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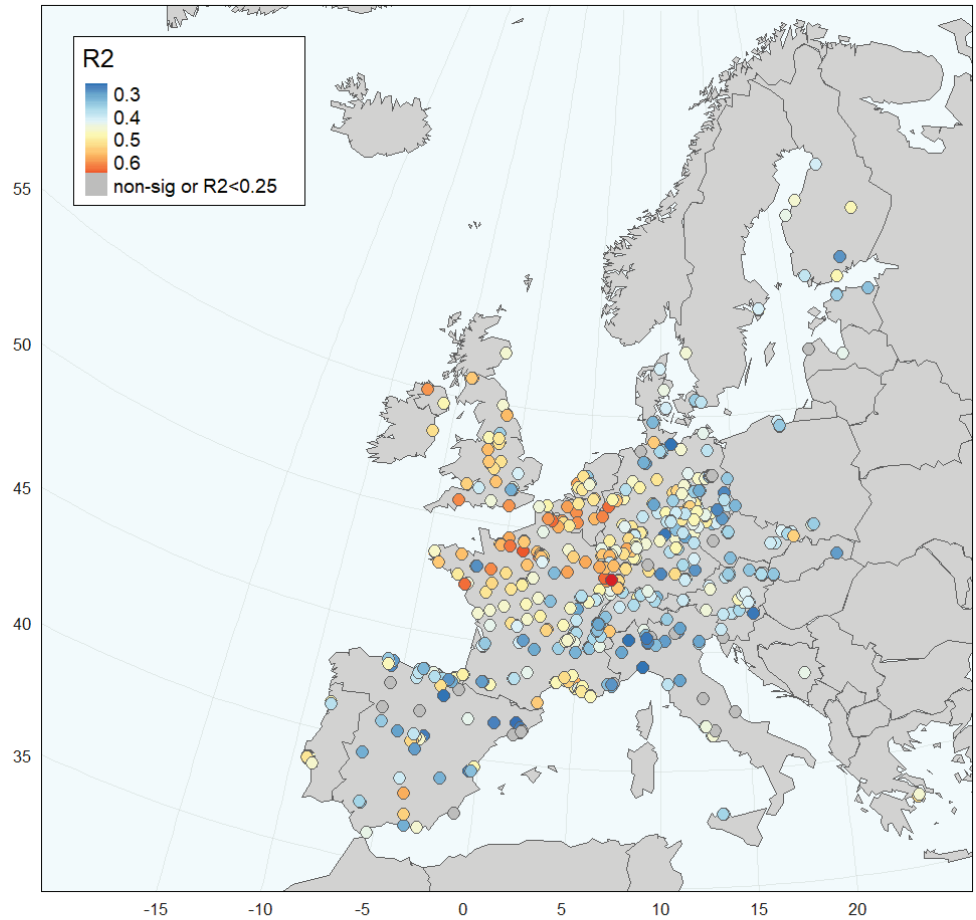
Trends in measured NO_2 and PM

Discounting the effect of meteorology

December 2018



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Cover photo – *r^2 for the GAM predicted daily NO_2 concentrations during winter months for the period 2000-2010 for EEA's Airbase urban stations*

More information on the European Union is available on the Internet (http://europa.eu).

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1 Executive summary

This report documents a study on long-term trends in observed atmospheric levels of NO_2, PM_{10} and $PM_{2.5}$ based on data from the European Environmental Agency (EEA) Airbase v8 (EEA, 2018). The main aim is to evaluate to what extent the observed time series could be simulated as a function of various local meteorological data plus a time-trend by a Generalized Additive Model (GAM). The GAM could be regarded an advanced multiple regression model. If successful, such a model could be used for several purposes; to estimate the long-term trends in NO_2 and PM when the effect of the inter-annual variations in meteorology is removed, and secondly, to "explain" the concentration levels in one specific year in terms of meteorological anomalies and long-term trends. The GAM method was based on a methodology developed during a similar project in 2017 looking at the links between surface ozone and meteorology.

The input to the study consisted of gridded model meteorological data provided through the EURODELTA Trends project (Colette et al., 2017) for the 1990-2010 period as well as measured data on NO_2, PM_{10} and $PM_{2.5}$ extracted from Airbase v8. The measurement data was given for urban, suburban and rural stations, respectively. The analysis was split into two time periods, 1990-2000 and 2000-2010 since the number of stations differ substantially for these periods and since there is reason to believe that the trends differ considerably between these two periods.

The study was focused on the 4-months winter period (Nov-Feb) since it was important to assure a period of the year with consistent and homogeneous relationships between the input explanatory data (local meteorology) and the levels of NO_2 and PM. For NO_2, this period will likely cover the season with the highest concentration levels whereas for PM high levels could be expected outside this period due to processes such as secondary formation, transport of Saharan dust and sea spray.

When measured by the R2 statistic, the GAM method performed best for NO_2 in Belgium, the Netherlands, NW Germany and the UK. Significantly poorer performance was found for Austria and areas in the south. For PM_{10} there were less clear geographical patterns in the GAM performance.

Based on a comparison between the meteorologically adjusted trends and plain linear regression, our results indicate that for the 1990-2000 period meteorology caused an increase in NO_2 concentrations that counteracted the effect of reduced emissions. For the period 2000-2010 we find that meteorology lead to reduced NO_2 levels in the northwest and a slight increase in the south.

The amount of observational data is much less for PM than for NO_2. For the 1990-2000 period the number of sites with sufficient length of time series is too small to apply the GAM method on a European scale. For the 2000-2010 period, we find that the general performance of the GAM method is poorer for PM_{10} than for NO_2. With respect to the link between PM_{10} and temperature, the results indicate a marked geographical pattern with a negative relationship in central Europe and a positive relationship in Spain, southern France and northern Italy.

For PM_{10} during 2000-2010, the vast majority of the estimated trends are found to be negative. The difference between the GAM trend and the plain linear regression, indicates that meteorology lead to increased PM_{10} levels in the southern and central parts and decreased levels in the north.

For $PM_{2.5}$ it turned out that the amount of data in the entire period 1990-2010 was too small to use the GAM method in a meaningful way on a European scale. Only a few sites had sufficient time series and thus more recent data are required.

2 Introduction

Long-term air quality trends have been the topic of various ETC's technical reports the last years (Colette et al., 2015; Solberg et al., 2015; Colette et al., 2016; Solberg et al., 2018). This work has been linked to ongoing activity within EMEP TFMM and EURODELTA Trends (Colette et al., 2017), named EDT hereafter. More specifically, the role of interannual variations in meteorology versus the effect of changes in anthropogenic emissions for the trends has been studied in detail. A variety of methods for isolating these two effects on the long-term trends of air pollutants have been proposed in the scientific literature, commonly called "meteorological adjustment" of the trends.

The main aim of the work presented here is to evaluate whether a statistical method applied by the US-EPA to adjust for the effect of interannual variations in meteorology on surface ozone trends and applied last year on European ozone data (Solberg et al., 2018) could be applied to European NO_2 and PM data. The basic method was published by Camalier et al. (2007) and results (maps and time series) are presented at EPA's web page on an annual basis: https://www.epa.gov/air-trends/trends-ozone-adjusted-weather-conditions.

In principle this and similar methods could be used to adjust for the effect of interannual variations in meteorology both with respect to the long-term trends as well as for the evaluation of the air concentrations in a single year.

3 Methodology

The methodology in this work was based on the methodology developed in the 2017 project on ozone trends and meteorology (Solberg et al., 2018) and the reader is referred to that report for all details. The basic method is a so-called GAM, i.e. a generalized additive model, which is a statistical procedure that finds the optimum solution to an equation linking a number of input explanatory variables to an output function:

$$G(\mu_i) = \beta_0 + \sum_{j=1}^{n} \beta_j \left(x_{ij}\right) + \varepsilon_i. \qquad (1)$$

In our application in the present work, μ_i denotes the measured NO_2 or PM concentrations given as daily mean at day number *i* at one monitoring station. The $x_{i,j}$ variables denote the input explanatory variables like temperature, wind speed, etc. for $j = 1, \ldots, n$ number of variables at day number *i*. Both PM_{10} and $PM_{2.5}$ data were included in the analyses. It turned out, however, that the amount of long-term monitoring data on $PM_{2.5}$ was too little to provide a detailed study.

The GAM can be seen as an extension of a multiple linear regression (MLR) in which the β_j coefficients are smooth functions and not constants as in a MLR, and furthermore that the dependent variable μ_i is replaced by a link function, $G(\mu_i)$.

The choice of settings and parameters in the present study (Table 1) was closely related to the settings and parameters in the ozone trend study (Solberg et al., 2018) with some exceptions:

1. A log function was used for the link function for NO_2 and PM whereas a unity function was used in the ozone trend study. Whereas O_3 has a smaller range of concentrations nearly following a Gaussian distribution, NO_2 and PM could range over several orders of magnitudes and has a distribution closer to a Gamma distribution.

2. The weekday was added as an extra explanatory variable (i.e. Monday = 1, Tuesday = 2, etc.). This was done to reflect that NO_2 and part of the PM are emitted species (in contrast to O_3 being a secondary species) that are strongly influenced by road traffic and general working day effects.
3. We used the daily mean temperature in this study as opposed to the daily maximum temperature in the ozone trend study. The reason for this is that whereas ozone is linked to the photochemistry, NO_2 and PM are mostly determined by the vertical stability and dynamics.
4. Lastly, the focus of the present study was on the winter period (as opposed to the summer season for ozone), i.e. the four months period November – February.

For the other explanatory variables the same set as in the ozone study (Solberg et al., 2018) was used, i.e. relative humidity, global radiation, wind speed, height of the planetary boundary layer, day number in season and time in fraction of years (Table 1).

The choice of looking only at the winter months needs a certain comment. The use of a GAM relies on an overall assumption of homogeneity, i.e. that the links between the explanatory variables and the output are fairly homogeneous within the time period considered. At least for a component like PM which is of both primary and secondary origin one would expect that certain dependencies change between winter and summer. In winter one would expect a negative link between temperature and PM concentration reflecting situations with stable inversions, whereas in summer there may be a positive link due to secondary PM formation. If one included both seasons in the GAM, these signals would cancel each other and the response functions would be blurred and loose statistical significance. The choice of looking only at winter in the present study was based on the fact that it's the time of year with the highest levels of NO_2. For PM also spring and summer would be of interest due to the influence of biogenic and secondary formation, though. Ideally, the GAM could be applied for separate seasons for each of the individual species, but this was not done in the present study.

Table 1. List of explanatory variables used in the GAM (Eq. (1)) for NO_2 and PM in this study. The short names refer to the legends used in the map plots shown below (Chapter 6).

	Associated explanatory variable	**Short name in the plot legends**	**Function type**
x_1	Daily mean temperature	tem2	Smooth
x_2	Daily mean relative humidity	sreh	Smooth
x_3	Daily mean global radiation	swrd	Smooth
x_4	Daily mean 10 m wind speed	w10m	Smooth
x_5	Daily mean PBL height	hght	Smooth
x_6	Week day number	dayofweek	Smooth
X_7	Day number in season	dayofseason	Smooth
X_8	Continuous time in fraction of years (0.0 = 1 Jan at start of period). This is the trend term.	years	Linear

3.1 Use of GAM and openair model software

As in the ozone trend study, the GAM model (Eq. (1)) was fitted using the GAM library `mgcv` (Wood, 2017) in the statistical modelling system R (R Core Team, 2018) for each station and for each period 1990-2000 and 2000-2010 separately. We used all data in each period in order to estimate the long-term trend coefficient β_8 in Eq. (1) at each station. This was the basis for the estimation of the trends and the meteorology adjustments.

Through the GAM optimisation we calculated the β functions and their significance levels for each station/period as well as various measures of the GAM model evaluation performance such as RMSE, R^2, etc. For this latter part we used the `openair` library (Carslaw and Ropkins, 2012; Carslaw, 2015). As part of the output we also calculated the GAM fit, i.e. the predicted log (NO_2) or log (PM) each day (the log of the daily mean values) as well as the slope of the meteorologically adjusted trend over the period (β_8 in Eq. (1)). The results are presented in plots and tables below.

As discussed in the ozone trend study we applied smooth β functions for all the explanatory variables except the trend term ($\beta_8(x_{i,8})$) in Table 1 for which we assumed a linear relationship, i.e. that β_8 is a constant. The smoothness of the β functions is predefined and set as input to the GAM routine by specifying the "degrees of freedom" for each of these functions. These settings are a matter of subjectivity and knowledge of the physical processes. Using too many degrees of freedom for a variable implies a risk of overfitting meaning that the functions are tied too closely to every explanatory data point and that the value of the GAM as a prediction tool is reduced. On the other hand, setting too few degrees of freedom implies that the flexibility of the GAM is reduced. The experience from the ozone trend study was that allowing too many degrees of freedom in the trend term produced unphysical results and that parts of the error term (ε_i in Eq. (1)) was incorporated into the trend term.

In order to see how well the GAM model performed in each separate year we made additional calculations where we excluded the data for that year, i.e. the "target year", and fitted the GAM model (Eq. (1)) to the rest of the data (the left out years). This means that e.g. when fitting the GAM model to predict the daily concentration levels in 2008, we skipped the input data for 2008, but used the data for the other years in the 2000-2010 period to calculate the β coefficients. Then, these β coefficients were used together with the meteorological data for 2008 to predict the daily NO_2 and PM concentration levels in 2008. It should be noted that these β coefficients would differ slightly from the β coefficients used when all 11 years were included.

4 Input data

4.1 Surface monitoring data

The study was based on EEA's Airbase data for the period 1990-2010. RIVM assisted in data extraction and provided a subset of Airbase containing hourly measurement data on NO_2, PM_{10} and $PM_{2.5}$ as well as daily measurement data for PM_{10} and $PM_{2.5}$ for the whole period 1990-2010 together with information on site locations and monitoring history. The latter information was needed in cases with overlapping data (e.g. concurrent daily and hourly monitoring at a site), parallel sampling (several monitors at the same site), etc. Based on the concentration values as well as the monitoring history and recommendations provided by RIVM one time series of daily mean data for 1990-2000 and 2000-2010, respectively, was constructed for every station with sufficient data. We used a requirement of at least 75 % valid data capture in at least 75 % of the years in the period based on the season actually used in the GAM modelling. For every year *y* this was defined as the period November year *y-1* to February year *y*. The number of stations fulfilling this criteria for each species, time period and station type (urban/suburban/rural) is given in Table 2.

Table 2. Number of monitoring stations for each species and time periods with sufficient data for use in the GAM model (at least 75 % data capture). Only EEA's Airbase stations were used in this work.

		Number of sites 1990-2000	Number of sites 2000-2010
Urban	NO_2	182	642
	PM_{10}	7	338
	$PM_{2.5}$	0	6
Suburban	NO_2	96	288
	PM_{10}	1	135
	$PM_{2.5}$	0	1
Rural	NO_2	75	210
	PM_{10}	4	91
	$PM_{2.5}$	0	7

As seen from Table 2, the number of stations with data varies strongly with time period and species. In general there are less PM data than NO_2 data, partly because the history of PM monitoring is shorter.

4.2 Meteorological data

Meteorological data were extracted in the same way as for the ozone trend study (Solberg et al., 2018) from the data set used in the EDT project (Stegehuis et al., 2015) which covers the geographical domain from 30°-70° N and 25°W-45°E in a latitude-longitude grid with a grid spacing of 0.25° latitude and 0.40°, corresponding approximately to 25 x 25 km^2. Details of the set-up and design of the EDT project is given by Colette et al. (2017).

Based on the gridded fields of hourly meteorological data we calculated annual time series containing daily mean values of T, RH, global radiation, 10 m wind speed and PBL height for every monitoring station for 1990-2010 as indicated by Table 1.

The time series of daily meteorological data were then paired with the time series of the daily mean values of NO_2 and PM by selecting the model grid square containing the monitoring site. No interpolation of neighbouring grid cells was applied to the model data.

5 Description of output

The GAM was applied to each of the station types, species and time periods in Table 2 separately, and the results are presented in individual subchapters below. In contrast, the original work by Camalier et al. (2007) was focussed on urban agglomerates in the US and was thus merging data within each urban area.
With eight explanatory variables, two time periods, three types of stations and three species combined with many hundreds of stations and numerous statistics, the output is overwhelming and in the following we have tried to present the results in a condensed way on geographical maps.

We present three types of output:

- Statistical metrics for evaluating the performance of the GAM shown as coloured dots on maps
- The value of each of the explanatory variables in Eq. (1) as linearized β values as explained below shown as coloured dots on maps

- Daily and annual time series together with various statistical plots for some selected sites

The response curves (the β_j functions in Eq. (1)) are smooth functions as indicated by Table 1 and could therefore not be presented as single values. For mapping these functions, we calculated so-called linearized β-values using a linear fit to the interior of the data ranges, defined by the area between the 25- and 75-percentiles of these data. This is identical to the EPA approach (see Figure 5 in Camalier et al., 2007). This means that we fit a straight line to the middle part of each of the curves in Figure 1. The slopes of these straight lines are thus just indicative of the overall relationship between the explanatory variables and the dependent variable (NO_2 and PM). When the interior range of the response curves are approximately linear, these linearized β values are good representatives of the underlying β functions. For highly non-linear responses, however, the linearized values are less informative.

5.1 Statistical metrics

An overview of the statistical metrics used to evaluate the performance of the statistical method is given in Table 3. The metrics are further explained and defined in the following.

Table 3. Statistical metrics used to evaluate the GAM performance at each station

Abbreviation	Explanation
n	Number of stations
R2	Coefficient of determination (r squared)
RMSE	Root mean square error
MB	Mean bias

Coefficient of determination, R2

The coefficient of determination is the proportion of variance in the observations that can be explained by the model predictions. It is calculated as $\mathrm{R2} = r^2$, where r is the correlation between observed and predicted values.

Root mean squared error, RMSE

The root mean squared error is a commonly used statistic that provides a good overall measure of how close the predicted values are to the observed values. Unit: µg m^{-3}. It is calculated as

$$\mathrm{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}\left\{O_i - P_i\right\}^2},$$

where O_i and P_i are the observed and predicted values respectively for $i = 1,\dots,n$, and *n* is the number of observations.

Mean bias, *MB*

The mean bias provides a good indication of the average of over- or underestimation of predictions as compared with observations. Unit: µg m^{-3}. It is calculated as

$$\mathrm{MB} = \frac{1}{n}\sum_{i=1}^{n}\left\{O_i - P_i\right\},$$

where again O_i and P_i are the observed and predicted values.

5.2 Importance of the explanatory variables

As explained above, we used smooth βj functions for the explanatory variables j=1 ,…, 7 in Eq. (1). An example of these functions for NO_2 at the French urban station Gerardmer (FR30022) for the period 2000-2010 is given in Figure 1. The curves show the partial responses of log(NO_2) on the y-axis to each of the individual explanatory variables on the x-axis based on daily data in the winter season (November-February). The lines in the diagrams mark the GAM method's best fit smooth functions, i.e. the βj functions in Eq. (1). The shaded areas represent uncertainty regions, i.e. ±2 standard errors confidence regions, for the underlying fitted smooth curves. Note that these plots are normalised, i.e. having an overall mean value of zero for the partial derivatives for each variable.

Furthermore, the curves shown in Figure 1 represent the normalised perturbations when assuming mean values for all the other variables. The down-sloping curves for wind speed (w10m) in particular as well as for temperature (tem2) and mixing height (hght) reflects that the NO_2 levels tends to be lower when wind speed, temperature and mixing height is increased at this site. The up-sloping curve for day of season indicates that at this site, the NO_2 concentration tends to increase during winter provided that all the other variables are at mean levels.

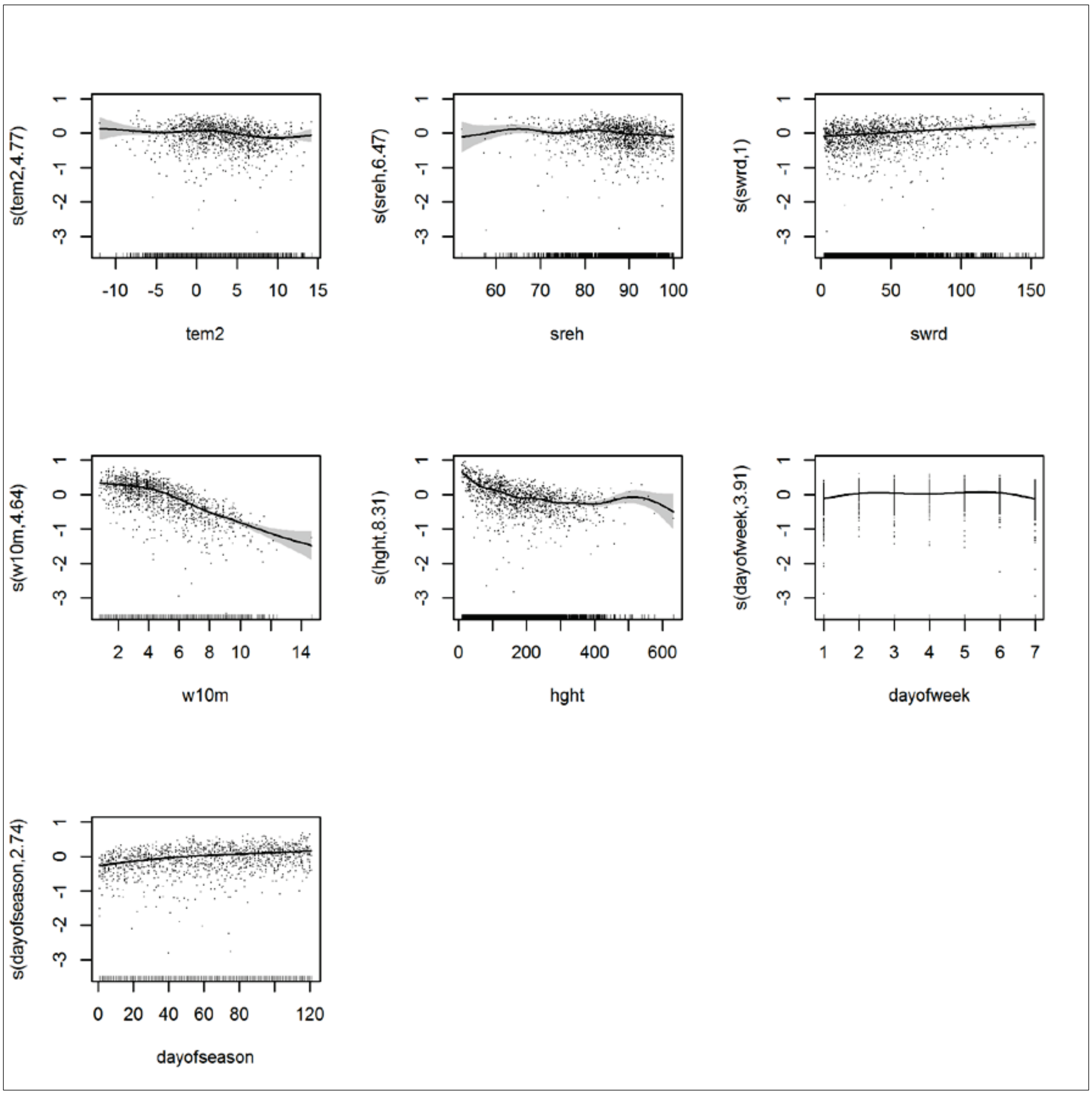

Figure 1. Response curves for FR30022 (Gerardmer) for 2000-2010 showing scatter plots of the daily values of the normalized log(NO_2) on the y-axis vs each of the explanatory variables on the x-axis. The normalization implies that the mean = 0. The black curves show the best fit smooth functions (the beta functions in Eq. (1)), whereas the shaded grey areas indicates ±2 standard errors confidence regions.

6 Results

In the following the output results as outlined above is presented separately for each species (NO_2, PM_{10} and $PM_{2.5}$), each time period (1990-2000 and 2000-2010) and each station type (urban, suburban and rural) in individual sub-chapters. Various statistical metrics were calculated to evaluate the method at each site, and based on these results it was decided to use the R2 statistic based on daily data to classify the performance. For sites where the method produced an R2 < 0.25, we regarded the method as not applicable, and for these sites all results are only shown as grey markers (corresponding to "non-significant") in the following. It should be said, though, that the 0.25 criteria is a subjective choice based on evaluations of the observed and predicted time series. Examples of time series and model fit for various levels of R2 are shown in Ch. 7 below to illustrate how good the GAM predictions are.

6.1 NO_2

Map plots of the results for NO_2 for the individual time periods and station types are shown in the sub-chapters below. Table 4 gives a summary of the overall performance of the GAM as indicated by the fraction of sites with an R2 > 0.25 as well as certain mean performance statistics (R2, RMS and MB) based on the accepted sites only.

Table 4. Summary statistics for the GAM performance for NO_2 showing the total number of sites, the percentage of "accepted sites" defined as R2 > 0.25 and the mean of R2, RMSE and MB for the accepted sites.

		Total # sites	% accepted	R2	RMSE	MB
1990-2000	Urban	182	68	0.359	13.69	-0.22
	Suburban	96	81	0.376	11.75	-0.16
	Rural	75	65	0.380	10.55	-0.12
2000-2010	Urban	642	81	0.397	11.35	0.05
	Suburban	288	74	0.408	10.22	0.08
	Rural	210	57	0.376	8.40	0.04

Table 4 shows that the highest fraction of sites within the acceptance criteria (R2 > 0.25) is seen for the urban and suburban sites. Furthermore, the mean R2 is higher for these two categories of stations compared to the rural station in the 2000-2010 period. The smallest RMSE and MB is, however, observed at the rural sites, presumably reflecting that the concentration levels are lower at these sites. For the 2000-2010 period it seems that the GAM performance is fairly similar for the urban and suburban stations based on the mean statistics of R2, RMSE and MB. Furthermore, the overall performance is also better in the last decade than the first one. The difference in MB for the two time periods is somewhat unexpected. Since the GAM is centred for each time period, one would expect that MB would be fairly similar. The reason why this is not the case is not clear, but could possibly reflect larger changes during the 1990s compared to the 2000s.

6.1.1 NO_2 in the period 1990-2000

Performance statistics for the GAM method at urban, suburban and rural NO_2 sites for 1990-2000 are shown in Figure 2 - Figure 4, respectively. First of all, the network of sites with sufficient monitoring history in this period is only covering a limited area of the European continent that could be named “central Europe”, namely UK, Belgium, Netherlands, Germany, Czech Republic, Austria and Switzerland. There are no sites in France or the Mediterranean and just a very few in the north (Scandinavia) or the east.

Next, there is a clear pattern that the area with the best R2 score are found in Belgium, northwest Germany and some parts of the UK both for urban, suburban and rural sites. Significantly poorer performance as measured by R2 are seen in Austria and Switzerland. With respect to bias (as measured by RMSE and MB) it is a less clear spatial pattern although the same area as for R2 (Belgium and NW Germany) also shows low RMSE and MB. Low bias is also seen at sites in Switzerland and some sites in Austria.

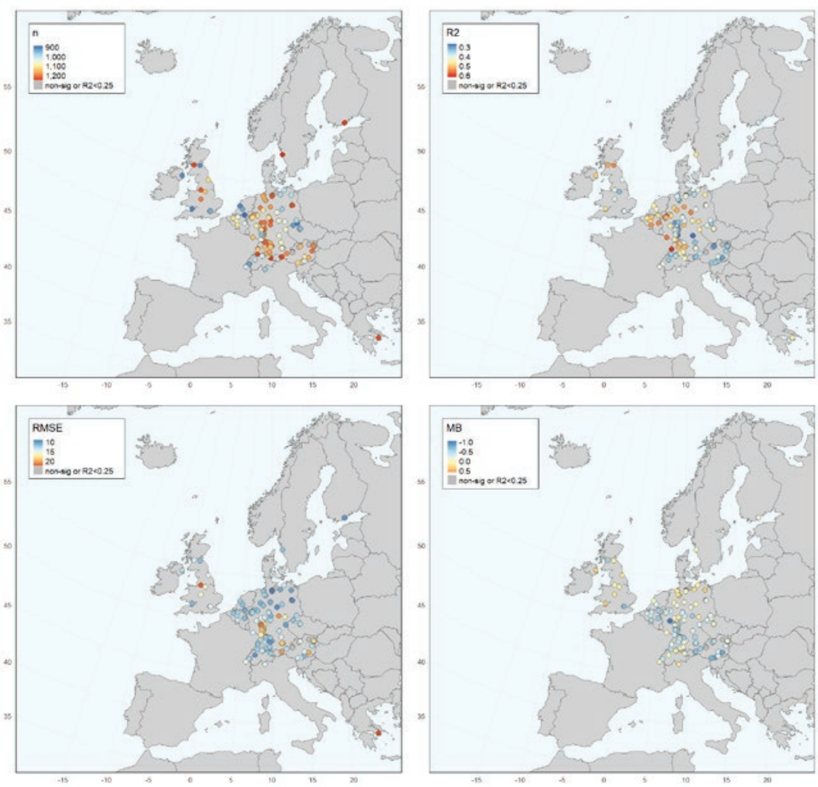

Figure 2. Performance statistics for urban NO_2 data 1990-2000. ‘n’ in the upper left panel shows the total number of days with data during the 11 year’s period used in the GAM model.

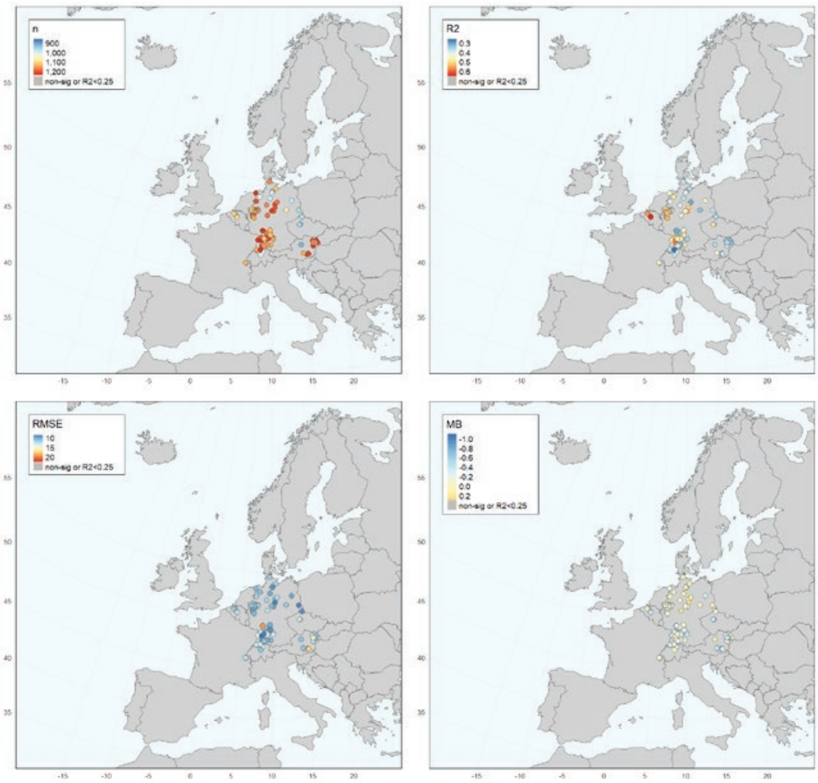

Figure 3. Performance statistics for suburban NO_2 data 1990-2000.

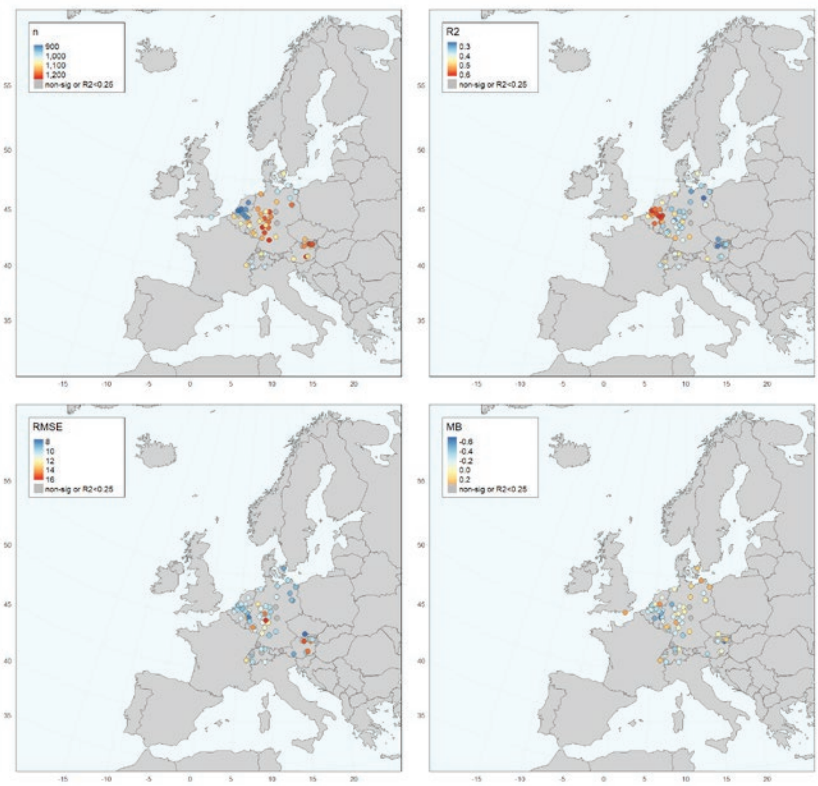

Figure 4. Performance statistics for rural NO_2 data 1990-2000.

The linearized β coefficients as defined above for the explanatory variables in Eq. (1) are shown in Figure 5 - Figure 7 for the urban, suburban and rural NO_2 sites, respectively. Note that these values refer to the log transformed data, i.e. they have the unit [log (µg m^{-3})] except the trend coefficients. The log transformation makes the interpretation of the results more difficult but was necessary due to the distribution of NO_2 levels that was closer to a Gamma distribution than a Gaussian distribution. Note also that the weekday number (a discrete number going from 1 to 7) is not included in these plots since the linearized β coefficient is meaningless for that parameter. The weekday number is included to take into account the weekend effect, whereas the linearized β coefficient would only reflect the relationship between the dependent variable and the middle range (i.e. weekdays between 2 and 5).

The trend coefficients, "beta_years", "beta_linear" and "diff_trend" at the bottom row of the panel are explained in the following way:

- beta_years: This is the percentage change in mean NO_2 concentration during 1990-2000 relative to 1990 based on the GAM and thereby adjusted for the meteorological variability.
- beta_linear: This is the percentage change in the mean concentration over the same period calculated by a plain linear regression without taking into account the meteorological variability
- diff_trend: This is just the difference (beta_years – beta_linear) and thus representing the influence of meteorology for the trend in measured concentrations.

For the urban data (Figure 5) there is a negative relationship between NO_2 level and both wind speed, PBL height and temperature for most sites. This is as expected and reflects that high levels of NO_2 is linked to conditions characterized by little wind and a cold and stable surface layer with small vertical mixing. For short wave radiation and relative humidity, many of the sites show non-significant numbers. It seems, though, that there is a positive relationship between short wave radiation and NO_2 at most sites.

The trend calculated by the GAM and adjusted for the influence of the meteorology (beta_years) shows negative values (i.e. reduced NO_2 levels) for most sites, but with positive values (increased levels) at a number of sites. The plain linear regression ("beta_linear") also shows negative values at most sites and many more non-significant numbers than the meteorologically adjusted trend. The difference between the two quantities shows slightly positive values at all sites. This indicates that during this period the downward trend in NO_2 levels due to changes in emissions and other boundary conditions were actually stronger than the observed trend in NO_2 since meteorology caused an increase in NO_2 counteracting the effect of reduced emissions. This meteorological effect was apparently not very large, though, but amounting to about 5 % relative to the 1990 levels.

For the suburban data (Figure 6) there are less sites than for the urban data, and the results are somewhat more mixed. Note though, the dynamical scaling of the color scale, meaning that the colors differ. Also for the suburban data, there is a clear negative relationship between wind speed and NO_2, and between PBL height and NO_2. With respect to the negative relationship between temperature and NO_2, this seems to be somewhat less marked for the suburban compared to the urban sites. As for the urban sites, the meteorologically adjusted trends from the GAM show more significant downward trends for the suburban sites than the plain linear regression, and the difference between them indicate that the meteorology lead to a slight increase in NO_2 as for the urban sites.

As for the urban and suburban sites, we find a systematic negative relationship between wind speed and PBL height on one side and NO_2 on the other side for the rural sites (Figure 7). The link between temperature and NO_2 is, however, not so clear for the rural sites showing both positive and negative values. The meteorologically adjusted trends from the GAM shows an overweight of negative values (reduced NO_2) although strong increases are seen at some sites. The difference compared to the plain linear regression indicate a positive (increasing) influence from meteorology on NO_2 also for most of the rural sites.

Note also that the results in Figure 5 - Figure 7 indicate a positive relationship between the day number ("day of season") and the NO_2 level. The day number is a numeric variable ranging from 1 at Nov 1 to 120 at the end of February. Thus, the positive relationship indicates that on average the episodes with the highest NO_2 levels tend to occur in the late part of the season. Note though, that the data plotted in Figure 5 - Figure 7 are showing the so-called linearized β coefficients that are based on the interior part of the distribution only as explained above. For relationships between the explanatory variables and NO_2 that are very non-linear and thus varies strongly with the magnitude of the explanatory variable, the linearized coefficients are not well suited and could thus be misleading (see e.g. Fig 5 in Camalier et al., 2007).

Table 5. The percentage of significant trends in NO_2 calculated by the GAM and the linear regression. The percentage of positive and negative significant trends are given as well together with the overall mean trend given in %/decade relative to the start year (1990 or 2000, respectively).

		Total nr of sites	Sites with significant GAM trends (%)	Sites with significant linear trends (%)	Sites with positive GAM trends (%)	Sites with negative GAM trends (%)	Sites with positive linear trends (%)	Sites with negative linear trends (%)	Mean GAM trend (%/dec)	Mean lin. trend (%/dec)
1990-2000	Urban	182	78	57	8	69	1	56	-17	-18
	Suburban	96	87	66	8	79	4	62	-18	-18
	Rural	75	70	50	8	62	5	45	-18	-18
2000-2010	Urban	642	74	36	24	50	12	24	-5	-4
	Suburban	288	72	29	19	52	9	19	-8	-5
	Rural	210	62	33	16	46	12	21	-8	-2

Table 5 summarizes the trend statistics for NO_2. It lists the fraction of significant trends identified by the GAM and the linear regression as well as the fraction of positive and negative significant trends, respectively and the overall mean trend based on all significant trends. Note that the monitoring sites are unevenly distributed and thus in some areas like in Austria, there may be many neighboring sites that are overlapping in the map plots and therefore it could be difficult to judge the fraction of significant sites from these plots directly. As seen from Table 5 nearly all significant trends were found to be negative (downwards) for the 1990-2000 period, independent on the station type.

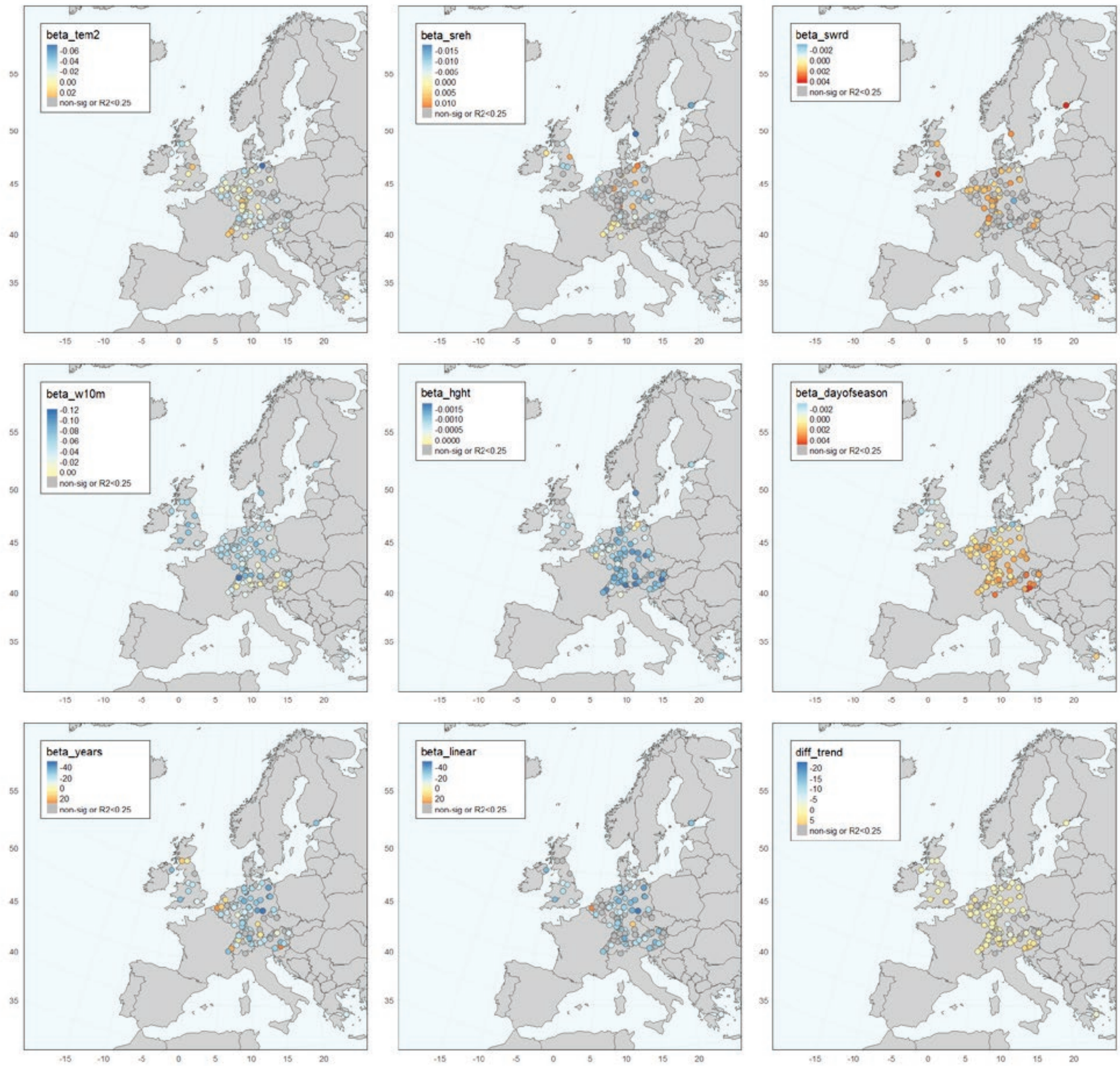

Figure 5. Linearized β coefficients for the individual explanatory variables for the GAM method applied to urban NO_2 sites 1990-2000. Note that the values refer to the log-transformed coefficients. See Table 1 for an explanation of the abbreviated names in the legends. At the bottom row, the coefficient "beta_years" shows the estimated change 1990-2000 as calculated by the GAM (adjusted for meteorology) in percentage relative to 1990 whereas "beta_linear" shows the plain linear trend without meteorological adjustment, also in percentage relative to 1990. "diff_trend" to the right shows the difference (beta_years – beta_linear) which reflects the percentage change 1990-2000 caused by meteorology.

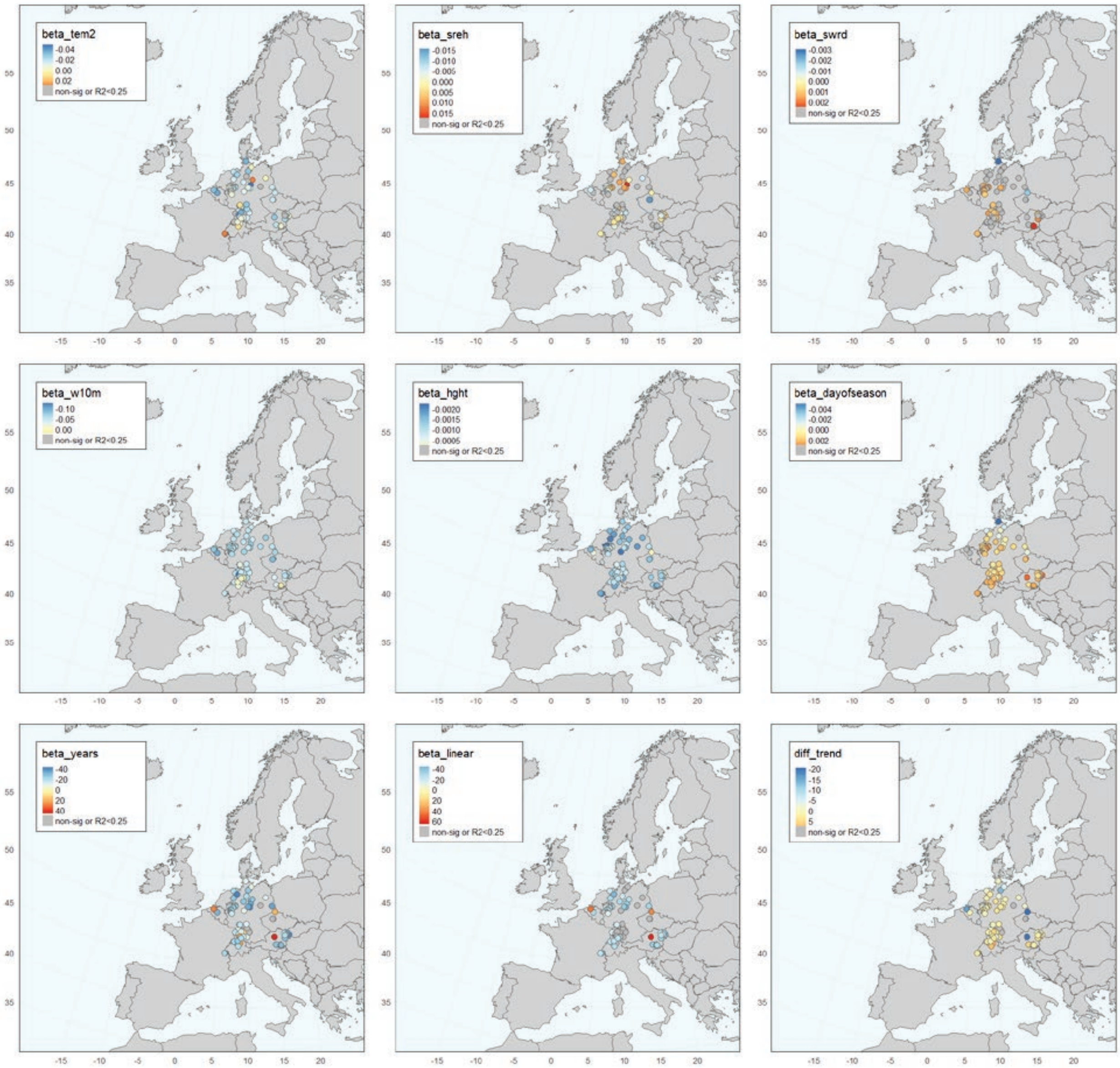

Figure 6. Same as Figure 5 for suburban NO_2 sites.

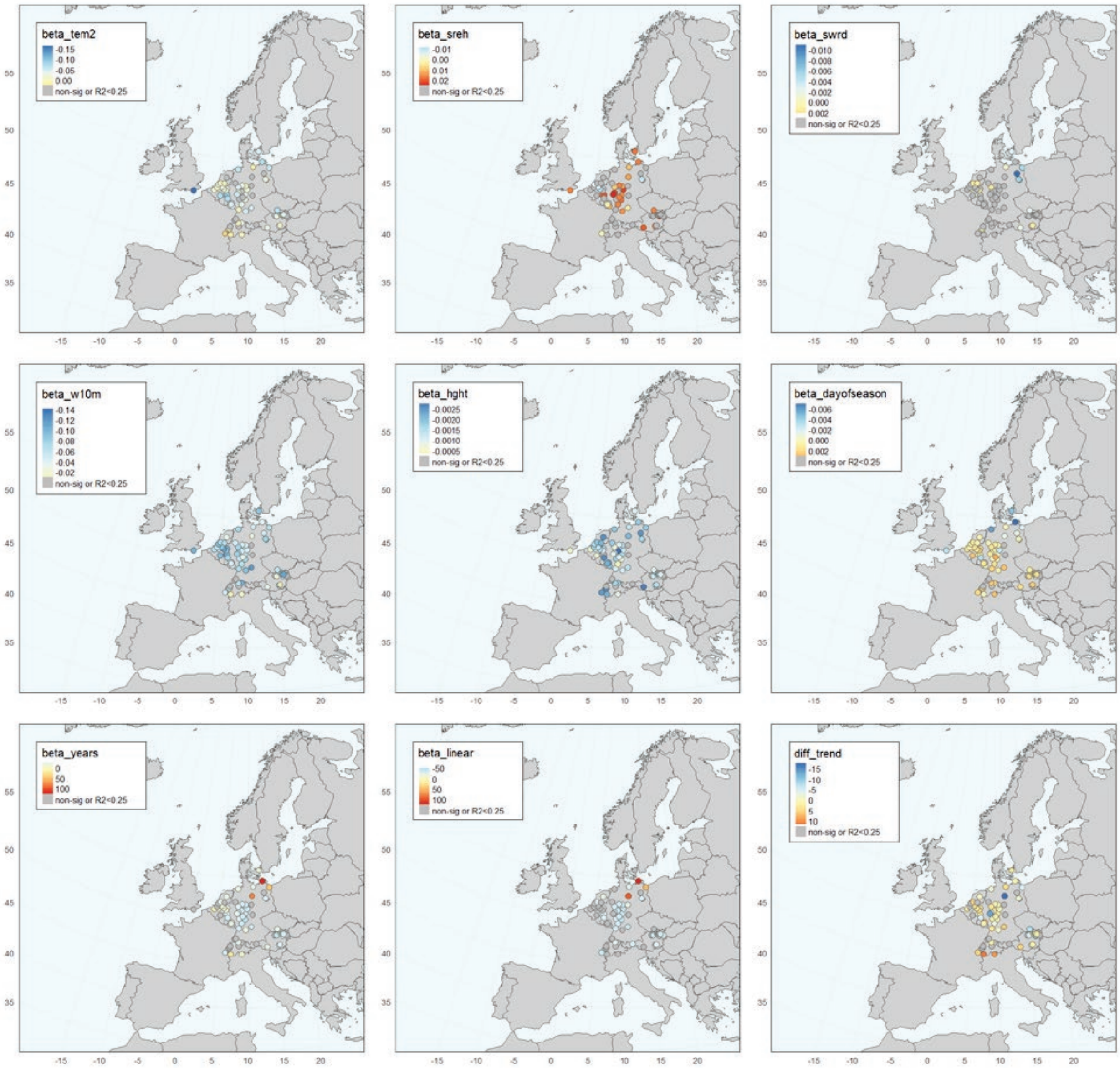

Figure 7. Same as Figure 5 for rural NO_2 sites.

6.1.2 NO_2 in the period 2000-2010

Performance statistics for the GAM method at urban, suburban and rural NO_2 sites for 2000-2010 are shown in Figure 8 - Figure 10, respectively. These plots show a much better geographical coverage than for the 1990-2000 period although Eastern Europe is still poorly covered. Furthermore, as seen also for 1990-2000, there is a clear geographical pattern in the GAM performance with substantially better performance in the UK, Belgium, Netherlands, Northern France and Western Germany compared to the rest of the continent. This is valid for all three types of stations, i.e. urban, suburban and rural. R2 ranges from 0.5-0.6 at sites in the best fit areas compared to around 0.3 in other areas. The poorest performance as measured by R2 is found in N-Italy, E-Germany and Spain. For RMSE and MB there are less clear spatial patterns. A number of strong outliers in RMSE and MB are seen though.

The reason for the systematic geographical differences in the performance for NO_2 is not clear. The area with the highest performance corresponds broadly with the region with the strongest NO_x emissions in Europe and thus, a reason for the regional patterns in performance could be that the GAM method in general performs best for NO_2 near the emission sources. Since the GAM method is based on local relationships and does not take long-range transport into consideration, one could expect this kind of relationship.

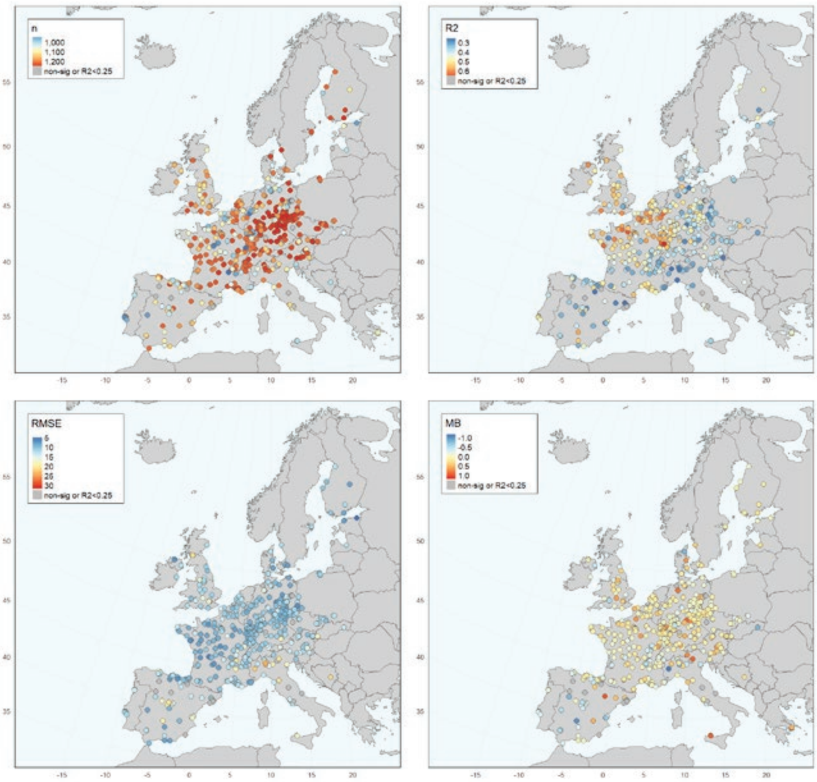

Figure 8. Performance statistics for urban NO_2 data 2000-2010.

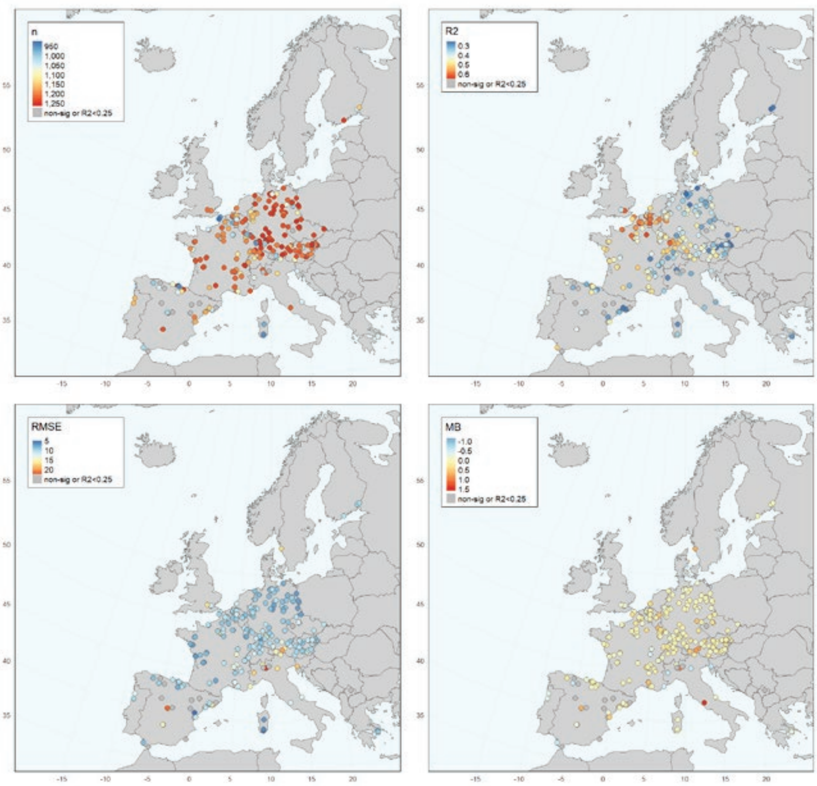

Figure 9. Performance statistics for suburban NO_2 data 2000-2010.

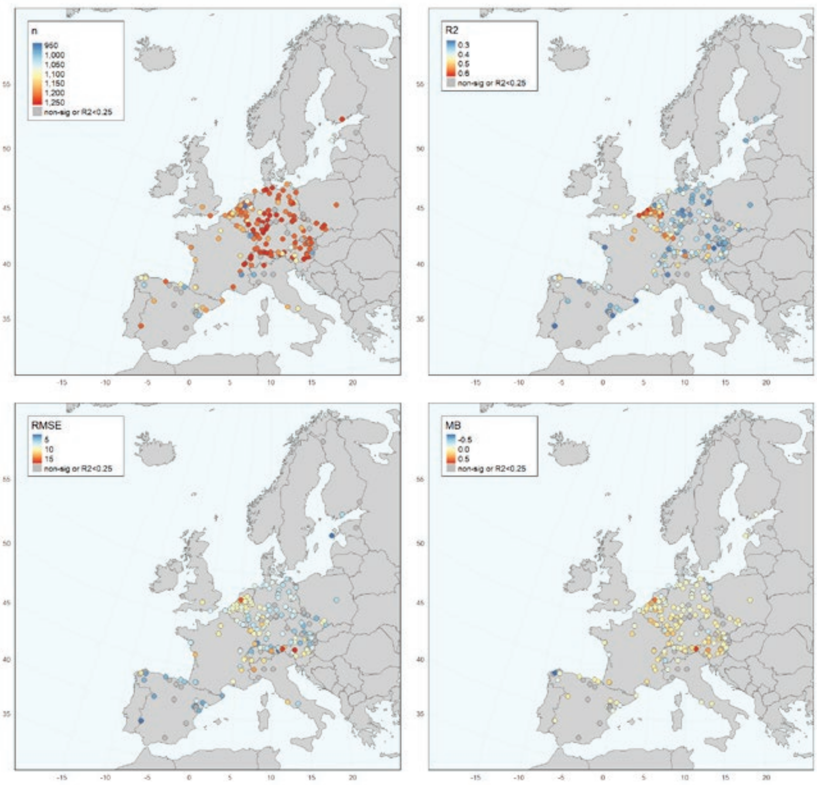

Figure 10. Performance statistics for rural NO_2 data 2000-2010.

The linearized β coefficients for the explanatory variables in Eq. (1) are shown in Figure 11 - Figure 13 for the 2000-2010 urban, suburban and rural NO_2 sites, respectively. As noted above, these values refer to the log transformed data, i.e. they have the unit [log (µg m^{-3})], except for the trend terms as already explained.

As seen from the 1990-2000 period, the GAM method finds a negative relationship between temperature, wind speed and mixing height on one side and NO_2 level on the other for the urban sites when looking at the areas with the best GAM performance (as seen from Figure 8). In other areas, e.g. Switzerland, Austria and Italy, the GAM actually indicates a positive relationship between temperature and NO_2 and between wind speed and NO_2. Whether this apparent relationship is an artefact in areas with a lower GAM performance or reflects certain physical relationships is difficult to judge. A link between higher temperatures (and wind speed) and NO_2 levels may seem contra-intuitive and it's not clear why such a relationship should exist.

The difference between the meteorologically adjusted trend from the GAM and the plain linear regression show negative values in Northwest Europe and positive values in certain areas further south. This indicates that over the 2000-2010 period, meteorological variability lead to reduced NO_2 levels in the northwest thereby strengthening the decline due to emission reduction whereas in areas in the south meteorology counteracted the decline by causing a slight increase in NO_2.

Table 5 shows that the GAM found significant trends at many more sites than the plain linear regression for all kind of sites, especially the urban and suburban sites. Furthermore, as for 1990-2000, the majority of sites in the 2000-2010 period show negative (downward) trends in NO_2, although not as marked as in the former decade. As seen from Table 5 both the GAM trend and the plain linear trend gives downward trends in NO_2 for all three stations types and for both decades, but with systematically smaller change in the 2000-2010 period compared to 1990-2000.

For all three station types, there are systematic differences in the geographical pattern of the GAM trend showing areas with positive (increasing) trends mainly in parts of central Europe like Austria, S-Germany and some sites in S-France and Spain, and negative (decreasing) trends elsewhere. Table 5 shows that the urban sites have on average a slightly smaller magnitude of the GAM trends. Note though the varying color scale. For the plain linear trends, the results indicate that the smallest magnitude is seen at the rural sites for the 2000-2010 period.

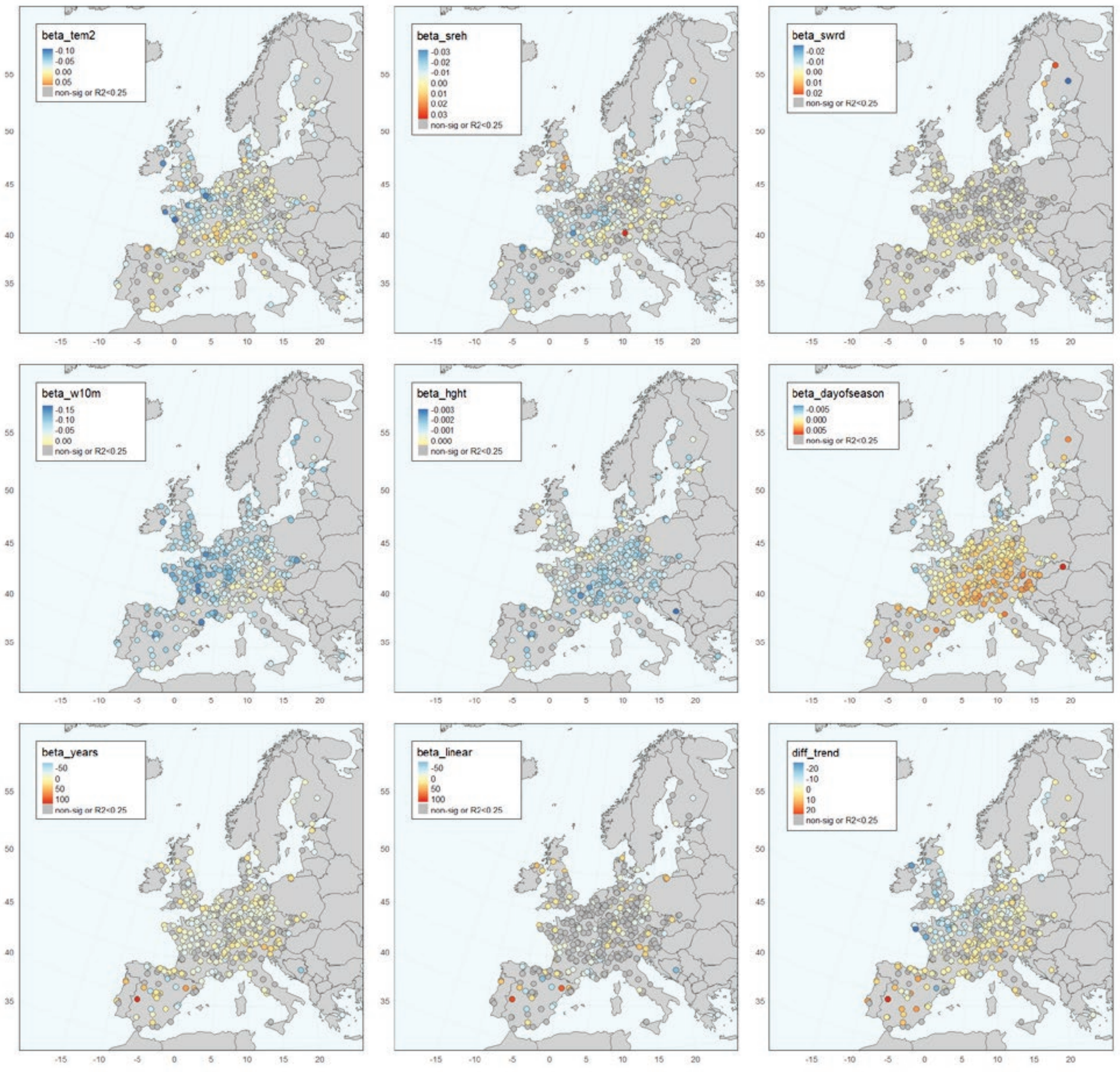

Figure 11. Linearized β coefficients for the individual explanatory variables for the GAM method applied to urban NO_2 sites 2000-2010. Note that the values refer to the log-transformed coefficients. See Table 1 for an explanation of the abbreviated names in the legends. At the bottom row, the coefficient "beta_years" shows the estimated change 1990-2000 as calculated by the GAM (adjusted for meteorology) in percentage relative to 1990 whereas "beta_linear" shows the plain linear trend without meteorological adjustment, also in percentage relative to 1990. "diff_trend" to the right shows the difference (beta_years – beta_linear) which reflects the percentage change 1990-2000 caused by meteorology.

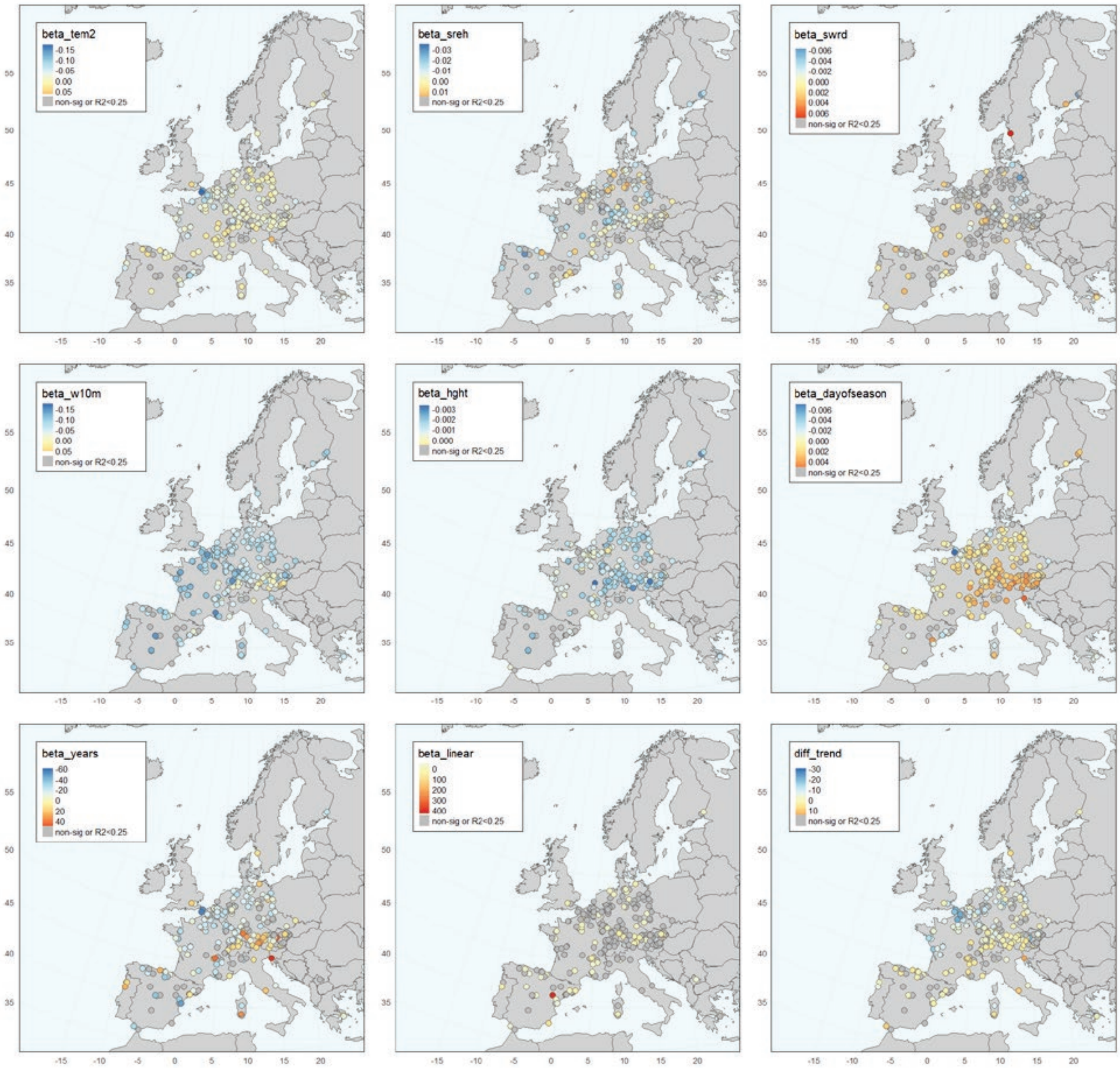

Figure 12. Same as Figure 11 for suburban NO_2 sites.

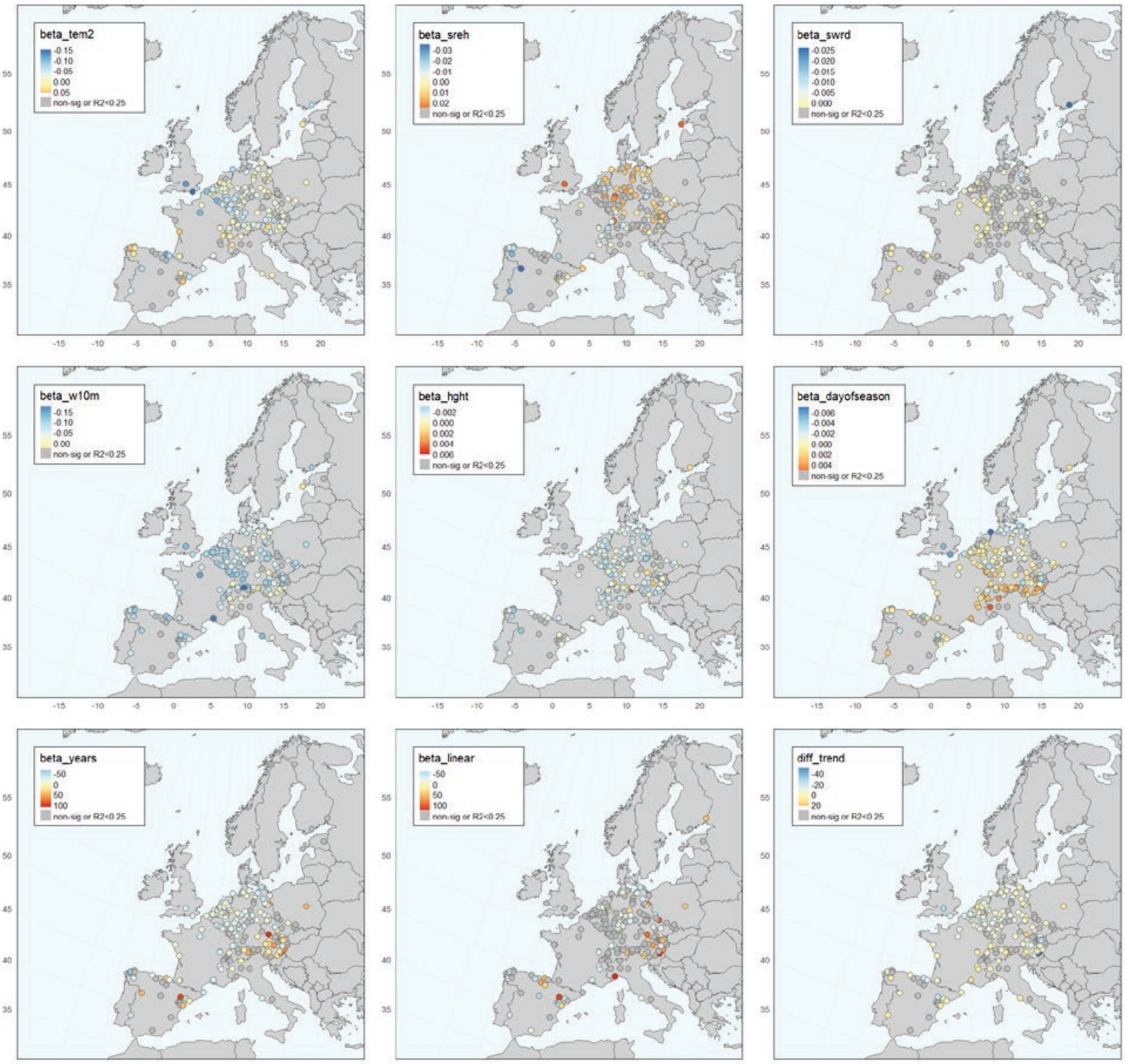

Figure 13. Same as Figure 11 for rural NO_2 sites.

6.2 PM_{10}

Compared to NO_2 the monitoring history and length of the time series for PM_{10} is much shorter. Table 6 gives an overview of the GAM performance for the different station types and time periods for PM_{10}. For the 1990-2000 period, there are very few sites with sufficient length of the time series and thus the statistics are just relevant for those sites. For the period 2000-2010 there are more data available, particularly from urban sites.

In general the GAM performance is poorer for PM_{10} than for NO_2 (see Table 4 and Table 6) as indicated both by the fraction of accepted sites (i.e. sites with R2 > 0.25) and by the statistical metrics themselves. Furthermore, R2 indicates that the performance is better for the urban and suburban data compared to the rural sites. This probably reflects that the concentration levels at sites closer to the emission sources are more tied to the local meteorology (as given by the GAM input) than sites in rural, background areas which presumably are more dependent on long-range transport.

Table 6. Summary statistics for the GAM performance for PM_{10} showing the total number of sites, the percentage of "accepted sites" defined as R2 > 0.25 and the mean of R2, RMSE and MB for the accepted sites.

		Total # sites	% accepted	R2	RMSE	MB
1990-2000	Urban	7	86	0.367	19.11	-0.39
	Suburban	1	100	-	-	-
	Rural	4	75	0.315	21.63	-0.27
2000-2010	Urban	338	61	0.339	16.79	-0.09
	Suburban	135	64	0.352	15.77	-0.03
	Rural	91	36	0.321	15.20	-0.10

6.2.1 PM_{10} in the period 1990-2000

Performance statistics for the GAM method at urban and rural PM_{10} sites for 1990-2000 are shown in Figure 14 and Figure 15, respectively. There were only one suburban site with sufficient PM_{10} data for this period, and thus we have no map for that. Furthermore, the few sites with sufficient monitoring history of PM_{10} in this period only stems from Belgium, the Netherlands and one site in the UK and even for these very few sites, the statistical metrics differ strongly as seen from Figure 14 and Figure 15. Thus, we conclude that the amount of PM_{10} data from 1990-2000 is too little to give any meaningful evaluations of the trends in that period.

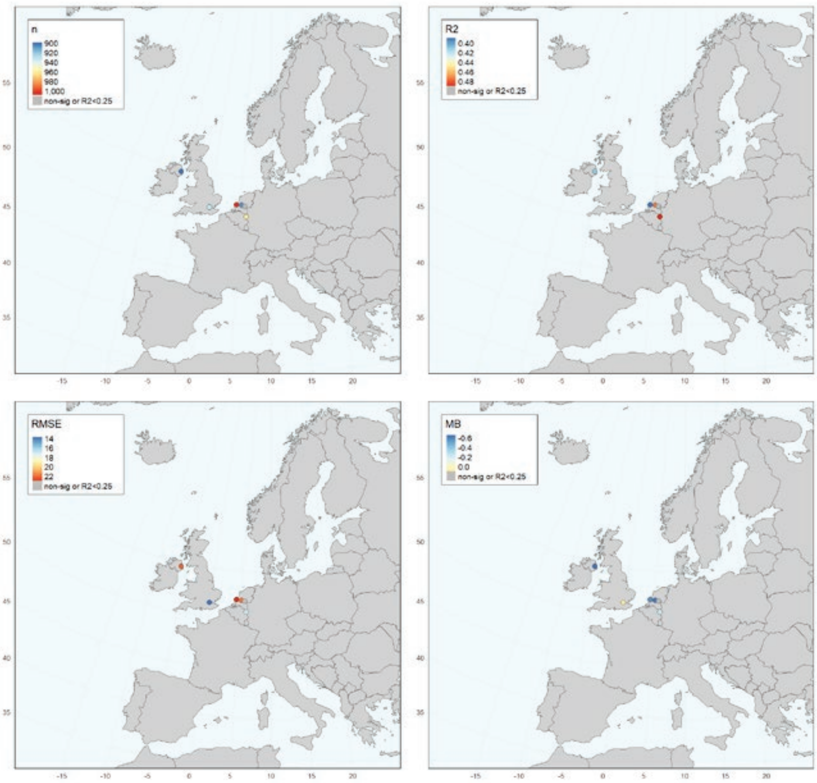

Figure 14. Performance statistics for urban PM_{10} data 1990-2000.

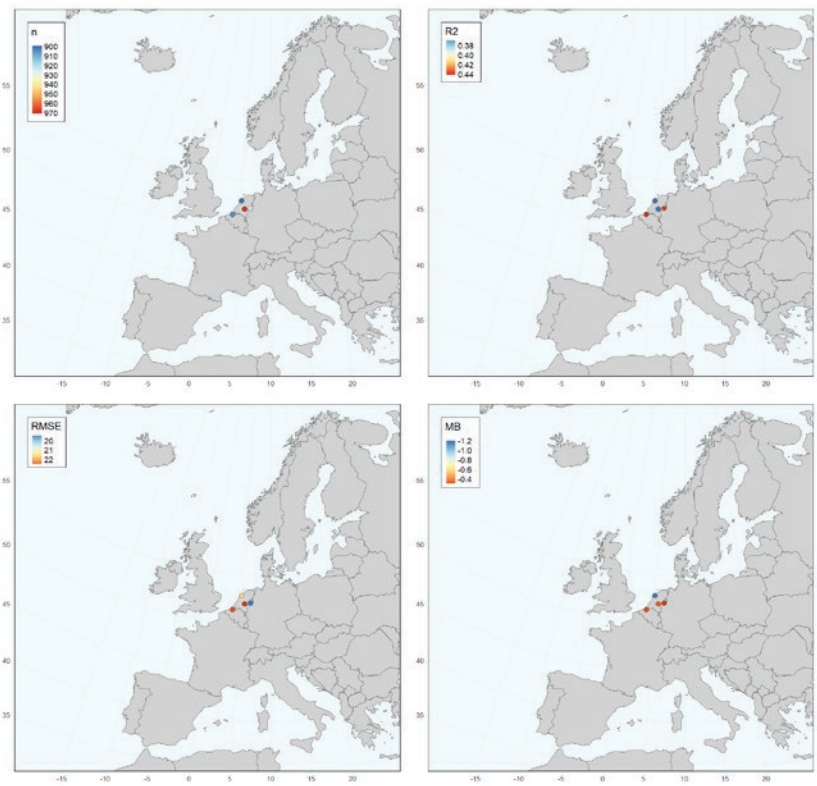

Figure 15. Performance statistics for rural PM_{10} data 1990-2000.

6.2.2 PM_{10} in the period 2000-2010

Performance statistics for the GAM method at urban, suburban and rural PM_{10} sites for 2000-2010 are shown in Figure 16 - Figure 18, respectively. For urban and suburban sites, the PM_{10} data from this period cover many parts of the continent except the south eastern area. France and southern part of Italy is poorly covered with PM_{10} sites in the 2000-2010 period. Furthermore, the number of rural sites with PM_{10} monitoring is much smaller than for the other station types.

Compared to the results for NO_2, there is a less clear pattern in the GAM performance for PM_{10}. The statistics for urban and suburban sites indicate best performance in Belgium, southern Germany and Switzerland. The sites with the highest R2 score are, however, found at some sites in Spain and (for the suburban data) in Portugal although the overall GAM performance for sites on the Iberian plateau is highly variable. Since the PM levels from Spain and Portugal are believed to be significantly influenced by sources not taken into account in the GAM model, in particular Saharan dust, it is somewhat unexpected with the best GAM scores in these countries. It should be said, though, that the number of sites and the variation in the GAM performance is so large that it is difficult to draw firm conclusions about the regional pattern.
The results for PM_{10} at the rural sites (Figure 18) indicates in general a fairly poor GAM performance except at a few sites, at least when measured by R2.

Table 7 shows the main trend statistics for PM_{10}, listing the fraction of significant trends identified by the GAM and the linear regression as well as the fraction of positive and negative significant trends, respectively and the overall mean trend based on all significant trends. This shows that the vast majority of the significant trends were found to be negative (downward) with estimated mean trends that are surprisingly similar for all three station types and for both the GAM trend and the plain linear regression; of the order of 20 % reduction during 2000-2010 relative to 2000 as the reference year.

Table 7. The percentage of significant trends in PM_{10} calculated by the GAM and the linear regression. The percentage of positive and negative significant trends are given as well together with the overall mean trend given in %/decade relative to 2000.

		Total # sites	# sig GAM trends (%)	# sig lin. trends (%)	# pos. GAM trends (%)	# neg. GAM trends (%)	# pos. lin trends (%)	# neg. lin trends (%)	Mean GAM trend (%/dec)	Mean lin. trend (%/dec)
2000-2010	Urban	338	68	52	7	61	4	48	-20	-20
	Suburban	135	67	40	8	58	3	36	-19	-17
	Rural	91	53	40	5	48	5	35	-20	-17

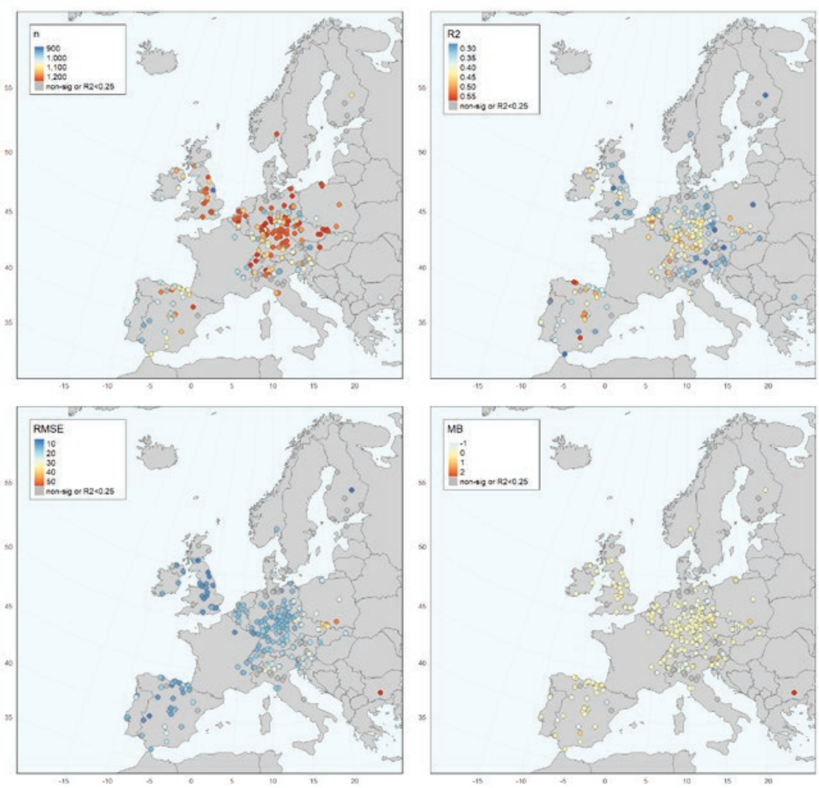

Figure 16. Performance statistics for urban PM_{10} data 2000-2010.

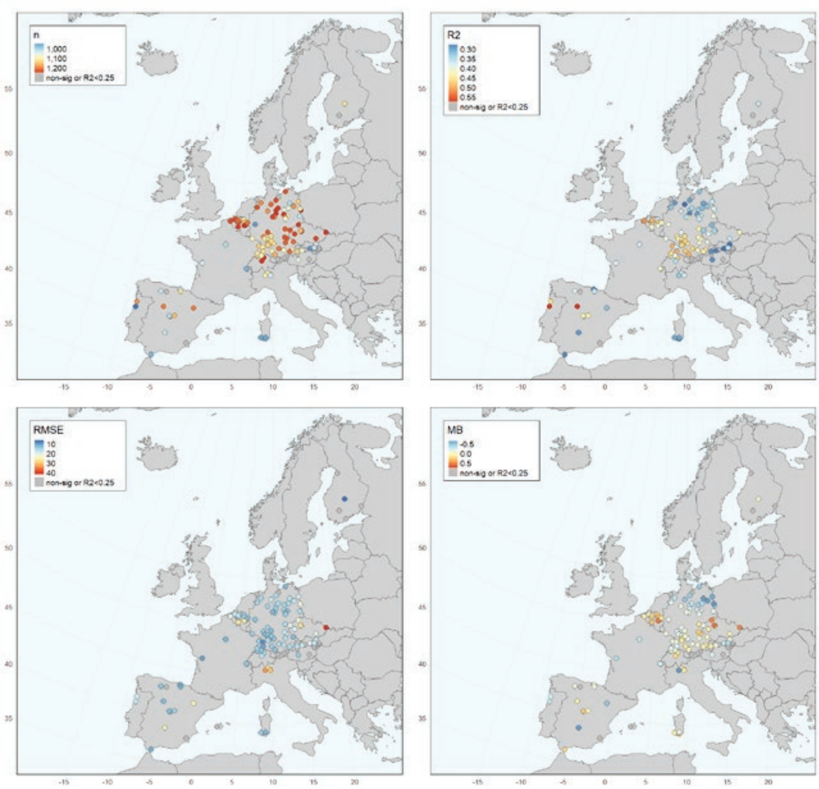

Figure 17. Performance statistics for suburban PM_{10} data 2000-2010.

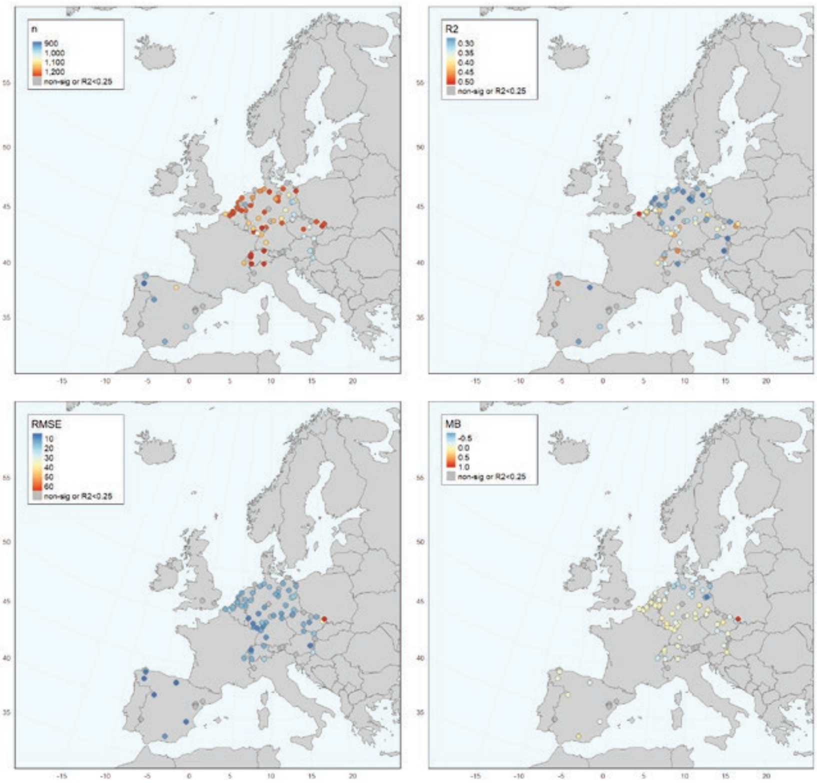

Figure 18. Performance statistics for rural PM_{10} data 2000-2010.

The linearized β coefficients for the explanatory variables in Eq. (1) are shown in Figure 19 - Figure 21 for the 2000-2010 urban, suburban and rural PM_{10} sites, respectively. As noted above, these values refer to the log transformed data, i.e. they have the unit [log (µg m^{-3})], except for the trend terms.

For all three station types, there is a clear negative relationship between wind speed and PBL height on one hand and PM_{10} levels on the other, i.e. high levels of PM_{10} is linked to situations with low wind speeds and a shallow mixing layer. This was also seen for NO_2 and is simply reflecting that the highest concentration levels are coupled to episodes with the least atmospheric mixing.

As for NO_2, the PM_{10} results indicates a positive relationship between day of season and concentration level, indicating that the strongest PM_{10} episodes tend to occur in the last part of winter. Furthermore, for all three types of stations, the results show a positive relationship between relative humidity in areas closer to the sea around Germany, Belgium and the Netherlands whereas there is a negative relationship elsewhere. This might reflect an effect of sea salt in coastal areas and an effect of wash-out of PM in continental areas. Without more detailed investigations of the data, these are speculations though.

With respect to the link between PM_{10} and temperature, both the urban, suburban and rural data indicate a marked geographical pattern with a negative relationship in central Europe and a positive relationship in Spain, southern France and northern Italy. This indicates that there are distinct differences in the sources of PM_{10} and the associated weather conditions in south Europe compared to central Europe. One possible reason for this could be a dominance of secondary PM in the south and primary PM pollution in central Europe.

As mentioned above, the vast majority of the estimated trends are found to be negative. At a few sites, an increasing trend is found without a very clear pattern though. For the urban sites, the French sites plus a few Spanish sites show increasing trend in PM_{10} as seen by the GAM. The difference between the GAM trend and the plain linear regression, indicate that meteorology lead to increased levels in the southern part and – for the suburban and rural sites – also the central part of the continent, and decreased levels in the north (mainly N-Germany) during 2000-2010.

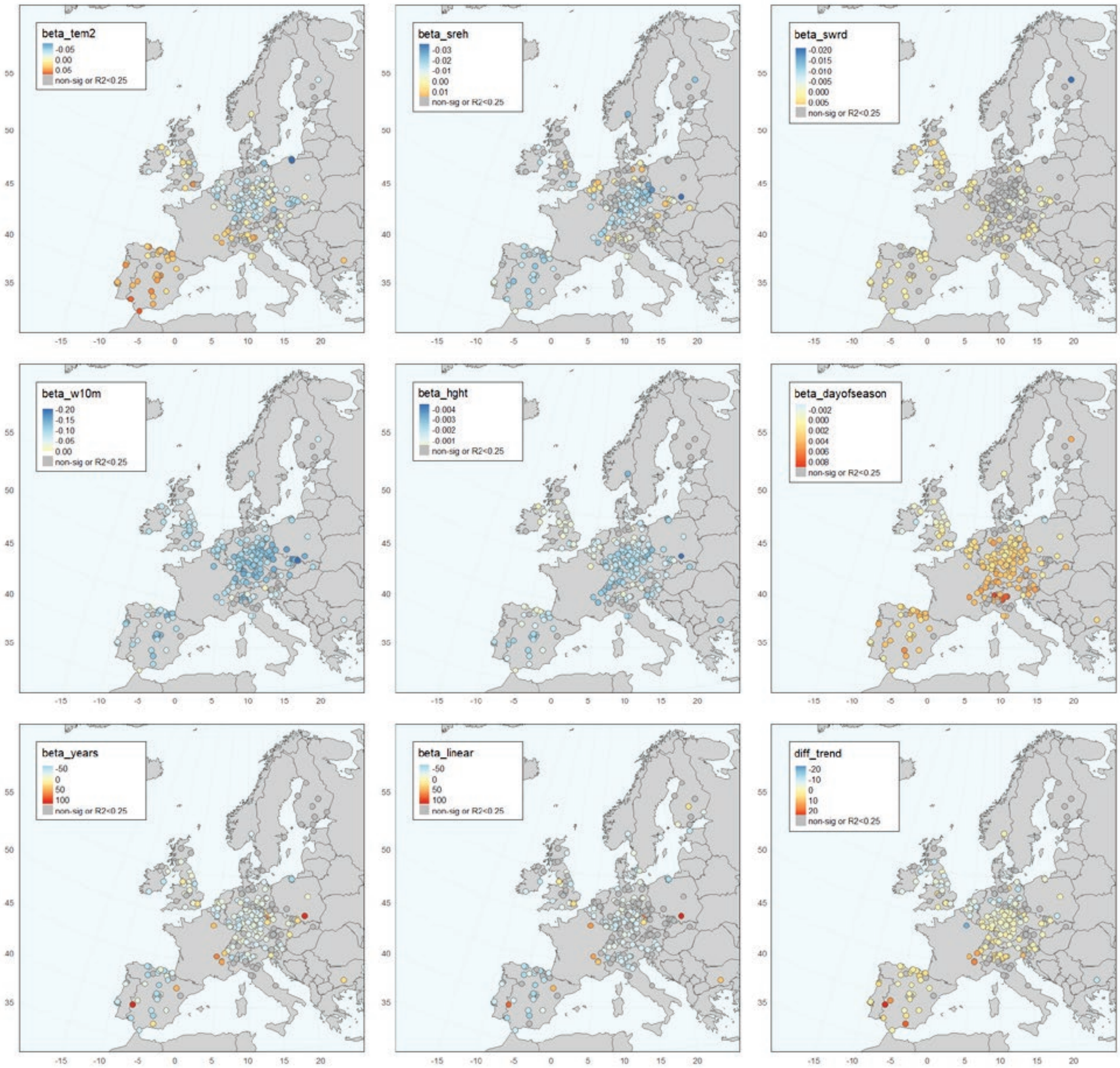

Figure 19. Linearized β coefficients for the individual explanatory variables for the GAM method applied to urban PM_{10} sites 2000-2010. Note that the values refer to the log-transformed coefficients. See Table 1 for an explanation of the abbreviated names in the legends. The coefficient "beta_years" show the estimated linear trend as calculated by the GAM (with meteorological decomposition) whereas "beta_linear" show the plain linear trend without any meteorological decomposition.

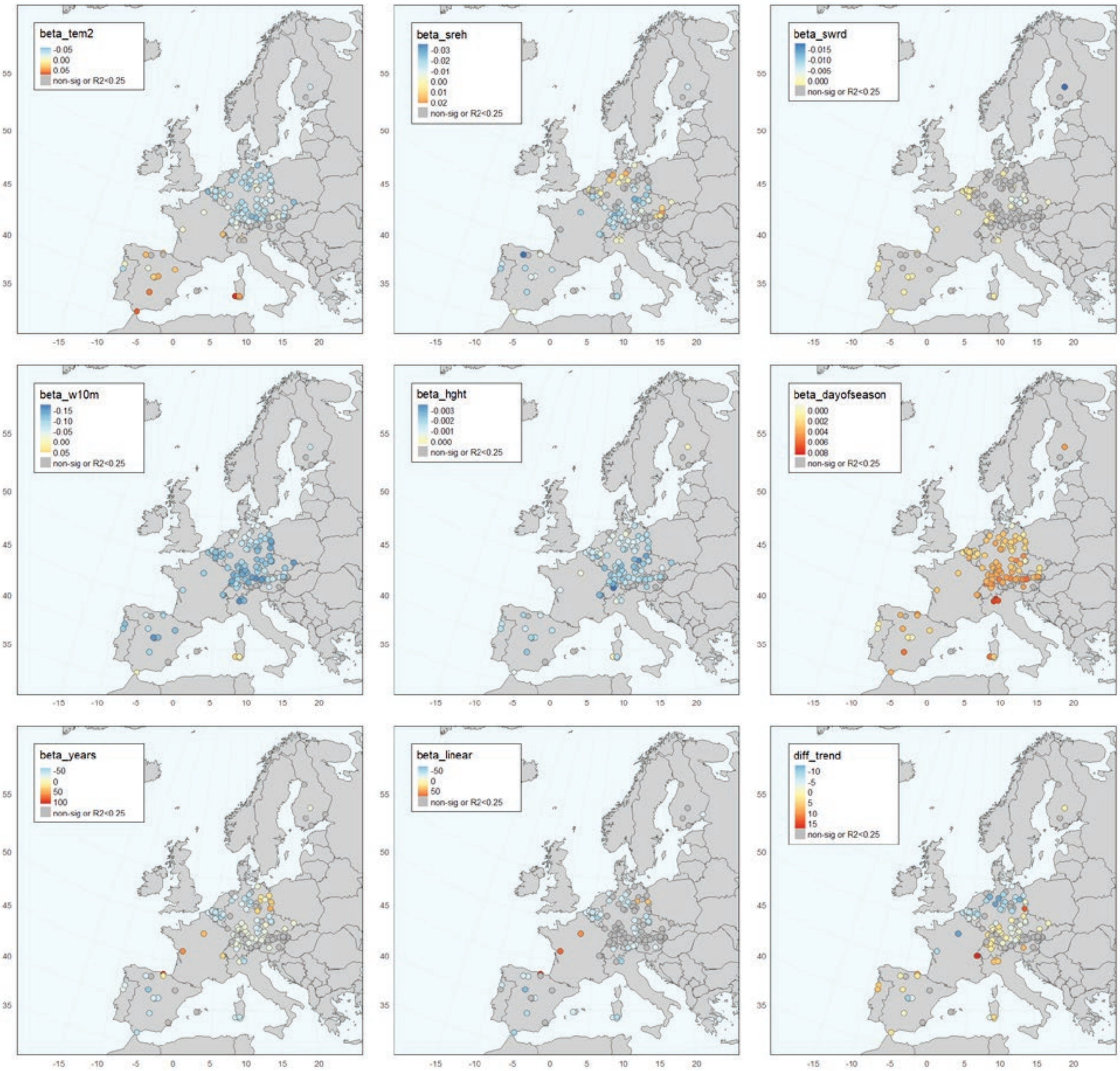

Figure 20. Same as Figure 19 for suburban PM_{10} data 2000-2010.

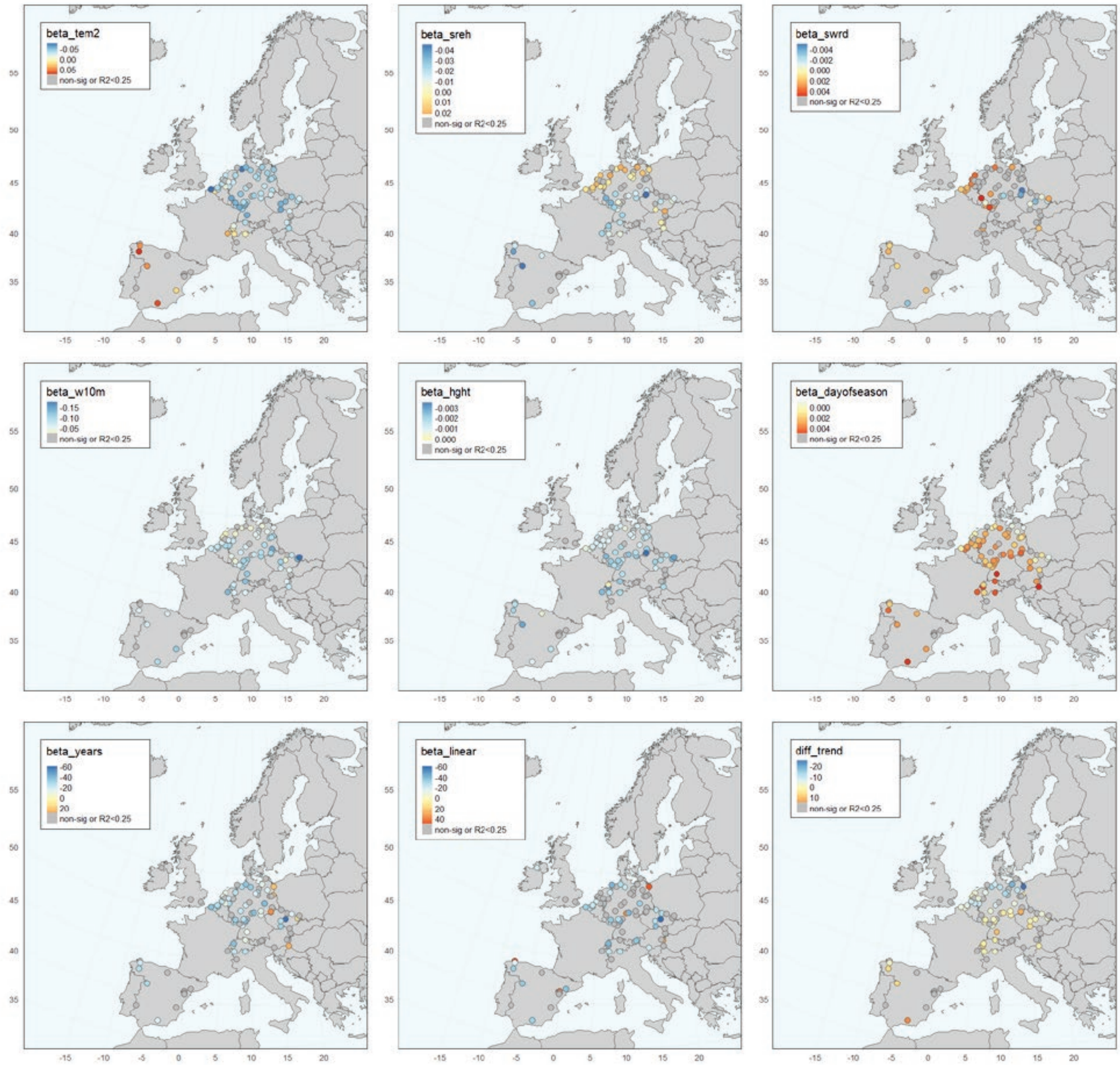

Figure 21. Same as Figure 19 for rural PM_{10} data 2000-2010.

6.3 $PM_{2.5}$ in the period 2000-2010

As seen from Table 2, the number of sites with sufficient length of the time series with $PM_{2.5}$ data is very small. An overview of the GAM performance for $PM_{2.5}$ for the period 2000-2010 is given in Table 8. The number of urban and rural $PM_{2.5}$ sites for this period is only 6-7 and for around half of the sites, the GAM performance was too low to be accepted based on the criteria R2 > 0.25 (Figure 22 - Figure 23). Thus, without more sites, we can't really make any further evaluation of the trends and the GAM method for $PM_{2.5}$.

Table 8. Summary statistics for the GAM performance for $PM_{2.5}$ for 2000-2010 showing the total number of sites, the percentage of "accepted sites" defined as R2 > 0.25 and the mean of R2, RMSE and MB for the accepted sites.

		Total # sites	% accepted	R2	RMSE	MB
2000-2010	Urban	6	50	0.367	9.16	-0.04
	Suburban	1	100	-	-	-
	Rural	7	57	0.341	6.39	0.05

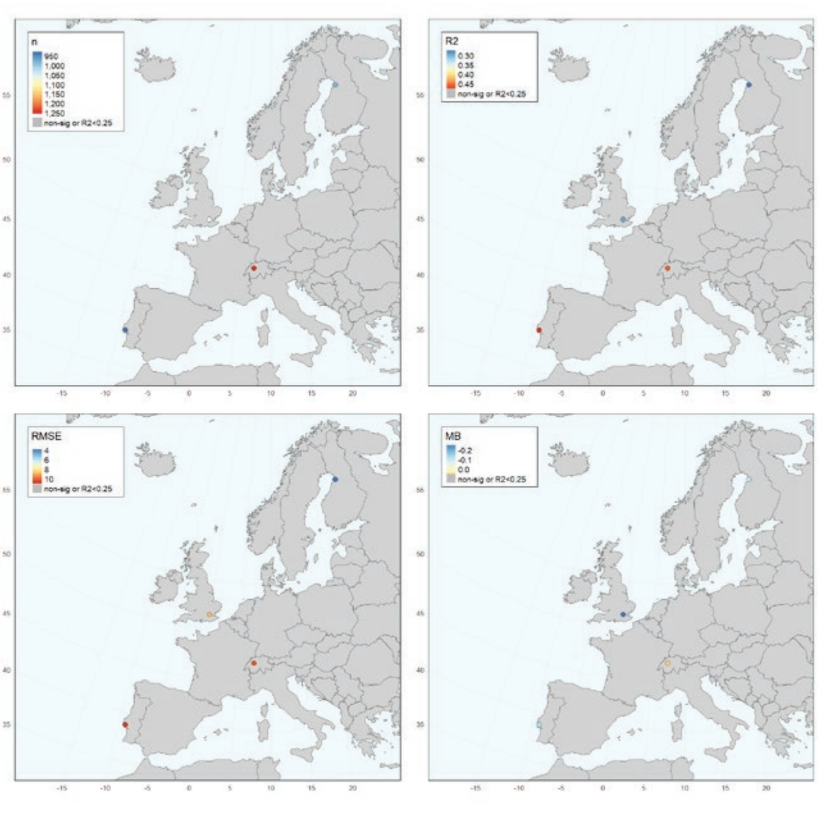

Figure 22. Performance statistics for urban $PM_{2.5}$ data 2000-2010.

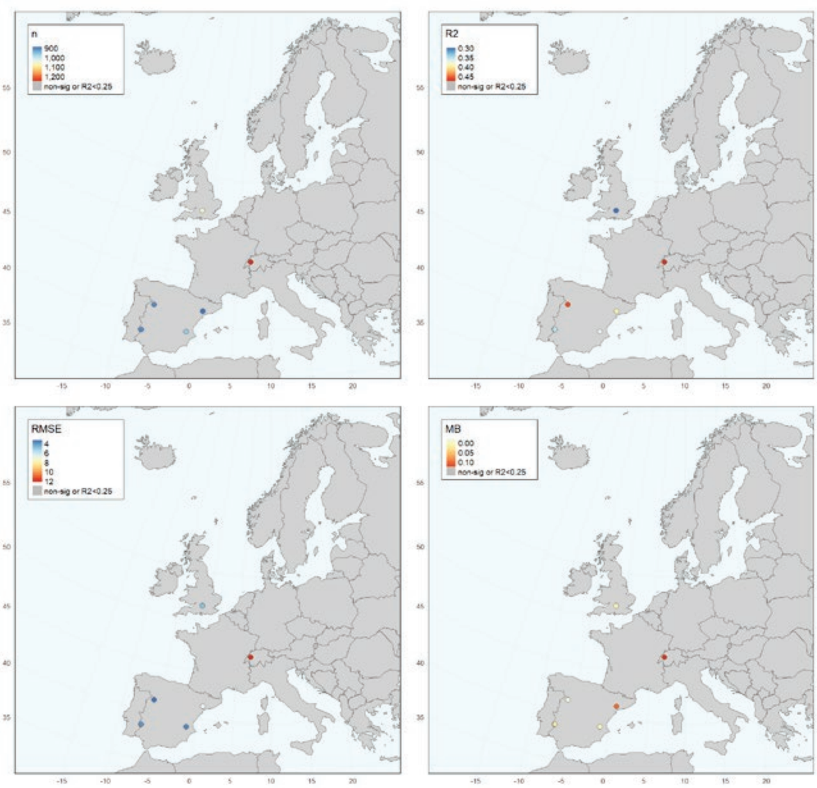

Figure 23. Performance statistics for rural $PM_{2.5}$ data 2000-2010.

7 Trends and time series at selected example sites

The rationale for the present work is the issue of meteorological influence on long-term trends in NO_2 and PM. While the air pollutant concentrations are the combined result of emissions, atmospheric transport and chemical transformation, we would like to separate these processes in order to evaluate the separate influence of emissions on one side and other, natural processes (i.e. meteorology) on the other side.

A main assumption of the present method is that the daily mean measured pollutant levels at a monitoring site could be estimated by a linear combination of local gridded daily meteorological parameters plus a long-term trend. In the discussion above, we have shown how our method (the GAM) performs on a daily basis as measured by standard statistical metrics like R2, RMSE, MB, etc.

In the following we show examples of observed and predicted daily time series at some selected sites as well as the estimated trends. As mentioned, the amount of data is overwhelming and therefore just a few selected examples could be presented. The panels showing daily data are indeed small and hard to read, but the main purpose of these is to show the general agreement between measured and modelled data through the season.

The GAM performance for the urban NO_2 data for the period 2000-2010 was ordered and sorted based on the R2 statistic. Below are shown the results for some selected sites.

7.1 NO_2 at Gerardmer (FR30022) 48.087°N, 6.873°E, 660 m asl, background urban

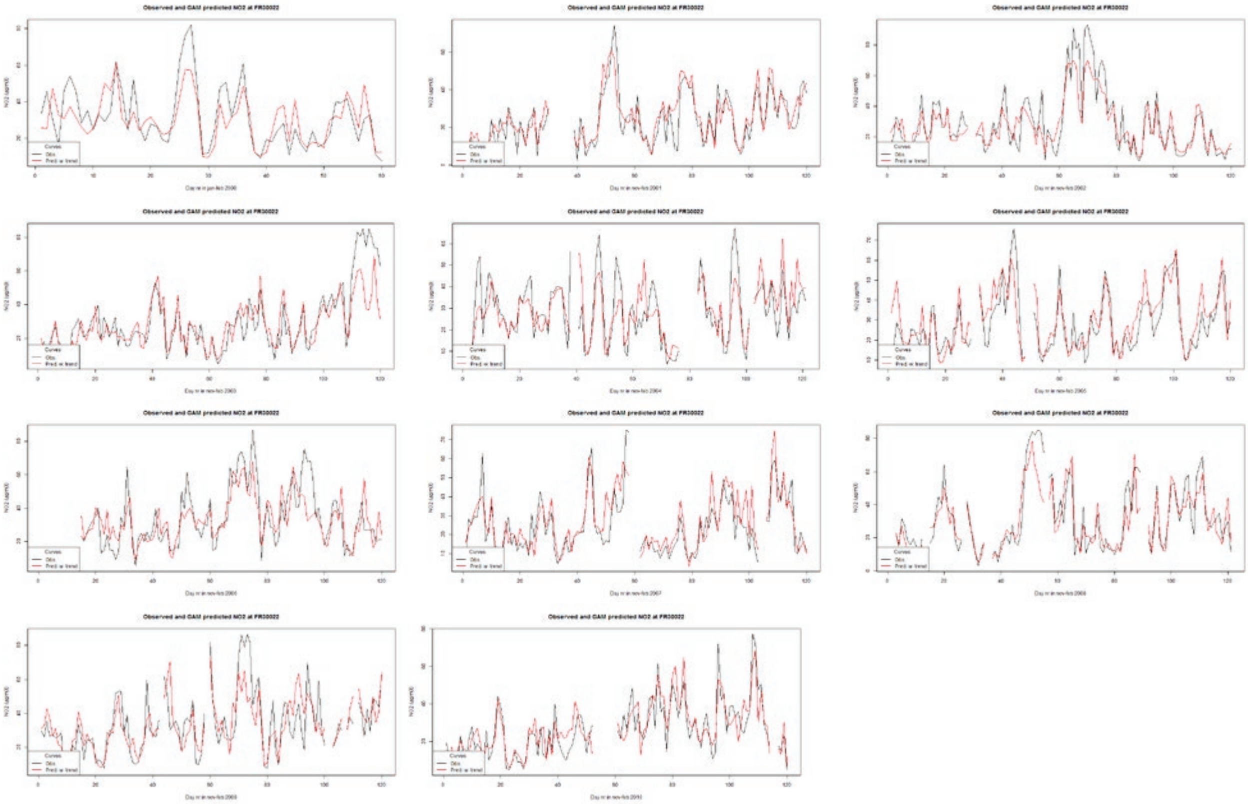

Figure 24. Daily mean measured (black) and predicted (red) NO_2 concentrations at FR30022 winter months 2000-2010. The years are arranged from left to right. Note that the panels show data for 1 Nov the previous year to end of Feb.

This site showed the highest R2 score (R2 = 0.668) of the urban NO_2 sites for the 2000-2010 period. The station is located in the eastern part of France. As indicated by Figure 24 there is a close agreement between observed and predicted values in all years although the peak episodes tend to be underestimated by the GAM. Figure 25 shows that nearly all the inter-annual variability is due to the meteorology whereas the trend term is negligible (indicated by the difference between the two red lines). The PBL height, wind speed and day of season are the dominant explanatory variables as shown by Figure 25. The right plot in Figure 25 is based on modelling using the R package deweather ver. 0.5 (Carslaw and Taylor, 2009). This package uses a boosted regression tree method which is a nonparametric method rather than GAM to model the relationship between NO_2 and the explanatory variables. However, similar relationships between the variables was found using this method as for the GAM.

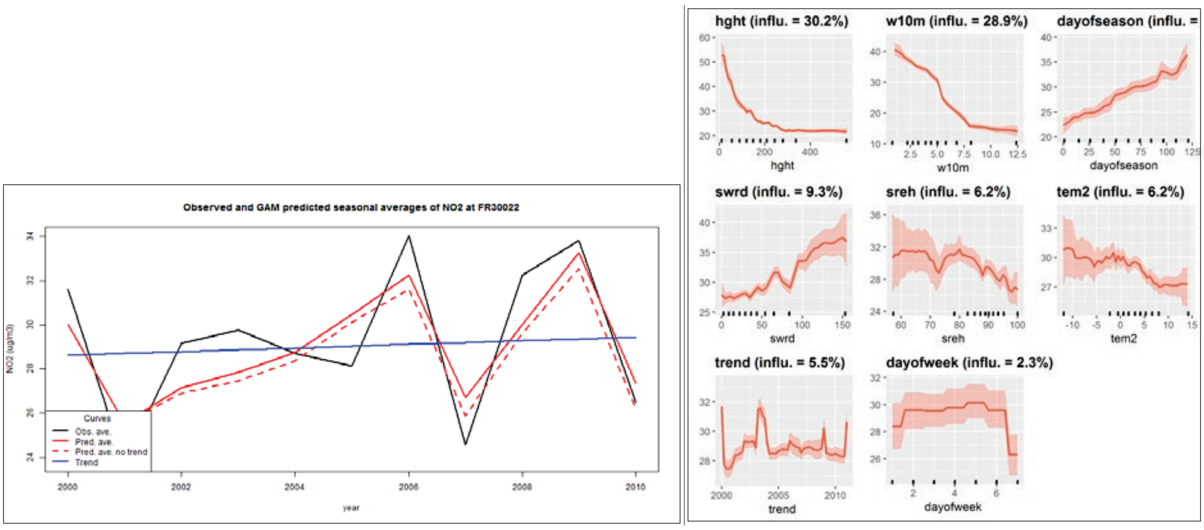

Figure 25 Left: Measured (black) and predicted (red) seasonal mean NO_2 concentrations. Red dashed line mark the predicted values subtracted the trend term (blue). Right: Relative importance and levels of the explanatory variables.

7.2 NO_2 at Garches (FR04145) 48.846°N, 2.189°E, 134 m asl, background urban

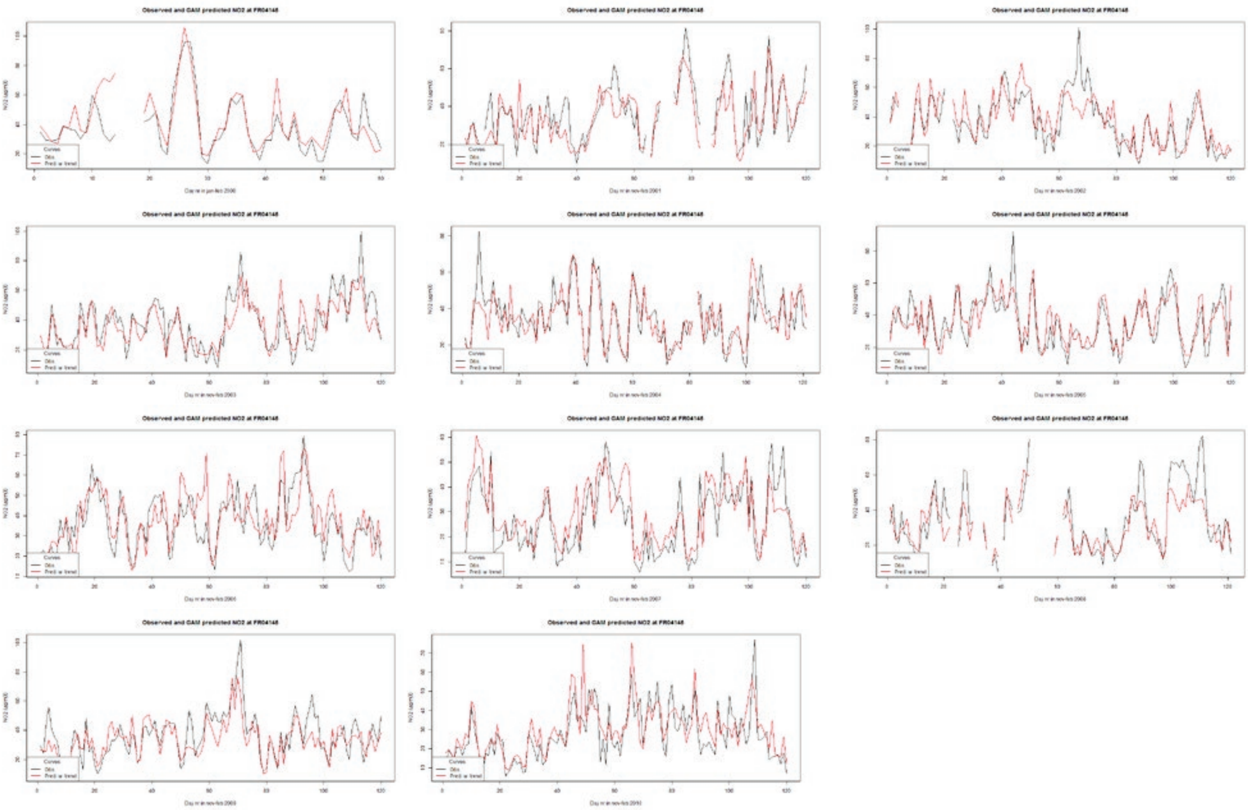

Figure 26. Daily mean measured (black) and predicted (red) NO_2 concentrations at FR04145 winter months 2000-2010. The years are arranged from left to right. Note that the panels show data for 1 Nov the previous year to end of Feb.

This site showed the next highest R2 score (R2 = 0.633) of the urban NO_2 sites for the 2000-2010 period. The station is located near the centre of Paris, somewhat to the west of the main centre. As indicated by Figure 26 there is a close agreement between observed and predicted values in all years also for this site. Some peak episodes are underestimated by the GAM, though. Figure 27 indicates a strong downward trend in NO_2 as well as inter-annual meteorological variability leading to peaks in 2006 and 2009. The wind speed, temperature and trend term are the dominant explanatory variables as shown by Figure 27.

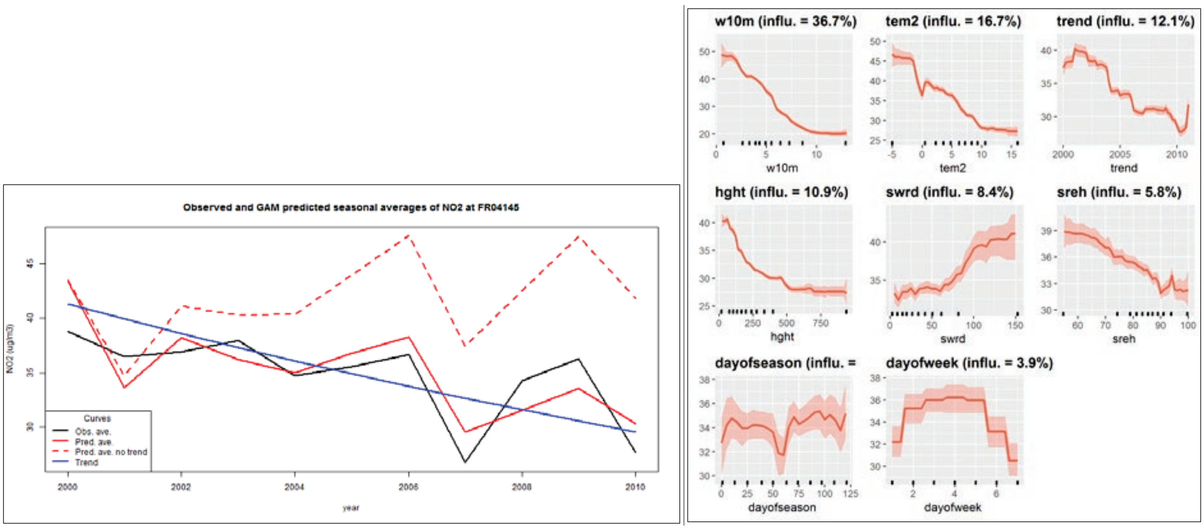

Figure 27. Left: Measured (black) and predicted (red) seasonal mean NO_2 concentrations. Red dashed line mark the predicted values subtracted the trend term (blue). Right: Relative importance and levels of the explanatory variables.

7.3 NO_2 at Evreux Centre (FR25039) 49.0213°N, 1.1483°E, 65 m asl, background urban

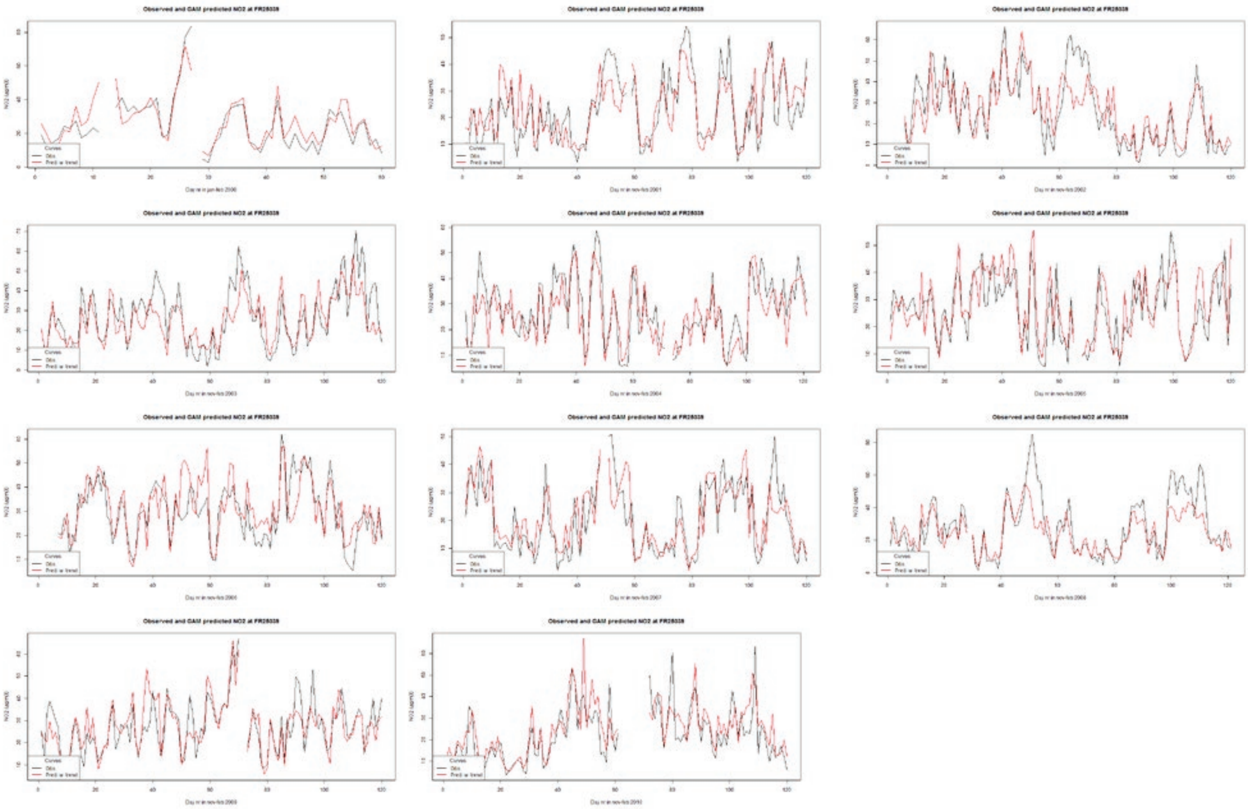

Figure 28. Daily mean measured (black) and predicted (red) NO_2 concentrations at FR25037 winter months 2000-2010. The years are arranged from left to right. Note that the panels show data for 1 Nov the previous year to end of Feb.

This site showed the third highest R2 score (R2 = 0.622) of the urban NO_2 sites for the 2000-2010 period. The station is located approximately 85 km NW of the centre of Paris. Figure 28 indicates a close agreement between observed and predicted values although some peak episodes are underestimated by the GAM. Figure 29 indicates a downward trend in NO_2 when adjusted for the meteorological variability. Based on the NO_2 data alone, one would presumably not find any significant trend at this site. It is interesting though, to note the discrepancy in the observed and predicted NO_2 seasonal mean values in Figure 29 given the close agreement in the daily values. The wind speed, PBL height and day of week are the dominant explanatory variables as shown by Figure 29.

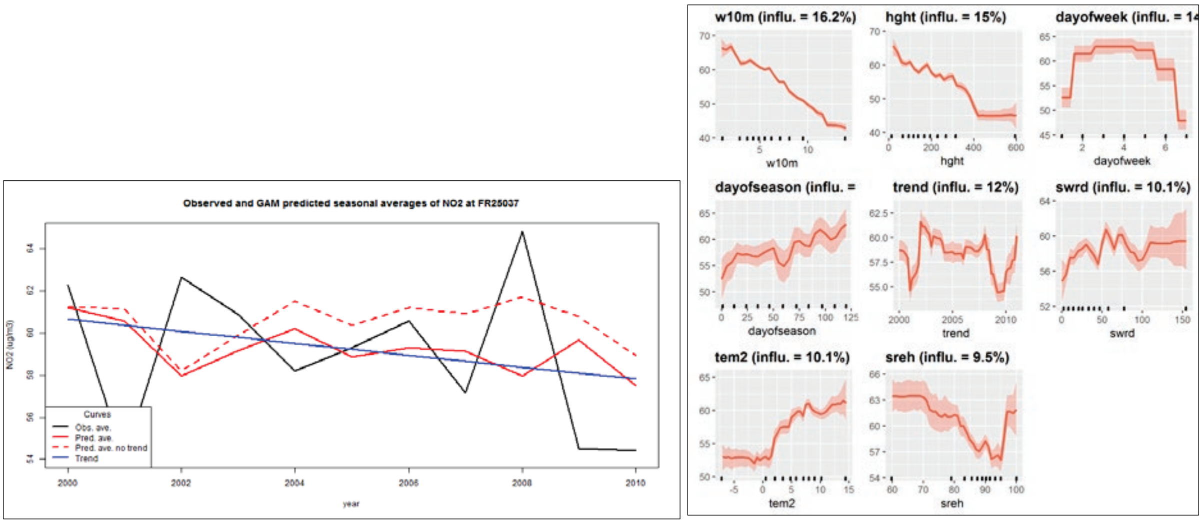

Figure 29. Left: Measured (black) and predicted (red) seasonal mean NO_2 concentrations. Red dashed line mark the predicted values subtracted the trend term (blue). Right: Relative importance and levels of the explanatory variables.

7.4 NO_2 at London Kensington N (GB0620A) 51.5211°N, 0.2134°W, 5 m asl, background urban

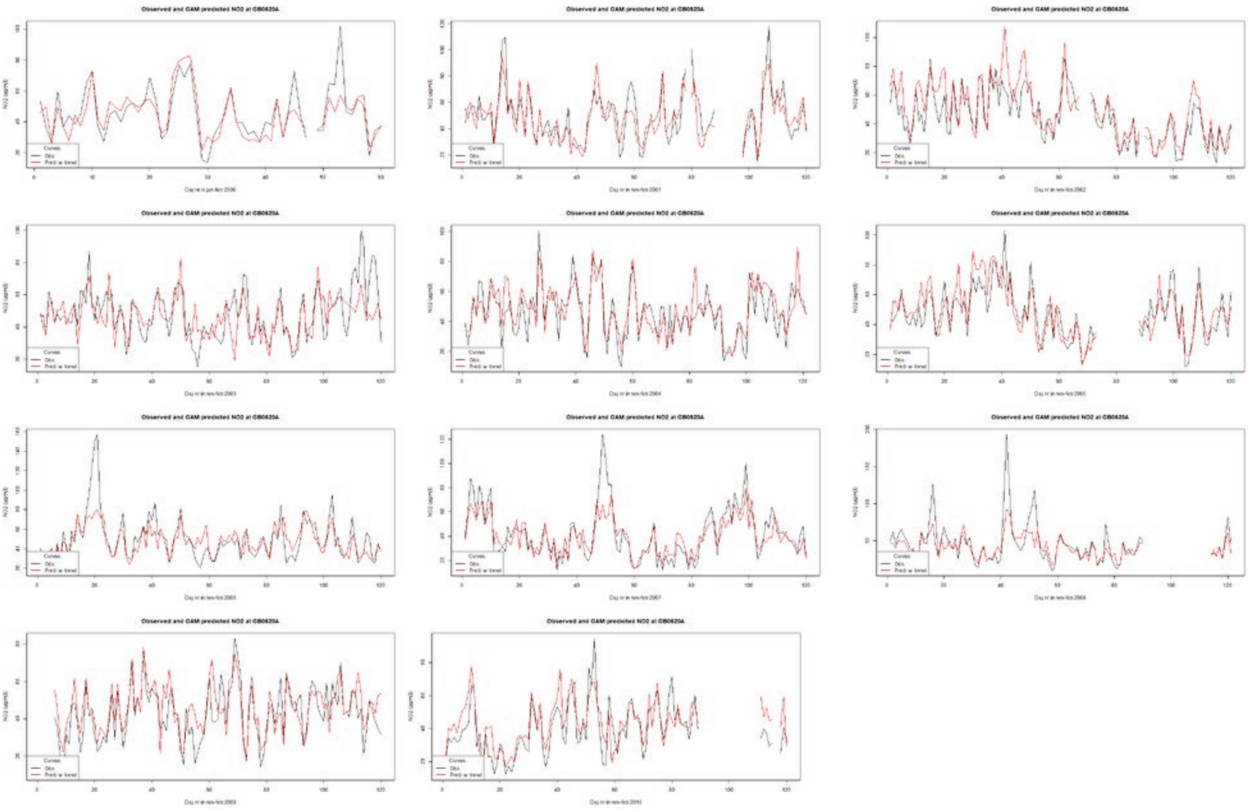

Figure 30. Daily mean measured (black) and predicted (red) NO_2 concentrations at GB0620A winter months 2000-2010. The years are arranged from left to right. Note that the panels show data for 1 Nov the previous year to end of Feb.

This site showed the seventh highest R2 score (R2 = 0.596) of the urban NO_2 sites for the 2000-2010 period. The station is located in central London and experiences peak levels above 100 µg m^{-3} in the daily mean NO_2 (even approaching 200 µg m^{-3} in 2008). Figure 30 shows a very good agreement between observed and predicted values for episodes up to around 80 µg m^{-3} whereas the highest peaks are strongly underestimated. The long term changes during 2000-2010 is apparently the net result of a strong overall decline with significant inter-annual variability due to variations in meteorology as seen from Figure 31. Wind speed, PBL height, trend and day of season are the most important explanatory variables at this site.

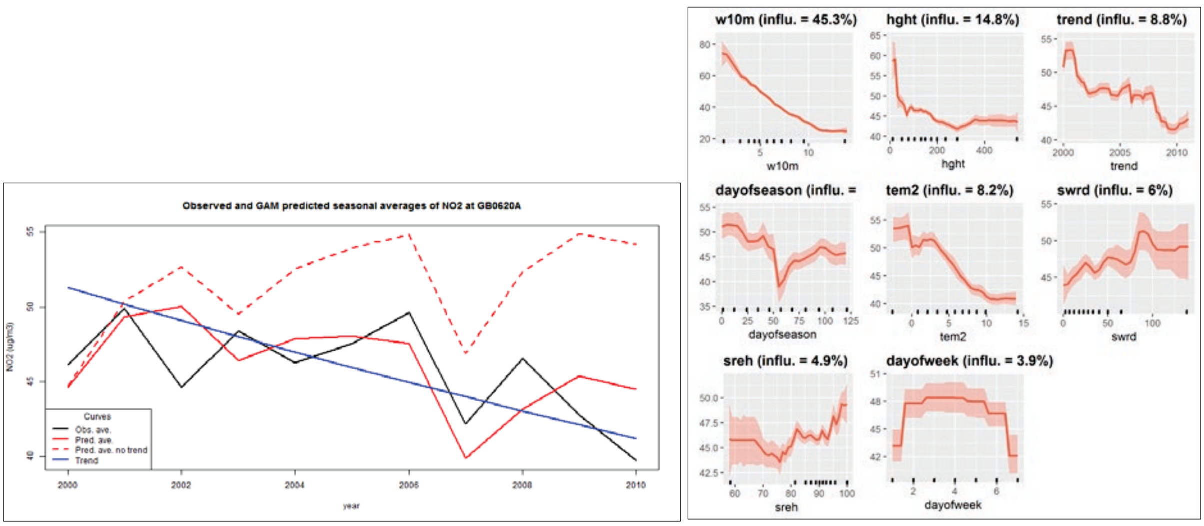

Figure 31. Left: Measured (black) and predicted (red) seasonal mean NO_2 concentrations. Red dashed line mark the predicted values subtracted the trend term (blue). Right: Relative importance and levels of the explanatory variables.

7.5 NO_2 at Maranon (ES0116A) 40.4378°N, 3.6908°W, 669 m asl, traffic urban

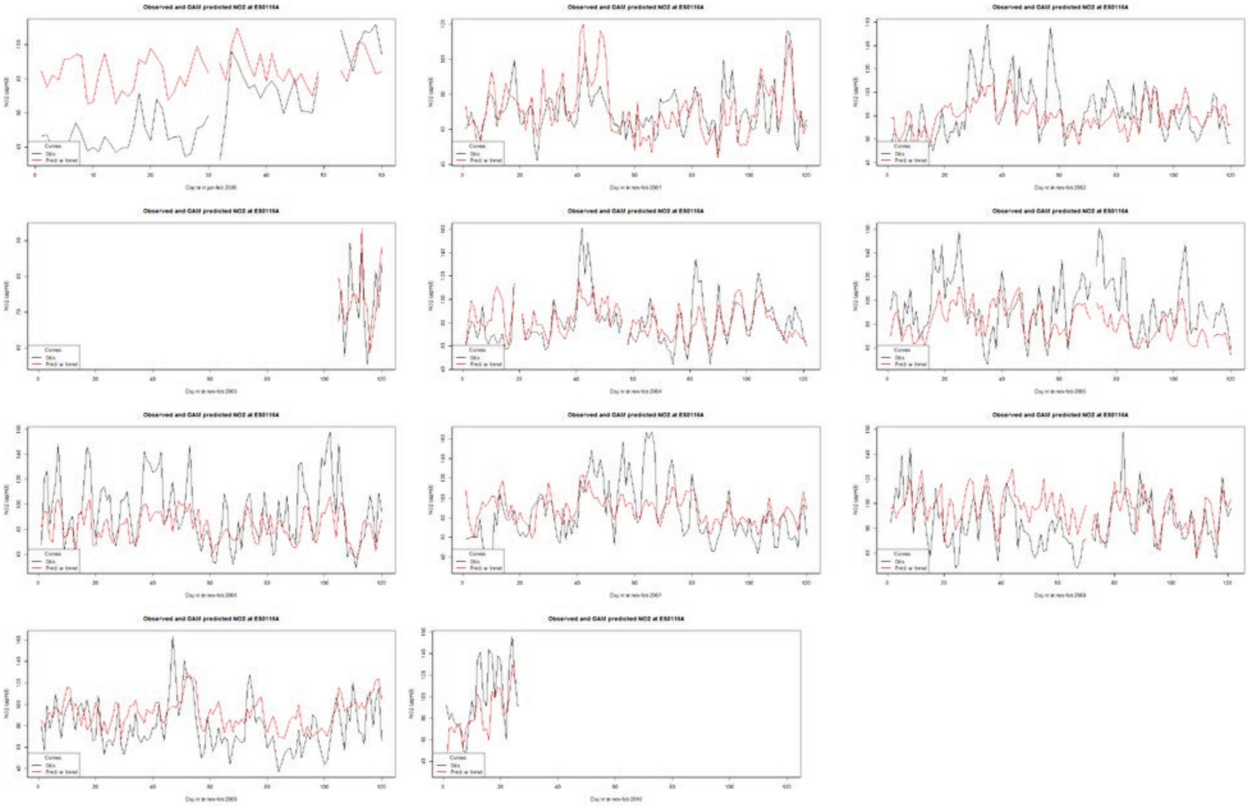

Figure 32. Daily mean measured (black) and predicted (red) NO_2 concentrations at ES0116A winter months 2000-2010. The years are arranged from left to right. Note that the panels show data for 1 Nov the previous year to end of Feb.

This site is included as an example of sites on the other end of the scale, having an R2 = 0.258 which is just above the acceptance criteria of 0.25. The Maranon site is located at the side of a road in the centre of Madrid with levels of daily mean NO_2 of the same order as the London Kensington site discussed above. Figure 32 shows that the variation from day to day is not that bad predicted, but that many of the peak episodes are strongly underestimated while the GAM overestimates the levels in some other parts. The data also includes some extended periods with missing data. Figure 33 shows a marked increasing trend, both in the observations and in the meteorologically adjusted trend estimated from the GAM with some inter-annual meteorological variability on top. The underestimation of the concentration levels in 2005 and 2006 is clearly seen.

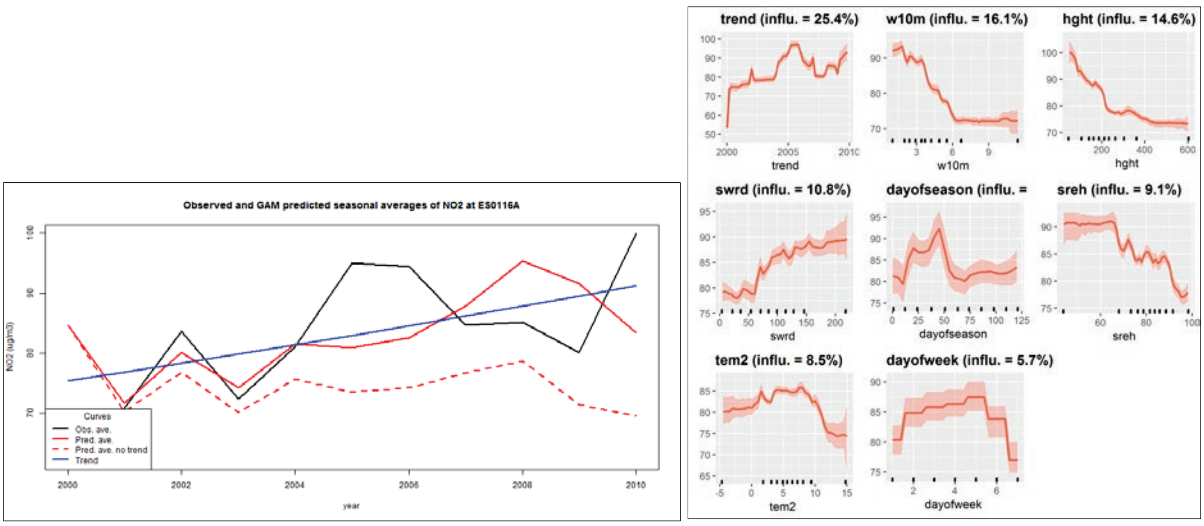

Figure 33. Left: Measured (black) and predicted (red) seasonal mean NO_2 concentrations. Red dashed line mark the predicted values subtracted the trend term (blue). Right: Relative importance and levels of the explanatory variables.

7.6 PM_{10} at Bailén (ES1253A) 38.0929°N, 3.7839°W, 368 m asl, industrial urban

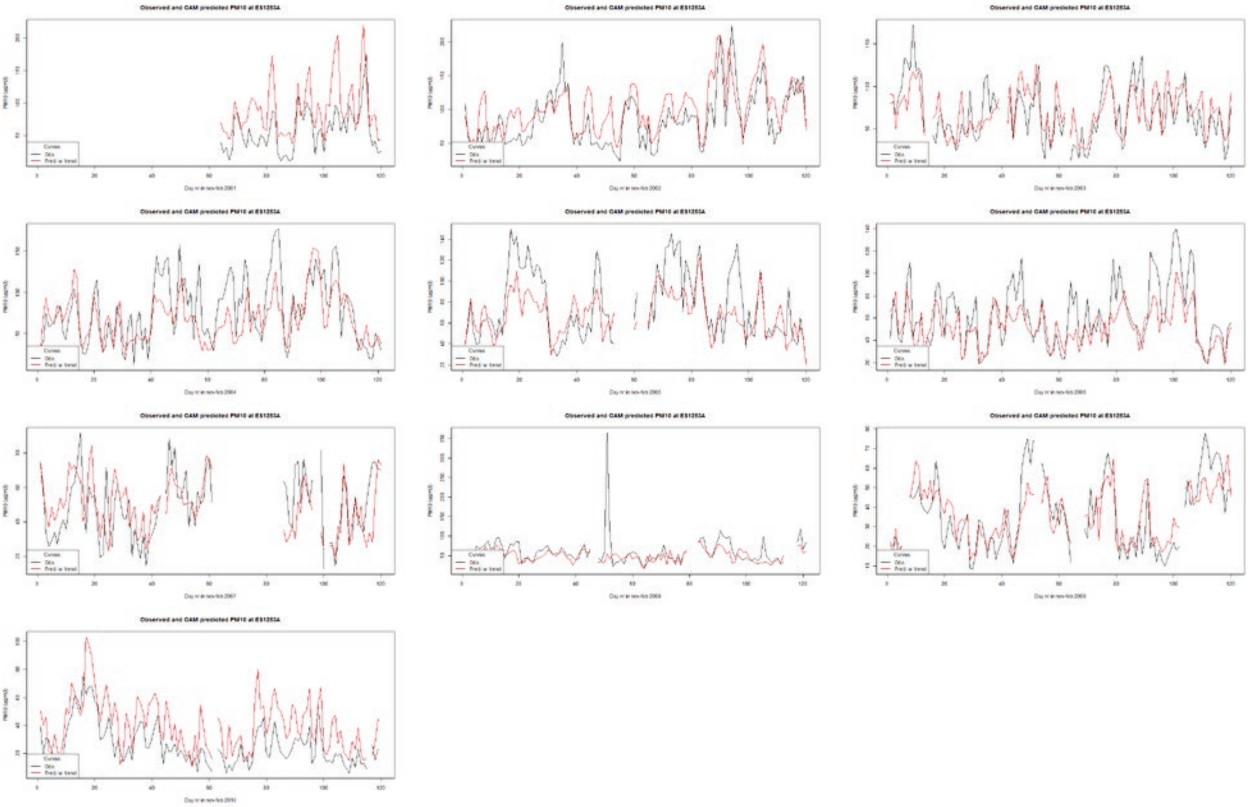

Figure 34. Daily mean measured (black) and predicted (red) PM_{10} concentrations at ES1253A winter months 2001-2010. The years are arranged from left to right (no data for 2000). Note that the panels show data for 1 Nov the previous year to end of Feb.

This site is one of the PM_{10} sites with the highest R2 score (R2 = 0.499). The station is located in the town of Bailén in southern Spain in the eastern part of the community of Andalusia. Figure 34 shows that the temporal variation is fairly well predicted although the GAM underestimates the levels in some years and overestimates them in 2010. One extreme episode in 21 Dec 2007 with 365 µg m^{-3} is probably linked to Saharan dust. The weather map show a situation with strong advection from the south this day. The seasonal averages (Figure 35) indicates a very strong decline in the PM_{10} levels during the period 2001-2010 at this site, while the inter-annual variation due to meteorology is small in comparison.

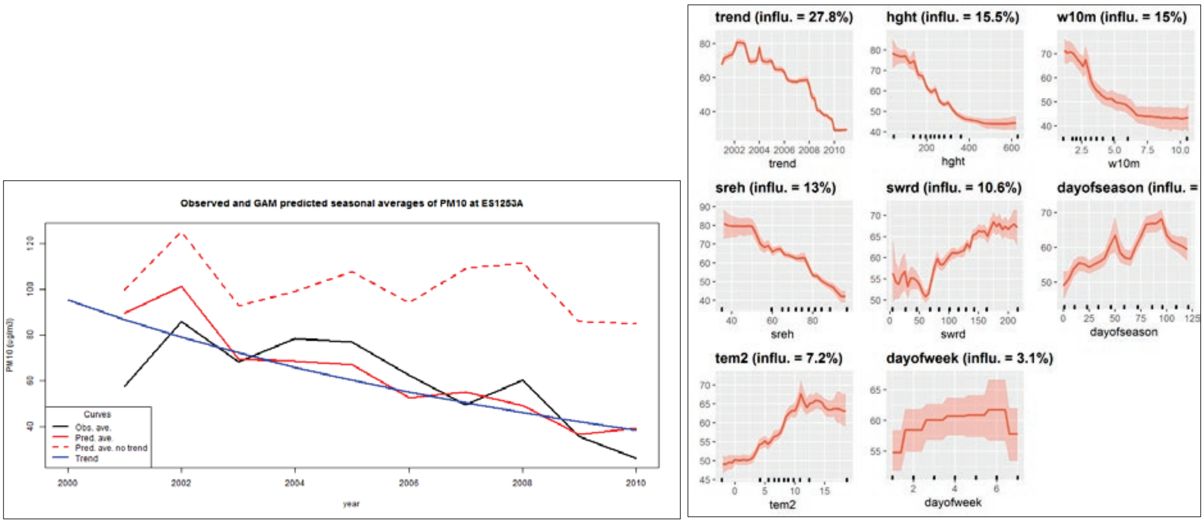

Figure 35. Left: Measured (black) and predicted (red) seasonal mean PM_{10} concentrations. Red dashed line mark the predicted values subtracted the trend term (blue). Right: Relative importance and levels of the explanatory variables.

8. Discussion and conclusions

This report has presented the results applying a GAM (Generalized Additive Model) to NO_2 and PM_{10} monitoring data in order to explain the trends in these species with respect to meteorological variability and other effects such as emissions, respectively. Two time periods were studied: 1990-2000 and 2000-2010 for which gridded meteorological data from the EuroDelta Trends projects were available. For the air pollutant data we used EEA's Airbase data allocated to three types of site locations; urban, suburban and rural. For each station separately, we constructed time series consisting of daily values, i.e. daily means of the measured species as well as daily means of the gridded meteorological model data. Then, for each station individually, we applied the GAM for the two time periods separately.

It turned out that the general performance of the GAM when measured by the R2 statistic showed clear geographical differences. Best performance for NO_2 was found in the areas typically associated with the main European emission area, i.e. Belgium, the Netherlands, NW Germany and the UK. Significantly poorer performance was found for e.g. Austria and areas in southern Europe. Marked differences were also found for PM_{10} although there was a less clear spatial pattern.

The difference between the meteorological adjusted trend from the GAM and the plain linear regression was used to estimate the impact of meteorology on the trends. This indicated that in the period 1990-2000, the meteorological variability lead to a slight increase in NO_2 levels for the whole region covered by the monitoring stations, thereby counteracting the benefit of the significant decline due to emission reductions to some extent. For the period 2000-2010 the results indicate a north-south difference in this effect; in the northwest the results indicate that meteorology caused a slight decline in NO_2 levels, thereby strengthening the downward trend induced by the emission reductions, whereas for certain areas in the south meteorology caused a small increase in NO_2 counteracting the decline due to emission reductions.

Overall, the GAM method identified many more significant trends than the plain linear regression. This is as expected, since when using the GAM method the inter-annual meteorological variability is removed. In comparison, the plain linear trend that are based on uncorrected data are to some extent masked by the "noise" induced by the meteorological variations.

9. A look forward. Feasibility of a regular procedure to be run annually

The work and results given in the present report is a continuation of related work in previous years on the same topic: The issue of long-term trends and the role of inter-annual variations in meteorology vs emissions and boundary effects for the estimated trends. With the experience and methods developed through this activity the question of establishing a regular procedure for this issue is now raised. Such a procedure could typically be used on a regular basis every year to analyse last year's monitoring data with two questions in mind:

- To what extent could the concentration levels that year be explained by the meteorological conditions and by the emissions (and boundary conditions), respectively?
- How much of the trend in the concentration levels when looking at a certain time period could be explained by inter-annual variations in meteorology vs other parameters like the surface emissions?

Additionally, the experience has told us (Solberg et al., 2018) that the methods developed could also be used for a third issue:

- Could we identify time series with "odd" concentration levels and time trends possibly reflecting errors in the monitoring data?

The experience is based on application of the GAM method (Generalized Additive Model) to surface O_3 monitoring data as well as NO_2 and PM_{10} for the period 1990-2010 and gridded meteorological data from the EURODELTA Trends project. The amount of $PM_{2.5}$ data in this period turned out to be too small to make any evaluations, but if extended to more recent years, the data availability is presumably significantly increased.

The results so far have indicated that the statistical method could be applied and used to answer the three questions above to a varying degree. It is clear that the performance of the GAM method varies with geographical region so this needs to be taken into account when running a standard procedure. Next, both for O_3, NO_2 and PM_{10} there is a tendency for peak values to be underestimated, which is often seen by chemical transport model calculations as well. E. g. Watson et al. (2015) looked at the standard deviation in model predictions relative to the standard deviation of observed data and found a value of less than 1 for all models, indicating a smaller variability in model results compared to observations. They also found that the models tended to underestimate NO_2 levels and explained this by the coarse grid resolution in the models.

This effect has consequences for the statistics to be investigated. In this report we have studied daily mean values whereas air quality standards are also linked to hourly peak values (for NO_2 and O_3) or annual mean values (for NO_2 and PM). It is possible that other statistical methods (e.g. quantile regression) could be more suited for the prediction of hourly peak values, but that remains to be checked since the present work has been focussed on the GAM procedure. Furthermore, it seems the performance of the GAM method depends on the component studied with generally poorer performance for e.g. PM_{10} compared to O_3.

For a standardized procedure to be designed, one could think of various developments of the method applied so far:

- Ox = O_3 + NO_2 is in principle a more conserved quantity than O_3 and NO_2 individually, and thus it would be of interest to look at the GAM performance for Ox vs the performance for the individual species.
- So far, the method has been applied to individual monitoring sites separately. It would, however, be of interest to design a method based on spatial domains (e.g. gridded) either by merging or in other ways combining the station data prior to the statistical calculations or by aggregating the GAM results by post-processing.
- The use of more recent data (compared to the 1990-2010 period) is an obvious follow-up of the present work.
- The list of explanatory variables could be extended to include boundary conditions (e.g. hemispheric baseline levels varying with season). For PM the use of data for sea salt and dust could make an important difference if such data are available.
- Adding some kind of air mass trajectory data as input to the method is a possibility although that would significantly increase the work load.
- Finally, the question of uncertainty and in broader perspective – quality assurance – could be addressed more rigorously. One could for example construct artificial time series with known dependencies and look at how the GAM are able to reproduce these dependencies.

With the developments mentioned above, we would actually be one step closer to building a full chemical transport model (CTM). It is important to be aware that the GAM could be seen as a very simplified CTM. Although statistical models such as the GAM is fitted to match the observations at the monitoring sites, they could never be used for detailed process studies and analyses in the way today's state-of-the-art CTMs are used. The main limitation with the statistical models is that they rely on the assumptions of local relationships, i.e. that the measured levels of an atmospheric constituent (NO_2, PM, O_3) could be modelled as a function of the local conditions. This assumption is certainly not true at most sites, and the reason the statistical models anyway work (to some extent) is that the local conditions in turn are linked to large scale meteorological patterns; winter-time cold spells with low wind speeds and a shallow mixing height is typically associated with anticyclonic conditions leading to high levels of primary pollutants and so on.

The advantage of statistical models is that they are based on observational data, but that is also their main limitation, because only local effects are taken into account. Their relative simplicity makes them attractive, but they remain a model, whose performances should be compared to more comprehensive chemistry transport models.

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