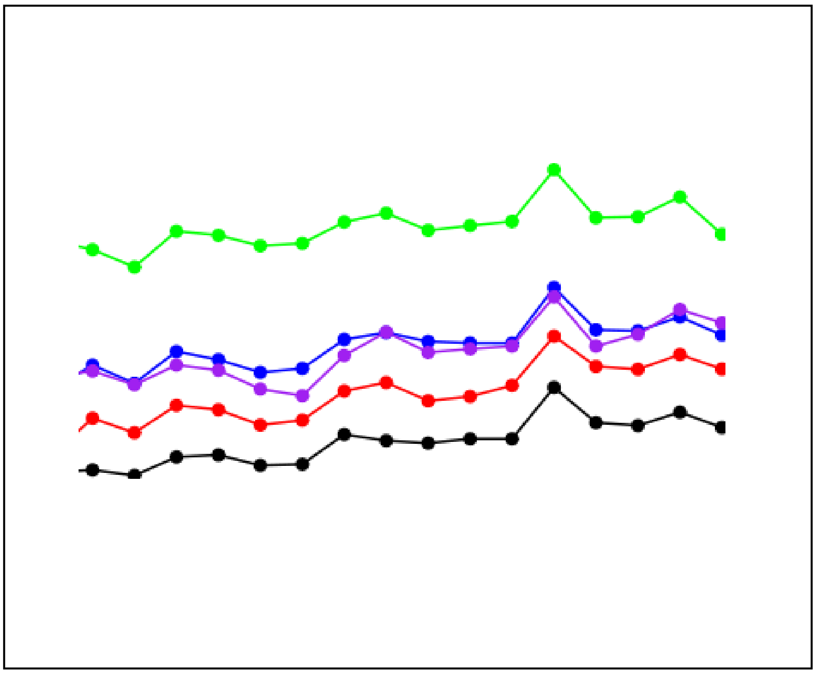
Air Quality Trends in AIRBASE

in the context of the LRTAP Convention



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European Topic Centre on Air Pollution and Climate Change Mitigation

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Front page picture:
Schematic of ozone trends at various European sites

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Abstract

The airbase repository is used for an assessment of air quality trends over the past 20years. After having developed and applied an automated geostatistical outlier detection algorithm for background rural, suburban and urban sites and checked record completion criteria for all background stations, as well as industrial and traffic stations, various trend diagnostics are computed and analysed in the present report.

For nitrogen dioxide:

- Concentrations are steadily decreasing over Europe, at an average pace of 42, 44, 48% at urban, suburban and rural background sites over 1990-2012, which is in good agreement with the 51% reductions of European emissions between 1990 and 2012;
- The rate of change was larger in the 1990's compared to the 2000's;
- A qualitative analysis indicates that the spatial variability of the trends over the last decade does not exhibit any particular pattern;
- At stations closer to emission sources, and in winter, NO_2 decreases are less pronounced because in such conditions, background ozone (which did not change much) plays an important role;
- At background stations NO_2 decreased faster in urban and suburban sites, than over rural areas.

For ozone:

- There was an increase in annual mean ozone over the 1990's followed by a flattening of the trend over recent years;
- For ozone peaks, a somewhat flat trend was already observed over the 1990's, with a slight decrease over the 2000's;
- The slight decrease of ozone peaks in the 2000's is quite spatially homogeneous over Europe;
- Ozone trends tend to be frequently more positive at urban background sites than rural sites for both ozone annual average and summertime peaks and for both 1990's and 2000's time period because of the titration effect induced by NOx emission reduction;
- Increase in hemispheric ozone levels could also influence the flatter trends for annual mean ozone, as suggested by the increase in springtime ozone.

For particulate matter:

- The completeness of the record only allow for a trend assessment of PM10, in the second part of the period (the 2000s);
- The average rate of decrease in relative terms is of the order of 30% for background sites in the 2000s, with a limited spatial variability;
- The trend is more pronounced close to the sources (traffic, industrial and urban background sites) than in rural areas and in summer compared to winter

1 Introduction

The amount and quality of the air quality data included in the Airbase database has increased substantially over the recent years, making it a precious source of information for air quality assessments in Europe. Amongst such assessment we find trend analyses that are instrumental in documenting the efficiency of air quality legislation over recent years.

The Geneva Convention on Long Range Transboundary Air Pollution is presently undertaking an Assessment of the efficiency of the 1999 Gothenburg Protocol, which used as a reference year 1990. More specifically, the Task Force on Measurement and Modelling (TFMM) is performing a comprehensive 20 year Trend Assessment, which will ultimately lead to the publication of a dedicated report, in early 2016. It is expected that this report will provide an overview of observed trends in the key compounds targeted by the Gothenburg Protocol together with selected experiments involving numerical models to assess the relative contribution to the net trends of (i) European emissions of precursors, (ii) long range transport and (iii) meteorological variability.

Given that the TFMM will primarily focus on trends observed at EMEP stations which are located in relatively uncontaminated rural background locations, the EEA proposed a complementary contribution based on the Airbase dataset that would also include trends observed at different type of station (background, or in the proximity of traffic or industrial sources), and different type of area (rural, suburban, or urban).

Acknowledging that the data included in Airbase originate mostly from regulatory monitoring networks (mainly to comply with the monitoring obligations in the Air Quality Directives) that are not specifically designed for trend assessments, a prior selection of appropriate sites was performed. The details of this selection are presented in an earlier 2015 ETC/ACM Working Paper[1], the rationale relies on (i) data completeness criteria (75% of the considered time period in the given year, and 75% of the years for the considered temporal span), (ii) automated detection of outliers using a geostatistical algorithm. For background stations both filters are applied, while for traffic and industrial sites, the geostatistical approach is irrelevant due to their lack of spatial covariance, and only the criteria (i) on data completeness is used. Note that a minimum annual coverage of 85% for each year was used in the Working Paper, hence the slight differences here.

The total number of stations passing the annual completeness criteria (before and after outlier detection using the geostatistical algorithm for background sites) are displayed in Figure 1. As explained in the earlier 2015 Working Paper, the outlier detection algorithm is more sensitive to the chemical compounds with less spatial variability (i.e. O_3 and PM10 compared to NO_2). The turn of the century saw a dramatic increase of the network coverage, especially for PM10 so that we will exclude the 1990s from the present analysis for this compound. We

[1] Screening procedures to evaluate the quality and consistency of measurement time series in Airbase+ for trend analysis, A. Colette, M. Beauchamp, L. Malherbe, 2015, http://forum.eionet.europa.eu/etc-acm-consortium/library/subvention-2015/task-deliveries-ap2015/task-1121-air-quality-report/b.-final-drafts-approval-eea/wp-station-selection-trend-assessment

will also be cautious with the comparison between trends in the 1990s and in the 2000s for O_3 and NO_2.

The statistical method applied for the trend detection is Mann-Kendall (with a p-value of 0.05) and we compute the actual slope using the Sen-Theil approach. Both techniques differ from the more classical least square regression in the fact that they focus on the distribution of pairs of changes, aggregating their sign for Mann-Kendall, or using the median of differences for Sen-Theil. They are thus less sensitive to outliers, but also to autocorrelation, hence their widespread use in trend assessments. Furthermore, these methods don't require that the original data follow a normal distribution.

The purpose of the present working paper is to discuss the key features of the air quality trend assessment that was performed on the basis of the Airbase data repository. It is organised by compound, so that NO_2 trends are presented in Section 2, ozone trends are presented in Section 3, and PM10 trends are presented in Section 4.

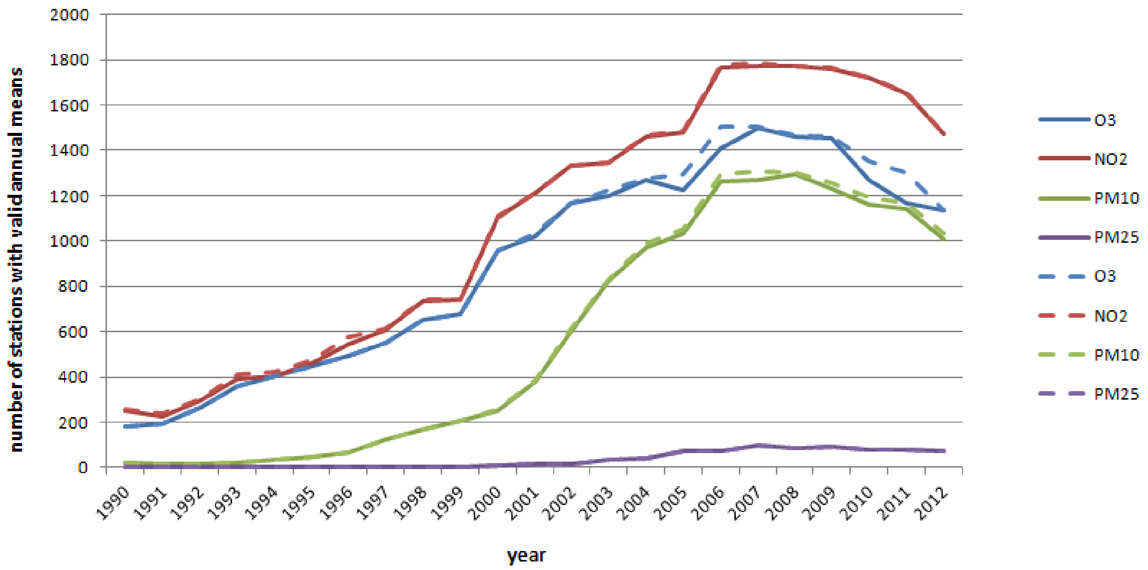


Figure 1: Number of stations (traffic, industrial, and rural, suburban and urban background) with valid annual means sorted by pollutant (solid lines: with the geostatistic outlier detection; dashed lines: without the geostatistic outlier detection)

2 Nitrogen Dioxide

2.1 Selected Stations

The network coverage of NO_2 is the densest of all compounds covered in the present report. It can reach almost 1800 sites over recent years (Figure 1) but the number of sites passing completeness and quality criteria over the past is more limited. Only 184 sites offer an appropriate annual coverage for 75% of the years in the 1990s, and most of them are located in Benelux, Germany, Austria and the UK. In the 1990s, twice as many station would pass the data completion criteria if it were based on percentage of a given season instead of percentage over the whole year. Over the full 23yr period (1990-2012), the network is also very inhomogeneous, but over the last decade, the spatial coverage is more satisfactory, with about 1000 sites considered appropriate for a trend analysis in the 2000s.

Table 1: Number of NO_2 monitoring stations in the selection for the three time periods (1990-2012, 1990-2001 and 2002-2012). The number in brackets is for the stations that comply with completion criteria in both 2002-2012 and 1990-2001.

		1990-2001	2002-2012	1990-2012
NO_2	year	184	969	255 (98)
	spring	367	1373	391
	summer	362	1346	395
	fall	371	1371	399
	winter	336	1371	399

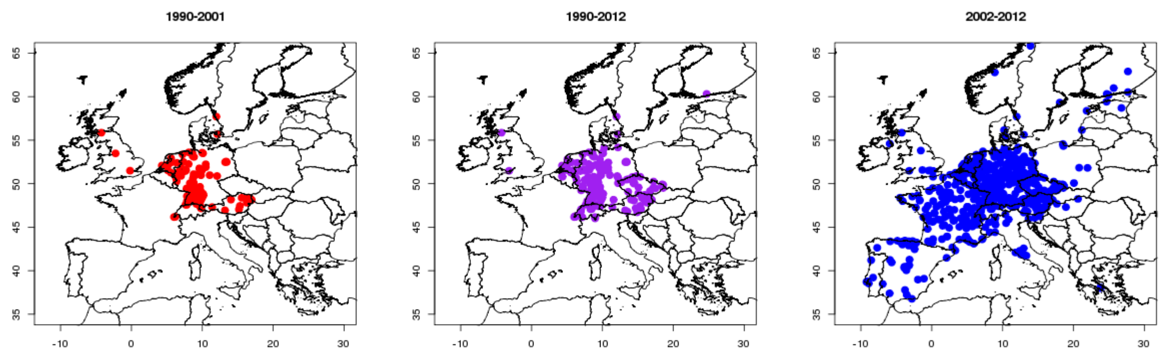


Figure 2: Location of NO_2 urban, suburban and rural background monitoring stations passing quality and completeness criteria for the three time periods (1990-2012, 1990-2001 and 2002-2012).

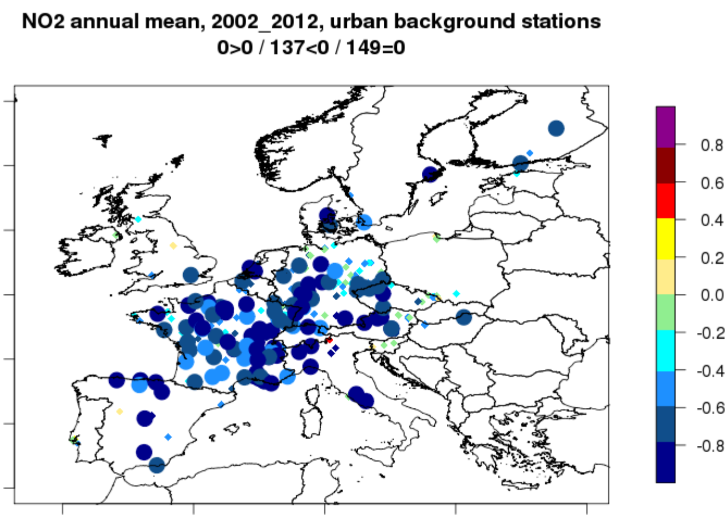
2.2 Overview of the NO_2 trends

Observed NO_2 concentration in Europe exhibit an overall strong decline throughout Europe over the past 20 years. The following subsections will present in more details the variability of the trend in space, time, and for the different type of stations and areas.

2.3 Spatial variability over 2002-2012

The **spatial variability of NO_2 trends over the last decade (2002-2012) does not exhibit any particular pattern**, apart from a slightly lower magnitude of the decrease over the North-Eastern part of the area covered by the network (Eastern Germany and Poland). More accurate aggregation techniques such as spatial block kriging should be implemented to confirm this feature (Working Paper on Spatial Aggregation for Trend Analysis, ETC/ACM 2013 working paper [2]).

A notable exception to this downward trend over 2002-2012 is for the suburban station GB0642A which is located near the Heathrow Airport. A quick view of the daily NO_2 time series recorded at that station over the whole period is provided in Annex. It illustrates that this trend does not seem influenced by any obvious flaw in the record. Further investigation with local experts would be needed to better understand this trend.



[2] http://forum.eionet.europa.eu/etc-acm-consortium/library/subvention-2013/task-deliveries-ip2013-3/1022-spatial-aq-data-and-assessments/b.-final-drafts-approval-eea/subtask-3-trend-analysis-methodology-exploration-plana-spatblockrig-planb-pwc/final-draft-working-paper-spatial-aggregated-trends-analysis-clean-version

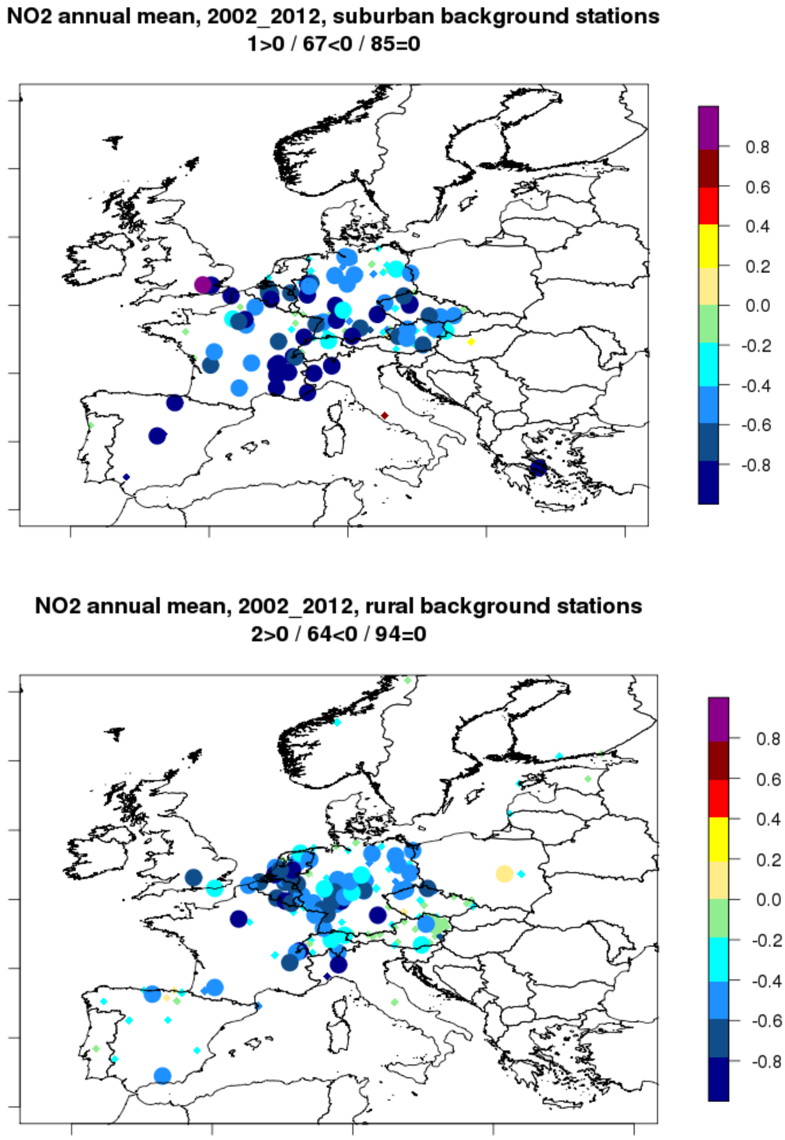


Figure 3: Trend in annual mean NO_2 (in µg/m^3/yr) at urban, suburban and rural background sites over the 2002-2012 time period. Smaller dots are for stations where the trend is not significant. In the title of each map, we provide the number of sites where the trend is significantly positive (">0"), significantly negative ("<0"), and non-significant ("=0") (p-value <0.05).

2.4 *Temporal variability*

The composite time series of NO_2 over the 255 sites that comply with the 75%/75% completion criteria for annual coverage and number of year covered over the 1990-2012 period is displayed on Figure 4. It consists of a simple arithmetic average and does not account for spatial inhomogeneities of the network so that the arithmetic average will be biased by countries better covered over the long run (Germany, Austria, Benelux) . Traffic and industrial stations (including all type of areas: urban, suburban, or rural) are included in this aggregate time series if they comply with the completeness criteria, while background sites are further required to pass the geostatistical filtering.

From this plot it appears that **downward NO_2 trends are stronger over the 1990s compared to the 2000s** for all station and area type. For both traffic and industrial stations, sharp decreases were recorded over the early 1990s. The completeness criteria requires that 75% of the years within 1990-2012 are covered. A lower number of valid stations over the first four years of the period could therefore in theory introduce an artefact on this part of the time series. That is why we also included on that plot the time series for the subset of stations including data for 100% of the years of the period (dotted lines). Of course this 100% completion criteria is much more stringent so that we reduce the spatial representativeness of the result, e.g. for traffic sites, we have only 5 stations located in Germany and Austria with valid data for all 23 years. But all of them exhibit this sharp decrease in the early 1990s, showing that this more important decline at the beginning of the 1990s is not an artefact of the station selection.

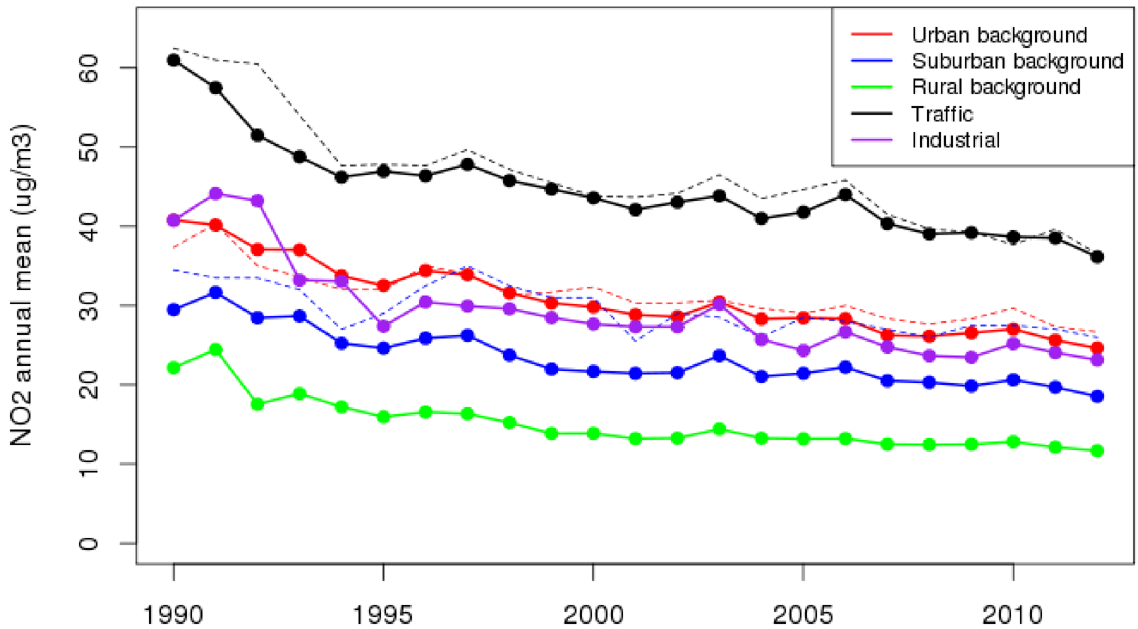


Figure 4: Average NO_2 time series ($\mu g/m^3$) over the subset of stations passing the completeness criteria (for traffic, industrial and background sites) and the geostatistical filtering (for background sites) over the 1990-2012 time period for all station types. The dotted lines is for the subset of station with valid data for 100% of the years (instead of 75%) of the period, if any.

A comparison of trends in the 1990's and in the 2000's using a consistent set of stations is given in Figure 5 (suburban sites displayed here for illustration purposes, similar features are found for other station and area typologies). It shows that the downward trend was much more pronounced in the 1990's. In relative terms (using the median concentration of the selected time period as reference, which changes when considering the 1990s, the 2000s, or the 23 years), NO_2 levels at suburban sites decreased by about 38% (±18.5%, standard deviation across the monitoring network) on average in the 1990s, and this decrease was about 17% (±16.7%) in the 2000s (Table 2). Over the full 23 year time period between 1990 and 2012, the average relative change is 44% (±33.8%), which is consistent with the 51% decline in NOx emissions over Europe between 1990 and 2012[3].

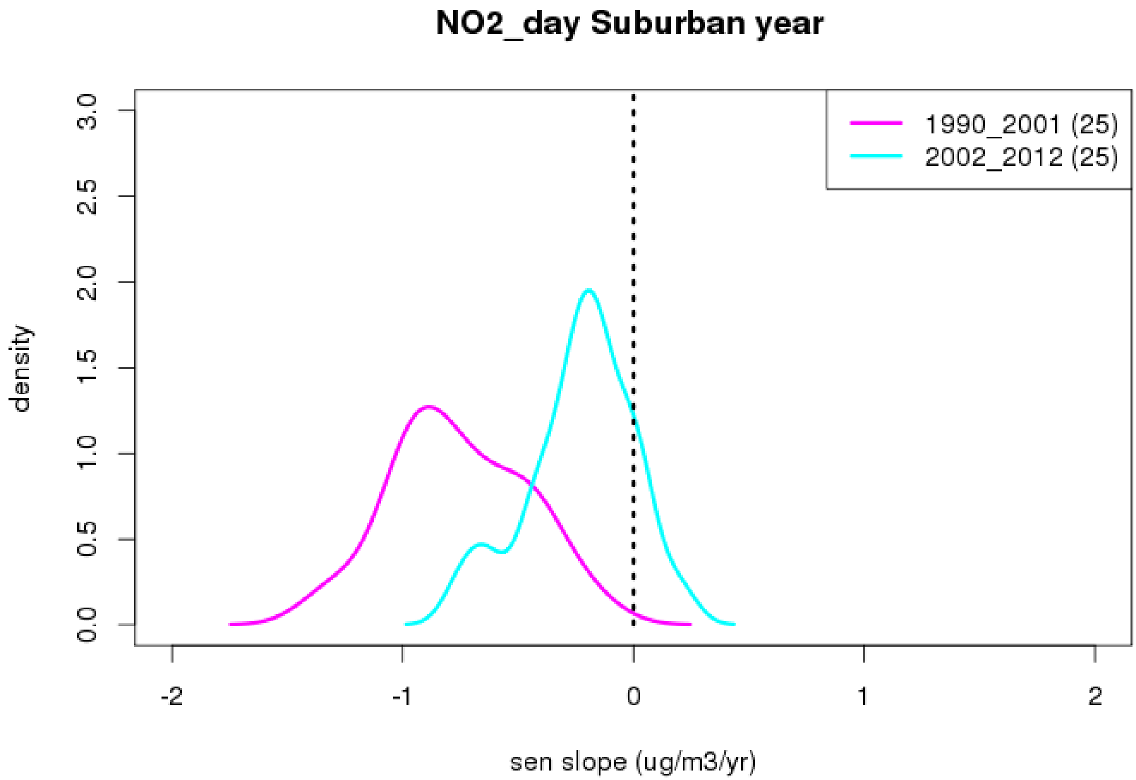


Figure 5: Distribution, as the probability density function (unit less), of trends in NO_2 annual mean at suburban background sites in the 1990's and in the 2000's using a consistent set of 25 stations.

[3] http://www.eea.europa.eu/data-and-maps/data/data-viewers/air-emissions-viewer-lrtap

Table 2: Spatial average and standard deviation of the relative trend (%) of annual NO_2 for each station typology in the 1990s and in the 2000s (reference taken as the median concentration over the selected time period).

	1990-2001		2002-2012		1990-2012	
	avg	std dev	avg	std dev	avg	std dev
urban background	-32.2	20.5	-20.5	17.7	-41.9	18.8
suburban background	-38.1	18.5	-17.6	16.7	-44.0	33.8
rural background	-44.6	13.3	-21.7	27.5	-48.0	27.2
traffic (all areas)	-32.6	20.8	-17.9	17.9	-34.1	26.9
industrial (all areas)	-24.5	22.5	-24.4	29.5	-36.0	34.7

2.5 Typology

The distribution of slopes for each station typology (background urban/suburban/rural, as well as traffic and industrial sites) is given in Figure 6. For background sites, we find that **NO_2 decreased faster in urban and suburban sites, than over rural areas**. The trend at industrial sites is similar to those of urban background stations, but trends at traffic sites exhibit a larger variability (standard deviation of the distribution) compared to other typologies.

These features are similar over the first and second decade of the period, the only slight difference regards traffic sites, whose downward trends appear less pronounced than urban stations in the 1990’s and more pronounced in the 2000’s. We have to point out that, in order to maximise the representativeness of the distributions, all stations passing the selection criteria for either the 1990’s or the 2000’s were included in these figures. The drawback is that different set of stations are used for each period. Using a consistent, yet less representative, subset of stations valid for both the 1990’s and the 2000’s we could not reproduce this differences in trends between urban background and traffic sites.
The average downward NO_2 trend was -1.15µg/m3/yr and -0.87µg/m3/yr at traffic and urban background sites in the 1990’s, while in the 2000’s these numbers were -0.68 µg/m3/yr and -0.39 µg/m3/yr, respectively. We conclude that NO_2 trends behave similarly for urban background and traffic sites and difference displayed in Figure 6 are – at least partly –due to changes in the spatial distribution of the network between both decades.

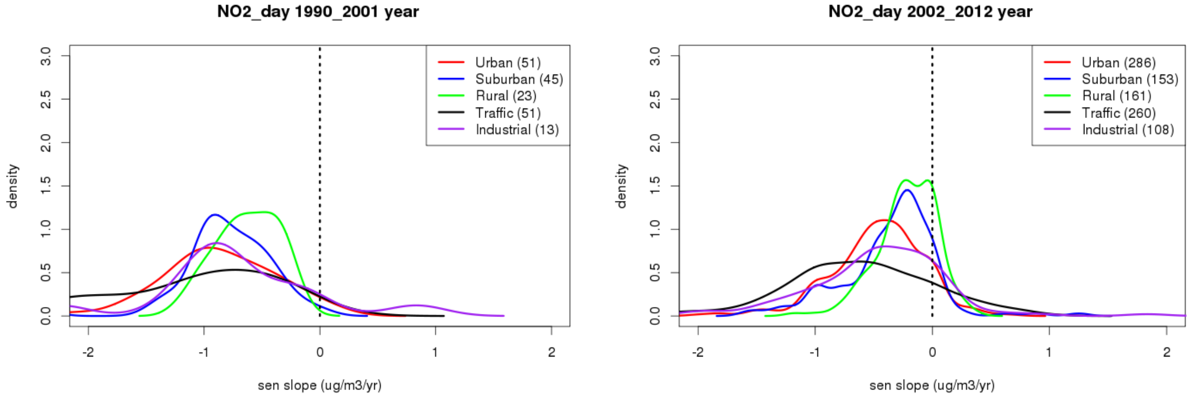
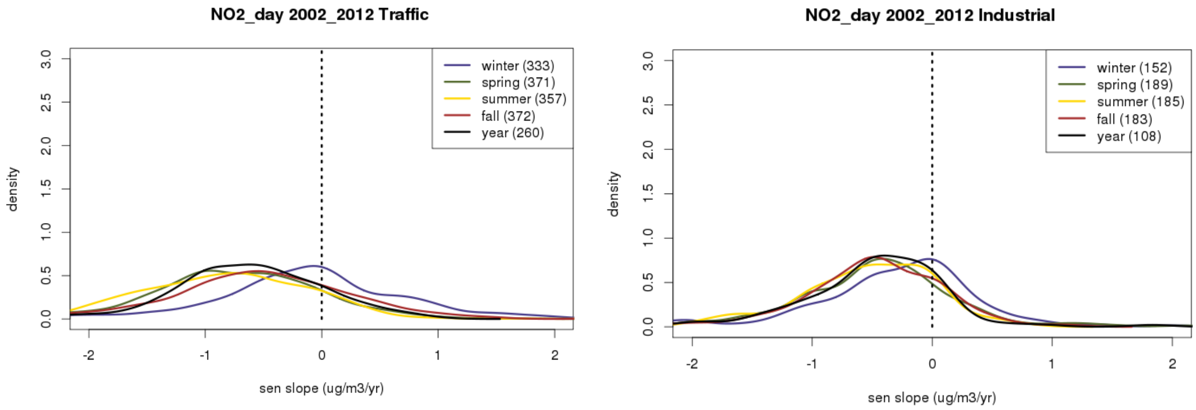


Figure 6: Distribution of trends by site and area typology for the first and second half of the time period (note that network coverage differs between both panels). The number in brackets is for the number of sites in the distribution.

2.6 Seasonality

The distributions of NO_2 trends for the four seasons and the annual trend are very similar at rural background sites, but at suburban and urban background sites, as well as at traffic and industrial sites a lower magnitude of the trend is found in winter. Because at polluted sites, wintertime NO_2 levels are controlled to a large extent by background ozone, this feature must be put in perspective with the trends in ozone background levels introduced in Section 3, where we will see that the trend in baseline is somewhat flat, which will yield a flat trend in NO_2 in wintertime close to the sources, even if local emissions have been reduced.



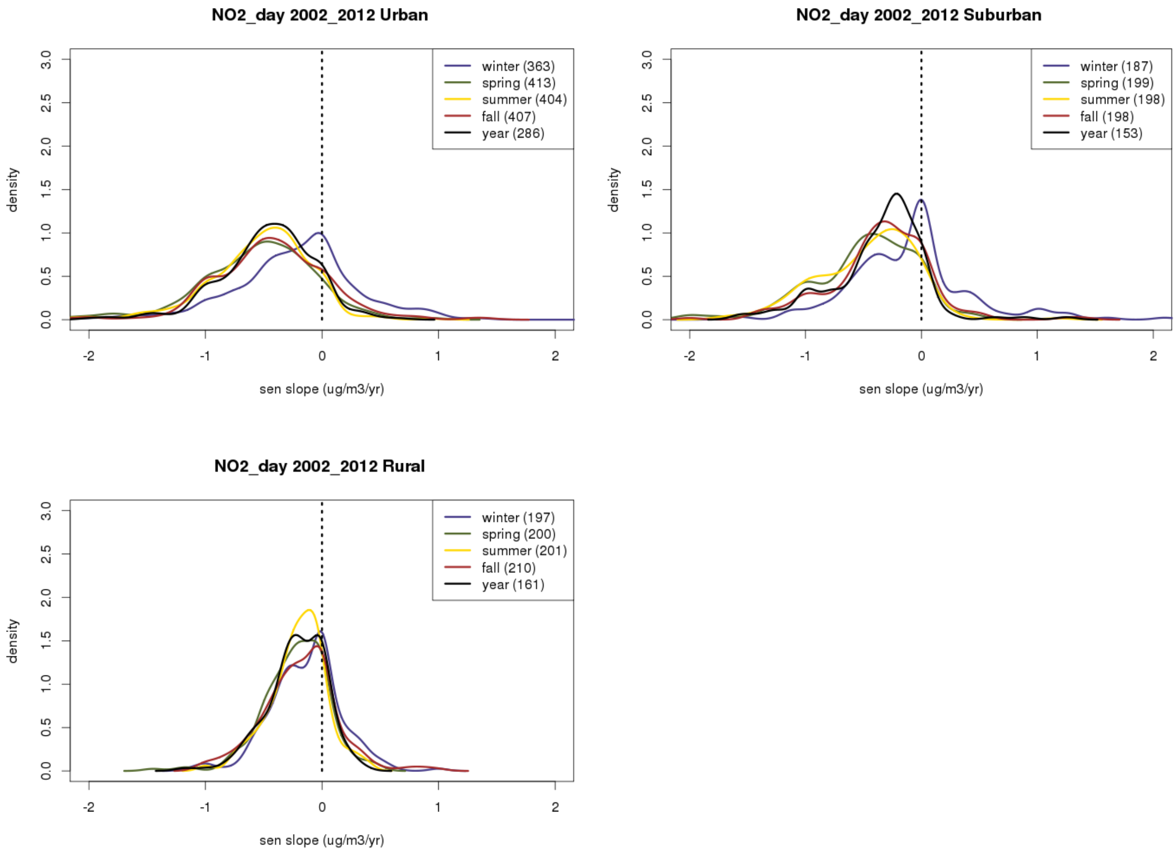


Figure 7: Distribution of trends over the 2002-2012 time period at each typology of area and station for the four seasons, and for the annual trends. The number in brackets is for the number of sites in the distribution.

3 Ozone

3.1 Selected Stations

Ozone is the second best covered atmospheric pollutant after NO_2 in the monitoring network. In the 1990s its coverage was as good as NO_2. It has today slightly less stations than NO_2, which is legitimate because of the smaller spatial variability of ozone. Again, we find that the spatial distribution of the network is biased towards Benelux, Germany and Austria in the 1990's. Over 800 sites can be considered as appropriate in terms of data coverage and quality to document annual mean ozone trends, this number can increase to more than 1100 sites where focusing on summertime trends in the 2000s.

Table 3: Number of O_3 monitoring stations in the selection for the three time periods (1990-2012, 1990-2001 and 2002-2012). The number in brackets is for the stations that comply with completion criteria in both 2002-2012 and 1990-2001.

		1990-2001	2002-2012	1990-2012
O_3	year	202	827	245 (129)
	spring	331	1142	331
	summer	366	1148	345
	fall	350	1192	359
	winter	269	1086	314

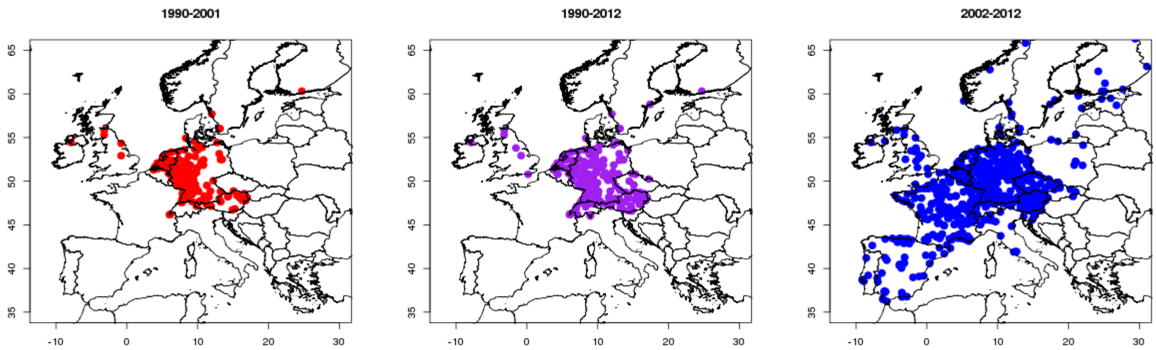


Figure 8: Location of O_3 urban, suburban and rural background monitoring stations passing quality and completeness criteria for the three time periods (1990-2012, 1990-2001 and 2002-2012).

3.2 *Overview of the O_3 trends*

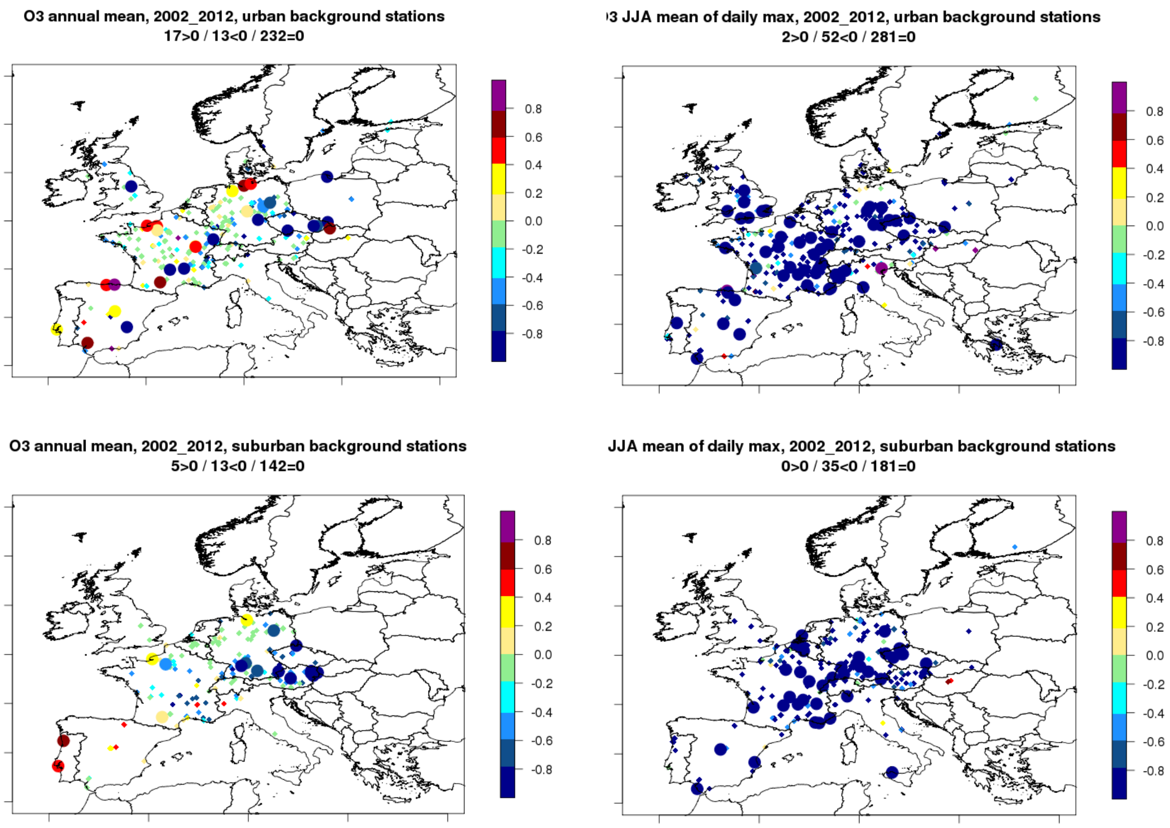
Ozone is a secondary pollutant largely influenced by long range transport and meteorological factors in addition to the emissions of precursors. Even if ozone peaks have been decreasing over recent years, the trend is not as large as reported for instance for NO_2 above.

3.3 *Spatial variability over 2002-2012*

Annual mean ozone trends exhibit a higher spatial variability (Figure 9) at background urban sites compared to suburban and rural background sites in the 2000’s. At rural sites, there is a slight zonal gradient, with more pronounced downward trends over Central and Eastern Europe compared to the Atlantic-facing countries although, again, more robust aggregation techniques are needed to give some statistical ground to this feature.

As far as summertime ozone peaks (as the JJA mean of daily maxima computed from 8hr running means) are concerned, **a quite spatially homogeneous downward trend of ozone peaks is found**. There remain however some geopraphical variability when comparing sites where the trend is significant or not, with less significant trends on the Atlantic-facing part of Europe.

There are a couple of outlying stations in Italy with substantially increasing trends (the IT0992A rural background site and the IT1590A urban background site), quick views are provided in the Annex and illustrate that no obvious flaw occurs in the network and only further investigation with local experts would allow understanding such trends.



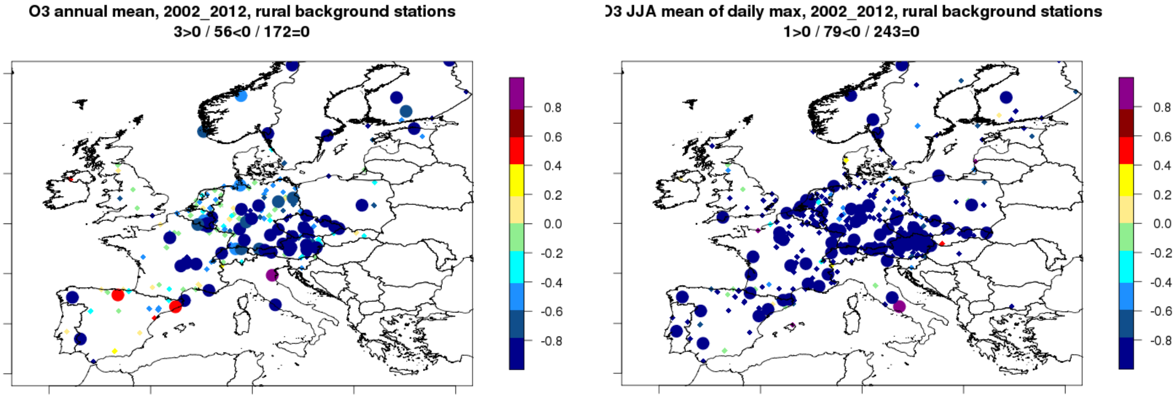
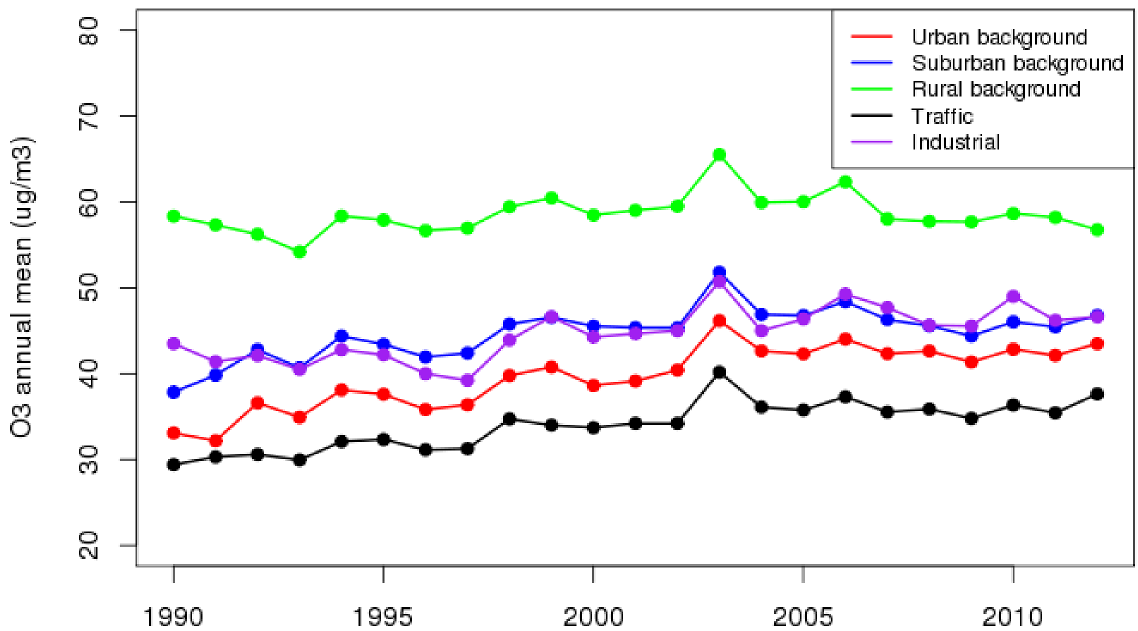


Figure 9: Trend in: left: annual mean O_3 and right: summertime mean daily maxima of the 8-hr running mean (in µg/m^3/yr) at urban, suburban and rural background sites over the 2002-2012 time period. Smaller dots are for stations where the trend is not significant. In the title, we provide the number of sites where the trend is significantly positive (“>0”), significantly negative (“<0”), and non-significant (“=0”) (p-value <0.05).

3.4 Temporal variability

The spatial composites (temporal evolution of the average in space over the whole network for each station and area type) in Figure 10 illustrate the **increase in annual mean ozone over the 1990s followed by a flattening out of the trend over recent years.** This behaviour is less pronounced at rural background sites, because the reduced titration is more important at other station type and location. **For ozone peaks, a somewhat flat trend was already observed over the 1990s, with a decrease over the 2000s**.

The outstanding high ozone years of 2003 and 2006 appear clearly on these plots, especially for summertime ozone peaks (lower panel of Figure 10). 1993 was an outstanding low ozone year compared to usual levels in the 1990s.



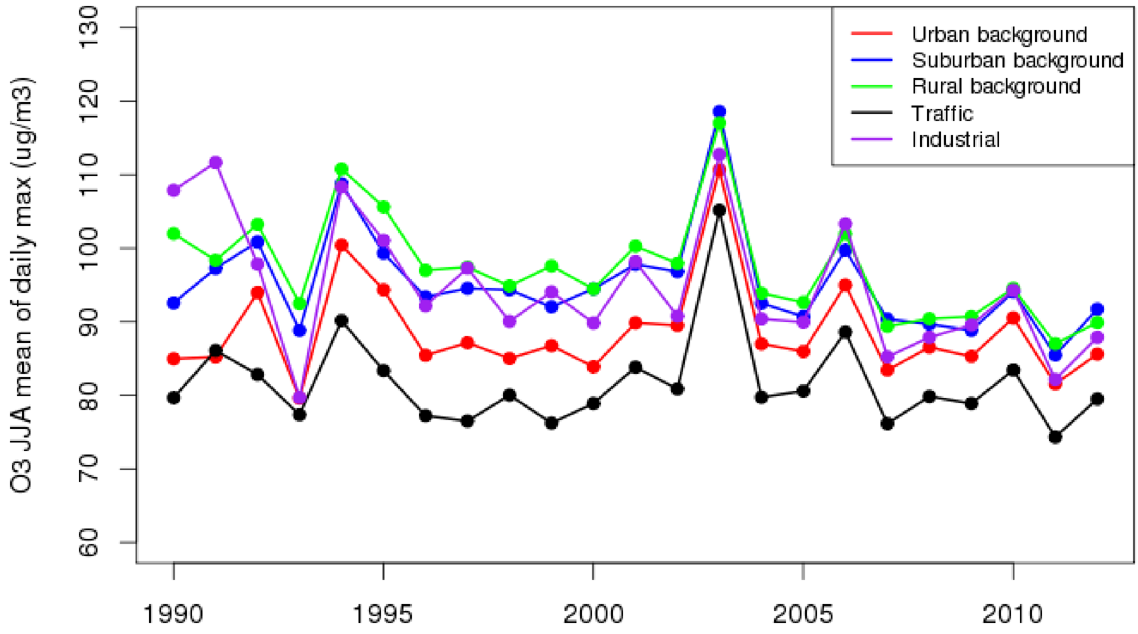


Figure 10: Average O_3 time series (µg/m^3) (top: annual mean, bottom: summertime mean of daily max) over the subset of stations passing the completeness criteria (for traffic, industrial and background sites) and the geostatistical filtering (for background sites) over the 1990-2012 time period for all station types.

A comparison of the trends in the 1990's and 2000's is shown in Figure 11 using a consistent set of rural background stations. It shows that the main features presented above on the basis of the arithmetic composite is well supported for the whole distribution of trends. Annual ozone levels increased over the 1990's while they slowed down or even decreased over the 2000's. For ozone peaks the distribution of trend is centred on negative values, although relatively flat for the 1990's while it decreases substantially over the 2000's.

In relative terms (Table 4), in the 1990's, annual ozone increased about 5.1% (±10.2%) at rural sites, while this increase was reaching 14.7% (±11.2%) at urban sites, showing that the increased cannot be considered significant over the network of rural background sites. Over the 2002-2012 period, annual mean ozone decreased at all background sites, up to -7% (±9.0%) at rural sites, although the decrease is only -0.3% (±10.2%) at urban sites. The important spread of trends over the network reflected by the 1-sigma standard deviation in brackets and in Table 4 emphasises that the trend of annual mean ozone is not significant over the network.

Daily ozone maxima were already declining on average in the 1990s', but at rural sites the relative trend was only -7.6% (±12.2%) while it reaches -13.6% (±8.7%) over the 2002-2012 period.

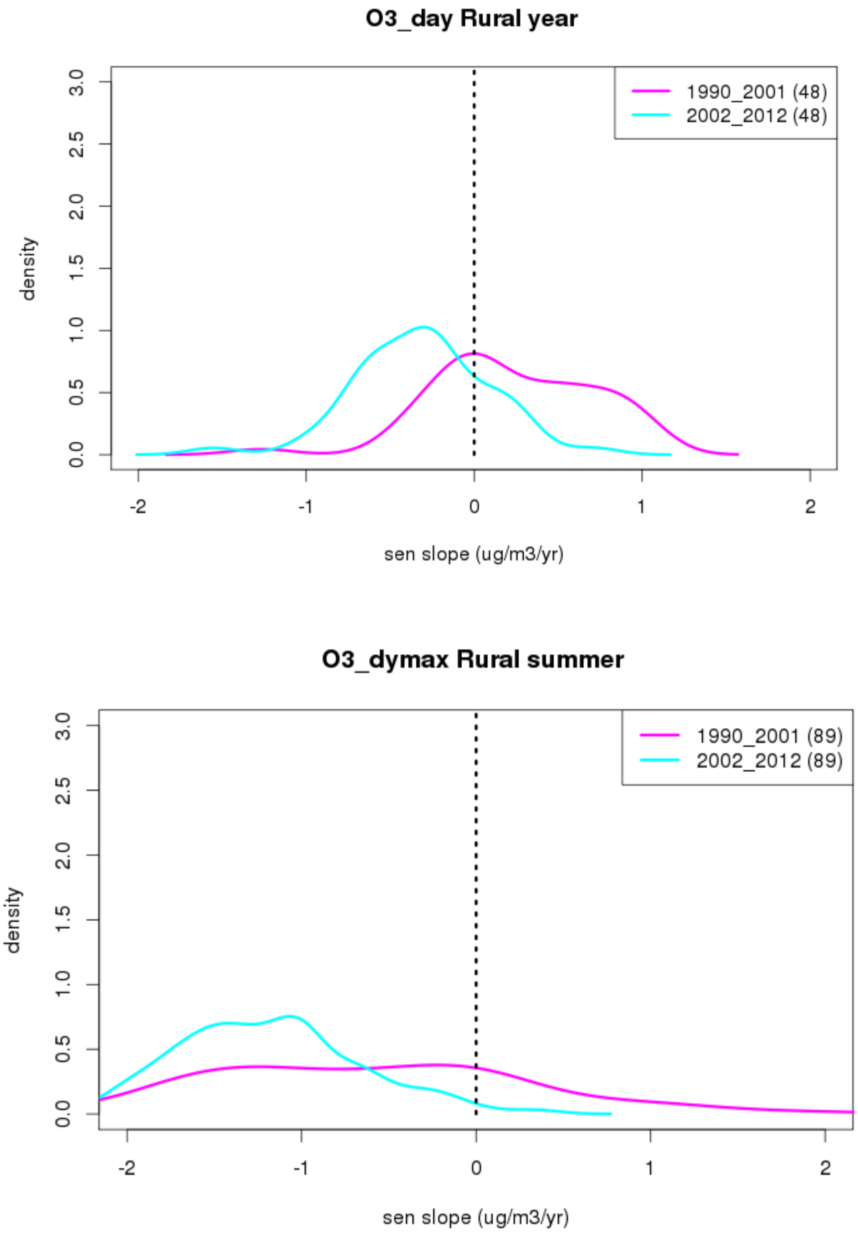


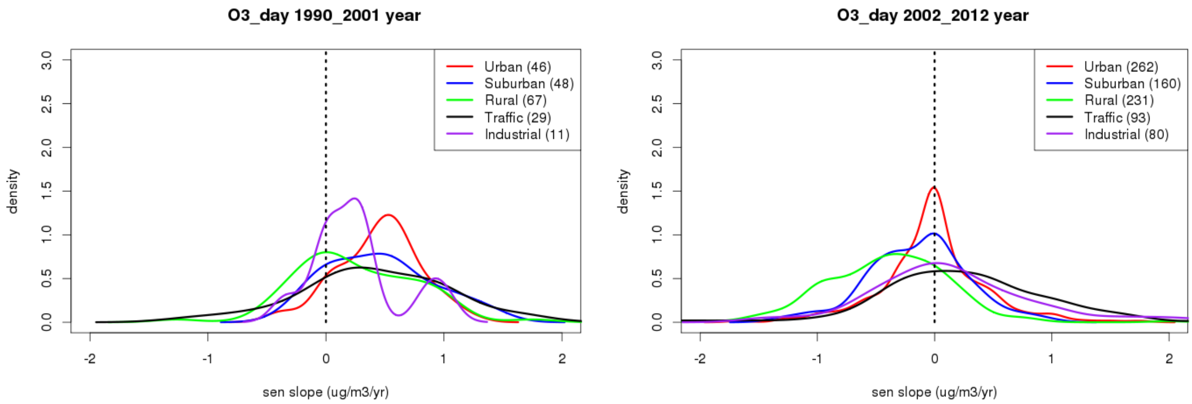
Figure 11: Distribution of O_3 trends (top: annual daily means, bottom: summertime daily max) at rural background sites in the 1990's and in the 2000's using a consistent set of stations.

Table 4: Spatial average and standard deviation of the relative trend (%) of annual daily mean O_3 and summertime daily max O_3 for each station type and area in the 1990s and in the 2000s (reference taken as the median over the selected time period).

		1990-2001		2002-2012	
		avg	std dev	avg	std dev
O3 daily mean (annual)	urban background	14.7	11.2	-0.3	10.2
	suburban background	13.5	12.9	-3.2	9.6
	rural background	5.1	10.2	-7.0	9.0
	traffic	15.8	25.6	9.1	28.3
	industrial	7.0	10.9	7.1	20.6
O3 daily max (summer)	urban background	-4.8	12.1	-12.1	10.2
	suburban background	-5.2	11.0	-13.3	8.1
	rural background	-7.6	12.2	-13.6	8.7
	traffic	0.3	27.7	-3.0	22.0
	industrial	-13.8	11.3	-6.4	19.0

3.5 Typology

Ozone trends tend to be more frequently positive at background urban sites than background rural sites for both ozone annual average and summertime peaks and for both 1990's and 2000's time period (Figure 12**)**. This is because the ozone titration effect close to the sources in NOx saturated environments where NOx declines tends to increase ozone concentrations. Trends at traffic and industrial sites for all type of areas can be even larger. The variability of the trend in ozone peaks is notable, especially in the 1990's.



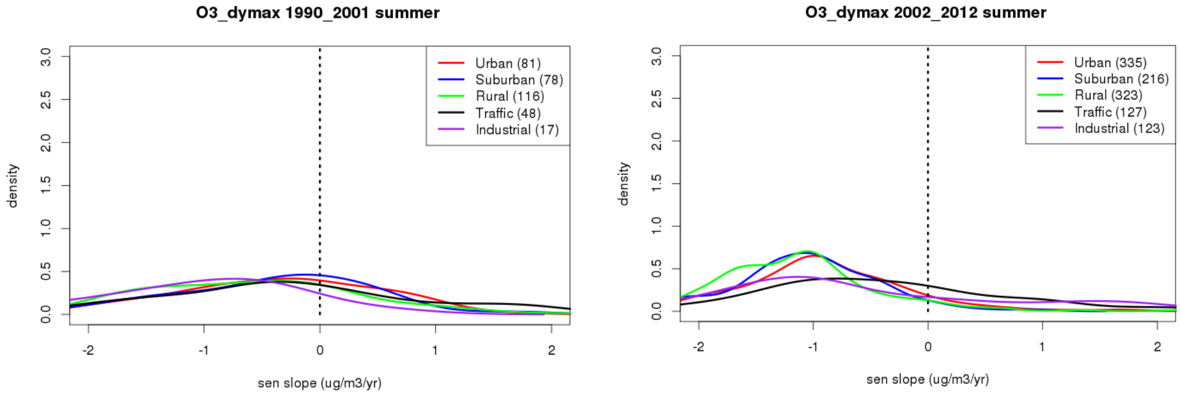
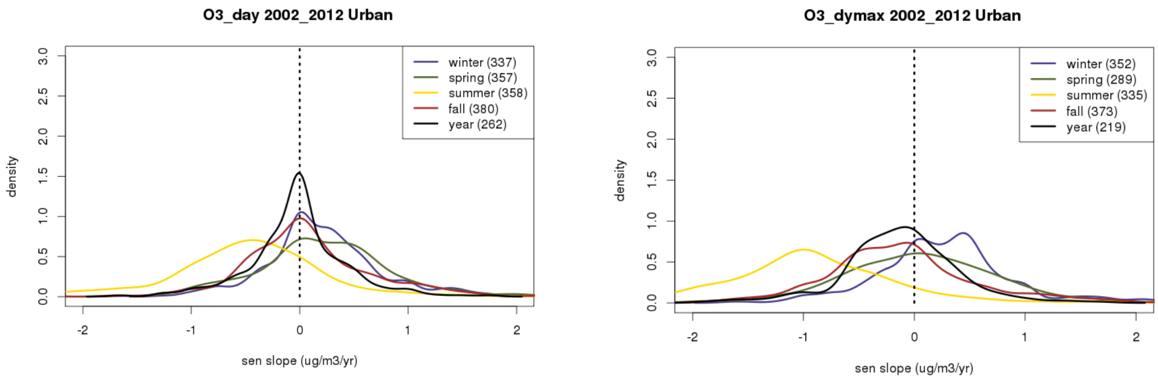


Figure 12: Distribution of O_3 trends (top: annual mean, bottom: summertime peaks) for background rural, suburban and urban sites as well as traffic and industrial stations for the first and second half of the time period (note that network coverage differ between both panels). The number in brackets is for the number of sites in the distribution.

3.6 Seasonality

For both ozone annual means and daily maxima, downward trends are more pronounced over summer and less pronounced over winter in the 2000's (Figure 13). This feature is beneficial in terms of exposure since it is of course in summer that ozone levels are highest.

Trends in low level ozone during intermediate seasons should however be further investigated in terms of exposure indicators (e.g. by looking at trends in AOT40 or SOMO10) to ascertain a reduction of exposure to detrimental ozone levels.



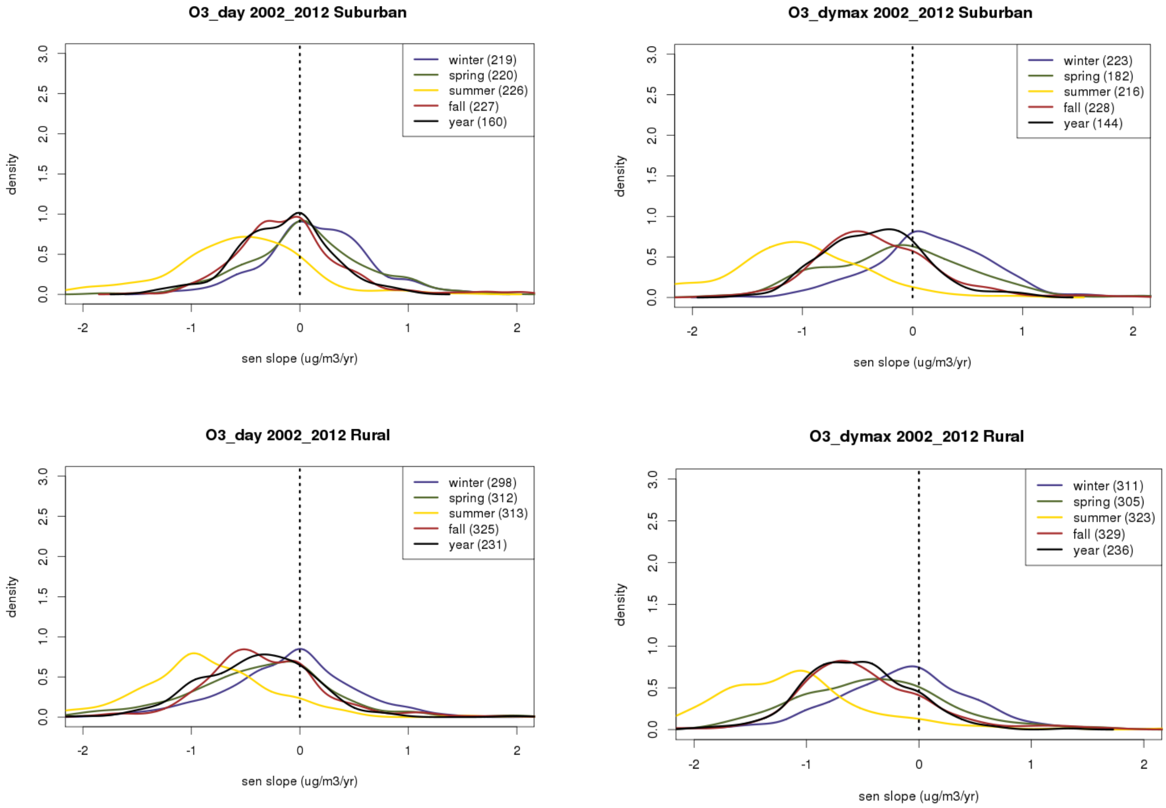


Figure 13: Distribution of ozone annual mean (left) and ozone peaks (right) trends over the 2002-2012 time period at background stations for the four seasons and for the annual trends. The number in brackets is for the number of sites in the distribution.

These changes in trends by season cycle are reflected when looking at average seasonal cycles, such as in Figure 14 where the average cycle over the 1990s and the 2000s is shown. An increase in the second period over winter and spring is found, whereas summertime levels decrease. Note that the change appears limited compared to trends in Figure 13 because we aggregate 1990-2001 and 2002-2012, thereby minimising differences. As detailed in (Wilson et al., 2012), changes in wintertime and summertime ozone are mostly related to local effects: (i) increase in winter due to the titration effect, (ii) reduction in summer due to less efficient photochemical ozone production. On the other hand, the increase in spring, that appears clearly in the airbase data, is related to long range transport and global ozone background levels as well as stratosphere-troposphere exchanges (Monks, 2000).

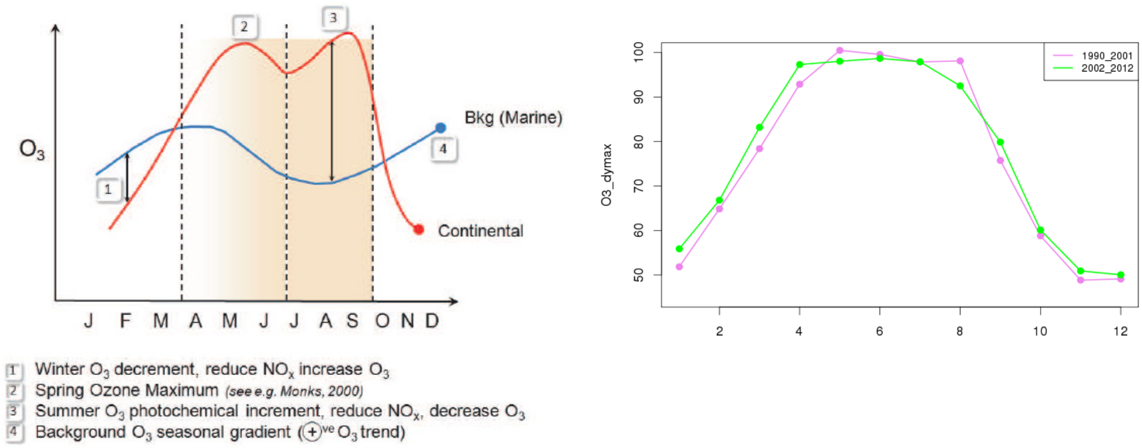


Figure 14: Left: schematic diagram of idealized marine ozone seasonal cycle (from (Wilson et al., 2012)), right: average seasonal cycle of daily ozone maxima at rural sites over the 1990s and the 2000s.

4 Particulate Matter

4.1 Selected Stations

The airbase network for particulate matter is much less favourable for trend analyses compared to NO_2 or O_3. Very few sites pass the quality criteria for PM25 in the 2000's and none over the 1990s.

For PM10, the coverage is relatively good in the 2000's with 500 stations passing the criteria for the annual mean. The geographical distribution of the network is presented in Figure 15. Records in the 1990's are not dense enough to allow any representative trend analysis.

Note that the measurement technique changed in France in 2007, to avoid discarding too many sites, a correction factor of 1.3 was applied to all French stations before that year.

Finally, after investigating stations for which outstanding trends were observed it appeared that a number of stations exhibited suspicious PM10 records and had to be discarded from the present analysis. A sample of such record is given in Annex. In total 47 stations[4] were further discarded on the basis that the minimum annual mean PM10 concentration could not be smaller than the 80th quantile of annual means divided by four.

Table 5: Number of PM monitoring stations in the selection for the three time periods (1990-2012, 1990-2001 and 2002-2012). The number in brackets is for the stations that comply with completion criteria in both 2002-2012 and 1990-2001.

		1990-2001	2002-2012	1990-2012
PM_{10}	year	9	520	7 (2)
	spring	20	909	25
	summer	19	853	23
	fall	24	897	30
	winter	17	761	21
$PM_{2.5}$	year	0	10	0 (0)
	spring	0	49	0
	summer	0	46	0

[4] The discarded stations are NL00641 IT1706A GB0682A DEST081 DETH039 DERP020 DETH072 DERP032 DERP029 DEHH026 DEHH070 DETH043 DESN061 DETH011 BEPWOL1 BEMR801 RO0002A IT0859A ES0777A ES0339A BELZL01 BEMGK02 BEMBE02 NL00241 IT1277A GB0658A GB0646A GB0613A GB0620A GB0036R GB0641A GB0567A DETH060 DEUB005 DEUB004 DETH005 DETH042 DETH018 DETH027 DEUB030 DETH041 DETH061 DETH036 DETH009 DETH020 DETH013 DETH026

	fall	0	43	0
	winter	0	22	0

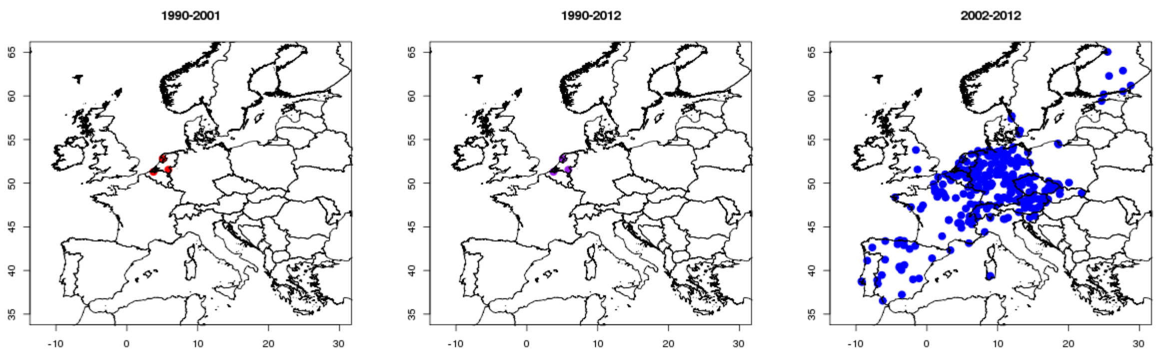


Figure 15: Location of PM10 urban, suburban and rural background monitoring stations passing quality and completeness criteria for the three time periods (1990-2001, 1990-2012, and 2002-2012) for the annual mean.

4.2 Overview of PM10 trends

Because of the recent development of the network, PM10 trends can only be documented over the second time period considered (2002-2012). Substantial reduction in PM10 concentration were reported over that decade throughout Europe.

4.3 Spatial Variability over 2002-2012

The spatial variability of the magnitude of PM10 trends is limited. Slightly lower trends are found in France but the use of a simple correction factor in that country questions the reliability of this feature.

Some stations do not exhibit a significant trend while neighbouring sites do show a significant trend. This spatial variability of the significance of the trend should be investigated.

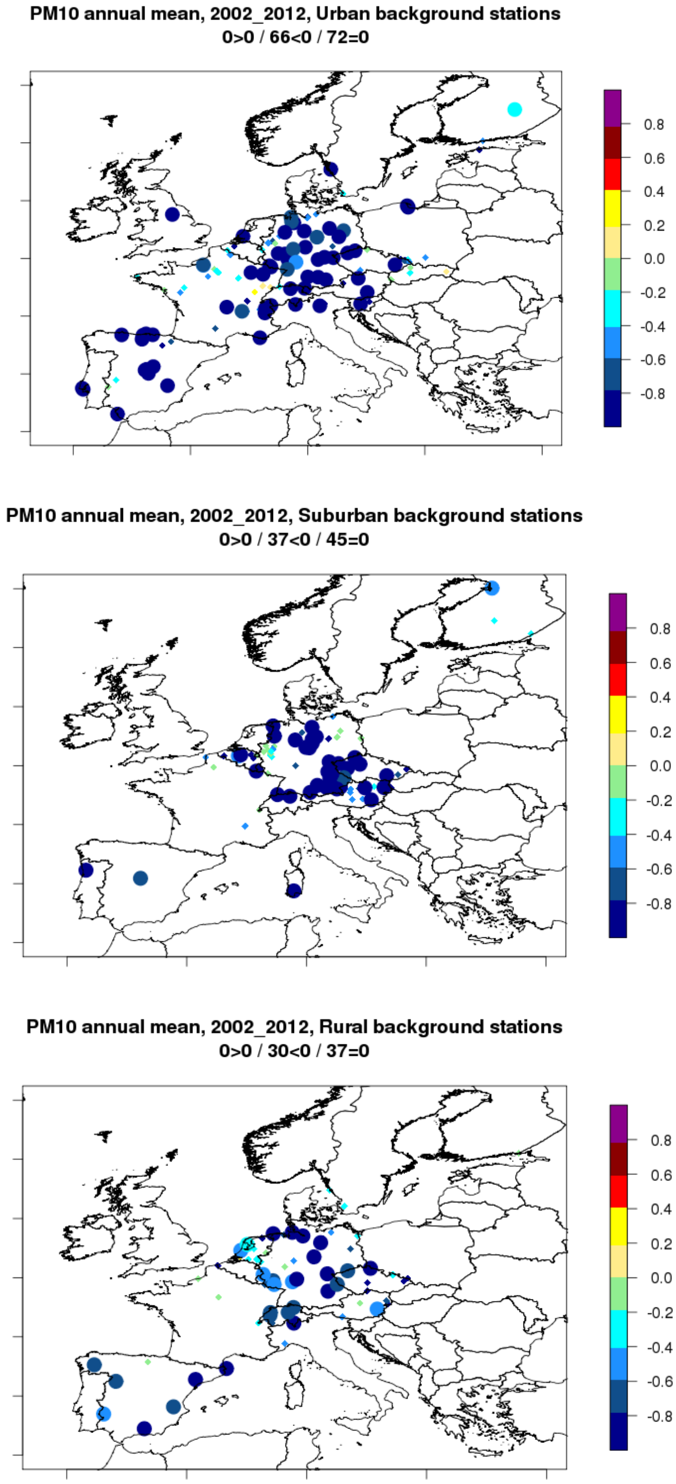


Figure 16: Trend in annual mean PM10 (in µg/m^3/yr) at urban, suburban and rural background sites over the 2002-2012 time period. Smaller dots are for stations where the trend is not significant. In the title, we provide the number of sites where the trend is significantly positive (">0"), significantly negative ("<0"), and non-significant ("=0") (p-value <0.05).

4.4 Temporal variability

A composite time series can only be produced for the second decade of the time period. An overall decline is found. There are a few features that appear on all station types in this timeseries that would deserve further investigation: (i) the sharp decrease in 2004, (ii) the higher levels in 2006, (iii) the steady increase between 2008 and 2011.

The **average rate of decrease in relative terms is of the order of 30% for background sites in the 2000's.** This trend is in satisfactory agreement with the 14% reduction of primary PM10 emission, supplemented by the 54%, 30%, and 8% reduction of SOx, NOx and NH3 precursors gases, respectively over the 2003-2012 time period (EEA, 2014).

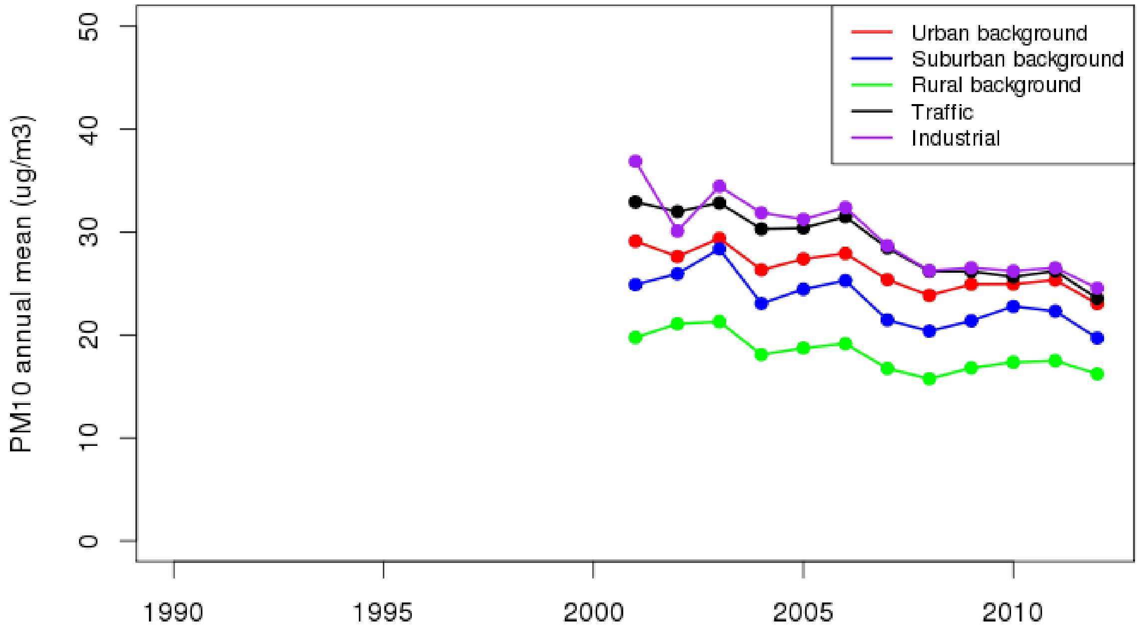


Figure 17: Average PM10 time series (µg/m^3) over the subset of stations passing the completeness criteria (for traffic, industrial and background sites) and the geostatistical filtering (for background sites) over the 2001-2012 time period for all station types.

Table 6: Spatial average and standard deviation of the relative trend (%) of annual PM10 for each station typology in the 2000's (reference taken as the median over the selected time period).

	2002-2012	
	avg	std dev
Urban background	-29.9	23.1
Suburban background	-33.6	28.0
Rural background	-30.6	26.2
traffic	-35.4	22.3
industrial	-40.3	33.7

4.5 Typology

The distribution of PM10 trends exhibits a larger spread for industrial sites, and also to some extent for urban background sites. Overall **the trend is more pronounced close to the sources (traffic, industrial and background urban sites) than in rural areas**.

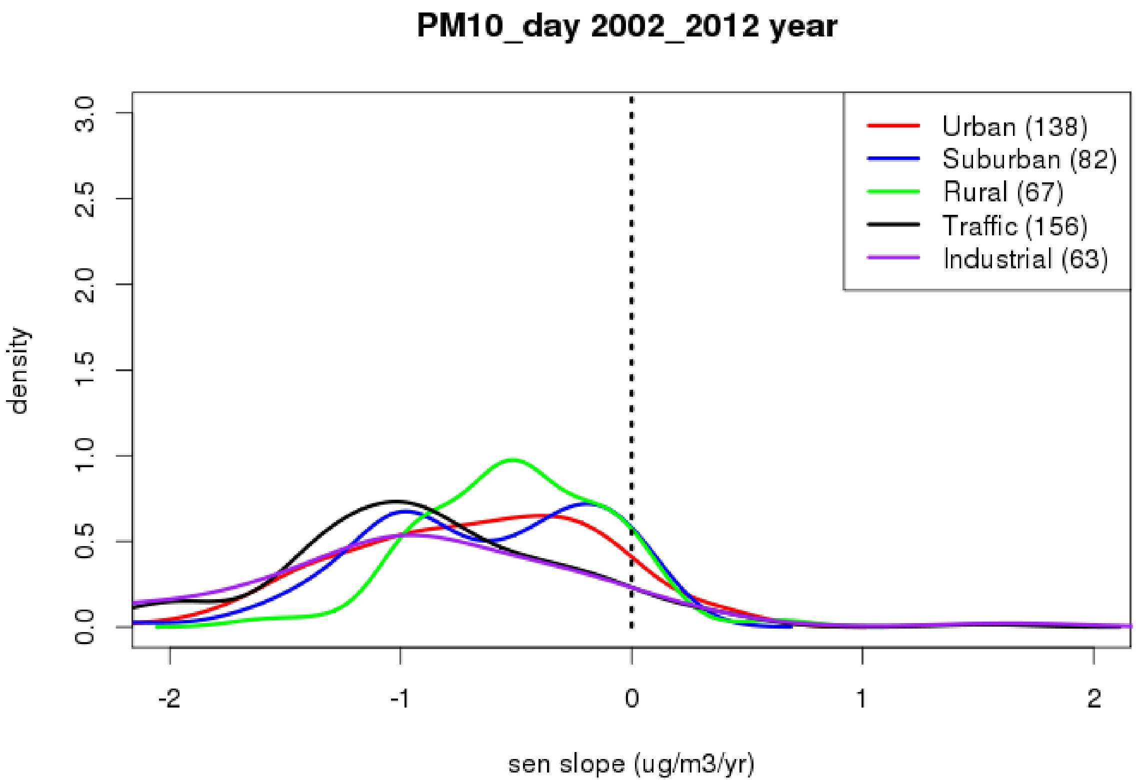


Figure 18: Distribution of PM10 trends by location and site typology. The number in brackets is for the number of sites in the distribution

4.6 Seasonality

The distributions of trends are very similar throughout the year for industrial stations, which is expected close to a source that does not exhibit very specific seasonal cycle. At traffic stations, negative trends are less pronounced in winter. This feature could be due to a meteorological effect (with more wintertime stagnation episodes towards the end of the period), or to a more efficient reduction in precursors of secondary aerosol (mainly formed in warmer seasons) in traffic emissions compared to primary emissions.

This **lower decrease of PM10 in winter compared to summer is also observed at background sites**. In urban areas, one could also suspect an increase in the use of low performance solid biomass devices for residential heating. The responsibility of this source is supported by the somewhat limited trends in fall compared to summer and spring. Note that springtime PM10 levels decrease in a similar fashion as summertime PM10.

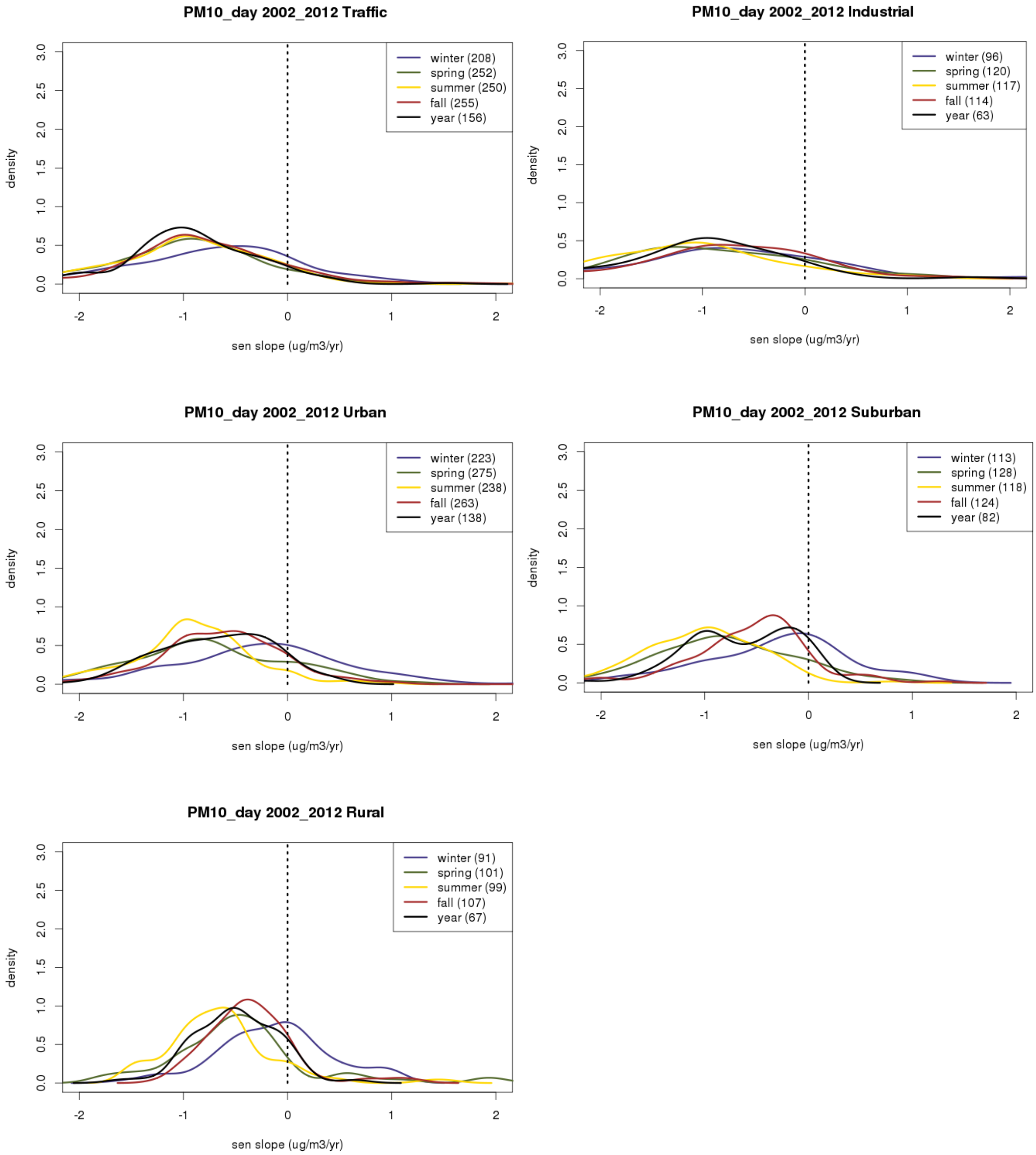


Figure 19: Distribution of PM10 trends over the 2002-2012 time period at each typology of area and station for the four seasons, and for the annual trends. The number in brackets is for the number of sites in the distribution.

5 Conclusion

Applying trend diagnostics to a cleansed version of the Airbase repository allow drawing relevant conclusions on the evolution of air quality in Europe.

Main Findings :

- We found that air quality improved overall in Europe. Negative trends are found for NO_2 and Particulate Matter, as well as ozone peaks;
- For annual mean ozone, the trend is more limited especially at urban background sites and in winter (because of the titration effect), but also in spring, suggesting an influence of long range transport;
- For all compounds, downward trends tend to be larger close to the sources, and also in summer compared to winter.

Perspectives:

- Some open questions remain on the representativeness of spatial aggregation presented here. Because the monitoring network is highly variable in space, more sophisticated geostatistical aggregation approaches should be considered to produce a representative European air quality trend;
- The data filtering approach was not 100% successful in filtering out dubious PM10 records and further quality checks should be developed;
- The trends discussed in the present report exclude trends in exposure (SOMO35, AOT40) or regulatory indicators (exceedances), which should be considered in further work;
- Last, using exclusively measurements, it is difficult to conclude on the relative role of European air pollutant emissions policies and externalities such as intercontinental transport or meteorological variability. Sensitivity simulations using chemistry-transport models could help filling this gap.

References

EEA: Air Quality in Europe, Copenhagen, 2014.

Monks, P. S.: A review of the observations and origins of the spring ozone maximum, Atmospheric Environment, 34, 3545-3561, 2000.

Wilson, R. C., Fleming, Z. L., Monks, P. S., Clain, G., Henne, S., Konovalov, I. B., Szopa, S., and Menut, L.: Have primary emission reduction measures reduced ozone across Europe? An analysis of European rural background ozone trends 1996-2005, Atmos. Chem. Phys., 12, 437-454, 2012.

Annex – Sample Time Series for Outlying stations

This annex provides quick views of air quality records presenting outstanding behaviour as pointed out in the main text to illustrate that such behaviour cannot be readily attributed to obvious flaws in the record.

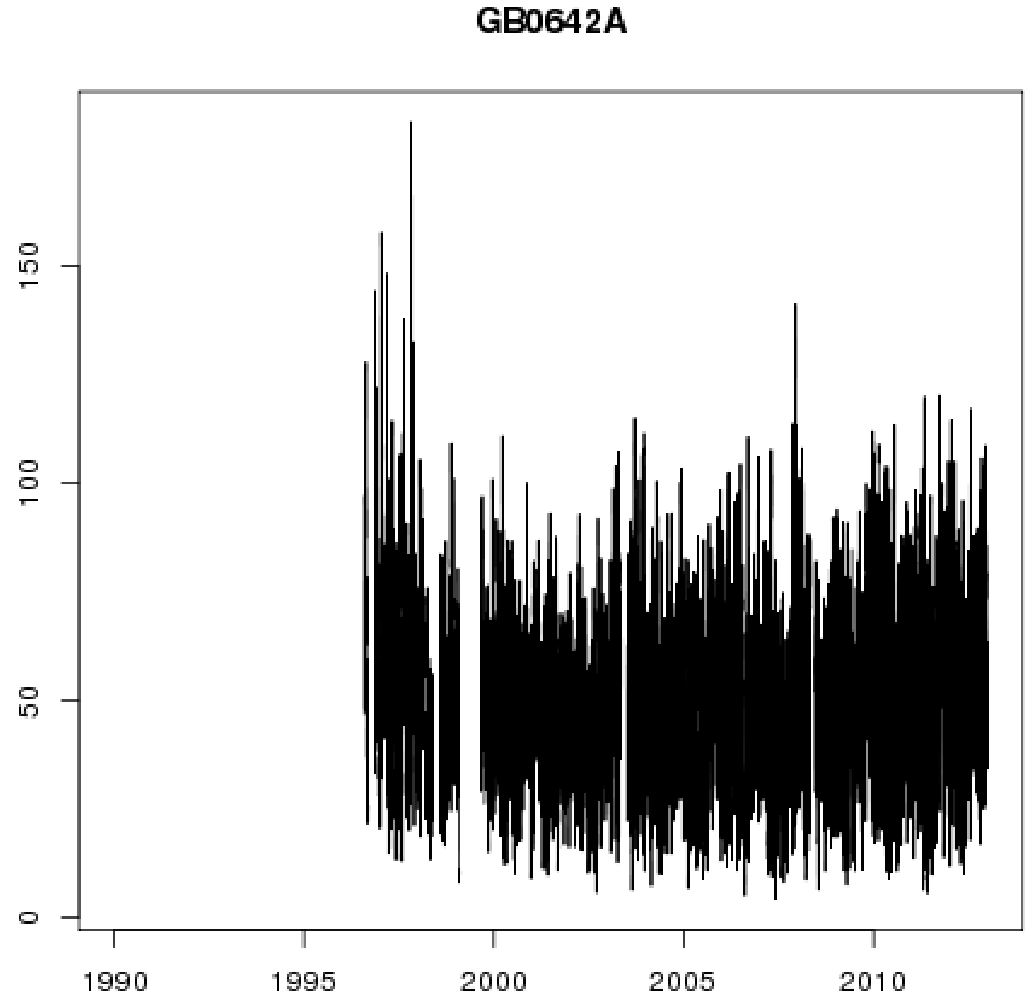


Figure 20: Time series of daily NO_2 (µg/m^3) at the GB0642A site.

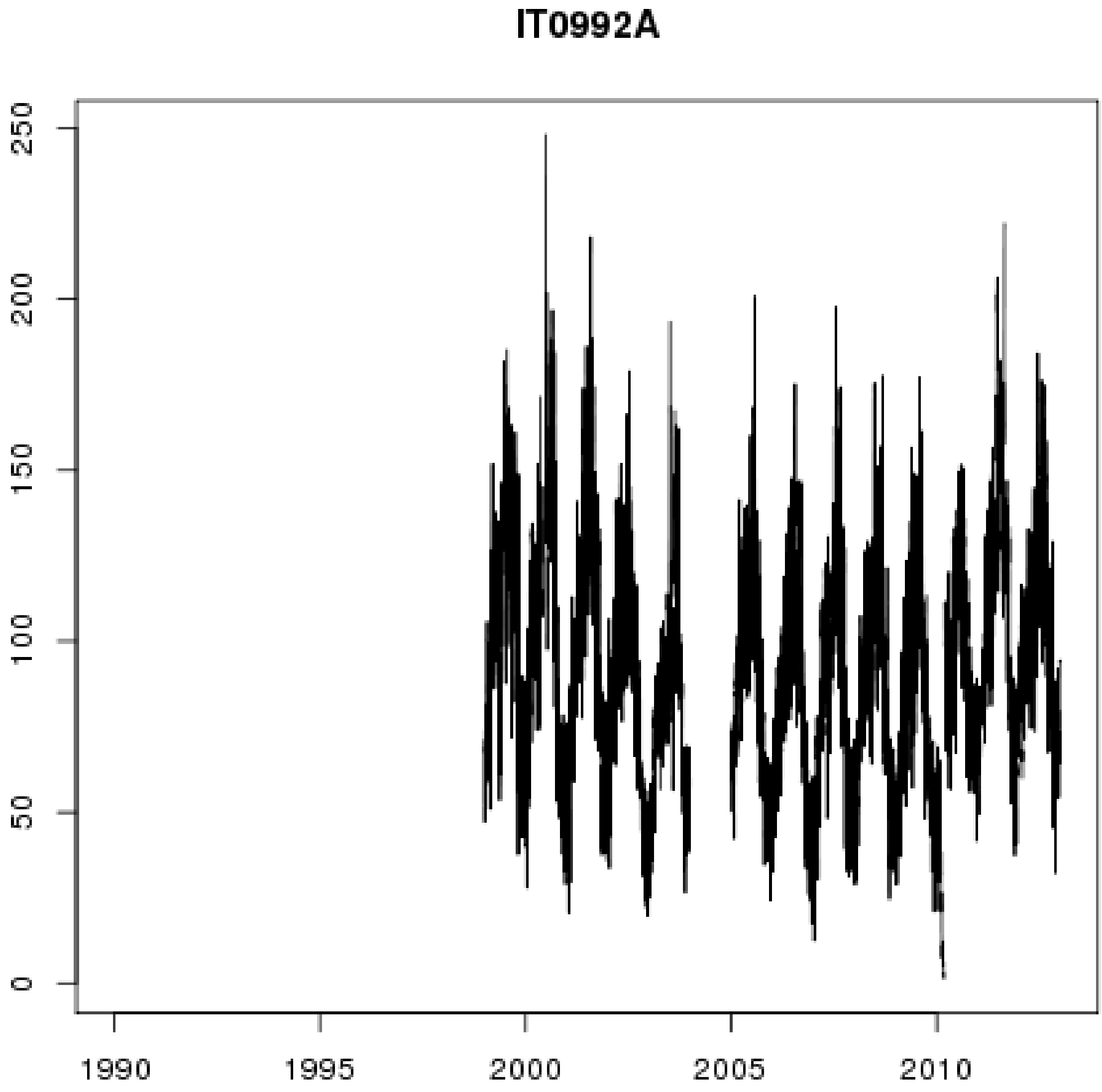


Figure 21: Time series of daily maxima O_3 (μg/m^3) at the IT0992A site.

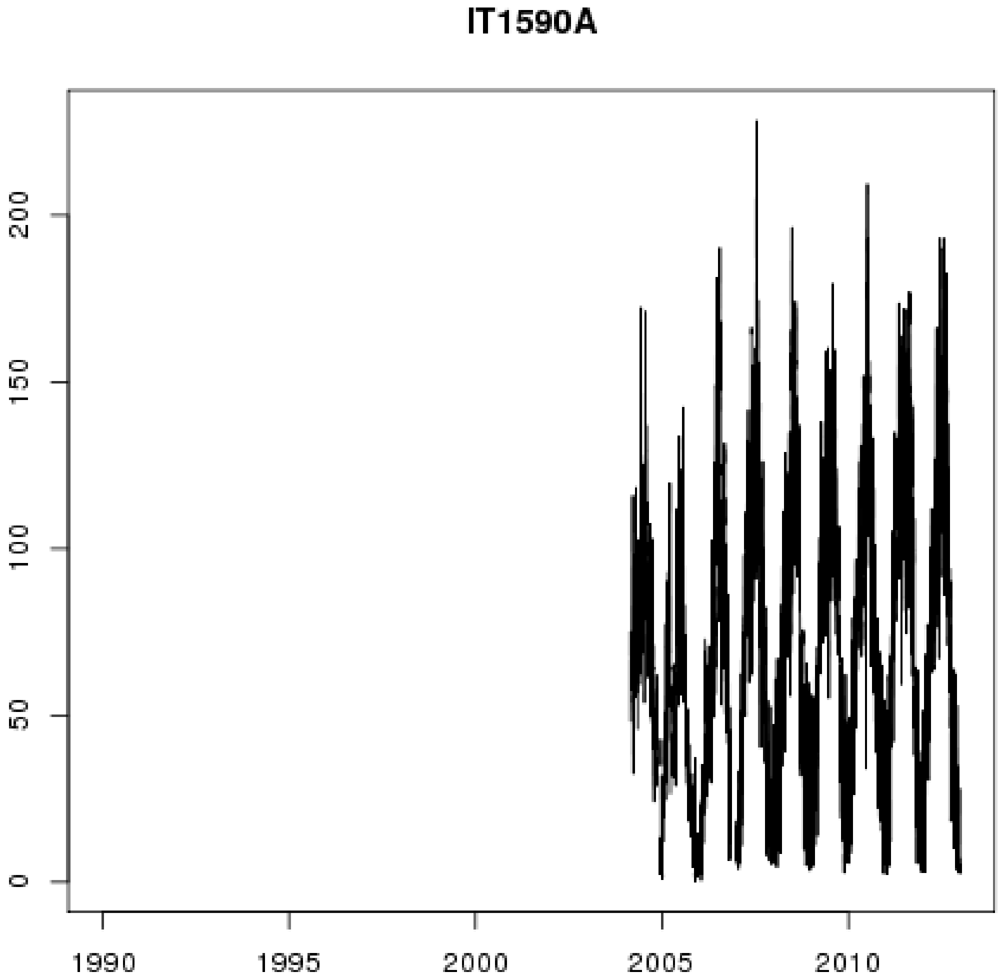


Figure 22: Time series of daily maxima O_3 (µg/m^3) at the IT1590A site.

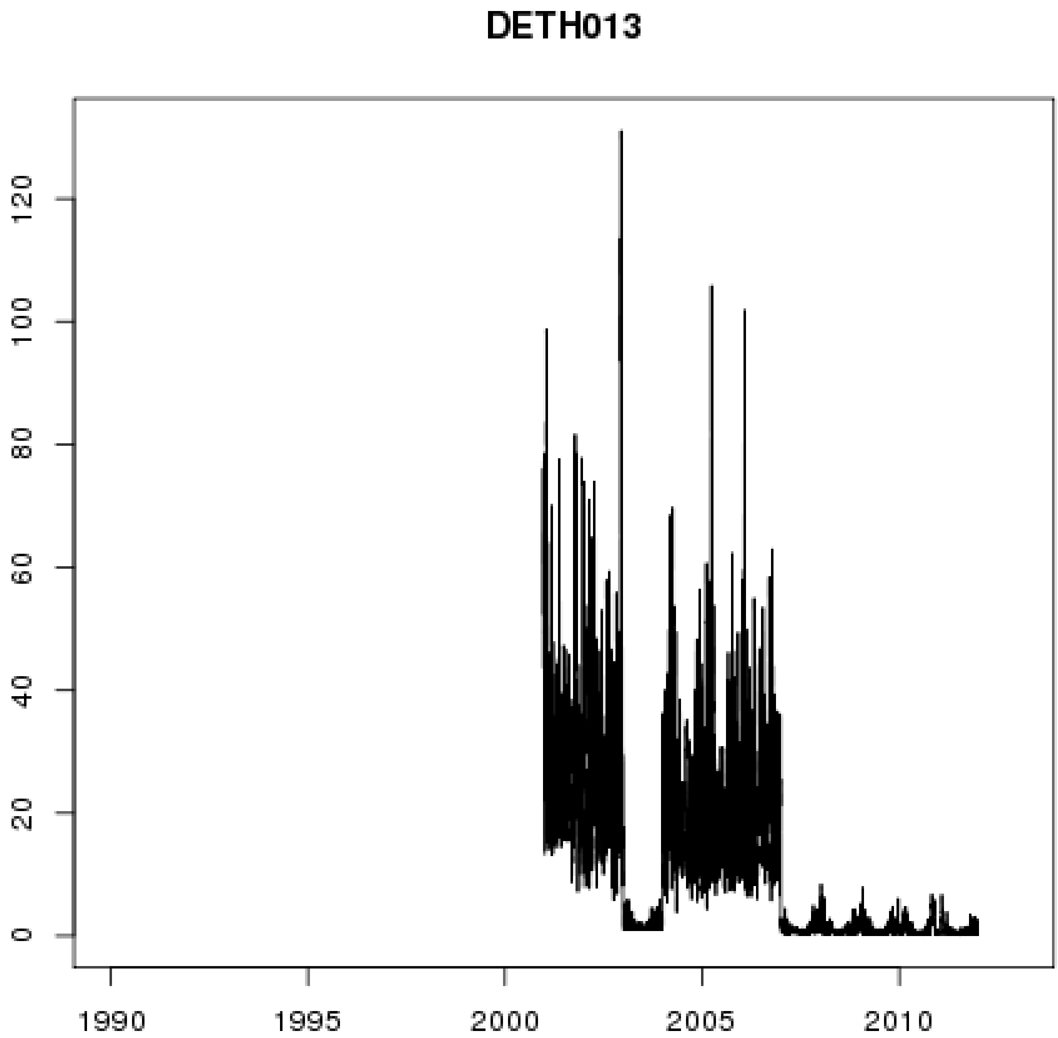


Figure 23: Time series of suspicious time series of daily mean PM10 (µg/m³) at the DETH013 site.