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Analysis of exceedances in the daily PM_{10} mass concentration ($50\,\mu g\,m^{-3}$) at a roadside station in Leipzig, Germany

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Abstract

Five years of PM_{10} and $PM_{2.5}$ ambient air measurements at a roadside, an urban, and a regional background site in Leipzig (Germany) were analyzed for violations of the legal PM_{10} limit value (1999/30/EC, 1999). The annual mean PM_{10} concentrations at the three sites were well below the legal threshold of 40 µg m^{-3} (32.6, 22.0 and 21.7 µg m^{-3}, respectively). However, at the roadside site, the daily maximum value of 50 µg m^{-3} was exceeded on 232 days (13 % of all days) in 2005–2009, which corresponds to 57 days more than warranted by the EC directive. We analysed the meteorological and local source factors that eventually led to these surplus exceedences. Not surprisingly, the highest pollutant concentrations and most exceedance days were observed in winter. Average concentrations for exceedance and non exceedance days of 64 and 28 µg m^{-3} at roadside and 40 and 19 µg m^{-3} in the regional background were observed, suggesting urban contributions of 24 and 8 µg m^{-3}, respectively. Statistical and back trajectory cluster analysis yielded the essential result that PM_{10} concentrations were regionally enhanced during high pressure conditions, characterized by very low temperature, dry air masses, very low wind speeds, and stable stratification. The latter factor was instrumental in generating high PM_{10} concentrations at roadside as well as in the regional background through pollution trapping below the atmospheric inversion. During winter exceedance days, the highest organic and elemental carbon mass concentrations were measured. The fewest exceedance days were observed during fast moving air masses from the west, characterized by slightly unstable stratification and lower air pressure. During wintrily exceedance days, about half of PM_{10} at roadside was originating from regional transport and half from the urban-related sources. This result indicates that both are equally important in generating exceedance days in case of favourable meteorological conditions and cannot be separately considered. Our conclusion is that a combined effort of local, national and international reduction measures could most likely avoid systematic exceedances of the daily limit value in the future.

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1 Introduction

Since 2005, legal limit values apply to environmental particulate matter (PM) within the European Community (Air Quality Framework Directive (96/62/EC) and First Air Quality Daughter Directive (1999/30/EC)). PM_{10} denotes the total mass concentration of suspended particles with aerodynamic diameters smaller than 10 µm. The metric of PM_{10} (and also $PM_{2.5}$) is simple in that it can be determined in air quality networks with reasonable costs using on-line instrumentation (TEOM, beta-gauge, OPC), or off-line methods (filter collection and gravimetry) that serve as the reference method. On the other hand, it is evident that especially PM_{10} encompasses a wide range of particle types regarding size (coarse, fine, ultrafine), chemical composition (dust, combustion particles, marine primary particles, secondary organic aerosol, secondary inorganic aerosol), and sources (natural, traffic, industry, domestic households, secondary processes). This complex composition hampers the understanding of PM_{10} as a function of local sources, long-range transport and meteorology for a given site. In practice, exceedances of the legal limit values, particularly the daily limit value of 50 µg m^{-3} have frequently occurred at air quality monitoring stations in many EC member states. Due to the scientific evidence of health effects as a result of airborne particulate matter exposure, health scientists have called for a more serious consideration of efficient abatement measures (Annesi-Maesano et al., 2007).

The actual reasons for the PM_{10} exceedances are manifold, and vary from region to region. In the UK, which may be taken as representative for Western Europe, the advection of continental air masses as well as regional secondary aerosol formation seem to be responsible for the majority of exceedances (Charron et al., 2007). In the metropolitan area of Berlin, Germany, 50 % of the PM_{10} mass concentration is estimated to originate from regional and long-range transport rather than local sources (Lenschow et al., 2001). In arid regions like Spain, wind-blown mineral dust – partially imported from Africa, partially from agricultural soils, may have a major impact (Escudero et al., 2007). Even as far north as Germany, Saharan dust may enhance PM

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concentrations at ground level several times a year (Bruckmann et al., 2008). Agricultural dust is sometimes assumed to be responsible for the differences between measured and modelled PM_{10} concentrations in urban areas, and significant amounts of it can originate from long-range transport (Vautard et al., 2005; Birmili et al., 2008). Putaud et al. (2004) concluded for Europe that mineral aerosol is the main component in the $PM_{10-2.5}$ aerosol fraction, but of minor importance in $PM_{2.5}$. Particular sources of PM_{10} in urban environments are traffic, domestic heating and cooking, construction sites, industries, power generation or wind-blown resuspended dust (e.g., Querol et al., 2004). The traffic induced contribution includes abrasion (e.g., brake wear and tyre wear) (Sanders et al., 2003; Weckwerth, 2001), resuspension of road dust (Sternbeck et al., 2002; Amato et al., 2009), diesel soot particles (Kittelson, 1998; Rose et al., 2006) and nucleation mode particles (Kittelson, 1998) from exhaust gas.

According to official inventories, the emissions of primary particles and precursors of secondary particles in Europe have declined significantly over the past 20 yr, particularly in Central and Eastern Europe as a result of political and economic changes since 1990. Until the year 2000, these emission reductions have reflected themselves in clear corresponding trends of PM_{10}, for example in Germany (Spindler et al., 2004; UBA, 2009). Since the year 2000, however, the PM_{10} concentrations in Germany seem to stagnate, and feature some mere inter-annual fluctuations (UBA, 2009). A similar stagnation of PM_{10} since 2000 has also been reported for other areas in Europe, such as the UK (Harrison et al., 2008), Switzerland (Barmpadimos et al., 2011), Belgium, Czech Republic, Italy and Norway. This is in notable contrast to the continued reductions in PM emissions. Germany, for example, reported a 20 % decrease in PM_{10} emissions between 2000 and 2010 (updated information from UBA (2009)). The stagnating ambient levels of PM_{10} are therefore not fully understood and merit detailed examination. A better understanding on the sources and concentrations of PM_{10} is highly relevant, because the problem of exceedances in the daily limit value of PM_{10} seem to continue.

This work is concerned with the analysis of the the meteorological situations leading to exceedances of the daily limit value of PM_{10} (50 µg m^{-3}) in the city of Leipzig, East Germany. The main tool is back trajectory cluster analysis. In order to minimize subjectivity, a k-means cluster algorithm was applied in this study. Trajectory coordinates were used as the clustering variables the first time by Moody and Galloway (1988). Also Kemp (1993) and Mukai and Suzuki (1996) have analysed aerosol data based on air mass trajectories. Ozone concentration data have been interpreted by trajectory clustering methods by Brankov et al. (1998) or Cape et al. (2000). The long-range transport of aerosol particle concentrations was estimated by Buchanan et al. (2002) and Dutkiewicz et al. (2004) investigated the long-range transport of sulphate. Similar to the procedure presented here, Abdalmogith and Harrison (2005) were clustering the back trajectories and than assigning pollutant concentrations like the PM_{10} mass to the defined clusters. They found the highest sulfate, nitrate and PM_{10} mass concentrations and the lowest chloride mass concentrations in continental air masses and reversely the lowest ones in fast moving marine air masses. Higher particle mass concentrations were also observed in winter and spring compared to summer and fall. Beddows et al. (2009) applied a completely mathematical cluster analysis without the use of back trajectories for rural, urban and, kerbside atmospheric particle size data to determine temporal and special trends of the particle size distributions. Baker (2010) used a cluster analysis to analyze the long-range air transport and associated particle mass concentrations. The method used here was already successfully applied for the interpretation of transboundary anthropogenic pollution previously (Engler et al., 2007; Birmili et al., 2010).

2 Observation sites and data set

In this work, PM_{10} mass concentrations and exceedances of the daily limit value of 50 µg m^{-3} in the city of Leipzig, Germany, were analyzed over a period of five years (January 2005 to December 2009). The observation sites include a roadside site

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("Leipzig-Mitte") and an urban background site ("Leipzig-West") operated by the Saxonian Office for the Environment, Agriculture, and Geology, and an additional regional background site ("Melpitz") operated by the Leibniz Institute for Tropospheric Research (IfT).

The traffic site Leipzig-Mitte is situated near a ring road around the city center in immediate vicinity of the main station (Fig. 1). Of the three stations, it shows the highest mass concentrations of particles and nitrogen oxides. About 20 m north to the measurement container, three main roads merge at an intersection, which traffic volumed around 44 000 vehicles/day (48 000 vehicles/day during week days) in 2008. The station itself is located at a road being connected to the ring road with traffic lights. This leads frequently to traffic jams only few meters distant to the aerosol inlets of the measurement site. Additionally, there have been several constructions in the vicinity of the site during 2007 and 2008. The influence of these constructions was estimated to be about 10 $\mu g\,m^{-3}$ in the monthly average of PM_{10} for December 2007 and January 2008 (Stadt Leipzig; Dezernat Umwelt, Ordnung, Sport/Amt für Umweltschutz/Abteilung Umweltvorsorge, 2009).

The observation site Leipzig-West is located in the western suburban part of Leipzig, and can be classified as an urban background station. The measurement container is installed on hospital premises and the vicinity is dominated by green spaces. A minor road passes by the station around 30 m west of the site.

The research station Melpitz is located 50 km NE of Leipzig. The site is surrounded by flat grass lands, agricultural pastures and woodlands within several tens kilometres. Thus, the area can be considered as typical for the Central European regional aerosol (Engler et al., 2007; Spindler et al., 2010). The overall distance to the sea is about 400 km to the northwest and 1000 km to the west. Melpitz is part of the networks EUSAAR (European Supersites for Aerosol Research), EMEP (European Monitoring and Evaluation Programme) and GUAN (German Ultrafine Aerosol Network; Birmili et al., 2009).

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The 24 h mass concentrations of PM_{10} and $PM_{2.5}$ at Leipzig-Mitte and Melpitz were determined by using DIGITEL High Volume Samplers (HVS, Walter Riemer Messtechnik, Germany) (Gnauk et al., 2005). PM_{10} mass in Leipzig-Mitte and Melpitz as well as $PM_{2.5}$ in Melpitz were available daily, but $PM_{2.5}$ in Leipzig-Mitte only every second day. The 24 h filter samples of Melpitz were additionally analyzed for daily chemical particle composition, considering the main ions (sulfate, nitrate, ammonium, chloride, sodium, calcium, magnesium, potassium) as well as organic and elemental carbon. The measurement uncertainty for the gravimetrical mass determination is about 1 to 2 $\mu g\,m^{-3}$ and for the ion analysis less than 10 % (Neusüß et al., 2000; Brüggemann et al., 2005). In Leipzig-West, the PM_{10} concentrations were measured with the TEOM instrument (Tapered Element Oscillating Microballance) with a time resolution of 30 min. This method is relatively uncertain, because the high operation temperature of 50 °C causes systematic under determination up to 50 % by evaporation of volatile compounds such as ammonium nitrate (Charron et al., 2004; Spindler et al., 2010).

Furthermore, local meteorological measurements were conducted at all three observation sites. 72 h back trajectories were calculated using the NOAA-HYSPLIT4 (HYbrid Single-Particle Lagrangian Integrated Trajectory) model (Draxler and Hess, 2004) and radio soundings from the meteorological observatory Lindenberg, operated by the German Weather Service (DWD), were used. Lindenberg is situated about 150 km north east of Leipzig and is suitable for the analysis of vertical temperature profiles due to the flat orographic structure of eastern Germany.

3 Methodology

3.1 Statistical methods

The daily average values of PM_{10} were classified at all sites into exceedance and non-exceedance days according to the EC directive. Lenschow et al. (2001) described the local PM_{10} concentration at a roadside site as the sum of three source

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type contributions: the regional background (or long-range transport), the urban background increment (related to sources inside the same city, but not immediately nearby), and the local (or traffic related) contributions of vehicular traffic within a radius of a few 10th of meters. These contributions as well as the absolute traffic plus additional urban background contribution to the local PM_{10} concentration at roadside, which was defined as

$$\Delta PM_{10} = PM_{10,\mathrm{roadside}} - PM_{10,\mathrm{rural}}, \tag{1}$$

were calculated for each day of the measurement period. The occurrence rates and average values for these different source contributions and the PM_{10} concentrations at roadside and the regional site were calculated separately for exceedance and non-exceedance days. The average chemical composition was calculated, too, using 24 h samples of PM_{10} at the regional station. Furthermore, the Mann-Whitney-U-Test, a non-parametric significance tests, was applied. It is used for testing whether two populations are equal or not. It was proposed initially by Wilcoxon (1945) for equal population sizes and extended to arbitrary sizes and in other ways by Mann and Whitney (1947). In this work, the test was applied with a significance level of $\alpha = 95\,\%$ to check, whether the observed PM_{10} values and other meteorological parameters are significantly different for exceedance days and non-exceedance days.

3.2 Back trajectory cluster analysis

Back trajectories have been recognized as a valuable tool to investigate the large-scale origin of air pollutants (Stohl, 1998). Back trajectory cluster analysis combines similar trajectories into distinct groups (cluster). Consequently, back trajectory cluster analysis was applied in this study. Some major advantages of the cluster method are that it can be automated, and provides a high degree of objectiveness.

We applied a custom-made k-means cluster algorithm, which was developed in analogy to the approach first reported by Dorling et al. (1992), and divided the entire dataset

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into a predetermined number of trajectory clusters. The groundwork of the cluster analysis were back trajectories calculated using the HYSPLIT4 model. In addition, vertical profiles of pseudo potential temperature calculated from regular radiosonde ascents at the DWD station Lindenberg were used. Profiles of pseudo potential temperature have been incorporated in the variables to be clustered because vertical temperature profiles characterize the degree of vertical atmospheric stratification, which is of elementary importance to any surface measurement of atmospheric constituents. We applied this method to two aerosol data sets before (Engler et al., 2007; Birmili et al., 2010), and could confirm that the inclusion of the vertical temperature profile enhanced the separation of the long-term data set into different, meteorologically defined trajectory clusters. The following Euclidian distances were computed to express the spatial separation between two back trajectories *i* and *j*:

$$L_{i,j} = \frac{1}{k} \sum_{k} \sqrt{\sum_{l=1}^{4} a_l (x_{li} - x_{lj})^2}. \tag{2}$$

Here, k is the number of trajectory points, which was chosen as $k = 72$ to represent hourly back trajectory positions over 3 days. The choice of back trajectory length of 72 h was supported by the life time of several secondary species (Wojcik and Chang, 1997).

l is the dimension of the vector to be clustered, which includes the four variables of geographical latitude and longitude (x, y; both in Cartesian coordinates), height above ground (z) and pseudo potential temperature (Θ_e). To create four variables of equitable magnitude, the weights a_l are required and were chosen as $a_1 = a_2 = 1\,^{\circ\,-1}$, $a_3 = 10^{-5}\,\mathrm{m}^{-1}$, and $a_4 = 2\,\mathrm{K}^{-1}$.

Notably, the choice of a_3 and a_4 required prior test runs, because the temperature is a variable entirely dissimilar from geographical position. In practice, the cluster algorithm was run for a range of cluster numbers between 3 and 14. The deviation of the average PM_{10} concentrations (and the other aerosol and meteorological data) between the clusters was calculated for each test run and used for the choice of the weighing parameters. Selecting a small number of clusters will generate larger, more representative sub-sets of the data. On the contrary, using a higher number of clusters will resolve more features of different aerosol types. The decision, how many clusters to use for the final discussion of the PM_{10} data reflected a compromise between simplicity of display (low n) and a more visible separation of the data clusters (high n), and yielded in 9 clusters.

4 Results

4.1 Description of the measurement period

Figure 2 presents the full time series of PM_{10} concentration at the three observation sites during the entire measurement period from January 2005 to December 2009 (24 h average). The horizontal lines indicate the average values over all five years as well as the limit value of 50 µg m^{-3} for 24 h averaged concentrations (black). At roadside, the mass concentrations were usually higher (32.6 µg m^{-3} on average) compared to the background, but the difference between urban and regional background was insignificantly low (22 and 21.7 µg m^{-3}, respectively), especially considering the measurement uncertainties (see Sect. 2). These values are in good agreement with the PM_{10} mass concentrations of other central European observation sites (Putaud et al., 2010). In general, four different types of days were observed: (a) all stations show PM_{10} mass concentrations clearly below the limit value of 50 µg m^{-3}, (b) the roadside mass concentrations were closely above the limit value, but significantly lower at the background stations, (c) exceedance of the limit value at roadside and background mass concentrations only very closely below and (d) exceedances at roadside as well as in the background. Many exceedance days were characterized by enhanced background mass concentrations, but below the limit value (type c). In every year of the observation period, at least one of type (d) episodes with varying length was found as

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well. This leads to the question, which conditions are favourable for exceedances and what causes these high concentrations.

The number of limit exceedings for all three observation sites is shown in Table 1. At roadside, more than 35 exceedances occurred particularly in the years 2005 to 2007.

4.2 Statistical comparison of exceedance and non-exceedance periods

4.2.1 General characteristics of exceedance and non-exceedance days

In total, 1788 out of 1826 possible days could be classified (data coverage of 98 % during 5 yr), resulting in 232 exceedance and 1556 non-exceedance days (13 and 87 %, respectively). To obtain a more detailed impression of the variability of the data, Fig. 3 shows the occurrence rate as a bar chart for PM_{10} at roadside and in the regional background as well as the absolute traffic and the relative regional contribution.

At roadside, the average PM_{10} mass concentration was found being 64 µg m^{-3} for exceedance days, which means an exceedance of the limit value by 28 %. Most exceedance days showed concentrations between 50 and 70 µg m^{-3}; considerably higher values (maximum: 130 µg m^{-3}) were scarcely found. In case of no limit exceedance, the average value was 28 µg m^{-3} and most days showed concentrations between 20 and 40 µg m^{-3}. In the regional background, the average PM_{10} mass concentrations were 40 µg m^{-3} (which is already 80 % of the daily mean limit value) and 19 µg m^{-3} for exceedance and non-exceedance days, respectively. Compared to roadside, the distribution in the regional background is much broader for days with limit exceedances, with high frequencies of occurrence between 20 and 50 µg m^{-3}. The highest values with significant occurrence were found up to 100 µg m^{-3}. The frequency maximum for non-exceedance days was 20 µg m^{-3}, being exactly the same as for roadside.

The absolute traffic contribution to the local PM_{10} mass concentration at roadside was on average 24 for exceedance and 8 µg m^{-3} for non-exceedance days. The distribution is broader for exceedance days with maximum values between 15 and 35 µg m^{-3} compared to 0 to 15 µg m^{-3} for non-exceedance days. For exceedance

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days, a considerable amount of PM_{10} mass was thus originating in traffic related sources. The histograms of the relative contribution of regional transport look very similar; the distributions are equally broad and only slightly shifted to smaller values for exceedance vs. non-exceedance days (on average 62 and 69 %, respectively). This leads to the assumption, that how large ever the total concentrations were, the relative contributions and thus the origins or at least the mixture of sources were similar for PM_{10} particles. If this was the case, the chemical composition should show similarities as well (see Fig. 7).

In order to discuss this more detailed, the connection between PM_{10} mass concentrations at roadside and in the regional background was analyzed, which is shown in Fig. 4. Each square symbolizes one day of the measurements; the ones below the horizontal line at 50 µg m^{-3} stand for days with no limit exceedance at the roadside station, the ones above stand for exceedance days. The red stars indicate the average values of both sub data sets. A linear fit was added to the graph, resulting in a slope of 1.43 ($r^2 = 0.62$). This means, on average 70 % of the roadside PM_{10} mass concentration are resulting from the regional background already. High background concentrations make definitely a significant contribution to limit exceedances at roadside, but they are not solely responsible for them.

4.2.2 Seasonal dependency (exceedance vs. non-exceedance days)

To scrutinize the reasons for the PM_{10} exceedances, a variety of factors, such as season, local meteorological parameters as well as long-range back trajectories computed by the NOAA HYSPLIT model were used. Figure 5 illustrates the seasonal differences in the occurrence of limit exceedances, with most affected days found in winter, spring and also fall but only very few summer days associated with exceedances. Possible reasons could be stronger emissions during the colder seasons on one hand, but on the other hand also an enhanced accumulation of aerosol particles emitted at ground level because of more frequent inversion conditions and thus missing vertical transport. Another reason is middle-range transport from eastern neighbouring countries playing an important role considering the PM_{10} mass concentrations in eastern Germany.

In order to quantify the distinct contributions of different source types, Fig. 6 shows the average PM_{10} as well as PM_{coarse} (defined as PM_{10}–$PM_{2.5}$) mass concentrations for exceedance and non-exceedance days at the roadside and the regional background station. Clearly higher concentrations were observed during exceedance days compared to non-exceedance days. At roadside, the PM_{10} mass concentration was on average 64 and 28 $\mu g\,m^{-3}$ and PM_{coarse} 23 and 13 $\mu g\,m^{-3}$; in the regional background, PM_{10} was 40 and 19 $\mu g\,m^{-3}$ and PM_{coarse} 6 and 4 $\mu g\,m^{-3}$, respectively. Even the standard deviations for PM_{10} (both roadside and regional) were above the average value over the entire measurement period. During exceedance days, the regional background contributes on average already 80 % to the limit value of PM_{10} (50 $\mu g\,m^{-3}$) not containing any local urban sources yet. Except for the PM_{10} mass concentrations during exceedance days, which were about 10 $\mu g\,m^{-3}$ lower in summer compared to winter, no clear seasonal variation could be observed.

The chemical particle composition of PM_{10} in the regional background is shown in Fig. 7. The main components are nitrate, sulfate, ammonium, organic, and elemental carbon. In winter, the highest total mass concentrations were measured. This is probably caused by several facts: in summer, there are less primary (anthropogenic) particle sources; there is more turbulence, which causes a higher atmospheric boundary layer and thus increased dilution of the emissions and possibly the fraction of transported particles is higher in winter because of more frequent easterly advection situations. During exceedance days, a higher mass fraction of the main compounds could be found in the particles. Considering the relative PM_{10} composition, nitrate was increased mostly in spring and winter, ammonium in spring, fall and winter, sulfate throughout the year, and organic carbon in summer, fall and winter. Volatile compounds such as ammonium nitrate are present in the gas phase during summer time but in colder air in the particle phase (Flechard and Fowler, 1998). The lower chloride masses in summer are explainable by the photochemical exchange of chloride by nitrate under participation of sulfuric acid (Pio and Lopes, 1998). Sodium and chloride were barely present during exceedance days, which supports the hypothesis of exceedance days occurring not

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(or only scarcely) in westerly (marine) air masses. To investigate this hypothesis further, a cluster analysis considering back trajectories was carried out (see Sect. 4.3). In winter, a higher mass fraction could be analyzed, which is probably due to less crustal material, dispersed in the air. The unidentified fraction is assumed to contain crustal material as silicates and insoluble carbonates (Espinosa et al., 2002). This assumption is supported by the slightly increased coarse particle mass concentration in summer (see Fig. 6). This is in agreement with Birmili et al. (2010), who also found a maximum of coarse particles in summer, which was concluded to originate from resuspension.

In the UK, nitrate was identified as the dominating compound of PM_{10} during exceedances (Yin and Harrison, 2008). They found contributions of up to 40 % at kerbside. In this study, only contributions of about 20 % were found for exceedance days, but nevertheless it was one of the main contributors to PM_{10} mass concentration, too. The highest OC and EC concentrations were identified during winter exceedances. The “low-emission-zone” in the city of Leipzig, will probably reduce this contribution to PM_{10}, including toxicologically relevant substances, attached to these particles since the particle emission of Euro 3 diesel vehicles will be reduced. But whether the total PM_{10} mass can be reduced as well is unclear, since there could be more traffic (but not exhaust) related particle emissions due to, e.g., abrasion and resuspension (Harrison et al., 2008). Also Dijkema et al. (2008) reported a more effective reduction of PM_{10} at roadside due to speed limit reduction (and thus less resuspended road dust) compared to the impact of reduced emission vehicles. However, the non-exhaust-related traffic emissions, which increase with increasing traffic volume independent of exhaust emission regulations, are of environmental significance, since they may contain trace metals causing negative health effects as well (Weckwerth, 2001; Sternbeck et al., 2002). These non-exhaust emissions are more or less constant during the year but depending on the traffic volume, the type of road, the pavement and the meteorological conditions. They are reduced by rainfall of at least 2 mm h^{-1}, probably because this results in a relatively long period with wet road surface and thus reduced resuspension (Keuken et al., 2010).

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For characterizing the meteorological conditions leading to exceedances of the limit value, Table 2 shows the average values of the main meteorological parameters for exceedance vs. non-exceedance days for the entire observation period as well as for each season. Values in bold indicate the sub-datasets of exceedance days being significantly different compared to non-exceedance days. This was tested using the Mann-Whitney-U-test with a significance level of $\alpha = 0.95$. On average, exceedance days were colder than non-exceedance days and also colder than the climate mean, especially in winter. The dew point was, except for summer days, also lower for exceedance days, which can also be seen in a lower absolute humidity. The relative humidity does not show significant differences. Exceedance days were also characterized by higher air pressure, low wind speeds and only very little precipitation. The trace gas concentrations have been analyzed as well and higher concentrations were found for SO_2, NO_2 and NO, while lower values of O_3 were measured. The PM_{10} mass concentration at roadside was clearly above the limit value of 50 µg m^{-3} for exceedance days (59 to 69 µg m^{-3}). Except for summer, almost 40 µg m^{-3} could be found in $PM_{2.5}$, already (55 to 65 %). During non-exceedance days, PM_{10} mass concentrations below 30 and $PM_{2.5}$ of about 15 µg m^{-3} were found. For exceedance days, a seasonal variation of about 10 µg m^{-3} could be found for winter compared to summer both in PM_{10} and in $PM_{2.5}$, but the seasonal influence was low for non-exceedance days. The urban and regional background PM_{10} mass concentrations were almost equal; the difference was 2 µg m^{-3} at the most. During exceedance days, mass concentrations of 29 to 44 µg m^{-3} for PM_{10} and 22 to 39 µg m^{-3} for $PM_{2.5}$ were observed at the regional station, which means a $PM_{2.5}$ contribution of 80 to 90 % to PM_{10}. Also in the background a clear seasonal difference of about 15 µg m^{-3} between summer and winter was found for both PM_{10} and $PM_{2.5}$. During non-exceedances, about 20 µg m^{-3} PM_{10} and 15 µg m^{-3} $PM_{2.5}$ were observed with no clear seasonal variation. For non-exceedance days, the $PM_{2.5}$ contribution to PM_{10} mass was higher (70 to 85 %) at the regional site compared to the roadside site, which points to sources of coarse particles connected to traffic, probably due to resuspension of road dust and tire and break abrasion. The absolute

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traffic contribution to the local PM_{10} mass concentration at roadside PM_{10} shows a clear difference between exceedance and non-exceedance days, especially in summer (7 and 30 µg m^{-3}, respectively). This is due to the elevated PM_{10} concentrations at roadside and in the background, since they are known to be connected to each other (see Fig. 4).

After Lenschow et al. (2001), PM from regional transport (RT), urban background sources (urban impact, UI), and local traffic related sources (traffic impact, TI) are the three main components that make up total PM_{10} mass concentration at an individual urban site. These contributions were estimated for each day by subtracting the mass concentration values in the regional background, urban background, and at roadside. ($RT = PM_{10,reg}/PM_{10,road}$; $UI = (PM_{10,reg}\text{-}PM_{10,urb})/PM_{10,road}$; $TI = (PM_{10,urb}\text{-}PM_{10,road})/PM_{10,road}$) The relative contribution of the local traffic was the highest for low regional background concentrations; nevertheless, the regional transport contribution was between 50 and 75 % of the local concentration. The lowest values were found for exceedance days (49 to 64 %), indicating both, regional transport and local sources playing an important role in the formation of exceedance days. To investigate the regional contribution more detailed, the origin of the air masses advected to the measurement sites have to be taken into account.

4.3 Back trajectory cluster analysis

A cluster analysis using a combination of daily back trajectories and vertical temperature profiles (similar to the earlier application of particle size distributions in Engler et al., 2007) was performed for the entire dataset. Both back trajectories and radio sounding data are sensitive indicators for the large scale weather condition. A run with a relatively strong weighing on the temperature profiles and with the result of 9 clusters was chosen for this analysis and the resulting clusters are shown in Fig. 8. Displayed are the geographical origin and the thermal properties of the air masses, the number of days as well as exceedances of the daily PM_{10} limit value and the seasonal distribution

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in each cluster. Cluster 1 represents slowly moving southerly air masses, which were slightly stable and occurred in fall and winter. Cluster 2 is representing slow easterly advection with anti-cyclonic character and unstable layering and is occurring mostly in spring and summer. Very stably layered and slowly moving air masses during winter times are grouped to cluster 3. The days in cluster 4 showed slow advection from north-easterly direction, were of anti-cyclonic character, neutrally layered and could be found mainly in fall and winter. Cluster 5 is representing slow westerly advection and unstably layered air occurring in spring and summer. Cluster 6 consists of fast moving air masses from the west, is slightly stable and occurring in fall and winter. The trajectories in cluster 7 were fast again and arriving from north-westerly directions, slightly cyclonic and unstably layered and occurred during all seasons. Cluster 8 shows similar trajectories as cluster 6, is unstably layered and occurring mostly in spring and summer. Cluster 9 is representative for very fast moving air masses from the west, with similar stability as cluster 8 and occurring mostly in winter and spring.

Table 3 summarizes the frequency of occurrence of each cluster, average values of meteorological parameters, trace gases, PM_{10} and $PM_{2.5}$ mass concentrations and source type contributions for the entire data set as well as each cluster. Again, a Mann-Whitney-U-test was performed for all considered parameters to compare the values of each cluster with the entire data set. A bold value in the table indicates a statistical difference between the days belonging to the cluster and the entire measurement period. The season index is a value between −1 (midwinter, 21 December) and 1 (midsummer, 21 June), indicating the seasons, contriuting to the individual clusters. A value close to −1 points to mainly winter days, close to 1 mainly summer days and close to 0 mainly spring and fall days. The seasonal distribution can be seen in Fig. 8d as well. The occurrence rate of exceedance days is calculated as the ratio of exceedance days to all days belonging to one cluster (see Fig. 8c; stars to columns). With other words, it is the probability of an exceedance if the weather condition of this cluster is occurring. Additionally, Fig. 9 shows the PM_{10} and PM_{coarse} mass concentrations at roadside and in the regional background as well as standard deviations for each cluster, and furthermore the average values of these four parameters over the entire measurement period.

The most discernable case is cluster 3 with the highest pollutant concentrations (particulate mass as well as SO_2 and nitrogen oxides) compared to the other clusters. This winter-cluster is characterized by temperatures clearly under the climate average, low dew point and absolute humidity, very low wind speed, no precipitation and the highest air pressure compared to the other clusters. A pronounced high pressure system during winter months causes very low temperatures and thus very stable layering with low vertical and horizontal exchange of the air. Due to the high stability and low vertical extend of the boundary layer, the air was trapped under an inversion layer and not transported horizontally. This is why the pollutants PM_{10}, $PM_{2.5}$, sulfur dioxide and the nitrogen oxides were accumulated close to the ground. Inversion situations and a low mixing layer height, which defines the dilution volume of air pollutants emitted, are strongly related to high concentrations of gases and particulates also in other European (Kukkonen et al., 2005) or German cities (Schäfer et al., 2006). Due to the low wind speed, the PM_{coarse} mass concentration was very low and the main contribution to PM_{10} came from particles smaller 2.5 µm, which are known to be emitted mostly by anthropogenic sources. The probability of an exceedance of the daily limit value was 60 % during days with the described meteorological situation, but only 4 % of all days were allocated to cluster 3 during the considered period of 5 yr. The probability of an exceedance day is very high for wintrily inversions, but since their occurrence is seldom compared to other weather situations, they contribute absolutely equally to exceedances as more frequent meteorological conditions like cluster 1, 2, and 4 do.

Cluster 1 is representing air masses arriving from the south with possible accumulation of aerosol mass over south-western Europe. Cluster 2 and 4 consist of air masses from the east and north-east. All three of them are neutrally or slightly unstably layered, quite slowly moving air masses with low wind speed and a small but considerable amount of precipitation and occurred throughout the year. The origin of the air masses are assumed to be responsible for regionally already elevated aerosol mass

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concentrations and the low wind speed and ineffective removal of particles by only low precipitation are assumed to be the reasons for their accumulation. However, the probability of limit exceedances is lower (22 %) in these clusters compared to the wintry inversions.

In the UK (which is representative for western Europe as well), the advection of air masses from the continental Europe and the regional formation of secondary aerosol were responsible for the majority of exceedance days, too (Charron et al., 2007). They also found an increased regional background contribution for exceedance days. In Spain, a compareable number of exceedance days was found by Escudero et al. (2007), but the reason was only seldomly identified in continental air masses. In southern Europe, Saharan dust plays a much more efficient role in the generation of exceedances compared to central Europe. However, anticyclonic conditions in winter drive the accumulation of pollutants also over the Iberian peninsula. As reported by Viana et al. (2007), the most accute episodes of PM_{10} pollution in western Europe were connected to air masses with European origin, too. They concluded the origin of the exceedances being anthropogenic, but may be long-range transport and not only local pollution. Anyhow, eastern European trajectories frequently coincide with anticyclonic scenarios, leading to air mass stagnation.

In contrast to the “exceedance”-clusters, cluster 9 describes the weather situation with fewest exceedance days. Fast moving trajectories arriving from the west indicate marine (and thus relatively clean) air masses and a slightly unstable layered boundary layer. The air was chilly because of the winter and early spring days, however, considerably warmer than the climate average for the same time of the year. A higher amount of precipitation and thus an increased deposition of existing aerosol particles reduces the msss concentration. The air pressure was lowest and the wind speed highest compared to the other clusters, which means a stronger dilution of at ground level emitted particles. Nitrogen oxides and sulfur dioxide showed the lowest concentrations and so did the PM_{10} mass both at roadside and in the regional background. However, the coarse particle fraction was highest compared to the other clusters, which was probably

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caused by resuspension of mineral dust due to the high wind speed.

Unlike the clusters 1 to 4, there are some weather situations, which inhibit the development of exceedance days just because of meteorology, which becomes apparent in a very low probability of exceedance days (2 to 4 %). Clusters 7 to 9 are characterized by fast moving air masses from the west or north-west and thus by higher wind speed and more precipitation than the exceedance clusters, causing more effective removal of aerosol particles out of the atmosphere. Due to the higher boundary layer, all pollutants do not accumulate close to the sources at the ground, resulting in lower PM_{10} mass and gaseous pollutant concentrations. Marine air masses from westerly directions are also supposed to be much less polluted than continental easterly air masses considering both, industrial and traffic related sources. Thus, much lower concentrations of sulfur dioxide as well as nitrogen oxides could be observed.

Clusters 5 and 6 are neither associated with exceedance nor with non-exceedance conditions, but somewhere in between. These two intermediate clusters are characterized by trajectories similar to the clean cluster 9, but slower. They are also slightly unstably layered and warmer than the climate average with considerable amounts of precipitation. Wind speed, air pressure, trace gas and PM_{10} mass concentrations, and the coarse mode contribution were around the average values. The probability of this intermediate weather condition was 26 % and the probability of a limit exceedance during this weather situation about 10 %.

The relative contribution of local traffic-related sources was highest for low background concentrations; nevertheless, the regional contribution was between 58 and 73 % of the local PM_{10} mass concentration. This is in good agreement with the results by Lenschow et al. (2001) in the city of Berlin, who found 50 % of urban PM_{10} originating from the regional background. They reported the traffic as the most important group of sources causing high PM_{10} concentrations at kerbside, with up to 15 % of PM_{10} mas concentration resulting from resuspended soil. These contributions show the horizontal origin of an air mass not being sufficient to cause exceedance days at the traffic site, but it plays an important role, anyway. However, the source contributions from regional,

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urban background and traffic related sources were surprisingly similar for all clusters, independent of the weather situation. This is why we suppose, that both regionally and locally close to ground emitted particles are accumulated in the boundary layer in case of favorable meteorological conditions, but the sources are nevertheless similar. This would result in different absolute masses but the same relative contributions. To investigate this more detailed, the chemical composition has to be considered as well, which is displayed in Fig. 10 for the regional background site.

Especially in clusters 1, 3 and 6, more ammonium and nitrate have been analyzed, because these compounds are volatile at higher ambient temperatures, and thus in the gas phase during summer months like clusters 2, 5 and 8 (Flechard and Fowler, 1998). The chloride concentration is low as well, due to the photochemical exchange by nitrate (Pio and Lopes, 1998). Clusters 1 to 5 show almost no sodium chloride since they are associated with continental air masses and not marine as other clusters. Particularly in the marine clusters 7 and 9, a considerable amount of sodium chloride was determined. Clusters 6 and 8 are of marine origin but already influenced by the continent, which results in some sodium chloride. Clusters 7 to 9 show a similar relative chemical composition as the non-exceedance days (see Fig. 7); cluster 7 resembles a mixture of all seasons, cluster 8 is alike spring, and cluster 9 alike a mixture of winter, spring and fall. However, the total mass was even lower for the clusters, since the clean marine air masses were separated from the intermediate polluted (continentally influenced) air masses by the clustering. Cluster 3 with the highest mass concentration shows almost the same chemical composition as the winterly exceedance days (see Fig. 7). Most apparent is the high sulfate concentration probably caused by industrial sources in Eastern Europe. The most organic compounds were found in clusters 2, 3 and 1, whereas the highest contribution of elemental carbon was found in cluster 3. Anyhow, as seen already in the seasonal analysis, also the relative chemical composition of each cluster was very similar, indicating also similar source type contributions. Certain meteorological conditions can then be the determining factors leading to PM_{10} limit exceedances at the observation sites.

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5 Summary and conclusions

Five years of PM_{10} and $PM_{2.5}$ measurements at three observation sites (roadside, urban and regional) have been analyzed concerning PM_{10} limit exceedances according to the EC directive 1999/30/EC. Especially in 2005 to 2007 more than 35 exceedances per year have been observed. Statistical and back trajectory cluster analysis yielded the essential result, that PM_{10} concentrations were regionally enhanced during high pressure conditions, characterized by very low temperature (clearly below the climate mean), dry air masses without any precipitation, very low wind speeds and stable stratification. The latter factor was instrumental in generating high PM_{10} mass concentrations at roadside as well as in the regional background through accumulation below the atmospheric inversion. In case of this weather situation, the probability of a limit exceedance at the roadside station in Leipzig was 60 %, but it was very seldom (4 % of all days). Because of only limited vertical and horizontal transport and missing wet deposition of emitted pollutants, PM_{10}, $PM_{2.5}$, sulfur dioxide and the nitrogen oxides could accumulate regionally in the ground layer. Additionally, the emission of primary particles is stronger in winter than in summer and during winter times, the advection from air masses from highly polluted regions in eastern Europe is more frequent leading to higher mass concentrations of transported primary particles as well as gaseous precursors and thus secondary particles, too.

Furthermore, in winter, more ammonium, nitrate and chloride have been found in PM_{10}. During exceedances and in winter, an increased mass fraction was identified due to less crustal material, dispersed in the air, which led to a lower coarse particle fraction for exceedance days compared to non-exceedance days for both, the roadside and the regional station. Consequently, $PM_{2.5}$ was the main contributor to the local PM_{10} mass during exceedances, which is mostly originating from anthropogenic sources. The coarse fraction at the roadside site was clearly higher than at the regional site. This points to sources of coarse particles connected to traffic, probably caused by resuspension of road dust, break and tire abrasion.

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Due to the very low wind speeds, the coarse particle fraction was lower for exceedance days compared to non-exceedance days for both, the roadside and the regional station. Nitrate contributions of about 20 % were found for exceedance days, but nevertheless it was one of the main contributors to PM_{10} mass concentration. During winter exceedances, the highest OC and EC concentrations were identified. The implementation of a "low-emission-zone" in the city of Leipzig, will probably reduce this contribution to PM_{10}, including toxicologically relevant substances, since the particle emission of Euro 3 diesel vehicles will be reduced. But whether the total PM_{10} mass can be reduced as well is unclear, since this reduction could be compensated by the emissions of other traffic (but not exhaust) related particles via, e.g., abrasion and resuspension.

During slowly moving air masses from the south, east or north east, characterized by neutral stratification, the origin (south-western and eastern Europe) was responsible for elevated concentrations of pollutants and the low wind speeds and ineffective removal (by only little precipitation) are reasons for even further accumulation. The probability of limit exceedances during these conditions was about 22 %. Generally, very low concentrations of sodium and chloride were found during exceedance days, indicating exceedances occurring only scarcely during westerly air mass advection. However, trajectories from eastern Europe frequently coincide with anticyclonic conditions, leading to air mass stagnation.

The fewest exceedances were observed during fast moving air masses from the west, where significant amounts of sodium and chloride have been identified in the chemical particle analysis. Marine air masses are known to be relatively clean considering both, industrial and traffic related sources. The slightly unstable stratification, the warmer air compared to the climate mean and the lower air pressure cause a higher boundary layer than during typical exceedance weather situations, which results in a larger dilution volume. Due to this fact, at ground level emitted particles cannot accumulate in the lower layers. Moreover, the precipitation plays a role by effectively removing existing particles and the higher wind speed causes stronger horizontal dilu-

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tion and a higher coarse fraction by, e.g. resuspension.

Three source type contributions (traffic related, diffuse urban, regional transport) have been calculated for each day of the period of concern. 50 to 70 % of the local PM_{10} mass could be identified as regional transport. In this study, the traffic related contributions were largest for low background concentrations. For wintrily exceedance days, about half of PM_{10} at roadside was originating from regional transport and half from the local traffic related sources. This result indicates both playing an important role in generating exceedance days and cannot be separately considered. Neither the horizontal transport of an air mass nor the traffic alone are sufficient to cause limit exceedances. However, the similar relative contributions to and the only marginal differences of the relative chemical composition of the local PM_{10} mass between exceedance and non-exceedance days lead to the conclusion that both, regionally and locally emitted particles, can accumulate in the boundary layer in case of favourable meteorological conditions, but the emissions originated in a comparable mixture of sources. The meteorological conditions can then be critical for dilution or accumulation of the particles and thus influence the generation of exceedances. This highlights local, national as well as EC-level action being required to substantially reduce the number of exceedances at the observation sites in Leipzig.

Four different weather situations have been recognized as favourable for limit exceedances in the city of Leipzig. Each of them makes an absolute contribution of about 18 %, thus 72 % of all limit exceedance days could be explained by one of them. Exceedance days were noticeably frequent during wintry inversion situations, since they were much more seldom compared to the other weather conditions leading to exceedances of the daily limit value. However, meteorological fluctuations may also have an effect on the annual average surface concentrations of air pollutants and the climate change could thus affect the concentrations as well (Velders and Matthijsen, 2009). Finally, it could be shown, the cluster analysis being an objective method, which separates the air masses as well as the PM_{10} data well.

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In this study, PM_{10} mass concentrations were regionally enhanced during high pressure conditions, characterized by very low temperature (clearly below the climate mean), dry air masses without any precipitation, very low wind speeds and stable stratification. The stratification was identified as the most important factor leading to high PM_{10} mass concentrations at roadside as well as in the regional background through accumulation of pollutants (such as primary particles or precursor gases) below the atmospheric inversion. The wintry inversions accounted for as many exceedance days as each of the other three weather conditions responsible for limit exceedances, even though they were in total much more seldom. This means, there is one meteorological condition, during which limit exceedances were conspicuously frequent, but they were occurring in other conditions as well. There was not *a single one* condition reliably leading and *a single one* condition reliably *not* leading to limit exceedances. However, the occurrence of days with low or high wind speed, air pressure and temperature corresponds to the natural weather pattern. The meteorological conditions are not the cause of high pollutant concentrations und hence exceedances. No more than the large-scale pollutant emissions by human activities in densely developed and low ventilated areas let a stable stratification become a problem. The meteorological condition is no more than a factor leading to accumulation of man-made emissions.

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Table 1. Number of PM_{10} limit exceedances and valid measurement days (in brackets) per year.

year	roadside	urban	rural
2005	70 (354)	9 (358)	8 (356)
2006	71 (348)	17 (350)	11 (350)
2007	38 (356)	6 (359)	10 (359)
2008	28 (337)	5 (337)	6 (353)
2009	25 (363)	18 (362)	11 (355)

Table 2. Mean meteorological parameters and trace gas concentrations for each season (non-exceedance vs. exceedance days; bold: significantly different, otherwise significantly equal).

parameter	all (232)	spring (79)	summer (11)	fall (59)	winter (83)
temperature [°C]	**11.1 / 5.9**	10.3 / 8.7	**18.7 / 20.9**	10.6 / 10.2	**2.9 / −1.9**
Δ T (climate mean) [°C]	**0.6 / −0.5**	0.31 / 0.87	**0.25 / 3.6**	0.7 / 1.1	**1.2 / −3.5**
dew point [°C]	**7.7 / 3.1**	**6.2 / 4.4**	**13.6 / 16.8**	8.1 / 8.1	**1 / −3.8**
rel. humidity [%]	**79.4 / 82.3**	75.5 / 74.4	72.2 / 77.4	**84.4 / 86.9**	87.3 / 87.2
abs. humidity [g/kg]	**8 / 5.9**	**7.2 / 6.4**	**11.6 / 14.1**	8.2 / 8.3	**5.2 / 3.7**
air pressure [hPa]	**1006 / 1011**	**1003 / 1008**	1007 / 1010	**1008 / 1011**	**1006 / 1015**
wind speed [m/s]	**2.5 / 1.5**	**2.4 / 1.5**	**1.9 / 1.1**	**2.6 / 1.5**	**3.3 / 1.5**
precipitation [mm/day]	**1.3 / 0.2**	**1.3 / 0.2**	1.8 / 1.8	**1.2 / 0.1**	**0.9 / 0.1**
SO_2 [µg m^{-3}]	**2.7 / 5**	**2.7 / 4.5**	2.3 / 2.5	**2.6 / 3.4**	**3.3 / 6.8**
O_3 [µg m^{-3}]	**61 / 48**	70 / 72	79 / 87	**46 / 34**	**45 / 31**
NO_2 [µg m^{-3}]	**9.1 / 15.6**	**8 / 12.2**	**6.5 / 7.9**	**10.2 / 13.9**	**12.4 / 21.1**
NO [µg m^{-3}]	**1.4 / 2.7**	**1.3 / 1.8**	1.2 / 1.1	**1.4 / 2.9**	**1.6 / 3.5**
PM_{10} (roadside) [µg m^{-3}]	**28 / 64**	**29 / 63**	**26 / 59**	**29 / 59**	**28 / 69**
$PM_{2.5}$ (roadside) [µg m^{-3}]	**15 / 38**	**16 / 40**	**13 / 31**	**16 / 36**	**18 / 37**
PM_{coarse} (roadside) [µg m^{-3}]	**13 / 23**	**13 / 22**	**12 / 26**	**13 / 23**	**12 / 26**
PM_{10} (urban) [µg m^{-3}]	**19 / 42**	**20 / 42**	**19 / 29**	**19 / 37**	**19 / 46**
PM_{10} (rural) [µg m^{-3}]	**19 / 40**	**20 / 40**	**19 / 29**	**18 / 35**	**19 / 44**
$PM_{2.5}$ (rural) [µg m^{-3}]	**15 / 33**	**17 / 31**	**13 / 22**	**14 / 29**	**16 / 39**
PM_{coarse} (rural) [µg m^{-3}]	**4.2 / 6.4**	**3.9 / 8**	**5.4 / 6.6**	**4.5 / 6.5**	**3.1 / 5**
ΔPM_{10} [µg m^{-3}]	**9 / 24**	**9 / 23**	**7 / 30**	**10 / 24**	**9 / 25**
traffic impact [%]	**31 / 35**	32 / 33	**26 / 51**	35 / 37	32 / 33
urban impact [%]	0 / 3	**0 / 4**	1 / 0	1 / 3	0 / 3
regional transport [%]	**69 / 62**	**70 / 63**	**73 / 49**	64 / 59	67 / 64
PM_{coarse} (roadside) [%]	**46 / 38**	45 / 35	**49 / 46**	**47 / 39**	**42 / 41**
PM_{coarse} (rural) [%]	**22 / 17**	18 / 21	28 / 21	**24 / 19**	**17 / 11**

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Table 3. Occurrence rates, mean meteorological parameters and trace gas concentrations for each cluster (bold: significantly different, otherwise not significantly different from the overall mean). The season index is a number between −1 (midwinter, 21 December) and 1 (midsummer, 21 June), giving an impression of the seasons contributing to the individual clusters.

parameter / cluster	all	1	2	3	4	5	6	7	8	9
air mass		S	E	st.	NE	WSW	WSW	NW	WSW	W
season index		−0.46	0.43	-0.69	−0.12	0.44	−0.46	0.13	0.29	−0.23
occurrence rate [%]		7	13	4	12	17	9	16	14	8
exceedance days	232	46	42	40	39	27	18	11	6	3
probability of exceedance [%]	13	35	18	61	18	9	11	4	2	2
temperature [°C]	10.4	**5.8**	**16.1**	**−2.1**	**5.1**	**15.9**	**7.3**	**8.3**	**14.2**	**8.8**
Δ T (climate mean) [°C]	0.5	0.6	**1.9**	**−4.3**	**−1.8**	**1.5**	**2.3**	**−2.2**	**1.5**	**2.9**
dew point [°C]	7.1	**4.2**	**10.7**	**−4**	**3**	**11.8**	**5.1**	**5**	**10.2**	**5.5**
rel. humidity [%]	80	**90**	70	**87**	**86**	**76**	**86**	79	**77**	80
abs. humidity [g/kg]	7.7	**6.4**	**9.6**	**3.6**	**5.9**	**10.4**	**6.8**	**6.7**	**9.4**	**6.9**
air pressure [hPa]	1007	**1005**	1010	**1015**	**1012**	**1004**	1006	1007	**1005**	**1002**
wind speed [m/s]	2.4	**2**	**1.7**	**1.8**	2.1	**1.7**	**2.8**	2.5	**2.9**	**4.3**
precipitation [mm/day]	1.2	1	**0.8**	**0.1**	**0.6**	1.6	1.5	0.9	**1.9**	**1.5**
SO_2 [µg m^{-3}]	3	**3.6**	**3.2**	**8**	**3.5**	2.6	3	2.1	**2.4**	**2.1**
O_3 [µg m^{-3}]	59	**31**	**80**	**31**	**46**	**73**	**41**	59	**67**	60
NO_2 [µg m^{-3}]	10	**15.6**	**7.8**	**18.9**	**11.2**	9.2	**13.7**	7.8	**7.9**	**8.4**
NO [µg m^{-3}]	1.6	**2.6**	1.4	**3.2**	**1.6**	1.6	**1.9**	1.1	**1.1**	**1**
PM_{10} (roadside) [µg m^{-3}]	32.6	**45.7**	38.8	**56.7**	**37.3**	32.8	31.5	**26.2**	**23.4**	**22.9**
$PM_{2.5}$ (roadside) [µg m^{-3}]	18.2	**28.1**	**21.6**	**33.1**	**23.7**	20.3	16.7	11	**12.6**	**10.6**
PM_{coarse} (roadside) [µg m^{-3}]	13.6	15.1	**16.6**	10	13.6	15.4	12.6	12.7	**10.4**	12.6
PM_{10} (urban) [µg m^{-3}]	22	**31.1**	**30.2**	**46.2**	**25.8**	**23.6**	**20.5**	**16.2**	**17.4**	**14.9**
PM_{10} (rural) [µg m^{-3}]	21.7	**31**	**27**	**40.5**	**24.5**	**22.1**	20	**15.1**	**17.1**	**14**
$PM_{2.5}$ (rural) [µg m^{-3}]	17.4	**26.6**	**21**	**36**	**20.2**	17.8	16	**11.5**	**12.8**	**9.9**
PM_{coarse} (rural) [µg m^{-3}]	4.6	5.6	**6.1**	4.5	4.4	**5.3**	**4.1**	**3.8**	4.5	4.1
Δ PM_{10} [µg m^{-3}]	11.2	**14.6**	**11.9**	**16.2**	**13.3**	11.1	11.6	11.1	**6.7**	**9.2**
traffic impact [%]	29	32	**22**	**18**	31	28	**35**	**38**	**26**	35
urban impact [%]	4.9	**0.2**	8.3	10	3.4	1.5	1.7	4	1.5	**4**
regional transport [%]	66	68	**70**	**72**	66	67	63	58	**73**	**61**
PM_{coarse} (roadside) [%]	42	**33**	43	**18**	**36**	47	40	**49**	45	**55**
PM_{coarse} (rural) [%]	21.5	**15.4**	22.2	**11.5**	**18.1**	**22.7**	**19**	**23.1**	**25.5**	**27.2**

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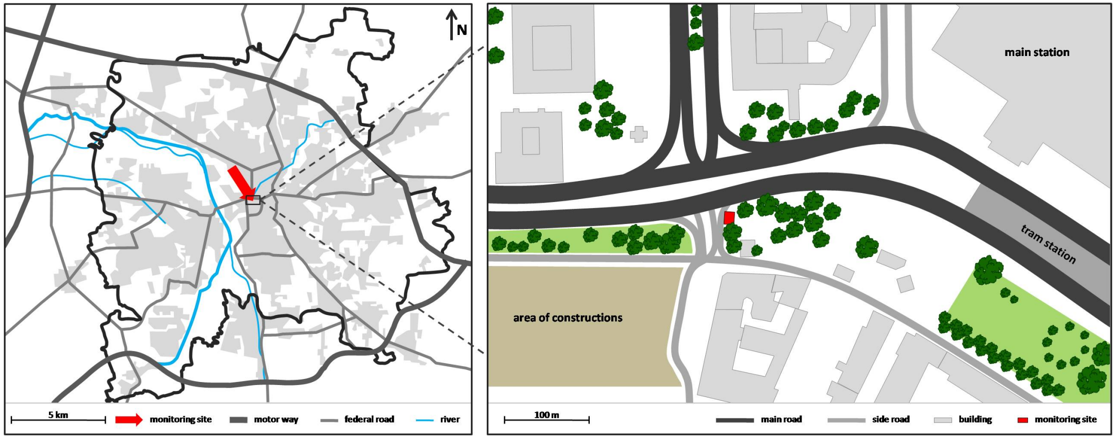

Fig. 1. Regional map of Leipzig, Germany, including the detailed surroundings of the LfULG monitoring site Leipzig-Mitte.

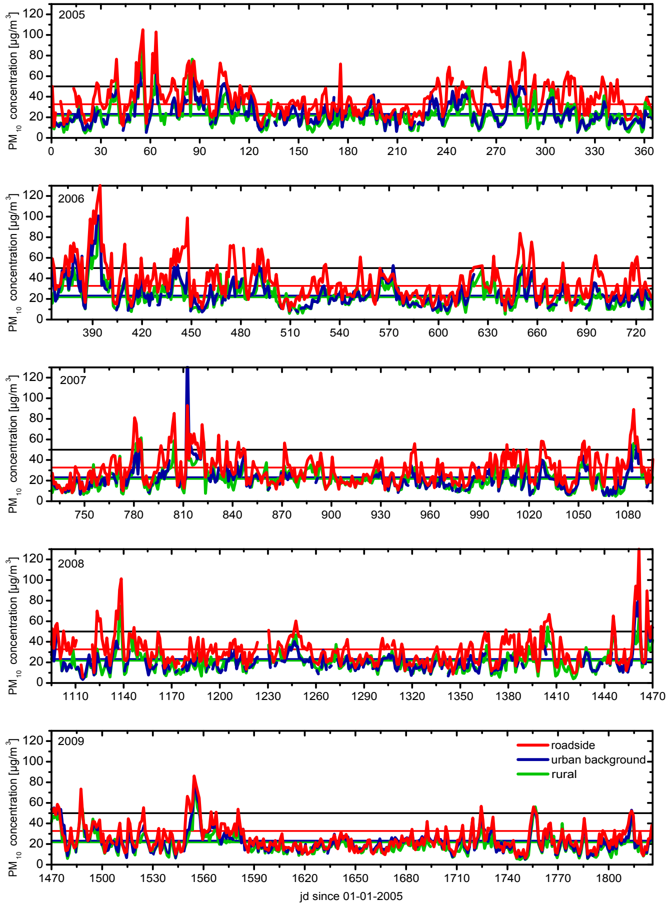

Fig. 2. PM_{10} concentrations at all three measurement sites (roadside, urban background and rural background) during five years of observation (2005 to 2009). Colored lines indicate the average values over the entire period for each station; the black line displays the limit value of 50 µg m^{-3}.

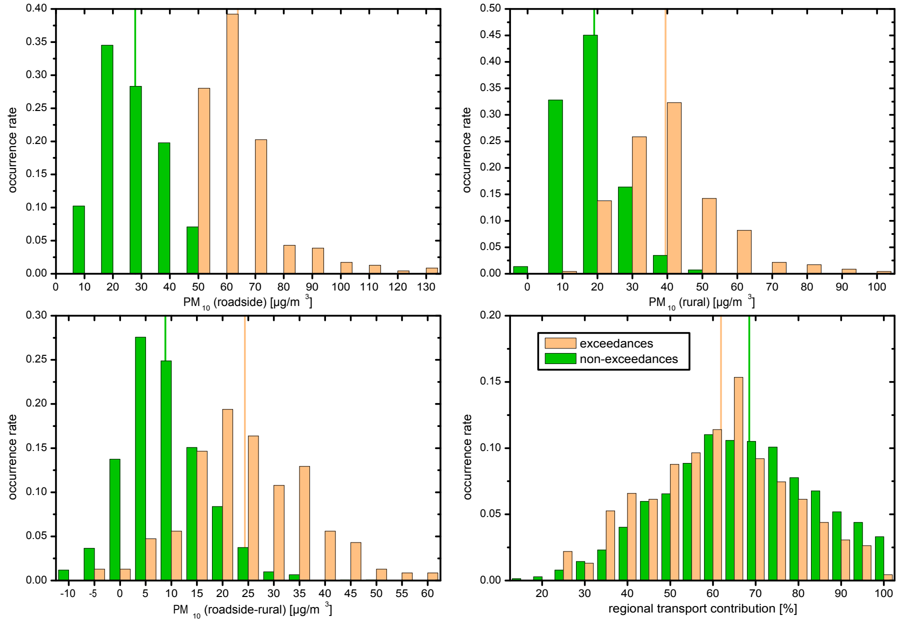

Fig. 3. Relative frequency of occurrence of **(a)** PM_{10} concentration at roadside, **(b)** PM_{10} concentration in the rural background, **(c)** absolute traffic related contribution to PM_{10} mass, and **(d)** regional contribution (relative) for exceedance and non-exceedance days at roadside. Colored vertical lines indicate corresponding median values.

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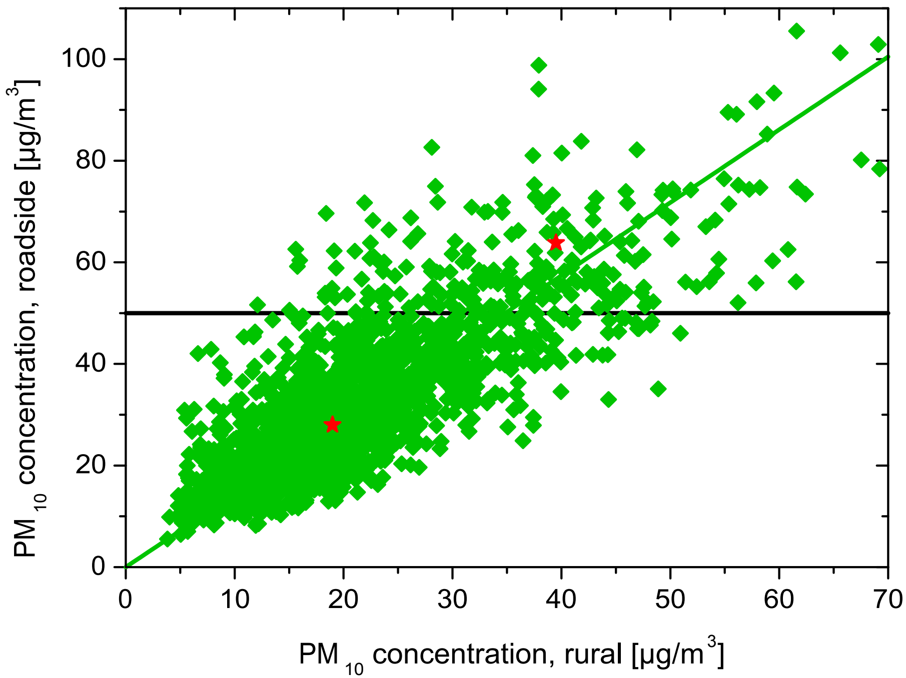

Fig. 4. Regional background vs. close-to-traffic PM_{10} mass concentration. Linear fit to all data yields in a slope of 1.43, which means, on average about 70 % of the close-to-traffic concentration can be found in rural background already. Red stars indicate the average values for exceedance and non-exceedance days, respectively.

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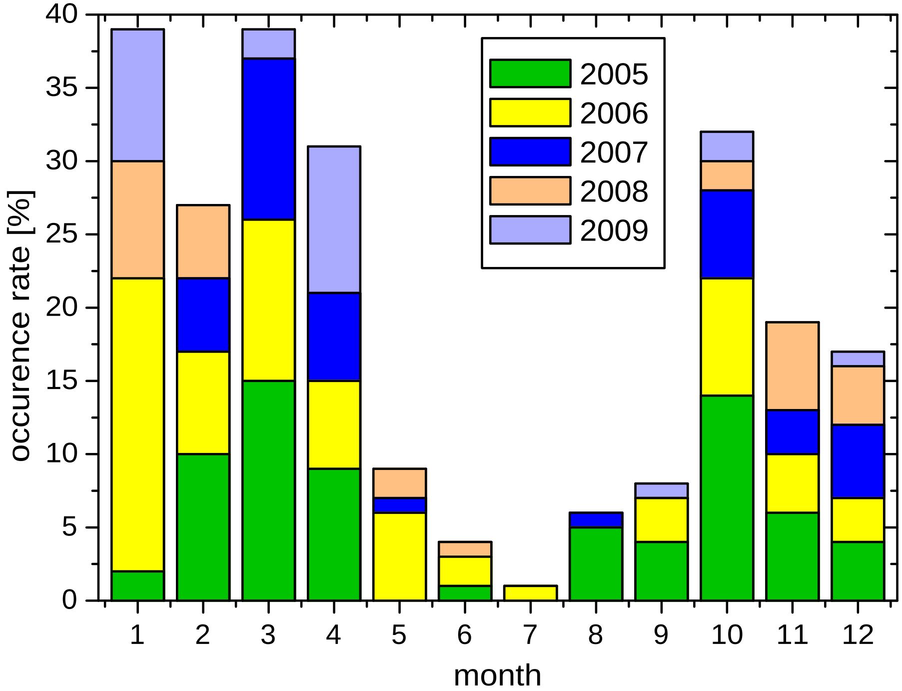

Fig. 5. Frequency of exceedances of the daily PM_{10} limit value at the roadside site Leipzig-Mitte (2005 to 2009) keyed after month of the year.

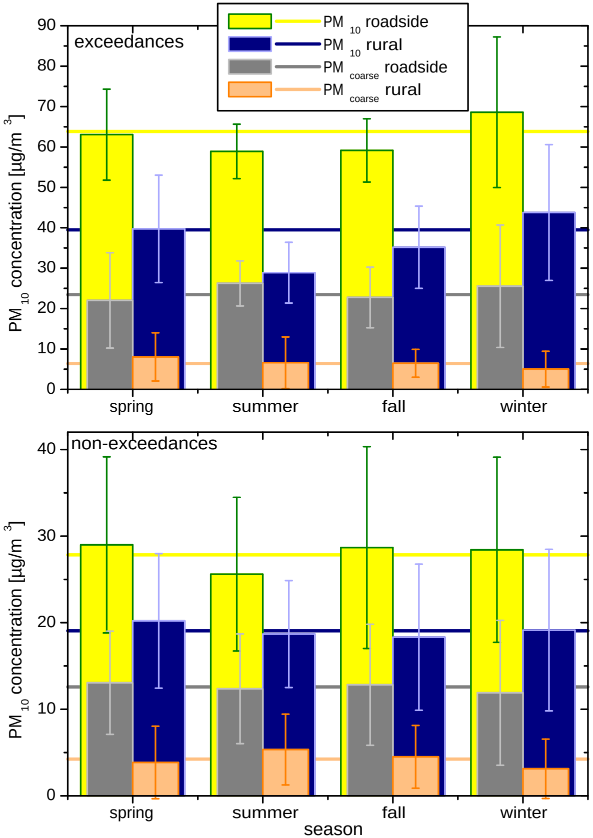

Fig. 6. Average PM_{10} and PM_{coarse} ($PM_{10} - PM_{2.5}$) concentration at roadside and regional background as well as standard deviation for **(a)** exceedance and **(b)** non-exceedance days (at the roadside station) and each season. Horizontal lines indicate the average values over the entire time period.

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Analysis of PM_{10} exceedances at roadside in Leipzig, Germany

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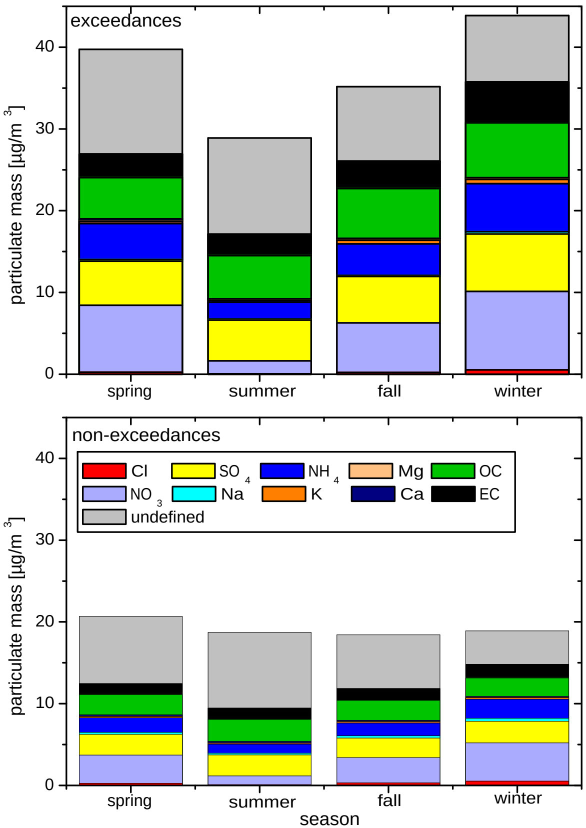

Fig. 7. Chemical composition of rural background PM_{10} for **(a)** exceedance days and **(b)** non-exceedance days (at the roadside station) and each season.

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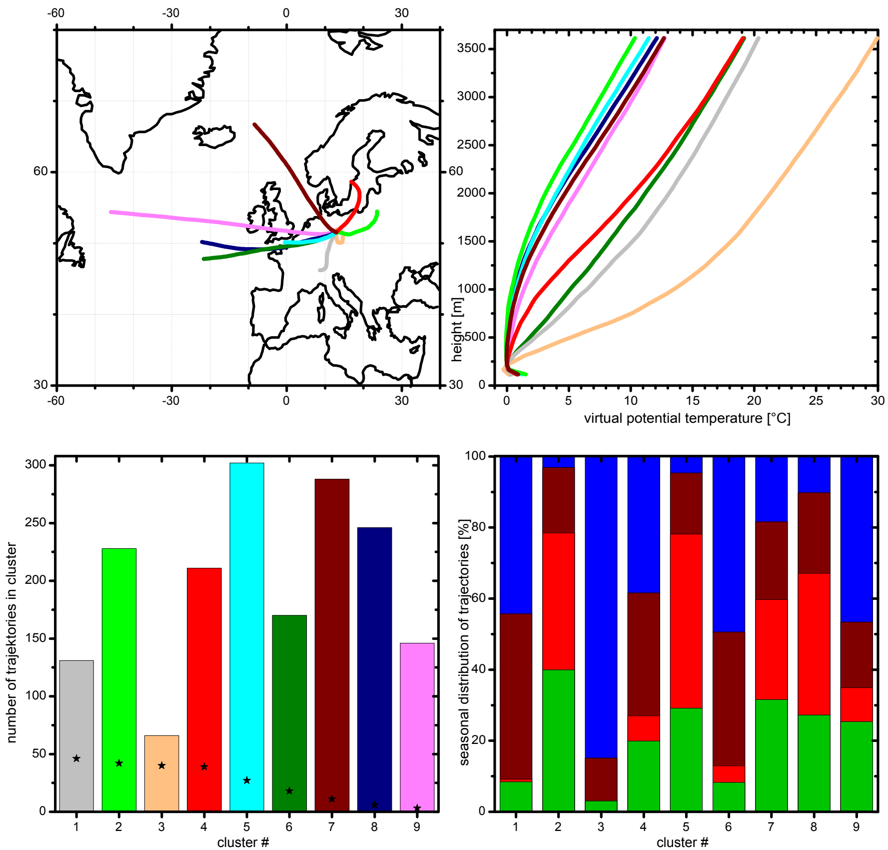

Fig. 8. Results of the cluster analysis for exceedance days. **(a)** Trajectories, **(b)** virtual pseudo potential temperature profiles, **(c)** number of days, **(d)** seasonal distribution of each cluster (green: spring, red: summer, brown: fall, blue: winter).

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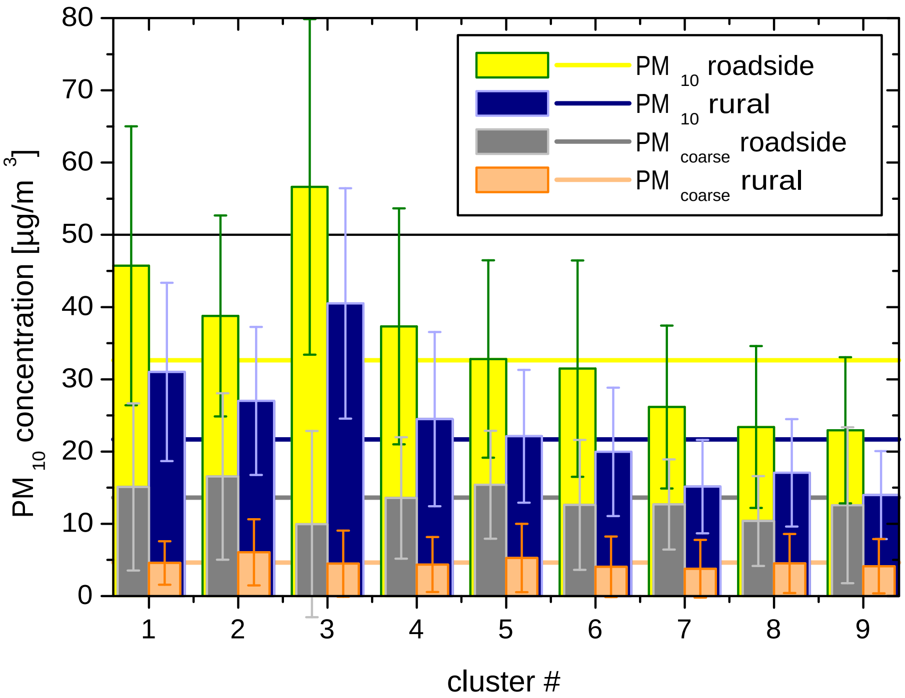

Fig. 9. PM_{10} and PM_{coarse} ($PM_{10} - PM_{2.5}$) concentration at roadside and regional background as well as standard deviation for each cluster. Horizontal lines indicate the average values over the entire time period.

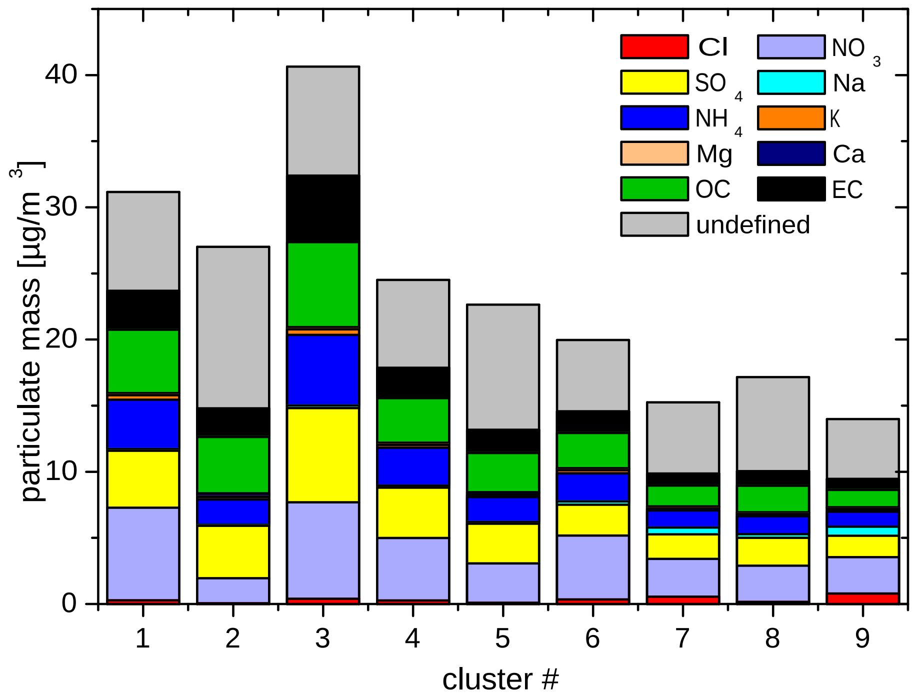

Fig. 10. Chemical composition of PM_{10} in the rural background during each cluster.

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