

Concentration gradient patterns of aerosol particles near interstate highways in the Greater Cincinnati airshed

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The objective of this study was to determine if there is an exposure gradient in particulate matter concentrations for people living near interstate highways, and to determine how far from the highway the gradient extends. Air samples were collected in a residential area of Greater Cincinnati in the vicinity of two major highways. The measurements were conducted at different distances from the highways by using ultrafine particle counters (measurement range: 0.02–1 µm), optical particle counters (0.3–20 µm), and PM2.5 Harvard Impactors (0.02–2.5 µm). The collected PM2.5 samples were analyzed for mass concentration, for elemental and organic carbon, and for elemental concentrations. The results show that the aerosol concentration gradient was most clearly seen in the particle number concentration measured by the ultrafine particle counters. The concentration of ultrafine particles decreased to half between the sampling points located at 50 m and 150 m downwind from the highway. Additionally, elemental analysis revealed a gradient in sulfur concentrations up to 400 m from the highway in a residential area that does not have major nearby industrial sources. This gradient was qualitatively attributed to the sulfate particle emissions from diesel engine exhausts, and was supported by the concentration data on several key elements indicative of traffic sources (road dust and diesel exhaust). As different particulate components gave different profiles of the diesel exposure gradient, these results indicate that no single element or component of diesel exhaust can be used as a surrogate for diesel exposure, but more comprehensive signature analysis is needed. This characterization is crucial especially when the exposure data are to be used in epidemiological studies.

Introduction

As the particulate air pollution generated by industry and power plants has decreased over the years, road traffic has become one of the major sources for airborne particle mass in urban areas.¹ For example, a recent emission inventory in London showed that 67% of PM10 (particles smaller than 10 µm) originate from diesel-based road transportation.² At the same time, several epidemiological studies have linked air pollutants emitted from traffic to an increase in respiratory symptoms.^{3–6} These studies used either the distance from the road or local traffic density as a measure of exposure. Studies focusing specifically on the airborne concentrations and fate of particles in the air near highways are few and their results somewhat inconsistent.

Janssen *et al.*⁷ compared the mass concentration and elemental composition of particles sampled at two sites near major roads (within 0.5 m from a busy street) and at two sites at background locations (1 000–2 000 m away from the nearest busy street). The authors concluded that PM10 and PM2.5 (particles smaller than 2.5 µm) concentrations were on average only 1.3 times higher near the road compared with the background, and elemental carbon readings were 2.6 times higher. Another study examined PM10, PM2.5, and black smoke (measured as a reflectance of a filter sample) at four different distances (15, 100, 150, and 300 m) from a major motorway.⁸ It was concluded that there was no significant

decrease in concentration of PM10 and PM2.5 with increased distance from the highway. Black smoke, however, decreased to about half between 15 and 300 m, with the largest decrease within the first 100 m from the highway. These gradients were more evident in periods when the sampling points were downwind at least 33% of the time. Kingham *et al.*⁹ measured PM10, PM2.5 and black smoke inside and outside of 13 homes located near a major road (<50 m) and in 14 homes located more than 50 m away from a major road. Their data show only weak spatial variation with distance from the road among the measured pollutants. They concluded, however, that black smoke in the PM2.5 fraction had the highest correlation between indoor and outdoor measurements. A group in the Netherlands studied the respiratory health of children attending schools located within 400 m of motorways.¹⁰ They reported that PM2.5 and “soot” (measured as the reflectance of filters) concentrations significantly increased with increasing truck density and significantly decreased with increasing distance from the highway. At an average traffic density at 13 146 trucks per weekday and average percentage of time downwind equal to 38%, PM2.5 concentration at 50 m was about 20% higher than at 400 m. The respective percentage for “soot” was 50%. A recent study by Zhu *et al.*¹¹ established gradients of the aerosol concentration near a highway in the Los Angeles area by measuring concentrations of particles as a function of distance from a highway. A steep gradient was found in total particle concentration, black carbon and CO concentrations up to distances of 150 m from an elevated highway. The concentration of ultrafine particles (6–220 nm) decreased exponentially with downwind distance from the

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highway and reached the background level at a distance of 300 m.

The effect of wind direction on the transport of traffic pollutants was studied by Hitchins *et al.*¹² They measured fine and ultrafine particle counts at 15, 55, 95, 135, 215, 295, and 375 m from a major road. The concentration of fine and ultrafine particles decayed to around half of the maximum at a distance of 100–150 m from the highway when the wind was blowing directly from the road toward the sampling sites. When the wind was blowing parallel to the road, the reduction to half of the concentration occurs at 50–100 m. No influence of vehicle emissions was observed when the wind was blowing towards the road from the sampling points.

Several ambient air quality studies have been performed in the Greater Cincinnati area,^{13–15} with the primary focus on the contribution of stationary sources. A 1998 investigation found that residents in the Greater Cincinnati metropolitan area were exposed to higher levels of smog more often than residents in Boston and New York and had higher rates of smog-related respiratory hospital admissions including asthma attacks and emphysema.¹⁶ In many urban areas in the USA, transportation sources contribute more than 50% of the total ambient PM2.5.¹ The Greater Cincinnati area has a heavy use of a concentric network of interstate highways forming major corridors for east–west and north–south deliveries (135 000–150 000 vehicles per day per interstate highway of which 12 000–16 000 are trucks), an extensive public bus system, and a heavily traveled railway network. This region, therefore, is ideal for characterizing and assessing exposures related to transportation sources.

While there are several studies that have evaluated the role of vehicular traffic on ambient pollution levels in the vicinity of highways, these studies have either primarily focused on gaseous pollutants, or on total mass concentrations or particulate matter number concentrations. Furthermore, the contribution of diesel engine exhausts has not been established due to the limited characterization of the particulate matter samples, especially in the Greater Cincinnati area. The aim of this study was to determine if there is a transversal gradient in the PM concentration and specific elemental concentrations in the vicinity of an interstate highway. Measurements were conducted at different distances from the highways by using ultrafine particle counters, optical particle size spectrometers, and PM2.5 Harvard Impactors. The samples were analyzed to determine PM2.5 levels and elemental concentrations (inorganic elements, elemental carbon and organic carbon) as a function of distance from the highway. The paper reports the results of a study, which has been conducted in support of a larger epidemiological investigation on the effect of traffic pollution on the incidence of atopy in newborn to age four children living near interstate highways.

Materials and methods

The study was performed in residential areas in the northern part of Greater Cincinnati near the I-75 and I-71 interstate highways. These highways run from south to north and the sampling locations were selected in an eastern direction from the highway. Since the prevailing wind direction in the Cincinnati area is from west and southwest, this configuration allowed us to determine the concentration gradient downwind from the PM source. The sampling locations were selected at different distances from the highway ranging from 50 to 1600 m (as measured from the edge of the highway). The first location represented the highest exposure, *i.e.*, the closest realistic distance of homes from a highway, and the last location represented suburban background exposure. At each location, the sampling was performed at a height of 1.5 m from the ground and at least 2 m away from any obstructions, such as

buildings and trees as recommended in the EPA PM2.5 sampling protocol.¹⁷

Aerosol particle measurements were performed at different distances from the highway during three periods, each lasting 2–5 days. Number concentrations of particles in the size range of 0.02–1 µm were determined with portable condensation nucleus counters, CNC (P-Trak, model 8525; TSI Inc., St. Paul, Minnesota). As most of the particles in the measured size range are expected to be smaller than 0.1 µm in size, the CNC results are referred to as ultrafine particles in this paper. The coarse fraction of airborne particles (0.3–20 µm) was measured with portable size-selective optical particle counters, OPC (Portable dust monitor, model 1.108; Grimm Technologies Inc., Douglasville, Georgia). Both the CNC and OPC were operated at a mode that saved concentration data in 1 min intervals in the data-logger. The PM2.5 aerosol fraction was collected on filters using Harvard Impactors (MS&T area sampler; Air Diagnostics Inc., Harrison, Maine) for subsequent gravimetric analysis and elemental analysis of the filter samples.

The first sampling period included OPC and CNC measurements performed at 50, 100, 200, 400, 800, and 1600 m east from I-75 and I-71 highways. The measurements were conducted for two days representing two different wind conditions. During the first day, the wind was blowing from the west, perpendicular to the highway, representing the typical wind condition in the Cincinnati area and allowing transversal measurements downwind from the highway. During the second day, the wind was blowing from the north, parallel to the highway. During the measurements, one CNC and one OPC were kept at 50 m from the highway, while the second CNC and OPC were moved to the other distances in random order. The measurements at each location lasted 10 min, and the last 5 min period at each location was used for the data analysis allowing time for stabilizing the instruments. The data were normalized by dividing all the concentration results by the corresponding result determined at a distance of 50 m.

The second and third sampling periods lasted 5 days each and included CNC and PM2.5 measurements at three distances from the I-71 highway: 80, 400, and 1600 m during the second sampling period, and 400, 800, and 1600 m during the third sampling period. The area near the I-71 highway was selected because traffic was anticipated to be the major source of the particulate matter air pollution, as there were no major industrial sources nearby. The particle count measurements with CNC were performed twice a day, at noon (12–1 pm) and during the afternoon rush hour (5–6 pm). For each test, the overall measurement time in the three sampling locations did not exceed 45 min, including 5–10 min in each location and the time needed to move the particle counters to the next location. Three PM2.5 impactors operated simultaneously at the three sampling locations. The samples were collected at a flow rate of 20 l min⁻¹ for 24 h onto Teflon filters (diameter = 37 mm, pore size = 2.0 µm; SKC Inc., Eighty Four, Pennsylvania). One trip blank was obtained for every 10 real PM2.5 samples (10%) by taking the filters to the field and back to the laboratory without exposing them. The gravimetric analysis was performed by weighing the filters before and after the sampling with a micro-balance (model A-160, Fisher Scientific, Pittsburgh, Pennsylvania). The filters were stabilized by keeping them in the refrigerator for 12 h before weighing. The elemental analysis was first performed for all the samples using PIXE (Proton Induced X-ray Emission) by Element Analysis Corporation (Lexington, Kentucky). Selected samples were reanalyzed using XRF (X-ray fluorescence analysis) performed by Chester Labnet (Portland, Oregon). During the third sampling period, additional samples were collected on quartz filters (Whatman, Maidstone, Kent, UK) for analyses of elemental and organic carbon. These analyses were performed by the Sunset Laboratory, Inc. (Forest Grove, Oregon) using thermal optical analysis.¹⁸

For quality control purposes, all the Harvard impactors were operated simultaneously at one sampling site for 5 consecutive 24 h measurements. The coefficient of variation of the measured PM_{2.5} concentration was 2.5%, showing that the instruments measure the PM_{2.5} concentration with very low variability.

During the first sampling period, the wind direction and speed were measured with a portable weather station at the 50 m location (6150 Vantage Pro, Davis Instruments Corp., Hayward, California). For the second and third sampling periods, hourly averages for wind speed and direction during the particle measurements were obtained from the local environmental monitoring station. This station was about 1 km south of the sampling site located at 80 m from the I-71 highway.

Results and discussion

Fig. 1 shows the results on the normalized counts of ultrafine (CNC) and coarse (OPC) particles measured at different distances downwind from the highway when the wind was perpendicular to the highway. The CNC results demonstrate a steep gradient in the ultrafine particle concentration. The concentration of ultrafine particles decayed to half between the 50 m and 150 m locations. It further decreased to 30% at the 400 m location and to 10% at the 1600 m location compared to the concentration measured at the 50 m location in the vicinity of both I-75 and I-71. In contrast, a minimal gradient is observed in the coarse particle concentrations as measured by the OPC. The concentration of coarse particles decreased to only 85% at the 400 m location, and to about 80% at the 1600 m

location compared to one measured at the 50 m location. The dispersion and dilution of particles near an emission source such as a highway is a complex process. The concentration of particles decreases as a function of distance from the highway due to gravitational settling, air dilution and deposition on surfaces due to interception, diffusion and turbulent motion. All these mechanisms are particle size sensitive. The chemical reactions, condensation, and coagulation contribute towards changes in the aerosol particle concentration and size distribution. The lack of gradient for coarse particles in our study could be due to the lower concentration of these particles emitted from traffic sources, and the fact that the coarse particles (such as entrained dust) settle at a much faster rate, within the first 50 m from the highway. Therefore, only CNC measurements were included in the subsequent sampling periods. Fig. 2 compares the results of CNC measurements during two wind directions: perpendicular and parallel to the highway. The average wind speeds during these two measurement periods were 1.3 and 0.6 m s⁻¹, respectively. At 50 m, the absolute concentration of ultrafine particles was 174 000 particles per 1 near I-75 and 71 000 particles per 1 near I-71 when the wind was perpendicular to the highway. The respective concentrations were 28 000 and 27 000 particles per 1 when the wind was parallel to the highway. The concentration at 1600 m varied from 11 000 to 32 000 particles per 1. When the wind was parallel to the highway the concentration of ultrafine particles decreased to 75–85% at the 400 m location and to 65–75% at the 1600 m location compared to the concentration measured at the 50 m location (Fig. 2). A steeper gradient was seen when the wind was perpendicular to the highway since the respective percentages were 30% and 10%, as described above.

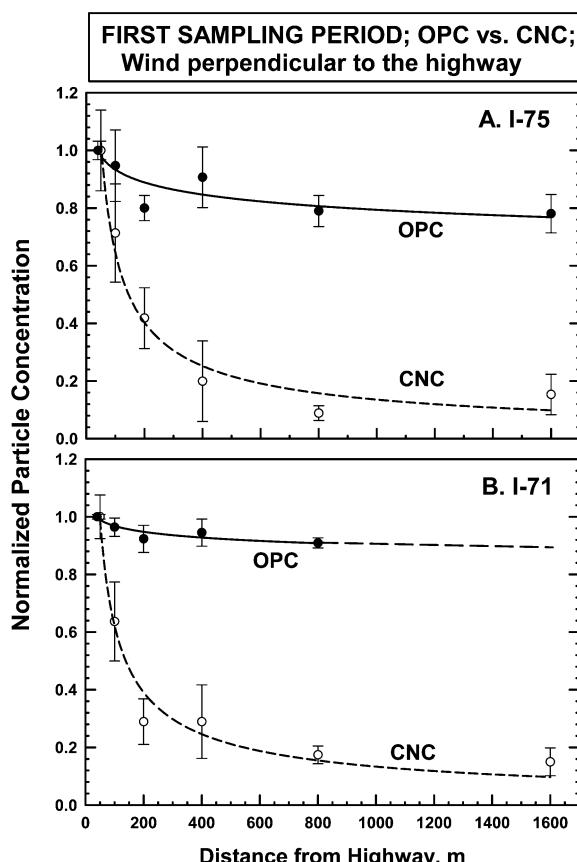


Fig. 1 Normalized particle concentration measured with an optical particle counter (OPC) and a condensation nucleus counter (CNC) at different distances downwind of the highway I-75 and I-71 when the wind was perpendicular to the highway. Each data point represents an average and standard deviation of five 1 min measurements.

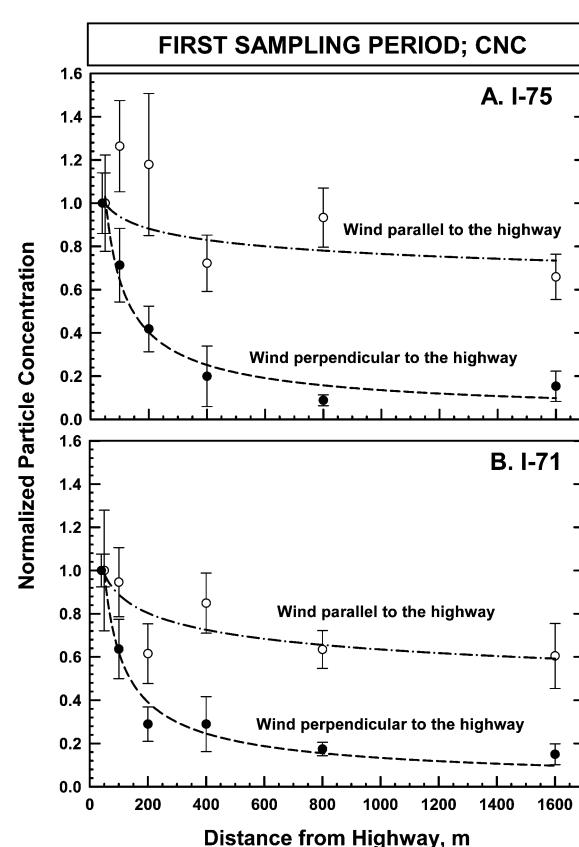


Fig. 2 Normalized particle concentration measured with a condensation nucleus counter (CNC) at different distances of highway I-75 and I-71 (the same sampling points as in Fig. 1) during two wind conditions: when the wind was parallel and when it was perpendicular to the highway. Each data point represents an average and standard deviation of five 1 min measurements.

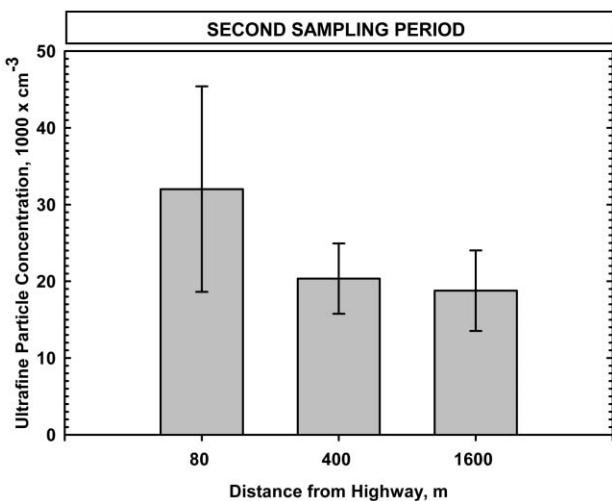


Fig. 3 Ultrafine particle concentrations measured with a condensation nucleus counter at three distances from the highway during the second sampling period. Each bar represents an average of five 1 min measurements performed in the afternoon (5–6 pm) during four consecutive days. Error bars represent standard deviations.

These results show that the ultrafine particles are transported further away from the highway when the wind is perpendicular compared to the situation when it is parallel to the highway. This supports the previous results by Hitchins *et al.*¹² who found similar particle concentration profiles and showed that the effect of the highway can be seen further away if the wind is blowing from the highway towards the sampling sites than if the wind is blowing parallel to the highway.

Ultrafine particle concentrations measured during the second sampling period are presented in Fig. 3. These measurements were performed at three distances from the highway in the afternoons (5–6 pm) of four consecutive days. During the monitoring the winds were blowing from the highway towards the sampling locations 60% of the time and the average wind speed was 1.8 m s^{-1} . As shown in Fig. 3, the sampling location closest to the highway had a higher concentration of ultrafine particles than the two other locations: the concentration at 400 and 1600 m had decreased to about 60% from the concentration measured at 80 m. A similar trend was seen in the ultrafine particle concentrations measured at noon (12–1 pm); an example of which is presented in Fig. 4. These trends support the results presented in Fig. 1. In addition to the real-time particle count measurements, PM2.5 samples were collected to obtain 24 h averaged particulate mass concentrations and the elemental concentrations. Table 1 (second column) presents the PM2.5 mass concentration. There was a slight decrease in the average PM2.5 concentration with increased distance: at 1600 m, the average concentration decreased to about 80% from the level measured at 80 m. However, there was a considerable daily variation in PM2.5 mass as shown by the high standard deviations. Thus, no significant PM2.5 gradient was observed

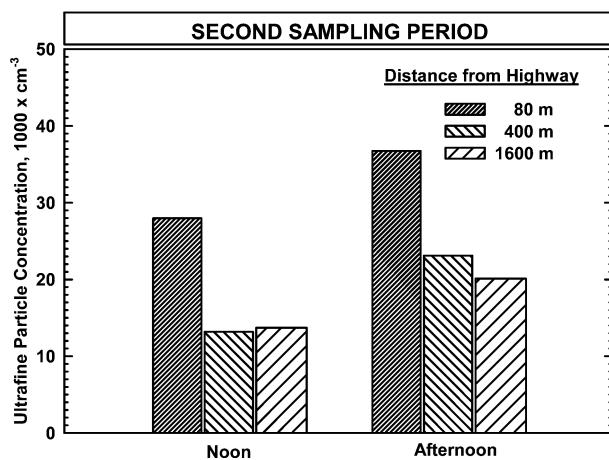


Fig. 4 Ultrafine particle concentrations measured with a condensation nucleus counter at three distances from the highway at noon (12–1 pm) and in the afternoon (5–6 pm) of the same day during the second sampling period. Each bar represents an average of five 1 min measurements.

between the sampling points located at different distances from the highway.

PIXE analysis detected 7 elements in the PM2.5 samples: aluminium, calcium, iron, potassium, silicon, sulfur, and zinc. The sulfur concentrations were examined more closely because this was the predominant element, and it is believed to be reflective of diesel exhausts.¹⁹ The sulfur concentrations measured in this study ranged from 0.07 to $3.77 \mu\text{g m}^{-3}$. Cincinnati is in the Ohio River Valley and is impacted by several coal-fired power plants. Due to this and the topography, the background "sulfate" concentration (measured as sulfur by PIXE and XRF) is high.¹⁴ For example, sulfur concentrations measured in St. Louis, Missouri (about 600 km west of Cincinnati) were typically lower, except for the days when the wind is directed from the Ohio River Valley.²⁰

Table 1 (third column) shows the average sulfur concentrations (analyzed by PIXE method) for the three sampling sites located at different distances from the highway. As seen from Table 1, the sulfur concentrations were higher at the locations 80 m and 400 m from the highway than in the background sampling point (1600 m) during the second sampling period. One of the mechanisms of fine particle formation in diesel engines involves nucleation of sulfuric acid and water, followed by particle growth by condensation of organic species. Fine particles are also known to form from the vapor species in the atmosphere in the vicinity of the engines.²¹ Thus, the observed sulfur gradients could be reflective of diesel engine emissions in areas where traffic is the major source of air pollution.

To investigate how the concentration decreases between the sites located at 400 m and 1600 m, the sampling was performed during a third period including the distance of 800 m as a sampling site between the above-mentioned ones. The third sampling period did not include the site located at 80 m from

Table 1 Average and standard deviation of PM2.5 mass concentration ($\mu\text{g m}^{-3}$) and sulfur mass concentration ($\mu\text{g m}^{-3}$) obtained using PIXE analysis on PM2.5 samples. The samples were collected simultaneously at three distances from the highway during three consecutive 24 h sampling days

Distance from the Highway/m	Second sampling period		Third sampling period	
	PM2.5	Sulfur	PM2.5	Sulfur
80	22 ± 14	0.42 ± 0.28	Not measured	Not measured
400	20 ± 5	0.67 ± 0.37	22 ± 7	2.73 ± 1.48
800	Not measured	Not measured	29 ± 16	1.24 ± 0.74
1600	17 ± 12	0.11 ± 0.06	17 ± 5	1.04 ± 0.83

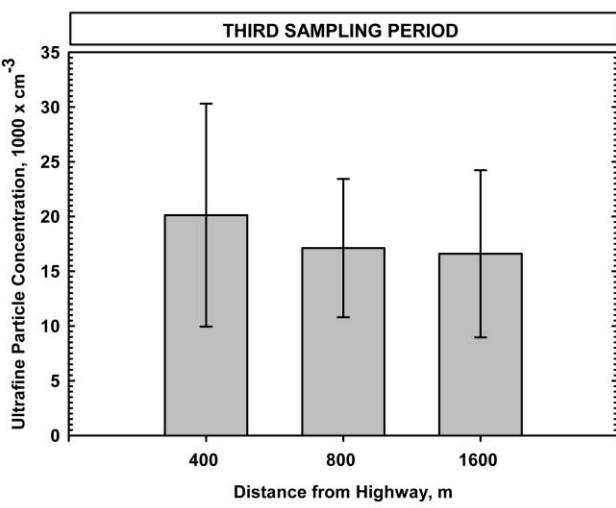


Fig. 5 Ultrafine particle concentration measured with a condensation nucleus counter at three distances from the highway during the third sampling period. Each bar represents an average of five 1 min measurements performed in the afternoon (5–6 pm) during four consecutive days. Error bars represent standard deviations.

the highway; thus, the total number of monitoring sites remained equal to three. During the third sampling period, the wind was blowing from the highway towards the sampling locations 75% of the time and the average wind speed was 2.5 m s^{-1} . The ultrafine particle counts during the third sampling period are presented in Fig. 5. The concentrations at the 800 m and 1600 m locations were about 85% of the concentration measured at the 400 m location. Thus, at these distances, the measurements at the 400 m location showed only a slightly higher concentration than at the two other distances, which again agrees well with the trend presented in Fig. 1. Similarly to the results obtained during the second sampling period, the PM2.5 mass concentrations did not show any clear trend as a function of distance (Table 1, fourth column). The sulfur concentration measured during the third sampling period was again higher at 400 m than at 1600 m, see Table 1, fifth column. Furthermore, the sulfur concentration at 800 m location was closer to the background level than to the value measured at the 400 m location. These findings provide preliminary evidence that in this region, the exposure to traffic pollution can be detected at distances of up to 400 m from the highway if sulfur is used as an indicator.

Selected samples ($n = 9$) were also analyzed by XRF to determine if additional elements could be detected, as the use of multiple indicators is usually needed to confirm the role of a specific source. This confirmation is also important as sulfur can be emitted from other industrial sources (coal combustion) and can be formed by conversion of sulfur dioxide to particulate sulfates in the atmosphere. The XRF analysis allowed identification of several more species in the samples in comparison to the PIXE method (35 species were identified in comparison to 7 by PIXE). Table 2 summarizes the results for

Table 2 Comparison of XRF and PIXE methods for the elemental analysis of PM2.5 samples. The results in each column represent an average and standard deviation of nine measurements

Element	Concentration measured with XRF/ ng m^{-3}	Concentration measured with PIXE/ ng m^{-3}	XRF/PIXE
Al	3.9 ± 4.0	45 ± 82	0.2 ± 0.1
Si	41 ± 28	86 ± 40	0.5 ± 0.2
S	340 ± 320	470 ± 390	0.7 ± 0.2
K	29 ± 21	25 ± 22	1.4 ± 0.9
Fe	70 ± 12	50 ± 14	1.4 ± 0.4

the nine samples that were analyzed with both methods. Average concentrations as ng m^{-3} and the average ratio of the one measured by XRF to the other measured by PIXE (XRF/PIXE) are listed for five elements that were detected with both methods in at least four of the nine samples. Half of the detection limit was assigned for the data points below the detection limit. As seen in Table 2, the XRF/PIXE ratio varied between 0.2 and 1.4. Aluminium, silicon, and sulfur were detected at higher concentrations by PIXE than by XRF, whereas for potassium and iron, the situation was the reverse. The largest discrepancy between XRF and PIXE results was observed with aluminium with the average XRF/PIXE ratio being 0.2. There are several reports in the literature on the inter-comparison of PIXE and XRF. While PIXE is known to be more sensitive for certain heavy/nuclear elements and useful in detection of elements bound to other elements,²² it is generally recognized that XRF is the more sensitive technique.^{23,24}

Several of the elements that were detected by XRF are commonly used for traffic signature in chemical mass-balance models e.g. refs 1, 25, and 26. The elemental concentration of selected species commonly found in road dust (aluminium, silicon, chlorine, and potassium) and diesel exhausts (sulfur, titanium, vanadium, iron, nickel, and lead) are plotted in Fig. 6. The concentrations of all of the elements presented were at the same level or higher near the highway (<400 m) than in the background location (1600 m). These results suggest that the XRF-data can be used for receptor modeling to establish the fraction of diesel exhaust and road dust particles in the ambient PM samples. A larger data set is needed, however, to accurately apply the chemical mass balances model, which would allow estimating the contribution of diesel exhaust and the road dust for the total particle load.

Elemental and organic carbon have been used as markers for traffic pollution in several studies,^{26,27} and were also measured in this study. The results plotted in Fig. 7 indicate a slight decrease in the average elemental carbon concentrations occurring when the distance increased from 400 to 1600 m. The average concentration decreased to 87% at the 800 m location and to 77% at the 1600 m location (both in percentage of the concentration at the 400 m location). There was no difference, however, in the organic carbon concentrations between 400 and 1600 m, which indicates the contribution of local gasoline-powered vehicles. Previous studies by Roorda-Knappe *et al.*⁸ and Janssen *et al.*⁷ have reported differences in

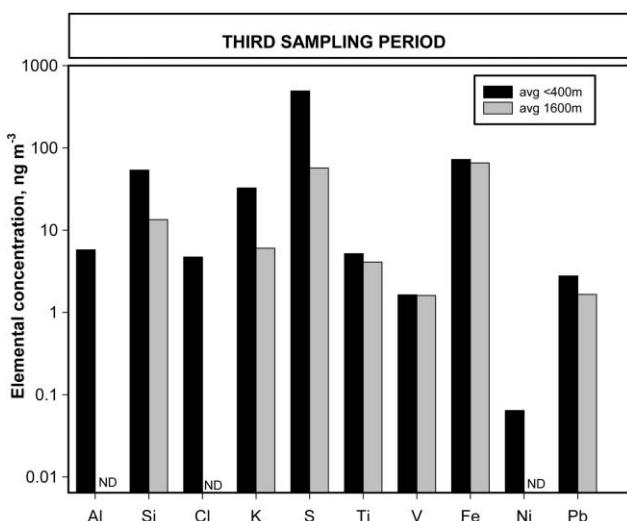


Fig. 6 Concentration of selected key elements typical for road dust and DEP particles. The samples were collected simultaneously at three sampling sites for 24 h using PM2.5 samplers and subsequently analyzed by XRF. The results represent an average of the two sampling sites close to the highway (<400 m) and the sampling site far away from the highway (1600 m). ND = not detected.

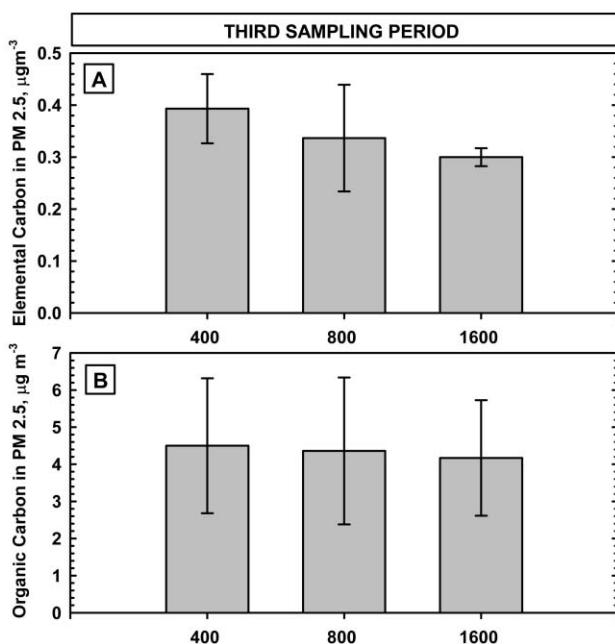


Fig. 7 Results on carbon measurements obtained during the third sampling period: (A) elemental carbon in PM2.5 fraction and (B) organic carbon in PM2.5 fraction. Each bar represents an average of three consecutive 24 h sampling days. Error bars represent standard deviations.

elemental carbon at distances much closer to the highway: 0.5–50 m versus >50 m.

Conclusions

A study was undertaken to determine the concentration gradients in the vicinity of two interstate highways slicing through a major metropolitan residential area. The project was designed in support of a larger epidemiological study on the effect of traffic pollution on the incidence of atopy in newborns followed longitudinally to age four. The results indicate that the aerosol concentration gradient was most clearly seen with respect to the particle number concentration measured in the ultrafine particle size range. The concentration of ultrafine particles decreased to half between the sampling points located at 50 m and 150 m downwind from the highway. Additionally, a gradient was seen in diesel engine emission levels up to about 400 m from the highway based on the gradient in particulate sulfur concentrations. Concentrations of several other key elements indicative of traffic sources (road dust and diesel exhaust) were also found elevated up to 400 m. In this pilot study, traffic related particles could not be distinguished from the background levels beyond 400 m from the highway. This was confirmed by comparing elemental and organic carbon concentration data at these distances. The background concentrations in this area are influenced by long-range aerosol transport and by local traffic. XRF analysis identified more elemental species than the PIXE analysis, including several indicator species for diesel exhaust. Data collection will be continued and a more detailed evaluation by receptor modeling is planned to firmly establish the contribution of diesel exhausts. Due to the complexity of transport and dispersion of traffic related pollutants, other models to accurately predict

spatial variations are needed for better estimates of diesel exposure.

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References

- J. A. Gillies and A. W. Gertler, *J. Air Waste Manage. Assoc.*, 2000, **50**, 1459–1480.
- R. N. Colvile, E. J. Hutchinson, J. S. Mindell and R. F. Warren, *Atmos. Environ.*, 2001, **35**, 1537–1565.
- M. Wijst, P. Reitmeir and S. Dold, *BMJ*, 1993, **307**, 596–600.
- S. K. Weiland, K. A. Mundt, A. Rückmann and U. Keil, *Ann. Epidemiol.*, 1994, **4**, 243–247.
- J. Edwards, S. Walters and R. K. Griffiths, *Arch. Environ. Health*, 1994, **49**, 223–227.
- P. van Vliet, M. Knape, J. de Hartog, N. A. H. Janssen, H. Harssema and B. Brunekreef, *Environ. Res.*, 1997, **74**, 122–132.
- N. A. H. Janssen, D. F. M. Van Mansum, K. Der Jagt, H. Harssema and G. Hoek, *Atmos. Environ.*, 1997, **31**, 1185–1193.
- M. C. Roorda-Knape, N. A. H. Janssen, J. J. De Hartog, H. N. Van Vliet, H. Harssema and B. Brunekreef, *Atmos. Environ.*, 1998, **32**, 1921–1930.
- S. Kingham, D. Briggs, P. Elliot, P. Fisher and E. Lebret, *Atmos. Environ.*, 2000, **34**, 905–916.
- N. A. H. Janssen, P. H. N. van Vliet, F. Aarts, H. Harssema and B. Brunekreef, *Atmos. Environ.*, 2001, **35**, 3875–3884.
- Y. Zhu, W. C. Hinds, S. Kim and C. Sioutas, *J. Air Waste Manage. Assoc.*, 2002, **52**, 1032–1042.
- J. Hitchins, L. Morawska, R. Wolff and D. Gilbert, *Atmos. Environ.*, 2000, **34**, 51–59.
- W. K. Merz and P. Biswas, *Proc. Air Waste Mgmt. Assoc.*, Annual Meeting, Paper 92-104.01, Kansas City, June 1992.
- S. Mukerjee and P. Biswas, *Environ. Technol.*, 1992, **13**, 635–646.
- S. Mukerjee and P. Biswas, *Environ. Manage.*, 1993, **17**, 531–543.
- Ohio Environmental Council (OEC), Ohio Valley Environmental Coalition, Regional Coalition for Ohio Valley Environmental Restoration, Ohio Valley-Ozone Alley, pp. 1–35, February 2000.
- US Environmental Protection Agency, *Quality Assurance Guidance Document 2.1.2. Monitoring PM2.5 in Ambient Air Using Designated Reference or Class I Equivalent Methods. Quality Assurance Handbook, Vol. II*, Part II; US Government Printing Office, Washington, DC, 1998.
- M. E. Birch and R. A. Cary, *Aerosol Sci. Technol.*, 1996, **25**, 221–241.
- D. B. Kittelson and W. Watts, in *NSF-ESF Nanoparticle Symposium Proceedings*, Dublin, Ireland, September 2000, pp. 17–21.
- J. Turner, Washington University, St. Louis, Missouri, Personal Communication, November 2002.
- D. B. Kittelson, *J. Aerosol Sci.*, 1998, **29**, 575–588.
- M. E. Magarino, A. R. De Arellano, M. A. Rodriguez, A. R. Conde and P. J. S. Soto, *Bol. Soc. Esp. Ceram. V.*, 2002, **41**, 531–538.
- S. M. Pyle, J. M. Nocerino, S. N. Deming, J. A. Palasota, J. M. Palasota, E. L. Miller, D. C. Hillman, C. A. Kuharic, W. H. Cole, P. M. Fitzpatrick, M. A. Watson and K. D. Nichols, *Environ. Sci. Technol.*, 1996, **30**, 204–213.
- W. M. Kwiatek, B. Kubica, C. Paluszakiewicz and M. Galka, *J. Alloys Compd.*, 2001, **328**, 283–288.
- J. G. Watson, J. C. Chow, Z. Lu, E. M. Fujita, D. H. Lowenthal and D. R. Lawson, *Aerosol Sci. Technol.*, 1994, **21**, 1–36.
- J. J. Schauer and G. R. Gass, *Environ. Sci. Technol.*, 2000, **34**, 1821–1832.
- L. S. Hughes, J. O. Allen and M. J. Kleeman, *Environ. Sci. Technol.*, 1999, **33**, 3506–3515.