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One decade of parallel PM_{10} and $PM_{2.5}$ measurements in Europe: trends and variability

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Abstract

The trends and variability of PM_{10}, $PM_{2.5}$ and PM_{coarse} concentrations at seven urban and rural background stations in five European countries for the period between 1998 and 2010 were investigated. Collocated or nearby PM measurements and meteorological observations were used in order to construct Generalized Additive Models, which model the effect of each meteorological variable on PM concentrations. In agreement with previous findings, the most important meteorological variables affecting PM concentrations were wind speed, wind direction, boundary layer depth, precipitation, temperature and number of consecutive days with synoptic weather patterns that favor high PM concentrations. Temperature has a negative relationship to $PM_{2.5}$ concentrations for low temperatures and a positive relationship for high temperatures. The stationary point of this relationship varies between 5 and 15 °C depending on the station. PM_{coarse} concentrations increase for increasing temperatures almost throughout the temperature range. Wind speed has a monotonic relationship to $PM_{2.5}$ except for one station, which exhibits a stationary point. Considering PM_{coarse}, concentrations tend to increase or stabilize for large wind speeds at most stations. It was also observed that at all stations except one, higher $PM_{2.5}$ concentrations occurred for east wind direction, compared to west wind direction. Meteorologically adjusted PM time series were produced by removing most of the PM variability due to meteorology. It was found that PM_{10} and $PM_{2.5}$ concentrations decrease at most stations. The average trends of the raw and meteorologically adjusted data are $-0.4\,\mu g\,m^{-3}\,yr^{-1}$ for PM_{10} and $PM_{2.5}$ size fractions. PM_{coarse} have much smaller trends and after averaging over all stations, no significant trend was detected at the 95 % level of confidence. It is suggested that decreasing PM_{coarse} in addition to $PM_{2.5}$ can result in a faster decrease of PM_{10} in the future. The trends of the 90th quantile of PM_{10} and $PM_{2.5}$ concentrations were examined by quantile regression in order to detect long term changes in the occurrence of very large PM concentrations. The meteorologically adjusted trends of the 90th quantile were significantly larger (as an absolute value) on average over all stations ($-0.6\,\mu g\,m^{-3}\,yr^{-1}$).

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One decade of parallel PM_{10} and $PM_{2.5}$ measurements in Europe

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1 Introduction

Airborne particles of aerodynamic diameters less than 10 µm (PM_{10}) and less than 2.5 µm ($PM_{2.5}$) have well-established adverse impacts on human health (Nel, 2005). Epidemiological (Brunekreef and Forsberg, 2005; Chang et al., 2011) and toxicological (Becker et al., 2003) evidence suggest that particles with aerodynamic diameter in the 2.5–10 µm size range (PM_{coarse}) have negative health effects too, although they have been investigated less extensively. European legislation so far has been focusing on PM_{10} and $PM_{2.5}$ particles. Fine and coarse particles have different sources, are often poorly correlated and have different health effects. This suggests that separate regulation should be considered for PM_{coarse}, in addition to existing regulation for PM_{10} and $PM_{2.5}$ (World Health Organisation, 2004; Clean Air Scientific Advisory Committee, 2010). The technical means to reduce PM_{coarse} emissions are not as developed as for PM_{10} and $PM_{2.5}$. However, some possibilities such as improving road conditions or regulating vehicle brake emissions (e.g. by using ceramic instead of metallic brake pads) do exist.

To design and implement appropriate policies for the mitigation of particulate matter air pollution, information on airborne particulate matter (hereafter referred to as PM) trends and variability is needed. PM_{10} has been measured on a regular basis in Europe since the beginning of the 1990s. This has allowed for the investigation of PM_{10} trends at certain European countries (Liu and Harrison, 2011; Hoogerbrugge et al., 2010) as well as on a pan-European scale (Colette et al., 2011). Regular $PM_{2.5}$ measurements, although a more recent development, are available from many European stations as well (Yttri et al., 2010). Decade-long parallel PM_{10} and $PM_{2.5}$ measurements at certain sites provide for the first time the opportunity to study the trends and the variability of PM_{coarse} (Liu and Harrison, 2011).

Among the most important factors influencing the trends and the variability of all gaseous and aerosol species in the atmosphere are meteorological conditions (Elminir, 2005; Zelenka, 1997; Rao et al., 1997). Therefore, proper quantification of these trends and variability requires the consideration of meteorology. Various statistical modeling methodologies have been applied to adjust the observed PM mass concentrations for the effect of meteorological variables. These include multi-linear regression (Hien et al., 2002), generalized additive models (Barmpadimos et al., 2011a) and neural networks (Hooyberghs et al., 2005). Periodic variations of concentrations with time (e.g. weekly and seasonal cycles) have to be taken into account as well. This can be done by filtering these periodic patterns before statistical modeling (Wise and Comrie, 2005), by treating each season separately (Ordonez et al., 2005), or by including additional time variables into the modeling process (Barmpadimos et al., 2011a).

The European Union (and other regulators around the world) does not only pose limits on PM concentrations in terms of average values, but also in terms of number of exceedances of a certain threshold (European Parliament and Council of the European Union, 2008). Consequently, it is important to monitor the time evolution of higher quantiles of PM concentrations in addition to the mean or the median.

The aim of this study is to investigate the trends and the variability of PM_{10}, $PM_{2.5}$ and PM_{coarse} during the 2000–2010 decade at certain European stations. Statistical modeling by means of generalized additive models is used to determine the relationship between PM and certain meteorological variables. The resulting relationships are used to adjust PM concentrations and variability for the effect of meteorology.

2 Data

PM measurements were obtained from five rural sites, which are part of the EMEP Co-operative Program for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe and from two additional European urban/suburban background sites. See Table 1 for a list of the sites and Fig. 1 for their location. All stations provide parallel PM_{10} and $PM_{2.5}$ measurements for approximately 10 yr (see Fig. 6 for the time span of the measurements). PM_{coarse} was calculated by subtracting $PM_{2.5}$ from PM_{10}. Measurements at all sites except Harwell and Bloomsbury are gravimetric according to the European standards EN-12341 for PM_{10} and EN-14907 for $PM_{2.5}$. Gravimetric PM_{10} measurements at Payerne and Basel since 2001 are obtained every fourth day and are complemented by high resolution parallel beta monitor or TEOM-FDMS measurements to obtain daily resolution (see Barmpadimos et al. (2011b) for details).

Measurements at Harwell and Bloomsbury were carried out using the TEOM method with heated (50 °C) inlets. PM_{10} data were multiplied by 1.3 as an approximate correction for losses of volatile material whereas for the $PM_{2.5}$ data only the non-volatile (at 50 °C) fraction is reported. The PM_{10} and $PM_{2.5}$ measurement methods are not considered to be equivalent to the European reference method. It is deemed however that the data are still suitable for trend analysis, under the assumption that there are no significant changes in their volatile fraction in the long-term.

The analysis of the effect of meteorology on PM concentrations requires meteorological observations. The surface observations were obtained from the weather station which was closest to the examined air quality station. The Payerne, Basel, Langenbruegge/Waldhof and Illmitz sites are collocated with meteorological stations whereas Harwell, Bloomsbury and Penausende have respective distances of 17, 2.5 and 48 km from the closest surface station with sufficient available meteorological data. The meteorological stations used are Benson RAF, London Weather Centre and Salamanca. The meteorological variables that were used from the surface stations are daily average wind speed, wind direction, temperature, relative humidity, atmospheric pressure and daily total precipitation. A further important meteorological variable for air quality applications is boundary layer depth. This was calculated using data from the closest sounding station using the simple parcel method put forward by Seibert et al. (2000). In addition, the synoptic weather conditions were taken into account by including the Hess-Brezowsky European synoptic weather regime known as *Grosswetterlage* (GWL) (Gerstengarbe et al., 1999) for each day.

PM concentrations during a certain day do not only depend on the weather conditions on the considered day but also on the recent history of weather. To account for this effect, two additional variables were constructed; the amount of precipitation of the

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previous day and the number of consecutive days with weather conditions enhancing PM. To derive the latter, two histograms with the distribution of GWL variable were constructed for each station. One histogram included all data and the second only included the days where PM concentrations belonged in the upper 20th sample quantile of the data. Then the relative difference between the two histograms was estimated. For example, if the frequency of occurrence of a certain GWL category was 0.1 in the total dataset and 0.15 in the days where concentrations belonged in the upper 20th sample quantile of the data, then the relative difference would be $(0.15-0.1)/0.1 = 0.5 = 50\,\%$. Let GWL_1 be the subset of GWL for which this difference exceeded 100 %. The count of consecutive days during which one of GWL_1 was present at the considered site forms the variable hereafter referred to as "high-PM GWL".

A further factor influencing PM concentrations is time. PM concentrations have in principle a weekly cycle, a seasonal cycle and long-term changes. These three time dependencies were represented in the analysis as additional time variables. Variables *day of the week* and *season* were included as categorical variables and variable *Julian day* (defined as the number of days since a defined date) was included as a numerical variable.

3 Methodology

3.1 Statistical model

The statistical modeling procedure described in the following is similar to the one used in (Barmpadimos et al., 2011a). Generalized Additive Models (GAMs) (Wood, 2006; Hastie and Tibshirani, 1990) were used to construct relationships between the logarithm of PM_{10} and $PM_{2.5}$ and meteorological variables. The computations were carried out using package *mgcv* (Wood, 2011) of programming language R (R Development Core Team, 2010). Logarithmic transformation was used because it improved the characteristics of the model residuals. GAMs were developed for each size (PM_{10} or $PM_{2.5}$), each season as well as for the complete yearly data for each station. This yielded a total of 70 GAMs. The relationships have the general formula

$$\ln PM_x = a + s_1(A_1) + s_2(A_2) + \ldots + b_{11}B_{11} + b_{12}B_{12} + \ldots + b_{21}B_{21} + b_{22}B_{22} + \ldots + \varepsilon \quad (1)$$

where

PM_x: PM_{10} or $PM_{2.5}$,

a: intercept,

$s_1(A_1) + s_2(A_2) + \ldots$: smooth non-parametric functions s_i of continuous covariates A_i,

$b_{11}B_{11} + b_{12}B_{12} + \ldots + b_{21}B_{21} + b_{22}B_{22}$: B_{ij} denotes categorical variables. Index i denotes the kind of categorical variable, which in this study is either day of the week or synoptic weather regime. Season is also included as a categorical variable in the yearly models. Index j denotes the category. For example, j has 7 possible values for the day of the week variable. B_{ij} is equal to 1 when the day in question is classified under category B_{ij} and 0 otherwise. b_{ij} is the corresponding coefficient;

ε: error term.

Several possibilities exist in terms of statistical modelling of the response of a variable as a function of explanatory variables. One possibility is generalized linear models, which have already been successfully used for datasets from Switzerland (Ordonez et al., 2005). GAMs were preferred over generalized linear models because they can estimate non-linear relationships between the target variable and the explanatory variables (in this case PM concentrations and meteorological variables). However, GAMs do involve the assumption that the relationship between PM and meteorological variables is additive (and after the logarithmic transformation multiplicative). Other statistical modeling methods such as neural networks are even more flexible, they require fewer assumptions than GAMs and they tend to have somewhat better predictive skill. However, the fact that they do not provide functional relationships between the target variable and the explanatory variables makes the interpretation of the results rather difficult (Venables and Ripley, 2002). In the present study we focus on the diagnosis and interpretation of PM trends rather than the prediction of PM concentrations and therefore GAMs were deemed more suitable.

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A stepwise forward variable selection algorithm was used to select the most important explanatory variables. After the addition of each variable, the Bayesian Information Criterion (BIC) for the resulting model was calculated and the addition of variables stopped when BIC was minimized. The variable selection is designed in such a way that over-fitting is avoided and relatively high percentages of the observed variance are explained by the models. If Julian day was not selected in the aforementioned process, it was added as a last explanatory variable. Julian day represents PM trends due to any influence that does not include the considered meteorological variables. By including Julian day it is ensured that the considered GAMs have a random model error without any inter-annual structure. A more detailed account of the variable selection process can be found in Barmpadimos et al. (2011a).

The smooth function of the Julian day variable of the constructed GAMs amounts to the PM trends after adjustment for the effect of meteorology. Therefore, the meteorologically adjusted trends were calculated using the relationship

$$\ln \mathrm{PM}_{x,\mathrm{adj}} = a + s(\text{Julian day}) \,. \tag{2}$$

The performance of the GAMs was evaluated using the *proportion deviance explained*. This follows the definition

$$\text{proportion deviance explained} = \frac{\text{null deviance} - \text{residual deviance}}{\text{null deviance}} \tag{3}$$

Deviance is a measure of discrepancy between the GAMs and the PM measurements. It can be interpreted in the same way as the *residual sum of squares* for ordinary linear modeling, although it is calculated differently (Wood, 2006). Small values of deviance imply better model performance. In Eq. (3), *null deviance* refers to the deviance of a model with just a constant term and *residual deviance* refers to the deviance of the fitted model. For an ideal model, proportion deviance explained (hereafter simply referred to as *deviance explained*) equals to unity.

GAM runs were also performed for yearly $\mathrm{PM}_{\mathrm{coarse}}$ data in order to estimate relationships between $\mathrm{PM}_{\mathrm{coarse}}$ and meteorological variables (Sect. 4.1). However, the

PM_{coarse} model runs had relatively low (about half) deviance explained compared to the PM_{10} and $PM_{2.5}$ runs (see Sect. 4.2). That is mostly because PM_{coarse} values are obtained indirectly by subtracting PM_{10} and $PM_{2.5}$ measurements and therefore have larger uncertainty. For the trend analysis in Sect. 4.3 adjusted PM_{coarse} were simply obtained as the difference between adjusted PM_{10} minus adjusted $PM_{2.5}$.

3.2 Very large values

The evolution of very large values is investigated by quantile regression (Koenker and Bassett, 1978). Let Y be a random variable with distribution function $F(y) = P(Y \leq y)$. The τ_{th} quantile of Y is defined as $Q(\tau) = F^{-1}(\tau) = \inf\{y : F(y) \geq \tau\}$. The best-known example is $Q(0.5)$, which is the median. Assume an independent variable X. The conditional τ_{th} quantile of Y given X is $Q_{Y|X}(\tau)$. Let $Q_{Y|X}(\tau)$ be a linear function of X according to equation $Q_{Y|X}(\tau) = X'\beta(\tau)$, where X' is the model matrix and $\beta(\tau)$ the vector containing the unknown model parameters. Parameter estimates $\hat{\beta}(\tau)$ can be obtained by solving

$$\begin{aligned}\hat{\beta}(\tau) &= \underset{\beta \in \mathbf{R}^p}{\arg\min} \sum_{i=1}^{n} \rho_\tau(y_i - x_i'\beta) \\ &= \underset{\beta \in \mathbf{R}^p}{\arg\min} \left[(\tau - 1) \sum_{y_i - x_i'\beta < 0} (y_i - x_i'\beta) + \tau \sum_{y_i - x_i'\beta > 0} (y_i - x_i'\beta) \right], \qquad (4)\end{aligned}$$

where ρ_τ termed the *loss function* is given by $\rho_\tau(y) = u(\tau - I(y < 0))$ and I is the *indicator function.* The idea behind the estimation of the linear parameters $\hat{\beta}(\tau)$ is that one changes the values of β until the quantity in the square brackets is minimized. The quantity in the squares brackets in turn, represents the "distance" of the points on a straight line with parameters β from points y_i. However, the distance is weighted according to the selection of quantile τ by quantities τ and $\tau - 1$. In practice, the calculations are done using principles of linear programming. Note that the problem is

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formulated in a similar fashion to ordinary least squares, except that the square function in the sum of Eq. (4) has been replaced by the loss function.

Fitting to the PM time series a straight line by quantile regression is in some ways similar to ordinary least squares (OLS) regression. By means of an OLS regression, a line that represents the mean value is fitted to the data, whereas by means of quantile regression a line that represents a certain quantile is fitted instead. We consider the 90th sample quantile of the data as an indicator of the magnitude of the upper portion of PM ambient concentrations, excluding extreme and relatively rare events and we examine the trend of the 90th quantile with time. Quantile regression computations were carried out using package *quantreg* (Koenker, 2011) of R programming language.

4 Results

4.1 Important explanatory variables

The contribution of each explanatory variable to the total modeled PM concentrations can be expressed as an additive factor using Eq. (1). By exponentiation of this relationship, the additive factors on the right hand side of Eq. (1) become multiplicative factors (hereafter referred to as "PM factor"), which contribute to an increase of PM if greater than 1 and a decrease if less than 1. In the following, the relationship of PM factors to PM will be discussed.

Table 2 shows the most frequently chosen explanatory variables for each season. These results refer to the $PM_{2.5}$ GAM runs. The results of the PM_{10} GAM runs were similar both in terms of the selected explanatory variables and in terms of the relationships between PM and each explanatory variable. The most prominent explanatory variables are convective boundary layer depth, wind speed, wind direction and temperature and they appear in all seasons. The selected variables did not vary considerably between different sites.

While some variables, such as convective boundary layer depth, have a monotonic relationship with $PM_{2.5}$, some others have relationships with stationary points. Temperature has a negative relationship with $PM_{2.5}$ in winter and a positive relationship in summer (Fig. 2, left). The winter relationship of $PM_{2.5}$ with temperature can be indirectly explained by the fact that space heating emissions are larger in that season. Space heating by wood burning has been shown to have a large influence in winter aerosol concentrations in Switzerland (Szidat et al., 2007; Sandradewi et al., 2008). The PM_{coarse} factors have a positive relationship with temperature. This could be attributed to the fact that higher temperatures are often associated with drier soil conditions, which in turn can lead to enhanced dust resuspension (Vardoulakis and Kassomenos, 2008). Moreover, primary biological PM_{coarse} emissions of pollen are likely enhanced at higher temperatures. The negative relationship between the $PM_{2.5}$ factor and temperature was observed for the winter $PM_{2.5}$ model runs at the continental sites of Central Europe (Basel, Payerne, Illmitz and Langenbruegge). At the remaining sites, temperature was either not selected as an explanatory variable (Bloomsbury and Penausende), or it was selected but it did not exhibit a negative relationship with $PM_{2.5}$ (Harwell). In contrast, the positive relationship between $PM_{2.5}$ and temperature in summer can be attributed to fast production of secondary aerosols that happens with high solar radiation coincident with high temperatures (Barmpadimos et al., 2011a). As shown in Fig. 2 (left), the stationary point of this relationship varies between 5 and 15 °C depending on the station. PM_{10} factors (not shown) are similar to the $PM_{2.5}$ factors.

A further explanatory variable that can exhibit stationary points is wind speed. The relationship between PM and wind speed involves dilution, resuspension and production of marine aerosol. The latter process is highly relevant for the UK sites (Harwell and Bloomsbury) (Jones et al., 2010). For low wind speeds dilution is the dominant process and thus PM concentrations have a negative relationship with wind speed. For high wind speeds resuspension of soil material and production of marine aerosol becomes more important and the PM vs. wind speed relationship is positive. This is particularly true for PM_{coarse}. PM resuspension depends on the soil condition and wind

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speed (Gillette and Passi, 1988). In addition, the effect of marine aerosol depends on the location of the site and wind speed. Therefore, the position of stationary points in the relationship between PM and wind speed varies from site to site. Figure 3 shows the $PM_{2.5}$ and PM_{coarse} wind speed factors for all sites. The Bloomsbury and Harwell sites exhibit stationary points for PM_{coarse} concentrations when wind speed is 3.5 and 3.9 m s^{-1}, respectively. These values are in line with Harrison et al. (2001) who report that the stationary point for the relationship between PM_{coarse} and wind speed is at approximately 3.8 m s^{-1} at an urban background site in Birmingham, UK. The Bloomsbury and Harwell sites also have higher average wind speeds (3.5 and 3.4 m s^{-1}, respectively) compared to all other sites whose average wind speeds range between 2.0 and 2.5 m s^{-1}. The $PM_{2.5}$ relationship for Bloomsbury has no stationary point whereas for Harwell the stationary point is at 5.7 m s^{-1}. Jones at al. (2010) have identified chloride ions from marine aerosol as the PM component with a positive relationship with wind speed at Harwell. The absence of a stationary point or the requirement of higher wind speed for one to occur for $PM_{2.5}$ is the result of the fact that most of the soil and marine aerosol are in the PM_{coarse} fraction. Querol et al. (2004) estimated from measurements at EMEP sites in Spain that mineral dust accounts for 8–21 % of the total $PM_{2.5}$ mass. The sites at Basel, Payerne and Penausende exhibit negative monotonic relationships of PM_{coarse} with wind speed. The same relationship for Illmitz becomes approximately constant for large wind speeds. At Langenbruegge/Waldhof an almost constant relationship for all available wind speeds was found. However, the behavior of PM_{coarse} concentrations for high wind speed at this site could not be identified because the maximum wind speed was only 6.1 m s^{-1}. The position of the stationary point is probably affected by the emission mechanism of PM_{coarse} too. For example, traffic-induced turbulence enhances resuspension of PM_{coarse}. Barmpadimos et al. (2011b) report that ambient PM_{coarse} in an urban background location (Zurich, Switzerland) mostly originate from traffic.

Wind direction is one of the most important explanatory variables. Although its relationship to PM depends on the site, some common patterns among different sites could

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be identified. Wind direction cannot substitute more comprehensive methodologies, such as trajectory models for the spatial identification of pollution sources, especially at large distances from the site of interest. It is deemed however that some preliminary conclusions can be drawn from long-term wind direction observations from meteorologically representative locations. The $PM_{2.5}$ and PM_{coarse} factors for wind direction are shown in Fig. 4. The Illmitz site shows considerably higher $PM_{2.5}$ (and PM_{coarse}) concentrations for east wind direction compared to west. This indicates that air masses coming from the west tend to be cleaner than air masses coming from the east, either because of their maritime origin or because of lower levels of air pollution in Western Europe compared to Eastern Europe (see also discussion for Illmitz in Sect. 4.3). A similar wind direction response function was found at the Harwell, Bloomsbury and Penausende sites and a similar distinction can be made between maritime clean air masses from the west vs. continental polluted air masses from the east. The response function for $PM_{2.5}$ for the Payerne site does not allow discerning a very clear pattern. The Payerne site is located in the Swiss Plateau and surrounded by the Alps on the East and Jura mountains on the West. It is hypothesized that long-range transport at that site is largely altered by topography. A similar consideration could apply to the Basel site, which has a local maximum for south-west wind direction for the $PM_{2.5}$ component but not for the PM_{coarse} component. The PM factors for wind direction do not only depend on long-range transport but they can also be affected by local sources. For example, the local maxima observed for north-west and north wind direction at the Basel and Illmitz sites, respectively are possibly attributed to the influence of the cities of Basel and Vienna, which are located north of the measurement stations.

Considering the remaining meteorological variables, their relationship to PM is similar for different size fractions, different sites and different seasons. Variables "GWL" "wind direction" and "relative humidity" are an exception as they depend strongly on the site. A further investigation of the relationships between PM and various meteorological variables can be found in Barmpadimos et al. (2011a).

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4.2 Model performance

Table 3 shows some important statistical quantities for the evaluation of the GAMs performance for the $PM_{2.5}$ runs. The number of covariates averaged over all stations was slightly above 7 for all seasons and 10.0 for the yearly dataset. Deviance explained (Eq. 3) ranges between 50 and 65 % depending on the season. GAM performance is somewhat poorer for spring and autumn, which indicates some difficulty in modeling the effect of meteorology on PM during transitional periods. Model performance in terms of deviance explained is considered to be adequate. GAMs developed by Pearce et al. (2011) to investigate the effect of meteorology on air quality in Melbourne, Australia, explained 21.1 % of the observed PM_{10} variability. In the study of Aldrin and Haff (2005), GAMs were used for the same purpose for a number of sites at Oslo, Norway. These models could explain between 48 and 80 % of the observed variability.

The GAM performance had relatively small variations between different stations (Table 4). The stations located in Switzerland and Austria (Basel, Payerne and Illmitz) have somewhat larger deviance explained and number of covariates than the other stations. Deviance explained is somewhat low for the sites Harwell and Penausende, which do not have collocated meteorological data.

4.3 Trends of PM

The linear PM trends were quantified using ordinary least squares (OLS) regression of the PM daily concentrations versus time. This was done for the raw PM data and for PM values adjusted for meteorology (Eq. 2). The resulting slopes expressed daily changes and they were multiplied by 365 to represent yearly changes. The confidence interval of the slopes has been calculated using the t-statistic (Yan and Su, 2009). Figure 5 shows a summary of all the slopes for all stations using the full year $PM_{2.5}$ (left) and PM_{coarse} (right) data. Note that PM trends after adjustment for meteorology have narrower confidence intervals than the trends of the measured data. Therefore, the adjusted data enable the detection of small trends with shorter time series. In order to get an overview of the yearly variation of $PM_{2.5}$, the yearly median time series for all

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sites is shown in Fig. 6. In addition, the slopes of the raw and adjusted time series of PM_{10}, $PM_{2.5}$, PM_{coarse} and the PM_{coarse}/PM_{10} ratio are given in Table 5.

In general, $PM_{2.5}$ concentrations have a small decreasing trend in the last decade. Most stations have a decreasing trend and the average trend is $-0.4\,\mu g\,m^{-3}\,yr^{-1}$ for both the raw and adjusted data. The rural background station at Harwell is an exception as concentrations there have no significant trend. Despite the absence of a significant trend at the Harwell site, the Bloomsbury urban site located in London has a decreasing trend ($-0.3\,\mu g\,m^{-3}\,yr^{-1}$), which indicates some reduction in the urban emissions of $PM_{2.5}$ and its precursors. The changes at Bloomsbury did not affect the Harwell site, which is located mostly upwind (about 80 km west) of Bloomsbury. Note that at the end of the 1990 decade and the beginning of the 2000 decade Bloomsbury had considerably greater $PM_{2.5}$ concentrations than Harwell. However, $PM_{2.5}$ concentrations at the two sites tend to converge towards the end of the 2000 decade (Fig. 6). A further pair of stations from the same country is the suburban background site at Basel and the rural background site Payerne in Switzerland. The difference in the trends between the two sites is very small. This is in agreement with the findings for PM_{10} of Barmpadimos et al. (2011a). In addition, the site at Basel is located at the outskirts of a relatively small city (with a population of about 170 000) far from the immediate influence of traffic. Therefore, trends at this site are not directly affected by changes in traffic although a regional influence from a decrease in traffic emissions does exist (Barmpadimos et al., 2011a).

The largest $PM_{2.5}$ changes ($-1.0\,\mu g\,m^{-3}\,yr^{-1}$) are observed at the rural background site Illmitz in Austria. According to Spangl and Nagl (2010), high levels of PM are associated with high-pressure weather systems over Eastern Europe in winter that on one hand lead to relatively stagnant weather conditions and on the other hand to transport of relatively polluted continental air masses from Eastern Europe to Austria. Conversely, low-pressure weather systems in Western and Northwestern Europe during winter facilitate transport of relatively clean air masses from Western Europe and the Atlantic and are associated with frequent fronts, which remove effectively airborne PM.

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The wind direction response function plotted in Fig. 4 supports this hypothesis, since wind directions between 190° and 300° are associated with considerably lower $PM_{2.5}$ concentrations that wind directions between 340° and 150°. In the same report, both low PM_{10} concentrations observed in 2004, 2007, 2008 and 2009 and high PM_{10} concentrations observed in 2003 and 2006 are mostly attributed to weather conditions. The $PM_{2.5}$ time series has similar features. Indeed, the meteorologically adjusted $PM_{2.5}$ trend is considerably reduced (in absolute terms) to $-0.6\,\mu g\,m^{-3}\,yr^{-1}$ and the meteorologically adjusted values for Illmitz have less year-to-year variability (Fig. 6). The Hess-Brezowsky European synoptic weather regimes which are associated with high levels of $PM_{2.5}$ and which were used for the construction of the "high-PM GWL" variable mentioned in Sect. 2 are WW, NWA, NZ, HNZ, TB and HNFZ (see Gerstengarbe et al. (1999) for an explanation of the weather regimes). Figure 7 shows how many days one of these weather regimes occurred each year. Comparison of Fig. 7 and the Illmitz yearly medians in Fig. 6 shows that GAMs tend to correct upwards $PM_{2.5}$ concentrations in years with low occurrence (e.g. less than 45 times) of one of the aforementioned weather regimes whereas the opposite happens in years 2002, 2003 and 2006, where these weather regimes occur frequently.

As seen in Fig. 5, another site with considerable meteorological adjustment is Payerne. An examination of the trends for each season at this site (not shown) shows that all seasons have a negligible meteorological adjustment except the winter data whose raw slope is not significantly different from zero whereas the adjusted slope is $-0.3\,\mu g\,m^{-3}\,yr^{-1}$. This is shown to be the case at a number of sites in the Swiss plateau (Barmpadimos et al., 2011a). Figure 8 shows the winter and summer yearly median values for Payerne. The year-to-year variability of the winter raw data is reduced heavily after the meteorological adjustment. One of the largest adjustments occurs in winter 2003, which had very high concentrations of PM in Switzerland. The summer of the same year had high levels of air pollution in large parts of Western Europe and this is reflected in the Payerne and Illmitz measurements. The GAMs for Payerne and Illmitz adjust substantially the summer data for 2003 as well.

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Annual mean concentrations at Penausende, Spain decrease at a rate of $-0.4\,\mu g\,m^{-3}\,yr^{-1}$. Although this slope is not particularly large compared to the other stations, it represents a considerable decrease for a station with average $PM_{2.5}$ concentrations of $8\,\mu g\,m^{-3}$ (Table 1). Dividing the slope by the grand average yields an annual decrease of about 5 % or a decrease by 45 % over the 9 yr of available data. Saharan dust episodes play an important role in the total amount and the variations of PM loads in Spain and mineral dust has a moderate contribution of an estimated 8–21 % of the total $PM_{2.5}$ mass (Querol et al., 2004). These episodes however have not been identified as a significant contribution to the annual PM_{10} concentrations at sites in Northern Iberia, where Penausende is also located (Fig. 1) (Querol et al., 2008). Although the number of occurrences of such episodes has seen a substantial increase in Penausende in the last decade (Querol et al., 2009), the $PM_{2.5}$ concentrations were decreasing. Pérez et al. (2008) also found a fast decrease of PM_{10} and $PM_{2.5}$ at a regional background site in Northeastern Spain (Montseny). As pointed out in the same study, the observed decrease is the result of a number of factors, which however are difficult to identify and it is suggested that both meteorology and anthropogenic emissions are possible major influences. In the present study the meteorological adjustment for Penausende does not change significantly the observed trends. Therefore we conclude that a decrease in anthropogenic emissions is more important as a driving factor for the observed decrease than meteorology. Penausende is an elevated background site and as such it is affected considerably by long-range transport. Thus, the observed decrease possibly reflects a decrease in background $PM_{2.5}$ concentrations in Spain in general and possibly in other nearby European and North-African countries. Like Penausende, all continental European sites used in this study show a decreasing trend (Fig. 5).

Langenbruegge/Waldhof has, in absolute terms, a considerably lower $PM_{2.5}$ trend than all other continental European sites ($-0.1\,\mu g\,m^{-3}\,yr^{-1}$). Given that 65 % of the area at the site is covered by coniferous forest and 30 % is covered by farmland, the observed trend can be the result of changes in anthropogenic and/or biogenic emissions.

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Biological emissions of primary aerosol and emissions of biogenic volatile organic compounds (which act as secondary organic aerosol precursors) are large during summer and small or negligible during winter (Karl et al., 2009; Winiwarter et al., 2009). Considering the winter raw and meteorologically adjusted trends at the site, when the influence of primary biological and biogenic secondary aerosol on the total measured PM mass is minimal, no significant trend is identified. Therefore, no considerable reduction of background aerosol of anthropogenic origin seems to have taken place in the area during the winter season. The small decreasing trend in the yearly data is mainly the result of a decrease of $PM_{2.5}$ in summer ($-0.2\,\mu g\,m^{-3}\,yr^{-1}$), although it is unclear to what extent this decrease is of anthropogenic or natural origin. If there is any natural contribution to the $PM_{2.5}$ summer trend this would be secondary organic aerosol, which is in the $PM_{2.5}$ size range. That is because the trend of the adjusted PM_{coarse} in summer is slightly increasing ($0.1\,\mu g\,m^{-3}\,yr^{-1}$). This is also the case for all other seasons except spring, which has no significant trend. The uncertainty in quantifying the role of natural emissions arises from the fact that the contribution of primary biological and biogenic secondary aerosol to the local PM concentrations is unknown. The contribution of primary biological aerosol to ambient PM_{10} on a European level is an estimated 2–3 % (Winiwarter et al., 2009) but this value is expected to be considerably larger at forest sites (Yttri et al., 2011). Production of biogenic precursors of secondary organic aerosol in Northern Germany is also relatively low (Karl et al., 2009) but this does not rule out locally large influences in the proximity of forests. Things are further complicated by the fact that biogenic secondary organic aerosols do not only depend on emissions of biogenic volatile organic compounds but also by anthropogenic emissions of NO_x, SO_x, NH_3, reactive non-methane carbon and primary carbonaceous particulate matter, which react with biogenic emissions to give biogenic secondary organic aerosol (Carlton et al., 2010).

Further insight into the causes of the observed PM trends can be gained by examining trends of certain PM fractions. Long term speciated measurements of PM are still uncommon. Regarding the sites used in this study, decade-long time series of

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sulfate are available for Illmitz, Payerne and Penausende (Fig. 9). The average sulfate contribution to $PM_{2.5}$ is 16 %, 15 % and 24 % for Illmitz, Payerne and Penausende, respectively. The average sulfate concentrations at the same sites are 3.1, 2.2 and 1.8 $\mu g\,m^{-3}$. The $SO_4^{2-}/PM_{2.5}$ ratio has no considerable changes at any site. The SO_4^{2-} concentrations at Payerne and Illmitz show a small decrease of $-0.02\,\mu g\,m^{-3}\,yr^{-1}$. This can be attributed to a small decrease in the European sulfur emissions (1–2 % yr^{-1}) in the 2000 decade (Monks et al., 2009). Gianini et al. (2011) also report a decrease of sulfate concentrations for the 1998/1999–2008/2009 period at Payerne. Yearly average ambient SO_4^{2-} concentrations have considerable year-to-year variability and the overall trend appears to be mainly the result of a relatively large decline in years 2006 and 2007. At Penausende SO_4^{2-} concentrations have less variability and exhibit a consistent decline in the last decade ($-0.05\,\mu g\,m^{-3}\,yr^{-1}$). From the above one can conclude that sulfate concentrations have contributed to the observed $PM_{2.5}$ decrease to a small extent at Payerne and Illmitz and to a larger extent at Penausende.

Differences between summer and winter PM trends are observed not only at Langenbruegge/Waldhof but at most sites. These differences however become insignificant after meteorological adjustment except the Langenbruegge/Waldhof and the Payerne sites, where the differences persist after the meteorological adjustment. The respective trends at Langenbruegge/Waldhof are $+0.1\,\mu g\,m^{-3}\,yr^{-1}$ (non-significant) and $-0.2\,\mu g\,m^{-3}\,yr^{-1}$ for winter and summer while at Payerne are -0.3 and $-0.5\,\mu g\,m^{-3}\,yr^{-1}$ for the same seasons. The site at Basel, which is a suburban background station also located in Switzerland has no significant difference between the winter and summer adjusted trends. Payerne is mostly surrounded by farmland. It can therefore be hypothesized that the larger summer changes in Payerne are the result of larger summer decrease in agricultural activities and/or natural biogenic emissions. Langenbruegge/Waldhof also has considerable agricultural local and regional emissions, in addition to forest emissions discussed in the previous paragraph.

The slopes of the PM_{coarse} raw and adjusted data are shown on the right panel of Fig. 5. These slopes are rather small and the average over all stations is zero. The failure to reduce PM_{coarse} is attributed to the fact that, unlike PM_{10} and $PM_{2.5}$, PM_{coarse} is not explicitly regulated in any European country. The fact that $PM_{2.5}$ concentrations decrease while PM_{coarse} concentrations do not, implies that the PM_{coarse} fraction of PM_{10} increases. Indeed, as shown in Table 5, most sites exhibit a small but significant increase of the PM_{coarse}/PM_{10} fraction. At the Payerne site, the observed meteorologically adjusted increase of 1.7 % yr^{-1} is considerably larger than the average of 0.6 % yr^{-1}. Given the rural location of this site, it is hypothesized that the increase of PM_{coarse} and PM_{coarse}/PM_{10} fraction at that site is due to increased agricultural PM_{coarse} emissions in the area. These emissions are deemed to be rather local because the other site in the swiss site, Basel, does not show any considerable increasing trend in the PM_{coarse} fraction. Table 5 also shows the PM_{10} trends. Since PM_{coarse} trends are rather small, the largest part of PM_{10} trends is attributed to the $PM_{2.5}$ trends.

4.4 Variability of PM

The variability of PM and in particular, the long-term changes of very large values is examined by calculating the slope of 90th quantile regression line. The 90th quantile slopes for all size fractions and stations are summarized in Table 6. The slopes of the raw 90th quantile data are negative and larger in absolute terms than their OLS regression counterparts for all stations except Harwell (see Tables 5 and 6). Nevertheless, the differences do not seem to be significant as their 95 % confidence intervals largely overlap (not shown). Penausende is an exception because the 90th quantile raw slope is significantly lower than the OLS slope.

Considering the slopes of the 90th quantile of the $PM_{2.5}$ adjusted data, they are significantly lower than the OLS $PM_{2.5}$ slopes at Illmitz and Penausende. This is not found to be the case for PM_{coarse}. The daily data with the OLS, the 50th quantile and the 90th quantile regression lines are shown in Fig. 10. The meteorological adjustment decreases the absolute value of the 90th quantile slopes for Illmitz and Penausende (Table 6), which indicates that changes in local weather and transport patterns are responsible to some extent for the observed changes of very large concentrations.

However, the confidence intervals of the 90th quantile slopes are rather large and the difference between the raw and adjusted values is not statistically significant. Therefore, the degree to which long range transport, local meteorology and regional emissions contributed to these changes is difficult to quantify.

5 Conclusions

The trends and variability of PM_{10}, $PM_{2.5}$ and PM_{coarse} at seven European sites were investigated. Statistical modeling by means of Generalized Additive Models was used to estimate the effect of several meteorological variables to PM concentrations and estimate PM concentrations adjusted for the effect of meteorology. The estimated relationships between PM and meteorology were reasonable and consistent with previous results (Barmpadimos et al., 2011a). The most important meteorological variables affecting PM concentrations were boundary layer depth, wind speed, wind direction, temperature, precipitation and synoptic weather pattern (represented by the "high-PM GWL" variable). The meteorologically adjusted PM concentrations had much less variability than the original data. The available meteorological and time variables could explain between 50 and 65 % of the null deviance, depending on the season.

PM_{10} and $PM_{2.5}$ trends are decreasing at most sites and on average over all sites ($-0.4\,\mu g\,m^{-3}\,yr^{-1}$ for both size fractions). PM_{coarse} have small trends of mixed signs at different sites and not significantly different from zero on average over all sites. Therefore, the observed decrease in PM_{10} is mostly attributed to the decrease of $PM_{2.5}$ concentrations. The effect of the meteorological adjustment varies between stations. However, PM trends were significantly negative after the meteorological adjustment at all sites, except Harwell. This indicates that the PM_{10} and $PM_{2.5}$ have reduced considerably in the previous decade because of non-meteorological factors. This decrease is present at all seasons ($-0.3\,\mu g\,m^{-3}\,yr^{-1}$ in autumn and winter and $-0.4\,\mu g\,m^{-3}\,yr^{-1}$ in spring and summer). The decrease of the mean PM concentrations is followed by a decrease in very large values as represented by the 90th sample quantiles. At two sites

(Illmitz and Penausende) the decrease of the meteorologically adjusted 90th quantiles is considerably faster than the decrease in the average. Further research is required to identify what changes in the emissions or possibly unaccounted for meteorological processes lead to the reduced variability.

Although PM_{10} concentrations decrease in the 2000 decade, the rate of reduction is slower compared to the 1990 decade (Barmpadimos et al., 2011a) and it does not correspond to the decrease in the emissions in Europe (Harrison et al., 2008). A number of possible explanations have been suggested for this (Harrison et al., 2008). The evidence put forward in this study supports the conclusion that meteorological conditions have not changed in favor of higher levels of PM, with the exception of Payerne for $PM_{2.5}$ (Fig. 5). It was also shown that $PM_{2.5}$ and PM_{coarse} play different roles in the development of PM_{10} trends: $PM_{2.5}$ decreases at most European sites, whereas PM_{coarse} does not. This also implies that the PM_{coarse} fraction in PM_{10} increases, the rate of increase being 0.6 % per year on average over all stations. Therefore, in order to keep reducing effectively PM_{10} in the future, air pollution abatement strategies should not only target $PM_{2.5}$ but also PM_{coarse}.

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Table 1. List of considered air quality sites. PM_{10} and $PM_{2.5}$ values are averaged over all available data.

Code	Name	Country	Altitude (m)	Type	Sampling	Mean PM_{10} ($\mu g\,m^{-3}$)	Mean $PM_{2.5}$ ($\mu g\,m^{-3}$)
BAS	Basel	Switzerland	365	Suburban background	Gravimetric	22	17
BLO	Bloomsbury	UK	20	Urban background	TEOM	28	14
HAR	Harwell	UK	137	Rural background	TEOM	19	11
ILL	Illmitz	Austria	117	Rural background	Gravimetric	25	20
LAN	Langenbruegge/ Waldhof	Germany	74	Rural background	Gravimetric	17	13
PAY	Payerne	Switzerland	489	Rural background	Gravimetric	20	17
PEN	Penausende	Spain	985	Rural background	Gravimetric	12	8

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Table 2. List of explanatory variables selected in GAMs for $PM_{2.5}$. Explanatory variables that were chosen for at least five (out of seven) stations are listed. Positive signs (+) next to variable names indicate a positive relationship between $PM_{2.5}$ and the explanatory variable, whereas negative signs (–) represent the opposite. Use of both signs (+/–) indicates relationships with turning points or variables whose behavior depends on the station. More frequently chosen variables are displayed first.

Winter	Spring	Summer	Autumn	Year
CBL depth (–)	CBL depth (–)	Wind speed (–)	CBL depth (–)	CBL depth (–)
Wind direction (+/–)	Wind direction (+/–)	Julian day (+/–)	Wind speed (–)	Wind direction (+/–)
Wind speed (–)	Wind speed (–)	Temperature (+)	Temperature (+)	Wind speed (–)
Precipitation (–)	Julian day (+/–)	CBL depth (–)	Wind direction (+/–)	Season
High-PM GWL (+)	Precipitation (–)	High-PM GWL (+)	High-PM GWL (+)	Temperature (+)
Pressure (+)	Temperature (+)	Wind direction (+/–)	Precipitation (–)	Julian day (+/–)
Temperature (–)	Previous-day Precipitation (–)		Relative humidity (+/–)	Precipitation (–)
				Previous-day Precipitation (–)

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Table 3. Deviance explained (%) and number of covariates averaged over all stations for each season. Numbers in parentheses indicate the minimum and the maximum.

	Deviance explained	Number of covariates
Spring	50 (28, 57)	7.3 (5, 8)
Summer	65 (61, 74)	7.1 (4, 9)
Autumn	57 (49, 68)	7.3 (6, 9)
Winter	60 (48, 75)	7.6 (6, 9)
Year	58 (49, 69)	10.0 (8, 11)

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Table 4. Deviance explained (%) and number of covariates averaged over all seasonal and yearly values for each station. Numbers in parentheses indicate the minimum and the maximum.

	Deviance explained	Number of covariates
Basel	67 (54, 75)	9.0 (8, 11)
Bloomsbury	58 (55, 63)	6.4 (5, 10)
Harwell	53 (49, 61)	7.8 (6, 9)
Illmitz	59 (48, 69)	8.2 (6, 10)
Langenbruegge/Waldhof	50 (28, 59)	7.2 (5, 11)
Payerne	67 (57, 74)	8.4 (7, 11)
Penausende	53 (50, 62)	6.0 (4, 8)

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Table 5. Raw and meteorologically adjusted trends of PM_{10}, $PM_{2.5}$, PM_{coarse} and the PM_{coarse} / PM_{10} ratio at all sites. The PM trends are given in $\mu g\,m^{-3}\,yr^{-1}$ and the PM_{coarse}/PM_{10} ratio trends are given in $\%\,yr^{-1}$. Trends whose 95 % confidence intervals do not overlap with zero are given in bold. A graphical depiction of the 95 % confidence interval of the $PM_{2.5}$ and PM_{coarse} trends is provided in Fig. 4. The last row is the average over all different stations.

	PM_{10}		$PM_{2.5}$		PM_{coarse}		PM_{coarse}/PM_{10}	
	Raw	Adj.	Raw	Adj.	Raw	Adj.	Raw	Adj.
BAS	**–0.5**	**–0.6**	**–0.4**	**–0.5**	**–0.08**	**0.03**	**0.4**	**0.5**
BLO	–0.2	**–0.3**	**–0.3**	**–0.4**	**0.2**	0.07	**0.9**	**0.9**
HAR	**0.2**	**0.2**	–0.04	**0.1**	**0.1**	**0.1**	0.2	0.1
ILL	**–1.3**	**–0.9**	**–1.0**	**–0.6**	**–0.2**	**–0.2**	**0.3**	–0.1
LAN	–0.1	0.04	**–0.1**	**–0.1**	**–0.06**	**0.07**	0.01	**0.7**
PAY	**–0.4**	**–0.4**	**–0.4**	**–0.6**	**0.1**	**0.3**	**1.3**	**1.7**
PEN	**–0.6**	**–0.5**	**–0.4**	**–0.4**	**–0.1**	**–0.1**	**0.9**	**0.7**
Aver.	**–0.4**	**–0.4**	**–0.4**	**–0.4**	–0.01	0.02	**0.6**	**0.6**

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Table 6. Raw and meteorologically adjusted yearly PM trends of the 90th quantile in $\mu g\,m^{-3}\,yr^{-1}$. Numbers in bold font indicate trends whose 95 % confidence interval does not include zero. Numbers in italic font indicate trends that are significantly different than the corresponding OLS trends in Table 5 at the 95 % level of confidence.

	PM_{10}		$PM_{2.5}$		PM_{coarse}	
	Raw	Adj.	Raw	Adj.	Raw	Adj.
BAS	**–0.8**	**–0.8**	**–0.4**	**–0.7**	***–0.2***	***–0.1***
BLO	–0.2	**–0.6**	**–0.4**	**–0.5**	0.0	–0.1
HAR	0.0	**0.2**	–0.1	–0.1	0.1	0.2
ILL	**–2.3**	***–1.6***	**–1.9**	***–1.4***	**–0.2**	**–0.4**
LAN	–0.2	–0.1	–0.1	–0.2	**–0.1**	0.0
PAY	***–0.8***	**–0.6**	**–0.4**	**–0.7**	**0.2**	**0.4**
PEN	***–1.4***	***–1.1***	***–0.9***	***–0.7***	**–0.5**	**–0.2**
Aver.	***–0.8***	***–0.6***	**–0.6**	***–0.6***	–0.1	0.0

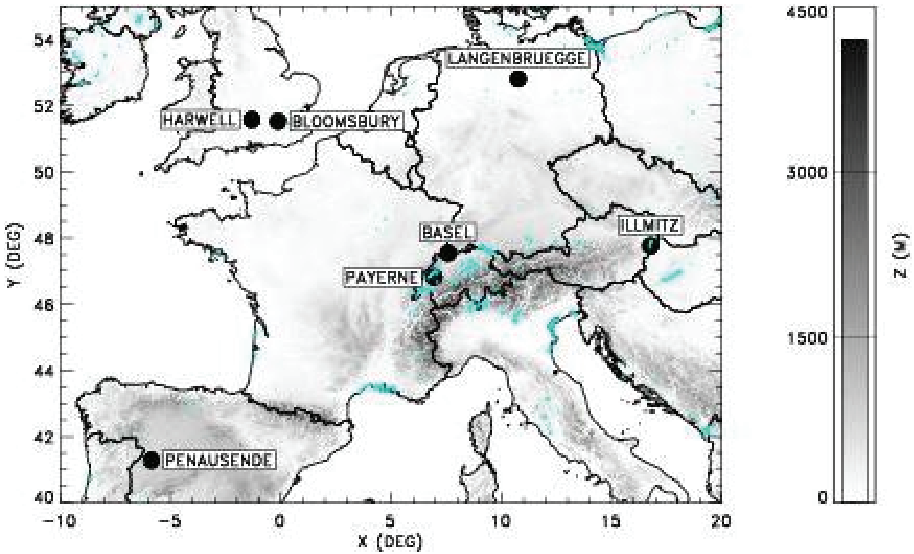

Fig. 1. Locations of the sites used in this study. Sidebar indicates altitude in meters.

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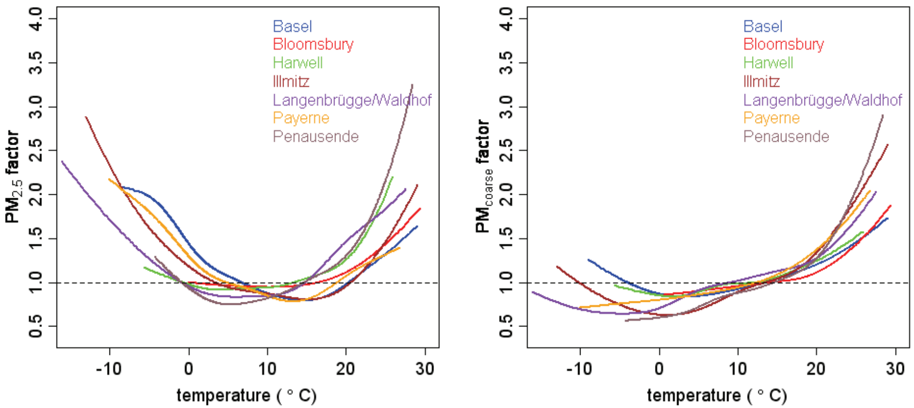

Fig. 2. $PM_{2.5}$ (left) and PM_{coarse} (right) factors for temperature at all sites.

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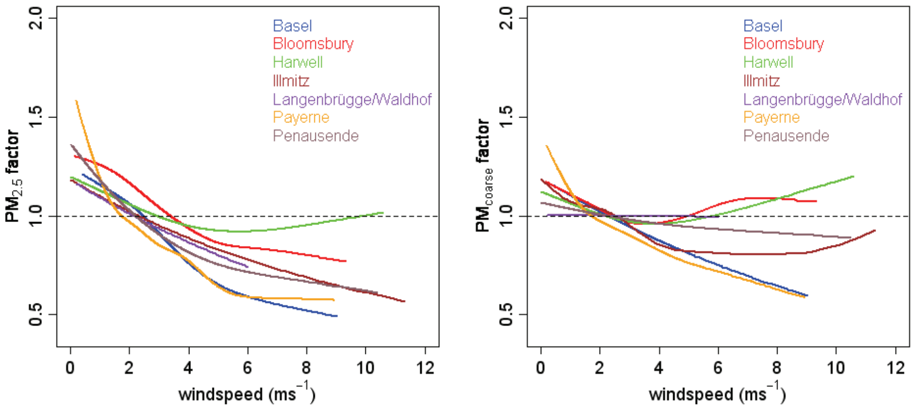

Fig. 3. $PM_{2.5}$ (left) and PM_{coarse} (right) factors for wind speed at all sites.

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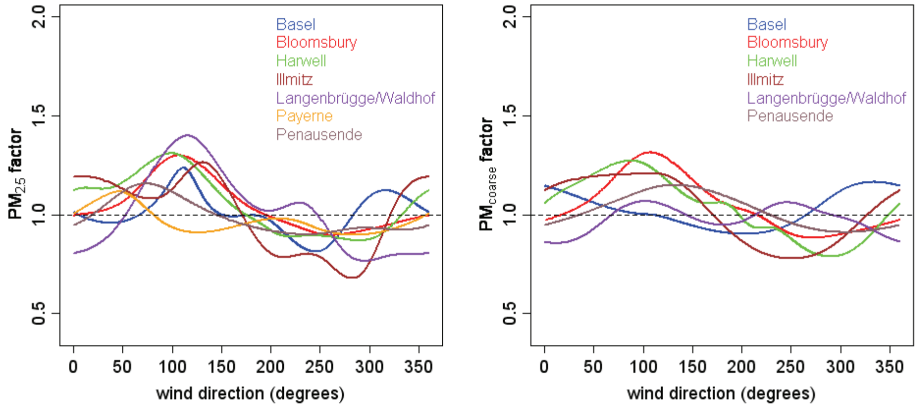

Fig. 4. $PM_{2.5}$ (left) and PM_{coarse} (right) factors for wind direction at all sites. Payerne is missing from the PM_{coarse} factor plot because wind direction was not selected for that site.

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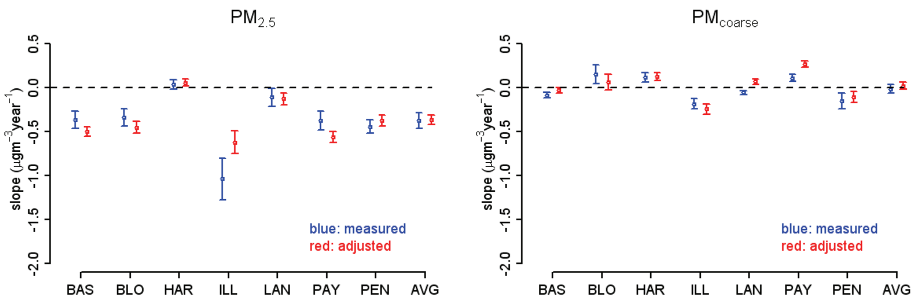

Fig. 5. Linear regression slopes and 95 % confidence intervals of the raw and meteorologically adjusted $PM_{2.5}$ time series (left) and for PM_{coarse} (right) for all sites. The average over all sites is shown last along with error bars calculated by error propagation.

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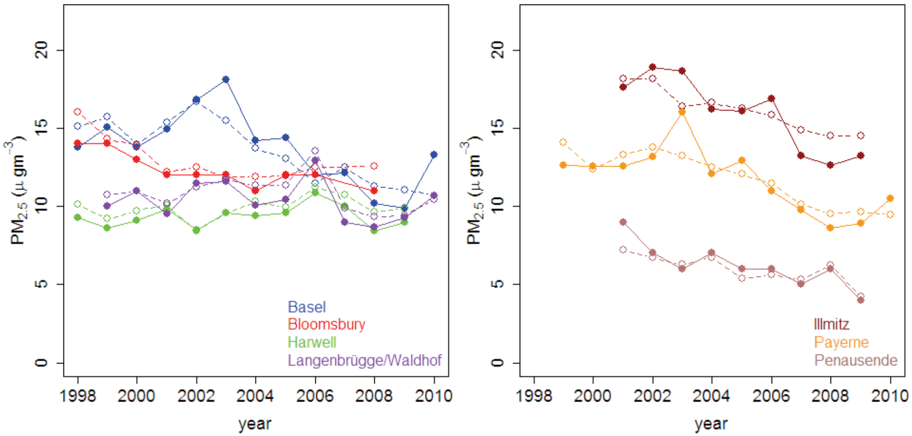

Fig. 6. $PM_{2.5}$ yearly median time series for all sites. Solid lines indicate raw data and dashed lines indicate meteorologically adjusted data.

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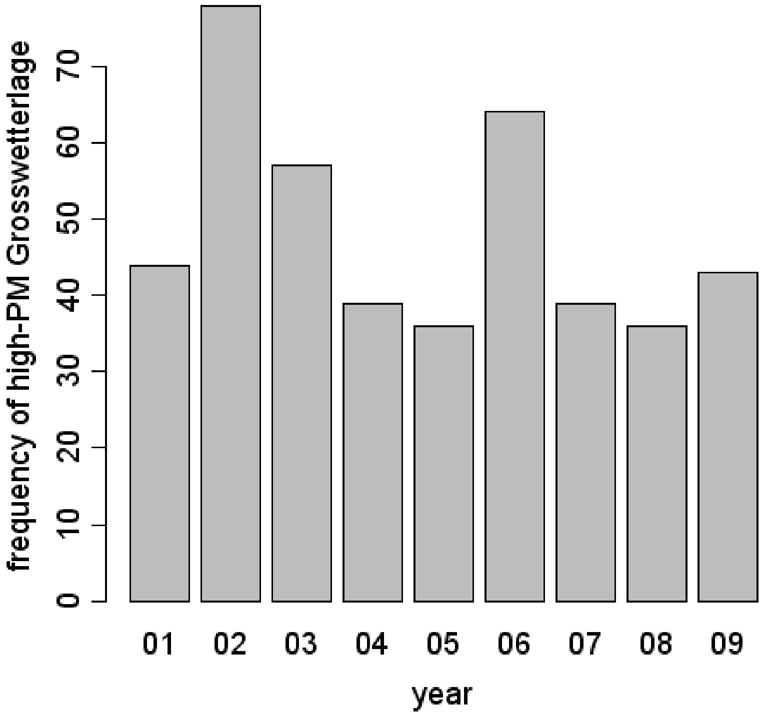

Fig. 7. Frequency of occurrence of high-PM GWL for each year at Illmitz.

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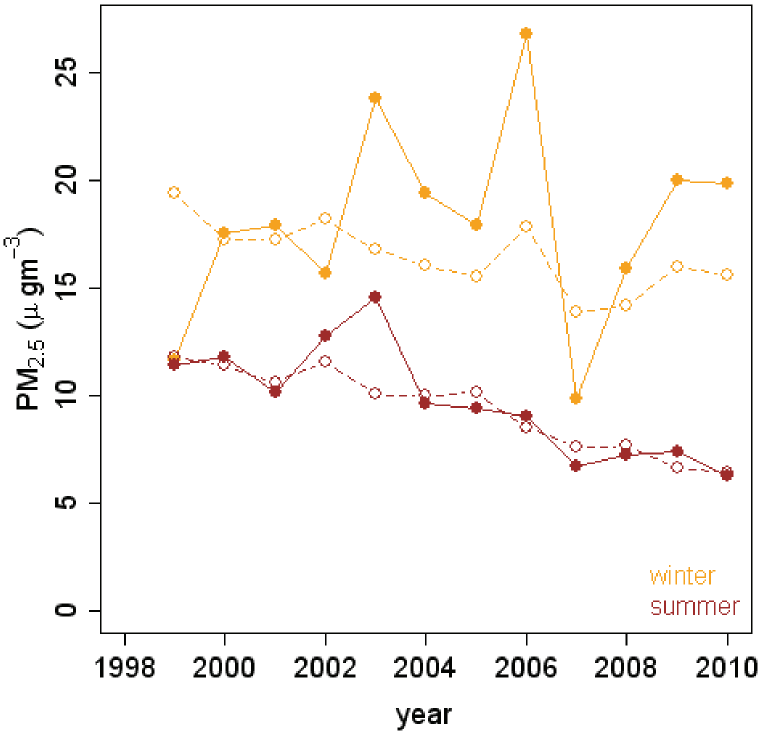

Fig. 8. Seasonal median $PM_{2.5}$ concentrations at Payerne for winter and summer data. Solid lines indicate raw data and dashed lines indicate meteorologically adjusted data.

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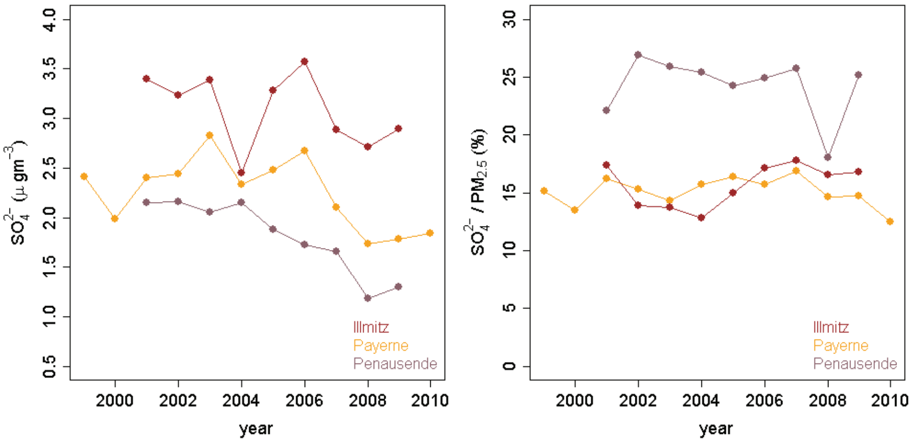

Fig. 9. Yearly average SO_4^{2-} concentrations (left) and $SO_4^{2-}/PM_{2.5}$ ratios (right) for Illmitz, Payerne and Penausende.

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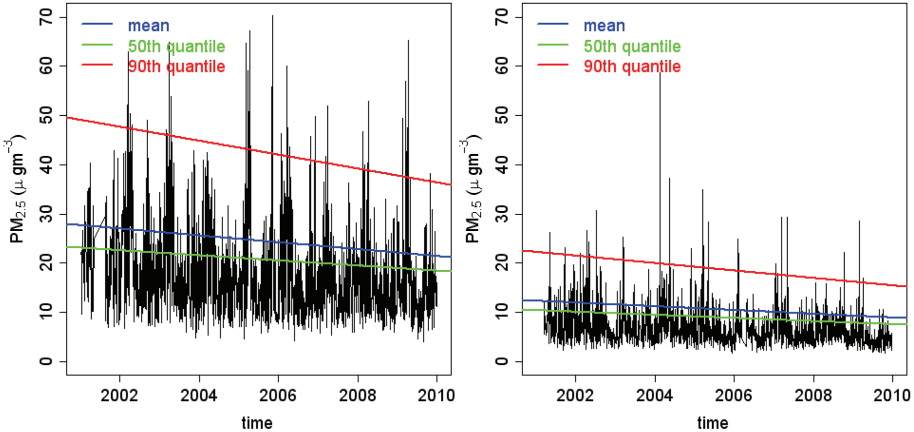

Fig. 10. Daily $PM_{2.5}$ time series for Illmitz (left) and Penausende (right). The OLS, 50th quantile and 90th quantile regression lines have been added.

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