Clustering of NO_2 trends applied to urban, suburban, and rural background stations in Spain. Top left: map of stations falling in classes 0,1,2. Top right: distribution (%) of stations of either urban, suburban, and rural background typology in each of the three classes. Bottom left distribution of NO_2 concentrations estimated in 2005 at stations belonging to each of the three classes. Bottom right: distribution of the relative trends of NO_2 concentrations between 2005 and 2021 at stations belonging to each of the three classes

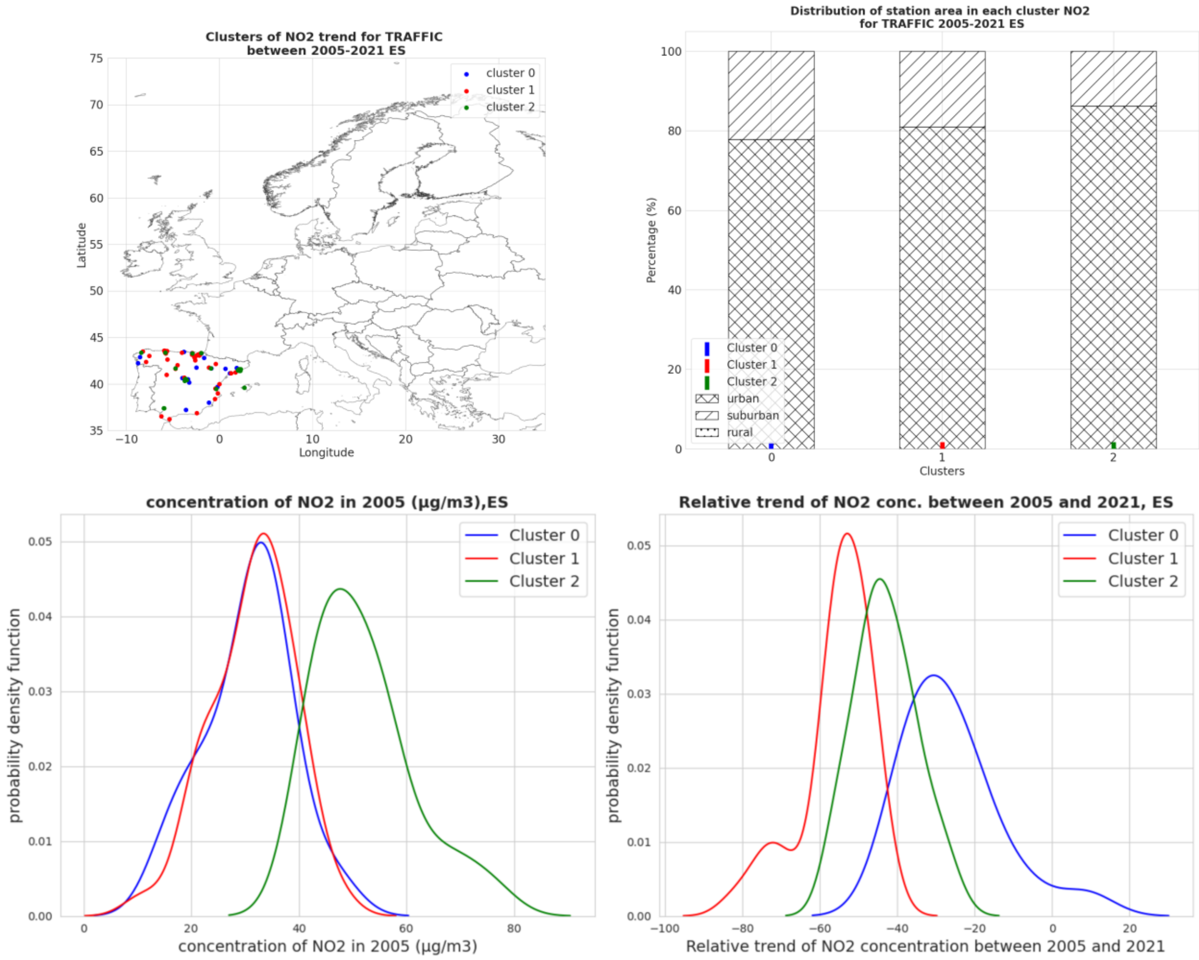


Clustering NO_2 at traffic stations in Spain

A similar analysis is repeated for traffic stations in Spain (Figure 4.6). Here the distributions of station types are quite evenly distributed between urban and suburban traffic sites. In cluster 2, the concentrations were much higher than in the other two in 2005 suggesting they could be located closer to major roads. The relative trends in that cluster fall in the middle of the distribution.

Here the specificity which would deserve further investigation in relation to local measures would be to analyze the list of stations falling in cluster 0 and 1: the values of concentrations where very similar in 2005 inferring somewhat similar traffic conditions. But the relative trends are very different: in cluster 1, the distribution is centered on a 50 % relative decrease over the period, while in cluster 0, the decrease in the middle of the distribution is only 30 %. The question is therefore to find out if any specific plans were introduced in the areas where stations in cluster 1 are located.

Figure 4.6: Same as Figure 4.5, but for traffic stations and NO_2 in Spain

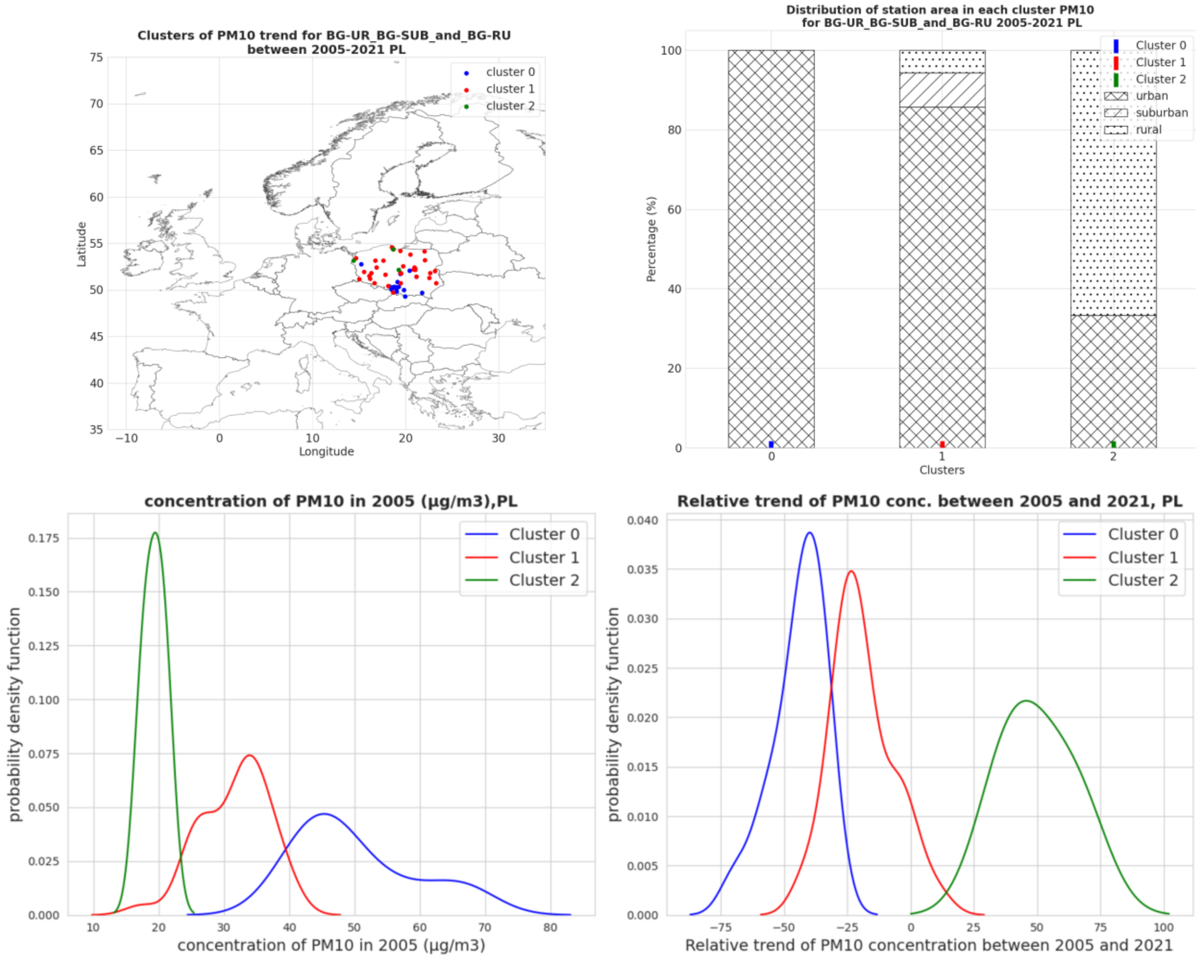


Clustering PM_{10} background stations in Poland

For the cluster of PM_{10} at urban, suburban and rural sites, we selected a case study for the Polish stations (Figure 4.7:). Here cluster 2 is dominated by rural stations, but it should be noted that it is constituted of very few stations. The concentrations in 2005 were relatively low, which is expected because of the larger fraction of rural sites. But the most peculiar feature is that relative trends actually increased in those areas. This increase could be further investigated with the national experts, for instance to inquire whether the influence of nearby pollution source might have changed in time.

Cluster 0 is constituted exclusively of urban sites, while cluster 1 is largely dominated by this typology but also include a couple of other station types. The concentrations were highest in cluster 0 at the beginning of the period, and it is also the stations for which the relative declines where largest (centered about -40 % against -25 % for cluster 1). It would be interesting to confirm if stations in cluster 0 were located in air quality zones targeted by specific mitigation plans compared to cluster 1.

Figure 4.7: Same as Figure 4.5, but for PM_{10} urban, suburban and rural background stations in Poland

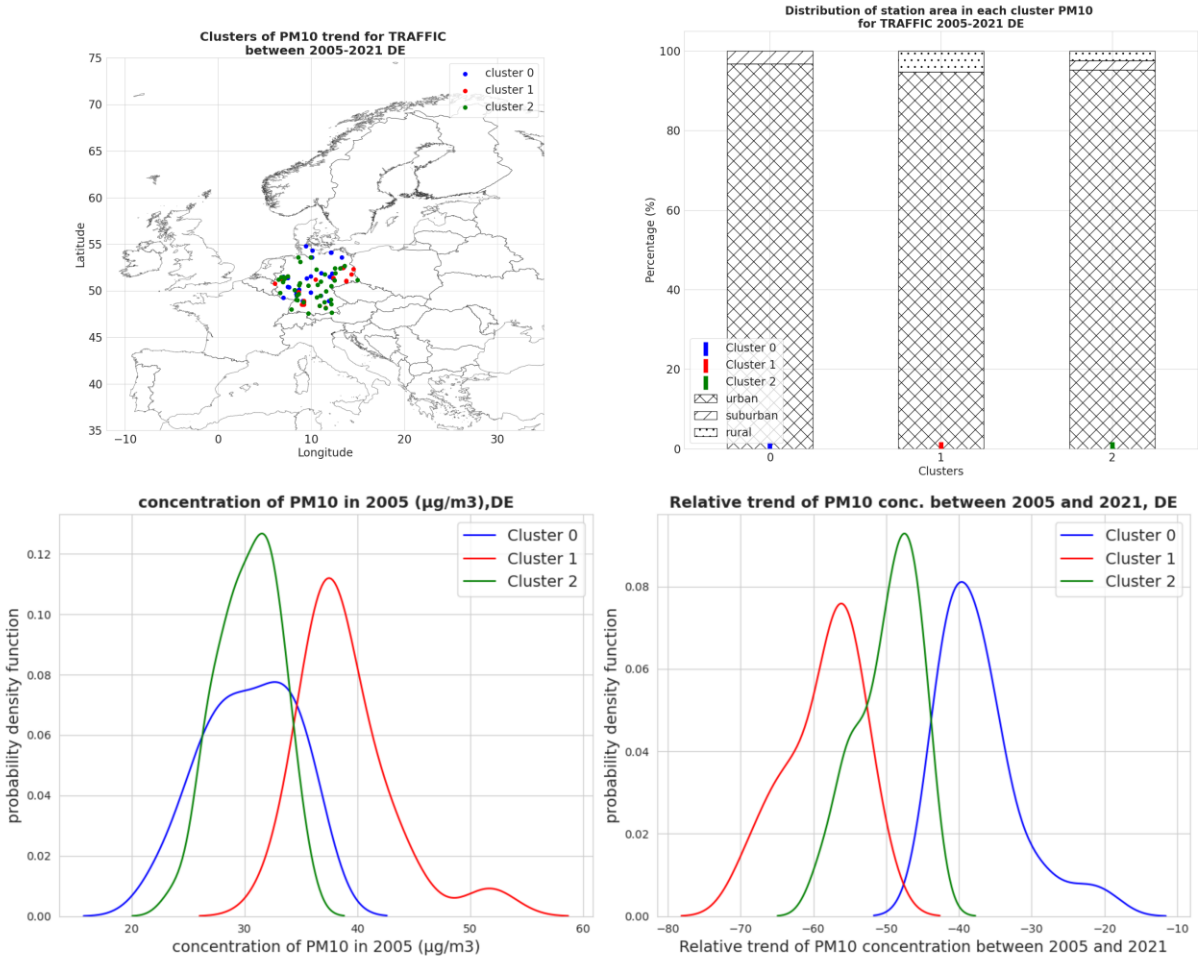


Clustering PM_{10} at traffic stations in Germany

Another illustration is provided in Figure 4.8 for the trend of PM_{10} at traffic stations. Here we picked Germany to highlight the relevant clustering information. As expected, traffic stations are predominantly found at urban areas for all classes. The stations where the largest concentrations were found are within cluster 1, which is also the cluster where the strongest relative declines are found. The question is therefore: is it only because such stations are located to major roads or large metropolitan areas, or can we also relate them to specific local plans.

Cluster 0 and 2 are also worth investigating, while the levels were very similar in 2005, the relative trends are very different: while in cluster 0, the decreases are centered on -40 %, they are centered on -50 % and sometimes reach much larger declines in cluster 2.

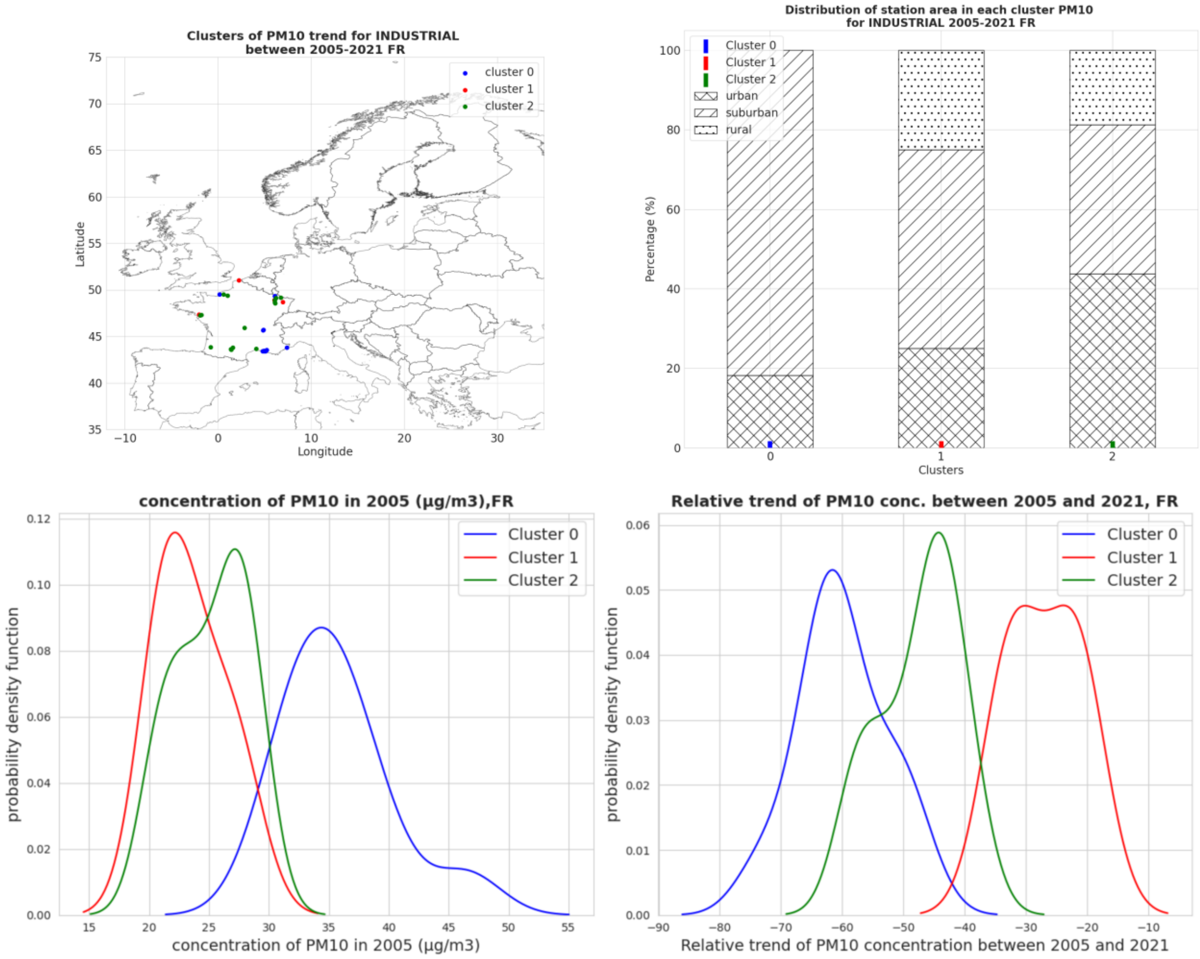
Figure 4.8: Same as Figure 4.5, but for PM_{10} traffic stations in Germany



Clustering PM_{10} at industrial stations in France

The example for PM_{10} trends at industrial stations is based on France (Figure 4.9). Most industrial sites are located in suburban or urban environments. Cluster 0 is dominated by stations where concentrations were high in 2005, and the relative trends are also strongly decreasing. The specificity which would be worth investigating in priority is why the trends in stations at cluster 1 are only about -25 %, while in cluster 2 they are centered on -50 % to -40 %, whereas the concentrations of PM_{10} at those sites were very similar in 2005.

Figure 4.9: Same as Figure 4.5, but for PM_{10} at Industrial sites in France



Clustering results for all pollutants, stations typology, and countries

The results for the other scenarios of clustering by country or over Europe are provided in supplementary material of this report for the periods 2005-2021 as well as 2000-2021 (see. Annex 3 and 4). This includes graphical clustering of all possible scenarios presented as a 'pdf' file attachment and the associated 'CSV' files. Each 'CSV' file contains cluster IDs along with corresponding metadata for identifying the station and feature data used for clustering. For the countries, each file is named in the following format: "clustering_<<pollutant name>>_<<station typology>>_<<trend period>>_<<country>>.csv". For country-wide clustering, only the countries with more than 10 stations in each typology are selected for the clustering. Over Europe, the file format is simply "clustering_<<pollutant name>>_<<station typology>>_<<trend period>>.csv". The variables 'var_fit_bgn', 'var_str', and 'emis_str' in the "csv" files correspond, respectively, to the concentration estimated in 2005 (or 2000), the relative trend of concentration between 2005 (or 2000) and 2021, and the relative trend of emissions between 2005 (or 2000) and 2021. For PM2.5, the relative trend of concentration and emissions starts from 2008.

5 Summary and conclusions

Detailed trend calculations of air pollutants for the periods 2005-2021 based on a descriptive statistics and machine learning methods have been performed. We find that the trends of sulphur dioxide present the largest decrease of all pollutants. The decrease is such that at industrial sites, the levels in 2021 are comparable to that of rural sites at the beginning of the period (2005). A reduction in concentrations of the order of 62-68 % (depending on the station typologies: urban, suburban, and rural background sites, as well as traffic and industrial) is found. At all background level, emissions of SO_2 decreased by 76 %, and the recorded concentrations showed a reduction of 61 %, which is a mismatch. However, the agreement between reported emission data and measured concentrations are relatively good up to the year 2008.

For NO_2, a mismatch between the trend in air concentrations and NO_x emissions is found. While the overall NO_x emissions are reported to be reduced by around 55 % over the period, the measured NO_2 data indicate a decline of around only 38 %. Differences between countries are found, though, with better agreement between emissions and measurements in some areas (typically Germany, Hungary, Sweden and Poland), which suggest a potential influence of the reporting methodology used in those countries. The relative changes in NO_2 are found to be largest for the lower/medium percentiles whereas smaller reductions are found for the highest peaks.

For O_3, our findings are in line with earlier studies. During the period 2005-2021, at background sites the annual mean ozone concentration has slightly increased between 2 % and 11 % while the high peaks have been reduced (by 6 % to 9 %). The increase in mean levels is explained by hemispheric transport and reduced titration by NO because of reduced NO_x levels. The reduction in high ozone peaks is expected when the general NO_x level in Europe is reduced.

For PM data (PM_{10} and $PM_{2.5}$) we find an opposite mismatch, meaning that the PM concentrations show stronger downward trends than the reported emissions. This is likely an effect of the importance of secondary aerosols which are mitigated by other activities than the direct PM emissions, and thus leading to larger reductions than the primary emissions alone. Depending on statistical approach, station type etc, we estimate an overall reduction in PM_{10} about 42 % at background sites during 2005-2021. The rate of concentration is larger than that of primary PM_{10} emissions which were reduced by about 36 %. A similar difference is found between concentration and emission in the $PM_{2.5}$ fraction. This difference is attributed to the additional benefit of reducing other precursors of secondary PM.

The evolution of air quality in Europe is assessed using an adaptation of the EEA air quality index (AQI) methodology, categorizing pollutant concentrations into six ranges. Despite limited stations covering all pollutants over a long period, a median index is computed for each country and EU27, revealing a slight improvement in overall air quality. While NO_2, PM_{10}, and $PM_{2.5}$ show increased "good" days, ozone's distribution remains consistent, and SO_2 already had good daily categories. The station network's long-term evolution indicates no significant sampling biases, except for the PM monitoring development which might have led to an underestimation of PM air pollution over the first part of the period.

A specific analysis was undertaken to identify the main drivers of air pollution changes in Europe for the main air pollutants. Besides the comparison with emission reported at national scale, we also conduct a clustering analysis for European urban, suburban, rural, traffic, and industrial monitoring stations considering explanatory factors such as the concentrations at the beginning of the period (2005) and the relative concentration trends.

We present selected case studies of such clustering analysis for various types of air pollutant and monitoring stations in Spain, Poland, Germany, and France. In each case we were able to identify a subset of stations with specific features in terms of long-term evolution which could not be explained by their geographic localisation or the station typology. Such outstanding groups of stations are provided for each pollutant and each country in supplementary material of this report. It will serve as a basis for a follow up analysis aiming at relating those clusters to the implementation of local air quality plans in European Member States.

6 List of abbreviations

Abbreviation	Name	Reference
AOT40	accumulated ozone over 40 ppbv	
EEA	European Environment Agency	www.eea.europa.eu
INERIS	Environnement Industriel et des Risques	
O_3	Ozone	
NILU	Norwegian Institute for Air Research	
NO_2	Nitrogen dioxide	
$PM_{2.5}$	Particulate Matter less than 2.5 micrometers	
PM_{10}	Particulate Matter less than 10 micrometers	
SOMO10	sum of ozone daily maxima in excess of 10 ppbv	
SOMO35	sum of ozone daily maxima in excess of 35 ppbv	
SO_2	Sulfur dioxide	
4MDA8	fourth highest daily peak	

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Annex 1: National trends in air concentrations as calculated from in-situ measurements 2005-2021

[See separate pdf attachment].

Annex 2: National trends in air concentrations as calculated from in-situ measurements 2000-2021

[See separate pdf attachment].

Annex 3: Clustering of air quality trends 2005-2021

[See separate pdf and zipped csv file attachments named CSV 2005-2021].

Annex 4: Clustering of air quality trends 2000-2021

[See separate pdf for Annex 4 and zipped csv file attachments named CSV 2000-2021].

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