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Intensive Short Term Measurements of the Ambient Aerosol in the Greater Cincinnati Airshed

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As part of a larger study undertaken in the Greater Cincinnati area to determine if diesel truck emissions are adjuvant to naturally occurring bioaerosols in the initiation of allergies in children, a more detailed intensive measurement campaign was undertaken to elucidate the characteristics of the ambient aerosol and compare to the regular, integrated measurements being conducted. The mass concentration, total number concentration, size distributions, and morphologies were established at several locations including a residential area far from major traffic (Mernic), a suburban area on both sides of a major highway (I-275, Blue Ash), a site in the city center very close to the highway (I-75, Findlay), and an enclosed oval track at a Truck Driving School.

Differences between real-time tapered element oscillating microbalance (TEOM) average mass concentrations and integrated Harvard impactor (HI) measurements were observed, with the magnitude of the difference being dependent on location and the organic compounds (OC) concentrations in the sample. Qualitative variation of the peaks in real-time PM 2.5 concentrations were observed with variation in truck traffic at the Findlay site; and no peaks in real-time PM 2.5 levels were observed at Mernic. Minimal variation in PM 2.5 was observed with distance from the highway at the Blue Ash site (fewer trucks). The site at Mernic had a smaller fraction of aggregated particles in comparison to the other sites. The two-dimensional fractal dimensions measured at the Findlay, Blue Ash, and Truck Driving School sites were statistically identical (1.58–1.61) but were higher than that measured at the Mernic site (1.41). Implications of the intensive measurement campaign vis-à-vis the epidemiological study are discussed briefly.

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INTRODUCTION

The inhalation of the fine fraction of the ambient aerosol may cause adverse health effects, as documented by several researchers (Schwartz and Dockery 1992; Oberdorster et al. 1994, 1995; Dab et al. 2001). Of specific concern are the potential health risks to children and the elderly living in urban areas. In particular, traffic-related aerosol emissions are receiving significant attention (Zhu et al. 2002b; Reponen et al. 2003). Among vehicular exhausts, diesel engine emissions are of particular interest due to their small particle size, chemical composition, and shape (morphology). The United States Environmental Protection Agency (USEPA) has recently classified diesel emission particles as being carcinogenic with chronic exposure and an irritant with acute exposures (USEPA 2002).

The State of Ohio is the region with the fifth highest number of miles of Federal Highways in the United States (FHWA 2001); recent studies indicate there will be as much as a 60% increase in heavy duty vehicles that pass through the region (FHWA 2001; ODOT 2002a). A large epidemiological study is underway in the Greater Cincinnati area to determine if diesel truck emissions are adjuvant to naturally occurring bioaerosols in the initiation of allergies in children. The initial results of the sampling study have pointed out trends in PM2.5 concentrations in regions close to the highway (Reponen et al. 2003). A follow up study was designed to explore the factors that cause the spatial and temporal variations in integrated PM2.5 aerosol and its elemental concentrations in this urban environment (Martuzevucius et al. 2004). A low spatial variability was observed in the PM2.5 concentrations during all measurement cycles. However, a noticeable seasonal variation of the PM2.5 concentration was observed with higher concentrations in the summer, probably due to enhanced formation of the secondary aerosol from SO_2 and VOCs.

In most urban monitoring sites, an integrated 24 h sample is collected and evaluated, as the current regulations are based on a

24 h particulate matter (PM) standard. Such measurements also provide a sufficient mass to perform a detailed elemental speciation using conventional techniques. While an integrated measurement is probably adequate for relating to long-term health effects, it does not provide details on the spatial or short-term temporal variations, or for that matter an acute or short-term health response. Several of the EPA Supersites are collecting near-real-time data, and the observations are also resulting in interesting interpretations of atmospheric fine-particle characteristics (McMurry and Woo 2002; Solomon et al. 2003).

The epidemiological study that is being performed in the Greater Cincinnati area would be tracking the health effects of as many as 879 children (CCAAPS 2003). While it is understood that real-time personal sampling is the most accurate indicator, due to practicality considerations this is not always possible or even economically feasible. As the impact on longer term health effects are to be identified, integrated measurements may suffice in establishing the broader correlations. However, to obtain additional information of the ambient aerosol over shorter time scales, a detailed intensive measurement campaign was undertaken in the Greater Cincinnati area using a battery of real time and integrated instruments (a tapered element oscillating microbalance (TEOM), an optical particle counter, a condensation particle counter (CPC), a scanning mobility particle sizer (SMPS), and an electrostatic aerosol sampler). A description of real-time measurements (mass, number, and morphology) that were performed at a few locations in the Greater Cincinnati airshed (a site close to the interstate in the city center (Findlay), a residential site close to the interstate (Blue Ash), a site distant from major roadways (Mernic), and a site dominated only by truck traffic (Truck Driving School)) is given in this article. A comparison of the results of this intensive campaign with the longer term integrated measurements (using Harvard Impactors (HI)) that are being routinely used in the epidemiological study is provided; and qualitative discussion of trends with traffic counts is described.

EXPERIMENTAL METHODS

The intensive campaign measured mass concentration, total particle number concentration, particle size distributions, particle morphology, and meteorological parameters. Mass concentrations were measured with both the TEOM and the HI. The TEOM (Rupprecht & Patashnick, Series 1400a, Albany, NY, USA) was used to measure PM2.5 mass concentration on a real-time basis. The TEOM operated with a filter temperature of 50°C. Readings were obtained between every 5 and 300 s, depending upon the site and the length of sampling for that day. All TEOM measurements were then averaged over a 10 min period. Two parallel HIs (MS&T Area Sampler, Air Diagnostics and Engineering, Harrison, ME, USA) operating on separate pumps were used to measure the integrated mass concentration. Sample duration varied between 5 and 24 h depending upon the site and the sampling day (Table 1). The impactor flow rate was measured and recorded at the beginning and end of the sampling period, from which the total volume sampled was calculated. Samples were obtained on both Quartz (Whatman, 1851-037, Kent, ME) and Teflon (Pall Gellman, R2PL037, Ann Arbor, MI) 37 mm filters at 20 l/min flow through the HI, corresponding to a cutpoint of 2.5 μm. Quartz and Teflon filter data were always collected at the same time, with one filter of each type in each of the two impactors. The filters were conditioned in a humidity chamber and weighed both prior to and after sampling to obtain the PM2.5 mass concentration. The Quartz filters were analyzed for elemental and organic carbon by the NIOSH 50–40 method (Sunset Laboratory, Hillsborough, NC, USA). Teflon filters were analyzed via X-Ray Fluorescence to determine elemental concentrations (Chester Lab Net, Tigard, OR, USA).

Meteorological data was collected during sampling with a Vantage Pro Weather Station (Model 6150, Davis Instruments, Baltimore, MD, USA). Temperature, wind speed, wind direction, relative humidity, barometric pressure, and other parameters were recorded at 5 min time resolution at a height of 2 m above the ground. Wind directions were calculated by aligning the Weather Station with magnetic North. The measurements were not corrected for magnetic declination, which was small (4.5°W) (NOAA 2003).

Particle size distributions between 15 and 660 nm were obtained with a differential mobility analyzer (DMA) (Model #3080, TSI, St Paul, MN, USA), while size distributions between 4.6 and 157 nm were obtained with the nano-DMA (Model #3085, TSI). The DMAs were used in conjunction with a CPC (Model # 3025A, TSI) running in the SMPS mode. These distributions were measured periodically at every sampling site. A portable CPC (Model 8525, TSI, range of "0.02 to greater than 1 μm" (TSI 2002)) measured total aerosol number concentrations, while a portable optical particle counter (Model 1.108, Grimm Technologies, Douglasville, GA, USA) measured the number concentrations of particles ranging from 0.3 to 3.0 μm in 8 bins (greater than 0.3, 0.4, 0.5, 0.65, 0.8, 1.0, 1.6, 2.0, and 3.0 μm). In addition, aerosol samples were collected on Lacey electron microscope grids (Model # 01883, Ted Pella Inc., Redding, CA, USA) via an electrostatic aerosol sampler (Model 3100A, TSI) for morphological analysis.

As there was only one set of equipment for measurements, data collected at different sites was taken on different days. An attempt was made to keep collection times at each location as close as possible to those at other locations. All measurements were taken on weekdays. For the Blue Ash location, measurements were taken at different distances from the highway by moving the equipment from location to location.

SITE SELECTION

Four sites were chosen for this campaign (Figure 1a). The Findlay site (Figure 1b) was chosen because of its proximity to the City Center and Interstate 75. It is situated in a lower income area in the downtown section of the city. The samplers were

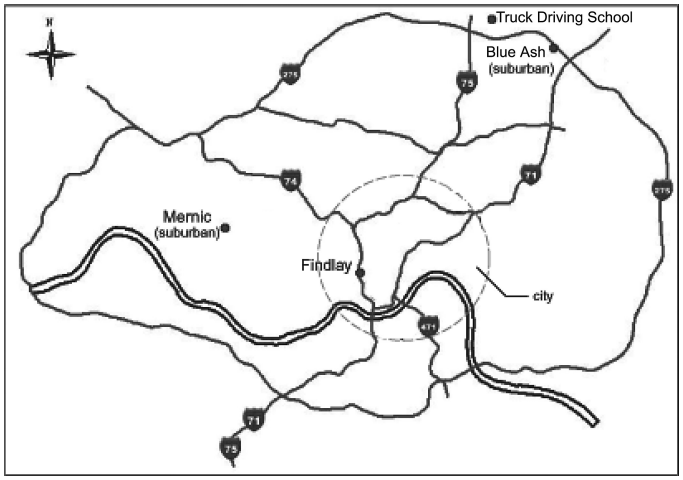
Table 1
Measured PM2.5, organic carbon, elemental carbon, and elemental mass concentrations at each of the four sites

	Blue Ash	Findlay	Truck test track	Mernic*	
Samples	2	2	1	2	
Collection time	9 am–6 pm	9 am–9 am	9 am–4 pm	9 am–9 am	Limit of detection[1,2]
PM2.5	7300	27000	89000	14000	350
EC	680	1800	2200	450	60
OC	3900	5400	10000	3000	60
Na	37	120	ND	65	1.22
Mg	4.8	18	730	12	0.73
Al	41	76	980	34	4.04
Si	120	220	3200	97	1.84
S	880	3800	2500	2300	0.6
K	34	80	500	82	1.45
Ca	89	280	7800	110	2.07
Ti	8.5	10	80	4.7	3.88
V	0.61	1.0	ND	0.24	1.22
Cr	0.046	1.7	2.0	0.77	0.69
Mn	2.1	5.6	34	1.9	0.18
Fe	100	280	970	74	0.16
Ni	0.5	1.2	1.3	0.7	0.14
Cu	2.2	5.3	ND	1.0	0.16
Zn	13	53	14	13	0.23
Ge	0.22	0.29	ND	0.31	0.25
As	0.92	2.2	ND	1.1	0.18
Se	2	3.3	4.4	2.4	0.16
Br	1.5	4.3	3.5	2.5	0.14
Rb	0.32	ND	3.6	ND	0.16
Sr	0.85	1.5	13	0.08	0.25
Y	ND	ND	ND	0.013	0.28
Zr	0.48	1.4	ND	0.66	0.28
Mo	0.13	0.46	0.45	0.98	0.37
Pd	1.4	ND	ND	0.28	5.26
Ag	ND	0.31	3.0	0.81	4.64
Cd	ND	0.10	ND	ND	5.05
Sn	2.2	16.4	1.7	3.3	7.00
Sb	ND	1.0	ND	1.1	7.21
Ba	8.4	23	37	6.0	11.89
La	ND	5.9	ND	6.9	16.2
Hg	0.033	0.025	ND	0.36	0.34
Pb	0.77	7.4	5.8	2.8	0.34

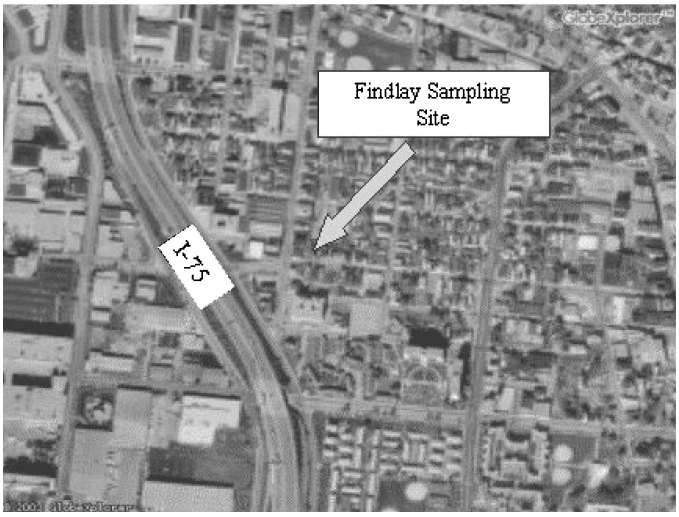
*The second Mernic sample was a 20 h average.
1, EC/OC LODs from NIOSH (1994).
2, XRF LODs from USEPA (1999).
Values given are in ng/m^3. ND, not detected.

placed on the roof of a building approximately 200 m east of one section of the highway and about 400 m south of another portion of the highway. Since the interstate is also elevated in this area, there was no appreciable difference in elevation between the samplers and the interstate. Sampling began on the evening of 24 July 2002 and ended in the morning on 27 July 2002.

The Mernic site was situated in a middle-income neighborhood. It was 4590 m from major highways and was not expected to be influenced by traffic-type sources. The samplers were set



(a)



(b)

Figure 1. (a) Locations of sampling sites within the Greater Cincinnati Airshed. (b) Photograph of the Findlay sampling area in relation to the major interstate highway I-75.

up in the backyard of a local residence, approximately 50 m from the street and approximately 300 m from the closest busy street. Sampling started on the evening of 30 July 2002 and ended two nights later.

To compare ambient emissions to those of a cluster of trucks, sampling was performed at a Truck Driving School (TDS). Large trucks drove in an oval track under all load conditions between 5 and 50 m from the samplers. Approximately 75 trucks passed the samplers every hour. Measurements were taken at the Truck Driving School in the late morning and early afternoon of 29 July 2002.

The Blue Ash location included two sampling sites directly across the highway from each other (Blue Ash South and Blue Ash North). The neighborhood is considered upper-middle class. The samplers were moved as close as 25 m and as far as 200 m on both sides of the highway. The sampling location on the south side of the interstate was about 10 m below the level of the highway, while the location on the north side of the interstate was at the same level as the highway. Sampling occurred on the south side on the morning of 23 July and on the north side in the afternoon of the same day.

RESULTS AND DISCUSSION

A comparison between the TEOM and HI measurements is discussed first. This is followed by a comparison of the results of all of the measurements at different sites. The similarities between the site that was close to the highway (Findlay) and the concentrated diesel source site (TDS) are then elucidated.

Mass Concentrations by TEOM and HI

Mass concentrations measured by the TEOM and wind speed and direction are plotted in Figures 2a and b for Findlay and Mernic, respectively. The averaged PM 2.5 TEOM concentration over a 24 h period is also shown along with the integrated 24 h PM2.5 as measured by the HI. Table 1 summarizes the elemental concentrations measured with the HIs. A comparison of the TEOM and HI measurements and the percent differences are summarized in Table 2.

At the Findlay and TDS sites, the TEOM measured PM2.5 concentrations that ranged from 18 to 42% of the HI Teflon values. The TEOM operates at 50°C, and it is well known that because of this high temperature volatile and semivolatile particles could be lost (Allen et al. 1997; Ayers et al. 1999; Mignacca and Stubbs 1999; Eatough et al. 2003). In contrast, the HI operates at ambient temperatures. The difference between the TEOM and HI measurements was less at Mernic (TEOM values 89–101% of HI values). The differences in measurements are due to the nature of the aerosol at these sites. The Findlay and the TDS sites are more influenced by traffic-type sources and have a larger amount of volatile particles such as organic carbon (OC). The Mernic aerosol, however, is dominated by a background sulfate aerosol (which is less volatile) and low OC levels (Table 1). The ratio of PM2.5 measured by the HI and TEOM was plotted as a function of the OC concentration for all of the samples (Figure 3) and a strong correlation was observed ($R^2 \sim 0.99$). The difference in TEOM and HI measurements at Findlay was greater (Figure 2a, 25 July) when the wind blew from the highway, further confirming that the traffic aerosol was a greater contributor of the volatile content. As a side note, mass concentrations measured by the HI using quartz filters were consistently lower than those measured using Teflon filters at all sites (Table 2), probably due to a random combination of artifacts as described by Chow (1995) and Turpin et al. (1994).

TEOM Concentrations and Traffic Data

As discussed in the previous section, there was a qualitative indication that traffic sources were affecting PM2.5 levels at the

Table 2
Twenty-four hour average PM2.5 concentrations (in $\mu g/m^3$) measured at three sites in Cincinnati

24 h averages measured by	Findlay 7/24		Findlay 7/25		TDS 7/29**		Mernic 7/30		Mernic 7/31***	
	Average ($\mu g/m^3$)	HI teflon value (%)	Average ($\mu g/m^3$)	HI teflon value (%)	Average ($\mu g/m^3$)	HI teflon value (%)	Average ($\mu g/m^3$)	HI teflon value (%)	Average ($\mu g/m^3$)	HI teflon value (%)
TEOM*	9.7 ± 7.4	42	13.2 ± 4.9	34	15.7 ± 10.7	18	13.3 ± 3.3	101	18.7 ± 7.3	89
HI teflon	23.1	100	39.2	100	88.8	100	13.2	100	21	100
HI quartz	19.8	86	31.3	80	64.8	73	4.7	36	16.9	80

*$\pm$ values represent one standard deviation within a day.
**Average value for the Truck Driving School is a 7 h average.
***Average value for Mernic on 7/31 is a 20 h average.

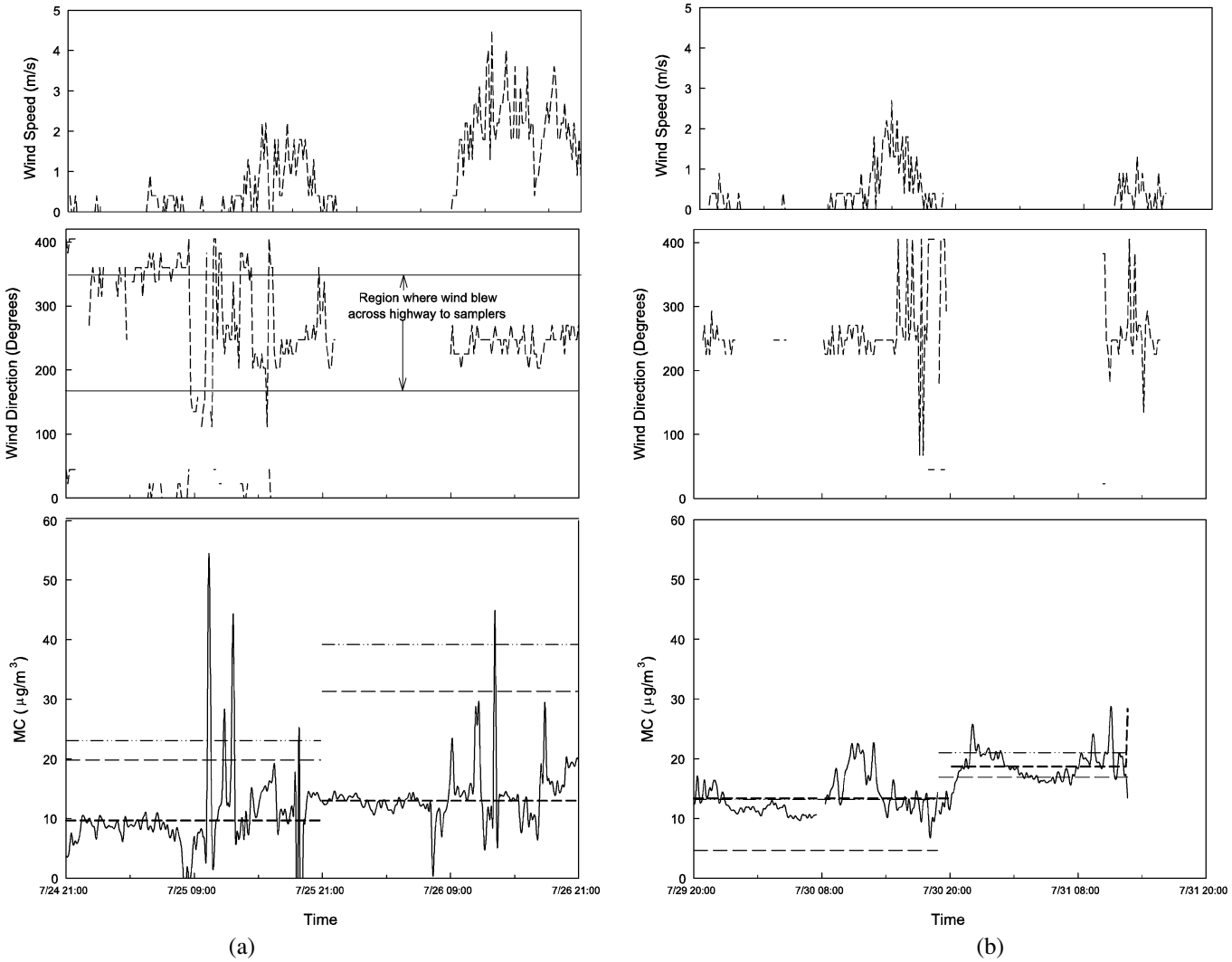


Figure 2. (a) Ten-minute and 24 h average mass concentrations at Findlay, measured by TEOM and HIs, as well as wind direction for the same days. The two solid lines in the wind direction figure indicate the time when the wind was directed from the highway to the sampling location: —- ●● —-, HI Teflon; —- — —-, HI quartz; **- - - - -**, 24 h TEOM average; — —, 10 min TEOM average. (b) Ten-minute and 24 h average mass concentrations at Mernic, measured by TEOM and Impactors, as well as wind direction for the same days: —- ●● —-, HI Teflon; —- — —-, HI quartz; **- - - - -**, 24 h TEOM average; — —, 10 min TEOM average.

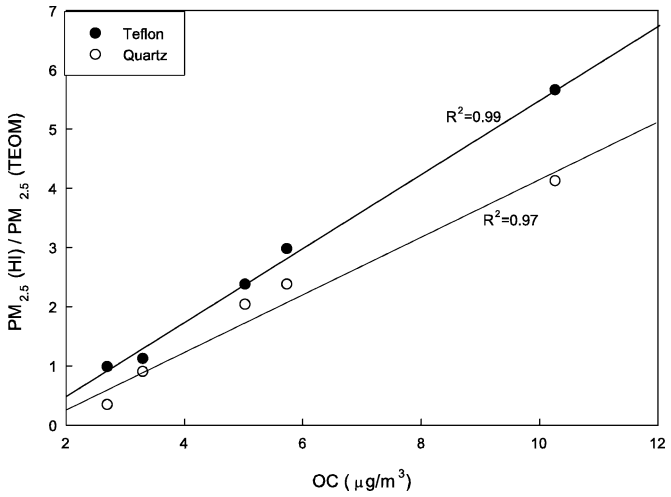


Figure 3. Ratio of PM2.5 measured by the HI (Teflon and quartz) and TEOM as a function of OC levels at all sites.

Findlay site. Figure 4 illustrates the temporal trends for automobile (defined as classes 1–3 of the Federal Highway Administration's vehicle classification scheme "F") and truck (classes 4–12 of the same scheme) traffic density (ODOT 2002b), along with PM2.5 mass concentrations for 25 and 26 July 2002 at the Findlay site. On both days of observation, truck traffic was highest between 9 am and 3 pm, with a second smaller peak between 6 and 9 pm. Automobile traffic was highest between 7 and 9 am during the morning rush hour, after which it decreased until the afternoon. In the afternoon, it then steadily increased until the evening rush hour around 5 pm. Ten-minute averaged TEOM mass concentrations peaked between 10 am and 1 pm, with a secondary peak between 6 and 7 pm. To ensure that the 10 min averaged mass concentration peaks were not simply small time scale peaks, the data was also smoothed to 1 h averages. The 1 h averages also had peaks during the same periods. Both 10 min and 1 h TEOM mass concentrations followed the truck pattern more closely than they followed the automobile pattern. While a qualitative match of the peak TEOM PM2.5 levels was observed with that of peaks in truck traffic, a detailed statistical correlation could not be done due to the mismatch on the temporal periods of the truck data that was available. On 25 and 26 July, correlation coefficients between PM2.5 (TEOM, 1 h average) and truck traffic were 0.38 and 0.24, respectively, while correlation coefficients between PM2.5 and automobile data on the same days were 0.31 and 0.21. These low correlation coefficients are probably due to the mismatch in the temporal scales of the traffic data and the TEOM PM2.5 levels.

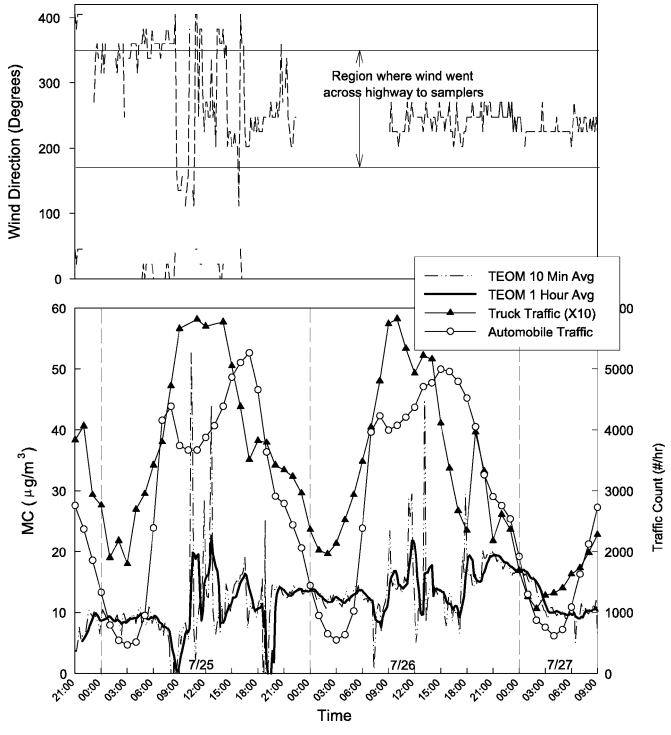


Figure 4. Automobile and heavy-duty truck traffic density and average mass concentrations (TEOM), versus time, at the Findlay sampling site (25 and 26 July 2002). The two solid lines in the wind direction figure indicate the time when wind was directed from the highway to the sampling location.

In summary, the TEOM measurements clearly elucidate that traffic-type sources are affecting short-term peak PM2.5 levels at the Findlay site. Furthermore, there is a qualitative indication that the peaks in PM2.5 are from truck-type traffic sources. The absence of these peaks at the Mernic site (Figure 2) further corroborate that the Findlay site is impacted by truck-traffic sources. The epidemiological study is relying upon integrated PM2.5 measurements, and it will be essential to use the elemental concentrations in conjunction with robust statistical receptor modeling methodologies to unravel the quantitative contribution of traffic-type sources; this will be the subject of a future study.

Morphology of the PM2.5 Samples

In addition to concentration and chemical measurements, aerosols were collected using an electrostatic collector on electron microscope grids at all of the sites to determine their morphology. The morphology of the particles may throw additional insights into the source of the particles and may also be important vis-à-vis health effects. The electrostatic collector assembly was connected to the outlet of a PM2.5 head, and details of the system are described elsewhere (McDonald and Biswas 2003). The collected particles were viewed in a scanning electron microscope, and the images were analyzed to establish the distribution of the particles by shape (spherical, fibrous, aggregate, and other) and to determine the fractal dimensions of the aggregates. The fractal dimensions of the aggregated aerosols were calculated using the method of nested squares (Katrinak et al. 1993; McDonald and Biswas 2003).

Table 3 lists the distribution of the shapes of the particles as classified into four categories. The only obvious difference

is that the Mernic site had a much smaller aggregate fraction (2%) in contrast to the other sites (approximately 11%). This low fraction of aggregate particles is another indicator that the Mernic aerosol is not influenced by traffic-type sources. The Mernic site has a larger fraction of spherical particles, which is probably due to the sulfate aerosol, and is expected to be spherical due to the high humidity cycles (ranges from 40 to 100%), with values being high in the morning hours. Other than Mernic, the other three sites are closer to the highway or influenced by traffic-type sources; the TDS site with the most direct influence of truck emissions had the highest fraction of aggregated particles (17%).

The aggregates (Figure 5) were further analyzed to determine their fractal dimensions; and the average two-dimensional (2D) fractal dimensions (D_f) measured at the Findlay, TDS, and Blue Ash sites were similar (1.60 $\pm$ 0.10, 1.58 $\pm$ 0.08, and 1.61 $\pm$ 0.08, averaged over 163, 20, and 27 samples, respectively). As indicated in the previous paragraph, these sites are all in the vicinity of major highways or clearly influenced by traffic-type sources (trucks at TDS). The average value of D_f

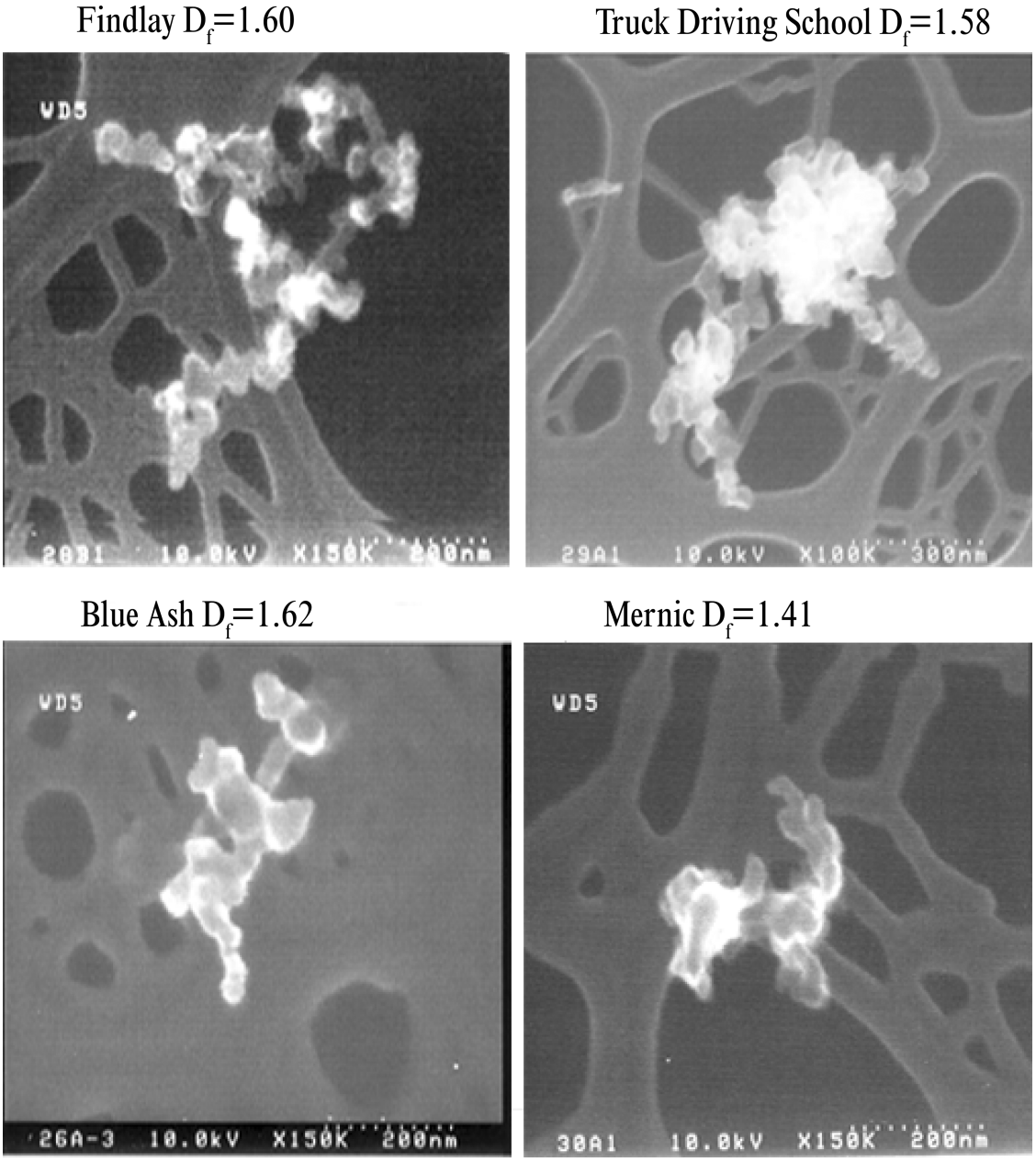


Figure 5. SEM images of aggregates collected at the four sites. Also listed are the average 2D fractal dimensions.

Table 3
Shape distributions of collected aerosols and average Df values of aggregates

Site	Fraction of aerosol				Aggregate D_f
	Spherical	Fibrous	Other	Aggregate	
Findlay	0.14	0.17	0.58	0.11	1.60 ± 0.10
Blue Ash	0.07	0.21	0.62	0.11	1.61 ± 0.08
Truck driving school	0.21	0.13	0.49	0.17	1.58 ± 0.08
Mernic	0.13	0.22	0.64	0.02	1.41 ± 0.09

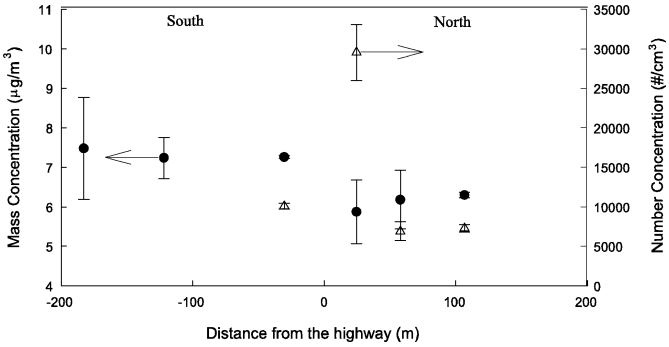


Figure 6. PM2.5 (TEOM) and number concentrations (CPC) measured at the Blue Ash site on 24 July 2002. Error bars represent one standard deviation. Sampling durations vary between 90 and 200 min.

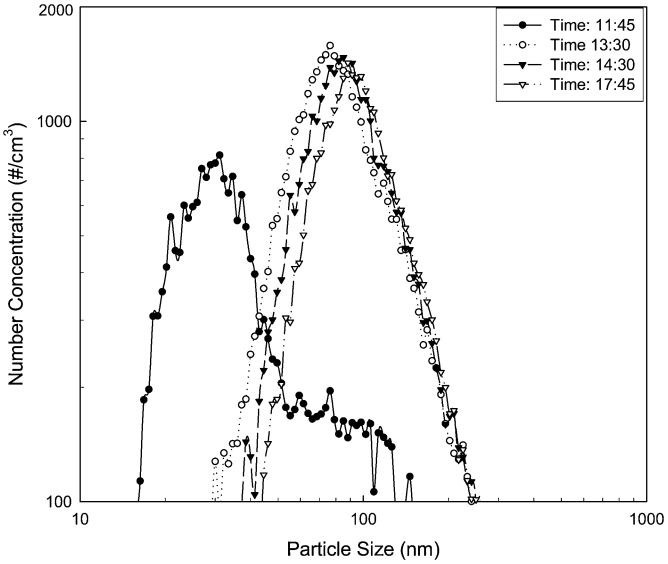
for the aggregates was lower at Mernic (1.41 ± 0.09, averaged over 12 samples). The Mernic site is not influenced by traffic-type combustion emissions, and the agglomerate particles are more a result of aggregation of freshly nucleated particles (that have not reached their self-preserving morphologies). While the sulfate aerosol is expected to be more dominant at this site, it is not expected to be agglomerated and is most likely spherical (confirmed by the larger fraction of spherical-shaped particles in comparison to other sites; Table 3); as nucleated sulfate particles are expected to be present in the liquid state at the higher humidities. The freshly nucleated particles could be formed by dry photochemical oxidation pathways—some of this has been observed in other studies (Jimenez et al. 2003)—or could also be emissions from other nontraffic combustion sources. The determination of the species that forms the aggregates at the Mernic site was not determined in this study.

Spatial and Temporal Variations

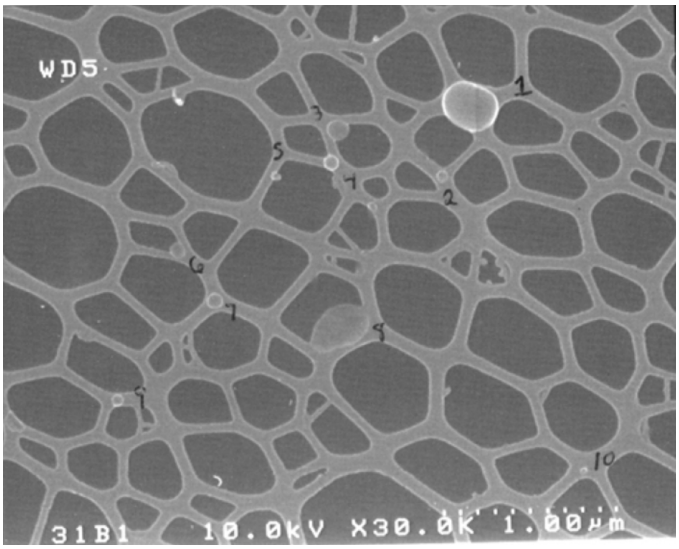
Spatial and temporal surveys of PM2.5 were conducted at the Blue Ash site by measuring the mass and number concentrations as a function of distance from the highway, on the south and north sides. The south side was upwind from the highway during sampling, the north side was downwind, and the wind direction stayed fairly constant on the day of the sampling. Sampling occurred at the Blue Ash South site in the morning and at the Blue Ash North site in the afternoon. The spatial variation of the PM2.5 mass concentration (measured by the TEOM) and number concentration (measured by the CPC) at the Blue Ash site is shown in Figure 6. PM2.5 mass concentrations did not significantly change with distance from the highway (as established by a single factor anova analysis, $p = 0.36$), nor did it change from one side of the highway to the other. In contrast, the number concentration was very high 25 m downwind of the highway (north side) and quickly dropped to near-constant levels 50 m from the highway. These results are consistent with other reports in the literature that have indicated that number concentration decays within 300 m of the roadway (Zhu et al. 2002a; Reponen et al. 2003).

The size distribution of the particles may be a more complete descriptor of the aerosol, and interesting temporal variations were observed. As an example, the measured size distribution at the Mernic site on 31 July 2002 is plotted in Figure 7a for various time durations. In the morning (11:45 am) the peak size (30 nm) and total number concentration (2.16×10^4 cm^{-3}) are both lower than in the afternoon (75 nm and 3.82×10^4 cm^{-3}). There could be several reasons for these observations. It could be that the wind was carrying in particles from another location. However, during this sampling period, meteorological parameters were fairly constant (wind ± 20 degrees, temperature increased 4 degrees, relative humidity decreased 15%), ruling out carryover from different locations. The other explanation is that a nucleation event could be taking place, resulting in the formation and growth of the particles. The presence of a large sulfate aerosol (Martuzevicius et al. 2004; see also Table 1) is supportive of this, as the gas-to-particle conversion to the sulfate would increase in the afternoon hours due to enhanced conversion with rise in temperature. The rapid nucleation is followed by particle growth, resulting in the larger mean size (75 nm). The particles were more spherical in shape (Figure 7b), indicating that it was probably growth of a hygroscopic sulfate aerosol. Clearly, more detailed and shorter time resolution size distribution measurements would be necessary to firmly point out the onset of the nucleation event.

On examining the ultrafine sizes (as measured by a nano-DMA), the number concentration of particles increased threefold (from 1×10^4 cm^{-3} to 3×10^4 cm^{-3}), along with an associated increase in the volume concentration (5.6×10^9 nm^3 cm^{-3} to 14.6×10^9 nm^3 cm^{-3}) between the 11:45 am and 1:30 pm measurements. After 1:30 pm, the number concentration of the ultrafine particles decreased, while the total volume concentration stayed constant, and the mean particle size increased. These clearly point out that coagulational growth maybe a dominant process in the afternoon hours.



(a)



(b)

Figure 7. (a) Size distributions measured with the DMA at Mernic on 31 July 2002. (b) Electron micrograph of numerous small spheres collected at the Mernic site.

CONCLUSIONS

A short-term intensive measurement study was conducted to support the long-term integrated measurements being conducted for the Cincinnati Childhood Allergy and Air Pollution Study. While the integrated measurements are being used as a basis of the exposure measurements, the short-term intensive study elucidated interesting behavior on the nature of the aerosol in the airshed. Though more extensive short-term measurement campaigns need to be undertaken to understand thoroughly the dynamics of the ambient aerosol, the short-term intensive studies resulted in several interesting findings. The role of traffic-type sources on specific locations was evident, and clear differences were deciphered between the residential site (Mernic) and those closer to the highway (Findlay).

The degree of volatility of the samples also varied from location to location, and this was clearly observed via the TEOM-measured mass concentrations. The excellent correlation between the TEOM/impactor deviation and OC content indicates that the negative artifact in the TEOM was most likely due to organic aerosol volatilization in the TEOM at the Findlay site. Locations closer to the highway (Findlay) or more influenced by truck-type sources (TDS) exhibited greater differences between TEOM and HI measurements. Qualitative variation of PM2.5 levels with associated traffic density peaks were observed at the Findlay site, though a direct correlation exhibited a low coefficient due to probable mismatch in the temporal values of the correlated data sets.

Interesting trends were also observed in the distribution of shapes of the particles at the various sites. The residential site (Mernic) that was not influenced by traffic sources had much fewer aggregated particles. Furthermore, there was a difference in the fractal dimension of these fewer aggregates at the Mernic site in comparison to the other sites. The other sites, Blue Ash, Findlay, and the TDS had aggregates with similar fractal dimensions. While distribution of shapes of particles and the aggregate characteristics such as fractal dimensions are not typically reported in ambient exposure studies, they may throw additional insights into the source of particles. Furthermore, they may also be important vis-à-vis their ultimate health effects. This is a subject that clearly deserves additional studies to develop conclusions firmly.

No significant mass concentration variations were observed as a function of distance from the highway; though the number concentrations showed clear peaks in the near vicinities of the highway (in the downwind directions). Size distribution measurements also enable the viewing of dynamic changes in the aerosol and establishing contribution of various growth events.

The cumulative set of measurements indicate that 24 h integrated PM2.5 mass concentrations by themselves would not be good indicators of traffic source contributions, especially as the sulfate aerosol is dominant in the entire Greater Cincinnati airshed. It will be imperative to use the chemical element signatures and additional measurements to decipher the contributions and spatial variations of traffic-type sources.

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