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Source apportionment of ambient fine particle size distribution using positive matrix factorization in Erfurt, Germany

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Abstract

Particle size distribution data collected between September 1997 and August 2001 in Erfurt, Germany were used to investigate the sources of ambient particulate matter by positive matrix factorization (PMF). A total of 29,313 hourly averaged particle size distribution measurements covering the size range of 0.01 to 3.0 μm were included in the analysis. The particle number concentrations (cm^{-3}) for the 9 channels in the ultrafine range, and mass concentrations ($ng\ m^{-3}$) for the 41 size bins in the accumulation mode and particle up to 3 μm in aerodynamic diameter were used in the PMF. The analysis was performed separately for each season. Additional analyses were performed including calculations of the correlations of factor contributions with gaseous pollutants (O_3, NO, NO_2, CO and SO_2) and particle composition data (sulfate, organic carbon and elemental carbon), estimating the contributions of each factor to the total number and mass concentration, identifying the directional locations of the sources using the conditional probability function, and examining the diurnal patterns of factor scores. These results were used to assist in the interpretation of the factors. Five factors representing particles from airborne soil, ultrafine particles from local traffic, secondary aerosols from local fuel combustion, particles from remote traffic sources, and secondary aerosols from multiple sources were identified in all seasons.

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1. Introduction

Numerous epidemiological studies have shown elevated morbidity and mortality in association with particulate air pollution (Dominici et al., 2006, HEI, 2003). However, the evidence about particle sources leading to the observed adverse effects is still limited. Only few studies have investigated the health effects of source-specific particulate matter (PM) (Laden et al., 2000, Ito et al., 2006, Mar et al., 2006). Thus, developing additional metrics that would permit the assessment of the health impacts of specific major sources would help further identify those sources that are most important to control if adverse human health effects are to be minimized.

Various methods can be used to investigate the PM sources. Positive matrix factorization (PMF) (Paatero, 1997) has been used successfully to identify sources of airborne fine particles in many studies (Lee et al., 1999, Ramadan et al., 2000, Polissar et al., 2001, Kim et al., 2003). These studies mainly focused on chemical composition of PM. To obtain the compositional data is often time-consuming and expensive. Besides the monitoring instruments for particle concentrations, additional capacities (such as Proton induced X-ray emission [PIXE] or X-ray fluorescence [XRF]) to determine the chemical composition are needed. Epidemiological studies often require long measurement periods that further increase the costs of this type of study.

Recently, PMF has been applied successfully on particle size distribution data to identify sources of PM in Seattle (Kim et al., 2004), Pittsburgh (Zhou et al., 2004, Zhou et al., 2005), Baltimore (Ogulei et al., 2006), Rochester (Ogulei et al., 2007a) and Buffalo (Ogulei et al., 2007b). This type of data is easier and more cost-efficient to collect and therefore, provides a cost-effective basis to explore health effects of source-specific PM in epidemiological studies.

The aim of this study was to identify possible sources of PM in Erfurt, Germany using particle size distribution data collected in Erfurt over a long measurement period from September 1997 to August 2001. Additional data of gaseous pollutants (O_3, NO, NO_2, CO and SO_2), particle composition (sulfate, organic carbon (OC) and elemental carbon (EC)) and meteorological parameters (wind direction and wind speed) were used to assist interpretation of the source factors. The results of this source apportionment were used used in an epidemiological study to explore adverse health effects of source-specific PM (Yue et al., 2007). This is the first instance of using source apportionment based on particle size distributions in health effects modeling.

2. Methods

2.1 Study area and measurement site

Erfurt is the capital of the German state of Thuringia and has roughly 200,000 inhabitants. It is located 200 m above sea level and surrounded by ridges 100 meters higher on three sides except towards the north. The monitoring site was situated in a residential area 1 km south of the city center and approximately 30 m west to the nearest major road (see Fig.1). The measurement station was surrounded to the south by neighborhood buildings that were about 10 m height.

2.2 Mobile aerosol spectrometer (MAS) data

Number concentrations (NCs) of PM in different size classes were determined by an mobile aerosol spectrometer (MAS) between September 1997 and August 2001. The MAS was a combination of two, separate, commercially available instruments that measure different size ranges. The system included: a differential mobility particle spectrometer (DMPS) and an optical laser aerosol spectrometer (PMS model LAS-X, Boulder, CO). These instruments used at the Erfurt site have been described in more detail elsewhere (Tuch et al., 1997, Tuch et al.,

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2000). Particles between 0.01 and 0.5 μm (geometric diameter) were quantified using the DMPS consisting of a differential mobility analyzer (DMA, TSI model 3071, TSI Inc., St. Paul, MN) combined with a condensation particle counter (CPC, TSI model 3010, TSI Inc., St. Paul, MN) at an aerosol flow rate of 0.61 l/min and a relative humidity (RH) of the sheath-air flow of 45±10% (Tuch et al., 2003). Particles with optical diameters between 0.1 and 3.0 μm were measured with the LAS-X. The overlap range (0.1-0.5 μm) was used to check the agreement between both size distributions (measurements were discarded if the measured values differed by more than 20 %). For the calculation of the final size distribution the DMPS measurements were used for particles with a diameter between 0.01 and 0.1 μm and LAS-X measurements for particles with a diameter between 0.1 and 2.5 μm.

The LAS-X classifies particles according to their scattered light into size-dependent channels. The intensity of light scattered by a particle depends on the particle diameter, shape, and optical properties such as the refractive index of the particle material. To classify ambient aerosol particles of unknown refractive indices by the LAS-X, a calibration procedure was applied using the combined MAS data. In the calibration, the DMA was used to select mono-mobile fractions of the ambient aerosol in the size range from 0.1 to 0.5 μm and then the single charged particles of these fractions were used to calibrate the LAS-X in terms of mobility equivalent diameters once a week (Liu et al., 1974, Knutson et al., 1976, Pitz et al., 2001, Tuch et al., 2003). The LAS diameter calibration curve was determined by the manufacturer for the polystyrene latex particles (PSL). Because the ambient particles have different refractive index compared to PSL particles, a mobility calibration was necessary. The deviation between the calibration curve for the PSL particles and the calibration curve for the ambient particles usually ranged between 10-15%.

As the refractive index of the ambient particles was not stable over the whole study period, we repeated the mobility calibration every week. We observed only minor variations between two adjacent mobility calibrations so that the new estimated calibration factor was assumed to be valid for the whole next week (until the next mobility calibration has been carried out). In addition, the performance of the LAS-X was tested monthly using mono-disperse polystyrene latex (PSL) particles with a diameter of 0.304 μm.

The particle NCs were measured every 6 minutes and averaged to hourly means. The mass concentrations (MCs) were determined from NCs assuming spherical particles of a constant mean density of 1.53 g cm^{-3} (Wichmann et al., 2000, Tuch et al., 1997).

Because of the mobility calibration, the size ranges of the 45 LAS channels normally have changed from calibration cycle to calibration cycle that was usually a week. Therefore, for the analysis of the data, 42 fixed particle size classes were created within the size range of the LAS. The actual LAS channel diameter was assigned to the closest particle size class. In case two LAS channels were assigned to one size class, a weighted mean was used. The 42nd size class (2.84-3.0 μm) was excluded because a large number of values were missing. Particles below 0.1 μm were described by the 9 DMPS channels that had fixed centroid diameters and fixed size ranges. As a result, 50 particle size classes were used in the PMF analysis in the size range from 0.01 to 2.84 μm (geometric diameter).

2.3 Additional data

Hourly means of gaseous pollutants (O_3, NO, NO_2, CO and SO_2) measured during the study period were obtained from a state-run station located 2 km away from the particle monitoring site. Nitrogen oxide concentrations were measured by chemiluminescence with an NO/NO_X monitor (Environment SA model AC31M). CO concentrations were measured by infra-red light absorption (Monitor Labs ML 8830). The sulfur dioxide concentrations were measured

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with a UV-fluorescence SO_2-monitor (Environment SA AF21M). All gaseous pollutants were measured continuously.

The particle composition data were only available during parts of the study period. Sulfate was measured every other day between September 1997 and December 1998. For the measurements of sulfate, a Harvard Impactor (Marple et al., 1987) having a sharp 50% cutoff at d_a = 2.5 μm was employed. The particles were sampled on Teflon membrane filter with a pore size of 2.0 μm. The sample volume was determined with a dry gas meter, and sample flow was checked with a calibrated rotameter. The filters were extracted with 0.001N $HClO_4$ and the extracts of the filters were analyzed for sulfate by ion chromatography using a Dionex Model DX 100 (Koutrakis et al., 1988, Cyrys et al., 1995).

Organic (OC) and elemental (EC) carbon values were measured hourly from January 2001 onwards. The measurements of EC/OC on $PM_{2.5}$ were done by an Ambient Carbonaceous Particulate Monitor (ACPM, R&P 5400) (Rupprecht et al., 1995). The R&P 5400 measures elemental and organic carbon contained in particulate matter at averaging times as short as one hour. Ambient air passes through a $PM_{2.5}$ size selective inlet before entering the instrument. The monitor contains two cartridges located in temperature-regulated ovens to collect the sampled particulate matter. While one cartridge is being used for particle collection, the instrument performs its thermal-CO_2 analysis upon the previously-collected particulate matter contained in the other collector. Using a direct thermal-CO_2 technique, the instrument differentiates between organic and elemental carbon particulate matter by oxidizing collected samples at an intermediate temperature (350°C) and at a high final burn temperature (750°C). In addition, hourly means of wind speed and wind direction were measured throughout the study period at our monitoring site.

2.4 Data quality assurance and control

We took great care in quality control. The data presented here is among the longest and best maintained records of continuous measurements of urban aerosols in Europe. However, even with the greatest care that is possible to be maintained over several years it can not be completely ruled out that some factors will affect the data. To limit their impact we performed extensive quality assurance procedures throughout the whole study period. They included daily check of all important parameters (e.g. flows, pressure drop, laser reference voltage, etc.) as well as the verification of the MAS performance during several side-by-side intercomparisons with other aerosol spectrometers (Tuch et al., 2000, Khlystov et al., 2001, Mirme et al., 2002). For quality control purposes we continuously used side-by-side integral measuring devices: number concentration was determined by a condensation particle counter (CPC, TSI model 3022) and the mass concentration by a Harvard $PM_{2.5}$ Impactor (Marple et al., 1987).

The squared correlation (R^2) between $NC_{2.5}$ measured by MAS and NC measured by CPC 3022 was 0.76 for the study period 1996 – 1999 and 0.91 for the study period 2000 – 2001 illustrating a strong correlation between the results obtained by the two different measurement methods. The regression equation was NC (MAS) = 0.63*NC (CPC) + 4300 for the years 1996 – 1999, and NC (MAS) = 0.63*NC (CPC) + 582 for 2000 – 2001. The lower NC levels obtained by MAS (DMPS) could be mainly attributed to the lower detection limit of the CPC3022 (7 nm) in comparison to the DMPS (10 nm) and to particle losses in the DMA.

The comparison between the $PM_{2.5}$ mass concentrations measured by MAS and the Harvard Impactor (HI) should be also considered separately for two measurement periods. Until October 6th, 1999 the Harvard Impactor collected $PM_{2.5}$ from 11 am the day before to 11 am same day, whereas the daily means for $PM_{2.5}$ derived from the aerosol spectrometer were computed from midnight to midnight. From October 7th, 1999 onwards the Harvard impactor filters were changed automatically at midnight to match the whole day as the basic unit used

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in the epidemiological analysis. The squared correlation (R^2) between $PM_{2.5}$ measured by MAS and $PM_{2.5}$ measured by HI varied between 0.77 and 0.83 from year to year in the first period (until October 6th, 1999), and was 0.81 for the second period from October 7th, 1999. It indicated a strong correlation between the LAS measurement technique and a gravimetrical method such as Harvard Impactor. The lower MC obtained by HI (data not shown) could be mainly attributed to losses of volatile and semivolatile components during a 24 hours sampling period on the Teflon filter (Harvard Impactor).

For the proper sizing of the ambient particles with unknown refractive indices by the LAS-X, the calibration procedures to the MAS described above was applied to the data.

The CO_2-sensor of the R&P 5400 was calibrated on a monthly base. A two-point calibration with a certified purge air gas bottle (20 % O_2 and 80 % N_2) and a certified CO_2 span gas bottle (4000 ppm) was performed. All devices for gaseous pollutants measurement were checked by the manufacturer on a regular base and calibrated with span gas once a week.

2.5 Positive matrix factorization (PMF)

PMF is an extended factor analytical method (Paatero, 1997). A two-way PMF model using robust mode was applied in this analysis. The application of PMF depends on the estimated uncertainties for each of the measured data. In this study, because the uncertainties were not provided by the measurement device, 10% of the measured concentrations were assumed as analytical uncertainties. For a few extreme outliers identified by visual inspection of the plots, the uncertainties were set to the respective concentration values to reduce their weights in the data analyses.

A total of 50 variables representing different particle sizes were included in the PMF analysis. The particle NCs (cm^{-3}) were used for the first 9 variables covering the ultrafine size range (0.01-0.10μm), and MC values (ng m^{-3}) were used for the 41 size classes of accumulation mode and fine particles (0.10-2.84 μm). The default FPEAK value of 0 was used in the PMF analysis.

The whole data set was divided into four seasons. Based on local temperature patterns, the seasons were defined as follows: spring (March, April and May), summer (June, July and August), autumn (September, October and November 1st -15th) and winter (November 16th-30th, December, January and February). Initially, we analyzed the data stratified for season and year to account for seasonal variations in the aerosol size distribution as well as to detect a potential trend over time in the aerosol size distribution. These initial analyses revealed a remarkable similarity of the factor patterns for each season over time. Therefore, we decided to derive and present results for the whole study period, stratified by season. Solutions with different numbers of factors were tested. Based on plausibility and interpretability of these solutions a model with five factors was selected for each season.

We refrained from including gaseous pollutants in the PMF analyses because for SO_2, more than 80 % of the measurements were close to the detection limit that might distort the factor patterns because of their low signal-to-noise ratio. Particle composition data (sulfate, EC, OC) were not included in the PMF analysis because they were available only for parts of the study period (EC, OC) or because they were obtained only on a daily basis instead of hourly basis (sulfate). These data were used as described in a subsequent section to aid in the interpretation of the PMF factors.

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2.6 Conditional probability function (CPF)

To assist interpretation of the source factors, contribution values were associated with wind directions using the conditional probability function (CPF) (Ashbaugh et al., 1985, Kim and Hopke, 2004). CPF values were calculated by the following equation:

$$CPF=\frac{m_{\Delta\theta}}{n_{\Delta\theta}} \quad (1)$$

where $m_{\Delta\theta}$ is the number of occurrences in the wind direction sector $\Delta\theta$ that exceeds the threshold which was defined as the upper 25th percentile of the fractional contribution from each source, and $n_{\Delta\theta}$ is the total number of wind occurrences in the same wind direction sector. When $n_{\Delta\theta}$ is below 10, the CPF value is set to zero. The width of wind direction sector was set to 10° and there were 36 directional sectors in total. Hours with calm wind (<1 m s^{-1}) were excluded from the CPF analysis (n=18,757, 58% of the total). The sources are likely to be located in the direction sectors with high CPF values.

2.7 Additional analyses

The correlations of five factors with gaseous pollutants (O_3, NO, NO_2, CO and SO_2) and particle composition data (sulfate, OC, EC) were utilized to assist in the interpretation of the sources. Correlations were classified as 'weak' (0.2-0.5), 'moderate' (0.5-0.8) or 'high' (≥0.8). Regression analyses between factor contributions and total particle number or mass concentration were performed to calculate the contribution of each factor to the total number or mass, respectively.

3. Results and Discussion

A total of 29,313 hourly samples (84% available), each with 50 different size channels covering the size range of 0.01 to 2.84 μm were included in the PMF analysis. The concentrations of particle size channels, gaseous pollutants and particle composition are summarized in Table 1. Five factors in each season were obtained from PMF analysis. The correlations of each factor with gaseous pollutants and particle composition can be seen in Table 2. All factors characterized by different size ranges in each season are shown in Fig.2. The regression results indicating the contribution of each factor to total NC and MC for each season are shown in Table 3. The distribution of the wind direction and the CPF values for each factor are presented in Fig.3 to indicate the possible direction of the sources. The wind comes predominantly from the west. However, the threshold used by the calculation of the CPGF values was more often exceeded when the wind comes from other direction as from the west. The plots clearly illustrate that air pollution levels should be elevated when the wind is coming from the east (which is not so often the case). Note that data from hours with no or little wind were not accounted by the plots. However, such data were also used to estimate the factors and might be responsible for the episodes with high concentrations.

In addition, the distributions of average 24 1-hour factor concentrations (Fig.4) shows the diurnal pattern of each factor on weekdays and weekends. Finally the characteristics of all five factors are summarized in Table 4.

Factor 1 was characterized by the larger particles in the size range between 1.0 and 2.84 μm. This factor did not correlate with the measured gaseous pollutants or particle composition. It did not contribute to the total NC because of its large size, but accounted for 8% of the total MC. The source of this factor was located southeast and northwest from the measurement station that coincided with the dominant wind directions at the monitoring site. As can be seen from the Fig. 4, factor 1 had similar diurnal patterns on weekdays and weekends. Based on its size distribution, factor 1 probably reflects the crustal content of fine particles that would be

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expected to come primarily from resuspended road dust, considering that the site is near a busy intersection with high traffic intensity what could contribute to the resuspension of the airborne soil in the roadway. The concentration peak around 10:00am of this factor supports the impact of human activities, especially of resuspended road dust.

Factor 2 was dominated by the ultrafine size range between 0.01 and 0.1μm. The shape was very consistent throughout all four seasons. This factor showed moderate Spearman correlation coefficients with NO, NO_2 and CO in all four seasons (Table 2). Factor 2 accounted for 78% of the total NC but only 4% of the total MC. Highest concentrations were observed during the morning and to a lesser extent during the afternoon rush hours when there was a higher mixed layer height. The concentration peaks on weekends were lower and occurred later than those on weekdays.

The presence of ultrafine particles and the observed correlations reflect traffic emission particles. Studies show that ultrafine particles are mainly due to motor vehicle emissions and exhibit a high correlation with NO_x and CO (Morawska et al., 1998a, Morawska et al., 1999, Shi et al., 1999, Cyrys et al., 2003, Jeong et al., 2004). The identified location of the source for factor 2 was to the southeast of the measurement station, where there is a major road about 30 m away from the monitoring site. This proximity supports the assignment of factor 2 to fresh motor vehicle emissions from local traffic such as gasoline-powered cars or light trucks. The pattern of this factor in Erfurt is similar to the local traffic factor found in the Pittsburgh particle size distribution study (Zhou et al., 2004, Zhou et al., 2005). Also the lower concentration and the later occurrence of the peak concentrations on weekends compared with weekdays are coinciding with the lower volume of traffic and later time schedule of human activities on weekends.

In spring, summer and autumn, factor 3 primarily included particles between 0.25 and 0.60 μm. In winter, its size ranged from 0.5 to 1.8 μm. It was weakly correlated with OC and EC. Note that no data on OC and EC was available for the autumn period and thus, correlation coefficients could not be calculated. It had weak correlations with NO_2 and CO in spring and summer and with SO_2 and sulfate in autumn and winter. This factor did not contribute to the total NC, but on average, represented 17% of the total MC. The directionality of this source was located in the two main wind directions of the monitoring site. It exhibited a relatively weak diurnal pattern and higher concentrations on weekends.

Factor 3 may reflect secondary aerosols including secondary sulfate and nitrate. It may also include OC and EC such as any transported wildfire smoke. Although the local SO_2 concentrations are lower in spring and summer, this factor might contain sulfate from distant sources.

In autumn and winter the hills surrounding the city of Erfurt favor temperature inversions that cause elevated air pollution levels. Therefore, this factor could also reflect secondary particles from local fuel combustion for space heating purposes. The local heating in winter is to a large extent provided from a power plant several kilometers northeast of the city center which also produces community heating. At the beginning of the study period, small amounts of surface coal (with 1.7% sulfur content) were still used as the fuel but were completely replaced by natural gas as of January 1999 (Ebelt et al., 2001, Vallius et al., 2005). When analyzing the data year by year, a trend was observed in the average particle diameter of Factor 3 towards smaller size ranges in winter similar to the other seasons (data not shown). It indicates that in winter, the secondary aerosol might be overshadowed by the heating emissions.

In addition, factor 3 exhibited an apparent anomaly at 0.63 μm that coincides with the transition between NC LAS channel 2 and channel 3. Therefore, it is possible that this factor also reflects to some extent the mobility calibration of the two measurement devices that might exhibit

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different sensitivities. However, no systematic interrelation could be established between mobility calibration days and potential anomalies in the time series of Factor 3.

Factor 4 peaked between 0.1 and 0.5 μm and between 0.01 and 0.1 μm. This factor was most often moderately correlated with NO, NO_2, OC, and EC. Except in the summer, there were also moderate or weak correlations with CO, SO_2, and sulfate. This factor contributed to both total NC (15%, except in summer) and MC (39%). It exhibited a pronounced morning peak. The peaks on weekends were less pronounced and later than on weekdays.

Thus, factor 4 could be also assigned to traffic-related particles, possibly rather aged particles from more distant areas of the city and from diesel engines such as delivery trucks and commuter buses. The correlation with EC supports this hypothesis. In addition, evidence from other studies has shown that diesel vehicles typically emit particles in the accumulation mode range (diameter ≤ 1 μm) (Morawska et al., 1998b) and a large number of ultrafine particles in the nuclei mode (diameter ≤ 0.05 μm) (Gertler et al., 2002). The principal mechanism producing the nuclei mode particles is shown to be Brownian coagulation between small primary particles during the combustion. The larger size modes are probably formed by the condensational growth of organic compounds by smaller mode particles (Kerminen et al., 1997). Uncontrolled diesel engines emit high concentrations of particles and NO_x and low concentrations of CO and hydrocarbons. Chemically, diesel particles consist primarily of OC and EC, followed by sulfate and various trace elements (Docekal et al., 1992, HEI, 2002). The characteristics of factor 4 showed similar size distribution and components as these previous study results. The CPF analysis indicated that this factor was from both east and west. There is a major road immediately east of the measurement station. Other major traffic routes are several hundred meters to the west of the measurement site. This factor is also similar to the traffic related factor found in the Seattle particle size distribution study (Kim et al., 2004).

During spring, autumn and winter, factor 5 peaked mainly in the accumulation mode around 0.3 μm. In summer, it exhibited three peaks in the ultrafine, accumulation and fine mode, respectively. This factor was not correlated with the gaseous pollutants or particle composition in spring and summer. In autumn and winter, there were limited correlations with the gases, sulfate, OC, and EC. This factor accounted for 31% of the total MC, but it did not contribute to the total NC except in summer (6%). Factor 5 exhibited a weak diurnal pattern and no differences between weekdays and weekends.

In summer, factor 5 showed two additional peaks in the ultrafine and fine mode. The peak in the ultrafine range might come from UFP that were assigned mainly to factor 4 in other seasons. In summer, however, factor 4 was loaded weaker on UFP. These findings suggest that Factor 5 is due to secondary particles, mainly from an easterly direction. In autumn and winter, the origin of this factor might be particles from combustion and traffic containing sulfate, OC, and EC, since the correlations with factor 3 (combustion-related) and factor 4 (traffic-related) in autumn and winter (r in the range of 0.44 to 0.58) are relatively high. In summer, factor 5 might also contain some contribution from the road dust or airborne soil that would explain the peak above 1 μm. Also, there was a significant correlation with factor 1 in summer (r=0.35).

The large amount of data with nearly 30,000 hourly samples within four years included in the analysis is a powerful strength of this study. The separated analysis within each season not only shows similar patterns of factors, but also indicates individual characteristics of particle sources in the different seasons. A combination of NC and MC for the different particle size channels used in the PMF analysis is a new approach in the particle size distribution source apportionment analysis. All five factors explained 94% of the total NC and 99% of the total MC.

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The use of a single monitoring site for the whole city of Erfurt may pose a limitation of these data to provide exposure estimates for the entire city population. However, this site has been demonstrated to be representative for the air quality within the city of Erfurt with respect to PM_{10} (PM diameter ≤ 10μm) and sulfate (Cyrys et al., 1998). The wind direction and weather condition at the monitoring site could more or less impact the observed dominant sources. Another limitation of the study is that the uncertainty values were not available. Thus, only estimated values based on reasonable assumptions were used in the PMF analysis. In addition, if two sources have very similar variations over time, factor analytical method may have difficulty to separate them explicitly. An example might be the considerable overlap between factors 3 and 5 in this study. Although these factors were named accordingly, they may still contain other unidentified sources.

Unlike in our previous studies (Wichmann et al., 2000, Kreyling et al., 2003), this study was not restricted to $PM_{2.5}$ (PM with aerodynamic diameter ≤ 2.5 μm) but the whole particle size distribution covered by the LAS with the exception of its largest channel, i.e. particle size distribution data between 0.01 and 2.84 μm (geometric diameter), was analyzed. This approach was used to better interpret coarse mode particles. This coarse mode fraction has been attributed principally to Factor 1. Note that Table 3 refers to the total mass or total number, respectively, summed over all channels analyzed in this study, i.e. up to 2.84 μm in geometric diameter that corresponds to aerodynamic diameters up to 3.5 μm. However, the contribution of the size range beyond 2.5 μm (aerodynamic diameter) to the total number is negligible. The contribution to the total mass is small: on average 2.6 % (95% percentile: 7.8%). Therefore, the total MC in this study deviates only marginally from the standard measure of $PM_{2.5}$. To better detect mechanically generated coarse mode particles in future source apportionment studies even larger size ranges up to 10 or 20 μm might be considered if such data is available.

4. Conclusions

In this study, particle size distribution data were included in the PMF analysis to investigate possible sources for ambient fine particles ($PM_{2.5}$) in Erfurt, Germany. The analysis was performed for each season separately. Five factors were identified that were similar for each season. In addition, diurnal patterns of factor scores, the correlations of the factor contributions with gaseous pollutants (O_3, NO, NO_2, CO and SO_2) and particle composition (sulfate, OC, and EC) data were used to assist interpretation of the sources. The five sources were identified as airborne soil, ultrafine particles from local traffic, secondary aerosols from local fuel combustion, remote traffic, and secondary aerosols from multiple sources. Further comparison of the analysis results with chemical composition data might provide more specific source information. The results will be used in epidemiological studies to explore adverse health effects of source-specific PM.

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References

Ashbaugh LL, Malm WC, Sadeh WZ. A residence time probability analysis of sulfur concentrations at Grand Canyon national park. Atmos. Environ 1985;19:1263–1270.

Cyrys J, Gutschmidt K, Brauer M, Dumyahn T, Heinrich J, Spengler JD, Wichmann HE. Determination of acidic sulfate aerosols in urban atmospheres in Erfurt (F.R.G.) and Sokolov (former C.S.S.R.). Atmos. Environ 1995;29(23):3545–3557.

Cyrys J, Heinrich J, Brauer M, Wichmann HE. Spatial variability of acidic aerosols, sulfate and PM10 in Erfurt, Eastern Germany. J. Expo. Anal. Environ. Epidemiol 1998;8:447–64.

Cyrys J, Stölzel M, Heinrich J, Kreyling WG, Menzel N, Wittmaack K, Tuch T, Wichmann HE. Elemental composition and sources of fine and ultrafine ambient particles in Erfurt, Germany. Sci. Total Environ 2003;305:143–56. [PubMed: 12670764]

Docekal B, Krivan V, Pelz N. Trace and minor element characterization of diesel soot. Fresenius J. Anal. Chem 1992;343:873–878.

Dominici F, Peng RD, Bell ML, Pham L, McDermott A, Zeger SL, Samet JM. Fine particulate air pollution and hospital admission for cardiovascular and respiratory diseases. J. Am. Med. Assoc 2006;295:1127–1134.

Ebelt S, Brauer M, Cyrys J, Tuch T, Kreyling WG, Wichmann HE, Heinrich J. Air quality in postunification Erfurt, East Germany: Associating changes in pollutant concentrations with changes in emissions. Environ. Health Perspect 2001;109:325–333. [PubMed: 11335179]

Gertler, AW.; Gillies, JA.; Pierson, WR.; Rogers, CF.; Sagebiel, JC.; Abu-Allaban, M.; Coulombe, W.; Tarnay, L.; Cahill, T. Res. Rep. Health Eff. Inst. Boston MA: 2002. Real-world particulate matter and gaseous emissions from motor vehicles in a highway tunnel. In: Emissions from Diesel and Gasoline Engines Measured in Highway Tunnels.

HEI Health Effects Institute. Res. Rep. Health Eff. Inst. Boston, MA: 2002. Research directions to improve estimates of human exposure and risk from diesel exhaust, A special report of the institute's diesel epidemiology working group.

HEI Health Effects Institute. Res. Rep. Health Eff. Inst. Boston, MA: 2003. Revised analyses of time-series studies of air pollution and health.

Ito K, Christensen WF, Eatough DJ, Henry RC, Kim E, Laden F, Lall R, Larson TV, Neas L, Hopke PK, Thurston GD. PM source apportionment and health effects: 2. An investigation of intermethod variability in associations between source-apportioned fine particle mass and daily mortality in Washington, DC. J. Expo. Sci. Environ. Epidemiol 2006;16(4):300–310. [PubMed: 16304602]

Jeong CH, Hopke PK, Chalupa D, Utell M. Characteristics of nucleation and growth events of ultrafine particles measured in Rochester, NY. Environ. Sci. Technol 2004;38:1933–1940. [PubMed: 15112791]

Kerminen VM, Makela TE, Ojanen CH, Hillamo RE, Vilhunen JK, Rantanen L, Havers N, Bohlen AV, Klockow D. Characterization of the particulate phase in the exhaust from a diesel car. Environ. Sci. Technol 1997;31:1883–1889.

Khlystov A, Kos GPA, ten Brink HM, Mirme A, Tuch T, Roth C, Kreyling WG. Comparability of three spectrometers for monitoring urban aerosol. Atmos. Environ 2001;35:2045–2051.

Kim E, Hopke PK, Edgerton ES. Source identification of Atlanta aerosol by positive matrix factorization. J. Air Waste Manage. Assoc 2003;53:731–739.

Kim E, Hopke PK, Larson TV, Covert DS. Analysis of ambient particle size distributions using Unmix and positive matrix factorization. Environ. Sci. Technol 2004;38:202–209. [PubMed: 14740737]

Kim E, Hopke PK. Comparison between conditional probability function and nonparametric regression for fine particle source directions. Atmos. Environ 2004;38:4667–4673.

Knutson, EO. Extended electrical mobility method for measuring aerosol particle size. In: Liu, B., editor. Fine Particles. Academic Press; New York: 1976. p. 739

Koutrakis P, Wolfson JM, Spengler JD. An Improved Method for Measuring Aerosol Strong Acidity: Results from a Nine-Month Study in St Louis, Missouri and Kingston, Tennessee. Atmos. Environ 1988;22:157–162.

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Kreyling WG, Tuch T, Peters A, Pitz M, Heinrich J, Stölzel M, Cyrys J, Heyder J, Wichmann HE. Diverging long-term trends in ambient urban particle mass and number concentrations associated with emission changes caused by the German unification. Atmos. Environ 2003;37:3841–3848.

Laden F, Neas LM, Dockery DW, Schwartz J. Association of fine particulate matter from different sources with daily mortality in six U.S. cities. Environ. Health Perspect 2000;108:941–947. [PubMed: 11049813]

Lee E, Chan CK, Paatero P. Application of positive matrix factorization in source apportionment of particulate pollutants in Hong Kong. Atmos. Environ 1999;33:3201–3212.

Liu BYH, Marple VA, Whitby KT, Barsic NJ. Size distribution measurements of airborne coal dust by OCP. Am. Ind. Hyg. J 1974;35(8):443–451.

Mar TF, Ito K, Koenig JQ, Larson TV, Eatough DJ, Henry RC, Kim E, Laden F, Lall R, Neas L, Stölzel M, Paatero P, Hopke PK, Thurston GD. PM source apportionment and health effects. 3. Investigation of inter-method variations in associations between estimated source contributions of PM2.5 and daily mortality in Phoenix, AZ. J. Expo. Sci. Environ. Epidemiol 2006;16:311–320. [PubMed: 16288316]

Marple V, Rubow KL, Turner W, Spengler JD. Low flow rate sharp cut impactors for indoor sampling: Design and calibration. J. Air. Pollut. Control. Assoc 1987;37:1303–1307.

Mirme A, Kreyling WG, Khlystov A, ten Brink H, Ruuskanen J, Tuch T, Pekkanen J. Intercomparison of aerosol spectrometers for ambient air monitoring. Aerosol Sci. Technol 2002;36(8):866–876.

Morawska L, Thomas S, Bofinger N, Wainwright D, Neale D. Comprehensive characterization of aerosols in a subtropical urban atmosphere: particle size distribution and correlation with gaseous pollutants. Atmos. Environ 1998a;32:2467–2478.

Morawska L, Bofinger ND, Kocis L, Nwankwoala A. Submicrometer and supermicrometer particles from diesel vehicle emissions. Environ. Sci. Technol 1998b;32:2033–2042.

Morawska L, Thomas S, Jamriska M, Johnson G. The modality of particle size distributions of environmental aerosols. Atmos. Environ 1999;33:4401–4411.

Ogulei D, Hopke PK, Zhou L, Pancras JP, Nair N, Ondov JM. Source apportionment of Baltimore aerosol from combined size distribution and chemical composition data. Atmos. Environ 2006;40(S2):396–410.

Ogulei D, Hopke PK, Chalupa DC, Utell MJ. Modeling Source Contributions to Submicron Particle Number Concentrations Measured in Rochester, NY. Aerosol Sci Technol 2007a;41:179–201.

Ogulei D, Hopke PK, Ferro AR, Jaques PA. Factor Analysis of Submicron Particle Size Distributions Near a Major United States-Canada Trade Bridge. J. Air Waste Manage. Assoc 2007b;57:190–203.

Paatero P. Least square formulation of robust nonnegative factor analysis. Chemometrics Intelligent Lab. Syst 1997;37:23–35.

Pitz M, Kreyling WG, Hölscher B, Cyrys J, Wichmann HE, Heinrich J. Change of the ambient particle size distribution in East Germany between 1993 and 1999. Atmos. Environ 2001;35:4357–4366.

Polissar AV, Hopke PK, Poirot RL. Atmospheric aerosol over Vermont: Chemical composition and sources. Environ. Sci. Technol 2001;35:4604–4621. [PubMed: 11770762]

Ramadan Z, Song XH, Hopke PK. Identification of sources of Phoenix aerosol by positive matrix factorization. J. Air Waste Manage. Assoc 2000;50:1308–1320.

Rupprecht, G.; Patashnick, H.; Beeson, DE.; Green, R.; Meyer, MB. A new automated monitor for the measurement of particulate carbon in the atmosphere. Particulate matter: health and regulatory issues. Proceedings of an International Speciality Conference; Air & Waste Management Association; Pittsburgh, PA. 30 -31; 1995.

Shi JP, Khan AA, Harrison RM. Measurements of ultrafine particle concentration and size distribution in the urban atmosphere. Sci. Total Environ 1999;235:51–64.

Tuch T, Brand P, Wichmann HE, Heyder J. Variation of particle number and mass concentration in various size ranges of ambient aerosols in Eastern Germany. Atmos. Environ 1997;31:4193–4197.

Tuch T, Mirme A, Tamm E, Heinrich J, Heyder J, Brand P, Wichmann HE, Pekkanen J, Kreyling WG. Comparison of two particle size spectrometers for ambient aerosol measurements in environmental epidemiology. Atmos. Environ 2000;34:139–149.

Tuch T, Wehner B, Pitz M, Cyrys J, Heinrich J, Kreyling WG, Wichmann HE, Wiedensohler A. Long-term measurements of size-segregated ambient aerosol in two German cities located 100 km apart. Atmos. Environ 2003;37:4687–4700.

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Vallius M, Janssen NAH, Heinrich J, Hoek G, Ruuskanen J, Cyrys J, Van Grieken R, de Hartog JJ, Kreyling WG, Pekkanen J. Sources and elemental composition of ambient PM2.5 in three European cities. Sci. Total Environ 2005;337:147–162. [PubMed: 15626386]

Wichmann, HE.; Spix, C.; Tuch, T.; Wolke, G.; Peters, A.; Heinrich, J.; Kreyling, WG.; Heyder, J. Res. Rep.Health Eff. Inst. Boston, MA: 2000. Daily mortality and fine and ultrafine particles in Erfurt, Germany part I: role of particle number and particle mass.

Yue W, Schneider A, Stölzel M, Rückerl R, Cyrys J, Pan X, Zareba W, Koenig W, Wichmann HE, Peters A. Ambient source-specific particles are associated with prolonged repolarization and increased levels of inflammation in male coronary artery disease patients. Mutation Research 2007;621:50–60. [PubMed: 17603085]

Zhou L, Kim E, Hopke PK, Stanier CO, Pandis S. Advanced factor analysis on Pittsburgh particle size-distribution data. Aerosol Sci. Technol 2004;38(S1):118–132.

Zhou L, Kim E, Hopke PK, Stanier CO, Pandis SN. data by positive matrix factorization. J. Geophys. Res 2005;110(7):1–15.art. No. D07S19, doi:10.1029/2004JD004707

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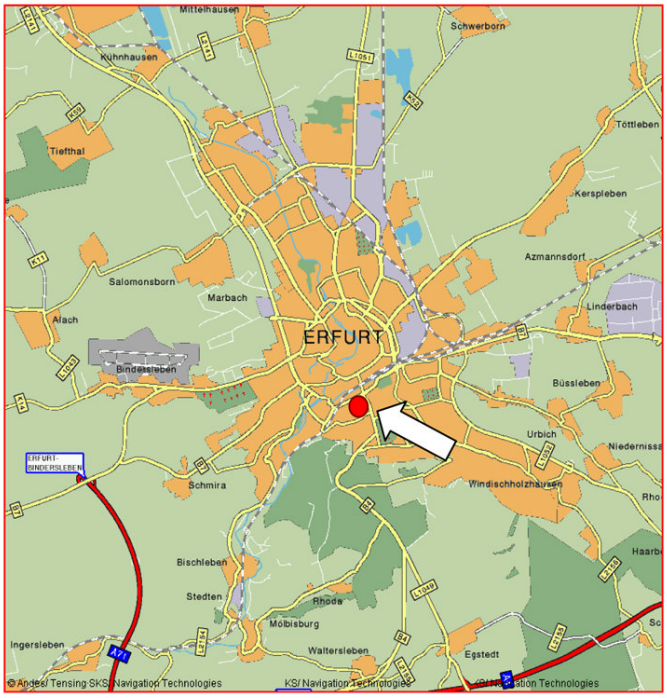
(a)



(b)

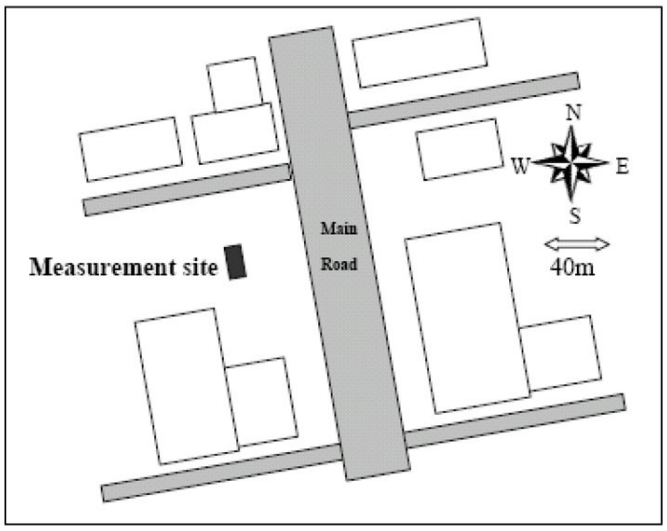


Figure 1.

(a) Erfurt city map and measurement site location (red circle);

(b) Schematic structure of measurement site.

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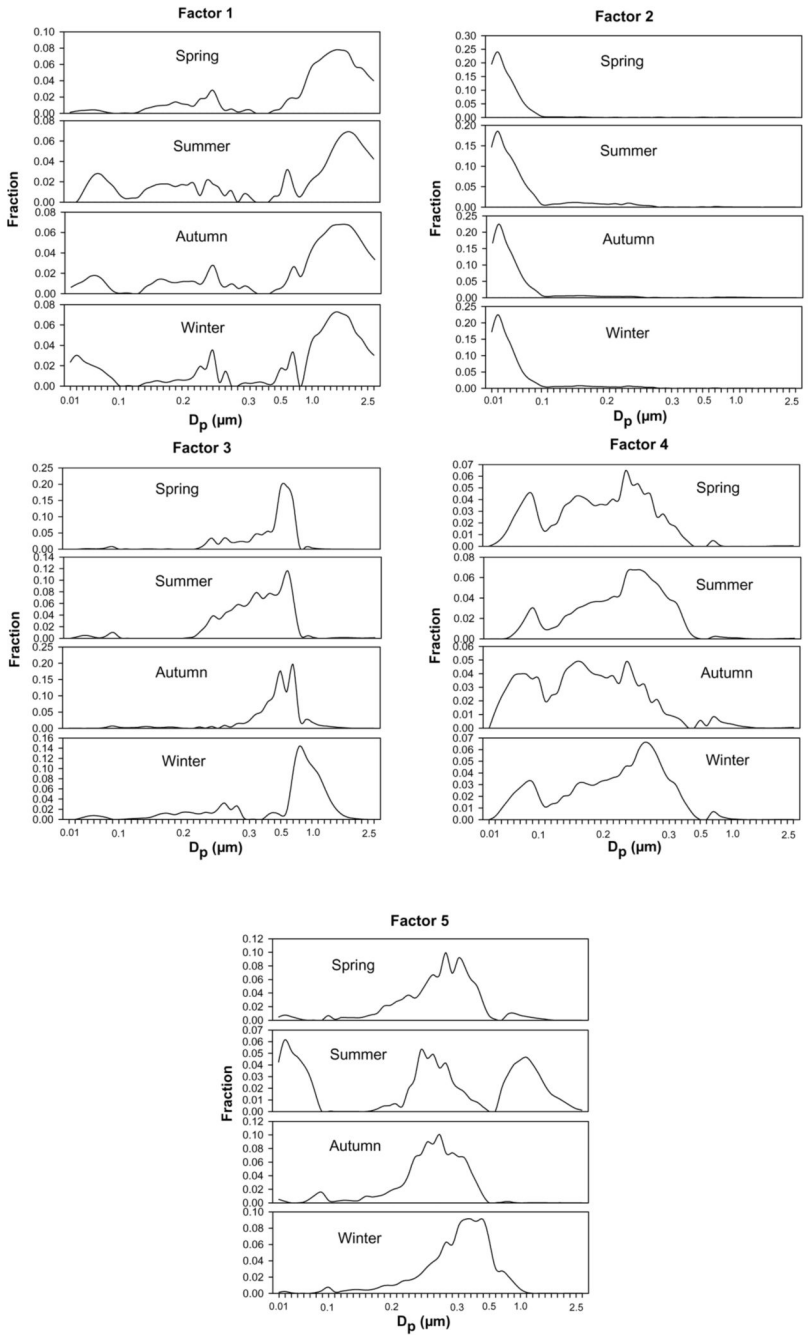

Figure 2.
Size distribution profiles of five factors in each season.

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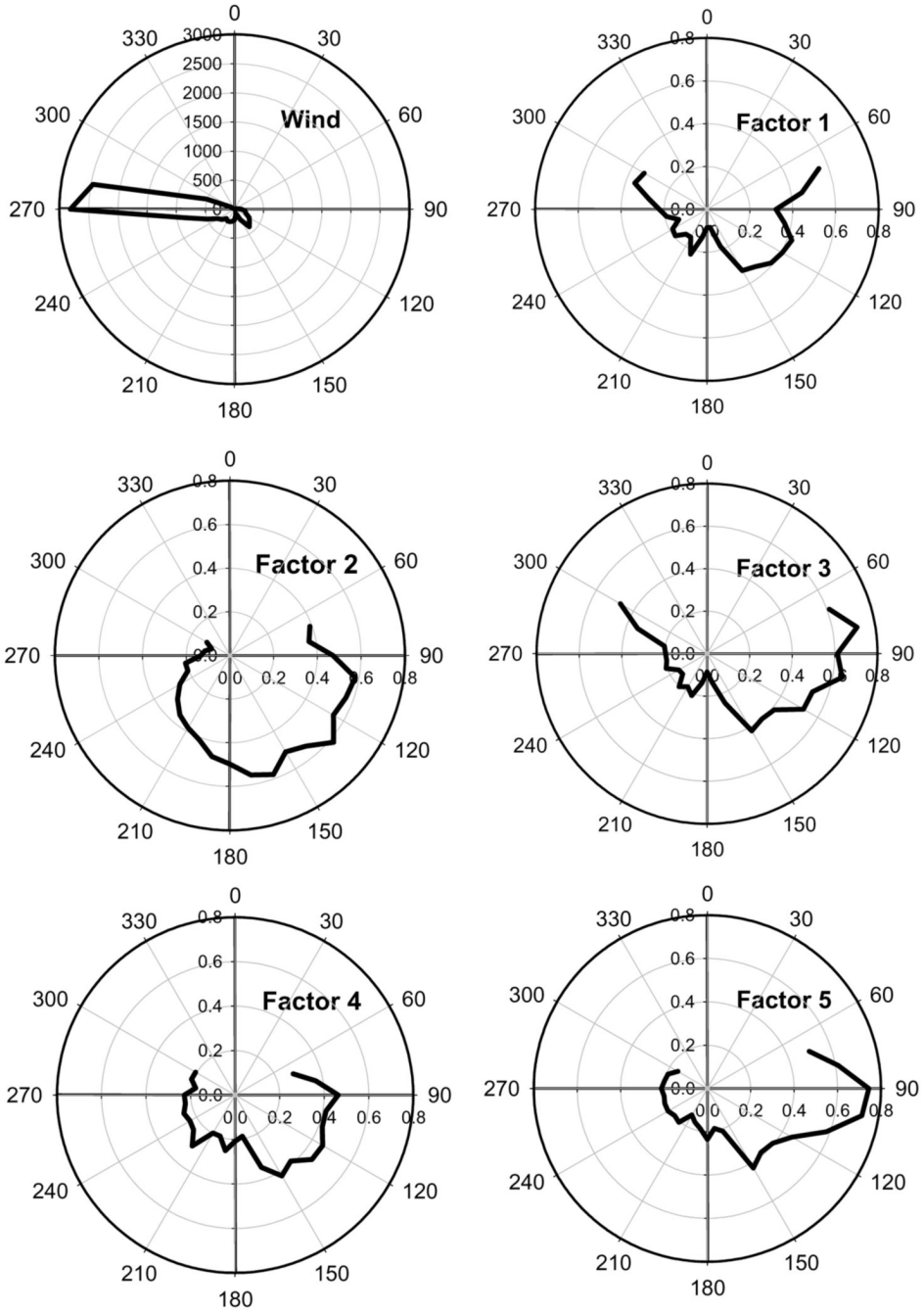

Figure 3.

Wind direction locations of five factors using conditional probability function (CPF) (Wind direction 310 ° −60 ° were considered as missing values due to each wind direction total frequency <10.).

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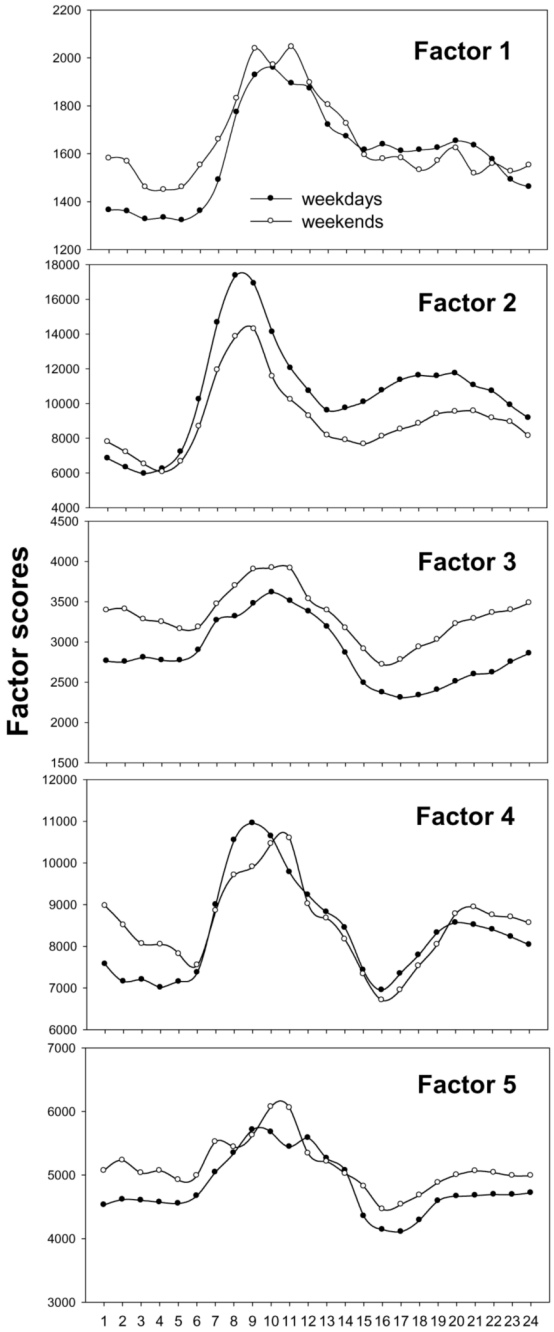

Figure 4.
24-hour diurnal pattern of five factors on weekdays and weekend

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Table 1

Description of the hourly average concentrations of particle size channels[a], gaseous pollutants[b] and particle composition[c] in Erfurt, Germany, 1997-2001.

	N	Mean	SD	Min	Max	Median	lower 25%	upper 25%	LOD[d]
$NC_{(DMPS)}$ (cm^{-3}) (1-9 channels) (0.01-0.10 μm	29313	12641	11516	308	184370	9121	5395	15803	0
$NC_{(LAS\ 1)}$ (cm^{-3}) (1-20 channels) (0.10-0.30 μm)	29313	11342	9760	300	200842	8897	5600	13824	0
$NC_{(LAS\ 2)}$ (cm^{-3}) (21-28 channels) (0.30-0.63 μm)	29313	5685	6920	56	203444	3297	1478	7009	0
$NC_{(LAS\ 3)}$ (cm^{-3}) (29-41 channels) (0.63-2.84 μm)	29313	2222	2983	32	87653	1476	824	2554	0
O_3 (μg m^{-3})	32560	40.9	29.3	2.0	182.0	39.0	16.0	59.0	1.0
NO (μg m^{-3})	32162	17.5	30.9	4.0	635.0	6.0	4.0	16.0	0.6
NO_2 (μg m^{-3})	32249	31.2	16.9	6.0	138.0	29.0	18.0	41.0	1.0
CO (μg m^{-3})	34334	374.2	362.5	100.0	8000.0	300.0	200.0	500.0	58.0
SO_2 (μg m^{-3})	32389	4.8	5.7	3.0	188.0	3.0	3.0	4.0	2.7
Sulfate (μg m^{-3}) [c]	233	3.6	3.2	0.4	22.3	2.5	1.6	4.2	0.35
OC (μg m^{-3})	5323	1.1	0.6	0.2	4.6	1.0	0.7	1.4	1.0
EC (μg m^{-3})	5323	1.5	1.9	0.1	22.1	0.8	0.5	1.6	1.0

[a] particle size channels data are hourly means covering the whole study period measured by DMPS and LAS

[b] gaseous pollutants (O_3, NO, NO_2, CO and SO_2) are hourly means covering the whole study period

[c] particle composition data are available only for parts of the study period. Sulfate is every other day means from Sep 1st, 1997 to Dec 31st, 1998; OC (organic carbon) and EC (elemental carbon) are hourly means from Jan 1st, 2001 to Aug 14th, 2001.

[d] LOD: limit of detection

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Table 2

Spearman correlation coefficients of factors with gaseous pollutants a and particle composition b in Erfurt, Germany for the whole study period (1997-2001) and stratified by season (correlation coefficients > 0.40 (negative and positive) are in bold)

	O_3	NO	NO_2	CO	SO_2	Sulfate	OC	EC
				Whole Study Period				
Factor 1	−0.02	0.11	0.15	0.13	0.10	0.15	0.05	0.07
Factor 2	**−0.47**	**0.51**	**0.61**	**0.44**	0.33	0.18	0.34	**0.44**
Factor 3	−0.11	0.08	0.19	0.18	0.14	0.28	0.38	**0.42**
Factor 4	−0.31	0.34	**0.47**	0.38	**0.41**	**0.45**	**0.47**	**0.55**
Factor 5	−0.40	0.30	0.34	0.34	0.33	0.40	0.26	0.28
				Factor 1				
Spring	0.05	0.09	0.10	0.15	0.24	0.25	−0.18	−0.15
Summer	−0.07	0.14	0.24	0.26	0.16	0.00	0.25	0.13
Autumn	−0.11	0.15	0.21	0.27	0.11	0.14	-	-
Winter	−0.12	0.23	0.20	0.19	0.15	0.24	0.19	0.24
				Factor 2				
Spring	**−0.41**	**0.47**	**0.63**	0.29	0.32	0.27	0.23	0.33
Summer	−0.35	0.25	**0.56**	0.09	0.07	−0.24	0.25	0.40
Autumn	**−0.49**	**0.55**	**0.55**	**0.50**	0.25	−0.07	-	-
Winter	**−0.49**	**0.59**	**0.61**	**0.56**	0.40	0.39	**0.58**	**0.60**
				Factor 3				
Spring	−0.28	0.17	0.34	0.25	0.21	0.39	**0.45**	**0.58**
Summer	−0.04	0.11	0.27	0.33	0.11	0.08	**0.54**	**0.54**
Autumn	−0.31	0.15	0.18	0.28	0.18	0.21	-	-
Winter	−0.19	0.15	0.18	0.24	0.27	**0.41**	**0.50**	**0.57**
				Factor 4				
Spring	−0.31	0.30	**0.52**	0.21	0.38	**0.54**	0.37	**0.51**
Summer	0.09	0.04	0.31	0.10	0.15	0.25	**0.63**	**0.71**
Autumn	**−0.45**	**0.47**	**0.53**	**0.45**	0.37	0.34	-	-
Winter	**−0.61**	**0.51**	**0.56**	**0.61**	**0.55**	0.39	**0.73**	**0.79**
				Factor 5				
Spring	−0.23	0.15	0.20	0.09	0.15	0.19	0.14	0.21
Summer	−0.04	0.10	0.07	0.03	0.05	0.09	−0.05	−0.30
Autumn	−0.40	0.22	0.28	0.37	0.27	0.39	-	-
Winter	**−0.53**	0.36	0.40	**0.45**	**0.49**	**0.68**	**0.55**	**0.66**

[a] gaseous pollutants (O_3, NO, NO_2, CO and SO_2) are hourly means covering the whole study period

[b] particle composition data are available only for parts of the study period. Sulfate is every other day means from Sep 1st, 1997 to Dec 31st, 1998; OC (organic carbon) and EC (elemental carbon) are hourly means from Jan 1st, 2001 to Aug 14th, 2001.

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Table 3

Mean contribution of each factor to total number concentration (NC) and mass concentration (MC) in regression analysis. The proportion of each factor in percent is given in parentheses.

Factors	Spring	Summer	Autumn	Winter	Whole period
			NC (cm^{-3}) (%)		
Factor 1	−17 (0)	98 (1)	−40 (0)	46 (0)	60 (0)
Factor 2	10261 (84)	6747 (79)	9956 (79)	12715 (75)	9831 (78)
Factor 3	−26 (0)	−72 (0)	51 (0)	97 (1)	32 (0)
Factor 4	1618 (13)	458 (5)	2331 (19)	3069 (18)	1844 (15)
Factor 5	−59 (0)	547 (6)	−48 (0)	−247 (−1)	177 (1)
Factor 0 [a]	435 (3)	752 (9)	336 (2)	1197 (7)	697 (6)
total NC	12211 (100)	8530 (100)	12586 (100)	16878 (100)	12641 (100)
			MC ($ng\ m^{-3}$) (%)		
Factor 1	1669 (9)	1416 (10)	2152 (11)	1240 (5)	1573 (8)
Factor 2	−292 (−2)	1020 (7)	508 (3)	1182 (5)	754 (4)
Factor 3	4297 (23)	3110 (22)	4211 (22)	2452 (10)	3354 (17)
Factor 4	6633 (36)	6547 (47)	4782 (25)	10645 (43)	7473 (39)
Factor 5	5469 (30)	1345 (10)	7985 (42)	8261 (33)	5920 (31)
Factor 0 [a]	679 (4)	437 (4)	−540 (−3)	1261 (4)	175 (1)
total MC	18456 (100)	13874 (100)	19098 (100)	25042 (100)	19249 (100)

[a] Factor 0: the residual of the regression analysis.

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Table 4

Summary of the characteristics of five factors

	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5
Source assigned	airborne soil	local traffic	secondary aerosols from local fuel combustion	remote traffic	secondary aerosols from multiple sources
Size range	1.0-2.84 μm	0.01-0.1 μm	0.25-0.60 μm (spring, summer and autumn); 0.5-1.8 μm (winter)	0.1-0.5 μm with a submode 0.01-0.1 μm	0.2-1.0 μm (additional 0.01-0.1μm, 1.0-2.5μm in summer)
Correlation with gaseous pollutants	no	NO, NO_2 and CO	NO_2 ,CO (spring and summer); SO_2 (autumn and winter)	NO and NO_2; CO and SO_2 (except summer)	weak correlations (autumn and winter)
Correlation with particle compositions	no	OC and EC	sulfate (autumn and winter); OC and EC	sulfate (except summer); OC and EC;	sulfate, OC and EC (autumn and winter)
Contribution to total NC [a] and total MC [b]	not to NC 8% to MC	78% to NC 4% to MC	not to NC 17% to MC	15% to NC 39% to MC	no to NC 31% to MC
Location by CPF [c]	southeast and northwest	southeast	southeast and northwest	east and west	east
Diurnal pattern	moderate	strong	weak	strong	weak
Weekdays/ weekends difference	no	obvious	obvious	moderate	no

[a] NC, number concentration

[b] MC, mass concentration

[c] CPF, conditional probability function.