Aerosol and Air Quality Research, 17: 1234–1243, 2017
Copyright © Taiwan Association for Aerosol Research
ISSN: 1680-8584 print / 2071-1409 online
doi: 10.4209/aaqr.2016.06.0242

The Association between Intermodal ($PM_{1-2.5}$) and PM_1, $PM_{2.5}$, Coarse Fraction and Meteorological Parameters in Various Environments in Central Europe

Jana Kozáková[1,2*], Petra Pokorná[2], Alena Černíková[2], Jan Hovorka[2], Martin Braniš[2], Pavel Moravec[1], Jaroslav Schwarz[1]

[1] *Institute of Chemical Process Fundamentals of the ASCR, v.v.i., Rozvojová 2, Prague 165 02, Czech Republic*
[2] *Charles University, Faculty of Science, Prague 128 01, Czech Republic*

ABSTRACT

Fine and coarse fractions of atmospheric aerosol overlap in the particle size range of about 1–2.5 μm (aerodynamic diameter). Sources of both fractions contribute to $PM_{1-2.5}$ to different extents due to meteorological and spatial conditions. Therefore, there is ongoing discussion as to whether $PM_{2.5}$ or PM_1 should be included for monitoring as a fine particulate pollutant by the national ambient air quality standard (NAAQS). The aim of the presented study is to examine the association between the intermodal and PM_1, $PM_{2.5}$, coarse fraction, and meteorological parameters in various environments. Outdoor 24-h mass concentrations of size-resolved PM and meteorological conditions were measured at 12 sites within 42 campaigns between 11/2005 and 3/2015. The data set was divided into 10 environments reflecting season, locality, total measured PM, and placement of the impactor. We used two types of statistic methods: nonparametric correlation analysis and multiple linear regression (MLR). Median $PM_{1-2.5}$ in PM_{10} or TSP percentages were 7% and 6% in summer and 7% and 9% in winter. On the other hand, $PM_{1-2.5}$ accounted for a higher mass portion of $PM_{2.5}$ during summer. Stronger positive correlation and relationship were identified between $PM_{1-2.5}$ and the coarse fraction than between $PM_{1-2.5}$ and PM_1 in all environments. MLR confirmed the dependence of $PM_{1-2.5}$ on PM_1 in only 3 environments. This study found that $PM_{1-2.5}$ in Central Europe represents mostly the "tail" of the coarse mode and probably has the same sources. Therefore, PM_1 should be considered by the NAAQS as a fine particulate pollutant in Central Europe.

Keywords: Intermodal fraction; Personal cascade impactor sampler; PMx; Rural aerosol; Urban aerosol.

INTRODUCTION

Based on the atmospheric aerosol size distribution described by Whitby *et al.* (1972, 1978), there are two fundamental categories of atmospheric aerosol: fine and coarse. These two particle modes are considered separate pollutants not only due to their size but also their different sources, behavior, health effects, chemical composition, etc. (e.g., Anlauf *et al.*, 2006; Herner *et al.*, 2006; Karanasiou *et al.*, 2007; Colbeck, 2008; Pérez *et al.*, 2008; Schwarz *et al.*, 2012). As is well known, the fine mode consists primarily of combustion particles and other particles emitted from processes involving condensation of hot vapors or those formed by gas to particle conversion (Whitby, 1978). The coarse mode is formed by mechanical attrition processes, and hence includes soil and mineral dust, sea spray, and many industrial dusts. Bioaerosol (pollen, spores, plant or animal residues, microorganisms, etc.) can be a significant component in the growing season (Whitby, 1978; Hinds, 1999; Colbeck, 2008).

Even so, the real dividing line between fine and coarse particles cannot be clearly defined. Both fractions overlap in the aerodynamic particle size range of 1–2.5 μm (up to 3) (aerodynamic diameter, d_a) - the intermodal fraction or intermediate range (Whitby *et al.*, 1972; Whitby, 1978; US EPA, 1996; Wilson and Suh, 1997; Hinds 1999; Baron and Willeke, 2001; Colbeck 2008).

During periods of high relative humidity, fine particles, specifically from the accumulation mode, can grow into the intermodal fraction (Geller *et al.*, 2004; Wang *et al.*, 2012; Tian *et al.*, 2014; Tan *et al.*, 2016). Conversely, coarse particles can occur in the particle size range of less than 2.5 μm (d_a) in arid, semi-arid areas, or dry conditions (Husar *et al.*, 1998; Claiborn *et al.*, 2000; Vallius *et al.*, 2000; Pérez *et al.*, 2008). It is evident that the particle size range between 1–2.5 μm (d_a) can include both particles of fine (specifically accumulation) mode origins and coarse particles formed by mechanical processes, such as resuspended dust, sea salt, primary biological particles, etc. In the US and European countries the national ambient air quality

* Corresponding author.
Tel.: 00420-220-390-130
E-mail address: kozakova@icpf.cas.cz

standards (NAAQS) define fine particles as $PM_{2.5}$ and particles less than 10 μm (d_a) as PM_{10}, which includes the coarse fraction ($PM_{2.5-10}$). According to studies mentioned above $PM_{2.5}$ and $PM_{2.5-10}$ are only approximations of real fine and coarse fractions of atmospheric aerosol.

It would appear that the intermodal fraction represents a small portion of the respirable fraction (d_a < 10 μm). But, as shown by some measurements, the intermodal fraction can account for a substantial mass portion (5–45%) of $PM_{2.5}$ (Lundgren *et al.*, 1997; Haller *et al.*, 1999; Geller *et al.*, 2004; Perez *et al.*, 2012). According to several studies performed mainly in dry environments, particles between 1–2.5 μm represent a "tail" of the coarse mode (Lundgren *et al.*, 1997; Haller *et al.*, 1999; Claiborn *et al.*, 2000; Kegler *et al.*, 2001). Opposite results concerning the similarity between the intermodal and PM_1 were described in three studies from Helsinki, Finland, Los Angeles basin, USA and Elche, Spain (Vallius *et al.*, 2000; Geller *et al.*, 2004; Galindo *et al.*, 2011). Perez *et al.* (2012) did not observe a significant correlation between the intermodal and coarse fraction ($PM_{2.5-10}$) or between the intermodal and PM_1 in Barcelona, Spain. Besides the physicochemical properties of the individual aerosol fractions, Jalava *et al.* (2006) investigated the biological effects of the fine ($PM_{1-0.2}$ and $PM_{0.2}$), intermodal, and coarse fractions but without any clear conclusions explaining the similarity of individual fractions.

There are at least two reasonable arguments for further and detailed investigation of the intermodal fraction. First, as mentioned above, there are several studies focused on investigation of the intermodal fraction in arid and semi-arid areas (mainly in USA and Spain), in areas with a higher average relative humidity, particularly within winter seasons (Vallius *et al.*, 2000), but no previous study that has examined this particulate fraction in middle latitude areas, such as Centrale Europe with various atmospheric aerosol sources and mild climatic conditions. Second, there is still ongoing discussion as to whether $PM_{2.5}$ or PM_1 should be included in NAAQS as fine fractions of atmospheric aerosol. Moreover, the extent to which $PM_{2.5}$ is influenced by the intermodal fraction is also an open question. The intrusion of crustal/soil aerosol particles into $PM_{2.5}$, as well as fine particles overgrowing into PM > 1 μm (d_a) during periods of high relative humidity, can cause problems not only in source apportionment but also with epidemiological and exposure studies. Therefore, it is necessary to describe under which conditions it is appropriate to consider PM_1 or $PM_{2.5}$ as fine fractions.

Our study aims to characterize the intermodal fraction and to examine its association with PM_1, $PM_{2.5}$, the coarse fraction, and meteorological parameters in various environments in Central Europe during winter and summer seasons.

METHODS

Sampling Sites and Instrumentation

Measurement of the size-resolved particulate matter was performed at twelve urban, suburban, and rural sites in the Czech Republic, Central Europe (Fig. 1). More details are indicated in Table S1 (Supplement).

24-h mass concentrations of size-resolved particulate matter were measured with a personal cascade impactor sampler (PCIS) that operates at a flow rate of 9 L min^{-1}. Particles were separated in the five aerodynamic particle diameter ranges (d_a) A: > 2.5 μm; B: 2.5–1 μm; C: 1–0.5 μm; D: 0.5–0.25 μm; P: < 0.25 μm (Misra *et al.*, 2002). Particles on the stages A–D were collected on PTFE 25 mm filters (Pall Corporation) used as impaction substrates and particles < 0.25 μm (stage P) on PTFE 37 mm backup filters (Pall Corporation, SKC Limited). At the Ostrava-Plesna, Ostrava-Radvanice, Prague-Suchdol, Prague-Benatska (campaigns in the years 2014 and 2015), and Celakovice sites, a cyclone was used to cut PM_{10} upstream of the inlet linked with the impactor. For these cases, impactor stage A separated particles 10–2.5 μm (d_a).

Meteorological data, including wind speed (WindSonic M,

Fig. 1. Outline map of Czech Republic with the sampling sites.

Gill), temperature, and relative humidity (Comet 200-80/E), were measured at the Brezno, Celakovice, Dobre Stesti, Laz, Ostrava-Radvanice, and Prague-Benatska measurement sites. Meteorology for other sites (Mokropsy, Prague-Mikulandska, Prague-Petriny, Prague-Suchdol, Prague-Benatska (campaign 2015), Ostrava-Plesna, and Svrcovec) was available from the nearest representative Automatic Imission Monitoring Station managed by the Czech Hydrometeorological Institute.

Gravimetric Analysis

The concentrations of atmospheric aerosol were assessed by gravimetric analysis. Before and after sampling the PTFE filters were preconditioned for at least 24 hours at 50 ± 5% relative humidity and 20 ± 2°C in weighing room. To dissipate any electrostatic charge, every filter was passed over a HaugU-electrode ionizer (PRXU27x18x27 200 radia; Haugh, GmbH&Co. KG, Germany) immediately before weighing with a microbalance (Mettler Toledo MX5; Mettler-Toledo, LLC, Ohio, USA).

Each filter was weighed at least 2 (25-mm filters) or 3 times (37-mm filters) and until the weight difference of the filter did not exceed 2 μg (25-mm filters) and 3 μg (37-mm filters) for 2 or 3 neighboring values, respectively. Final mass of the filter was calculated as an average of these weights. Sample weight equaled the weight change between the mass of filter before and after sampling.

For every measurement campaign, least 10% of the field blank filters were used to determine the limit of detection (LOD). Field blanks were exposed to the same conditions as the samples apart from the sampling period. The limit of detection of the weighing procedure was calculated from three times the standard deviation of the weight changes of all field blanks. The lowest level of the 24-h concentration that could be measured was determined as the ratio of LOD to the nominal volume of the air flowing through the impactor (12.96 m^3). The measured concentrations of atmospheric aerosol that were below the concentration lowest level were excluded from the analyzed data set (16% of the concentration data).

Total concentrations were determined as follows: TSP and PM_{10} (when the cyclone cutting PM_{10} was used) were determined from the sum of the aerosol weight from impactor stages (A–P), PM_1 from the sum of the weight of the aerosol from stages C–P (< 1 μm), $PM_{2.5}$ from a sum of the weight of the aerosol from stages B–P (< 2.5 μm), $PM_{1\text{-}2.5}$ were represented by stages B, and $PM_{2.5\text{-}10}$ (the cyclone upstream of the impactor) or $PM_{>2.5}$ (without the cyclone) were represented by stage A. In this study $PM_{2.5\text{-}10}$ and $PM_{>2.5}$ together are called the coarse fraction when the distinction between the two is not important.

Data Analysis

We divided the winter data set into 6 categories according to the environment (urban, suburban, rural), the impactor placement (outdoor, indoor; the outdoor atmospheric aerosol was collected using a vertical inlet (stainless steel, length 1.8 m, inner diameter 8 mm) that was connected to the impactor placed inside of the air-conditioned measurement station (20°C)), and use, or lack thereof, of cyclone cutting PM_{10} upstream of the inlet. The summer data set was divided into 4 categories according to the environment (urban, suburban, and rural) and the cyclone cutting of PM_{10}. We did not consider the impactor placement for summer data set due to small difference between outdoor and indoor temperature (Table 1).

For statistic evaluation the R program was used. Normality of variables: PM_1, $PM_{2.5}$, $PM_{1\text{-}2.5}$, $PM_{2.5\text{-}10}$, and $PM_{>2.5}$ concentrations were rejected using the Shapiro- Wilk test

Table 1. Dataset divided into 10 environmental categories.

Category of the environment	Season	Type of the locality	Sampling with (PM_{10}) or without (TSP) cyclone upstream of the impactor	Impactor placement	Site	Count of observation
urban_TSP_in	winter	urban	TSP	in	Prague Benatska	23
urban_TSP_out	winter	urban	TSP	out	Prague Petriny, Prague Mikulandska	77
urban_PM_{10}_in	winter	urban	PM_{10}	in	Celakovice, Prague Benatska (2015), Ostrava Radvanice	54
suburban_PM_{10}_out	winter	suburban	PM_{10}	out	Ostrava Plesna, Prague Suchdol	35
rural_TSP_in	winter	rural	TSP	in	Dobre Stesti, Laz, Brezno	46
rural_TSP_out	winter	rural	TSP	out	Mokropsy, Svrcovec	55
urban_TSP	summer	urban	TSP	-	Prague Benatska (2009, 2010), Prague Petriny, Prague Mikulandska	75
urban_PM_{10}	summer	urban	PM_{10}	-	Prague Benatska (2014)	11
suburban_PM_{10}	summer	suburban	PM_{10}	-	Prague Suchdol	10
rural_TSP	summer	rural	TSP	-	Brezno, Laz, Dobre Stesti, Mokropsy, Svrcovec	108

of normality. Therefore, we calculated nonparametric Spearman's rank correlation coefficients (r_s). The correlation test was calculated to detect statistically significant correlations between two variables (p-value < 0.05). Multiple linear regression (MLR) was used for exploration of the dependence between one dependent parameter and more than one independent variable in our dataset. The multiple regression model can be formulated as Eq. (1):

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + ... + \beta_k X_k + \varepsilon \quad (1)$$

where Y represents the dependent variable and X_j, j = 0, 1, ..., k represents the independent variable. The parameters β_j, j = 0, 1, ..., k, are called the regression coefficients and ε is a random error (Montgomery and Runger, 2003). For environments where the PM_{10} cyclone was used upstream of the impactor we used this model formula: $PM_{1\text{-}2.5} = \beta_0 + \beta_1 PM_1 + \beta_2 PM_{2.5\text{-}10} + \beta_3 RH + \beta_4 T + \beta_5 WS$ where $PM_{1\text{-}2.5}$, PM_1, and $PM_{2.5\text{-}10}$ represented concentrations of individual PM fractions; T is temperature; RH is relative humidity; and WS is wind speed. For other environments (without PM_{10} cyclone) the model formula was: $PM_{1\text{-}2.5} = \beta_0 + \beta_1 PM_1 + \beta_2 PM_{>2.5} + \beta_3 RH + \beta_4 T + \beta_5 WS$, where $PM_{1\text{-}2.5}$, PM_1, and $PM_{>2.5}$ again represented concentrations of individual PM fractions; T is temperature; RH is relative humidity; and WS is wind speed.

Before performing MLR, logarithmic transformation was applied to the PM concentration data sets to ensure equal variances. To find the optimal regression model, including variables with a significant effect on the dependent variable, the stepwise model selection was used with the backward/forward option and Bayesian information criterion (BIC).

RESULTS AND DISCUSSION

Meteorological Situation

During our measurements, the average winter and summer temperatures were 2°C and 16°C, respectively. The average relative humidity was 77% in winter and 69% in summer. We did not record high differences between wind speed during seasons (winter: 1.7 m s^{-1}, summer: 1.3 m s^{-1}). The average urban wind speed (2.0 m s^{-1}, 1.6 m s^{-1}) was slightly higher than the average rural (1.5 m s^{-1}, 1.3 m s^{-1}) and suburban (1.5 m s^{-1}, 1.0 m s^{-1}) wind speeds during winter and summer, respectively, due to different meteorological conditions at each of the measurement period.

Mass Portion of the Individual Fractions

Of the total measured atmospheric aerosol (TSP) the median mass portion of PM_1 in was 77%, 15% $PM_{>2.5}$, and 8% $PM_{1\text{-}2.5}$. The mass portion of PM_1 in PM_{10} was 85%, while $PM_{2.5\text{-}10}$ accounted for 8% and $PM_{1\text{-}2.5}$ 7% of the overall measurement. The median mass portions of PM_1 in TSP were 70% in summer and 85% during winter. The median mass portions of $PM_{>2.5}$ in TSP accounted for 21% in summer and 11% in winter. $PM_{1\text{-}2.5}$ in TSP constituted only 6% in summer and 9% in winter.

For campaigns where the PM_{10} cyclone was used upstream of the impactor mass portions of the coarse fraction were reduced. $PM_{2.5\text{-}10}$ constituted a median of 6% of PM_{10} in winter and 15% in summer. The mass portion of PM_1 in PM_{10} was 76% in summer and 87% in winter. The median mass of $PM_{1\text{-}2.5}$ was similar during both seasons (7%). The individual mass portions of each environment are shown in the Fig. 2.

In summer, PM_1 composed 15% less of the TSP than in winter, while $PM_{>2.5}$ and $PM_{1\text{-}2.5}$ were 10% and 3% higher, respectively. Similar differences were also seen in cases when PM_{10} cyclone was used, as PM_1 and $PM_{2.5\text{-}10}$ showed seasonal differences of 11% and 9%, respectively. We did not observe seasonal difference in mass portion of $PM_{1\text{-}2.5}$ in PM_{10}. Higher mass portion of the coarse fraction in summer indicated increased contribution of soil dust as evidence by Kegler *et al.* (2001) and Vecchi *et al.* (2004). On the other hand, the high relative humidity reduced the ability of dust resuspension in winter (Vallius *et al.*, 2000). Different mean portions of the intermodal fraction were found in Barcelona during almost 2 years of measurements (Perez *et al.*, 2012). Barcelona is located in dry Mediterranean region and it is strongly influenced by Saharan blown dust.

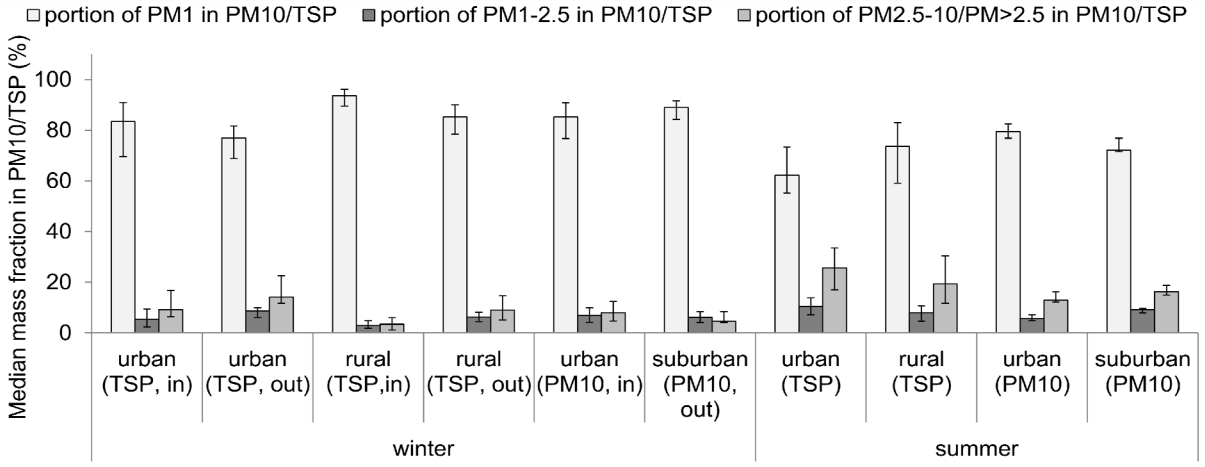

Fig. 2. Median mass portion of PM_1, $PM_{1\text{-}2.5}$, and $PM_{2.5\text{-}10}$ or $PM_{>2.5}$ in the total measured atmospheric aerosol, error bars represent 25th and 75th percentiles.

This source of coarse aerosol influences the proportion of individual fractions. The mass portion of $PM_{1-2.5}$ in PM_{10} reached 16% (excluded days with blown Saharan dust) and 22% (days with blown Saharan dust). The mass portions of $PM_{2.5-10}$ were also higher, with 34% and 31%. Conversely, the mass of PM_1 constituted lower portion of PM_{10} in Barcelona (50%, 47%) than during our measurements. Lundgren *et al.* (1997), who conducted a research study in Phoenix, USA, for 6 months (May–October), found a similar and nearly constant portion of $PM_{1-2.5}$ (8%) every month of their study. The highest portion of PM_{10} was $PM_{2.5-10}$ (69%) and PM_1 made up only 18%. The reverse portion of the coarse and fine fraction observed in Phoenix was probably caused by different meteorological conditions (semi-arid region) and the presence of dominant coarse aerosol source such as blown desert dust in Phoenix.

During our measurement, mass portion of $PM_{1-2.5}$ achieved, at most, 30% in summer and 31% in winter. On the other hand, PM_1 achieved a maximum of 98% during both seasons and $PM_{2.5-10}$ ($PM_{>2.5}$) constituted at most 23% (66%) of the total PM_{10} (TSP) during summer and 29% (49%) during winter. At a residential site in Spokane, USA, $PM_{1-2.5}$ constituted at most 36%, $PM_{2.5-10}$ 81%, and PM_1 95% of the total PM_{10} during the 1.5 year measurement (Haller *et al.*, 1999).

In general, the limitation of the result comparison is caused by different types of cascade impactors used in various studies. The main parameter connected with their design is the sharpness of the collection efficiency curve which influences particle distribution among individual impactor stages. Higher curve sharpness enables more accurate cut-off.

Particle bounce-off effect also alters the particle distribution, reduces collection efficiency, and increases wall losses (Chen and Yeh, 1979). Particle bounce depends on many factors, e.g., the nature of the impactor substrate (the material, using/not using of the coating material), the type of particles, particle loading on the impaction surface, and sampling conditions (Rao and Whitby 1978; Reischl and John 1978; Chen and Yeh, 1979; Hinds, 1999). The PCIS loaded with PTFE collection substrate used in our study was tested by Misra *et al.* (2002) and Singh *et al.* (2003). The laboratory evaluation of the 1.0 and 2.5 μm (d_a) impactor stages using polydisperse ammonium sulphate aerosol indicated that the 50% collection efficiency cut points were very close to the theoretical cut points.

Relationship between $PM_{1-2.5}$ and $PM_{2.5}$

In summer seasons, the highest median mass portion of $PM_{1-2.5}$ in $PM_{2.5}$ reached 11%. Considering the environmental categories, the highest median mass portion of $PM_{1-2.5}$ were recorded at urban and suburban sites during summer (urban_TSP 14% and suburban_PM_{10} 11%, for explanation see Table 1) due to increased contribution of resuspended soil dust (Kegler *et al.*, 2001; Vecchi *et al.*, 2004). The lowest mass portion occurred at the rural site during winter (rural_TSP_in 3%). Median mass portions of $PM_{1-2.5}$ in all environments are shown in the Fig. 3. Perez *et al.* (2012) observed higher mass portion of $PM_{1-2.5}$ in $PM_{2.5}$ at an urban site in Barcelona during days with blown Saharan dust (32%) and even during non-dust days (25%). In the Los Angeles Basin, USA, $PM_{1-2.5}$ accounted for a substantial portion of $PM_{2.5}$ (20–45%) at various types of sites (urban, residential, rural) during all seasons (Geller *et al.*, 2004) and in Phoenix, USA, $PM_{1-2.5}$ accounted for 31% of $PM_{2.5}$, on average, from May to October (Lundgren *et al.*, 1997). These listed sites are characterised by dry summers and mild, moist winters and influenced by desert blown dust.

To find some similarity between these two fractions we processed a correlation analysis. The Spearman correlation coefficients (r_s) suggested a statistically significant association between $PM_{1-2.5}$ and the $PM_{2.5}$ concentrations in almost all environments during winter (r_s = 0.34–0.79) and summer (r_s = 0.47–0.68; Table 2). For comparison, the correlation between $PM_{1-2.5}$ and the coarse fraction ($PM_{2.5-10}$ or $PM_{>2.5}$) was higher in winter, r_s = 0.54–0.81, and for summer, r_s = 0.34–0.81, for the majority of the environments (Table 2).

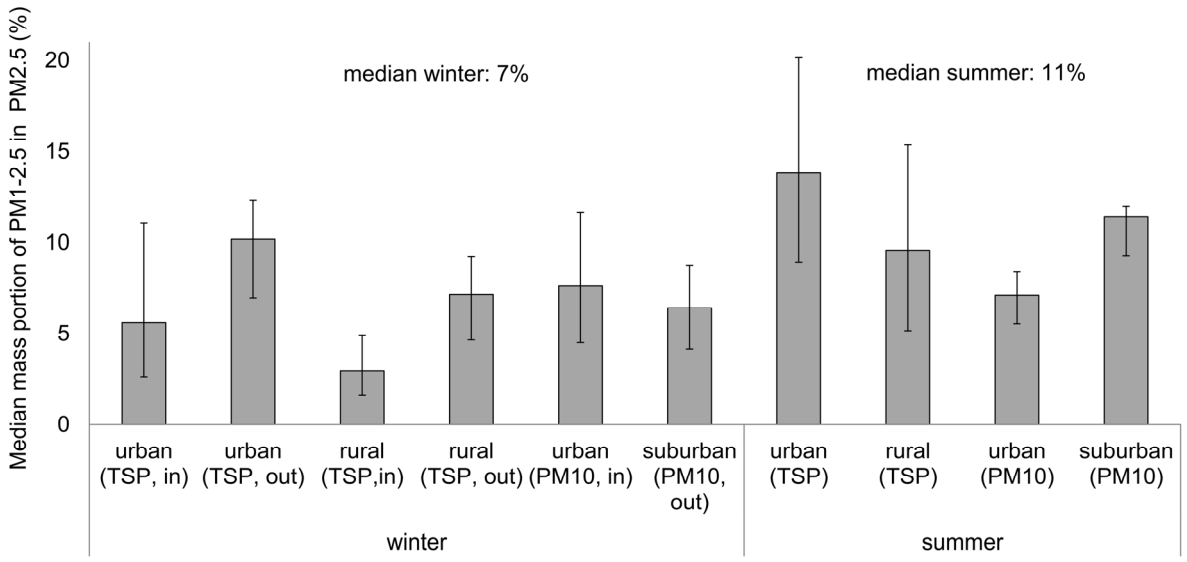

Fig. 3. Median mass portion of $PM_{1-2.5}$ in $PM_{2.5}$, error bars represent 25th and 75th percentiles.

Table 2. Spearman correlation coefficients between $PM_{1-2.5}$ and other monitored variables (statistically significant correlations in bold, p-value < 0.05) for every environment.

Category	$PM_{1-2.5}$									
	Winter						Summer			
	1	2	3	4	5	6	7	8	9	10
PM_1	0.36	**0.75**	**0.33**	**0.55**	**0.46**	**0.57**	**0.49**	**0.32**	0.62	–0.05
$PM_{2.5-10}/PM_{>2.5}$	**0.71**	**0.81**	**0.54**	**0.70**	**0.79**	**0.62**	**0.61**	**0.70**	**0.81**	0.34
$PM_{2.5}$	**0.46**	**0.79**	**0.34**	**0.61**	**0.53**	**0.68**	**0.64**	**0.47**	**0.68**	–0.05
Wind speed	–0.14	**–0.53**	**–0.43**	**–0.56**	–0.12	–0.28	0.18	–0.14	–0.25	0.49
Relative humidity	–0.05	–0.07	0.15	**0.32**	0.19	**0.43**	–0.22	**–0.38**	–0.27	–0.38
Temperature	**0.57**	–0.13	–0.19	0.16	**0.49**	0.25	0.01	**0.46**	–0.46	0.19
Count of observations	23	77	46	55	54	35	75	108	11	10

1-urban (TSP, in), 2-urban (TSP, out), 3-rural (TSP, in), 4-rural (TSP, out), 5-urban (PM_{10}, in), 6-suburban (PM_{10}, out), 7-urban (TSP), 8-rural (TSP), 9-urban (PM_{10}), 10-suburban (PM_{10}).

Vallius *et al.* (2000) also calculated a strong correlation coefficients between $PM_{1-2.5}$ and $PM_{2.5}$ (r_s = 0.77, 0.83) at an urban background site in Helsinki, Finland, for winter and spring, respectively.

Relationship between $PM_{1-2.5}$ and PM_1 Respective Coarse Fractions and Meteorological Parameters

The Spearman correlation coefficients between the mass concentrations of $PM_{1-2.5}$ and other monitored fractions and meteorological parameters are shown in the Table 2. The highest positive, statistically significant correlation coefficients (p-value < 0.05) were calculated between the $PM_{1-2.5}$ and $PM_{2.5-10}$ or $PM_{>2.5}$ (r_s = 0.54–0.81) in all environments except one (category 10). Additionally, somewhat weaker associations between $PM_{1-2.5}$ and PM_1 were found (r_s = 0.32–0.75) in many environments. Less significant correlations for $PM_{1-2.5}$–PM_1 relationship were found in categories when the impactors were placed inside of the air-conditioned measurement stations in winter. Temperature increase, flowrate, and also a type of the collection substrate during sampling can lead to evaporation of volatile/semi-volatile matter (particularly organic compounds, ammonium nitrate, and chloride) of atmospheric aerosol (Hering and Cass, 1999; Liu *et al.*, 2014). Organic matter is the major contributor to PM_1 but minor contributor to $PM_{>1}$ (Vecchi *et al.*, 2004). Water droplets can also evaporate even before deposition on collection substrate. These facts could influence the mass concentrations of particularly PM_1 negatively and then the correlations $PM_{1-2.5}$–PM_1.

Perez *et al.* (2009) presented a stronger correlation between $PM_{1-2.5}$ and $PM_{2.5-10}$ (correlation coefficient, r = 0.45) than between $PM_{1-2.5}$ and PM_1 (r = 0.24). Their research was conducted in an urban background site in Barcelona during almost two years of continuous measurements. Haller *et al.* (1999) found the stronger association between $PM_{1-2.5}$ and $PM_{2.5-10}$ at a residential site in Spokane, USA, during summer (r = 0.62) than during winter (r = 0.21). The associations between $PM_{1-2.5}$ and PM_1 were less significant during both summer (r = 0.25) and in winter (r = 0.27) in comparison with our data. This difference can be due to the fact that this area has hot and arid climate during summer, and very mild winter season.

Reverse observations were carried out at several different sites in the Los Angeles Basin, USA, (Geller *et al.*, 2004) where $PM_{1-2.5}$ correlated more significantly with PM_1 at receptor (situated downwind of the aerosol sources such as traffic and farming and livestock operations) and rural sites (r = 0.81, 0.86) than with the coarse fraction (r = 0.32, 0.37). At an urban source site with vehicles and construction emissions the correlation between $PM_{1-2.5}$ and PM_1 was also higher (r = 0.73) than between $PM_{1-2.5}$ and $PM_{2.5-10}$ (r = 0.33). At an urban traffic site the $PM_{1-2.5}$–PM_1 and $PM_{1-2.5}$–$PM_{2.5-10}$ correlations were similar (r = 0.69, 0.70). Galindo *et al.* (2011) found stronger correlations between $PM_{1-2.5}$ and PM_1 at a traffic site in the city Elche, Spain during summer and winter (r = 0.37 and 0.81, respectively). The correlation between $PM_{1-2.5}$ and $PM_{2.5-10}$ was significant only during winter (r = 0.72), but not in summer (r = 0.14). Vallius *et al.* (2000) observed a stronger association between $PM_{1-2.5}$ and PM_1 (r_s = 0.50 and 0.62) than between $PM_{1-2.5}$ and $PM_{2.5-10}$ (r_s = 0.17 and 0.24) at the urban background site in Helsinki, Finland, during winter and spring, respectively. In this case a growth of the fine mode into the intermodal fraction due to high relative humidity during cold period was probable reason for this correlation (Geller *et al.*, 2004; Guigliano *et al.*, 2005; Herner *et al.*, 2006; Wang *et al.*, 2012; Tian *et al.*, 2014; Tan *et al.*, 2016). According to these various observations, differences in correlation coefficients among individual sites and seasons are probably linked to seasonal changes in PM sources and the weather conditions at each site.

Negative association were observed between $PM_{1-2.5}$ and wind speed in several environmental categories during winter (see Table 2). The similar negative correlation between $PM_{1-2.5}$ and wind speed (r = –0.80) was also observed by Galindo *et al.* (2011) at a traffic site in the city Elche, Spain, in winter. They found the opposite condition in summer when the correlation was positive (r = 0.44). A different study conducted at a residential site in Spokane, USA, did not show a significant correlation between $PM_{1-2.5}$ and wind speed in either season (Haller *et al.*, 1999). According to Chaloulakou *et al.* (2003), negative associations between PM fractions (PM_{10}, $PM_{2.5}$, and $PM_{2.5-10}$) and wind speed can indicate the presence of dominant local source(s) of this fraction. Strong winds generally dilute pollution in the atmosphere, and low winds allow pollution level to rise.

Their study, conducted close to a busy street in Athens, Greece, showed the negative correlations between wind speed and PM_{10}, $PM_{2.5}$, and $PM_{2.5-10}$ in winter (r = –0.40, –0.55, –0.20) and summer (r = –0.41, –0.53, –0.20) during the 1-year measurement.

A significant positive associations between $PM_{1-2.5}$ and temperature were observed in some urban environments during winter (r_s = 0.57, 0.49) and rural in summer (r_s = 0.46) possibly due to increased resuspension and contribution of soil dust (Vallius *et al.*, 2000; Kegler *et al.*, 2001; Vecchi *et al.*, 2004). In addition, our data showed that increase of temperature led to decrease of relative humidity (r_s = –0.41, –0.44, and –0.72 for these winter and summer environments, respectively) and thus higher ability of dust resuspension. Haller *et al.* (1999) found a positive association between these two parameters in summer (r = 0.44), but not in winter (r = 0.01), at a residential site in Spokane, USA. Additionally, the study of Galindo *et al.* (2011) did not show any correlation (r = –0.02) at the traffic site in the city Elche, Spain.

A positive association between $PM_{1-2.5}$ and relative humidity was observed in two environments in winter (r_s = 0.32, 0.43); negative associations were observed in one environment in summer (r_s = –0.38). Galindo *et al.* (2011) observed a positive association (r = 0.37) for all measured data at the traffic site in the city Elche, Spain. High relative humidity can cause the growth of atmospheric particles due to their hygroscopicity, which shifts their size distribution towards larger particles (Geller *et al.*, 2004; Guigliano *et al.*, 2005; Herner *et al.*, 2006; Wang *et al.*, 2012; Tian *et al.*, 2014; Tan *et al.*, 2016). This fact can lead to the positive correlation between PM and relative humidity. On the other hand, the rainfall has negative effect to PM_1 and $PM_{2.5}$ level (Vecchi *et al.*, 2004) and thus lead to negative correlation between PM and relative humidity. To find the similarity between $PM_{1-2.5}$ and PM_1 or coarse fraction behavior we compared the correlation coefficients between individual size fractions and meteorological parameters in every environment (Table 2, Supplement: Tables S2 and S3). No statistically significant correlation between WS and either $PM_{1-2.5}$ or the coarse fraction was found during the summer season. Conversely, though, PM_1 negatively correlated with WS in two environments (r_s = –0.43, –0.82). This at least suggests more similarity between the behavior of the intermodal and coarse fractions during summer. During winter, all fractions negatively correlated with WS, which could have been caused by the same local PM source(s) and/or sources occurring in the same time period. Considering the different environments, both the intermodal and coarse fractions positively correlated with temperature in the winter_urban_PM_{10}_in environment and negatively with relative humidity in the summer_rural_TSP environment. The intermodal fraction and PM_1 positively correlated with the relative humidity in the winter_rural_TSP_out environment. All three fractions ($PM_{1-2.5}$, PM_1, coarse fraction) correlated with temperature in the summer_rural_TSP environment.

Likewise, in several environments involved in our study, similar behavior of $PM_{1-2.5}$ and PM_1, $PM_{1-2.5}$ and $PM_{2.5-10}$, or all three fractions together was observed at two studies (Haller *et al.*, 1999, Galindo *et al.*, 2011). Negative correlations between WS–$PM_{1-2.5}$, WS–PM_1, and WS–$PM_{2.5-10}$ (r = –0.80, –0.74, –0.69) were found at urban traffic site in Elche, Spain, in winter (Galindo *et al.*, 2011). Additionally, they also observed the similar high positive correlation in the RH–$PM_{1-2.5}$ (r = 0.37) and RH–PM_1 (r = 0.31) relationships. Haller *et al.* (1999) observed positive correlations in the T–$PM_{1-2.5}$, T–PM_1, and T–$PM_{2.5-10}$ (r = 0.44, 0.19, 0.57) relationships at a residential site in Spokane during a 1.5-year-long measurement.

Multiple Linear Regression (MLR) Analysis

The MLR analysis was selected to determine if $PM_{1-2.5}$ depended on other monitored parameters: PM_1, $PM_{2.5-10}$ or $PM_{>2.5}$, T, RH, and/or WS. Parameter $PM_{2.5}$ was not considered in the formula because $PM_{1-2.5}$ is already part of the $PM_{2.5}$ and thus it is not a separate independent variable. Detailed results from MLR analysis are summarized in Table 3.

In all environments, $PM_{1-2.5}$ depended on the coarse fraction ($PM_{2.5-10}$ or $PM_{>2.5}$) during winter and summer. This statement agrees with correlation analysis results that were presented in previous paragraph about correlation analysis. In contrast, the dependence of the intermodal fraction on PM_1 was observed in only two environments in

Table 3. Statistical significant independent variables upon which $PM_{1-2.5}$ was dependent in every environment.

	Category of the environment		Variables ($p < 0.05$)	Coefficient of multiple determination (R^2)
Winter	1	urban (TSP, in)	$PM_{>2.5}$, relative humidity, temperature	0.68
	2	urban (TSP, out)	PM_1, $PM_{>2.5}$[a]	0.64
	3	rural (TSP, in)	$PM_{>2.5}$, temperature[b]	0.72
	4	rural (TSP, out)	PM_1, $PM_{>2.5}$	0.56
	5	urban (PM_{10}, in)	$PM_{2.5-10}$	0.44
	6	suburban (PM_{10}, out)	$PM_{2.5-10}$	0.26
Summer	7	urban (TSP)	PM_1, $PM_{>2.5}$, temperature	0.50
	8	rural (TSP)	$PM_{>2.5}$	0.41
	9	urban (PM_{10})	$PM_{2.5-10}$, temperature	0.49
	10	suburban (PM_{10})	$PM_{2.5-10}$, wind speed	0.73

[a] Assumptions of normality of residuals and constant variability not met.
[b] Normality of model residuals was rejected.

winter (urban_TSP_out, rural_TSP_out) and one in summer (urban_TSP). In two winter environments, besides local sources such as traffic and/or domestic heating, humidity could play important role because the impactors were placed outside where humidity was high. Within high relative humidity as mentioned previously, fine particles can easily overgrow into PM > 1 (d_a) due to their hygroscopicity, which shifts their size distribution towards larger particles (Geller *et al.*, 2004; Guigliano *et al.*, 2005; Herner *et al.*, 2006; Wang *et al.*, 2012; Tian *et al.*, 2014; Tan *et al.*, 2016) than when the impactor is placed indoors. These results are in agreement with correlation analysis, the correlation coefficients found between $PM_{1\text{-}2.5}$ and PM_1 were higher in these categories than in others.

No dependence of $PM_{1\text{-}2.5}$ on PM_1 was observed in suburban winter environmental category (PM_{10}, out) when the impactor was also placed outside. This environment predominantly represented the Ostrava Plesna measurement site (67% of the dataset for environment winter_suburban_PM_{10}_out). Ostrava Plesna is located southwest and west of the industrial and coal combustion region (Junninen *et al.*, 2009) and it is strongly influenced by pollution transport from the northeast wind direction (Vossler *et al.*, 2015). Hence, the variable wind direction that was not included in the model could play important role in this case. This is also supported by the coefficient of multiple determination (R^2, see Table 3), which shows that only 26% of variance in the dependent (mass concentrations of $PM_{1\text{-}2.5}$) was explained by the independent variable (mass concentrations of $PM_{2.5\text{-}10}$).

For environments when the impactor inside the measurement station underwent higher temperature conditions than outside, drying occurred and, therefore, shrinkage of fine particles (Zhang *et al.*, 1993; Smolík *et al.*, 2008; Talbot *et al.*, 2016) before they were segregated inside the impactor, which caused the shift of their size distribution to sizes below 1 μm (Štefancová *et al.*, 2010). Thus, the relationship between the intermodal fraction and PM_1 was lost. In addition, as previously mentioned, the evaporation of volatile/semi-volatile matter particularly contributed to the fine fraction could cause a reduction of significance of $PM_{1\text{-}2.5}$-PM_1 relationship (Hering and Cass, 1999; Liu *et al.*, 2014).

The summer environmental category urban_TSP, where $PM_{1\text{-}2.5}$ also depended on PM_1, included measurement sites heavily influenced by exhaust emission from traffic and, thus, dominant PM_1 source (for instance: Guigliano *et al.*, 2005; Pérez *et al.*, 2010; Ondráček *et al.*, 2011; Cusack *et al.*, 2013). On the other hand, the second summer urban (urban_PM_{10}) environment represented only one measurement site directly situated closed to university botanical garden (35,000 m^2). Besides traffic (exhaust emission, brake, tire and road surface abrasion, and road dust resuspension reinforced by road paved with setts), bioaerosol (mainly pollen and plant debris) can be significant source of coarse particles mainly in summer during growing season (Hinds, 1999; Colbeck, 2008). Meteorological parameters, mainly temperature, were also significant in several environments.

In general, often used correlation coefficients indicate a predictive relationship between two variables but they do not necessarily imply the causality. Whereas MLR is a more sophisticated method and can determine interdependence among more than two variables. When we compared results obtained from correlation and MLR analyses for individual environments we found differences in association mainly between PM_1 and meteorological parameters. This phenomenon is caused mainly by the different mathematical calculations behind the two methods. First, multiple linear regression investigates the simultaneous effect of several variables, whereas the correlation coefficient only concerns two variables. When two independent variables have similar effect on the dependent variable, only one will be significant in the regression model. Second, Spearman correlation belongs to nonparametric methods, whereas linear regression is parametric. Quite often (depending on exact distribution of data), parametric tests have greater power (better ability to detect dependence, when it really occurs), which can create a significant variable in linear regression, even though its correlation is nonsignificant (Anděl, 1985; Montgomery and Runger, 2003).

It is necessary to mention the small count of observations for two summer environmental categories: urban_PM_{10} and suburban_PM_{10}. Since low number of observations decrease the power of the test, dependence was not proved as statistically significant, even though high correlation coefficients were observed.

CONCLUSIONS

$PM_{1\text{-}2.5}$ accounted for a substantial part of $PM_{2.5}$, and even PM_{10} or TSP on some days. The median mass portion of $PM_{1\text{-}2.5}$ in PM_{10} or TSP was nearly identical in summer and winter, in contrast with the fine and coarse fraction. Conversely, the median mass portion of the $PM_{1\text{-}2.5}$ in $PM_{2.5}$ was increased in summer due to intrusion of coarse (crustal/soil) aerosol.

The association between the intermodal and coarse fractions was strong in all environments. Nevertheless, a certain association between the intermodal fraction and PM_1 was observed in most of environmental categories, in particular, in winter. For these winter environments, besides local sources, humidity could have played a role, as the impactors were placed outside where humidity was high. This could have preserved their original ambient wet size distribution. When the impactors were placed indoors, the shift towards smaller particles caused decrease of the PM_1–$PM_{1\text{-}2.5}$ relationship.

Overall, the study found that the intermodal fraction represents the "tail" of the coarse mode in most cases, and probably has the similar sources in central Europe. Furthermore, the intermodal fraction may account for an important part of $PM_{2.5}$, with a higher percentage during summer. The intrusion of coarse (crustal/soil, industrial dust) aerosol particles into $PM_{2.5}$ can cause problems not only in the interpretation of source apportionment but also in epidemiological and exposure studies. Therefore, PM_1 should be considered when fine particle health effects are studied, if possible with $PM_{2.5}$ in parallel. $PM_{2.5}$ may be

strongly biased with the coarse mode tail particularly in summer, while during winter PM_1 is not sufficient to capture all fine particles and $PM_{2.5}$ sampling is necessary in Central Europe region.

ACKNOWLEDGMENTS

The study was supported by the Charles University in Prague, project GA UK No 274213 and the Czech Grant Agency, project No P503/12/G147.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at http://www.aaqr.org.

REFERENCES

Anděl, J. (1985). *Matematická Statistika*. Státní Nakladatelství Technické Literatury, Praha.

Anlauf, K., Li, S.M., Leaitch, R., Brook, J., Hayden, K., Toom-Sauntry, D. and Wiebe, A. (2006). Ionic composition and size characteristics of particles in the Lower Fraser Valley: Pacific 2001 field study. *Atmos. Environ.* 40: 2662–2675.

Baron, P.A. and Willeke, K. (2001). *Aerosol Measurement, Principles, Techniques and Applications*, John Wiley & Sons, Inc., New York.

Chaloulakou, A., Kassomenos, P., Spyrellis, N., Demokritou, P. and Koutrakis, P. (2003). Measurements of PM_{10} and $PM_{2.5}$ particle concentrations in Athens, Greece. *Atmos. Environ.* 37: 649–660.

Cheng, Y.S. and Yeh, H.C. (1979). Particle bounce in cascade impactors. *Environ. Sci. Technol.* 13: 1392–1396.

Claiborn, C.S., Finn, D., Larson, T.V. and Koenig, J.Q. (2000). Windblown dust contributes to high $PM_{2.5}$ concentrations. *J. Air Waste Manage. Assoc.* 50: 1440–1445.

Colbeck, I. (2008). *Environmental Chemistry of Aerosol*, Blackwell Publishing, Ltd., Oxford.

Cusack, M., Pérez, N., Pey, J., Alastuey, A. and Querol, X. (2013). Source apportionment of fine PM and sub-micron particle number concentrations at a regional background site in the western Mediterranean: A 2.5 year study. *Atmos. Chem. Phys.* 13: 5173–5187.

Galindo, N., Varea, M., Gil-Moltó, J., Yubero, E. and Nicolás, J. (2011). The influence of meteorology on particulate matter concentrations at an urban Mediterranean location. *Water Air Soil Pollut.* 215: 365–372.

Geller, M.D., Fine, P.M. and Sioutas, C. (2004). The relationship between real-time and time-integrated coarse (2.5–10 μm), intermodal (1–2.5 μm), and fine (<2.5 μm) particulate matter in the Los Angeles Basin. *J. Air Waste Manage. Assoc.* 54: 1029–1039.

Giugliano, M., Lonati, G., Butelli, P., Romele, L., Tardivo, R. and Grosso, M. (2005). Fine particulate ($PM_{2.5}$-PM_1) at urban sites with different traffic exposure. *Atmos. Environ.* 39: 2421–2431.

Haller, L., Claiborn, C., Larson, T., Koenig, J., Norris, G. and Edgar, R. (1999). Airborne particulate matter size distributions in an arid urban area. *J. Air Waste Manage. Assoc.* 49: 161–168.

Hering, S. and Cass, G. (1999). Arising from volatilization of particulate nitrate from Teflon filters. *J. Air Waste Manage. Assoc.* 49: 725–733.

Herner, J.D., Ying, Q., Aw, J., Gao, O., Chang, D.P.Y. and Kleeman, M.J. (2006). Dominant mechanisms that shape the airborne particle size and composition distribution in central California. *Aerosol Sci. Technol.* 40: 827–844.

Hinds, W.C. (1999). *Aerosol Technology: Properties, Behavior, and Measurement of Airborne Particles*, John Wiley & Sons, Inc., New York.

Husar, R.B., Tratt, D.M., Schichtel, B.A., Falke, S.R., Li, F., Jaffe, D., Gassó, S., Gill, T., Laulainen, N.S., Lu, F., Reheis, M.C., Chun, Y., Westphal, D., Holben, B.N., Gueymard, C., McKendry, I., Kuring, N., Feldman, G.C., McClain, C., Frouin, R.J., Merrill, J., DuBois, D., Vignola, F., Murayama, T., Nickovic, S., Wilson, W.E., Sassen, K., Sugimoto, N. and Malm, W.C. (2001). Asian dust events of April 1998. *J. Geophys. Res.* 106: 18317–18330.

Jalava, P.I., Salonen, R.O., Hälinen, A.I., Penttinen, P., Pennanen, A.S., Sillanpää, M., Sandell, E., Hillamo, R. and Hirvonen, M.R. (2006). In vitro inflammatory and cytotoxic effects of size-segregated particulate samples collected during long-range transport of wildfire smoke to Helsinki. *Toxicol. Appl. Pharmacol.* 215: 341–353.

Junninen, H., Mønster, J., Rey, M., Cancelinha, J., Douglas, K., Duane, M., Forcino, V., Müller, A., Lagler, F., Marelli, L., Borowiak, A., Niedzialek, J., Paradiz, B., Mira-Salama, D., Jiminez, J., Hansen, U., Astorga, C., Stanczyk, K., Viana, M., Querol, X., Duvall, R., Norris, G., Tsakovski, S., Wåhlin, P., Horak, J. and Larsen, B. (2009). Quantifying the impact of residential heating on the urban air quality in a typical European coal combustion region. *Environ. Sci. Technol.* 43: 7964–7970.

Karanasiou, A.A., Sitaras, I.E., Siskos, P.A. and Eleftheriadis, K. (2007). Size distribution and sources of trace metals and n-alkanes in the Athens urban aerosol during summer. *Atmos. Environ.* 41: 2368–2381.

Kegler, S.R., Wilson, W.E. and Marcus, A.H. (2001). PM_1, intermodal ($PM_{2.5-1}$) mass, and the soil component of $PM_{2.5}$ in Phoenix, AZ, 1995-1996. *Aerosol Sci. Technol.* 35: 914–920.

Liu, C.N., Lin, S.F., Awasthi, A., Tsai, C.J., Wu, Y.C., and Chen, C.F. (2014). Sampling and conditioning artifacts of $PM_{2.5}$ in filter-based samplers. *Atmos. Environ.* 85: 48–53.

Lundgren, D.A., Hlaing, D.N., Rich, T.A. and Marple, V.A. (1996). PM_{10}/$PM_{2.5}$/PM_1 data from a trichotomous sampler. *Aerosol Sci. Technol.* 25: 353–357.

Matthias-Maser, S. (1998). Primary Biological Aerosol Particles: Their Significance, Sources Sampling Methods and Size Distribution in the Atmosphere. In *Atmospheric Particles*, Harrison, R.M. and van Grieken, R.E. (Eds.), John Wiley & Sons, Ltd., Chichester, Pp. 349–368.

Misra, C., Singh, M., Shen, S., Sioutas, C. and Hall, P.M. (2002). Development and evaluation of a personal cascade

impactor sampler (PCIS). *J. Aerosol Sci.* 33: 1027–1047.

Montgomery, D.C. and Runger, G.C. (2003). *Applied Statistics and Probability for Engineers*, Third Edition, John Wiley & Sons, Inc., New York.

Ondráček, J., Schwarz, J., Ždímal, V., Andělová, L., Vodička, P., Bízek, V., Tsai, C.J., Chen, S.C. and Smolík, J. (2011). Contribution of the road traffic to air pollution in the Prague city (busy speedway and suburban crossroads). *Atmos. Environ.* 45: 5090–5100.

Perez, L., Medina-Ramón, M., Künzli, N., Alastuey, A., Pey, J., Pérez, N., Garcia, R., Tobias, A., Querol, X. and Sunyer, J. (2009). Size fractionate particulate matter, vehicle traffic, and case-specific daily mortality in Barcelona, Spain. *Environ. Sci. Technol.* 43: 4707–4714.

Perez, L., Tobías, A., Querol, X., Pey, J., Alastuey, A., Díaz, J. and Sunyer, J. (2012). Saharan dust, particulate matter and cause-specific mortality: A case-crossover study in Barcelona (Spain). *Environ. Int.* 48: 150–155.

Pérez, N., Pey, J., Querol, X., Alastuey, A., López, J.M. and Viana, M. (2008). Partitioning of major and trace components in PM_{10}–$PM_{2.5}$–PM_1 at an urban site in Southern Europe. *Atmos. Environ.* 42: 1677–1691.

Pérez, N., Pey, J., Cusack, M., Reche, C., Querol, X., Alastuey, A. and Viana, M. (2010). Variability of particle number, black carbon, and PM_{10}, $PM_{2.5}$, and PM_1 levels and speciation: Influence of road traffic emissions on urban air quality. *Aerosol Sci. Technol.* 44: 487–499.

Rao, A.K. and Whitby, K.T. (1978). Non-ideal collection characteristics of inertial impactors-I. Single-stage impactors and solid particles. J. *Aerosol Sci.* 9: 77–86.

Reischl, G.P. and John, W. (1978). The collection efficiency of impaction surface. *Staub Reinhalt Luft*. 38: 55.

Schwarz, J., Štefancová, L., Maenhaut, W., Smolík, J. and Ždímal, V. (2012). Mass and chemically speciated size distribution of Prague aerosol using an aerosol dryer--the influence of air mass origin. *Sci. Total Environ.* 437: 348–62.

Singh, M., Misra, C. and Sioutas, C. (2003). Field evaluation of a personal cascade impactor sampler (PCIS). *Atmos. Environ.* 37: 4781–4793.

Smolík, J., Dohányosová, P., Schwarz J., Ždímal, V. and Lazaridis M. (2008). Characterization of indoor and outdoor aerosols in a suburban area of Prague. *Water Air Soil Pollut.* 1: 35–47.

Štefancová, L., Schwarz, J., Maenhaut, W., Chi, X. and Smolík, J. (2010). Hygroscopic growth of atmospheric aerosol sampled in Prague 2008 using humidity controlled inlets. *Atmos. Res.* 98: 237–248.

Talbot, N., Kubelová, L., Makeš, O., Cusack, M., Ondráček, J., Vodička, P., Schwarz J. and Ždímal, V. (2016). Outdoor and indoor aerosol size, number, mass and compositional dynamics at an urban background site during warm season. *Atmos. Environ.* 131: 171–184.

Tan, J., Duan, J., Zhen, N., He, K. and Hao, J. (2016). Chemical characteristics and source of size-fractionated atmospheric particle in haze episode in Beijing. *Atmos. Res.* 167: 24–33.

Tian, S., Pan, Y., Liu, Z., Wen, T. and Wang, Y. (2014). Size-resolved aerosol chemical analysis of extreme haze pollution events during early 2013 in urban Beijing, China. *J. Hazard. Mater.* 279: 452–460.

US EPA (1996). Review of the National Ambient Air Quality Standards for Particulate Matter: Policy Assessment of Scientific and Technical Information, Appendix A: Consideration in Selecting Particle Size Cut Point for Fine Particles. EPA-452\R-96-013.

US EPA (2004). Air Quality Criteria for Particulate Matter, Volume II. EPA/600/P-99/002bF.

Vallius, M. J., Ruuskanen, J., Mirme, A. and Pekkanen, J. (2000). Concentration and estimated soot content of PM_1, $PM_{2.5}$ and PM_{10} in a subarctic urban atmosphere. *Environ. Sci. Technol.* 34: 1919–1925.

Vecchi, R., Marcazzan G., Valli, G., Ceriani, M. and Antoniazzi, C. (2004). The role of atmospheric dispersion in the seasonal variation of PM_1 and $PM_{2.5}$ concentration and composition in the urban area of Milan (Italy). *Atmos. Environ.* 38: 4437–4446.

Vossler, T., Černikovský, L., Novák, J., Plachá, H., Krejčí, B., Nikolová, I., Chalupníčková, E. and Williams, R. (2015). An investigation of local and regional sources of fine particulate matter in Ostrava, the Czech Republic. *Atmos. Pollut. Res.* 6:454–463.

Wang, X., Wang, W., Yang, L., Gao, X., Nie, W., Yu, Y., Xu, P., Zhou, Y. and Wang, Z. (2012). The secondary formation of inorganic aerosols in the droplet mode through heterogeneous aqueous reactions under haze conditions. *Atmos. Environ.* 63: 68–76.

Whitby, K.T., Husar, R.B. and Liu, B.Y.H. (1972). The aerosol size distribution of Los Angeles smog. *J. Colloid Interface Sci.* 39: 177–204.

Whitby, K.T. (1978). The physical characteristics of sulfur aerosols. *Atmos. Environ.* 12: 135–159.

Wilson, W.E. and Suh, H.H. (1997). Fine particles and coarse particles: Concentration relationships relevant to epidemiologic studies. *J. Air Waste Manage. Assoc.* 47: 1238–1249.

Zhang, X.Q., McMurry, P.H., Hering, S.V. and Casuccio, G.S. (1993). Mixing characteristics and water content of submicron aerosols measured in Los Angeles and at the grand canyon. *Atmos. Environ.* 27: 1593–1607.

Received for review, June 13, 2016
Revised, December 21, 2016
Accepted, January 9, 2017