Spatial and temporal variations of PM$_{2.5}$ concentration and composition throughout an urban area with high freeway density—the Greater Cincinnati study

Dainius Martuzevicius[a,1], Sergey A. Grinshpun[a,*], Tiina Reponen[a], Rafał L. Górny[a,2], Rakesh Shukla[a], James Lockey[a], Shaohua Hu[b], Rafael McDonald[b], Pratim Biswas[b], Linas Kliucininkas[c], Grace LeMasters[a]

[a] *Department of Environmental Health, Center for Health-Related Aerosol Studies, University of Cincinnati, P.O. Box 670056, Cincinnati, OH 45267-0056, USA*
[b] *Environmental Engineering Science, Washington University in St. Louis, St. Louis, MO, USA*
[c] *Department for Environmental Engineering, Kaunas University of Technology, Kaunas, Lithuania*

Abstract

The PM$_{2.5}$ concentration and its elemental composition were measured in the Cincinnati metropolitan area, which is characterized by intense highway traffic. The spatial and temporal variations were investigated for various chemical elements that contributed to the PM$_{2.5}$ fraction during a 1-year-long measurement campaign (December 2001–November 2002). The ambient aerosol monitoring was performed in 11 locations around the city during nine measurement cycles. During each cycle, four Harvard-type impactors were operating in parallel in specific locations to explore various factors affecting the PM$_{2.5}$ elemental concentrations. The sampling was performed during business days, thus assuring traffic uniformity. The 24-h PM$_{2.5}$ samples were collected on Teflon and quartz filters. Teflon filters were analyzed by X-ray fluorescence (XRF) analysis while quartz filters were analyzed by thermal-optical transmittance (TOT) analysis. In addition to PM$_{2.5}$ measurements, particle size-selective sampling was performed in two cycles using micro-orifice uniform deposit impactor; the collected fractionated deposits were analyzed by XRF. It was found that PM$_{2.5}$ concentration ranged from 6.70 to 48.3 µg m^{-3} and had low spatial variation (median coefficient of variation, CV = 11.3%). The elemental concentrations demonstrated high spatial variation, with the median CV ranged from 38.2% for Fe to 68.7% for Ni. For traffic-related trace metals, the highest concentration was detected in the city center site, which was close to a major highway. The particle size selective measurement revealed that mass concentration of the trace metals, such as Zn, Pb, Ni, as well as that of sulfur reach their peak values in the particle size range of 0.32–1.0 µm. Meteorological parameters and traffic intensity were not found to have a significant influence on the PM$_{2.5}$ elemental concentrations.

*Corresponding author. Tel.: +1-513-558-0504; fax: +1-513-558-2263.

E-mail address: sergey.grinshpun@uc.edu (S.A. Grinshpun).

[1] On leave from Department for Environmental Engineering, Kaunas University of Technology, Kaunas, Lithuania.
[2] Currently at: Department of Biohazards, Institute of Occupational Medicine and Environmental Health, Sosnowiec, Poland.

1. Introduction

The fine fraction of the ambient aerosol may cause adverse health effects, especially respiratory problems in children and the elderly living in large metropolitan areas. Respiratory problems, including allergies and asthma, have been associated with the finer fractions of the $PM_{2.5}$ aerosol (particulate matter smaller than 2.5 μm in aerodynamic diameter). Epidemiological studies confirm the relationship between concentration of fine aerosol particles and human mortality (Dockery et al., 1993; Schwartz et al., 1996), asthma (Weiland et al., 1994; Duhme et al., 1998; Ostro et al., 1998), bronchitis (Braun-Fahrlander et al., 1997), allergy (Saxon and Diaz-Sanchez, 2000) and reduced lung function (Brunekreef et al., 1997).

Urban traffic-related aerosols that include various toxic metal compounds have been particularly linked to adverse health effects (ATSDR, 1999, 2000). Since freeways have much more intense traffic of heavy-duty diesel vehicles than local access roads, a freeway transport seems to play a critical role as a PM air pollution source. In many urban areas in the US, mobile sources are dominant in generating the $PM_{2.5}$ aerosol fraction (Gertler et al., 2000), which includes diesel engine exhaust particles (Morawska et al., 1998; Shi et al., 1999; Lepperhoff, 2001), spark ignition engine exhaust particles (Ristovski et al., 1998), and compressed natural gas engines (Ristovski et al., 2000) in the size range of 0.01–0.1 μm. The State of Ohio where this study was conducted ranks sixth in the US, and Cincinnati ranks 24th from 401 urbanized areas in miles of federal interstate highways (FHWA, 2001). The concentric network of interstate highways surrounds 1.9 million residents in the Cincinnati area. Recent studies show that the number of heavy-duty freight vehicles may increase by as much as 60% by 2020 (ODOT, 2002). Thus, a comprehensive investigation of highway-related traffic contribution to urban $PM_{2.5}$ aerosol is needed.

The above considerations have motivated an extensive study on factors affecting the $PM_{2.5}$ concentration levels and elemental composition of ambient aerosols in the Greater Cincinnati area. This region is characterized by a combination of considerable highway automobile and truck traffic and a significant percentage of the local population having respiratory disorders (OEC, 2000).

Human exposure to traffic-related pollutants can be adequately assessed if aerosol monitoring is performed at a sufficient number of sites and with a sufficient frequency of measurements and sampling periods. The characterization of the spatial variability of ambient aerosol ($PM_{2.5}$ and its compounds) is crucial for designing an efficient monitoring network and for an adequate assessment of population exposure variation as a function of the location or with respect to a specific source (Burton et al., 1996). If the source is predominant and has a relatively constant aerosol emission rate for a specific area, spatial and temporal variations should be quantified. Spatial factors usually reflect the background aerosol characteristics, while temporal variation represents the influence of meteorological conditions. The temporal variation of $PM_{2.5}$ and some of its chemical compounds or elements has recently been investigated for several urban areas (Weber et al., 2003; Ye et al., 2003). Since the ambient monitoring data are usually based on daily measurements, the temporal variation is often represented by day-to-day concentration changes (Wight, 2000; Watson and Chow, 2001).

Spatial factors, such as distance from a highway, topography, land surface roughness, and the presence of other pollution sources affect the pollutant concentration and composition. Time-related factors, such as local meteorology (wind speed and direction, stability of the atmosphere boundary layer, precipitation, etc.), as well as traffic intensity are known to essentially affect the pollutant dispersion and—as a result—human exposure. Hence, this study was designed to investigate some of these factors causing the spatial and temporal variations of $PM_{2.5}$ aerosol and its elemental concentrations in an urban environment with a dense freeway network.

2. Experimental methods

2.1. Sampling and analysis of $PM_{2.5}$

Harvard-type $PM_{2.5}$ impactors (MS&T Area Sampler, Air Diagnostics and Engineering, Inc., Harrison, ME, USA) were used for the ambient aerosol sampling. With these impactors the sampled particles are deposited on a filter uniformly allowing for accurate nondestructive analysis. Uniformity was defined as follows: the deposit density in a specific area of the filter is within $\pm 10\%$ of the average deposit on the filter. Four Harvard Impactors (three with an operational flow rate of $20\,l\,min^{-1}$ and one with $10\,l\,min^{-1}$) were used. All impactors had the same cut size of 2.5 μm. With the standard sampling period of 24 h (National Ambient Air Quality Standard for $PM_{2.5}$), the sampling flowrate of each impactor was measured before and after the sampling period and calibrated with a flow meter (DryCal DC-Lite, BIOS International Corporation, Butler, NJ, USA). Impactors were supported by aluminum tripod and had a rain cover. When collecting ambient particles for elemental analysis, the samplers operated with 37-mm Teflon membrane filters (pore size = 1 μm) with a support ring (Pall Corporation, Ann Arbor, MI, USA). When collecting for carbon analysis, 37-mm quartz filters (Whatman Inc., Clifton, NJ, USA) were utilized. Polyester "Drain disk" pads (Whatman

Inc.) were used with the filters to maintain their integrity.

Standard operating procedures for the filter media preparation, gravimetrical operations and sampling were followed. Filter preparation included conditioning in a humidity chamber at a relative humidity of 30–40% for at least 24 h, weighing and labeling. Each 24-h sampling session began in the morning, with the four impactors being turned on between 9 AM to 12 PM allowing for team travel from one monitoring site to another. The sampling was conducted during workdays (Mondays through Fridays), having a total of four samples per week per site. During the first week of each cycle, four measurement sites were operating in parallel, each equipped with a Harvard Impactor loaded with a Teflon filter. Once the four-site measurement cycle was completed, two Harvard impactors were relocated from their sites to two other sites. Then the 24-h paired samples were collected simultaneously for 2 days at these two sites (with two impactors in each, one of which operated with Teflon and one with quartz filters). Overall, for eight samples taken during a typical measurement cycle one was collected on quartz filters and seven on Teflon filters. Also, one transportation blank and one field blank were collected during each measurement cycle and analyzed accordingly.

After the 24-h sampling, the filters were weighed following the same pre-sampling conditioning procedures. Once the $PM_{2.5}$ mass concentration was determined, then Teflon filters were sent for X-ray fluorescence (XRF) analysis (Chester Labnet, Tigard, OR, USA) for a total of 39 detected elements (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sn, Sb, Ba, La, Hg, Pb, Bi). Quartz filters were analyzed for elemental and organic carbon with the thermal-optical transmittance (TOT) technique using NIOSH-5040 method (Sunset Laboratory Inc., Hillsborough, NC, USA). The XRF method was successfully used in our pilot study (Reponen et al., 2003), suggesting advantages of the XRF over the proton-induced X-ray emission (PIXE) method for trace metals as also demonstrated by Toro and Cortés (1996) and Benyaïch et al. (1997). The limit of detection (LOD) for the filters analyzed by XRF (deposition area $= 6.61 \, cm^2$, flowrate $= 20 \, l \, min^{-1}$) varied by elements and ranged from $34 \, ng \, m^{-3}$ for sodium to $0.19 \, ng \, m^{-3}$ for cobalt, nickel and copper. The LOD of the TOT analysis for elemental carbon was $600 \, ng \, m^{-3}$.

2.2. Particle size selective sampling and analysis

In order to determine the particle size distribution of the ambient aerosol and the elemental composition of different particle size fractions, the eight-stage micro-orifice uniform deposit impactor sampler (MOUDI,

Model 100, MSP Corp., Minneapolis, MN, USA) operating at a flowrate of $30 \, l \, min^{-1}$ was used for the size-selective aerosol sampling. The particle size distribution measurements were performed during Cycles VIII and IX (Table 1) at a site that showed relatively high aerosol concentration (the city center). The particles were collected on impaction stages with the following cut-off sizes: $d_{50} = 0.18, 0.32, 0.56, 1.0, 1.8, 3.2, 5.6,$ and $10 \, \mu m$. The 47-mm Teflon filters, prepared similarly to those used with the Harvard impactors, were utilized as a collection media and subsequently analyzed using gravimetric and XRF methods. A Teflon filter substrate was used for subsequent XRF analysis; effects of bounce were not considered. During each of the two measurement cycles, the MOUDI was continuously operated for 1 week allowing for sufficient deposit at each impaction stage. In addition to the MOUDI sampler, the ambient aerosol particle size distribution was measured for 2 days with an optical particle counter (Portable Dust Monitor, Model 1.108, Grimm Technologies, Inc., Douglasville, GA, USA), which detected the particles in 15 channels in the size range of 0.3–20 μm.

2.3. Area of monitoring

The Greater Cincinnati metropolitan area is located in the American Midwest, at the southwestern tip of Ohio. The climate is primarily continental, with an average annual temperature of $11.8°C$, but subject to frequent changes due to the passage of cyclonic storms in winter and spring, and thunderstorms during the summer. Prevalent wind direction is from south–southwest (33% of all winds for year 2001, (NCDC, 2003)). The prevalent wind direction was considered when choosing ambient aerosol measurement sites throughout the city area. The city center is located on the bank of the Ohio River and extends over two ranges of hills. The hill elevation is about 120 m above the river valley level. Although the valley is not very steep, it influences meteorological conditions and pollutant dispersion in the area, including downtown.

Greater Cincinnati is intersected by the network of highways—I-75, I-71, I-74, I-471, loop I-275 and local highways (Ronald Reagan and SR-562) (Fig. 1). Highway I-75 is a major north–south cargo transportation artery. A 17-mile stretch of I-75 in Cincinnati carries 124,000 vehicles including 14,000 heavy-duty diesel vehicles (daily average during year 2002). Interstates I-75 and I-71 form "a highway corridor" directed to the north-northeast, matching the predominant wind direction.

With very intense traffic, Greater Cincinnati also hosts several major industrial companies and many smaller ones, totaling 233 facilities that potentially emit air pollutants within the municipal area limits and hundreds more in suburban areas. The counties

Table 1
Purpose of measurement cycles by site

Measurement cycle #	Period	Sites	Aim
I	3–7 December 2001	Police, Highland, Water tower	$PM_{2.5}$ concentration (total, elemental) as a function of distance from highway
II	26 February–7 March 2002	Police, Highland, Water tower, Canyon	$PM_{2.5}$ concentration (total, elemental, carbon) as a function of distance from highway; compare inside/outside highway corridor concentrations
III	18 March–6 April 2002	Water tower, Canyon, Grooms, Findlay	$PM_{2.5}$ concentrations along the highway corridor as a function of distance from highway traffic intensity and meteorology.
IV	10–26 April 2002	Grooms, Findlay, Harrison, Mernic	$PM_{2.5}$ concentrations inside/outside the highway corridor as a function of distance from highway, traffic intensity and meteorology.
V	28 May–6 June 2002	Water tower, Findlay, Harrison, Oakbrook	$PM_{2.5}$ concentrations along the highway corridor and in "background" site
VI	10–18 June 2002	Findlay, Oakbrook	$PM_{2.5}$ concentrations in city center vs. rural area
VII	20–27 June 2002	Grooms	Sampler intercomparison (QC)
VIII	27 August–18 September 2002	Findlay, Harrison, Linn, Hays	$PM_{2.5}$ concentrations in the city center area, particle size distribution measurements
IX	7–21 November 2002	Findlay	Particle size distribution measurements

surrounding the Greater Cincinnati area host 873 industrial, agricultural and other facilities that produce air pollutants from 894 stacks and 1346 other point and area sources (US EPA EnviroFacts, 2003).

2.4. Location of sites

Overall, the ambient aerosol monitoring campaign included nine measurement cycles. In most of the cycles, four Harvard impactors were operating simultaneously in different sites. The 11 measurement sites distributed throughout the entire monitoring area are displayed in Fig. 1. The following considerations were applied for site selection: to cover different city areas; to establish different proximities to pollution sources including linear sources (highway and state routes) and point sources; and to test different locations relative to the interstate highway corridor (I-71 and I-75) and with respect to the predominant wind direction. The sites were located at various distances from major highways, ranging from 210 m (Findlay) to 4590 m (Mernic). Each combination of sites was selected to address the influence of a specific factor on $PM_{2.5}$ and its elemental concentrations, as indicated below (see Section 2.7).

2.5. Meteorological factors

The meteorological data during the measurement cycles were collected from the US National Weather Service operated weather stations at the Greater
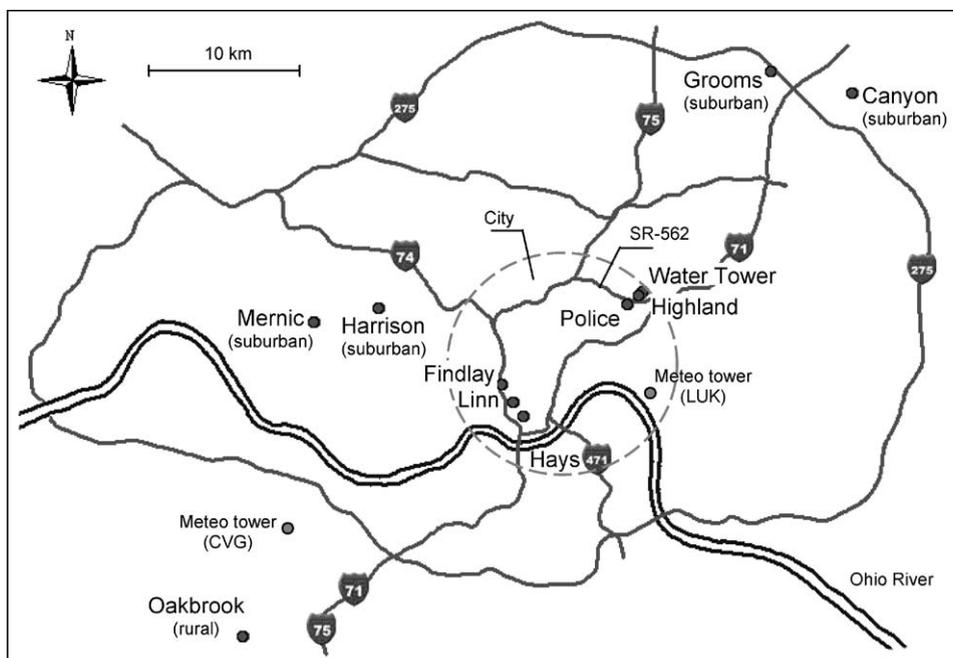
Fig. 1. The Greater Cincinnati highway system and location of PM$_{2.5}$ sampling sites.

Cincinnati/Northern Kentucky International Airport CVG (situated in the southern part of the Cincinnati metropolitan area) and at the Lunken Municipal Airport LUK (situated east of downtown Cincinnati in the Ohio River valley). The hourly recorded wind speed, wind direction, ambient temperature and humidity data were averaged over the samplers' 24-h operation time.

2.6. Traffic counts

The highway vehicle traffic counts were measured by the Ohio Department of Transportation (ODOT) counters, positioned on highways I-75 and I-275. The 24-h averages were calculated according to the starting and ending time points for each sampling period. Since the traffic was routinely counted only on one direction of the highway, the above 24-h average values were doubled in order to calculate approximately total numbers of the passing vehicles.

2.7. Measurement cycles

The annual measurement campaign was performed from December 2001 through November 2002. The measurement cycles are described in Table 1. During Cycle I, three Harvard impactors were positioned in the vicinity of the state highway SR-562 (Police, ~270 m from the highway, Highland, ~370 m, and Water Tower, ~480 m sites), serving as a connector for two major interstate highways (I-71 and I-75). In Cycle II,

another Harvard impactor was added in a presumably clean area further (~1.8 km) from a highway (Canyon site). The aim of Cycles I and II was to identify how the distance from SR-562 affects the PM$_{2.5}$ and its elemental concentrations. In Cycle III, the impactors were positioned along "the highway corridor", including the city center close to I-75 (Findlay site), the industrial area (Water Tower site), the northern suburban area close to I-275 (Grooms site), and the Canyon site, far from I-275. This cycle aimed at characterizing the PM$_{2.5}$ along the highway corridor (which approximately coincides with the vector of the predominant wind direction). Cycles IV and V aimed to compare the PM$_{2.5}$ concentration inside and outside the "highway corridor". During Cycle VI, more extensive sampling using Teflon and quartz filters in parallel was performed in the city center (Findlay) and in a rural area (Oakbrook). Cycle VII served for quality control purposes. All four Harvard impactors operated in a close distance from each other and the PM$_{2.5}$ concentration was measured for 5 consecutive days. The coefficient of variation (CV) of the PM$_{2.5}$ concentration was 2.5%, demonstrating low measurement variability. During Cycle VIII, three monitoring stations were positioned in the city center (Findlay, Linn, and Hays) along highway I-75 and at one station on the western side (Harrison) of the city. The size-selective particle sampling was performed in parallel to PM$_{2.5}$ measurements during Cycles VIII and IX, where two Harvard impactors and MOUDI were operating in parallel in the city center (Findlay site).

2.8. Statistical analysis

The statistical analysis of the collected data was performed using Microsoft Excel 2000 (Microsoft Corp., Redmond, WA, USA) and SPSS 11.0 (SPSS Inc., Chicago, IL, USA) software. To quantitatively characterize the temporal and spatial variation of $PM_{2.5}$ concentration, CV was calculated. The temporal CV was determined separately for each site using the daily measured values within a cycle. The spatial CV was calculated for each cycle and represented the variation among four simultaneously operated monitoring stations based on daily measurements. The single factor analysis of variance (ANOVA) was used to determine the significance of the variations of studied parameters. The relationship between meteorological factors, traffic intensity and the measured concentrations were characterized by Spearman correlation coefficient (R), Pearson correlation coefficient (r) and the coefficient of determination (r^2).

3. Results and discussion

3.1. $PM_{2.5}$ concentration

Overall, 219 daily samples of $PM_{2.5}$ aerosol were collected. The lowest concentration level of $6.70 \, \mu g \, m^{-3}$ was measured in the Harrison site on 11 September 2002, and the highest one of $48.3 \, \mu g \, m^{-3}$ occurred at the Linn site on 9 September. The average daily $PM_{2.5}$ concentration within a monitoring cycle ranged from 10.8 ± 2.9 to $28.6 \pm 11 \, \mu g \, m^{-3}$ (Table 2).

The lowest daily variation among all cycles and sites occurred during Cycle II (winter) in the Highland site, with CV = 10.9%. The highest variation was obtained during Cycle VIII (summer) in the Harrison site, with CV = 43.6%, when smog conditions were followed by clear and windy days. The $PM_{2.5}$ concentration was a subject of a pronounced seasonal variation with the peak occurring in summer.

Overall, the $PM_{2.5}$ concentration exhibited rather low spatial variation throughout all measurement cycles. The median value of CV among the sites was 11.3%. No significant influence of the distance from a highway on the $PM_{2.5}$ level was observed. The ANOVA test was performed for measurement data collected on the same day in four sites operating simultaneously in each cycle. During all the cycles the spatial variation was not significant ($p > 0.05$).

The highest average $PM_{2.5}$ concentration was observed in the city center. Once it was found that the $PM_{2.5}$ level in the Findlay site was higher than in most of the sites located in other areas, two more samplers were placed in the city center area at a distance of approximately 500 m between each other (Linn and Hays sites). Such design allowed allocating local sources that might have affected specifically the Findlay site data. Based on the $PM_{2.5}$ data collected on Findlay, Linn and Hays sites, the analysis of variance showed that the difference among the daily measured concentration levels was not significant ($p > 0.05$, CV = 8.1%), suggesting that the spatial distribution of the $PM_{2.5}$ concentration was relatively uniform in the city center.

There have been a few field-monitoring campaigns performed with Harvard impactors that can be used for the comparison with our results. The most recent study by Hoek et al. (2002) reports the $PM_{2.5}$ concentration measurements in three areas: Stockholm County (Sweden), Munich (Germany) and the main cities of The Netherlands. The 14-day sampling averages were taken in 40 sites in each area. The annual average $PM_{2.5}$ concentration ranged from 11 to $20 \, \mu g \, m^{-3}$ in Munich, from 8 to $16 \, \mu g \, m^{-3}$ in Stockholm County, and from 14 to $26 \, \mu g \, m^{-3}$ in the Dutch cities. The study by Houthuijs et al. (2001) assessed the mean annual concentration levels of $PM_{2.5}$ and PM_{10} in six Central and Eastern European countries: Bulgaria, the Czech Republic, Hungary, Poland, Romania and the Slovak Republic. The 24-h averaged concentrations were measured every 6th day on urban background sites. The $PM_{2.5}$ concentration ranged from 29 to $68 \, \mu g \, m^{-3}$, with the lowest concentrations found in the Slovak Republic and the highest in Bulgaria and Poland. Fischer et al. (2000) evaluated differences in concentration of air pollutants outside and inside of homes on streets with low and high traffic intensity in Amsterdam, The Netherlands. The measured average $PM_{2.5}$ concentration was $25 \, \mu g \, m^{-3}$ near homes, which were located in high traffic density areas, and $21 \, \mu g \, m^{-3}$ near homes that had a low-density traffic in their vicinity. Cyrys et al. (2002) performed a study on source apportionment of fine and ultrafine ambient particles in Erfurt, Germany, for the period of 1995–1998. For the ambient aerosol samples collected near a major urban road, the mean $PM_{2.5}$ concentration was $26.3 \, \mu g \, m^{-3}$ with a standard deviation of $20.8 \, \mu g \, m^{-3}$, thus, the $PM_{2.5}$ concentration levels found in the Greater Cincinnati area are similar to studies performed in Europe. These did not exceed the EPA 24-h average standard of $65 \, \mu g \, m^{-3}$. The average measured concentrations in some sites, however, were close to or exceeded the annual standard of $15 \, \mu g \, m^{-3}$. Our findings obtained with Harvard impactors were in a good agreement with $PM_{2.5}$ levels measured in local EPA monitoring sites using Thermo Andersen RAAS2.5-300 filter-based samplers (Thermo Electron Corp., Franklin, MA, USA: Federal Reference Method). The EPA reported the average $PM_{2.5}$ concentration range of $15–20 \, \mu g \, m^{-3}$ measured in the Greater Cincinnati area (US EPA AirData, 2002).

Similarly to our findings, many studies performed in urban areas report relatively low spatial variation of the

Table 2
The average (mean±standard deviation) measured values of PM$_{2.5}$ and its elemental concentrations obtained during the measurement campaign

Site	Measurement cycle	PM$_{2.5}$ concentration (µg m^{-3})	Elemental PM$_{2.5}$ concentration (ng m^{-3})								
			Al LOD = 2.2	Si 1.5	S 1.1	Ca 0.54	Mn 0.43	Fe 0.35	Ni 0.19	Zn 0.27	Pb 1.1
Police	I	14.6±4.0	39.2 ±15	115±45	982±400	168±73	4.15±3.0	129±40	0.227±0.19	15.9±7.7	5.77±4.4
	II	12.2±2.7	39.5±8.7	132±29	711±100	120±44	1.9±0.7	85.1±26	0.407±0.55	11.0±4.4	2.2±0.70
Highland	I	16.2±4.2	39.7±14	114±59	956±310	130±74	6.65±7.9	136±60	0.586±0.41	20.1±10	5.22±4.0
	II	10.9±1.1	32.2±5.9	97.8±27	683±150	80.7±36	2.07±1.3	69.6±26	0.486±0.72	10.2±3.6	2.33±1.1
Water tower	I	16.3±3.8	42.4±8	109±29	1080±390	125±49	9.01±10	156±63	0.82±0.46	24.8±15	7.69±4.8
	II	11.8±1.8	42.4±14	129±43	749±140	99.0±46	2.39±1.0	84.4±34	0.477±0.49	11.2±4.6	2.31±0.90
	III	13.6±4.1	41.7±25	96.2±64	1220±400	78.7±50	4.55±4.7	93.7±37	0.969±0.53	15.7±7.3	3.56±1.2
	V	23.9±7.4	42.3±14	130±68	1950±360	81.0±38	11.4±12	141±53	1.04±1.1	28.0±15	4.32±1.9
Canyon	II	10.8±2.9	44.4±19	127±50	711±190	90.4±34	1.92±0.7	72.4±29	0.507±0.70	8.30±2.5	1.96±0.80
	III	11.7±3.7	25.8±25	73.0±64	1170±350	47.3±35	2.07±0.9	56.6±23	0.382±0.22	10.4±3.2	2.8±1.2
Grooms	III	13.2±4.7	23.7±21	73.7±53	1260±400	52.6±26	2.33±1.0	70.4±24	1.13±1.4	14.7±8.2	2.57±1.4
	IV	12.5±4.1	48.8±24	129±73	1340±500	69.9±20	2.3±1.4	85.4±27	4.59±4.6	20.9±20	3.17±2.7
	VII	25.7±9.9	76±37	168±91	2560±680	98.7±58	2.72±1.0	119±45	3.37±2.32	11.2±3.1	2.23±1.1
Findlay	III	16.4±4.5	43.6±23	131±66	1220±360	156±82	3.77±0.8	143±48	0.667±0.32	24.6±19	5.00±2.2
	IV	17.8±4.9	74.2±31	223±110	1370±420	245±93	4.52±1.7	233±92	0.614±0.37	27.6±27.6	5.94±3.3
	V	24.2±6.8	38.9±10	139±55	2340±600	137±49	4.46±1.0	152±57	0.824±0.93	21.3±12.7	4.03±2.4
	VI	25.4±9.1	52.9±27	149±90	2450±490	174±110	4.22±2.4	197±150	0.975±0.57	67.6±92	9.29±11
	VIII	27.4±9.8	71.7±28	217±85	2780±1700	222±110	7.41±3.3	277±160	1.87±0.96	211±250	28.4±28
	IX	14.5±3.6	25.8±12	91.2±36	1020±270	107±62	2.46±1.5	127±75	0.630±0.35	24.8±13	4.28±1.8
Harrison	IV	13.4±3.8	55.6±29	158±100	1430±550	101±44	2.38±1.0	77.3±30	0.371±0.24	10.8±7.4	2.74±1.4
	V	22.4±6.3	33.5±14	129±64	2150±660	106±58	2.76±0.6	90.8±21	0.475±0.32	11.5±3.9	2.3±1.0
	VIII	23.2±10	47±16	125±53	3060±1700	84.4±32	3.85±1.7	112±53	1.21±0.49	50.1±68	6.66±3.8
Mernic	IV	17.5±5.6	35.5±14	96.3±37	1680±440	63.4±20	2.16±0.6	55.9±12	1.11±0.99	8.90±3.7	2.91±1.5
Oakbrook	V	20.8±6.4	27.2±8	111±56	2050±720	49.9±22	2.18±0.5	67.2±14	0.497±0.40	8.50±4.1	2.29±0.70
	VI	17.7±4.5	29.9±8.3	83.3±29	1850±650	62.3±40	1.56±0.6	48.7±15	0.376±0.27	8.40±3.3	1.79±0.40
Linn	VIII	28.6±11	158±74	484±240	2910±1900	411±160	8.90±3.5	342±160	1.71±0.56	157±230	19.7±20
Hays	VIII	25.0±10	62.2±32	183±94	3150±1900	187±110	6.05±2.9	207±130	1.81±0.95	139±250	17.7±22

PM$_{2.5}$. The low spatial variation is associated with high temporal variation (Brook et al., 1997; Orlic et al., 1999; Gehrig and Buchmann, 2003) and reflects the contribution of regional sources and long-range aerosol transport (Burton et al., 1996; Brook et al., 1997). High seasonal variation was emphasized in all studies, although in contrast to our findings in the Greater Cincinnati area, the PM$_{2.5}$ concentration measured in those studies was higher during the cold period of the year. These aerosol concentrations increases observed in other urban areas during winter may be attributed to increased fuel combustion from heating as well as to an increased frequency of thermal inversions. In Cincinnati, however, higher summertime concentration levels were likely induced by increased secondary aerosol formation form SO$_2$ and VOCs due to the relatively high ambient temperature, humidity and solar radiation.

3.2. Elemental concentrations of the PM$_{2.5}$ fraction

Among 39 elements identified by XRF analysis, few elements were selected for presentation purposes, including Al, Si, Ca, and Fe (crustal elements), S (from the secondary sulfate source, coal combustion and diesel sources), and Mn, Fe, Ni, Zn, and Pb (traffic-related trace elements, Cadle et al., 1998; Gilles and Gertler, 2000; HEI, 2002). The average elemental concentrations with standard deviations obtained in every cycle are presented in Table 2. For most elements the concentration was significantly greater than LOD, except for lead and nickel, some measurement data were comparable. Thus some of these variations described below may result from instrumental errors.

The concentration of crustal elements ranged from 23.7 ± 21 ng m^{-3} (Al, Grooms site, Cycle III) to 484 ± 240 ng m^{-3} (Si, Linn site, Cycle VIII). The 24-h measurement data show that the contribution of Al, Si, Ca and Fe to the PM$_{2.5}$ mass varies from 0.391% to 9.35%. The following median values of CV were obtained for the spatial variation among sites: $CV_{Al} = 23.8\%$, $CV_{Si} = 27.0\%$, $CV_{Ca} = 45.1\%$, and $CV_{Fe} = 37.5\%$. The highest daily variation was observed during Cycle III in the Canyon station (CV ranged from 73.2% for Ca to 96.8% for Al). This variation is likely due to local activities involving soil. The median values of daily variation during all the cycles were as follows: $CV_{Al} = 41.4\%$, $CV_{Si} = 49.5\%$, $CV_{Ca} = 46.1\%$, and $CV_{Fe} = 38.2\%$.

Sulfur showed the highest concentration among all elements analyzed by XRF, ranging from 683 ± 150 to 3150 ± 1900 ng m^{-3} (6.2–12.6% of total PM$_{2.5}$ mass). Although the sulfur concentration varied with season, the levels remained elevated throughout the year, which is likely attributed to the sulfur emissions from several coal power plants located along the Ohio River valley, followed by secondary conversion to sulfate. Moreover,

the sulfur concentrations in suburban and rural areas were of the same magnitude as those measured in the city center or near industrial pollution sources during the same cycles. This finding indicates that sulfur was predominant in the background aerosol. The median CV-value representing the spatial variation for sulfur was 6.5%. The sulfur concentration showed considerable seasonal variation but lower daily variation. The median CV-value representing the daily variation of sulfur among all cycles was 30.7%, approximately equal to the one obtained for the PM$_{2.5}$ concentration (28.5%).

The PM$_{2.5}$ levels of the selected trace metals (Mn, Ni, Zn, and Pb) ranged from 0.227 ± 0.19 ng m^{-3} (Ni, Police site, Cycle I) to 211 ± 250 ng m^{-3} (Zn, Findlay site, Cycle VIII). The overall contribution of these elements to the PM$_{2.5}$ mass ranged from 0.22% to 5.65%. Same-day measurements demonstrated that the contribution of the trace metals to the PM$_{2.5}$ concentration in the city center was higher by a factor of 1.2–1.9 compared to suburban area (Harrison) and by a factor of 1.5–2.3 compared to rural area (Oakbrook). Both the suburban and rural sites are distant from point pollutions sources, but the suburban site had more intense local street traffic, resulting in a higher trace metal concentrations (by a factor of 1.1–1.6). For these elements, the spatial variation among sites was calculated as follows: $CV_{Mn} = 37.6\%$, $CV_{Ni} = 61.9\%$, $CV_{Zn} = 33.1\%$, and $CV_{Pb} = 33.8\%$. Higher spatial variations of Mn, Ni, Zn, and Pb as compared to PM$_{2.5}$ and sulfur suggest that these elements originated mostly from local pollution sources. The trace metals also showed substantial daily and seasonal variations. The highest daily variation was observed for Zn and Pb concentrations measured in the Findlay site during Cycle VIII, with CV reaching 117% for Zn and 97.5% for Pb. The median CV value for trace metals ranged from 43.6% for Mn to 68.7% for Ni.

In addition to the above listed elements, several others were present at relatively high concentration levels. Those include alkaline metals, such as Na (0–960 ng m^{-3}, 0–5.6% of total PM$_{2.5}$ mass), K (20–210 ng m^{-3}, 0.13–0.97%), and Mg (0–50 ng m^{-3}, 0–0.35%), as well as halogen Cl (0–1780 ng m^{-3}, 0–10.6%). The other 26 elements (P, Ti, V, Cr, Co, Cu, Ga, Ge, As, Se, Br, Rb, Sr, Y, Zr, Mo, Pd, Ag, Cd, In, Sb, Sn, Ba, La, Hg, Bi) showed concentration levels close to the detection limits, with a minor contribution to the total PM$_{2.5}$ mass: from 0.10% to 1.13%.

Table 3 presents the Spearman correlation coefficient (R) matrices for PM$_{2.5}$ and concentrations for particular elements obtained in three different sampling sites representing the city center (Findlay), suburban area (Harrison) and rural area (Oakbrook). The values of $R \geqslant 0.8$ are shown in bold. As shown in Table 3, the sulfur and PM$_{2.5}$ concentrations correlate well in all

locations. The trace metal concentrations did not correlate well with the $PM_{2.5}$ concentration because of their small contribution to the total mass as compared to sulfur and carbon. Although it was found that the distance from a highway is not a key parameter affecting the $PM_{2.5}$ concentration, the proximity to a highway influenced the elemental composition of the ambient aerosol. Slightly different patterns of correlation between certain elements can be noticed in the city center, suburban sites, and the rural location, respectively (see Table 3). Elements, such as Al, Si and Ca correlate with each other in the three sites. This confirms that their origin comes from crustal materials. In suburban

(Harrison) and rural (Oakbrook) sites these elements are more likely to be a product of soil erosion, while in the city center their major source is likely the road dust aerosolized by traffic. In the vicinity of highways, the traffic-related trace metals exhibited higher correlation between each other compared to that found in other locations.

3.3. Elemental and organic carbon in the $PM_{2.5}$ fraction

The measurements of elemental and organic carbon were conducted to trace a possible contribution of the emission from heavy-duty diesel-powered vehicles to the

Table 3
Correlation matrices for elemental concentrations of the $PM_{2.5}$ aerosol (city center—Findlay, suburban—Harrison, and rural—Oakbrook)

	Al	Si	S	Ca	Mn	Fe	Ni	Zn	Pb	OC	EC
Findlay n = 56 (Al through Pb), n = 23 (EC and OC)											
$PM_{2.5}$	0.44	0.51	**0.85**	0.44	0.62	0.51	0.43	0.18	0.26	0.68	0.62
Al		**0.97**	0.27	**0.86**	0.68	**0.82**	0.25	0.05	0.26	0.79	0.77
Si			0.31	**0.90**	0.70	**0.86**	0.26	0.06	0.25	**0.83**	0.79
S				0.21	0.40	0.26	0.32	0.12	0.12	0.48	0.40
Ca					0.67	**0.92**	0.32	0.15	0.28	**0.86**	**0.85**
Mn						**0.80**	0.52	0.38	0.52	0.75	0.73
Fe							0.45	0.31	0.45	**0.88**	**0.91**
Ni								0.58	0.52	0.43	0.47
Zn									0.73	0.42	0.42
Pb										0.35	0.30
OC											**0.93**
Harrison n = 27 (Al through Pb)											
$PM_{2.5}$	0.07	0.35	**0.89**	0.25	0.46	0.55	0.49	0.16	0.17	a	a
Al		**0.82**	0.08	0.72	0.40	0.54	0.14	0.04	0.14	a	a
Si			0.30	**0.93**	0.37	0.60	0.09	−0.19	−0.06	a	a
S				0.17	0.46	0.45	0.56	0.20	0.35	a	a
Ca					0.34	0.54	0.05	−0.12	0.01	a	a
Mn						**0.84**	0.47	0.44	0.43	a	a
Fe							0.38	0.39	0.29	a	a
Ni								0.47	0.54	a	a
Zn									**0.81**	a	a
Pb										a	a
OC											a
Oakbrook n = 19 (Al through Pb), n = 6 (EC and OC)											
$PM_{2.5}$	0.49	**0.56**	**0.81**	0.43	0.24	0.39	0.39	−0.32	0.16	**0.90**	0.38
Al		0.62	0.46	0.67	0.27	0.28	−0.04	0.10	−0.42	0.77	0.37
Si			0.34	0.74	0.79	0.79	0.30	0.10	−0.16	0.43	0.54
S				0.32	0.07	0.24	0.17	−0.12	0.37	0.77	0.37
Ca					0.59	0.61	0.20	−0.02	−0.39	0.60	**0.89**
Mn						**0.94**	0.22	0.30	0.04	0.26	0.71
Fe							0.34	0.32	0.21	0.49	0.77
Ni								−0.32	−0.66	0.46	0.32
Zn									0.15	0.14	0.09
Pb										−0.17	−0.03
OC											0.60

Values denote Spearman coefficient of correlation (R). Significant correlations ($p < 0.05$) are shaded; $R \geqslant 0.8$ are marked bold.
[a]Sample size is not sufficient.

ambient $PM_{2.5}$ aerosol fraction. Elemental carbon is considered to be a good marker for the diesel-fueled engine exhaust emissions (Fowler, 1985; Gray and Cass, 1998; Birch and Cary, 1996; Funasaka et al., 1998).

The daily measured organic carbon (OC) and elemental carbon (EC) concentrations ranged from 6.25 to $36.0\,\mu g\,m^{-3}$ and from 1.04 to $12.6\,\mu g\,m^{-3}$, respectively. During Cycle VI, the carbon concentration was monitored in the city center (Findlay site) and the rural area (Oakbrook site) for 6 days. The average ratio of elemental to total carbon, $EC/(EC + OC)$, in the city center was 0.25 versus 0.12 in rural site. The difference between the two was found to be significant ($p < 0.001$). Moreover, in the city center site, which is close to a highway, the elemental carbon showed relatively high correlation with other traffic markers (trace metals and organic carbon), while no clear correlation pattern can be distinguished in suburban and rural sites (see Table 3). The EC concentration measured in the city center correlates well with the crustal elements concentration, reflecting the road dust contribution to the $PM_{2.5}$ mass concentration. The average EC contribution to total $PM_{2.5}$ concentration was 6.2% for the city center, 3.0% for the suburban area and 2.2% for the rural area. The values for OC were 22%, 22% and 23%, respectively.

3.4. $PM_{2.5}$ mass closure

The contribution of all the measured elements was evaluated for the same three urban areas: the city center (Findlay), suburban (Harrison) and rural (Oakbrook). The nitrogen compounds (nitrates and ammonia) were not measured during this study and were referred as "non-identified". Conventional correction factors were introduced in order to assess oxygen and hydrocarbon mass in oxides and hydrocarbons. It was assumed that all sulfur exist in sulfate form (Sweet and Gatz, 1998), and Si, Al, Ca, a part of Fe, and Ti are of a crustal nature and exist in oxides (Eldred et al., 1987; Marcazzan et al., 2001). Organic carbon concentration was corrected by a factor of 1.4 in order to assess total organic matter (Weber et al., 2003). The average $PM_{2.5}$ mass compositions for these three sampling sites are summarized in Table 4. The obtained values are typical of those in urban areas. It is clearly visible that elemental carbon and the trace metals have higher contribution to the $PM_{2.5}$ mass in the city center area than in suburban and rural locations, thus reducing relative contribution of sulfur.

3.5. Particle size-selective elemental concentrations

The particle size-selective data obtained with the 8-stage MOUDI during two measurement cycles (VIII and IX) are presented in Fig. 2. As seen from Fig. 2a, the

Table 4

Mass closure of the $PM_{2.5}$ aerosol elemental composition (city center—Findlay, suburban—Harrison, and rural—Oakbrook)

	Findlay	Harrison	Oakbrook
Sulfate	25	34	30
Other measured elements (including trace metals)	4	2	1
Organic matter	30	30	33
Elemental carbon	6	3	2
Crustal material	13	11	6
Non-identified elements (determined by difference)	22	20	28

Values denote percentage of the total $PM_{2.5}$ mass.

ambient aerosol concentration was about 1.5–6 times lower during Cycle IX than during Cycle VIII (when unfavorable meteorological conditions was noted). This finding is confirmed also by the data collected with Harvard impactors (Table 2, Findlay site). Although the $PM_{2.5}$ and the element-specific concentration levels obtained in the two cycles differed significantly, similar trends were observed in the particle size distributions shown in Fig. 2. The concentration values obtained in upper MOUDI stages (5.6 and $10\,\mu m$) may be of limited accuracy because the particle size is high enough to affect the inlet sampling efficiency, and may also result in the particle bounce.

During Cycle VIII, the peak of the particle size distribution was reached at d_{50} as low as $0.32\,\mu m$, possibly reflecting a high concentration of smog particles. Baik et al. (1996) indicated that during smog conditions in Seoul, a peak particulate matter concentration was determined at $0.8\,\mu m$. The study of Baik et al. (1996), however, was performed in a relatively clean area without extensive local air pollution sources, while in this study the aerosol samples were collected near a major highway. During Cycle IX, the peak mass concentration was reached at $d_{50} = 1\,\mu m$, demonstrating that the ambient air had lower concentration of fine particles than during Cycle VIII.

Relatively high particulate mass concentrations detected in the submicron size range imply that the number concentration of smaller particles ($< 0.3\,\mu m$) is about three to five orders of magnitude greater than the number concentration of larger particles ($> 1.0\,\mu m$). This finding was supported by 2 days of measurements with the Grimm Portable Dust Monitor. The average measured concentration of the particles in the range of $0.3–0.4\,\mu m$ was $9.18 \times 10^8\,particles\,m^{-3}$, while concentration of particles $> 1.0\,\mu m$ was $2.5 \times 10^5\,particles\,m^{-3}$.

Similarly to the ambient aerosol mass concentration determined by MOUDI, the elemental concentrations in different particle size fractions followed the same pattern in both measurement cycles (Fig. 2b–f). The elemental
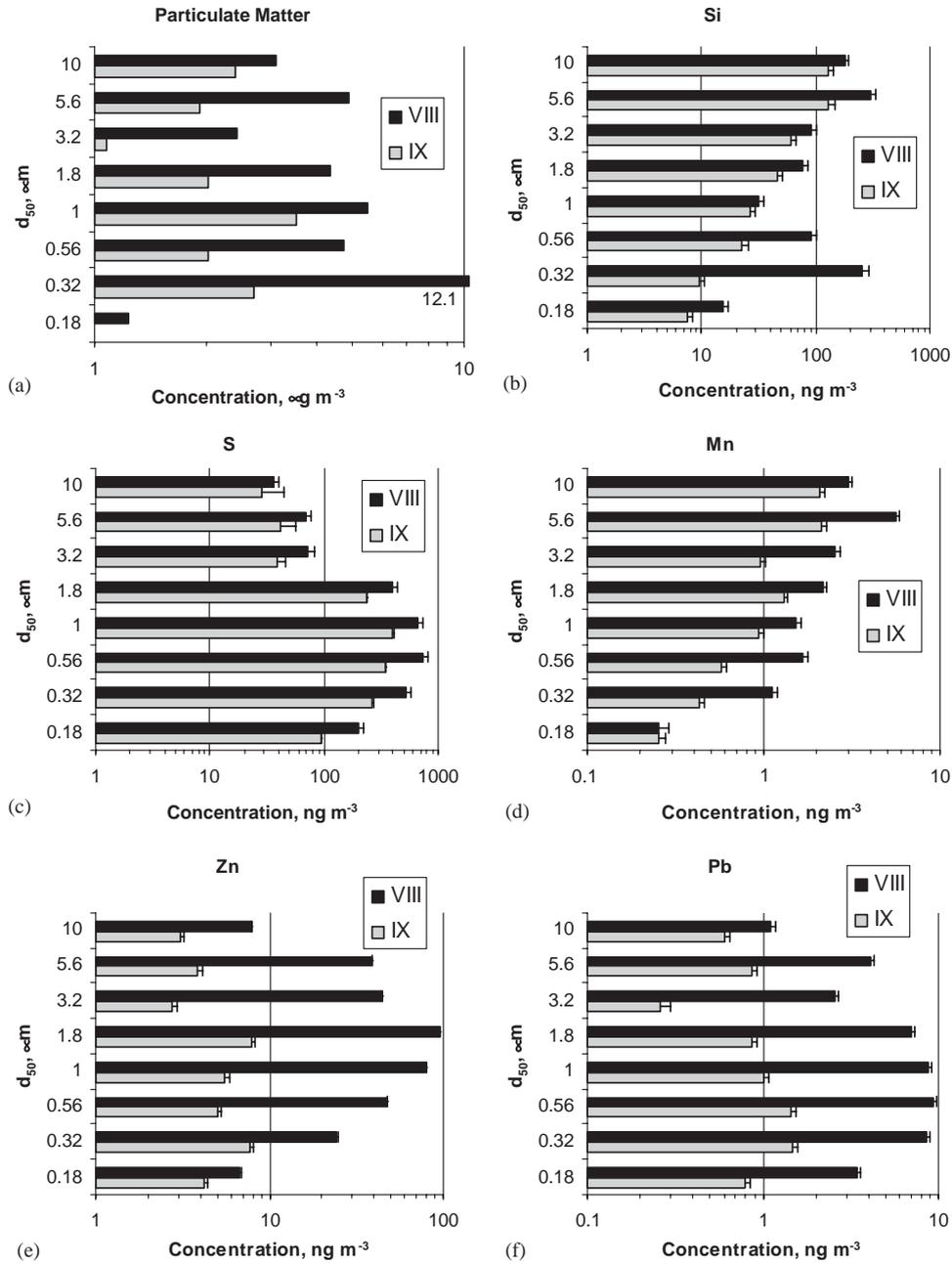
Fig. 2. The particle size-selective data obtained in the Findlay site (city center, close to a major highway) during the cycles VIII and IX.

analysis of the impaction substrates used for the particle size-selective collection showed mainly two types of the particle size distribution. The crustal elements, such as Si, Ca, Al and Fe, had a peak concentration in upper particle size range (5.6 μm). Sulfur and trace metals with the exception of Mn demonstrated their peak values in the lower size ranges (0.32–1.0 μm). Roadside manganese is a combustion product of methylcyclopentadienylmanganese tricarbonyl (MMT), which has replaced

by tetraethyl lead (TEL) as a gasoline fuel additive. Thus, there is a significant accumulation of Mn in road dusts near major freeways (Lytle et al., 1995). Higher concentration of Mn occurs in particles of a larger aerodynamic diameter because Mn is deposited and combined with larger particles of crustal matter (SiO_2, Al_2O_3 and others).

The ratio between elemental concentrations during both monitoring cycles differs for crustal elements and

trace metals. The size-fractional mass concentrations of silicon, aluminum, calcium and iron were approximately equal in both cycles, except for the particle diameters of 0.56 and $0.32\,\mu m$ for Si and Al. The trace metal concentrations were up to 10 times higher during Cycle VIII compared to Cycle IX.

3.6. Influence of meteorological factors

As stated earlier, the distance from a highway appeared to be an important but not the major factor affecting $PM_{2.5}$ concentrations and its elemental composition. Many authors regard meteorological parameters as important factors for the aerosol transport (Brunnemann et al., 1996; Louka et al, 1997; Wrobel et al., 1999; Marcazzan et al., 2002). In this study, the impact of the following parameters on the $PM_{2.5}$ and its elemental concentrations was analyzed: the wind speed, the ambient air temperature and humidity. The effect of the stability of the atmospheric boundary layer was not characterized because no measurements of the vertical temperature gradient were performed. The data available from the meteorological mast located outside the Cincinnati metropolitan area appeared to be of limited relevance because the vertical temperature gradient profile may not be representative.

During the annual monitoring campaign, the average daily temperature ranged from $-7.5\,^{\circ}C$ (Cycle II, winter) to $27.3\,^{\circ}C$ (Cycle VIII, summer). The average daily temperature in the Ohio River valley (LUK tower) was found to be very similar to that measured outside of city (CVG tower): the differences did not exceed $1.5\,^{\circ}C$. The relative humidity had also rather low spatial variability: the maximum difference was 3% during the warm period of the year. While temperature and relative humidity followed the same patterns between the two locations (Person correlation coefficients $r = 1.00$ and 0.92, respectively), the wind speed was noticeably higher outside of the city than in the city center: average values were $3.6\,m\,s^{-1}$ (as measured at the International Airport) and $2.7\,m\,s^{-1}$ (as measured at the Municipal Airport), with $r = 0.80$. The atmospheric pressure readings in the two meteorological sites were highly correlated ($r = 0.97$), although the pressure in the valley was significantly higher (an average of $10.7\,mm\,Hg$).

Table 5 presents the Spearman correlation coefficient matrices between the concentrations of $PM_{2.5}$ and 11 selected elements, compared to wind speed, temperature and relative humidity. The correlation between $PM_{2.5}$ concentration, the concentrations of elements and the wind speed was negative. The lower correlation coefficient for crustal elements illustrates the competition of two mechanisms: atmospheric dispersion and aerosolization from surfaces. With the increasing wind speed, the dispersion mechanism predicts decreasing aerosol concentration. At the same time, higher wind speed also causes greater release of dust particles due to erosion and re-suspension. The temperature changes did not significantly influence the ambient aerosol concentration, although considerable positive correlation was found between the ambient temperature and concentration of sulfur in the particles. The increased sulfur concentration during summer months may be attributed to an enhanced formation of a secondary aerosol due to higher temperature and humidity. The ambient temperature had little effect on the concentration of the crustal elements and higher effect on trace metals. The relative humidity showed a negative correlation with most of the elements, such as Al ($R = -0.34$), Si ($R = -0.37$) and Ca ($R = -0.44$), $p < 0.05$. This finding agrees with the data reported by Mirme and Ruuskanen (1996). During the high humidity episodes, the particle hygroscopic growth and condensation likely result in an increase of the coarse aerosol fraction; hence, fewer particles were collected as part of the $PM_{2.5}$ fraction.

Although clear trends with the meteorological variables have been revealed from the data analysis, it is acknowledged that the meteorological parameters were recorded far from the measurement sites. The effect of meteorological factors on $PM_{2.5}$ and its elemental composition could have been assessed more precisely if these parameters were measured in a close proximity to each sampler.

3.7. Traffic intensity

As mentioned in the Section 1, the metropolitan area of Greater Cincinnati is characterized by very intense highway traffic per capita, which is expected to further increase in the future. Our analysis shows that the variation of highway traffic intensity was low during all

Table 5
Correlation matrices for meteorological parameters and $PM_{2.5}$ and its elemental concentrations in the city center—Findlay site

Parameter	$PM_{2.5}$	Al	Si	S	Ca	Mn	Fe	Ni	Zn	Pb	EC	OC
Wind speed (m/s)	−0.59	−0.40	−0.42	−0.43	−0.40	−0.68	−0.62	−0.59	−0.44	−0.48	−0.75	−0.71
Temperature (°C)	0.59	0.46	0.45	0.72	0.29	0.37	0.29	0.13	−0.18	−0.03	0.37	0.43
RH (%)	0.27	−0.34	−0.37	0.29	−0.44	−0.16	−0.28	0.10	0.00	−0.08	0.02	0.04

Values denote Spearman coefficient of correlation (R). Significant correlations ($p < 0.05$) are shaded; $n = 56$.

the monitoring cycles, i.e., a traffic pattern appeared to be rather homogeneous. The influence of the traffic volume on the concentration of $PM_{2.5}$ and its elements was analyzed in two sites, Grooms and Findlay.

The variation of the traffic intensity, expressed by the CV of the total daily highway traffic was 5.1% near the Grooms site, with 4.2% for gasoline vehicles and 9.5% for diesel vehicles. The above variations near the Findlay site were 3.2%, 3.7% and 3.3% accordingly. The average total vehicle flow during the measurement days on I-75 was 158,767 vehicles per day, of which 89% were gasoline vehicles and 11% were diesel vehicles. The daily number of vehicles determined on I-275 was 129,576, with 91% of gasoline and 9% of diesel vehicles, respectively.

The regression analysis showed that in the Findlay site only 1% of the variation of the $PM_{2.5}$ mass concentration could be attributed to the traffic density fluctuation ($r^2 = 0.01$). This is most likely because this fluctuation itself was very low, which did not allow us to characterize the influence of this factor on the $PM_{2.5}$ aerosol. It was concluded that in urban areas with relatively uniform highway traffic pattern the meteorological factors and secondary sulfate produce a major effect on the variability of the $PM_{2.5}$ and its elemental concentrations.

4. Summary and conclusions

The ambient $PM_{2.5}$ concentration and elemental composition were measured during an annual period in a large metropolitan area with intense highway traffic. The spatial and temporal variations were determined. The following conclusions were made based on the gravimetrical, elemental and ECOC (carbon) analysis of ambient $PM_{2.5}$ aerosol. The $PM_{2.5}$ concentration showed low spatial variation (median CV = 11.4%) during all measurement cycles. No significant contribution from local (point and linear) pollution sources was noticed; however, the city center measurement site (Findlay) constantly showed slightly higher $PM_{2.5}$ concentration. There was a noticeable seasonal variation of the $PM_{2.5}$ concentration. The higher $PM_{2.5}$ concentrations occurred during the summer, when relatively high ambient temperature, humidity, and solar radiation resulted in the increased formation of the secondary aerosol from SO_2 and VOC. The analysis of elemental composition revealed that the major amount of $PM_{2.5}$ mass was contributed by organic carbon, followed by sulfate, elemental carbon, crustal elements, and trace metals.

The sulfur concentration showed low spatial variation but significant daily variation. The concentration of sulfur correlated well with the $PM_{2.5}$ concentration at all sampling sites ($R = 0.81 - 0.89$). This finding can be explained by the influence of regional coal power plants situated along the Ohio River valley. These power plants are potentially the major contributors to the $PM_{2.5}$ mass in the Greater Cincinnati area.

Trace metals showed high spatial ($CV_{Zn} = 33.1\%$ to $CV_{Ni} = 61.9\%$) and temporal variation ($CV_{Mn} = 43.6\%$ to $CV_{Ni} = 68.7\%$) reaching peak values in the city center site during an episode of smog. Higher concentrations of crustal elements were also observed in the city center and attributed to the resuspended road dust particles. The data collected in the city center showed high correlation between trace metals, elemental and organic carbon. In contrast, the data from suburban and rural sites showed no clear pattern of correlation between elemental and organic carbon. The good correlation observed in the city center reflects the influence of intensive highway traffic and hilly topography (the latter is unfavorable for dispersion).

The particle size-selective sample analysis performed in the city center showed that a significant amount of the $PM_{2.5}$ mass is contributed by particles of $d_{50} = 0.32 - 1.0 \, \mu m$. The elemental analysis of different size fractions revealed ordinary distributions for traffic related aerosol elements, with crustal elements having peak mass concentrations in the particle size range of $5.6 - 10 \, \mu m$ and trace metals and sulfur peaking at the size range of $0.32 - 1.0 \, \mu m$.

The meteorological factors showed fair correlation with the $PM_{2.5}$ concentration. The sulfur concentration was the most sensitive to the ambient temperature changes. The wind speed and relative humidity had low negative correlation with $PM_{2.5}$ and its elemental concentrations. The obtained low correlation values imply that local meteorological stations should be positioned near the air sampler in order to obtain stronger correlations.

The traffic intensity in Cincinnati highways showed low variation of daily values. The influence of traffic flow variations to the $PM_{2.5}$ and its elemental concentrations could not be estimated due to low variation of the mobile source intensity.

Resulting from this investigation, an ambient monitoring network was designed for a large epidemiological study, which aims at assessing children's exposure to traffic-related particles.

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