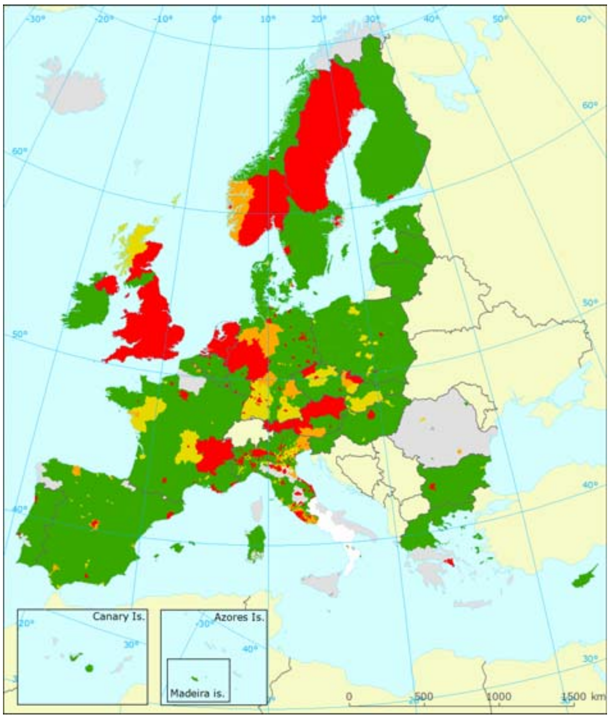
# Status and trends of $NO_2$ ambient concentrations in Europe

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European Topic Centre on Air and Climate Change

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***Front page picture:***
*Reported $NO_2$ annual concentrations in zones and agglomerations from 2006 to 2008, compared to the annual limit value (LV) of 40 µg/$m^3$ (Figure 6 of this report).*

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## DISCLAIMER




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# SUMMARY

Notwithstanding reduction of total $NO_x$ emissions in Europe, the trend in ambient air concentrations of $NO_2$ lags clearly behind the decreasing trend in the $NO_x$emissions. The present decrease in $NO_2$ concentrations is far from ensuring compliance with the $NO_2$ limit values (LV) in many European agglomerations and zones in the next few years. Exceedances of the annual $NO_2$ limit value (40 µg/m$^3$) were observed in nearly all countries at one or more stations in 2008. Further, compliance with this LV is expected for only a few of the cities which have been affected by exceedances in the last years, despite adopted measures both at local and at EU level. The main reasons for the non-achievement of compliance are assumed to be a late start of planning and implementation of measures and the underestimation of real world $NO_x$ emissions from road vehicles compared to legislative limits (emission standards), in combination with increasing primary $NO_2$ emissions from diesel vehicles, which lead to a significant overestimation of the emission reduction potential of the current measures.

Traffic stations across Europe have had both increasing and decreasing trends of the $NO_2$ annual mean in the past 10 years. In 25% of the traffic stations the trends were statistically significant and decreasing, especially in the North-west region (30%) and the North region (36%). For $NO_X$ annual mean concentrations, a significant decreasing trend was registered in 52% of the traffic stations. On the other hand, 8% of the European traffic stations registered a significant increasing trend for $NO_2$ (2% for $NO_X$), especially in the Central east region (11% for $NO_2$ and 2% for $NO_X$), but also in the North-west region (8% for $NO_2$, albeit 0% for $NO_X$) and in the South (6% for $NO_2$ and 1% for $NO_X$). For the majority of the background stations the trends are decreasing, 38% of the suburban- and 35% of the urban background stations had significant decreasing $NO_2$ annual mean trends, especially in the North-western region (56% and 47%, respectively). On the other hand, significant increasing trends were registered in 3% of the urban background stations in the Central and Eastern and Southern regions.

With the exception of industrial stations, an increase of the $NO_2/NO_X$ concentration ratio has been registered in average for all types of stations and especially at urban traffic sites. While the $NO_X$ concentrations in Europe show in average a downward trend at all types of stations and in all European regions, with the exception of the South[1], the proportion of $NO_X$ emitted directly as $NO_2$ from vehicles has been increasing since the early 2000s, as a result of an increased market penetration of diesel cars in some countries and the fitting of pollution control devices, e.g., particulate traps and oxidation catalysts for diesel EURO3 (and above). On the other hand, an increase of the $NO_2/NO_X$ concentration ratio must be expected when $NO_X$ concentrations decrease, simply due to a shift in the photostationary state, with constant ozone. The increase in the $NO_2/NO_X$ concentration ratio is therefore due to both an increase in the $NO_2/NO_X$ ratio of primary emissions and a decrease in $NO_X$, without an equivalent decrease in ozone concentrations.

[1] Note that there is a low number of stations (29) with $NO_X$ data available for the Southern region and most of the stations available in the region Centre and East are in Germany, which should be taken into consideration when analyzing the trends, which are considered to be geographically biased.

# CONTENTS

# INTRODUCTION

Notwithstanding reduction of total $NO_x$ emissions in Europe, the trend in ambient air concentrations of $NO_2$ lags clearly behind the decreasing trend in the $NO_x$ emissions. Current European legislation and emission standards aim at the reduction of $NO_x$ emissions, both from traffic (EURO standards) and from industrial sources (NEC Directive). These emission ceilings and standards do not differentiate between the $NO_2$ and NO fractions and regulate only the total $NO_x$ emissions. On the other hand, the CAFE Directive regulates the $NO_2$ concentrations in ambient air, to which the population is exposed. These two different legislative efforts to reduce oxides of nitrogen have apparently led to a gap between the $NO_x$ emission reduction trends and the $NO_2$ ambient air concentration trends in Europe. In addition, there is a gap between decreasing $NO_x$ concentration trends and $NO_2$ concentration trends in the EU27 that may be explained by: 1) the fact that measures to reduce the emissions of $CO_2$, CO, HC or particles from road traffic have had an impact on the primary $NO_2$ fraction emitted from vehicles (f-$NO_2$) and $NO_2/NO_x$ emission ratios; 2) the decrease in NOx, leading to an increase in the $NO_2/NO_x$ ratio, due to a shift in the photostationary state, with at least as much ozone as earlier available.

The present paper presents an analysis of the current $NO_2$ ambient levels (Chapter 1). The concentrations are compared with the limit values (to be met in 2010) and with the limit value (LV) plus margin of tolerance (MT). The analysis is based on the latest available data, which is the year of 2007 for the EMEP modelled data and the year 2008 for the analysis based exclusively on measurement data. The trends in the last 10 years in Europe, comparing with trends in $NO_x$ concentrations and emissions are discussed in Chapter 2. The paper further reviews several analyses of the reasons for these developments (Chapter 3) and of the applied measures for $NO_2$ compliance across Europe.

# 1. CURRENT STATUS OF NO2 CONCENTRATIONS IN EUROPE

An interpolated map of the $NO_2$ annual mean concentrations in 2007 across Europe is presented in Figure 1. The map is based on measured data, modelled concentrations with the EMEP model, population density, altitude and wind speed, both for rural and the urban background areas (Horálek et al., 2006). Figure 2 shows the $NO_2$ annual mean concentrations measured at European background stations in 2007 (AirBase – air quality database). The highest background concentrations are found in the Po Valley and in large cities, as well as in the Benelux countries and in the Rhein-Ruhr area. The main reason why some of the highest concentrations measured in background stations presented in Figure 2 are not seen in Figure 1 is that Figure 1's map resolution is 10 x 10 $km^2$ (i.e. calculated spatial average over the grid cell), while the geographic representativity of the urban and sub-urban background stations, where the highest concentrations are measured, is lower than 10 x 10 $km^2$. The other reason is that the kriging method used for the spatial interpolation slightly smoothes the values, in order to give better estimates in the points with no measurements. For details, see Horálek et. al (2006).

Figure 3 shows the $NO_2$ annual mean concentrations measured at European traffic and industrial stations in 2007 (AirBase). The highest concentrations are found at these types of stations. Exceedances occurred everywhere in Europe, especially in Italy, Germany and the Benelux countries, where the stations density is also higher.

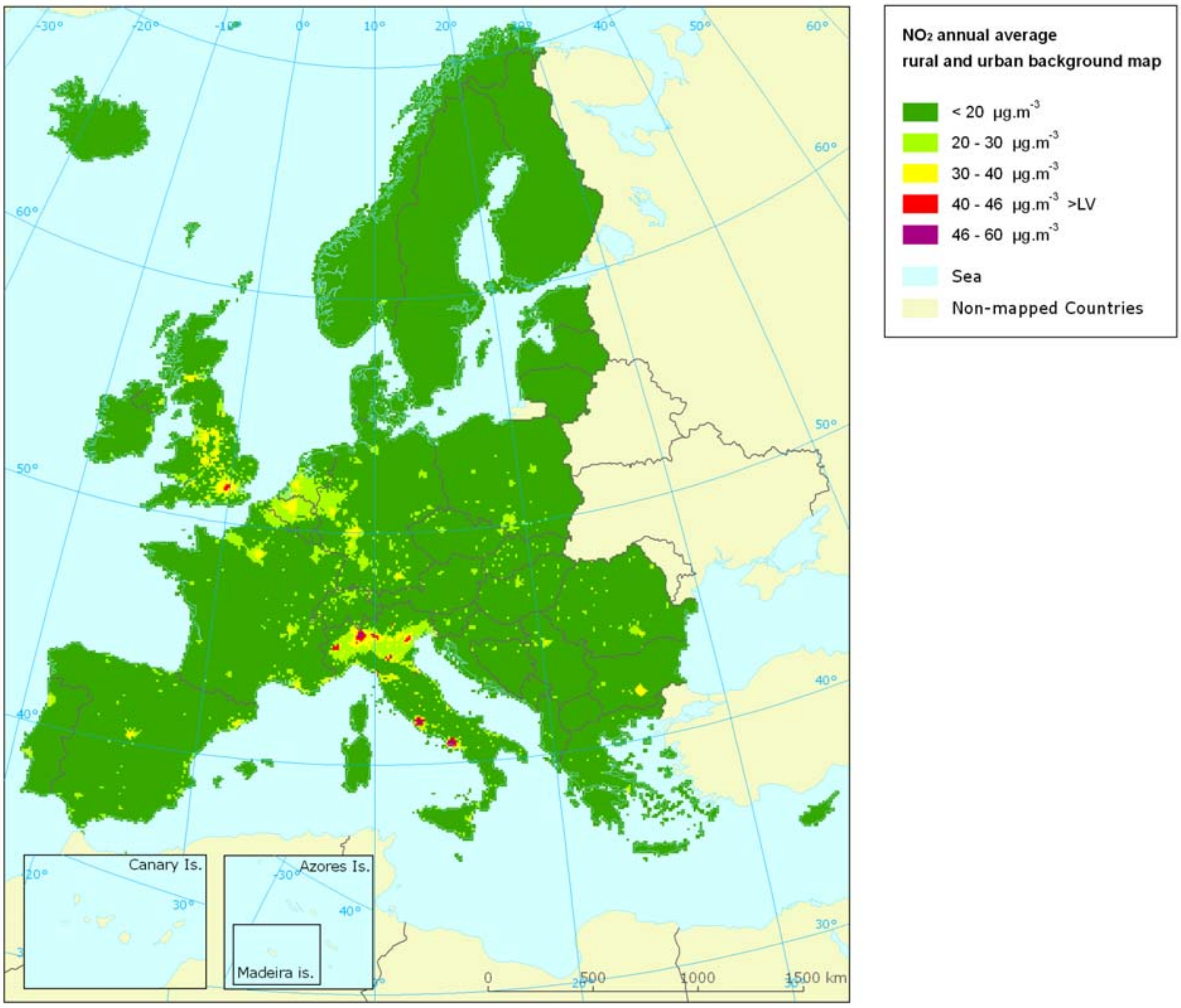



**Figure 1: Annual mean $NO_2$ concentrations over Europe in 2007 estimated with 10x10 $km^2$ resolution. Based on EMEP modelling and AirBase rural and urban background stations. The two highest concentration classes correspond to the limit value (40 µg/$m^3$) and limit value plus margin of tolerance in 2007 (46 µg/$m^3$), respectively.**

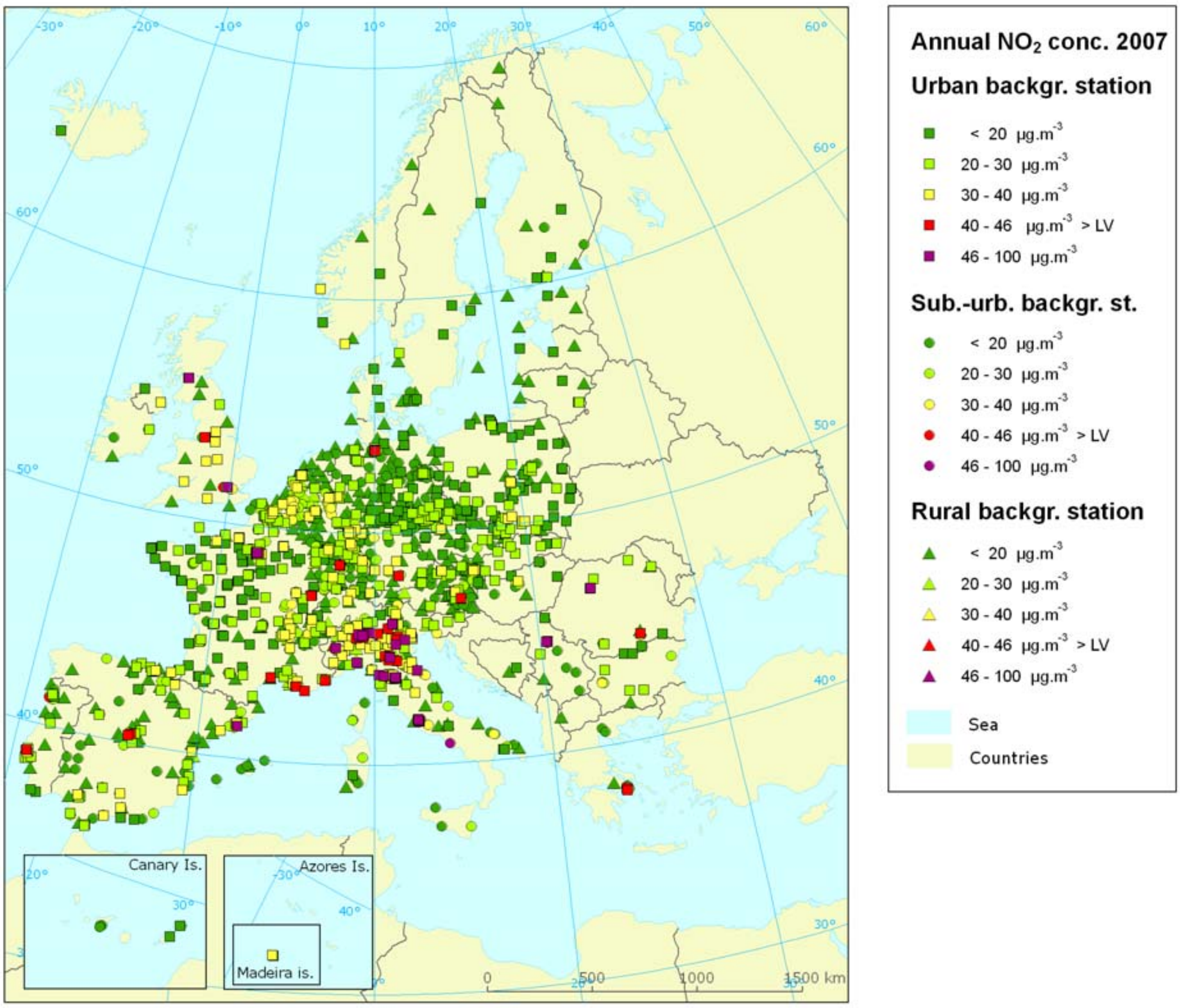



**Figure 2: Annual mean $NO_2$ concentrations over Europe in 2007. Based on AirBase background stations measurement data. The two highest concentration classes correspond to the limit value (40 µg/m$^3$) and limit value plus margin of tolerance in 2007 (46 µg/m$^3$), respectively.**

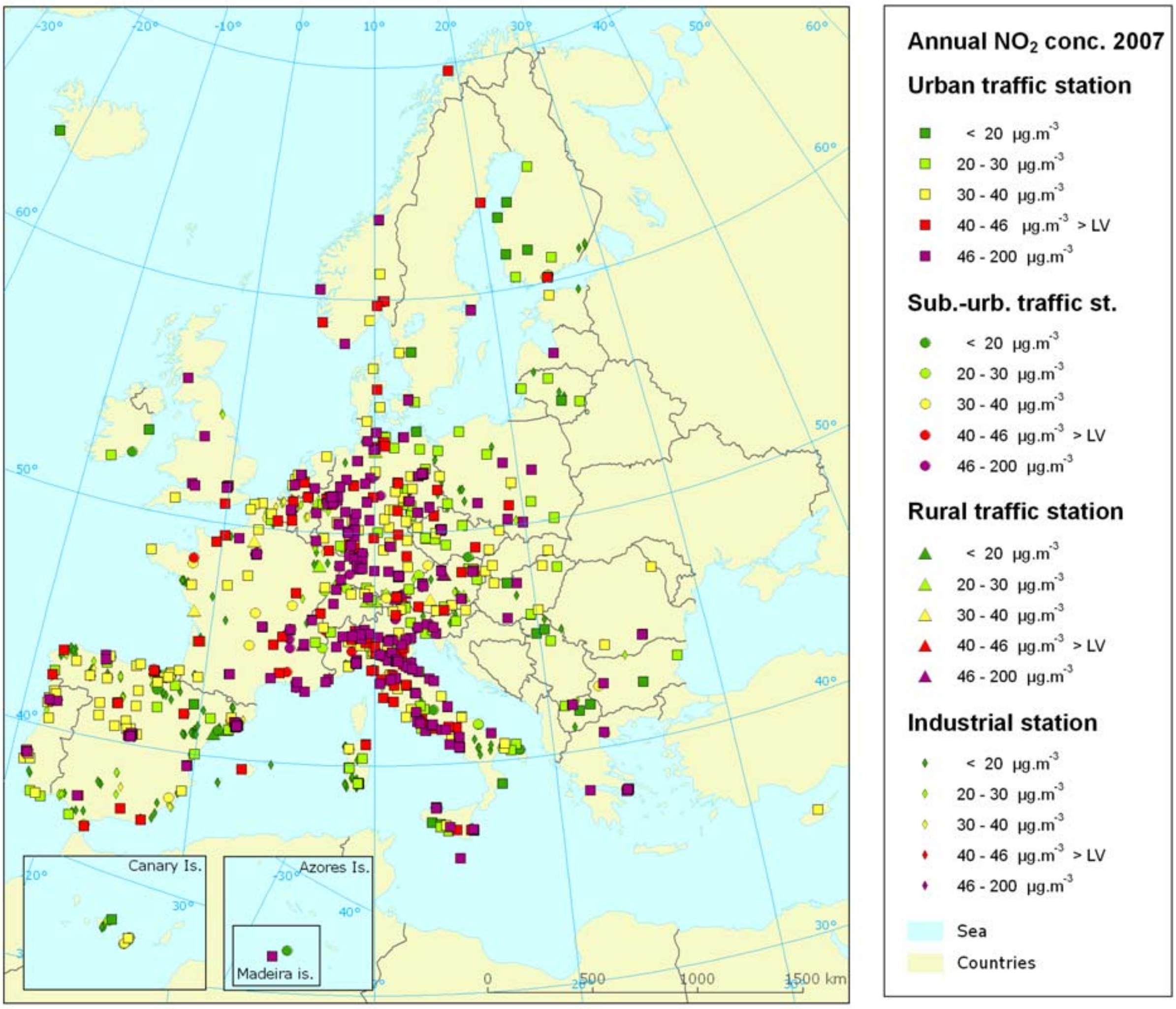



**Figure 3: Annual mean $NO_2$ concentrations over Europe in 2007. Based on AirBase traffic and industrial stations measurement data. The two highest concentration classes correspond to the limit value (40 µg/m$^3$) and limit value plus margin of tolerance in 2007 (46 µg/m$^3$), respectively.**

## 1.1. Distance to target

The targets for $NO_2$ are defined by the two limit values for the protection of human health:

- The long term limit value, corresponding to the annual mean limit value of 40 µg/m$^3$;
- The short term limit value, corresponding to the hourly mean limit value of 200 µg/m$^3$, which must not be exceeded more than 18 times a calendar year. To check the attainance of this limit value we refer to the 19$^{th}$ highest $NO_2$ hourly concentration.

*Exceedances in agglomerations and zones in 2008*

The annual mean limit value for $NO_2$ for the protection of human health is 40 µg/m$^3$ and had to be met in 2010. Before 2010, Member States had to meet this limit value plus a margin of tolerance (MT), which was 46 µg/m$^3$ in 2007 and 44 µg/m$^3$ in 2008. The hourly mean limit value for $NO_2$ is 200 µg/m$^3$ and must not be exceeded more than 18 times in a calendar year after 01.01.2010.

Based on the results of the Member States' (MS) reporting on ambient air quality assessment for 2008 (de Leeuw and Vixseboxse, 2009), 27% of the zones and agglomerations exceeded the annual mean limit value (LV) for $NO_2$ and 23% exceeded the limit value and margin of tolerance (44 µg/m$^3$ in 2008). In relation to the population, about 50% of the population of the EU27 live in a zone or agglomeration which was not in compliance with the health related $NO_2$ annual mean LV, in 21 Member States.

In terms of the hourly mean $NO_2$ limit value, 5% of the zones and agglomerations exceeded this LV in 2008, while 3% exceeded the (LV + MT) in 2008. About 13% of the population was exposed to levels above the health related hourly mean LV of $NO_2$ in 2008.

The member states' reported reasons for $NO_2$ annual mean LV exceedances are mostly attributed to local traffic (over 2/3), domestic heating, local industry and power generation, accidental industrial emissions and others (Vixseboxse and de Leeuw, 2009).

*Exceedances at monitoring stations in 2008*

Figure 4 shows the annual mean concentrations of $NO_2$ measured in 2008, including traffic stations. Exceedances of the $NO_2$ annual mean LV were observed in nearly all countries at one or more stations in 2008. The distance-to-target plots in Figure 4 show, both for the hourly and annual mean LVs, that the exceedances are observed most frequently at traffic stations, while the LVs are not exceeded at the rural background stations. The annual mean $NO_2$ LV was exceeded of 46% of the traffic stations, while the LV+MT (44 µg/m$^3$) was exceeded at 16% of the traffic stations (Mol et al., 2010). The hourly limit value of $NO_2$ is less stringent, with exceedances at about 1 and 6% of the (sub)urban and traffic stations, respectively. (Mol et al., 2010).

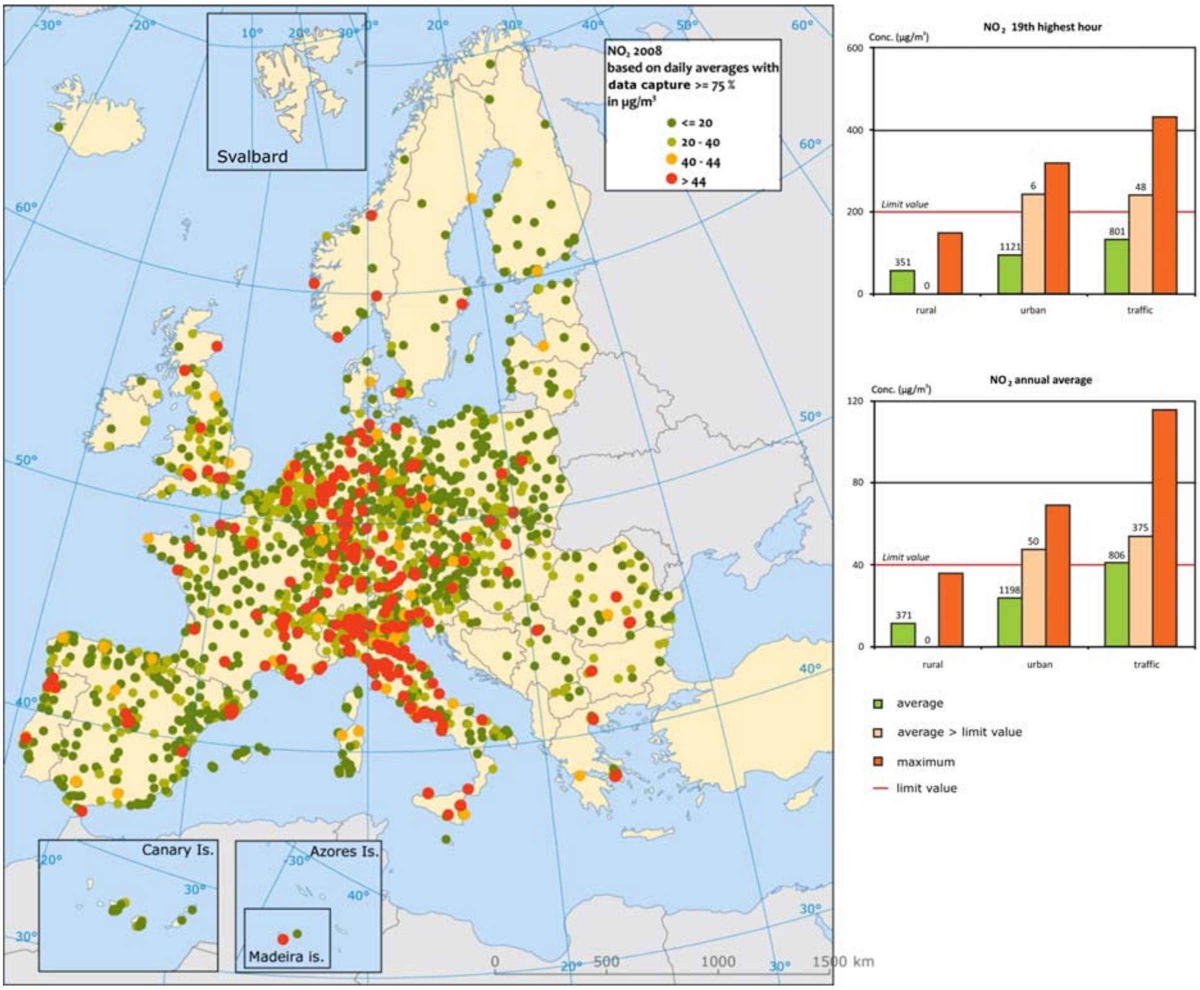



**Figure 4: Left map - Annual mean concentration map of $NO_2$ (µg/m³); the two highest concentration classes correspond to the limit value (40 µg/m³) and limit value plus margin of tolerance (44 µg/m³), respectively. From Mol et al. (2010). Right: distance to target plots for the hourly mean (top) and annual mean (bottom) $NO_2$ limit values. Numbers over the bars indicate the number of stations under (green) or over (pink) limit values.**

## 1.2. Persistent exceedances from 2006 to 2008

*Zones and agglomerations with persistent exceedances*

The persistence over the last three years (2006-2008) of the exceedances of the $NO_2$ annual and hourly mean limit values in zones and agglomerations, reported by the Member States, are shown in Figure 5 and Figure 7, respectively. Figure 6 and Figure 8 show the same, but compared to the limit values plus margin of tolerance.

Figure 5 shows that the exceedances of the annual mean $NO_2$ LV and of the LV + MT have been persistent over a considerable number of zones and agglomeration throughout Europe. Figure 8 shows, on the other hand, that the exceedances of the hourly $NO_2$ LV are less persistent, with the exception of the agglomerations around about 15 major European cities.

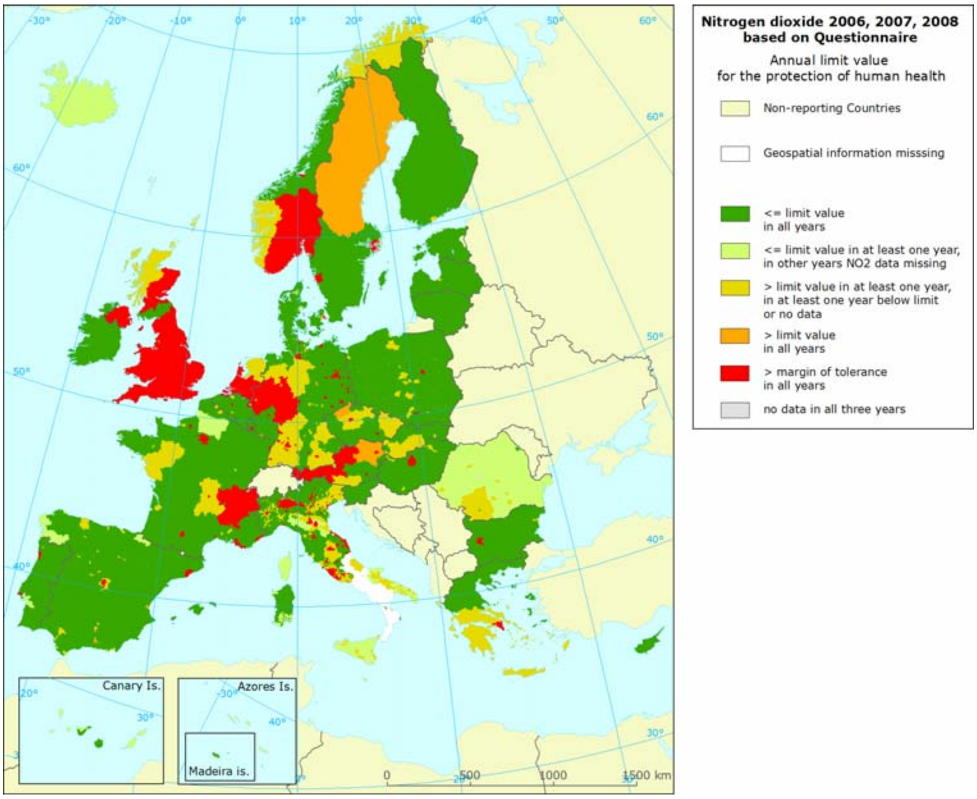



**Figure 5: Reported $NO_2$ annual concentrations in zones and agglomerations from 2006 to 2008, compared to the annual limit value (LV) of 40 µg/m$^3$ and LV plus margin of tolerance (MT).**

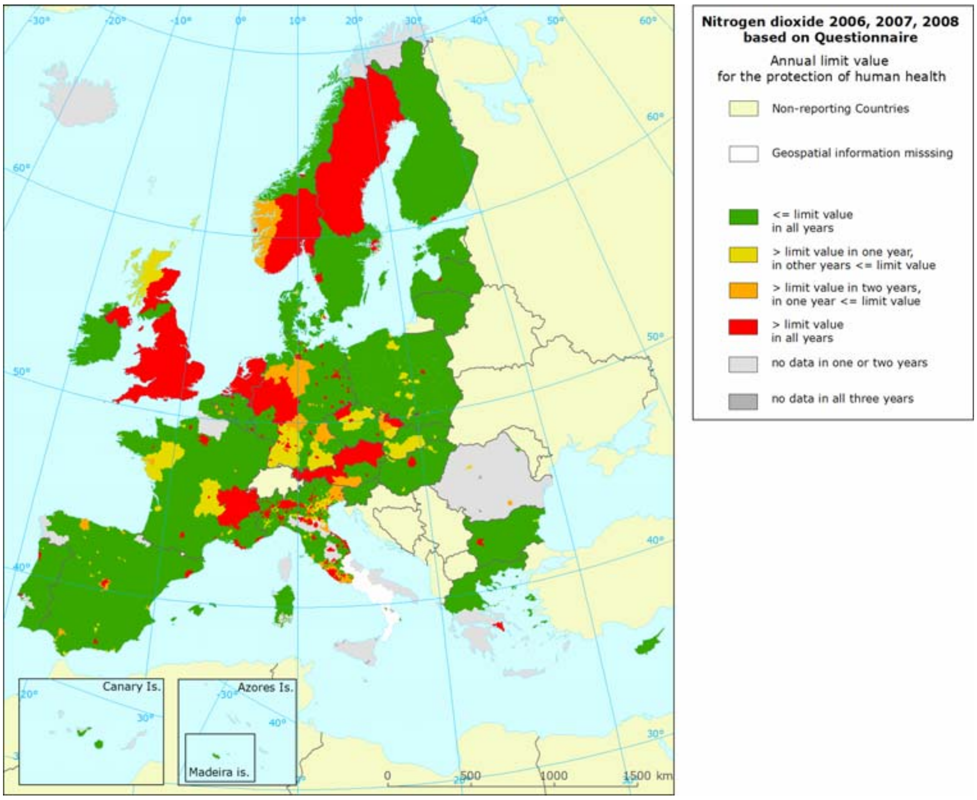



**Figure 6: Reported $NO_2$ annual concentrations in zones and agglomerations from 2006 to 2008, compared to the annual limit value (LV) of 40 µg/m$^3$.**

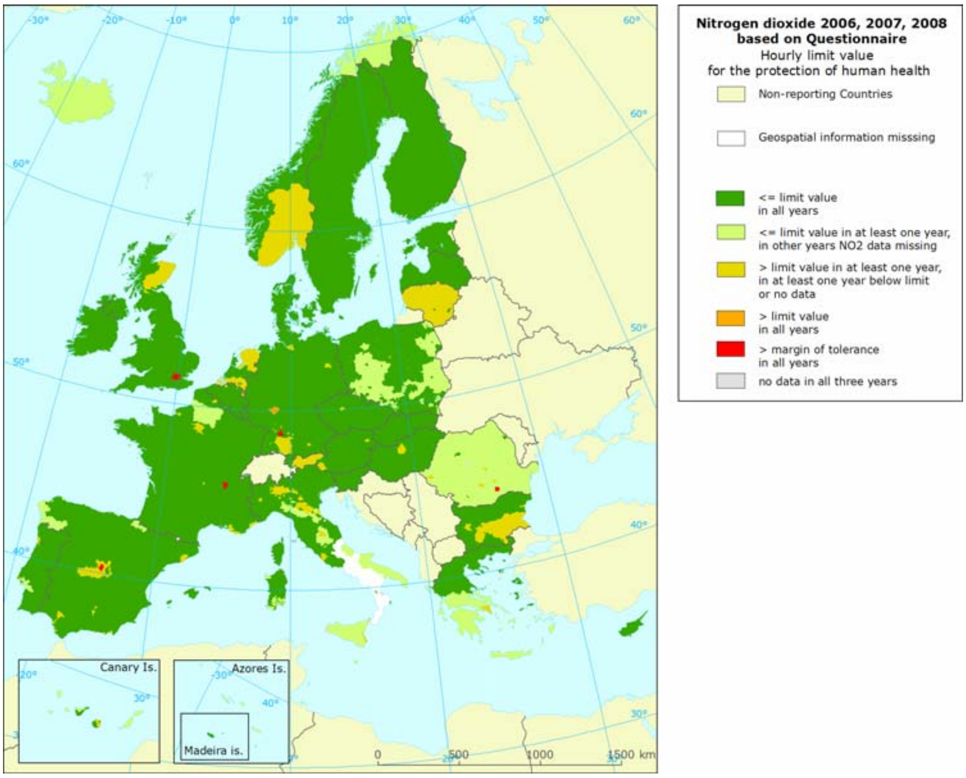



**Figure 7: Reported 19th highest hourly $NO_2$ concentrations in zones and agglomerations from 2006 to 2008, compared to the hourly limit value of 200 µg/m$^3$ and LV plus margin of tolerance (MT).**

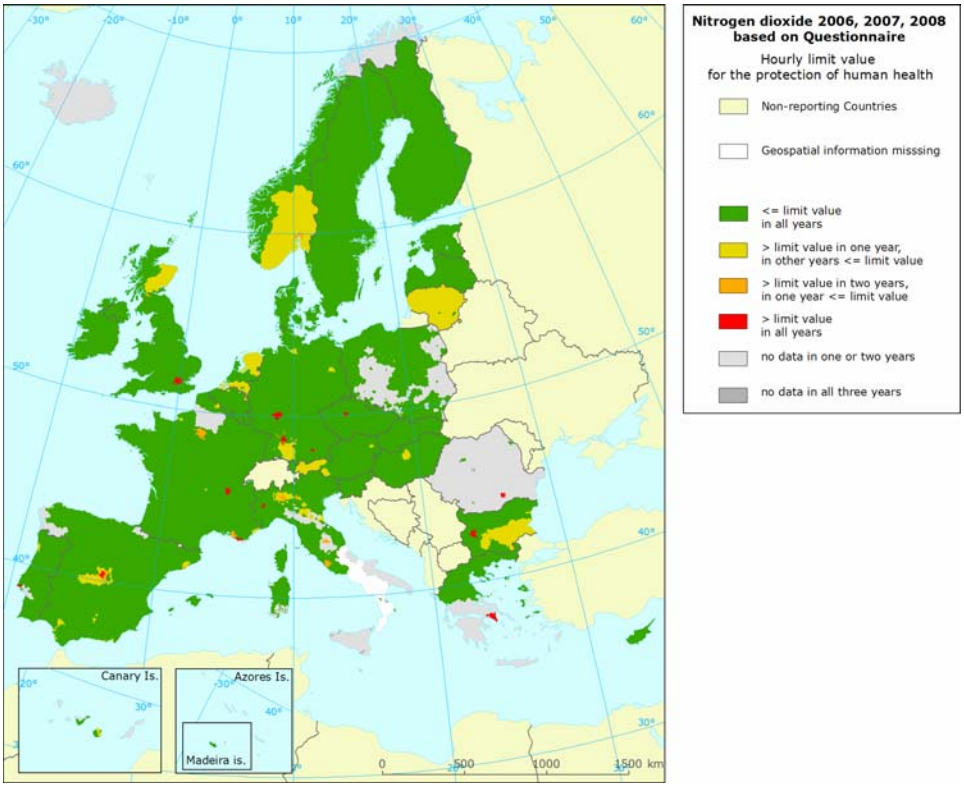



**Figure 8: Reported 19th highest hourly $NO_2$ concentrations in zones and agglomerations from 2006 to 2008, compared to the hourly limit value of 200 µg/m$^3$.**

*Hot spot stations with persistent exceedances*

The persistence of the $NO_2$ annual mean LV exceedances is also shown by the analysis of the AirBase monitoring data for 2006 to 2008. Mol et al. (2010) note that the 3-year averaged $NO_2$ concentrations are above the limit value at more than 420 stations operational in the period 2006-2008. These long-lasting exceedances are observed in 21 Member States, mostly at traffic stations (83%), but also at (sub)urban background stations (13%) and industrial stations (4%) (Mol et al., 2010).

On the other hand, the analysis of the AirBase measurement data confirms that the exceedances of the hourly $NO_2$ LV are less common and less persistent. Mol et al. (2010) note that the averaged number of exceedances of the hourly $NO_2$ LV (200 µg/m$^3$) over the last three years exceeds the allowed number of 18 at a limited number of stations (59 traffic, 8 (sub)urban background and 5 industrial).

Table 1 and
Table 2 show the list of the 25 monitoring stations with the highest concentrations of $NO_2$ annual and 19$^{th}$ highest hourly concentrations, respectively, over the three years. As one can see from Table 1, the 25 stations with the highest annual mean concentrations are traffic stations in urban or suburban areas, within or close to major European cities. The cities are distributed within 7 countries: Stuttgart and München in Germany; London in the UK; Bucharest in Romania; Paris, Marseille and Lyon in France; Athens in Greece; Firenze, Roma, Perugia, Genoa, Torino, Milano and Pescara in Italy and Madrid in Spain. The $NO_2$ annual means measured at these stations are between 185 to 280 % of the LV.

Table 2 shows that 25 stations with the 19$^{th}$ highest hourly $NO_2$ concentration over the last three years are not only traffic stations in urban or suburban areas, but also two industrial urban stations and even one urban background station, the latter in Belgrade. Getafe, Sofia, Prague, Toulon, Olbia and Lisboa are among the cities with the highest hourly $NO_2$ concentrations, which were not among the cities with highest annual means. The 25 highest 19$^{th}$ highest hourly concentrations are 14 to 80% higher than the LV. Thirteen stations are listed in both Table 1 and
Table 2. These 13 stations are all traffic stations and are located in the following cities: Stuttgart, Madrid, Paris, Lyon, London, Athens, Milano, Torino and Bucharest.

## Table 1: Worst 25 hot spot stations, based on the 2006, 2007 and 2008 $NO_2$ annual mean concentrations.

| Station name | Annual mean (µg/m3) 2006 | 2007 | 2008 | average | Type of station | area | city | Country | Street type | EoI code |
|---|---|---|---|---|---|---|---|---|---|---|
| Stuttgart Am Neckartor | 121 | 106 | 107 | 111 | Traffic | urban | Stuttgart | DE | Unknown | DEBW118 |
| London Marylebone road | 111 | 102 | 115 | 109 | Traffic | urban | London | UK | Unknown | GB0682A |
| Cercul Militar | 126 | 111 | 79 | 105 | Traffic | urban | Bucharest | RO | Unknown | RO0070A |
| Bd Periph Auteuil | 99 | 104 | 105 | 103 | Traffic | suburb. | Paris | FR | Unknown | FR04053 |
| Stuttgart Hohenheimer Straße (S) | 104 | 98 | 98 | 100 | Traffic | urban | Stuttgart | DE | Unknown | DE1605A |
| Place Victor Basch | 94 | 96 | 91 | 94 | Traffic | urban | Paris | FR | Unknown | FR04012 |
| Patision | 85 | 100 | 92 | 93 | Traffic | urban | Athens | GR | Canyon st.: L/H < 1.5 | GR0032A |
| München/Landshuter Allee | 98 | 89 | 85 | 91 | Traffic | urban | München | DE | Unknown | DEBY115 |
| Auto A1 -Saint-Denis | 91 | 91 | 89 | 90 | Traffic | suburb. | Saint-Denis, Paris | FR | Unknown | FR04058 |
| Marseille_Plombieres | 88 | 82 | 82 | 84 | Traffic | suburb. | Marseille | FR | Unknown | FR03004 |
| FI-GRAMSCI 904811 | 72 | 83 | 93 | 82 | Traffic | urban | Firenze | IT | Unknown | IT0861A |
| C.SO Francia 1205802 | 83 | 84 | 79 | 82 | Traffic | urban | Roma | IT | Wide st.: L/H > 1.5 | IT0825A |
| ES0116A-Marañón | 86 | 80 | 79 | 81 | Traffic | urban | Madrid | ES | Unknown | ES0116A |
| A7 Sud Lyonnais | 81 | 83 | 79 | 81 | Traffic | suburb. | Lyon | FR | Unknown | FR20013 |
| Fontivegge 1005402 | 87 | 83 | 71 | 80 | Traffic | urban | Perugia | IT | Wide st.: L/H > 1.5 | IT1101A |
| Ludwigsburg Friedrichstraße (S) | 80 | 81 | 76 | 79 | Traffic | urban | Ludwigsburg, Stuttgard | DE | Unknown | DEBW117 |
| Giardini Melis - Genova 701027 | 70 | 78 | 85 | 78 | Traffic | urban | Genova | IT | Unknown | IT1479A |
| Stuttgart-Mitte-Straße | 82 | 75 | 74 | 77 | Traffic | urban | Stuttgart | DE | Wide st.: L/H > 1.5 | DEBW099 |
| TO_1272_TO_Rebauden 100110 | 94 | 71 | 66 | 77 | Traffic | urban | Torino | IT | Wide st.: L/H > 1.5 | IT0470A |
| Milano - V.Le Marche 301526 | 78 | 76 | 74 | 76 | Traffic | urban | Milano | IT | Wide st.: L/H > 1.5 | IT0477A |
| Milano Via Zavattari 301544 | 76 | 73 | 78 | 76 | Traffic | urban | Milano | IT | Wide st.: L/H > 1.5 | IT0467A |
| Camden Kerbside | 72 | 77 | 76 | 75 | Traffic | urban | London | UK | Unknown | GB0636A |
| München/Stachus | 79 | 71 | 74 | 74 | Traffic | urban | München | DE | Unknown | DEBY037 |
| PE - Corso Vit. Emanuele 1306807 | 79 | 70 | 74 | 74 | Traffic | urban | Pescara | IT | Canyon st.: L/H < 1.5 | IT1422A |
| London Cromwell road 2 | 83 | 72 | 67 | 74 | Traffic | urban | London | UK | Unknown | GB0695A |

L/H is the street width to buildings' height ratio. Street canyons have L/H<1,5 and wide streets have L/H > 1,5.

## Table 2: Worst 25 hot spot stations, based on the 2006, 2007 and 2008 19th highest hourly $NO_2$ concentrations.

| Station name | 19th highest hourly conc. (µg/m3) 2006 | 2007 | 2008 | average | Type of station | area | city | Country | Street type | EoI code |
|---|---|---|---|---|---|---|---|---|---|---|
| Cercul Militar | 429 | 365 | 276 | 357 | Traffic | urban | Bucharest | RO | Unknown | RO0070A |
| ES0116A-Marañón | 332 | 311 | 338 | 327 | Traffic | urban | Madrid | ES | Unknown | ES0116A |
| Drumul Taberei | | 342 | 305 | 324 | Industrial | urban | Bucharest | RO | Unknown | RO0069A |
| Mihai Bravu | 378 | 286 | 283 | 316 | Traffic | urban | Bucharest | RO | Unknown | RO0067A |
| London Marylebone road | 294 | 300 | 319 | 304 | Traffic | urban | London | GB | Unknown | GB0682A |
| Stuttgart Am Neckartor | 317 | 262 | 268 | 282 | Traffic | urban | Stuttgart | DE | Unknown | DEBW118 |
| Stuttgart Hohenheimer Straße (S) | 293 | 260 | 260 | 271 | Traffic | urban | Stuttgart | DE | Unknown | DEBW116 |
| ES1192A-Alcalá Final | 244 | 296 | 231 | 257 | Traffic | urban | Madrid | ES | Unknown | ES1192A |
| Bd Periph Auteuil | 245 | 258 | 263 | 255 | Traffic | suburban | Paris | FR | Unknown | FR04053 |
| ES1804A-Getafe | | 272 | 229 | 251 | Traffic | urban | Getafe | ES | Unknown | ES1804A |
| AMS Orlov most-Sofia | 293 | 205 | 248 | 248 | Traffic | urban | Sofia | BG | Wide st.: L/H > 1.5 | BG0054A |
| TO_1272_TO_Rebauden 100110 | 276 | 261 | 199 | 245 | Traffic | urban | Torino | IT | Wide st.: L/H > 1.5 | IT0470A |
| ES1521A-Barrio del Pilar | 230 | 249 | 251 | 243 | Traffic | urban | Madrid | ES | Unknown | ES1521A |
| Milano - V.Le Marche 301526 | 273 | 235 | 218 | 242 | Traffic | urban | Milano | IT | Wide st.: L/H > 1.5 | IT0477A |
| Omladinskih brigada | 117 | 311 | 290 | 239 | Background | urban | Belgrade | RS | Wide st.: L/H > 1.5 | RS0007A |
| Pha2-Legerova | 228 | 257 | 230 | 239 | Traffic | urban | Prague | CZ | Wide st.: L/H > 1.5 | CZ0ALEG |
| Berceni | 216 | 216 | 282 | 238 | Industrial | urban | Bucharest | RO | Unknown | RO0068A |
| Toulon_Foch | 228 | 258 | 225 | 237 | Traffic | urban | Toulon | FR | Unknown | FR03068 |
| A7 Sud Lyonnais | 242 | 237 | 229 | 236 | Traffic | suburban | La Mulatière, Paris | FR | Unknown | FR20013 |
| Patision | 216 | 269 | 221 | 235 | Traffic | urban | Athens | GR | Canyon st.: L/H < 1.5 | GR0032A |
| Milano Via Zavattari 301544 | 240 | 239 | 220 | 233 | Traffic | urban | Milano | IT | Wide st.: L/H > 1.5 | IT0467A |
| Camden Kerbside | 216 | 258 | 223 | 232 | Traffic | urban | London | GB | Unknown | GB0636A |
| Place Victor Basch | 240 | 236 | 212 | 229 | Traffic | urban | Paris | FR | Unknown | FR04012 |
| CENS09 2009016 | 292 | 172 | 222 | 229 | Traffic | urban | Olbia, Sardegna | IT | Wide st.: L/H > 1.5 | IT1308A |
| Avenida da Liberdade | 258 | 220 | 204 | 227 | Traffic | urban | Lisboa | PT | Wide st.: L/H > 1.5 | PT03075 |

L/H is the street width to buildings' height ratio. Street canyons have L/H<1,5 and wide streets have L/H > 1,5.

# 2. TRENDS

In order to analyse the development in $NO_2$ and $NO_x$ concentrations in Europe, and relate to the $NO_x$ emission development, $NO_2$ and $NO_x$ concentrations measured between 1999 and 2008 were analyzed in regard to possible trends. The development in the number of stations and zones above the limit value and the margin of tolerance was also investigated. In addition, a trend analysis of the $NO_2/NO_x$ ratio was performed, to identify possible changes due to an increase in the primary $NO_2$ emissions ratio.

In order to find the general trends, a trend analysis was done for monitoring stations aggregated by type/area of station and within four European regions. In addition, trends were calculated for each individual station, in order to give information of the variability of trends within each group. More information on the methodology and input data used in the calculation of the trends, both for the average of stations and for the individual stations, is given in Annex A. Note that more stations were used for the individual stations trend analysis, compared to the average trend analysis. While only 8 years with 75% data capture was required for the individual stations trend analysis (stations in yellow and pink in Figure 9), 10 years with 75% data capture was required for the average trend analysis (stations in pink in Figure 9). These two different trend analyses are therefore not directly comparable.

## 2.1. Trend of $NO_2$

### 2.1.1 Trend at aggregated station level

The $NO_2$ indicators, annual mean and 19$^{th}$ maximum hourly mean, were calculated for each year, for different station area/types and for four European regions, as well as for the whole set of the stations with data capture over 75% for all the 10 years. Figure 9 shows in yellow the stations used for the $NO_2$ annual average trend analysis, as well as the European regions used in the analysis. The map with the stations used for the trend analysis of the 19$^{th}$ maximum hourly mean is very similar.

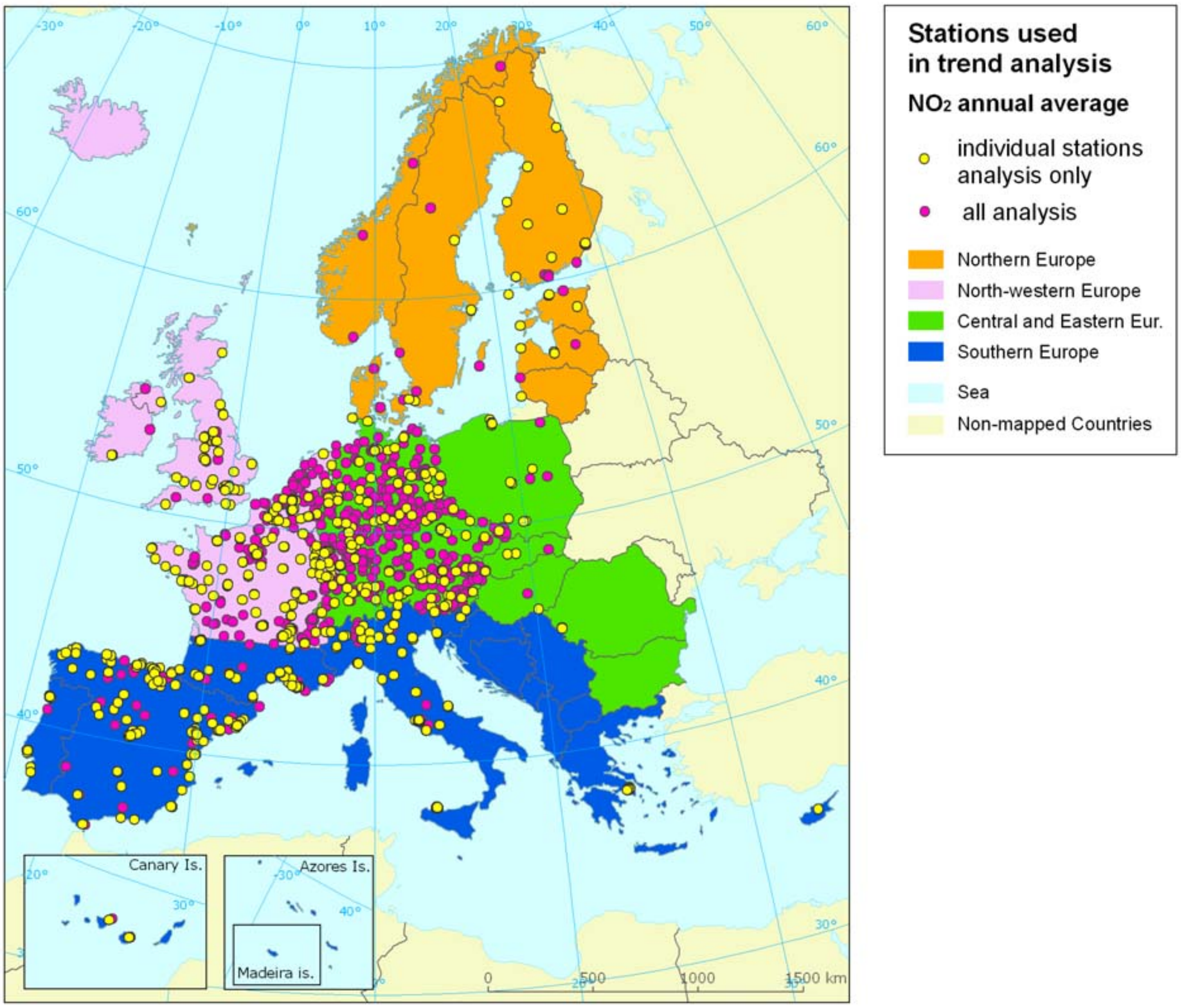



**Figure 9: Stations used in trend analysis of the $NO_2$ annual mean, both for the average of stations (stations in pink) and for the individual stations (all stations, both pink and yellow), and the four European regions used in the analyses.**

Table 3 and Table 4 present the calculated average $NO_2$ annual means and 19$^{th}$ maximum hourly means, respectively, as well as the estimated Sen's slope and the result of Mann-Kendall's test (Gilbert, 1987) for testing the presence of the trend.

**Table 3:** ***Trend analysis for $NO_2$ annual mean, for the years 1999-2008, for all stations together and separately for different station/area types and regions. The number of stations in relevant group, the average concentration values of the group (in µg.m$^{-3}$) for individual years, the estimated Sen's slope and the result of Mann-Kendall's test for testing of the presence of the monotonic increasing or decreasing trend are presented. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$.***

| group of the stations | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | [1]signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 755 | 29,8 | 28,5 | 28,2 | 28,1 | 30,1 | 27,7 | 28,1 | 28,4 | 26,9 | 26,2 | -0,27 | * |
| rural background | 158 | 13,0 | 12,6 | 12,5 | 12,5 | 13,5 | 12,3 | 12,2 | 12,5 | 11,6 | 11,5 | -0,14 | ** |
| suburban background | 159 | 26,1 | 25,2 | 24,7 | 24,5 | 26,4 | 24,0 | 24,3 | 24,3 | 22,8 | 22,4 | -0,35 | ** |
| urban background | 178 | 30,5 | 28,5 | 28,5 | 28,0 | 30,2 | 27,4 | 27,9 | 27,8 | 26,2 | 25,6 | -0,38 | ** |
| traffic (all area types) | 195 | 47,3 | 45,6 | 44,9 | 45,1 | 47,8 | 44,5 | 45,6 | 46,7 | 44,4 | 43,2 | -0,25 | no |
| industrial (all area types) | 64 | 24,9 | 24,3 | 24,8 | 24,4 | 26,0 | 23,9 | 24,3 | 24,3 | 23,4 | 22,5 | -0,18 | * |
| Northern Europe | 24 | 14,9 | 13,7 | 13,7 | 14,3 | 14,3 | 14,1 | 13,7 | 14,6 | 13,0 | 12,6 | -0,16 | no |
| North-western Europe | 207 | 33,7 | 32,3 | 32,2 | 31,4 | 34,4 | 31,3 | 31,3 | 30,6 | 29,7 | 29,3 | -0,42 | ** |
| Central and Eastern Eur. | 418 | 26,9 | 25,7 | 25,4 | 25,6 | 27,6 | 25,0 | 25,7 | 26,6 | 24,4 | 23,9 | -0,21 | no |
| Southern Europe | 106 | 36,7 | 35,7 | 35,2 | 34,8 | 35,1 | 34,3 | 34,7 | 34,7 | 34,5 | 32,6 | -0,27 | ** |
| rural background - N | 13 | 3,4 | 3,2 | 3,1 | 3,4 | 3,5 | 3,4 | 3,3 | 3,6 | 2,8 | 2,8 | -0,05 | no |
| rural background - NW | 31 | 19,6 | 18,9 | 19,5 | 18,9 | 21,3 | 19,1 | 18,5 | 18,0 | 17,3 | 17,3 | -0,26 | * |
| rural background - CE | 100 | 12,1 | 11,8 | 11,5 | 11,7 | 12,4 | 11,3 | 11,4 | 11,9 | 10,8 | 10,8 | -0,12 | * |
| rural background - S | 14 | 13,9 | 13,4 | 12,9 | 11,7 | 13,3 | 12,7 | 12,3 | 12,7 | 12,7 | 11,9 | -0,15 | + |
| suburban backgr. - N | 0 | no station with complete data | | | | | | | | | | | |
| suburban backgr. - NW | 62 | 29,5 | 28,1 | 27,9 | 27,1 | 29,6 | 26,5 | 26,5 | 25,6 | 24,6 | 24,1 | -0,53 | ** |
| suburban backgr. - CE | 85 | 23,1 | 22,3 | 21,6 | 21,8 | 23,6 | 21,5 | 22,0 | 22,8 | 20,5 | 20,6 | -0,21 | no |
| suburban backgr. - S | 12 | 30,2 | 31,1 | 29,8 | 29,5 | 29,5 | 29,6 | 29,5 | 29,2 | 29,2 | 25,8 | -0,18 | ** |
| urban background - N | 5 | 23,2 | 21,5 | 21,6 | 21,4 | 22,2 | 21,4 | 20,6 | 21,8 | 19,1 | 18,0 | -0,41 | * |
| urban background - NW | 52 | 33,6 | 31,5 | 31,4 | 30,3 | 32,8 | 29,7 | 29,5 | 28,6 | 27,9 | 27,7 | -0,55 | *** |
| urban background - CE | 102 | 29,3 | 27,5 | 27,5 | 27,2 | 29,3 | 26,4 | 27,1 | 27,6 | 25,1 | 24,7 | -0,36 | * |
| urban background - S | 19 | 30,3 | 27,6 | 27,7 | 28,4 | 30,0 | 28,4 | 29,3 | 28,3 | 29,0 | 26,5 | -0,06 | no |
| industrial - N | 1 | 7,5 | 6,9 | 7,6 | 6,9 | 7,6 | 8,9 | 9,6 | 11,1 | 14,1 | 13,1 | 0,77 | ** |
| industrial - NW | 23 | 28,0 | 27,8 | 28,8 | 28,1 | 30,6 | 27,5 | 27,6 | 27,2 | 26,0 | 25,0 | -0,30 | * |
| industrial - CE | 25 | 23,3 | 23,0 | 22,8 | 23,0 | 24,8 | 22,0 | 22,4 | 23,9 | 22,2 | 21,8 | -0,14 | no |
| industrial - S | 15 | 23,8 | 22,4 | 23,0 | 22,0 | 22,1 | 22,6 | 23,5 | 21,5 | 22,1 | 20,6 | -0,19 | + |
| traffic - N | 5 | 37,8 | 34,4 | 34,7 | 37,2 | 36,0 | 35,5 | 34,7 | 36,8 | 33,1 | 32,6 | -0,45 | no |
| traffic - NW | 38 | 55,8 | 53,8 | 52,7 | 52,3 | 57,4 | 53,5 | 54,5 | 53,8 | 52,6 | 52,1 | -0,29 | no |
| traffic - CE | 106 | 42,5 | 40,6 | 40,1 | 40,7 | 44,2 | 40,1 | 41,5 | 43,2 | 40,0 | 38,7 | -0,17 | no |
| traffic - S | 46 | 52,1 | 51,4 | 50,5 | 50,1 | 49,5 | 48,4 | 48,8 | 49,8 | 48,8 | 47,2 | -0,48 | ** |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1(meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

## It can be seen from

Table 3 that on average the $NO_2$ annual mean levels decreased by 0,27 µg.m$^{-3}$ per annum between 1999 and 2008. For most station types and regions there is a weak decreasing trend in the $NO_2$ annual means between 1999 and 2008. No significant trend is detected for: 1) the rural background stations of the Northern region; 2) the urban background stations of the Southern region; 3) the suburban background and 4) industrial stations of the Central and Eastern region.

The suburban background and industrial stations of the Northern region have no complete time series data or only have available data for one station, respectively. This single industrial station shows the increasing trend.

For the regions and type of stations where a trend is detected, the trend is negative and varies between -0.12 and -0.55 µg.m$^{-3}$/year. The strongest trend is detected at the urban background stations of the North-western region. These decreasing trends are deemed as too weak to ensure compliance in the non-compliant zones and agglomerations, especially at the traffic sites.

In Table 4 the results for the 19$^{th}$ maximum $NO_2$ hourly mean are presented.

**Table 4:** ***Trend analysis for the 19$^{th}$ maximum $NO_2$ hourly mean, for the years 1999-2008, for all stations together and separately for different station/area types and regions. The number of stations, the average concentration values of the group (in µg.m$^{-3}$) for individual years, the estimated Sen's slope and the result of Mann-Kendall's test for testing of the presence of the monotonic increasing or decreasing trend are presented. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$.***

| group of the stations | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 737 | 101,1 | 96,5 | 93,6 | 96,5 | 107,5 | 96,4 | 99,4 | 105,0 | 97,6 | 94,5 | -0,03 | no |
| rural background | 139 | 61,0 | 58,9 | 57,4 | 60,5 | 63,6 | 59,4 | 59,0 | 67,6 | 56,9 | 56,6 | -0,30 | no |
| suburban background | 160 | 92,4 | 90,5 | 86,0 | 89,3 | 100,6 | 89,3 | 91,0 | 97,7 | 88,2 | 86,0 | -0,34 | no |
| urban background | 178 | 102,1 | 94,9 | 92,7 | 96,9 | 110,3 | 95,5 | 99,5 | 102,5 | 96,4 | 93,0 | -0,11 | no |
| traffic (all area types) | 197 | 138,1 | 132,2 | 127,2 | 129,6 | 144,6 | 132,0 | 137,1 | 143,2 | 138,3 | 132,3 | 0,57 | no |
| industrial (all area types) | 62 | 92,6 | 87,5 | 90,5 | 89,1 | 97,4 | 86,7 | 90,9 | 92,7 | 87,9 | 85,7 | -0,29 | no |
| Northern Europe | 14 | 88,6 | 81,4 | 83,6 | 88,6 | 94,8 | 90,4 | 90,1 | 97,7 | 92,2 | 87,3 | 0,73 | no |
| North-western Europe | 207 | 110,2 | 107,5 | 105,2 | 105,6 | 118,5 | 105,4 | 105,2 | 108,9 | 108,5 | 106,9 | -0,09 | no |
| Central and Eastern Eur. | 410 | 88,7 | 84,0 | 80,6 | 86,2 | 98,6 | 85,5 | 90,4 | 97,8 | 84,8 | 81,7 | -0,26 | no |
| Southern Europe | 106 | 132,6 | 125,5 | 123,0 | 119,2 | 122,1 | 121,7 | 124,0 | 126,1 | 127,1 | 120,9 | -0,29 | no |
| rural background - N | 3 | 57,1 | 45,5 | 44,9 | 48,0 | 59,8 | 51,5 | 45,8 | 63,6 | 43,7 | 42,4 | -0,57 | no |
| rural background - NW | 30 | 76,4 | 74,1 | 76,1 | 75,1 | 84,6 | 77,6 | 74,5 | 78,2 | 75,4 | 74,2 | -0,11 | no |
| rural background - CE | 92 | 55,2 | 53,3 | 51,8 | 56,4 | 57,0 | 53,3 | 54,5 | 65,2 | 50,8 | 50,9 | -0,17 | no |
| rural background - S | 14 | 67,6 | 66,8 | 56,5 | 58,7 | 63,5 | 61,9 | 58,8 | 61,5 | 60,2 | 59,5 | -0,81 | no |
| suburban backgr. - N | 0 | no station with complete data | | | | | | | | | | | |
| suburban backgr. - NW | 63 | 103,9 | 101,2 | 96,3 | 97,8 | 108,7 | 95,8 | 94,1 | 99,8 | 97,8 | 95,0 | -0,76 | no |
| suburban backgr. - CE | 85 | 80,3 | 78,9 | 74,1 | 80,0 | 92,5 | 80,7 | 85,5 | 94,2 | 77,8 | 76,1 | 0,29 | no |
| suburban backgr. - S | 12 | 117,6 | 117,0 | 116,4 | 110,5 | 114,6 | 116,4 | 113,8 | 111,8 | 110,6 | 108,6 | -0,89 | ** |
| urban background - N | 5 | 88,0 | 82,4 | 89,7 | 87,7 | 98,8 | 90,6 | 90,4 | 96,2 | 88,3 | 82,1 | 0,13 | no |
| urban background - NW | 52 | 111,2 | 106,9 | 104,2 | 104,9 | 118,3 | 103,6 | 103,3 | 104,8 | 107,3 | 108,3 | -0,20 | no |
| urban background - CE | 102 | 94,0 | 87,3 | 84,4 | 91,2 | 105,9 | 88,4 | 94,8 | 99,2 | 86,7 | 82,2 | -0,55 | no |
| urban background - S | 19 | 124,5 | 106,4 | 107,1 | 108,3 | 115,0 | 113,4 | 116,9 | 115,5 | 120,3 | 112,2 | 0,98 | no |
| industrial - N | 1 | 66,0 | 53,0 | 71,0 | 45,0 | 62,0 | 65,0 | 78,0 | 77,0 | 99,0 | 85,0 | 4,00 | * |
| industrial - NW | 22 | 94,7 | 94,6 | 100,0 | 97,6 | 109,4 | 95,0 | 95,2 | 98,4 | 98,2 | 96,5 | 0,13 | no |
| industrial - CE | 25 | 79,6 | 77,3 | 74,3 | 77,3 | 91,4 | 76,6 | 81,3 | 88,0 | 73,8 | 73,4 | -0,41 | no |
| industrial - S | 14 | 114,4 | 97,3 | 105,7 | 99,8 | 91,7 | 93,3 | 102,2 | 93,5 | 95,9 | 90,7 | -1,57 | + |
| traffic - N | 5 | 112,6 | 107,7 | 103,4 | 122,7 | 118,3 | 118,7 | 118,7 | 123,8 | 123,8 | 119,9 | 1,40 | * |
| traffic - NW | 39 | 153,8 | 151,5 | 146,4 | 147,3 | 165,7 | 150,1 | 154,4 | 158,1 | 158,4 | 154,8 | 0,93 | no |
| traffic - CE | 106 | 121,7 | 113,3 | 108,5 | 114,5 | 134,2 | 116,7 | 123,5 | 129,9 | 120,5 | 114,6 | 0,84 | no |
| traffic - S | 47 | 164,5 | 161,3 | 156,1 | 149,6 | 153,5 | 152,7 | 155,5 | 163,1 | 163,2 | 154,9 | -0,16 | no |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1(meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

From Table 4 it can be seen that for the 19$^{th}$ maximum hourly mean no trend is detected in most cases, contrary to the annual mean. The variability in hourly concentrations is much higher than the variability in annually mean concentrations, due to the meteorological variability.Only for several combinations of station types and regions a trend is detected, namely the decreasing trend for the suburban background (and also industrial) stations of the Southern region, and an increasing trend for the traffic stations of the Northern region.

Figure 10 presents the calculated average $NO_2$ annual means and $19^{th}$ maximum hourly means from 1999 to 2008 for the different combinations of stations/area types and regions.

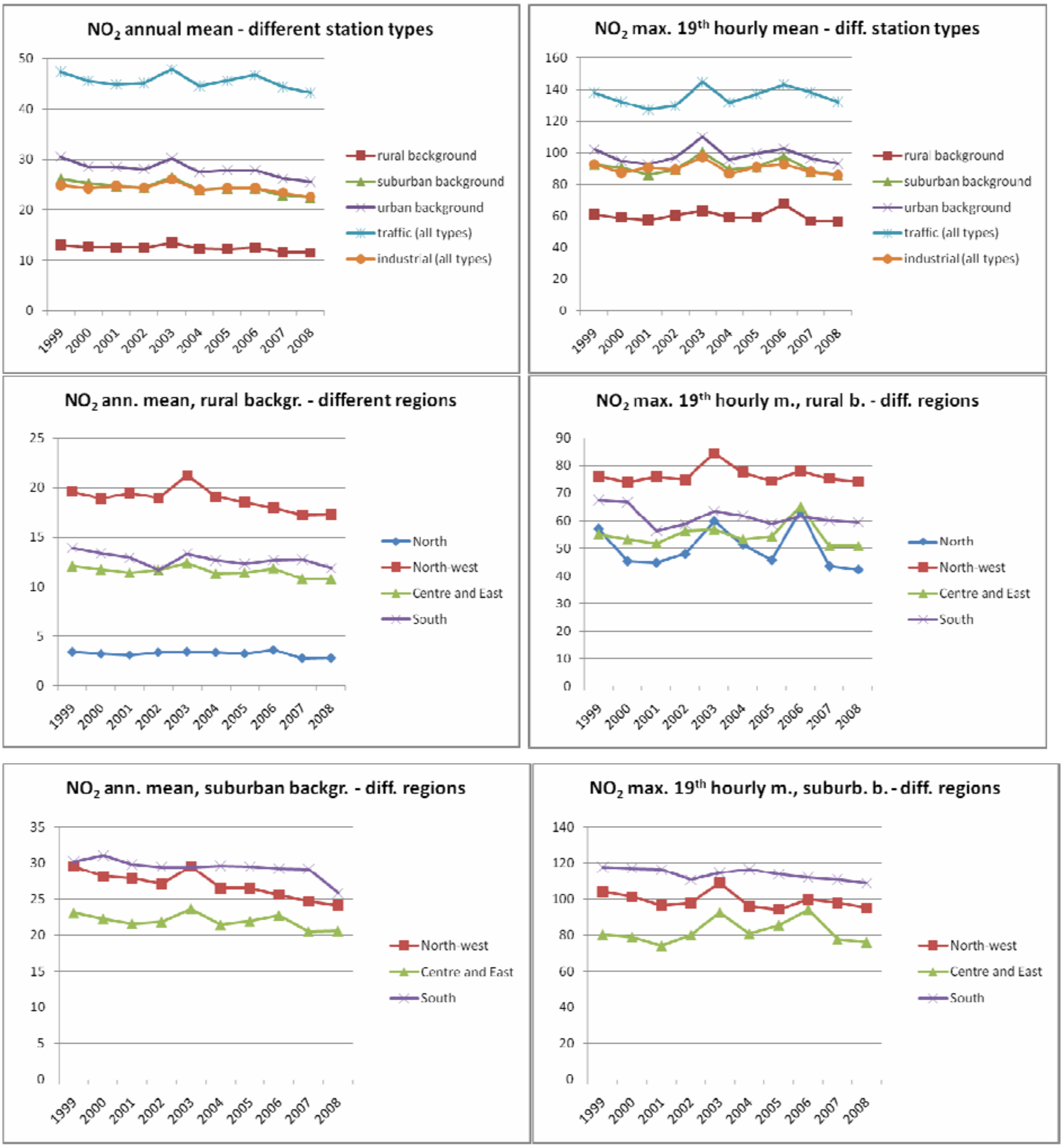



**Figure 10:** ***The average $NO_2$ annual means (left) and $19^{th}$ maximum hourly means (right) in µg.m$^{-3}$, for the years 1999 – 2008, for different combinations of station/area types and regions.***

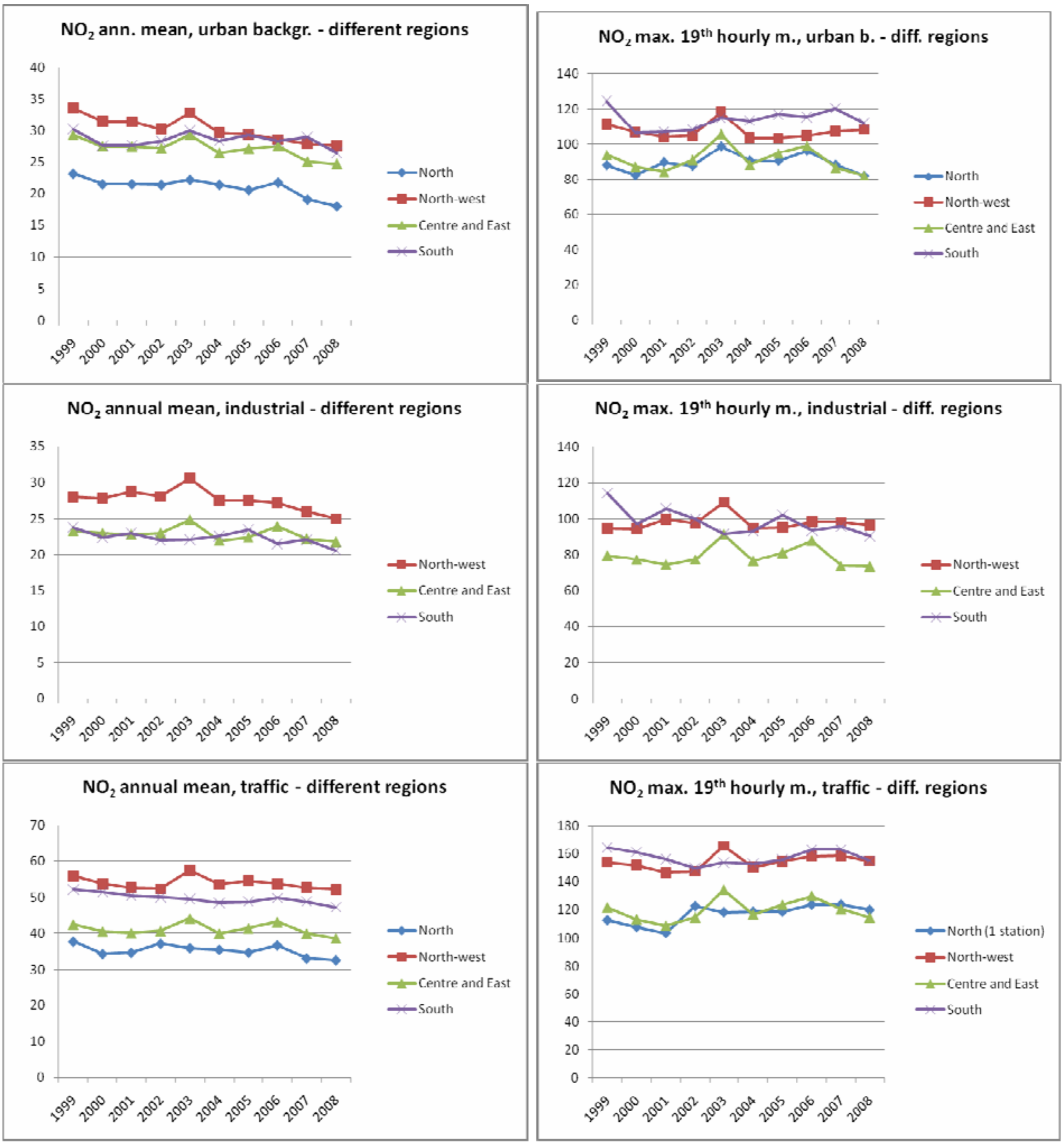



**Figure 10 cont.:** ***The average $NO_2$ annual means (left) and 19$^{th}$ maximum hourly means (right) in µg.m$^{-3}$, for the years 1999 – 2008, for different combinations of station/area types and regions.***

## 2.1.2 Trend at the individual stations

In addition to the estimation of the average trends for different types of stations and regions, the trends for individual stations were also calculated in order to investigate the variability of the trends within the regions or/and types of stations. Figure 9 shows in yellow and pink the stations used in this analysis. In Table 5 the distribution of the Sen's slope across the different groups of the stations is presented for $NO_2$ annual mean, separately for the significant and non-significant trends (based on Mann-Kendall's test).

**Table 5:** ***Fractional distribution of the Sen's slope at the individual stations for $NO_2$ annual mean, for the years 1999-2008, for all stations together and separately for different station/area types and regions. The number of the stations in relevant group, the percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. The Sen's slope represents the annual change of concentration, in $\mu g.m^{-3}$.***

| group of the stations | N | < -1.5 | | -1.5 - -1.0 | | -1.0 - -0.5 | | -0.5 - 0.0 | | 0.0 - 0.5 | | 0.5 - 1.0 | | 1.0 - 1.5 | | > 1.5 | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. |
| all stations | 1280 | 2,8 | 0,7 | 4,7 | 1,6 | 13,0 | 6,6 | 10,9 | 31,3 | 1,2 | 19,9 | 1,4 | 2,6 | 0,5 | 1,2 | 1,1 | 0,5 |
| rural background | 226 | 0,0 | 0,0 | 0,4 | 0,0 | 4,0 | 1,8 | 24,7 | 41,9 | 2,6 | 23,3 | 0,9 | 0,4 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburban background | 235 | 2,6 | 0,4 | 5,5 | 0,0 | 17,0 | 4,7 | 13,2 | 32,8 | 0,9 | 21,3 | 0,0 | 0,9 | 0,0 | 0,4 | 0,0 | 0,4 |
| urban background | 353 | 2,5 | 0,6 | 5,1 | 1,1 | 19,0 | 7,6 | 7,9 | 34,6 | 0,3 | 16,1 | 0,8 | 1,7 | 0,3 | 1,1 | 0,6 | 0,6 |
| traffic (all types) | 320 | 5,9 | 1,9 | 5,6 | 4,1 | 11,3 | 10,0 | 2,2 | 20,0 | 0,6 | 21,6 | 3,1 | 5,6 | 1,6 | 2,8 | 3,1 | 0,6 |
| industrial (all types) | 144 | 1,4 | 0,0 | 6,9 | 2,1 | 10,4 | 6,9 | 12,5 | 29,2 | 2,8 | 18,1 | 2,1 | 4,2 | 0,0 | 0,7 | 1,4 | 1,4 |
| Northern Europe | 48 | 0,0 | 0,0 | 0,0 | 0,0 | 14,6 | 0,0 | 18,8 | 39,6 | 2,1 | 20,8 | 2,1 | 2,1 | 0,0 | 0,0 | 0,0 | 0,0 |
| North-western E. | 380 | 3,9 | 0,5 | 7,4 | 0,5 | 20,5 | 6,8 | 13,9 | 29,2 | 0,3 | 13,4 | 1,1 | 1,1 | 0,5 | 0,5 | 0,3 | 0,0 |
| Central + Eastern E. | 536 | 0,9 | 0,0 | 2,2 | 0,6 | 11,2 | 4,1 | 13,2 | 34,5 | 2,1 | 23,9 | 1,7 | 3,2 | 0,6 | 0,7 | 1,1 | 0,0 |
| Southern Europe | 316 | 5,1 | 2,2 | 6,3 | 4,7 | 7,0 | 11,4 | 2,2 | 27,2 | 0,6 | 20,9 | 1,3 | 3,5 | 0,3 | 2,8 | 2,2 | 2,2 |
| rural backgr. - N | 20 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 20,0 | 50,0 | 0,0 | 30,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| rural backgr. - NW | 54 | 0,0 | 0,0 | 0,0 | 0,0 | 11,1 | 1,9 | 29,6 | 40,7 | 0,0 | 14,8 | 1,9 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| rural backgr. - CE | 121 | 0,0 | 0,0 | 0,0 | 0,0 | 1,7 | 0,8 | 27,3 | 41,3 | 4,1 | 24,0 | 0,0 | 0,8 | 0,0 | 0,0 | 0,0 | 0,0 |
| rural backgr. - S | 32 | 0,0 | 0,0 | 3,1 | 0,0 | 3,1 | 6,3 | 9,4 | 40,6 | 3,1 | 31,3 | 3,1 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - N | 1 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 100 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - NW | 96 | 5,2 | 0,0 | 8,3 | 0,0 | 29,2 | 6,3 | 13,5 | 22,9 | 1,0 | 13,5 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - CE | 100 | 0,0 | 0,0 | 2,0 | 0,0 | 6,0 | 2,0 | 17,0 | 44,0 | 1,0 | 28,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - S | 38 | 2,6 | 2,6 | 7,9 | 0,0 | 15,8 | 7,9 | 0,0 | 28,9 | 0,0 | 23,7 | 0,0 | 5,3 | 0,0 | 2,6 | 0,0 | 2,6 |
| urban backgr. - N | 13 | 0,0 | 0,0 | 0,0 | 0,0 | 30,8 | 0,0 | 15,4 | 38,5 | 7,7 | 7,7 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| urban backgr. - NW | 127 | 6,3 | 0,8 | 6,3 | 0,0 | 22,8 | 10,2 | 11,0 | 33,1 | 0,0 | 9,4 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| urban backgr. - CE | 141 | 0,0 | 0,0 | 4,3 | 0,7 | 19,9 | 5,0 | 6,4 | 37,6 | 0,0 | 19,9 | 2,1 | 2,8 | 0,0 | 0,7 | 0,7 | 0,0 |
| urban backgr. - S | 72 | 1,4 | 1,4 | 5,6 | 4,2 | 8,3 | 9,7 | 4,2 | 30,6 | 0,0 | 22,2 | 0,0 | 2,8 | 1,4 | 4,2 | 1,4 | 2,8 |
| industrial - N | 3 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 33,3 | 0,0 | 0,0 | 0,0 | 33,3 | 33,3 | 0,0 | 0,0 | 0,0 | 0,0 |
| industrial - NW | 41 | 2,4 | 0,0 | 12,2 | 0,0 | 17,1 | 4,9 | 19,5 | 29,3 | 0,0 | 12,2 | 2,4 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| industrial - CE | 34 | 0,0 | 0,0 | 0,0 | 0,0 | 8,8 | 0,0 | 23,5 | 26,5 | 8,8 | 26,5 | 0,0 | 5,9 | 0,0 | 0,0 | 0,0 | 0,0 |
| industrial - S | 66 | 1,5 | 0,0 | 7,6 | 4,5 | 7,6 | 12,1 | 1,5 | 31,8 | 1,5 | 18,2 | 1,5 | 4,5 | 0,0 | 1,5 | 3,0 | 3,0 |
| traffic - N | 11 | 0,0 | 0,0 | 0,0 | 0,0 | 27,3 | 0,0 | 9,1 | 36,4 | 0,0 | 27,3 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| traffic - NW | 61 | 1,6 | 1,6 | 11,5 | 3,3 | 13,1 | 6,6 | 3,3 | 19,7 | 0,0 | 21,3 | 3,3 | 6,6 | 3,3 | 3,3 | 1,6 | 0,0 |
| traffic - CE | 140 | 3,6 | 0,0 | 2,9 | 1,4 | 15,0 | 8,6 | 2,9 | 20,7 | 1,4 | 24,3 | 4,3 | 7,1 | 2,1 | 2,1 | 3,6 | 0,0 |
| traffic - S | 108 | 12,0 | 4,6 | 6,5 | 8,3 | 3,7 | 14,8 | 0,0 | 17,6 | 0,0 | 17,6 | 1,9 | 3,7 | 0,0 | 3,7 | 3,7 | 1,9 |

The same distributions are presented also as histograms in Figure 11 and Figure 12 for all stations together and separately for different station/area types and regions.

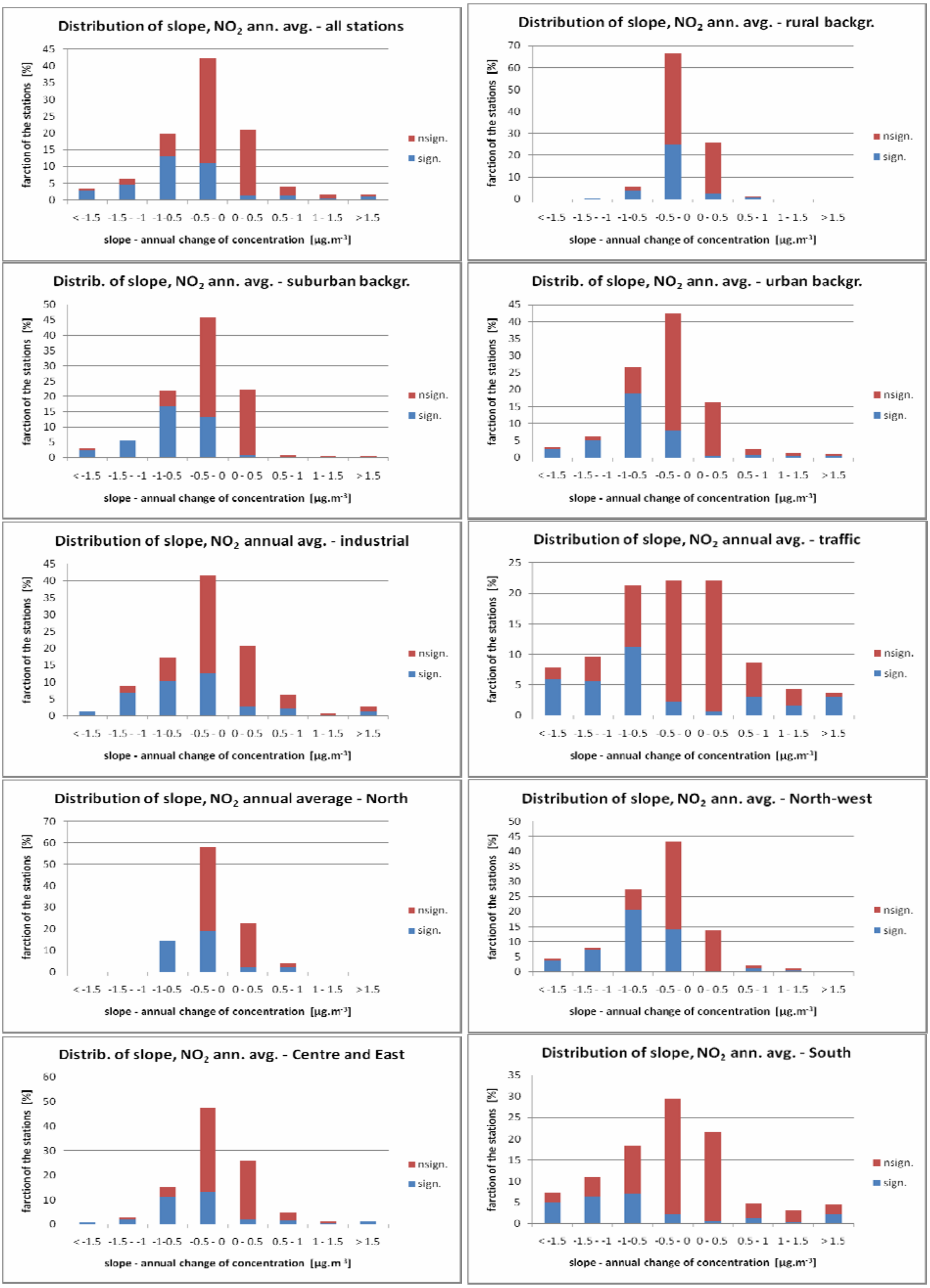



**Figure 11:** ***Distribution of the Sen's slope at the individual stations for $NO_2$ annual mean, for the years 1999-2008. The percentage of the stations of the relevant group in different slope intervals of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. Significant trend is in blue, non-significant trend is in red. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$***

In Figure 12, similar histograms for different combinations of stations/area types and regions are shown.

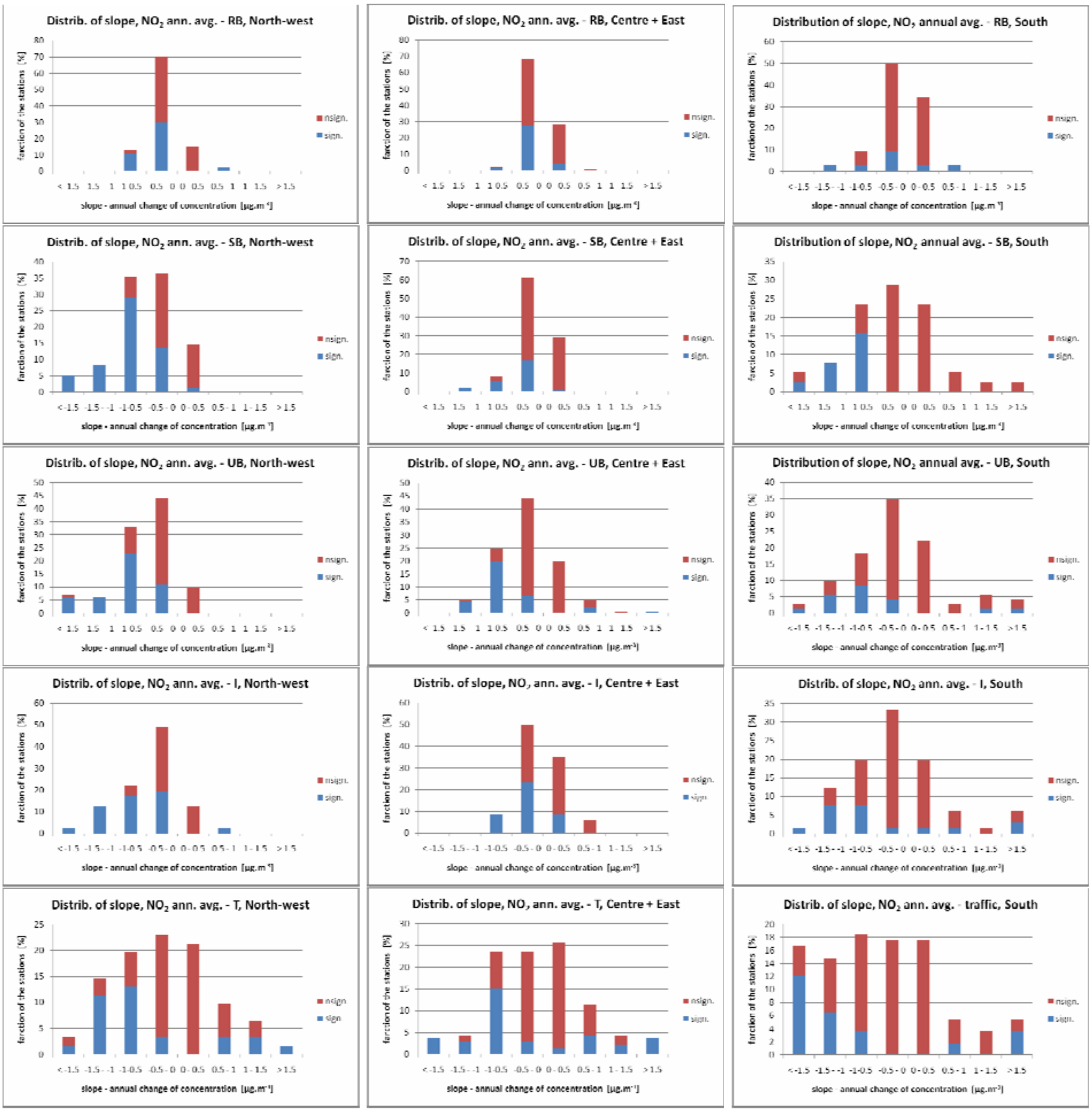



**Figure 12:** ***Distribution of the Sen's slope at the individual stations for $NO_2$ annual mean, for the years 1999-2008, separately for different station/area types and regions. (North region is not shown, due to small number of stations.) The percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. Significant trend is in blue, non-significant trend is in red.***

This analysis shows that traffic stations across Europe have had both increasing and decreasing trends in the past 10 years of the $NO_2$ annual mean. In 25% of the traffic stations the trends were significant and decreasing, especially on the North-west region (30%) and the North region (36%). On the other hand, 8% of the European traffic stations registered a significant increasing trend, especially in the Central east region (11%), but also in the North-west region (8%) and in the South (6%). This is also well illustrated by

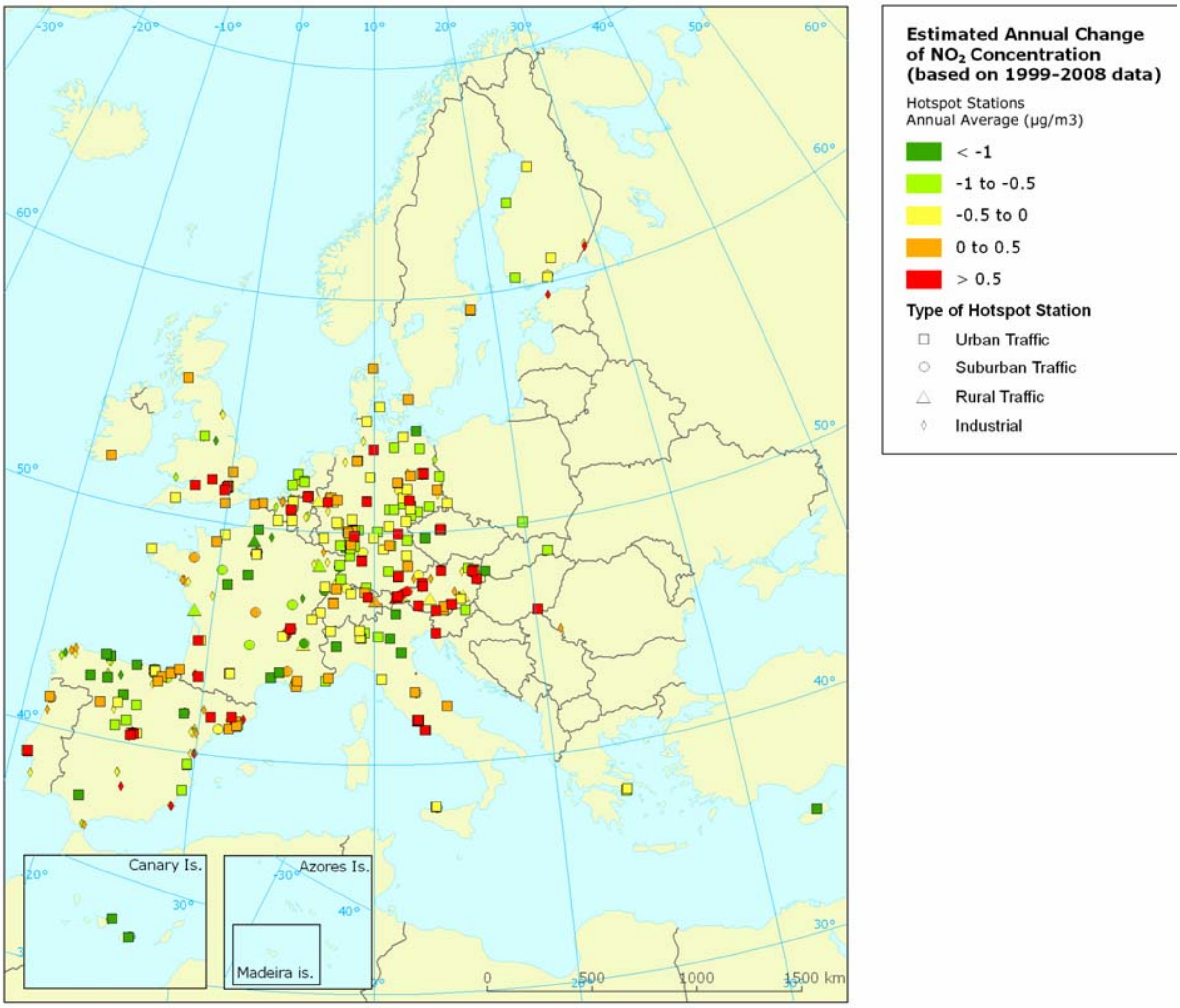

Estimated Annual Change of $NO_2$ Concentration (based on 1999-2008 data)
Hotspot Stations
Annual Average (µg/m3)
< -1
-1 to -0.5
-0.5 to 0
0 to 0.5
> 0.5
Type of Hotspot Station
Urban Traffic
Suburban Traffic
Rural Traffic
Industrial
Canary Is.
Azores Is.
Madeira is.
0 500 1000 1500 km

Figure 13.

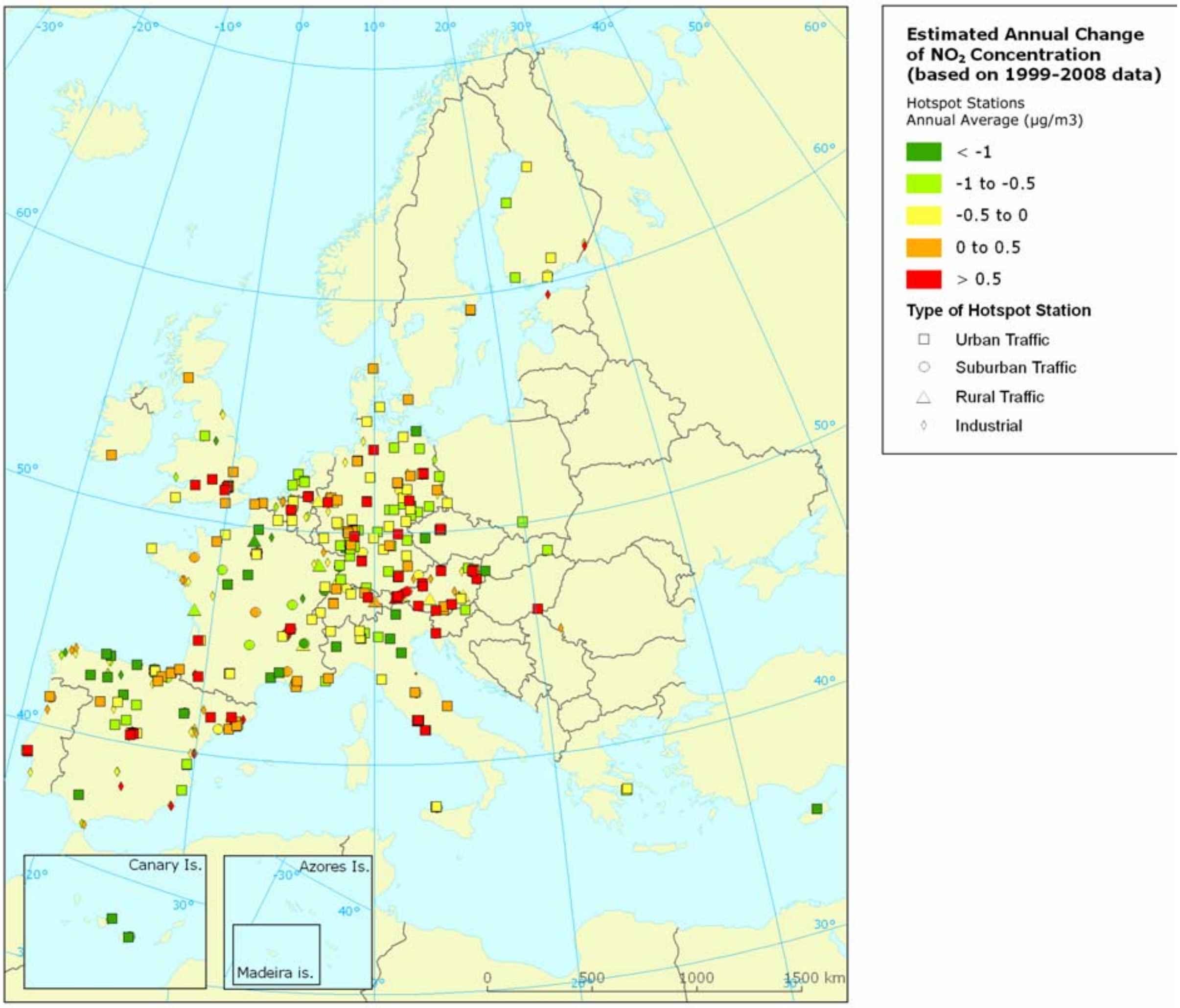



**Figure 13: Average annual change in $NO_2$ annual mean concentrations (in $\mu g.m^{-3}$) at traffic and industrial stations between 1999 and 2008.**

For industrial stations, significant decreasing trends were registered in the North-western region (51%), the Central and Eastern region (32%) and the Southern region (18%). On the other hand, some significant increasing trends were also registered in 9% of the industrial stations in Central & Eastern region, 6% in the Southern region and 2% in the North-western region.

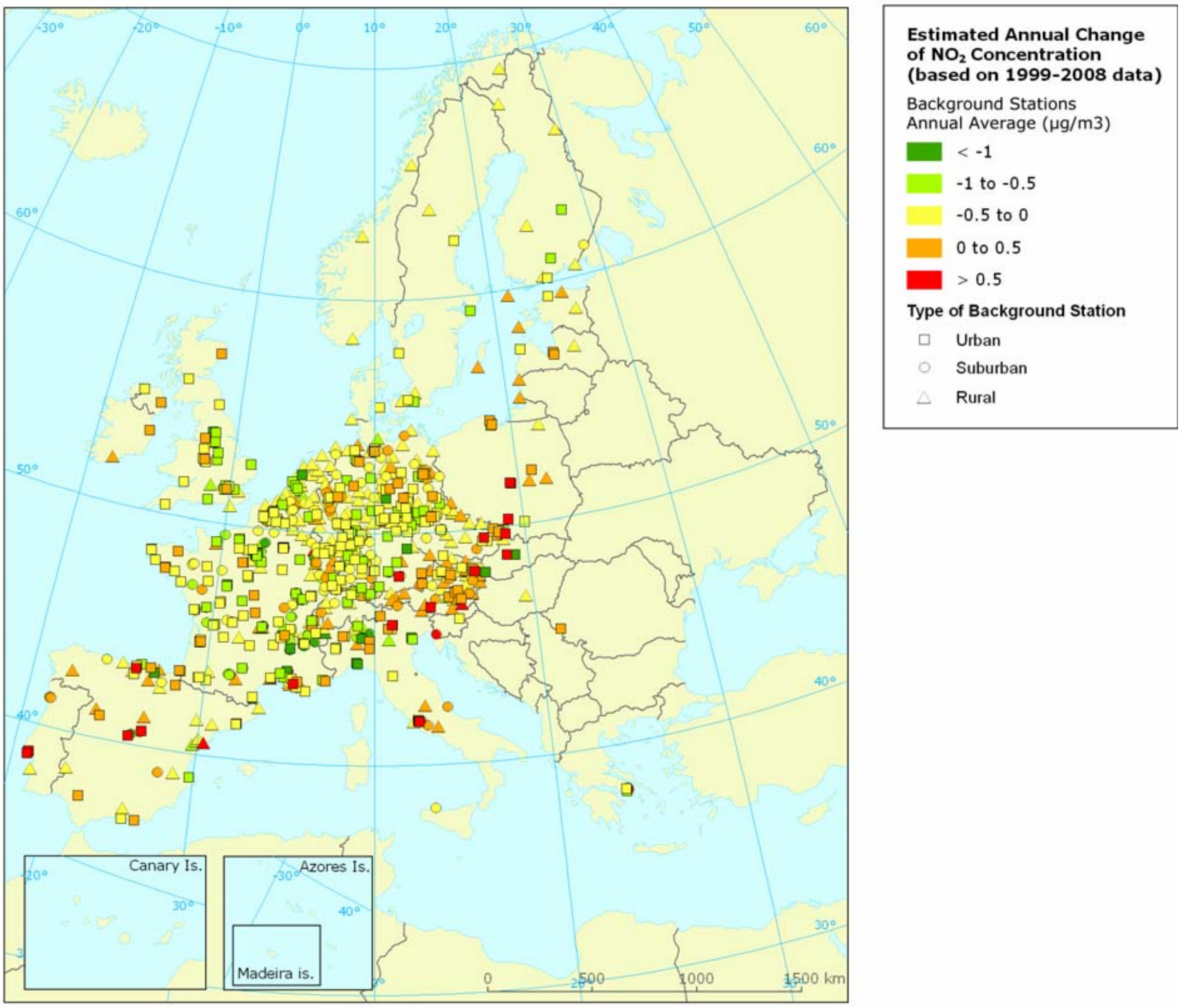



**Figure 14: Average annual change in $NO_2$ annual mean concentrations (in µg.m$^{-3}$) at background stations between 1999 and 2008.**

Figure 14 shows the average annual change in $NO_2$ annual mean concentrations at background stations between 1999 and 2008. For the majority of the background stations, the trends are decreasing, 38% of the suburban- and 35% of the urban background stations had significant decreasing trends. The North-western region had most background stations with significant decreasing trends, with 56% in sub-urban areas and 47% in urban areas. On the other hand, significant increasing trends were registered at 3% of the urban background stations in the Central and Eastern and Southern regions. At rural background stations significant increasing trends were registered at 6% of the southern stations, 4% of the Central and Eastern region stations and 2% of the North-western region stations.

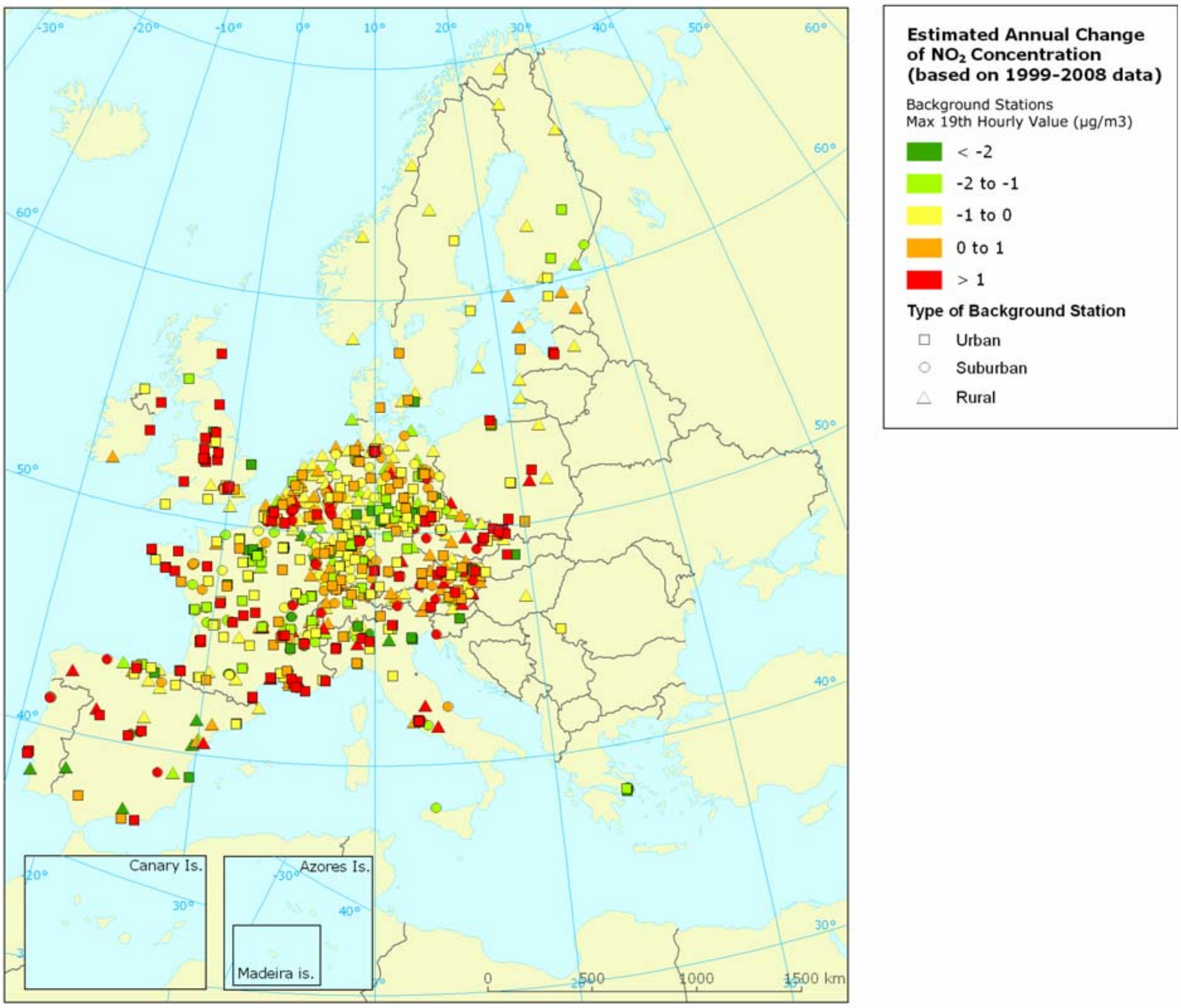



**Figure 15: Average annual change in the 19th highest hourly $NO_2$ concentrations (in µg.m$^{-3}$) at background stations between 1999 and 2008.**

Figure 15 and Figure 16 show the average annual change in the 19th highest hourly $NO_2$ concentrations between 1999 and 2008 at background and hot spot stations, respectively. Both figures show an important number of stations with an average increase of the 19th highest hourly $NO_2$ concentrations, distributed throughout Europe. The variability of the 19th highest hourly $NO_2$ concentrations in time is high and most of the calculated trends are not significant.

For background stations a significant increasing trend was detected at 13% of the stations and a significant decreasing trend at 10% of the stations (For more detailed information, see Annex B). 77% of the background stations did not have significant trends. For the traffic stations, a significant increasing trend was detected at 16% of the stations, while 9% had a significant decreasing trend between 1999 and 2008. It is never the less difficult to interpret these trends, since the meteorological conditions play a major role for the highest hourly concentrations and are likely to influence the trends more than year to year emission changes.

This analysis shows that there has been no general improvement of the 19th highest hourly $NO_2$ concentrations, despite improvements in the annual mean concentrations.

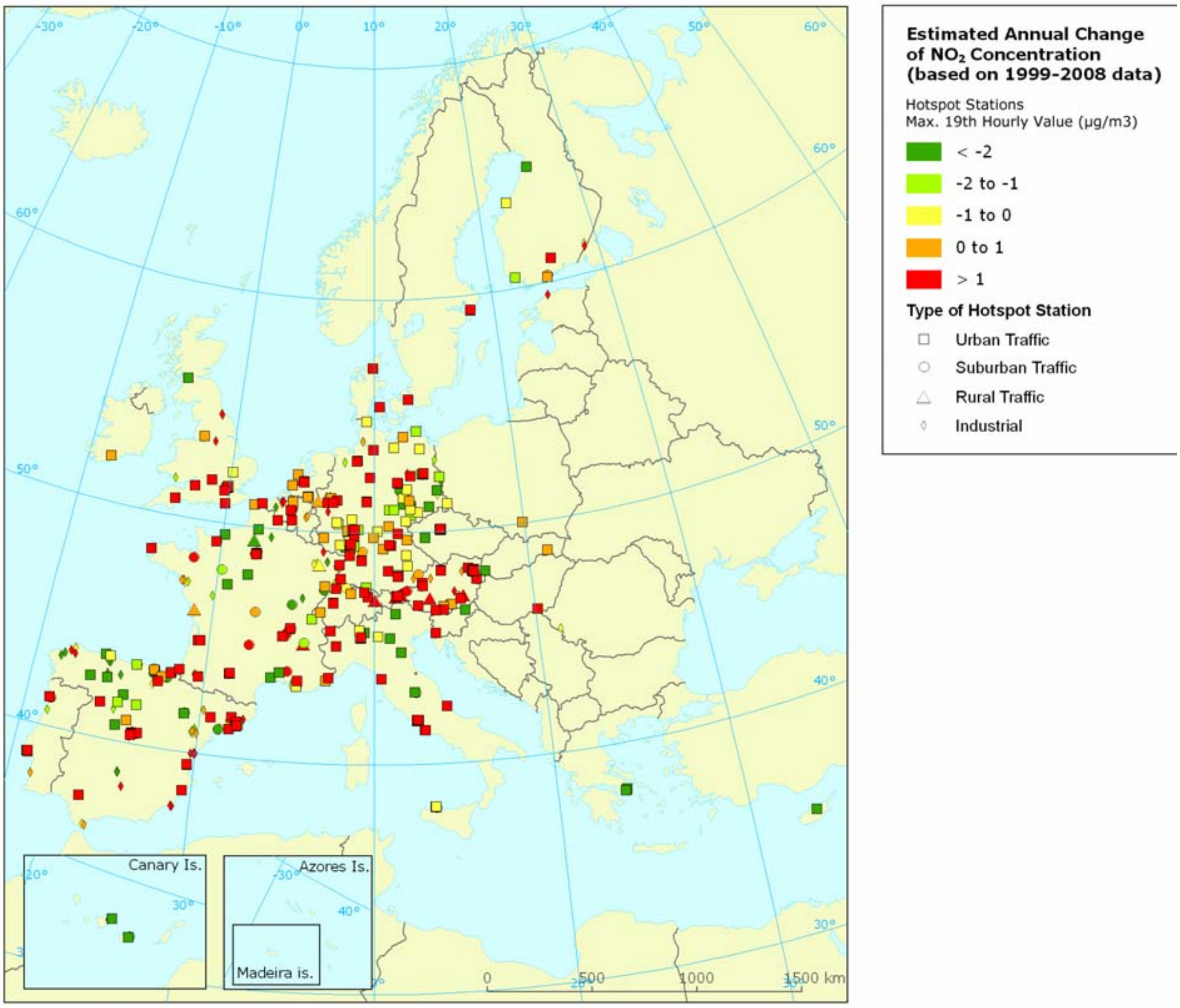



**Figure 16: Average annual change in the 19th highest hourly $NO_2$ concentrations (in $\mu g.m^{-3}$) at traffic and industrial stations between 1999 and 2008.**

## 2.2. Trend of $NO_X$ in comparison with $NO_2$

### 2.2.1 Trend at aggregated station level

A similar trend analysis to the one for $NO_2$ concentrations has been performed for the $NO_x$ annual mean concentrations. Figure 17 shows in yellow the stations used for the $NO_x$ annual average trend analysis, as well as the European regions used in the analysis.

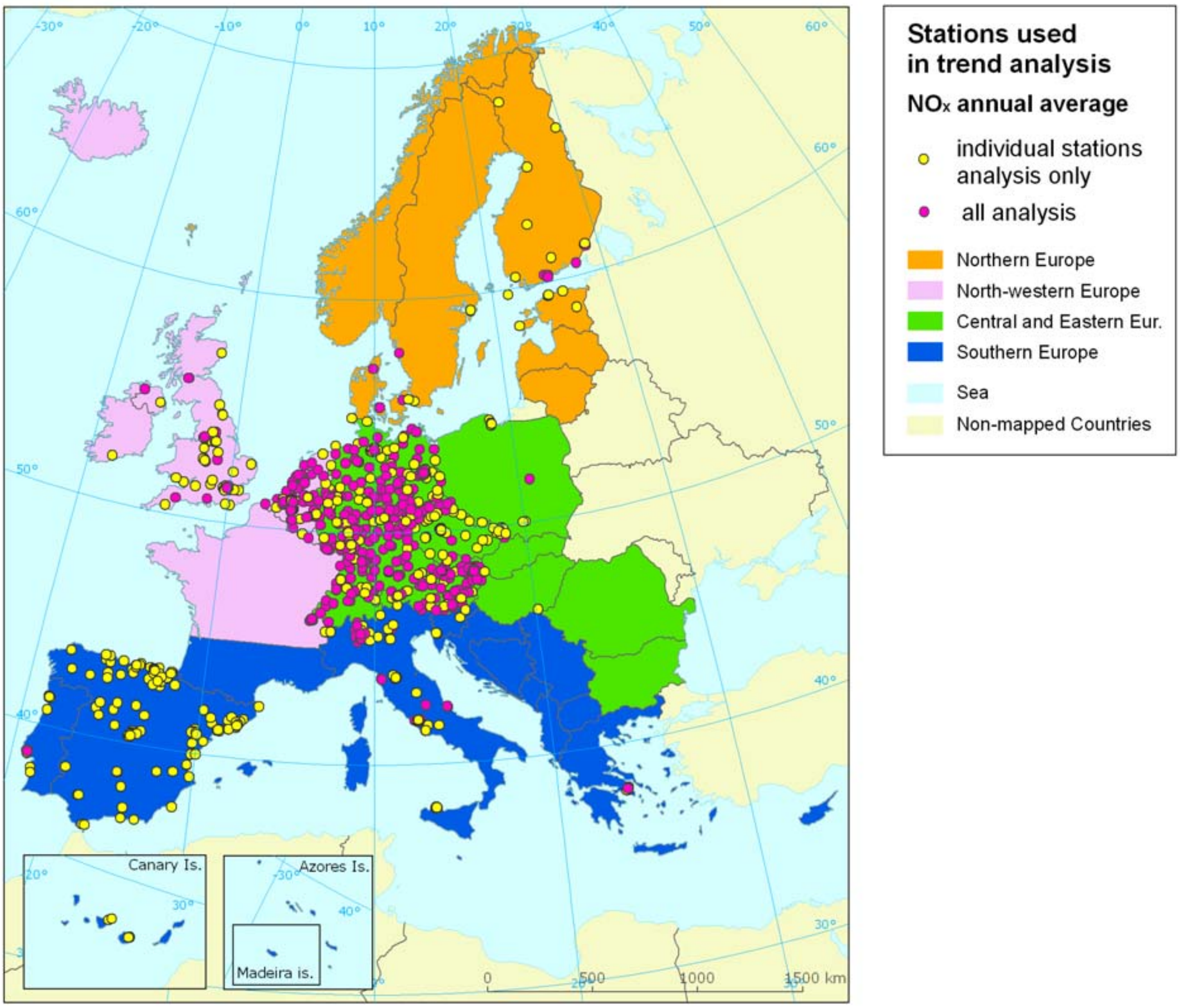



**Figure 17: Stations used in trend analysis of the $NO_x$ annual mean, both for the average of stations (stations in pink) and for the individual stations (all stations, both pink and yellow), and the four European regions used in the analyses.**

Table 6 shows the results of the trend analysis for the $NO_x$ annual mean concentrations measured between 1999 and 2008.

**Table 6:** ***Trend analysis for $NO_x$ annual mean, for the years 1999-2008, for all stations and separately for different groups of stations by area/type and region. The number of stations in relevant group, the average concentration values of the group (in µg.m$^{-3}$) for individual years, the estimated Sen's slope and the result of Mann-Kendall's test for testing of the presence of the monotonic increasing or decreasing trend are presented. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$.***

| group of the stations | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 473 | 63,7 | 60,8 | 59,1 | 60,0 | 61,1 | 57,4 | 56,0 | 56,6 | 52,5 | 50,1 | -1,29 | ** |
| rural background | 109 | 19,1 | 18,8 | 18,5 | 19,3 | 20,0 | 18,9 | 17,4 | 18,4 | 16,8 | 16,4 | -0,28 | * |
| suburban background | 81 | 42,2 | 41,0 | 39,7 | 40,0 | 42,2 | 39,0 | 38,2 | 39,8 | 36,7 | 36,0 | -0,64 | ** |
| urban background | 112 | 55,7 | 53,1 | 52,8 | 52,1 | 53,9 | 48,8 | 47,9 | 49,0 | 45,2 | 43,1 | -1,30 | ** |
| traffic (all area types) | 137 | 123,0 | 116,4 | 111,1 | 114,3 | 113,8 | 109,3 | 107,1 | 106,2 | 98,6 | 93,2 | -2,55 | *** |
| industrial (all area types) | 34 | 44,5 | 44,4 | 47,1 | 45,0 | 48,8 | 44,1 | 43,0 | 44,4 | 43,1 | 41,4 | -0,34 | * |
| Northern Europe | 13 | 57,9 | 52,0 | 49,8 | 52,0 | 51,0 | 49,4 | 47,2 | 46,6 | 44,0 | 42,0 | -1,61 | *** |
| North-western Europe | 70 | 75,8 | 72,2 | 75,0 | 68,6 | 74,4 | 68,1 | 64,1 | 61,9 | 62,3 | 60,5 | -1,72 | ** |
| Central and Eastern Eur. | 361 | 58,9 | 57,6 | 55,4 | 54,7 | 56,4 | 51,7 | 50,8 | 52,5 | 47,6 | 45,9 | -1,40 | ** |
| Southern Europe | 29 | 96,2 | 76,9 | 71,0 | 108,0 | 91,0 | 105,2 | 105,9 | 99,4 | 93,9 | 81,7 | 0,45 | no |
| rural background - N | 3 | 10,2 | 9,2 | 9,0 | 9,6 | 9,7 | 9,6 | 8,7 | 10,5 | 7,8 | 7,8 | -0,17 | no |
| rural background - NW | 21 | 26,8 | 25,7 | 29,5 | 27,9 | 30,5 | 28,3 | 24,9 | 25,0 | 24,1 | 24,0 | -0,37 | + |
| rural background - CE | 82 | 17,2 | 17,4 | 16,1 | 17,0 | 17,5 | 16,2 | 15,4 | 16,6 | 14,8 | 14,4 | -0,31 | * |
| rural background - S | 3 | 24,6 | 17,0 | 18,4 | 32,1 | 24,3 | 33,9 | 30,2 | 29,2 | 28,0 | 27,3 | 0,92 | no |
| suburban backgr. - N | 0 | no station with complete data | | | | | | | | | | | |
| suburban backgr. - NW | 8 | 52,4 | 48,0 | 55,0 | 48,2 | 55,0 | 49,2 | 46,9 | 45,2 | 48,6 | 44,2 | -0,75 | no |
| suburban backgr. - CE | 70 | 40,4 | 40,2 | 38,2 | 38,2 | 40,6 | 36,9 | 36,2 | 38,2 | 34,4 | 34,5 | -0,67 | ** |
| suburban backgr. - S | 3 | 58,5 | 39,0 | 33,6 | 59,1 | 45,9 | 61,3 | 61,5 | 63,3 | 59,7 | 48,2 | 0,80 | no |
| urban background - N | 4 | 35,7 | 32,3 | 32,9 | 30,6 | 34,3 | 33,8 | 32,1 | 34,5 | 29,7 | 27,2 | -0,54 | no |
| urban background - NW | 14 | 72,1 | 65,2 | 70,1 | 63,7 | 68,6 | 62,4 | 60,0 | 56,8 | 57,8 | 55,3 | -1,76 | ** |
| urban background - CE | 89 | 54,7 | 52,4 | 51,6 | 50,6 | 52,7 | 47,0 | 46,2 | 47,8 | 43,0 | 41,5 | -1,42 | ** |
| urban background - S | 5 | 43,5 | 48,0 | 42,5 | 61,9 | 49,0 | 54,5 | 56,9 | 59,9 | 61,6 | 48,9 | 1,99 | no |
| industrial - N | 1 | 11,5 | 11,8 | 12,2 | 10,1 | 13,0 | 15,5 | 16,5 | 19,1 | 32,0 | 29,3 | 1,98 | ** |
| industrial - NW | 10 | 57,6 | 57,0 | 64,0 | 57,2 | 64,7 | 57,5 | 55,7 | 54,1 | 54,8 | 52,9 | -0,53 | * |
| industrial - CE | 23 | 40,3 | 40,3 | 41,2 | 41,2 | 43,4 | 39,5 | 38,6 | 41,2 | 38,4 | 37,0 | -0,35 | no |
| industrial - S | 0 | no station with complete data | | | | | | | | | | | |
| traffic - N | 5 | 113,5 | 101,6 | 95,3 | 102,9 | 96,8 | 92,6 | 88,6 | 83,5 | 79,5 | 76,7 | -4,09 | *** |
| traffic - NW | 17 | 161,1 | 155,8 | 151,1 | 139,3 | 148,3 | 137,3 | 128,9 | 124,2 | 124,1 | 121,9 | -4,62 | *** |
| traffic - CE | 97 | 115,7 | 113,1 | 108,1 | 105,5 | 107,3 | 99,8 | 98,3 | 100,2 | 91,2 | 86,7 | -2,96 | *** |
| traffic - S | 18 | 129,1 | 101,2 | 93,9 | 141,6 | 121,3 | 138,5 | 139,5 | 128,1 | 119,5 | 105,5 | -0,43 | no |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1(meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

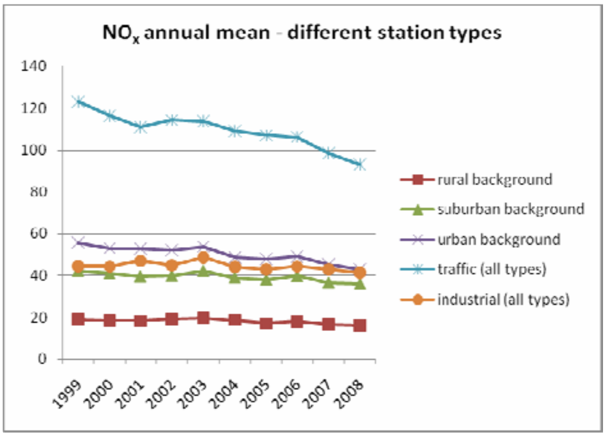
**In general, it can be seen that the annual mean concentrations of $NO_x$ have had a stronger decreasing trend than $NO_2$ (**

Table 3) between 1999 and 2008. The average decrease of $NO_x$ annual mean concentrations for all stations was of -1.29 µg.m$^{-3}$ per annum, while for $NO_2$ it was only -0,27 µg.m$^{-3}$ per annum. Note that in the $NO_2$ annual mean trend analysis 755 stations were included, while for the $NO_x$ annual mean trend analysis only 473 stations had available data. This impacts on the calculated trends, especially for the Southern European region, where few stations with $NO_x$ data for the studied period were available.

The strongest $NO_x$ trend is detected at the traffic stations (slope of -2.5 µg.m$^{-3}$), especially in the North-western region (slope of -4.6 µg.m$^{-3}$) and with the exception of southern Europe, where no trend was detected. On the other hand, traffic stations did not register in average a significant decreasing trend for $NO_2$ concentrations, with the exception of the southern region.

On a regional basis, negative trends of $NO_x$ are detected for North-Western (slope of -1.7 µg.m$^{-3}$), Central and Eastern Europe (slope of -1.4 µg.m$^{-3}$). For Northern Europe, the slope of the trend is -1.6 µg.m$^{-3}$, but it is calculated based on only 13 stations. The Southern region did not register a significant trend.
The average annual mean $NO_2$ trends were only significant for North-Western and Southern Europe and considerably less pronounced, with slopes of -0.4 and of -0.3 µg.m$^{-3}$, respectively.

In Figure 18, the graphs of the 1999 - 2008 $NO_x$ trends are presented for the different groups of type of stations and four European regions. In Figure 27 (Annex C) the same trends for different combinations of stations/area types and regions are shown. The high variability from year to year of the averaged $NO_x$ annual mean concentrations over the Southern region may be due to the low number of stations (29) with data available. As Figure 17 shows, most of the stations available in the region Centre and East are in Germany, which should be taken into consideration when analysing the trends.



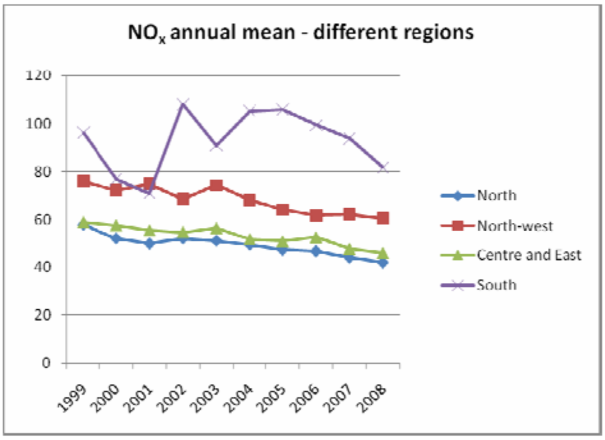




**Figure 18:** ***The average concentrations for different station/area types (left) and regions (right), for $NO_x$ annual mean in µg.m$^{-3}$, for the years 1999 – 2008.***

## 2.2.2 Trend at the individual stations

As for the analysis of the $NO_2$ trends, the trends of annual mean $NO_x$ concentrations for individual stations were also calculated in order to evaluate the variability of the trends within the regions or/and types of stations. Figure 17 shows in yellow and pink the stations used in this analysis, In Table 7 the distribution of the Sen's slope across the different group of the stations is presented for $NO_x$ annual mean, separately for the significant and non-significant trends (based on Mann-Kendal's test).

**Table 7:** ***Percentage distribution of the Sen's slope at the individual stations for $NO_x$ annual mean, for the years 1999-2008, for all stations together and separately for different station/area types and regions. The number of the stations in relevant group, the percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. The Sen' slope represents the annual change of concentration, in µg.m$^{-3}$.***

| group of the stations | N | < -4 | | -4 - -2 | | -2 - -1 | | -1 - 0 | | 0 - 1 | | 1 - 2 | | 2 - 4 | | > 4 | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. |
| all stations | 887 | 7,1 | 0,7 | 10,5 | 2,5 | 12,6 | 5,4 | 11,3 | 27,6 | 1,6 | 15,3 | 0,6 | 1,7 | 0,6 | 1,2 | 0,2 | 1,1 |
| rural background | 185 | 0,0 | 0,0 | 0,0 | 0,0 | 3,2 | 1,1 | 27,6 | 36,8 | 4,3 | 25,9 | 0,0 | 0,5 | 0,5 | 0,0 | 0,0 | 0,0 |
| suburban background | 129 | 0,8 | 0,0 | 3,9 | 0,0 | 10,9 | 5,4 | 17,8 | 43,4 | 0,0 | 13,2 | 0,0 | 1,6 | 0,0 | 2,3 | 0,8 | 0,0 |
| urban background | 213 | 2,8 | 0,0 | 12,2 | 1,9 | 26,8 | 5,2 | 6,6 | 23,9 | 0,0 | 14,1 | 1,4 | 2,3 | 0,9 | 0,9 | 0,0 | 0,9 |
| traffic (all types) | 252 | 21,8 | 2,4 | 19,8 | 6,3 | 9,5 | 8,7 | 0,8 | 14,7 | 0,8 | 7,5 | 0,0 | 1,6 | 0,8 | 2,0 | 0,0 | 3,2 |
| industrial (all types) | 108 | 0,9 | 0,0 | 11,1 | 1,9 | 10,2 | 5,6 | 9,3 | 30,6 | 3,7 | 20,4 | 1,9 | 2,8 | 0,0 | 0,9 | 0,9 | 0,0 |
| Northern Europe | 32 | 6,3 | 0,0 | 9,4 | 3,1 | 18,8 | 3,1 | 9,4 | 28,1 | 0,0 | 18,8 | 3,1 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| North-western E. | 130 | 10,8 | 0,0 | 13,1 | 2,3 | 15,4 | 7,7 | 10,0 | 34,6 | 0,0 | 5,4 | 0,8 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| Central + Eastern E. | 515 | 6,0 | 0,4 | 11,3 | 1,2 | 14,8 | 3,9 | 15,7 | 27,6 | 1,7 | 15,9 | 0,4 | 1,0 | 0,2 | 0,0 | 0,0 | 0,0 |
| Southern Europe | 210 | 7,6 | 1,9 | 7,1 | 5,7 | 4,8 | 8,1 | 1,4 | 23,3 | 2,4 | 19,5 | 0,5 | 4,8 | 1,9 | 5,2 | 1,0 | 4,8 |
| rural backgr. - N | 10 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 10,0 | 40,0 | 0,0 | 50,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| rural backgr. - NW | 33 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 3,0 | 27,3 | 57,6 | 0,0 | 12,1 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| rural backgr. - CE | 118 | 0,0 | 0,0 | 0,0 | 0,0 | 5,1 | 0,0 | 34,7 | 28,8 | 4,2 | 26,3 | 0,0 | 0,8 | 0,0 | 0,0 | 0,0 | 0,0 |
| rural backgr. - S | 24 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 4,2 | 0,0 | 45,8 | 12,5 | 33,3 | 0,0 | 0,0 | 4,2 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - N | 1 | 0,0 | 0,0 | 0,0 | 0,0 | 100 | 0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - NW | 13 | 0,0 | 0,0 | 7,7 | 0,0 | 23,1 | 7,7 | 7,7 | 53,8 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - CE | 96 | 1,0 | 0,0 | 3,1 | 0,0 | 8,3 | 6,3 | 22,9 | 44,8 | 0,0 | 12,5 | 0,0 | 1,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| suburb. backgr. - S | 19 | 0,0 | 0,0 | 5,3 | 0,0 | 10,5 | 0,0 | 0,0 | 31,6 | 0,0 | 26,3 | 0,0 | 5,3 | 0,0 | 15,8 | 5,3 | 0,0 |
| urban backgr. - N | 8 | 0,0 | 0,0 | 0,0 | 0,0 | 25,0 | 0,0 | 12,5 | 62,5 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| urban backgr. - NW | 37 | 8,1 | 0,0 | 21,6 | 8,1 | 32,4 | 8,1 | 2,7 | 13,5 | 0,0 | 5,4 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| urban backgr. - CE | 132 | 0,8 | 0,0 | 12,9 | 0,0 | 31,8 | 3,0 | 9,1 | 25,0 | 0,0 | 15,9 | 1,5 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| urban backgr. - S | 36 | 5,6 | 0,0 | 2,8 | 2,8 | 2,8 | 11,1 | 0,0 | 22,2 | 0,0 | 19,4 | 2,8 | 13,9 | 5,6 | 5,6 | 0,0 | 5,6 |
| industrial - N | 3 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 33,3 | 0,0 | 0,0 | 33,3 | 33,3 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| industrial - NW | 22 | 4,5 | 0,0 | 22,7 | 0,0 | 13,6 | 4,5 | 4,5 | 45,5 | 0,0 | 0,0 | 4,5 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| industrial - CE | 32 | 0,0 | 0,0 | 6,3 | 0,0 | 9,4 | 0,0 | 15,6 | 34,4 | 6,3 | 25,0 | 0,0 | 3,1 | 0,0 | 0,0 | 0,0 | 0,0 |
| industrial - S | 51 | 0,0 | 0,0 | 9,8 | 3,9 | 9,8 | 9,8 | 5,9 | 23,5 | 3,9 | 25,5 | 0,0 | 3,9 | 0,0 | 2,0 | 2,0 | 0,0 |
| traffic - N | 10 | 20,0 | 0,0 | 30,0 | 10,0 | 30,0 | 10,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| traffic - NW | 25 | 40,0 | 0,0 | 12,0 | 0,0 | 8,0 | 16,0 | 4,0 | 16,0 | 0,0 | 4,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 | 0,0 |
| traffic - CE | 137 | 21,2 | 1,5 | 26,3 | 4,4 | 12,4 | 7,3 | 0,7 | 15,3 | 1,5 | 7,3 | 0,0 | 1,5 | 0,7 | 0,0 | 0,0 | 0,0 |
| traffic - S | 80 | 17,5 | 5,0 | 10,0 | 11,3 | 2,5 | 8,8 | 0,0 | 15,0 | 0,0 | 10,0 | 0,0 | 2,5 | 1,3 | 6,3 | 0,0 | 10,0 |

The same distributions as in Table 7 are presented also as histograms in Figure 19.

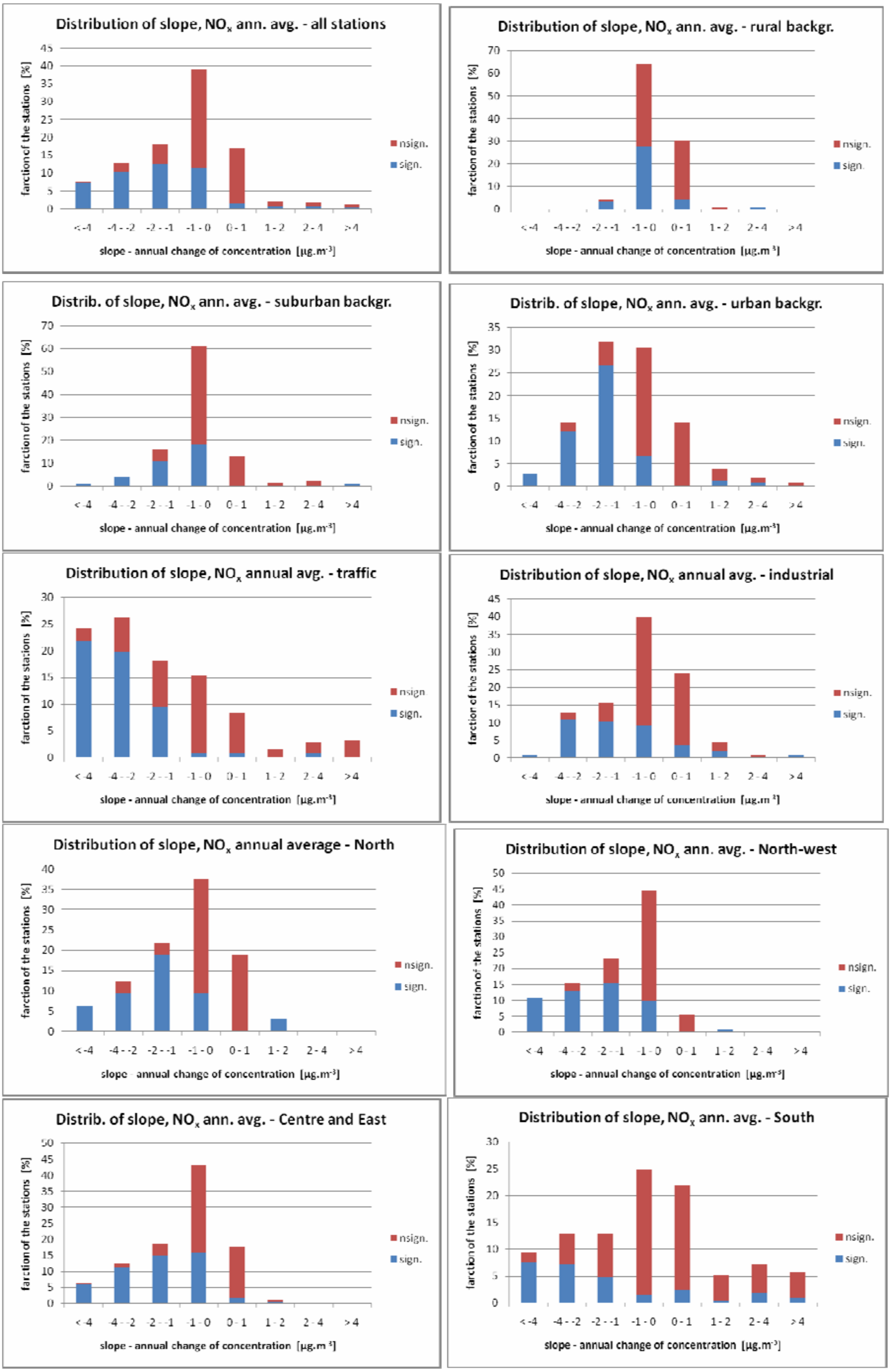



**Figure 19:** ***Distribution of the Sen's slope at the individual stations for $NO_x$ annual mean, for the years 1999-2008, for all stations together and separately for different station/area types and regions.***

In Figure 20 similar histograms for different combinations of stations/area types and regions are shown.

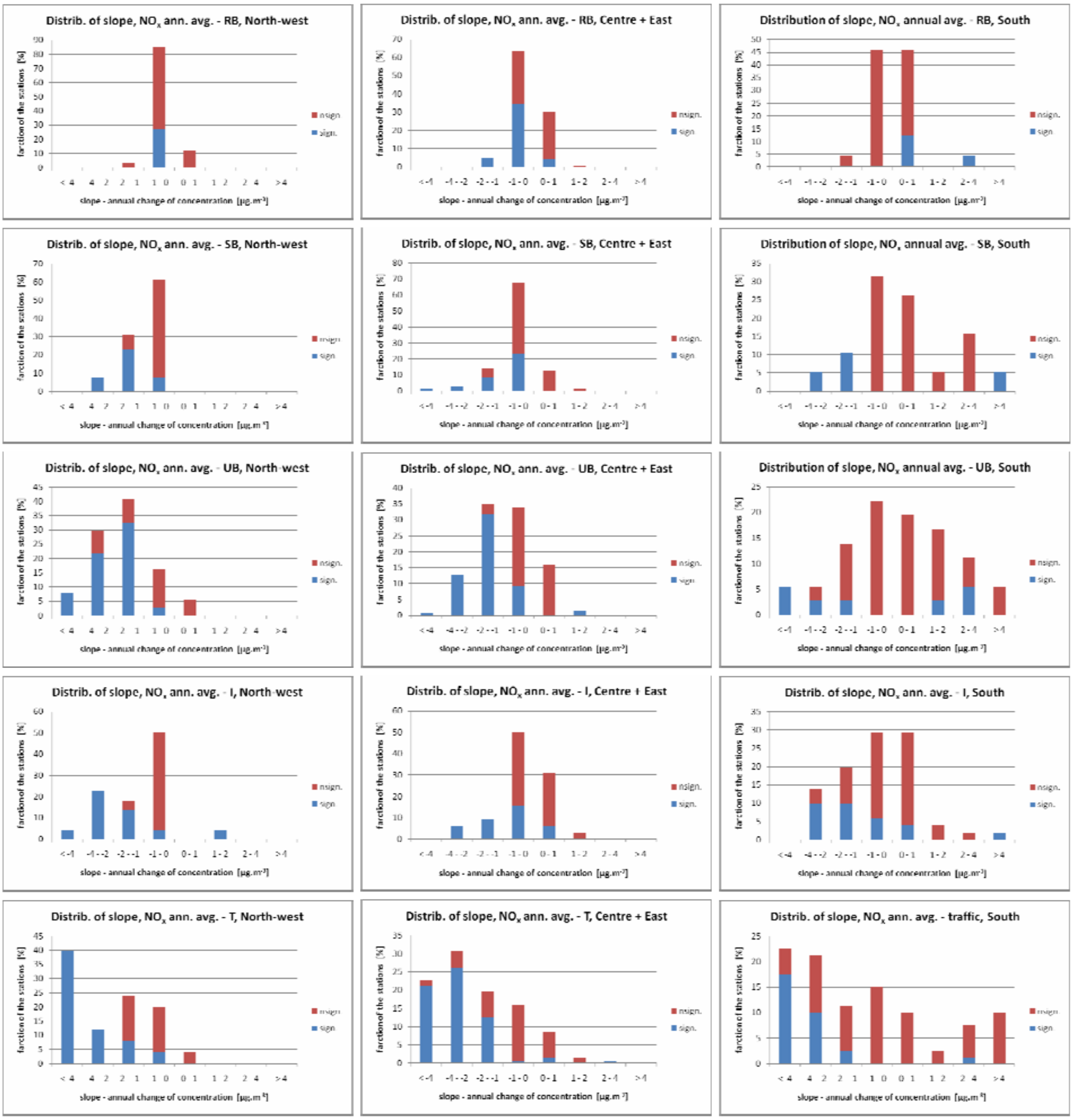



**Figure 20:** ***Distribution of the Sen's slope at the individual stations for $NO_x$ annual mean, for the years 1999-2008, separately for different station/area types and regions. (North region is not shown, due to small number of stations.) The percentage of the stations of the relevant group in different slope intervals and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. Significant trend is in blue, non-significant trend is in red.***

This analysis shows that many more traffic stations across Europe have had a significant decreasing trend of annual mean $NO_x$ concentrations (52%) than for $NO_2$ (25%). Concomitantly, a significant increasing trend was registered at 8.4% of the $NO_2$ traffic stations, while only at 1.6% of the $NO_x$ traffic stations. This tendency is registered in all analysed European regions, except in Southern Europe where the difference between $NO_x$ and

$NO_2$ stations is not so big. It is important to note that in total 40% of the $NO_2$ traffic stations had data for both $NO_2$ and $NO_x$ concentrations, while this is the case for only 10% of traffic stations in the Southern region and 26% in the North-western region (based on number of stations presented in Table 13). These results are consistent with the $NO_2/NO_x$ ratio trend analysis presented in Table 13, in chapter 2.4.

In the Northern, North-western and especially in the Central and Eastern region, there are also more stations registering decreasing trends for $NO_x$ than for $NO_2$ annual means. In the Central and eastern region 48% of the $NO_x$ stations registered a significant decreasing trend, while this only happened at 28% of the $NO_2$ stations. This is more clearly pronounced at the traffic stations, with 24% against 61% of the stations registering a significant decreasing trend for $NO_2$ and $NO_x$, respectively in the Central and eastern region. This analysis is also consistent with the trend analysis of the ratio $NO_2/NO_x$ presented in Table 13, in chapter 2.4.

On the contrary, in the Southern European region a higher percentage of stations has had an increasing trend (both significant and non significant) for $NO_x$ concentrations than for $NO_2$, especially at suburban and urban background stations, but also in rural background and at industrial stations. Only at traffic stations the percentage of stations with decreasing trends is slightly higher for $NO_x$ than for $NO_2$, as registered in other regions. It is never the less important to note that the number of stations that was the basis for the $NO_x$ trend calculations is about two thirds of the number of stations used for the $NO_2$ trend analysis. Furthermore, the number of stations with available data for both $NO_2$ and $NO_x$ is only 5% of the number of stations with available data for $NO_2$ (10% for traffic stations). The higher percentage of stations with increasing $NO_x$ concentration trends, compared to $NO_2$ stations, may be explained by the fact that more stations with $NO_x$ data were situated in areas where concentrations increased.

## 2.3. Development of the number of the stations and zones above LV and MT

The development of the number of the stations above the $NO_2$ limit values (LV) and margin of tolerance (MT) in the years 1999-2008 was also examined. In order to be consistent with the previous trend analysis, the fixed set of the stations with the valid data for all 10 years was used (see Annex A, Input data). The purpose of this examination is to analyse the development of the number of stations above LV and MT. The total number of analysed stations is limited to the ones with data available for the whole period and it does not correspond to the total number of stations above the LV or MT for each year.

Additionally, and based on this fixed set of stations, the development of the number of the zones above LV (resp. MT) was analysed based on the zones reported by Member states for 2008. Only the zones including at least one station during the period were considered.

In Table 8 and Table 9 the development of the stations and zones above limit value, for annual average and the 19$^{th}$ highest hour value is presented. It is clear from the analyses that there is a decreasing trend in the number of stations and in the number of zones in exceedance of the $NO_2$ annual mean LV in Europe. On the other hand, there has been no general improvement as to compliance with the hourly $NO_2$ limit value in Europe.

**Table 8:** ***Development of the number of stations (and type of stations) and the number of zones (also by region) above $NO_2$ annual average limit value 40 µg.m$^{-3}$ in the years 1999-2008, based on the fixed set of the stations with valid data in all 10 years and the zones as reported for 2008.***

| group of the stations/zones | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 755 | 159 | 131 | 137 | 126 | 158 | 120 | 125 | 126 | 107 | 98 | -4,71 | ** |
| rural background stations | 158 | 1 | 0 | 1 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 0,00 | no |
| suburban background st. | 159 | 8 | 6 | 5 | 3 | 7 | 1 | 2 | 2 | 1 | 0 | -0,80 | ** |
| urban background stations | 178 | 29 | 15 | 16 | 15 | 24 | 16 | 14 | 9 | 8 | 4 | -1,83 | ** |
| traffic stations | 195 | 119 | 108 | 112 | 105 | 125 | 102 | 106 | 113 | 97 | 93 | -2,00 | + |
| industrial stations | 64 | 2 | 2 | 3 | 3 | 2 | 1 | 2 | 2 | 1 | 1 | -0,13 | + |
| zones in Northern Europe | 24 | 2 | 1 | 1 | 1 | 1 | 1 | 1 | 2 | 1 | 1 | 0,00 | no |
| zones in North-western Europe | 206 | 53 | 42 | 47 | 37 | 53 | 37 | 38 | 33 | 28 | 25 | -3,00 | ** |
| zones in Central and Eastern Eur. | 418 | 66 | 54 | 50 | 51 | 70 | 51 | 54 | 60 | 45 | 40 | -1,50 | no |
| zones in Southern Europe | 106 | 38 | 34 | 39 | 37 | 34 | 31 | 32 | 31 | 33 | 32 | -0,80 | * |
| all zones | 238 | 77 | 60 | 69 | 64 | 77 | 58 | 64 | 62 | 53 | 51 | -2,17 | * |

**Table 9:** ***Development of the number of stations (and type of stations) and the number of zones (also by region) above the $NO_2$ maximum 19$^{th}$ hourly limit value 200 µg.m$^{-3}$ in the years 1999-2008, based on the fixed set of the stations with valid data in all 10 years and the zones as reported for 2008.***

| group of the stations/zones | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | [1]signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 737 | 21 | 23 | 16 | 14 | 21 | 16 | 20 | 21 | 22 | 14 | 0,00 | no |
| rural background stations | 139 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| suburban background st. | 160 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| urban background stations | 178 | 1 | 0 | 0 | 0 | 0 | 1 | 1 | 0 | 0 | 0 | 0,00 | no |
| traffic stations | 197 | 19 | 23 | 16 | 14 | 21 | 15 | 19 | 21 | 22 | 14 | 0,00 | no |
| industrial stations | 62 | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0,00 | no |
| zones in Northern Europe | 14 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| zones in North-western Europe | 206 | 6 | 7 | 6 | 6 | 10 | 6 | 6 | 6 | 9 | 6 | 0,00 | no |
| zones in Central and Eastern Eur. | 410 | 2 | 2 | 0 | 1 | 6 | 1 | 3 | 5 | 2 | 2 | 0,17 | no |
| zones in Southern Europe | 106 | 13 | 14 | 10 | 7 | 5 | 9 | 11 | 10 | 11 | 6 | -0,43 | no |
| all zones | 227 | 13 | 11 | 12 | 9 | 16 | 9 | 13 | 14 | 13 | 10 | 0,00 | no |

[1] Significance “no” means no significant trend was found.

In Figure 21, the development of the number of the stations and the zones above LV is presented.

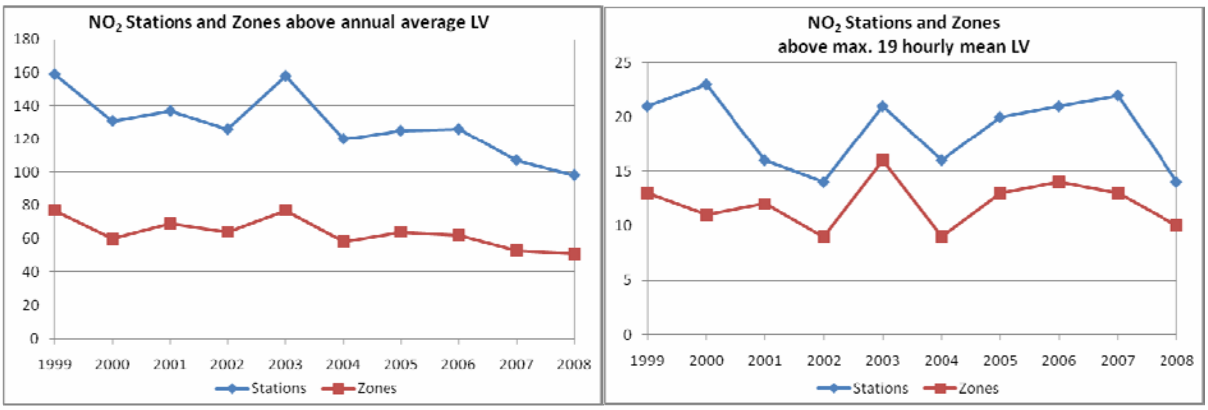



**Figure 21:** ***Development of the number of stations and zones above $NO_2$ annual average (left) and maximum 19$^{th}$ hourly limit value (right) in the years 1999-2008.***

In addition, the analysis of the development of the stations above the limit value including the margin of tolerance (MT) was examined, in order to see whether the decrease of $NO_2$ concentrations is able to satisfy the decrease requirements set by the decrease of the MT.

The margin of tolerance is decreasing constantly from the year 2001, as can be seen in Table 10.

**Table 10:** ***Development of the margin of tolerance in the years 1999-2008.***

| | LV | limit value (LV) including margin of tolerance (MT) | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 |
| annual average | 40 | 60 | 60 | 58 | 56 | 54 | 52 | 50 | 48 | 46 | 44 |
| maximum 19th hourly value | 200 | 300 | 300 | 290 | 280 | 270 | 260 | 250 | 240 | 230 | 220 |

The results are presented in Table 11 and Table 12.

**Table 11:** ***Development of the number of the stations and zones above $NO_2$ annual average limit value (LV) including margin of tolerance (MT) in the years 1999-2008, based on the fixed set of the stations with valid data in all 10 years and the zones as reported for 2008. The number of the stations above MT for all stations together and separately for different station/area types and regions, the number of the zones above MT, the estimated Sen's slope and the result of Mann-Kendall's test are presented.***

| group of the stations/zones | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 755 | 38 | 37 | 35 | 44 | 53 | 48 | 61 | 69 | 72 | 76 | 4,88 | ** |
| rural background | 158 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| suburban background | 159 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| urban background | 178 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 1 | 2 | 1 | 0,13 | * |
| traffic (all area types) | 195 | 38 | 37 | 35 | 44 | 52 | 48 | 61 | 68 | 70 | 74 | 4,50 | ** |
| industrial (all area types) | 64 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0,00 | no |
| Northern Europe | 24 | 0 | 0 | 0 | 1 | 0 | 0 | 0 | 1 | 1 | 1 | 0,00 | + |
| North-western Europe | 206 | 14 | 14 | 13 | 13 | 16 | 14 | 14 | 15 | 18 | 19 | 0,50 | * |
| Central and Eastern Eur. | 418 | 11 | 9 | 10 | 15 | 20 | 16 | 26 | 32 | 28 | 30 | 2,63 | ** |
| Southern Europe | 106 | 13 | 14 | 12 | 15 | 17 | 18 | 21 | 21 | 25 | 26 | 1,50 | *** |
| all zones | 238 | 22 | 19 | 19 | 26 | 29 | 25 | 32 | 33 | 34 | 40 | 2,00 | ** |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1(meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

**Table 12:** ***Development of the number of stations and zones above $NO_2$ maximum 19th hourly limit value (LV) including margin of tolerance (MT) in the years 1999-2008, based on the fixed set of the stations with valid data in all 10 years and the zones as reported for 2008. The number of the stations above MT for all stations together and separately for different station/area types and regions, the number of the zones above MT, the estimated Sen's slope and the result of Mann-Kendall's test are presented.***

| group of the stations/zones | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 737 | 3 | 1 | 0 | 0 | 1 | 3 | 2 | 8 | 8 | 9 | 1,00 | * |
| rural background | 139 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| suburban background | 160 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| urban background | 178 | 1 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0,00 | no |
| traffic (all area types) | 197 | 2 | 1 | 0 | 0 | 1 | 3 | 2 | 8 | 8 | 9 | 1,00 | * |
| industrial (all area types) | 62 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| Northern Europe | 14 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | | no |
| North-western Europe | 206 | 0 | 1 | 0 | 0 | 1 | 1 | 1 | 3 | 4 | 6 | 0,50 | ** |
| Central and Eastern Eur. | 410 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 1 | 0 | 0 | 0,00 | no |
| Southern Europe | 106 | 3 | 0 | 0 | 0 | 0 | 2 | 1 | 4 | 4 | 3 | 0,33 | + |
| all zones | 227 | 3 | 1 | 0 | 0 | 1 | 3 | 2 | 7 | 6 | 6 | 0,63 | ** |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1(meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

In Figure 22 the development of the number of stations and zones above MT is presented.

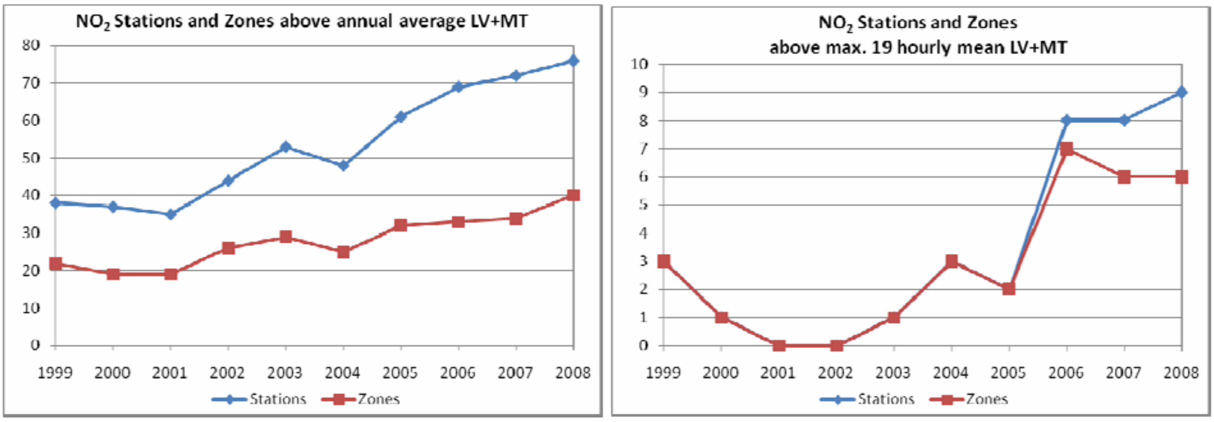



**Figure 22:** ***Development of the number of stations and zones above $NO_2$ annual average (left) and maximum 19th hourly mean (right) limit value including margin of tolerance (MT) in the years 1999-2008.***

It can be seen that the concentrations do not decrease as the margin of tolerance, and thus both the number of stations and zones above limit value plus the margin of tolerance increases, especially for annual average.

## 2.4. Trends in the concentration ratio $NO_2$ /$NO_x$

The trend of the ratio $NO_2$/$NO_x$ of measured concentrations in the years 1999-2008 was examined in order to see if there is an increasing trend is some regions and station types that may be related to a change in the primary $NO_2$ emissions from vehicles.

The ratios were calculated for all the individual stations and all the years, based on annual average $NO_2$ and $NO_x$ values. The ratio values above 1 were excluded. In the trend analysis, two sets of the stations are used: 1) for spatial average, only the stations with ratios for all the ten years were used; 2) for examining of individual stations, the stations with ratios for at least eight years were used. The number of the stations with sufficient ratio data is for (1) the spatial average 448, and for (2) the individual stations analysis 873 stations.

### 2.4.1 Trend of the average of the stations

The average annual mean $NO_2/NO_x$ ratios were calculated for all the years for different station/area types and four main European regions, as well as for the whole set of the stations. In Table 13 these values are presented, as well as the estimated Sen's slope and the result of Mann-Kendall's test for testing the presence of the trend. Figure 28 and Figure 29 in Annex D show the same results as figures.

**Table 13:** ***Trend analysis for $NO_2/NO_x$ ratio calculated based on the annual averages, for the years 1999-2008, for all stations together and separately for different station/area types and regions. The number of the stations in relevant group, the average ratio values of the group for individual years, the estimated Sen's slope and the result of Mann-Kendall's test for testing of the presence of the monotonic increasing or decreasing trend are presented. The Sen' slope represents the annual change of $NO_2/NO_x$ ratio.***

| group of the stations | N | 1999 | 2000 | 2001 | 2002 | 2003 | 2004 | 2005 | 2006 | 2007 | 2008 | slope | signif. |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| all stations | 448 | 0,58 | 0,58 | 0,58 | 0,58 | 0,59 | 0,59 | 0,61 | 0,61 | 0,61 | 0,63 | 0,01 | * |
| rural background | 102 | 0,75 | 0,75 | 0,75 | 0,74 | 0,75 | 0,74 | 0,78 | 0,77 | 0,78 | 0,78 | 0,00 | + |
| suburban background | 78 | 0,62 | 0,62 | 0,62 | 0,61 | 0,63 | 0,63 | 0,65 | 0,63 | 0,64 | 0,64 | 0,00 | * |
| urban background | 108 | 0,58 | 0,56 | 0,56 | 0,57 | 0,58 | 0,59 | 0,61 | 0,61 | 0,61 | 0,64 | 0,01 | ** |
| traffic (all area types) | 127 | 0,41 | 0,42 | 0,43 | 0,42 | 0,45 | 0,44 | 0,46 | 0,47 | 0,47 | 0,48 | 0,01 | *** |
| industrial (all area types) | 33 | 0,61 | 0,60 | 0,58 | 0,59 | 0,59 | 0,59 | 0,61 | 0,62 | 0,61 | 0,61 | 0,00 | no |
| Northern Europe | 13 | 0,60 | 0,60 | 0,61 | 0,63 | 0,61 | 0,61 | 0,63 | 0,63 | 0,62 | 0,64 | 0,00 | ** |
| North-western Europe | 68 | 0,60 | 0,61 | 0,56 | 0,58 | 0,59 | 0,60 | 0,62 | 0,63 | 0,62 | 0,63 | 0,01 | * |
| Central and Eastern Eur. | 350 | 0,58 | 0,57 | 0,58 | 0,58 | 0,59 | 0,59 | 0,62 | 0,61 | 0,62 | 0,63 | 0,01 | ** |
| Southern Europe | 17 | 0,46 | 0,66 | 0,68 | 0,43 | 0,51 | 0,47 | 0,48 | 0,48 | 0,49 | 0,53 | 0,00 | no |
| rural background - N | 3 | 0,85 | 0,82 | 0,83 | 0,87 | 0,84 | 0,86 | 0,89 | 0,87 | 0,87 | 0,89 | 0,01 | * |
| rural background - NW | 21 | 0,76 | 0,75 | 0,69 | 0,71 | 0,73 | 0,71 | 0,76 | 0,76 | 0,75 | 0,76 | 0,00 | no |
| rural background - CE | 77 | 0,75 | 0,74 | 0,76 | 0,74 | 0,75 | 0,75 | 0,78 | 0,77 | 0,78 | 0,79 | 0,00 | ** |
| rural background - S | 1 | 0,41 | 0,78 | 0,95 | 0,53 | 0,58 | 0,55 | 0,57 | 0,57 | 0,56 | 0,60 | 0,00 | no |
| suburban backgr. - N | 0 | no station with complete data | | | | | | | | | | | |
| suburban backgr. - NW | 8 | 0,62 | 0,65 | 0,58 | 0,62 | 0,62 | 0,63 | 0,65 | 0,67 | 0,62 | 0,65 | 0,01 | + |
| suburban backgr. - CE | 68 | 0,62 | 0,61 | 0,61 | 0,62 | 0,63 | 0,63 | 0,65 | 0,63 | 0,64 | 0,64 | 0,00 | ** |
| suburban backgr. - S | 2 | 0,56 | 0,92 | 0,96 | 0,52 | 0,57 | 0,61 | 0,61 | 0,59 | 0,58 | 0,64 | 0,00 | no |
| urban background - N | 4 | 0,69 | 0,57 | 0,59 | 0,57 | 0,60 | 0,59 | 0,62 | 0,60 | 0,61 | 0,59 | 0,00 | no |
| urban background - NW | 13 | 0,59 | 0,61 | 0,61 | 0,62 | 0,62 | 0,63 | 0,66 | 0,64 | 0,65 | 0,65 | 0,01 | ** |
| urban background - CE | 88 | 0,57 | 0,73 | 0,73 | 0,72 | 0,73 | 0,73 | 0,76 | 0,75 | 0,75 | 0,76 | 0,01 | * |
| urban background - S | 3 | 0,72 | 0,60 | 0,59 | 0,55 | 0,57 | 0,58 | 0,60 | 0,59 | 0,58 | 0,59 | 0,00 | no |
| industrial - N | 1 | 0,65 | 0,50 | 0,53 | 0,56 | 0,58 | 0,63 | 0,62 | 0,59 | 0,62 | 0,62 | 0,01 | no |
| industrial - NW | 10 | 0,61 | 0,56 | 0,55 | 0,56 | 0,57 | 0,58 | 0,60 | 0,60 | 0,60 | 0,70 | 0,01 | no |
| industrial - CE | 22 | 0,61 | 0,55 | 0,55 | 0,56 | 0,58 | 0,59 | 0,60 | 0,58 | 0,60 | 0,60 | 0,01 | no |
| industrial - S | 0 | no station with complete data | | | | | | | | | | | |
| traffic - N | 5 | 0,37 | 0,33 | 0,36 | 0,38 | 0,41 | 0,40 | 0,42 | 0,43 | 0,43 | 0,42 | 0,01 | ** |
| traffic - NW | 16 | 0,40 | 0,40 | 0,40 | 0,43 | 0,45 | 0,45 | 0,47 | 0,47 | 0,47 | 0,49 | 0,01 | *** |
| traffic - CE | 95 | 0,41 | 0,53 | 0,52 | 0,53 | 0,54 | 0,54 | 0,56 | 0,57 | 0,57 | 0,58 | 0,01 | *** |
| traffic - S | 11 | 0,37 | 0,46 | 0,47 | 0,42 | 0,45 | 0,45 | 0,46 | 0,47 | 0,47 | 0,48 | 0,01 | * |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1 (meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

The analysis shows that the average of the $NO_2/NO_x$ annual means ratio has an increasing trend of 0,8% a year for the period of 1999 to 2008 at traffic stations and urban background stations. At suburban background stations, the increasing trend in the $NO_2/NO_x$ ratio is of 0.3% a year and of 0.5% a year for the rural background stations. All the analysed regions in Europe registered an increasing trend, with the exception of southern Europe, where there was no significant average trend.

The traffic stations registered significant increasing average $NO_2/NO_x$ ratio trends for all the four regions, especially for the Northern and North-western regions, with 0.11% average $NO_2/NO_x$ ratio increase a year. This is probably due to two main reasons: 1) the decrease in $NO_x$, leading to an increase in the $NO_2/NO_x$ ratio, due to a shift in the photostationary state, with at least as much ozone as earlier available; 2) changes in the traffic $NO_2/NO_x$ emission ratios, due to an increase of diesel vehicles share in the European vehicular park and due to an increase of the $NO_2$ fraction of $NO_x$ emissions. The second reason is further discussed in Chapter 3.

The industrial stations did not register significant $NO_2/NO_x$ ratio trends. Background stations (rural, urban and suburban) registered in average a significant increase in the $NO_2/NO_x$ ratio, although not as significant and the trend registered in traffic stations.

## 2.5. Analysis of concentrations and emissions trends

A decreasing trend for $NO_2$ annual mean is detected for most of the European regions and stations/area types. Nevertheless, in the cases where the trend is detected it is rather low, varying between -0.123 and -0.553 $\mu g.m^{-3}$ $NO_2$ per annum. In general, the average annual decrease of $NO_2$ between 1999 and 2008 was about -0.3 $\mu g.m^{-3}$ per annum. Contrary to the annual mean, no trend is detected for the most of the regions and stations/area types for $NO_2$ indicator 19$^{th}$ maximum hourly mean.

The decreasing trend for $NO_x$ annual mean concentration is detected for most of the regions and stations/area types. In the cases where the trend is detected, the slope varies between -0.28 and -4.62. The average annual decrease of $NO_x$ in 1999-2000 was about 1.3 $\mu g.m^{-3}$.

In general, $NO_x$ concentrations have had a stronger decreasing trend than $NO_2$, which is also seen by the general increasing trend in the $NO_2/NO_x$ concentration ratio. The main reason for this is an increase of the $NO_2/NO_x$ ratio, especially at urban traffic sites, and a higher real life $NO_x$ emissions of diesel vehicles than originally expected. Recently published emission factors for road traffic sources suggest that the decline in $NO_x$ emissions has been less pronounced than expected in recent years. This gap is expected to exist also for future emission projections, due to the differences in $NO_x$ emission factors between the regulatory test cycle and the real world driving cycle for the Euro 6 standards (Umweltbundesamt & AEA Technology, 2010).

In contrast to the average decreasing trend of $NO_2$ levels in recent years, an increase was observed in some regions especially at traffic stations. 39% of the European traffic stations with data for the period 1999 to 2008 registered an increase in $NO_2$ annual mean concentrations in that period. 8,3% of these traffic stations had a significant increasing trend, especially in the Central east region (11%), but also in the North-west region (8%) and in the South (6%). The main reasons for these concentration increases are probably an increase in the traffic volume in some regions/countries and/or developments of the share of diesel vehicles in different countries. Ozone long range transport can play a role for the formation of $NO_2$ as well.

In addition, 9% of the industrial stations in Central and Eastern Europe and 6% in Southern Europe registered significant increasing trends of $NO_2$ annual mean concentrations between 1999 and 2008. In total, 32% and 18% of the industrial stations, with data for the period, registered some increase in measured concentrations (significant and non significant trends) in Central and Eastern Europe and in Southern Europe, respectively.

Trends in $NO_x$ emissions were calculated used the MAKESENS application (Salmi et al., 2002) and are presented in Table 14. Trends in $NO_x$ emissions showed a constant decrease in EU27 as a total since 1990 (Air Quality Expert Group, 2007). Nevertheless, some countries in the Central and Eastern Europe as Austria, Bulgaria, Romania and Poland, as well as in Southern Europe as Greece and Spain have not registered a decreasing trend in their total $NO_x$ emissions between 1999 and 2008 (Table 14).

Road traffic $NO_x$ emissions decreased in average with -3.5% per annum in the EU27 between 1999 and 2008 (Table 14), as reported by the member states. The reported emissions are based on the emission factors from the approved test cycles and not from real world driving cycles. Most countries registered a decrease in their road traffic $NO_x$ emissions, with the exception of Austria, Bulgaria, Hungary, Lithuania, Romania and Slovakia. In the eastern European countries this increase is most likely due to an increase in traffic volume. The countries with the strongest road traffic $NO_x$ emission decrease in the period were Germany (-6.7%), Finland (-6.5%), United Kingdom (-5.9%), Sweden (-5.8%) and Malta (-7.0%), as shown in Table 14.

**Table 14:** ***Trend analysis for $NO_x$ road traffic and total annual emissions, for the years 1999-2008, for the EU-27countries. The number of years with available emission data, the average emissions in the period, the estimated Sen's slope and the result of Mann-Kendall's test for testing of the presence of the monotonic increasing or decreasing trend are presented. The Sen's slope represents the annual change of emissions, in gigagrams (Gg). The Sen's slope % represents the annual change of emissions as a percentage of the average emissions in the period.***

| Country | Road traffic emissions | | | | | Total emissions | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | n | Signif. | Slope | Aver. emis. | slope % | n | Signif. | Slope | Aver. Emis. | slope % |
| Austria | 10 | no | 0,8 | 137 | 0,6 | 10 | no | 1,9 | 221 | 0,8 |
| Belgium | 10 | * | -4,4 | 126 | -3,5 | 10 | * | -8,3 | 280 | -2,9 |
| Denmark | 10 | *** | -2,3 | 74 | -3,1 | 10 | ** | -5,7 | 189 | -3,0 |
| Finland | 10 | *** | -4,1 | 64 | -6,5 | 10 | ** | -4,6 | 199 | -2,3 |
| France | 10 | *** | -28,4 | 824 | -3,4 | 10 | *** | -37,8 | 1510 | -2,5 |
| Germany | 10 | *** | -53,1 | 794 | -6,7 | 10 | *** | -58,2 | 1619 | -3,6 |
| Greece | 10 | * | -2,3 | 114 | -2,0 | 10 | * | 4,3 | 360 | 1,2 |
| Ireland | 10 | *** | -1,4 | 53 | -2,6 | 10 | ** | -2,8 | 125 | -2,3 |
| Italy | 10 | *** | -30,5 | 681 | -4,5 | 10 | *** | -50,5 | 1307 | -3,9 |
| Luxembourg | 0 | | | | | 7 | no | -0,0 | 1 | -3,9 |
| Netherlands | 10 | *** | -4,8 | 137 | -3,5 | 10 | *** | -12,9 | 340 | -3,8 |
| Portugal | 10 | ** | -2,0 | 124 | -1,6 | 10 | ** | -5,5 | 310 | -1,8 |
| Spain | 10 | *** | -9,2 | 517 | -1,8 | 10 | no | 3,6 | 1403 | 0,3 |
| Sweden | 10 | *** | -5,3 | 93 | -5,8 | 10 | *** | -7,0 | 188 | -3,8 |
| United Kingdom | 10 | *** | -36,7 | 622 | -5,9 | 10 | *** | -42,2 | 1707 | -2,5 |
| Bulgaria | 10 | ** | 1,6 | 37 | 4,4 | 10 | * | 3,2 | 141 | 2,2 |
| Cyprus | 10 | * | -0,2 | 10 | -2,1 | 10 | no | -0,1 | 20 | -0,4 |
| Czech Republic | 10 | ** | -4,5 | 113 | -4,0 | 10 | * | -13,4 | 321 | -4,2 |
| Estonia | 10 | ** | -0,3 | 14 | -2,0 | 10 | no | -0,3 | 37 | -0,8 |
| Hungary | 10 | ** | 1,4 | 109 | 1,3 | 10 | no | -0,7 | 190 | -0,4 |
| Latvia | 10 | no | -0,4 | 19 | -2,0 | 10 | no | -0,2 | 41 | -0,5 |
| Lithuania | 10 | * | 1,4 | 29 | 4,7 | 9 | * | 2,3 | 53 | 4,4 |
| Malta | 10 | ** | -0,2 | 3 | -7,0 | 10 | no | -0,1 | 9 | -1,1 |
| Poland | 1 | no | | 256 | 0,0 | 10 | no | 3,3 | 845 | 0,4 |
| Romania | 10 | ** | 6,1 | 111 | 5,5 | 10 | * | 7,7 | 337 | 2,3 |
| Slovakia | 10 | * | 1,3 | 38 | 3,3 | 10 | ** | -1,8 | 102 | -1,8 |
| Slovenia | 9 | no | -0,2 | 19 | -0,9 | 10 | no | -1,3 | 84 | -1,5 |
| **EU-27** | **10** | ******* | **-169,6** | **4886** | **-3,5** | **10** | ******* | **-229,2** | **11933** | **-1,9** |

[1] The four significance levels of the Mann-Kendal test are: + for 0.1(meaning 10% probability there is no trend), * for 0.05, ** for 0.01, and *** for 0.001 (meaning 0,1% probability there is no trend). "No" means no significant trend was found. For more information on the Mann-Kendal test, see Annex A.

# 3. Analysis of the influence of an increase of diesel cars to the $NO_2$ concentrations development

Traffic is known to be by far the largest contributor to nitrogen dioxide ($NO_2$) concentrations, both at traffic sites and at urban background sites. In contrast to $PM_{10}$, long range and transboundary transport only play a minor role in most regions. This was also found in a recent study on the traffic influence on urban air quality in European cities (Hak et al., 2009). Compliance with the annual $NO_2$ limit value of 40 µg/m$^3$ (attainment date 1 January 2010) was expected for only few of the cities which had been affected by exceedances in the last few years, although measures have been taken both at local level and at EU level. The main reasons for the non-achievement of compliance are assumed to be a late start of planning and implementation of measures (Umweltbundesamt, 2006) and the underestimation of real world emissions from road vehicles compared to legislative limits, in combination with increasing primary $NO_2$ emissions from a larger fleet of diesel vehicles, which lead to a significant overestimation of the emission reduction potential of the current measures.

While the urban $NO_x$ concentrations in Europe showed a downward trend since the early 1990s, the proportion of $NO_x$ emitted directly as $NO_2$ from vehicles (primary $NO_2$ fraction, f-$NO_2$, often expressed as percentage) has been increasing since the early 2000s, as a result of an increased market penetration of diesel cars in some countries and the fitting of pollution control devices, e.g., particulate traps and oxidation catalysts for diesel EURO3 (and above). Thus, there is increasing concern that Member States may experience difficulty complying with the annual mean limit value for $NO_2$ of 40 µg m$^{-3}$.

A typical f-$NO_2$ for petrol vehicles is < 5%, the historically typical f-$NO_2$ for conventional diesel vehicles was 10-12%, while values are in the range 20-70% for newer diesel vehicles. The f-$NO_2$ varies regionally and is dependent on the local vehicle fleet and traffic conditions. An assessment of the average (Europe-wide) increase of f-$NO_2$ showed a development from 8.6% in 2000 to 12.4% in 2004 at ten case study monitoring sites (Grice et al., 2007). Future projections for f-$NO_2$ resulted in 19.6% in 2010 and 32.0% in 2020 (Grice et al., 2007).

Grice et al. (2009) published a study on trends and projections of primary $NO_2$ emissions in ten case study countries with different fleet compositions. Their predictions for urban road traffic emissions show a steady decrease of $NO_x$ emissions from 1995 until 2020. $NO_2$ emissions, however, increase steeply between 2000 and 2010 and are projected to reach a maximum around 2015 and decrease after 2015. In the majority of countries considered, the f-$NO_2$ has been rising from 1995 and is predicted to continue until 2020, with a maximum slope between 2005 and 2015, primarily due to the fitting of exhaust after-treatment systems. Urban $NO_2$ emissions are predicted to increase from 2000 to 2010 in contrast to the decline in $NO_x$ emissions. They are then predicted to flatten off to 2015 and then decline to roughly equivalent 2005 values in 2020 for the baseline. By 2020, the decrease in $NO_x$ emissions is expected to be sufficient to offset the increase in f-$NO_2$. Changes in vehicle exhaust after treatment technology, particularly selective catalytic reduction, are expected to result in a decrease in the emissions of primary $NO_2$ and an improvement in roadside air quality by 2020. Future $NO_2$ concentrations have been estimated based on the combination of emission inventory calculations and projections of ambient air quality, where $NO_x$ emissions were estimated from the TREMOVE model and f-$NO_2$ and $NO_2$ roadside concentrations were obtained from the Netcen primary $NO_2$ model. Modelled $NO_2$ ambient concentrations have been decreasing since 2005 and are projected to further decrease in most countries studied. In the Czech Republic and France, however, an increase until 2010 was found, followed by decreasing $NO_2$ concentrations (Grice et al., 2009).

In France, for example, the fraction of diesel vehicles was high already in the 1990s compared to many other European countries. A French study on $NO_2$ emissions of light vehicles between 1990 and 2014 shows a distinct increase with the introduction of the diesel oxidation catalyst for diesel EURO2 vehicles in ~1997 until ~2002 (AFSSET, 2009).

In Norway, in order reduce $CO_2$ emissions from traffic, the government introduced a new taxation system in 2007 that largely favours the purchase of new diesel cars, rather than gasoline cars. This, combined with changes in the diesel and gasoline sale taxes, has led to a considerable increase in the percentage kilometres driven by diesel and gasoline vehicles in recent years. Between 2005 and 2009, the total amount of driven kilometres by gasoline vehicles dropped by 16%, while kilometres driven by diesel vehicles increased by 63%. The corresponding figures for private cars are a 15% decrease for gasoline and 118% increase for diesel driven vehicles. (Statistics Norway, http://www.ssb.no/emner/10/12/20/klreg/tab-2010-05-11-07.html).

**Technical aspects**

The increase in f-$NO_2$ can be ascribed to technical development on the diesel sector, while the f-$NO_2$ of petrol fuelled vehicles has remained around 3-4% for all technologies and emission standards. Exhaust after-treatment systems have been installed in diesel vehicles in order to reduce CO and HC or particle emission; the drawback for some of the after-treatment technologies is increased primary $NO_2$ emission. The effect of exhaust after-treatment on $NO_2$ emission strongly depends on the after-treatment technology used. The differences in design of diesel particulate filters (DPF) are based on the way the DPFs are regenerated. The most commonly used types in Europe are of the *continuously regenerating type* (e.g. CRT®). These systems usually use an oxidation catalyst to deliberately oxidise the NO in the exhaust to $NO_2$, a part of which then oxidises the soot trapped on the filter (and thereby regenerates the filter). *Diesel oxidation catalysts* are relatively inexpensive and durable catalyst devices which have been fitted on many light duty diesel vehicles to reduce CO and HC emissions. They were also found to achieve some reduction in PM emission. However, the same process oxidises NO to $NO_2$, resulting in f-$NO_2$ increasing to around 30%. $NO_x$ reduction technologies, whilst leading to a reduction in $NO_x$, typically of between 30 and 50%, often also lead to an increase in f-$NO_2$, with ratios up to 60% being observed for some new passenger car technologies. *Fuel-borne catalysts* (FBC) have been shown to have a beneficial effect on PM, $NO_x$ and $NO_2$ emissions, reducing f-$NO_2$. FBC are added into the fuel on board of the vehicle. *Selective Catalytic Reduction* (SCR) can be applied to diesel exhausts to reduce $NO_x$ and $NO_2$ emissions, leading to lower f-$NO_2$. It has mostly been applied to heavy duty vehicles and marine diesel engines. Combined SCR-CRT after-treatment systems have been developed to simultaneously reduce PM and $NO_x$. SCR is the technology favoured by the majority of engine manufacturers to meet Euro IV emission standards. A detailed description of after treatment systems and their effects is given by Air Quality Expert Group (2007).

# 4. ANALYSIS OF APPLIED MEASURES FOR $NO_2$ COMPLIANCE

Various efficient measures to reduce ambient $NO_2$ levels, regardless of cost considerations, are presented in a report by the Austrian federal environmental agency and have been discussed at recent workshops (Umweltbundesamt, 2006, 2010). They include tight emission limit values for industries, traffic restrictions dependent on EURO standard (in combination with retrofitting schemes), congestion charge, low emission zones, progressive scrapping of lorries with a rating below EURO3, mobility management and enforcement of vehicle inspection.

Since road traffic is the major source of $NO_2$, most measures applied for $NO_2$ compliance are on the traffic sector. Examples for widely applied measures on local to national scale are presented here under.

**Reduction of traffic volume**, e.g. the introduction of areas within a city, where access for motor vehicles is restricted (*low emission zones, LEZ*), which is an urban/local scale measure. The restrictions are usually coupled to certain emission criteria (EURO class). LEZs were established in many European cities. In London, by a reduction of $NO_x$ emissions from road traffic of 4% in 2010 and 10% in 2012, the area where $NO_2$ exceedances occur was expected to be reduced by 5% until 2010 and 16% in 2012, compared to the normal course of events (Transport for London, 2008). $NO_x$ emission reduction up to 10% was estimated for Stockholm and London, 4.6% for Munich. Another urban/local scale measure is the introduction of a *congestion charge,* which was for example introduced in London and Stockholm and reduced $NO_x$ emissions from traffic by 13% in London. In Stockholm, the traffic volume went down by more than 20%, leading to a decrease in average $NO_x$ levels by 5-10 $\mu g/m^3$. The *increase of taxes and charges*, which are in most cases measures at national level, e.g., road pricing, also lead to a reduction in traffic volume in some cities. Charges depending on EURO standard and charges dependent on distance are also considered. The *improvement of public transport* and *encouragement of cycling and walking* are also measures implemented to reduce the traffic volume, mostly at local/urban level. A ban of transport of specific goods (e.g. waste, rocks, soil, rubble, timber, cork, cars, steel, tiles) by heavy duty vehicles (HDV) on motorway, favouring transport on rails has also been implemented or considered, mostly at regional scale.

**Change type of vehicles.** *Incentives* for gas, electric, low emission vehicles and *scrappage schemes* for old vehicles stimulate a change of the vehicle fleet. Also the *increase of tax on diesel* or other differentiating taxes/charges aim at changing the composition of the vehicle fleet are examples of this type of measures at national level. Similarly, the introduction of *environmental zones* and *ban of certain vehicles* encourage replacing an old car by a newer emission technology at local/urban scale.

**Changing emission factors of vehicles**, e.g. by establishing *speed limits* for passenger cars (local to national measure). Emission reductions following lower speed limits were found to be largest for diesel passenger cars and LDVs ($NO_x$ reduction of 25% and 33%, respectively) on motorways when reducing the speed from 130 or 120 to 100 or 80 km/h, since emission factors increase rapidly from ~100 km/h up (Umweltbundesamt, 2006). Even higher reduction is expected if the measure is enforced by section control. Positive side effects of these measures are reduced noise and GHG emission and fewer accidents. *Retrofitting* of diesel engines (both marine and heavy duty automotive) with *Selective Catalytic Reduction* (SCR) is an effective measure on national level to reduce $NO_x$ and $NO_2$ emissions (40-90%), resulting in a lower f-$NO_2$, however it is not considered feasible for passenger cars. On the marine sector, also the increased shift to *shore-side electricity* in the harbour and the *change of fuel* (lower N-content) reduce $NO_x$ emissions at local to regional scale.

**Reallocation of emissions** only relieves the situation at the hotspot site, while the amount of emissions stays constant. *Bypass roads*, *tunnels*, *night-time ban of HDV* and *ban of through-traffic* are measures which are applied to reallocate emissions spatially or temporally.
Many of the measures applied are soft measures (information, guidance, campaigns), where the effects on emissions and air quality are difficult to quantify.

**Non-traffic measures** to reduce ambient $NO_2$ levels are mainly aimed at the reduction of emissions at the source, e.g. by introduction of local measures as *district heating*, new installation of *combined heat and power plants* (CHP) and national measures as *enforcement of best available technology* for stationary sources. In some countries, a *$NO_x$ charge for installations* has been introduced at national level.

# 5. CONCLUSIONS

The decrease in $NO_2$ levels is much lower than anticipated by the policy makers and the present development is far from ensuring compliance with the $NO_2$ LVs in all European agglomerations and zones the next few years. Compliance with the annual $NO_2$ limit value of 40 µg/m$^3$ is expected for only a few of the cities which have been affected by exceedances in the last few years, although measures have been taken both at local level and at EU level. The main reasons for the non-achievement of compliance are assumed to be a late start of planning and implementation of measures and the underestimation of real world $NO_x$ emissions from road vehicles compared to legislative limits, in combination with increasing primary $NO_2$ emissions from diesel vehicles, which lead to a significant overestimation of the emission reduction potential of the current measures.

This study reveals that an increase of the ambient $NO_2/NO_x$ concentration ratio took place in the past decade, especially at urban traffic sites. While the $NO_x$ concentrations in Europe show in average a downward trend at all types of stations and in all European regions, with the exception of the South, the proportion of $NO_x$ emitted directly as $NO_2$ from vehicles has been increasing since the early 2000s, as a result of an increased market penetration of diesel cars in some countries and the fitting of pollution control devices, e.g., particulate traps and oxidation catalysts for diesel EURO3 (and above). On the other hand, an increase of the $NO_2/NO_x$ concentration ratio must be expected when NOx concentrations decrease, simply due to a shift in the photostationary state, with constant ozone and where ozone is a limiting factor for the oxidation of NO into $NO_2$. The increase in the $NO_2/NO_x$ concentration ratio is therefore due to both an increase in the $NO_2/NO_x$ ratio of primary emissions and a decrease in NOx, without an equivalent decrease in ozone concentrations.

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# Annex A - Input data and methodology for trends calculations

## Input data

Measured $NO_2$ air quality data were extracted from the European monitoring database AirBase. The following components and their indicators are considered:

$NO_2$ – annual mean [$\mu g.m^{-3}$], years 1999 - 2008
– $19^{th}$ maximum hourly mean value [$\mu g.m^{-3}$], years 1999 – 2008
$NO_x$ – annual mean [$\mu g.m^{-3}$], years 1999 - 2008
NO – annual mean [$\mu g.m^{-3}$], years 1999 – 2008 (for the purposes of $NO_x$ calculation only)

For all the years, only stations with temporal data coverage of at least 75 percent are used. For $NO_2$ 4006 stations are used. In the case of $NO_x$, 1977 stations with sufficient $NO_x$ data in at least one year are reported in AirBase. However, at many other stations $NO_x$ is measured, but not reported as such but separately as NO and $NO_2$. For these stations reporting NO and $NO_2$ separately, the $NO_x$ annual average concentrations were derived from $NO_2$ and NO annual averages according the equation:

$$NO_x = NO_2 + 46/30.NO \tag{2.1}$$

where all components are expressed in $\mu g.m^{-3}$, with a molecular mass for NO of 30 and for $NO_2$ of 46 $g.mol^{-1}$.

These stations were added to the set with reported $NO_x$ values resulting in an extended set of 3479 stations.

For trend analysis two sets of the stations were used: For spatial average, only the stations with sufficient data for all the ten years were used. For examining of individual stations, the stations with sufficient data for at least eight years were used. For $NO_2$ 1286, resp. 759 stations were selected (1280, resp. 755 stations for annual average, and 1269, resp. 737 stations for $19^{th}$ maximum hourly mean); for $NO_x$ these were 887, resp. 473 stations.
For development of the number of the zones above limit value the zones as reported by the Member states for the year 2008 were used. Only the zones which include at least one station from the fixed set of the stations with valid data for all the ten years, i.e. 238 (from the total number 836) zones were considered.

## Methodology

For estimating the general trend of $NO_x$ concentrations, different groups of stations were handled together. The average of the relevant groups was examined. In addition, the trends of individual stations were investigated for variability within the group.

The set of stations was divided in two ways: based on the station and area types and based on the region. Five different stations (resp. area) types were considered: rural background, suburban background, urban background, industrial and traffic. Industrial and traffic stations are handled without regard to area type.

Four European regions were considered, namely *Northern Europe*: Sweden, Finland, Norway, Estonia, Lithuania, Latvia and Denmark; *North-western Europe*: United Kingdom, Ireland, Iceland, the Netherlands, Belgium, Luxembourg and France north of 45 degrees latitude; *Central and Eastern Europe*: Germany, Poland, Czech Republic, Slovakia, Switzerland, Liechtenstein, Austria, Hungary, Bulgaria and Romania; *Southern Europe*: Albania, Bosnia-Herzegovina, France south of 45 degrees latitude, Portugal, Spain, Italy, San Marino, Slovenia, Croatia, Greece, Cyprus, F.Y.R. of Macedonia, Montenegro, Serbia and Malta.

For different station/area types and regions the average of the relevant stations was calculated. In addition, trends for the individual stations were calculated.

For detecting and estimating the trends in time series of annual values the nonparametric Mann-Kendall's test for testing the presence of the monotonic increasing or decreasing trend is used.

The Mann-Kendall test is applicable in cases when the data values $x_i$ of a time series can be assumed to obey the model

$$x_i = f(t_i) + \varepsilon_i \qquad (1)$$

where *f(t)* is a continuous monotonic increasing or decreasing function of time and the residuals $\varepsilon_i$ can be assumed to be from the same distribution with zero mean. It is therefore assumed that the variance of the distribution is constant in time.

We want to test the null hypothesis of no trend, $H_0$, i.e. the observations $x_i$ are randomly ordered in time, against the alternative hypothesis, $H_1$, where there is an increasing or decreasing monotonic trend.

The significance of the Mann-Kendal test is calculated and shown as + for 0.1, * for 0.05, ** for 0.01, and *** for 0.001. The significance level 0.001 means that there is a 0.1% probability that the values $x_i$ are from a random distribution and with that probability we make a mistake when rejecting $H_0$ of no trend. Thus the significance level 0.001 means that the existence of a monotonic trend is very probable. Respectively the significance level 0.1 means that there is a 10% probability that we make a mistake when rejecting $H_0$.

In addition, the nonparametric Sen's method for estimating the slope of a linear trend is executed. To estimate the true slope of an existing trend (as change per year) the Sen's nonparametric method is used. The Sen's method can be used in cases where the trend can be assumed to be linear. This means that *f(t)* in equation (1) is equal to

$$f(t) = Qt + B \qquad (2)$$

where Q is the slope and B is a constant.

# Annex B - Trend of the 19th highest hourly $NO_2$ concentrations at the individual stations

## Trend of the 19th highest hourly $NO_2$ concentrations at the individual stations

In Table 15 the distribution of the Sen's slope across the different group of the stations is presented for $NO_2$ indicator 19$^{th}$ maximum hourly mean, separately for the significant and non-significant trends (based on Mann-Kendal's test), in order to provide a closer look to these groups.

**Table 15:** ***Percentage distribution of the Sen's slope at the individual stations for $NO_2$ indicator 19$^{th}$ maximum hourly mean, for the years 1999-2008, for all stations together and separately for different station/area types and regions. The number of the stations in relevant group, the percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$.***

| group of the stations | N | < -4 | | -4 - -2 | | -2 - -1 | | -1 - 0 | | 0 - 1 | | 1 - 2 | | 2 - 4 | | > 4 | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. | s. | ns. |
| all stations | 1269 | 2.2 | 1.9 | 4.0 | 3.6 | 3.1 | 10.6 | 0.8 | 22.8 | 0.5 | 24.1 | 1.7 | 9.7 | 3.0 | 6.9 | 3.3 | 1.9 |
| rural background | 214 | 0.0 | 0.5 | 1.4 | 0.9 | 5.1 | 7.9 | 2.3 | 39.3 | 0.9 | 31.3 | 1.9 | 4.7 | 0.9 | 1.9 | 0.9 | 0.0 |
| suburban background | 235 | 1.3 | 1.7 | 4.7 | 1.3 | 3.0 | 11.1 | 1.3 | 26.4 | 0.4 | 27.7 | 0.9 | 11.9 | 3.4 | 3.8 | 0.4 | 0.9 |
| urban background | 351 | 1.1 | 1.4 | 4.3 | 3.1 | 4.3 | 12.5 | 0.3 | 22.5 | 0.3 | 25.1 | 1.7 | 10.3 | 2.3 | 7.4 | 2.0 | 1.4 |
| traffic (all types) | 323 | 4.6 | 3.1 | 4.0 | 5.9 | 0.6 | 9.6 | 0.0 | 12.1 | 0.3 | 16.1 | 1.5 | 11.1 | 5.6 | 12.4 | 9.0 | 4.0 |
| industrial (all types) | 144 | 4.2 | 2.8 | 6.3 | 7.6 | 2.8 | 11.8 | 0.7 | 17.4 | 0.7 | 22.9 | 2.8 | 8.3 | 1.4 | 5.6 | 2.1 | 2.8 |
| Northern Europe | 38 | 0.0 | 0.0 | 2.6 | 0.0 | 2.6 | 13.2 | 2.6 | 26.3 | 0.0 | 28.9 | 0.0 | 10.5 | 10.5 | 0.0 | 2.6 | 0.0 |
| North-western E. | 382 | 1.6 | 1.6 | 3.7 | 4.2 | 2.1 | 11.8 | 0.5 | 25.9 | 0.5 | 25.4 | 1.6 | 9.7 | 2.9 | 6.5 | 1.3 | 0.8 |
| Central + Eastern E. | 530 | 0.2 | 0.4 | 3.6 | 0.8 | 3.6 | 10.4 | 0.9 | 26.4 | 0.6 | 28.5 | 1.7 | 10.6 | 2.1 | 6.8 | 3.0 | 0.6 |
| Southern Europe | 319 | 6.6 | 5.0 | 5.3 | 8.2 | 3.4 | 9.4 | 0.6 | 12.5 | 0.3 | 14.7 | 1.9 | 8.2 | 3.8 | 8.2 | 6.3 | 5.6 |
| rural backgr. - N | 10 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 10.0 | 10.0 | 40.0 | 0.0 | 40.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| rural backgr. - NW | 54 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 9.3 | 0.0 | 53.7 | 0.0 | 29.6 | 0.0 | 3.7 | 0.0 | 1.9 | 1.9 | 0.0 |
| rural backgr. - CE | 118 | 0.0 | 0.0 | 0.0 | 0.0 | 7.6 | 7.6 | 2.5 | 38.1 | 1.7 | 33.1 | 3.4 | 3.4 | 0.0 | 2.5 | 0.0 | 0.0 |
| rural backgr. - S | 32 | 0.0 | 3.1 | 9.4 | 6.3 | 6.3 | 6.3 | 3.1 | 18.8 | 0.0 | 25.0 | 0.0 | 12.5 | 6.3 | 0.0 | 3.1 | 0.0 |
| suburb. backgr. - N | 1 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 100 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 |
| suburb. backgr. - NW | 96 | 2.1 | 2.1 | 7.3 | 3.1 | 4.2 | 11.5 | 2.1 | 26.0 | 0.0 | 26.0 | 1.0 | 8.3 | 1.0 | 5.2 | 0.0 | 0.0 |
| suburb. backgr. - CE | 100 | 0.0 | 0.0 | 1.0 | 0.0 | 0.0 | 8.0 | 1.0 | 31.0 | 1.0 | 33.0 | 1.0 | 17.0 | 4.0 | 3.0 | 0.0 | 0.0 |
| suburb. backgr. - S | 38 | 2.6 | 5.3 | 7.9 | 0.0 | 7.9 | 15.8 | 0.0 | 15.8 | 0.0 | 18.4 | 0.0 | 7.9 | 7.9 | 2.6 | 2.6 | 5.3 |
| urban backgr. - N | 13 | 0.0 | 0.0 | 7.7 | 0.0 | 7.7 | 7.7 | 0.0 | 30.8 | 0.0 | 30.8 | 0.0 | 15.4 | 0.0 | 0.0 | 0.0 | 0.0 |
| urban backgr. - NW | 127 | 1.6 | 1.6 | 3.9 | 4.7 | 2.4 | 14.2 | 0.0 | 22.8 | 0.0 | 24.4 | 2.4 | 11.0 | 2.4 | 7.9 | 0.8 | 0.0 |
| urban backgr. - CE | 138 | 0.0 | 0.0 | 5.8 | 1.4 | 4.3 | 15.2 | 0.7 | 24.6 | 0.0 | 29.0 | 0.0 | 7.2 | 2.2 | 8.0 | 0.7 | 0.7 |
| urban backgr. - S | 73 | 2.7 | 4.1 | 1.4 | 4.1 | 6.8 | 5.5 | 0.0 | 16.4 | 1.4 | 17.8 | 4.1 | 13.7 | 2.7 | 6.8 | 6.8 | 5.5 |
| industrial - N | 3 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 33.3 | 0.0 | 0.0 | 0.0 | 33.3 | 33.3 | 0.0 | 0.0 | 0.0 |
| industrial - NW | 41 | 2.4 | 2.4 | 2.4 | 4.9 | 2.4 | 9.8 | 0.0 | 22.0 | 2.4 | 31.7 | 4.9 | 12.2 | 2.4 | 0.0 | 0.0 | 0.0 |
| industrial - CE | 33 | 0.0 | 0.0 | 9.1 | 0.0 | 9.1 | 9.1 | 0.0 | 18.2 | 0.0 | 39.4 | 0.0 | 9.1 | 0.0 | 3.0 | 3.0 | 0.0 |
| industrial - S | 67 | 7.5 | 4.5 | 7.5 | 13.4 | 0.0 | 14.9 | 1.5 | 13.4 | 0.0 | 10.4 | 3.0 | 4.5 | 0.0 | 10.4 | 3.0 | 6.0 |
| traffic - N | 11 | 0.0 | 0.0 | 0.0 | 0.0 | 0.0 | 18.2 | 0.0 | 9.1 | 0.0 | 27.3 | 0.0 | 9.1 | 27.3 | 0.0 | 9.1 | 0.0 |
| traffic - NW | 63 | 1.6 | 1.6 | 1.6 | 7.9 | 0.0 | 11.1 | 0.0 | 11.1 | 1.6 | 19.0 | 0.0 | 11.1 | 9.5 | 14.3 | 4.8 | 4.8 |
| traffic - CE | 141 | 0.7 | 1.4 | 5.0 | 1.4 | 0.7 | 9.9 | 0.0 | 17.0 | 0.0 | 18.4 | 2.8 | 15.6 | 2.8 | 12.8 | 9.9 | 1.4 |
| traffic - S | 108 | 12.0 | 6.5 | 4.6 | 11.1 | 0.9 | 7.4 | 0.0 | 6.5 | 0.0 | 10.2 | 0.9 | 5.6 | 4.6 | 12.0 | 10.2 | 7.4 |

In Table 16 the number and percentage of the stations with significant trend is summarized.

**Table 16:** ***Number and percentage of the stations for which the monotonic trend of $NO_2$ indicator $19^{th}$ maximum hourly mean for the years 1999-2008 (based on Mann-Kendall's test, for significance level 0.10) is significant, for all stations together and separately for different station/area types and regions. Increasing and decreasing trends are presented separately.***

| group of the stations | N | decreasing trend | | | | increasing trend | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | signif. | | non signif. | | signif. | | non signif. | |
| | | N | % | N | % | N | % | N | % |
| all stations | 1269 | 128 | 10.1 | 494 | 38.9 | 107 | 8.4 | 540 | 42.6 |
| rural background | 214 | 19 | 8.9 | 104 | 48.6 | 10 | 4.7 | 81 | 37.9 |
| suburban background | 235 | 24 | 10.2 | 95 | 40.4 | 12 | 5.1 | 104 | 44.3 |
| urban background | 351 | 35 | 10.0 | 139 | 39.6 | 22 | 6.3 | 155 | 44.2 |
| traffic (all types) | 323 | 30 | 9.3 | 99 | 30.7 | 53 | 16.4 | 141 | 43.7 |
| industrial (all types) | 144 | 20 | 13.9 | 57 | 39.6 | 10 | 6.9 | 57 | 39.6 |
| Northern Europe | 38 | 3 | 7.9 | 15 | 39.5 | 5 | 13.2 | 15 | 39.5 |
| North-western E. | 382 | 30 | 7.9 | 166 | 43.5 | 24 | 6.3 | 162 | 42.4 |
| Central + Eastern E. | 530 | 44 | 8.3 | 201 | 37.9 | 39 | 7.4 | 246 | 46.4 |
| Southern Europe | 319 | 51 | 16.0 | 112 | 35.1 | 39 | 12.2 | 117 | 36.7 |
| rural backgr. - N | 10 | 1 | 10.0 | 5 | 50.0 | 0 | 0.0 | 4 | 40.0 |
| rural backgr. - NW | 54 | 0 | 0.0 | 34 | 63.0 | 1 | 1.9 | 19 | 35.2 |
| rural backgr. - CE | 118 | 12 | 10.2 | 54 | 45.8 | 6 | 5.1 | 46 | 39.0 |
| rural backgr. - S | 32 | 6 | 18.8 | 11 | 34.4 | 3 | 9.4 | 12 | 37.5 |
| suburb. backgr. - N | 1 | 0 | 0.0 | 1 | 100.0 | 0 | 0.0 | 0 | 0.0 |
| suburb. backgr. - NW | 96 | 15 | 15.6 | 41 | 42.7 | 2 | 2.1 | 38 | 39.6 |
| suburb. backgr. - CE | 100 | 2 | 2.0 | 39 | 39.0 | 6 | 6.0 | 53 | 53.0 |
| suburb. backgr. - S | 38 | 7 | 18.4 | 14 | 36.8 | 4 | 10.5 | 13 | 34.2 |
| urban backgr. - N | 13 | 2 | 15.4 | 5 | 38.5 | 0 | 0.0 | 6 | 46.2 |
| urban backgr. - NW | 127 | 10 | 7.9 | 55 | 43.3 | 7 | 5.5 | 55 | 43.3 |
| urban backgr. - CE | 138 | 15 | 10.9 | 57 | 41.3 | 4 | 2.9 | 62 | 44.9 |
| urban backgr. - S | 73 | 8 | 11.0 | 22 | 30.1 | 11 | 15.1 | 32 | 43.8 |
| industrial - N | 3 | 0 | 0.0 | 1 | 33.3 | 1 | 33.3 | 1 | 33.3 |
| industrial - NW | 41 | 3 | 7.3 | 16 | 39.0 | 4 | 9.8 | 18 | 43.9 |
| industrial - CE | 33 | 6 | 18.2 | 9 | 27.3 | 1 | 3.0 | 17 | 51.5 |
| industrial - S | 67 | 11 | 16.4 | 31 | 46.3 | 4 | 6.0 | 21 | 31.3 |
| traffic - N | 11 | 0 | 0.0 | 3 | 27.3 | 4 | 36.4 | 4 | 36.4 |
| traffic - NW | 63 | 2 | 3.2 | 20 | 31.7 | 10 | 15.9 | 31 | 49.2 |
| traffic - CE | 141 | 9 | 6.4 | 42 | 29.8 | 22 | 15.6 | 68 | 48.2 |
| traffic - S | 108 | 19 | 17.6 | 34 | 31.5 | 17 | 15.7 | 38 | 35.2 |

The same distributions as in Table 15 are presented also as histograms in Figure 23.

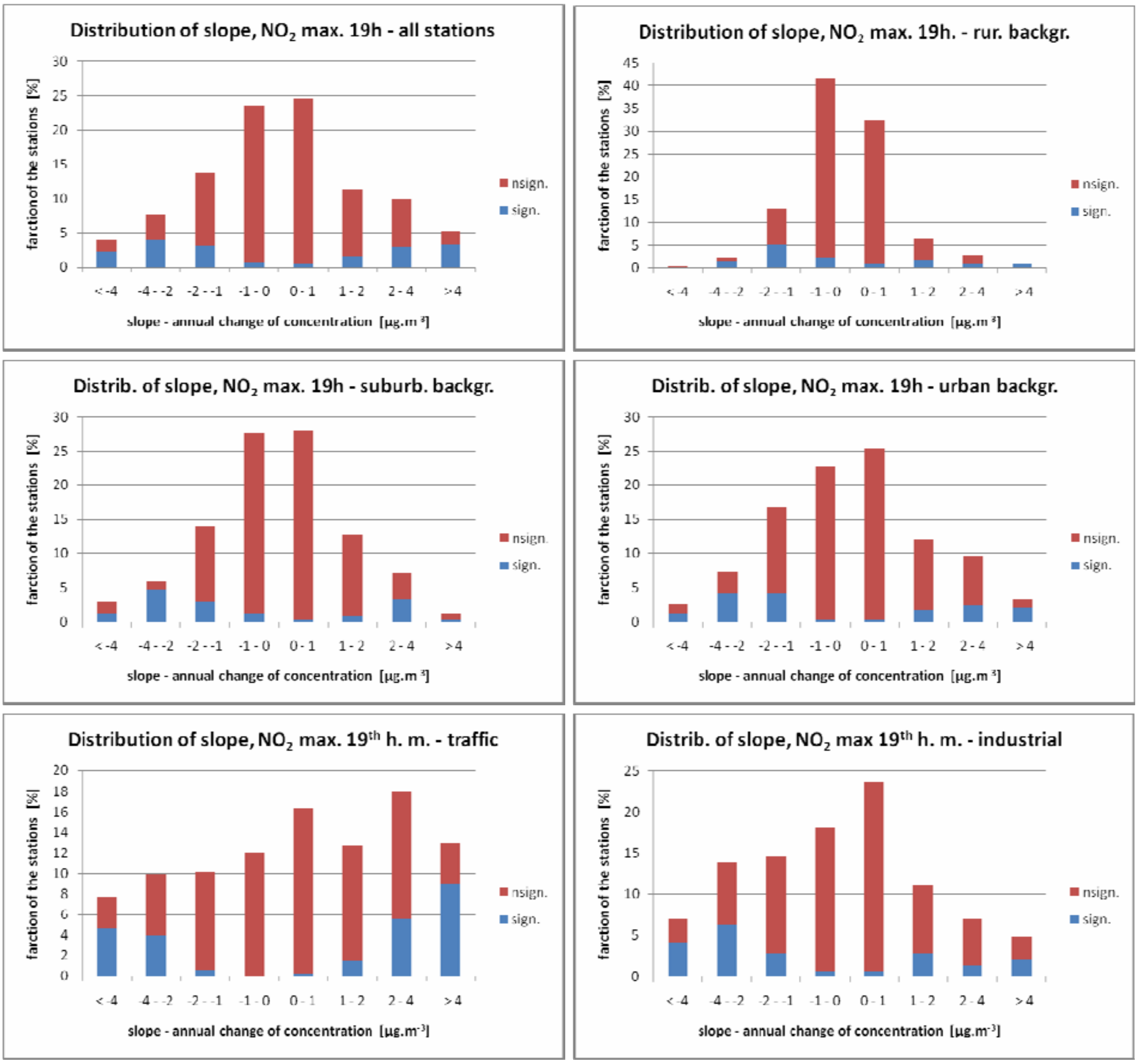


**Figure 23:** ***Distribution of the Sen's slope at the individual stations for $NO_2$ indicator $19^{th}$ maximum hourly mean for the years 1999-2008, for all stations together and separately for different station/area types. The percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. Significant trend is in blue, non-significant trend is in red. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$***

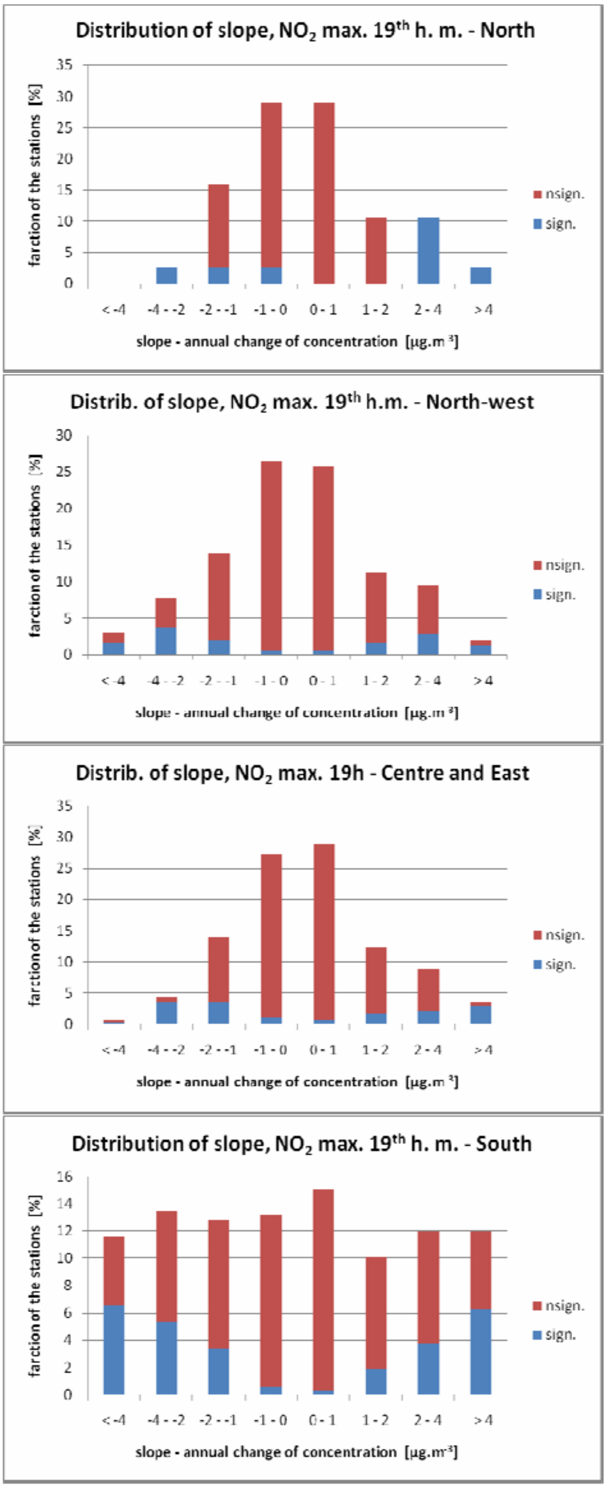


**Figure 23 cont.:** ***Distribution of the Sen's slope at the individual stations for $NO_2$ indicator 19$^{th}$ maximum hourly mean for the years 1999-2008, for different regions. The percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. Significant trend is in blue, non-significant trend is in red. The Sen's slope represents the annual change of concentration, in µg.m$^{-3}$***

In Figure 24 the similar histograms for different combinations of stations/area types and regions are shown.


**Figure 24:** ***Distribution of the Sen's slope at the individual stations for $NO_2$ indicator $19^{th}$ maximum hourly mean, for the years 1999-2008, separately for different station/area types and regions. (North region is not shown, due to small number of stations.) The percentage of the stations of the relevant group in different interval of and the indication of significance of the monotonic trend (based on Mann-Kendall's test, for significance level 0.10) are presented. Significant trend is in blue, non-significant trend is in red.***

# Annex C - Average annual change in $NO_x$ between 1999 and 2008

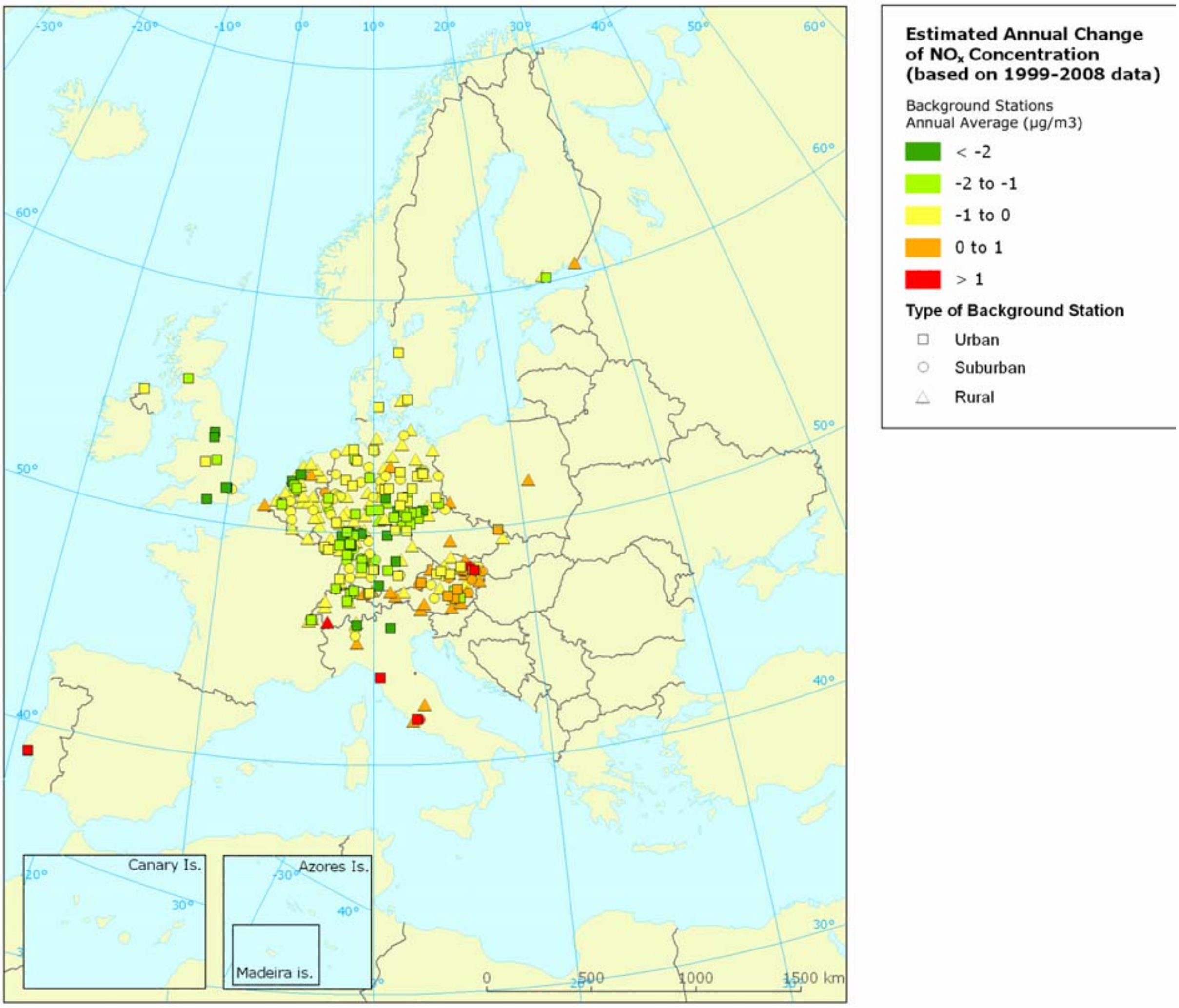


**Figure 25: Average annual change in $NO_x$ annual mean concentrations at background stations between 1999 and 2008.**

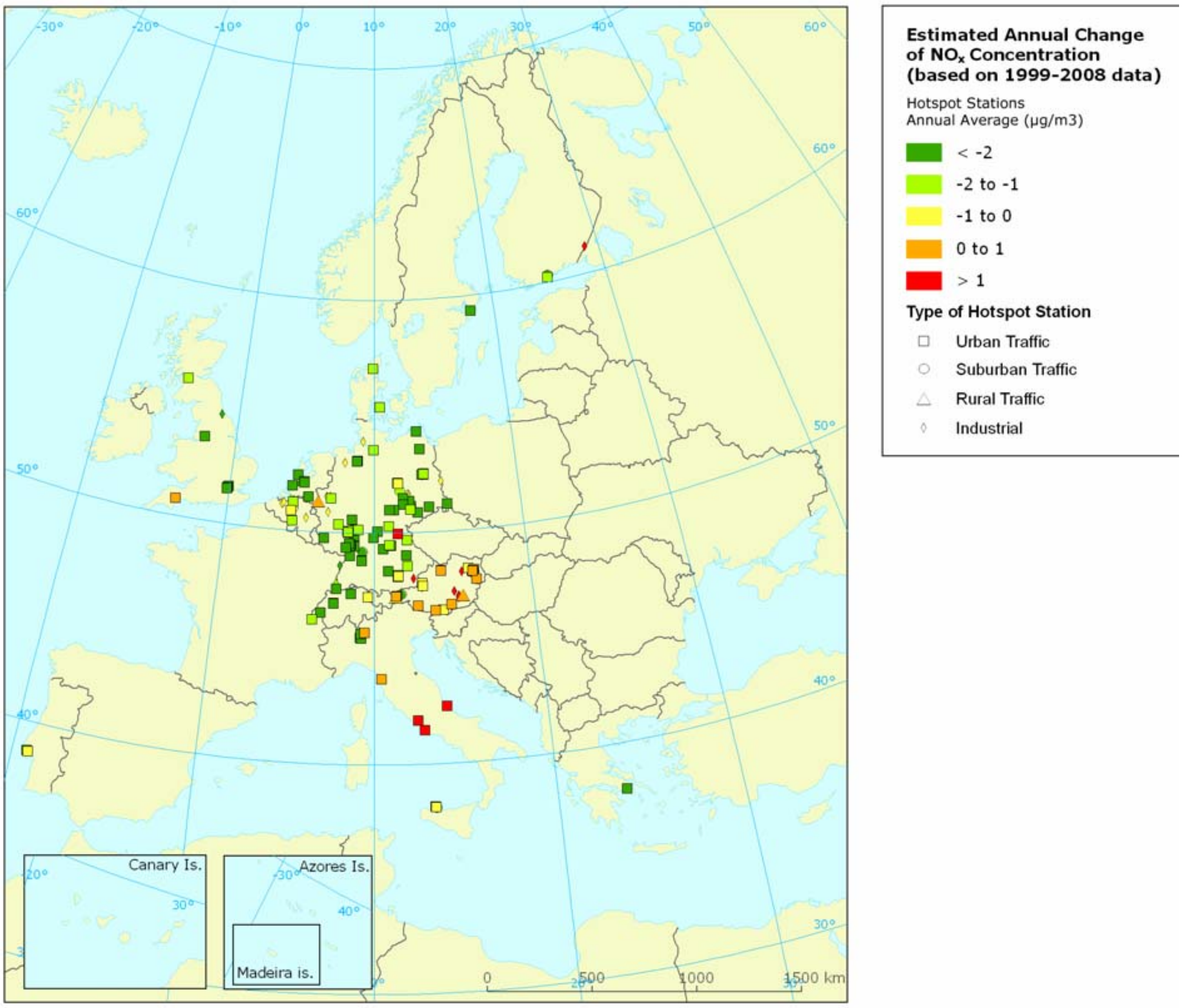


**Figure 26: Average annual change in $NO_x$ annual mean concentrations at traffic and industrial stations between 1999 and 2008.**

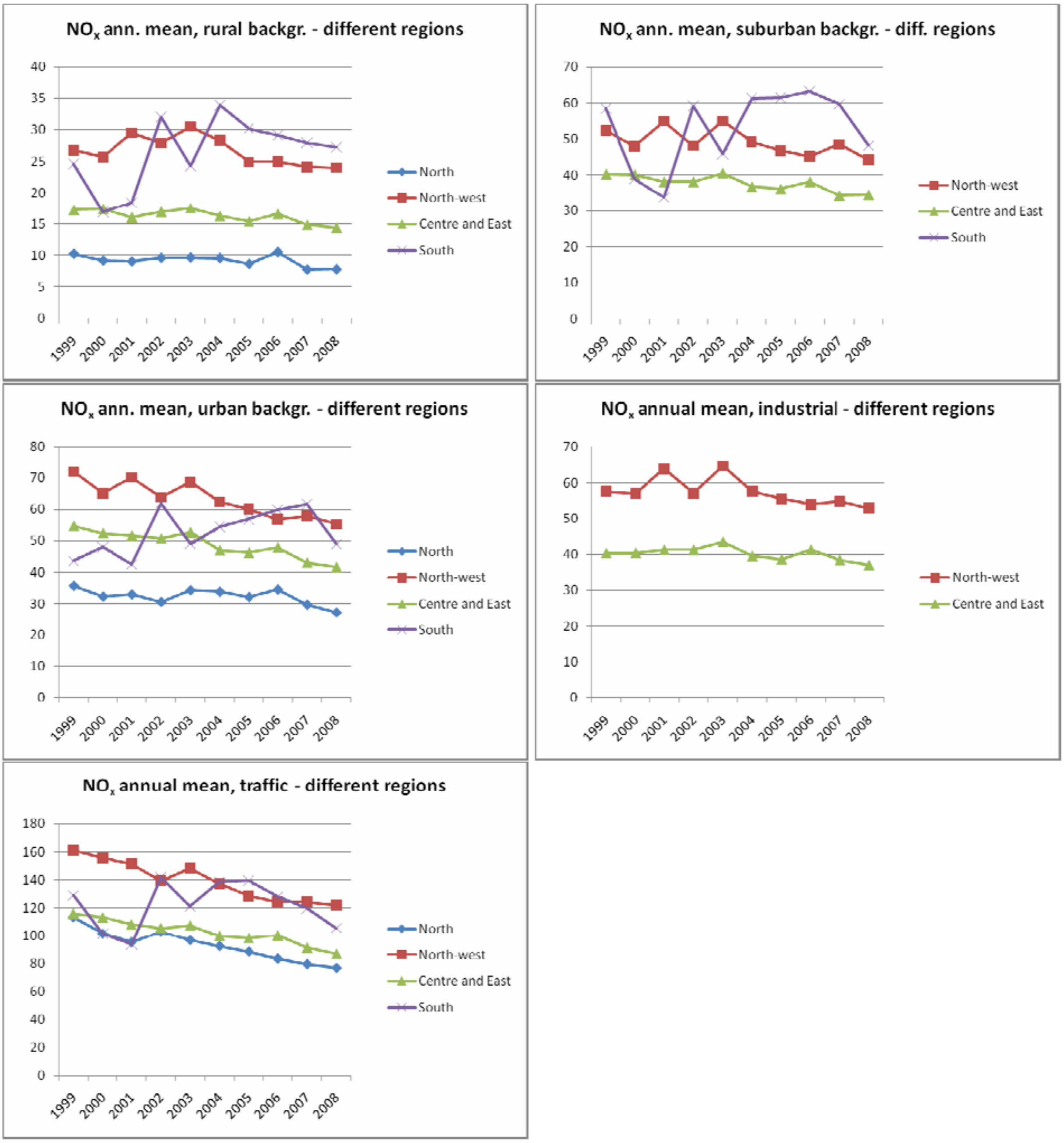


**Figure 27: The average concentration values for different combinations of station/area types and regions, for $NO_x$ annual mean in µg.m$^{-3}$, for the years 1999 – 2008.**

# Annex D - Average $NO_2$ /$NO_x$ ratio trends between 1999 and 2008

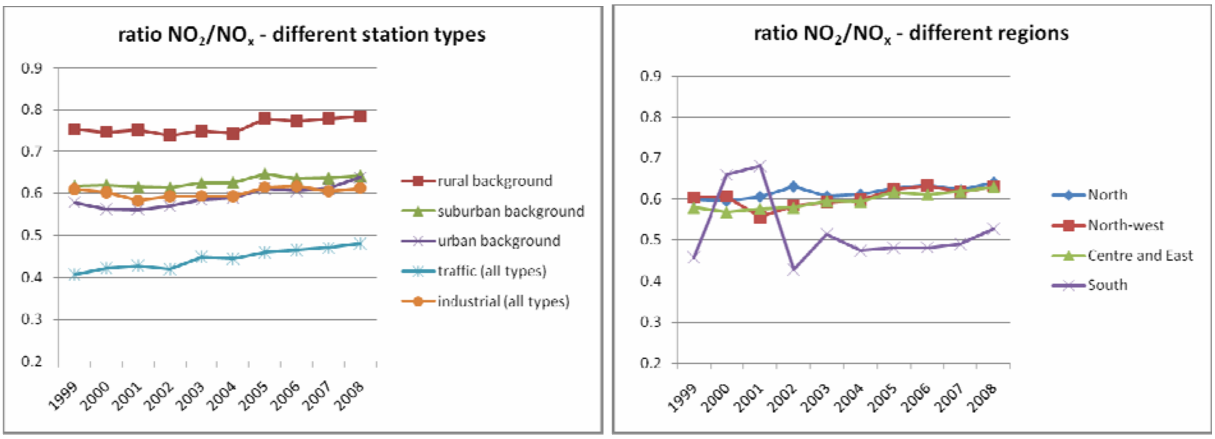


**Figure 28:** ***Average ratio $NO_2$ /$NO_x$ values for different station/area types (left) and regions (right), for the years 1999 – 2008.***

In the Figure 29 the same trends for different combinations of stations/area types and regions can be seen.

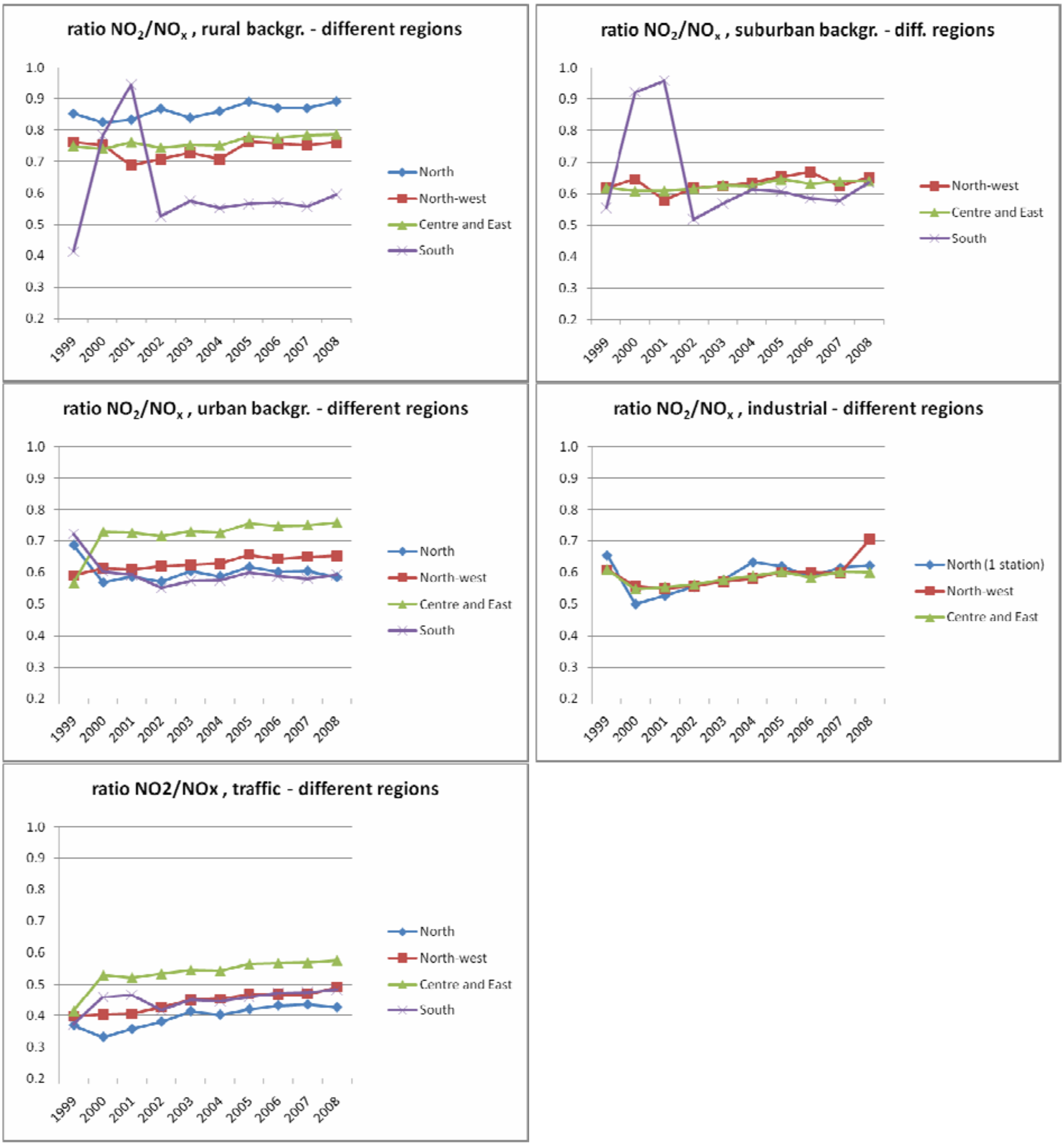


**Figure 29:** ***Average ratio $NO_2$ /$NO_x$ values for different combinations of station/area types and regions, for the years 1999 – 2008.***