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Traffic Particles in Ambient Air of a Major US Urban Area: Has Anything Changed over a Decade?

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ABSTRACT

Few studies have addressed the potential changes in particulate matter (PM) exposure occurring in major metropolitan areas over substantial time periods, e.g., 5 or 10 years. The present study examined changes in the $PM_{2.5}$ concentration and elemental composition between two monitoring campaigns carried out in 2002–2005 and 2010–2011 within the Greater Cincinnati area (USA). This area is recognized for high volume of diesel truck traffic (about 10 million trucks annually on regional freeways). The 24-hour filter samples were collected at four sites. General linear models were used to examine differences between the two data sets for elemental carbon (EC), organic carbon (OC), and EC/OC. The comparison was extended to the concentrations of $PM_{2.5}$ and its relevant elemental constituents. At one site, which was previously identified as a particularly hot spot for traffic/diesel air pollution, the concentrations of most traffic related elements as well as EC and EC/OC ratio significantly decreased ($p < 0.05$) between the two campaigns. No significant differences between carbon data generated in the two campaigns were observed at the other three monitoring stations. These findings did not depend on whether the comparison model accounted for wind speed and direction. The EC/OC determined in the recent campaign across all sites showed no significant differences between the Summer and Fall data but Winter values were significantly lower. The site with the highest traffic influence revealed no significant seasonal difference in $PM_{2.5}$ but essentially all relevant elements showed significant seasonal variations between the Fall (higher) and Winter (lower). The findings suggest that air quality and engine exhaust control policies implemented between 2005 and 2010 have not produced significant changes in metropolitan traffic air pollution levels. However, the decreasing trends in $PM_{2.5}$, Ti, V, Mn, Fe, Zn, Br, and Pb, EC, OC, and EC/OC may become sustainable over a longer time.

Keywords: $PM_{2.5}$; Carbon; Traffic; Spatial and seasonal variability.

INTRODUCTION

Traffic in urban areas significantly contributes to the ambient air pollution level, which has been linked to adverse health effects among children and adults. Several large population-based studies have been initiated over the past 10–15 years in the USA and worldwide to establish associations between the traffic (and particularly diesel exhaust) aerosol exposure and respiratory health effects (Jerrett *et al*., 2008; Barraza-Villarreal *et al*., 2008; Gehring *et al*., 2010; Carlsten *et al*., 2011; Ristovski *et al*., 2011). Among them, there is the Cincinnati Childhood Allergy and Air Pollution Study (CCAAPS) – a prospective birth cohort investigation (2001–2012) designed to determine if exposure to high levels of diesel exhaust particles (DEP) during early childhood increases the risk for developing allergic diseases. Resulting from the CCAAPS, wheezing, night cough and allergic rhinitis in infants residing in proximity to major roads and stop-and-go traffic was linked to exposure to elemental carbon attributable to traffic, which served as a marker for diesel exhaust particles (DEP) (Ryan *et al*., 2005, 2007, 2009; Sucharew *et al*., 2010). Similar to other studies, CCAAPS relied on the aerosol exposure data measured in the tested metropolitan area over a specific period of time (beginning with the pilot study in 2001 and extending into a full-scale sampling in 2002–2005).

The follow-up of the CCAAPS birth cohort is continuing to the present. Therefore, it is important to determine whether the initially collected exposure data are useful beyond the air monitoring period and are still relevant and representative. The above question is not trivial since various environmentally friendly policies have been recently implemented not only in Cincinnati but many urban areas for heavy-duty vehicle engine modification such as retrofitting

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and diesel fuel (composition changes in diesel fuel) that presumably reduce PM emissions. Additionally, efforts have been and are being made to reduce the number of traffic congestion zones. Interventions such as the nationwide "cash for clunkers" program helped remove millions of heavily emitting vehicles from the US roads. All these measures suggest that the concentration of traffic aerosols is decreasing. On the other hand, widening highways by adding lanes allows for higher traffic volume, and more heavy-duty vehicles are currently on the US roads compared to a decade ago (US Department of Transportation, 2013). All this may increase the traffic aerosol emission in major urban areas. Air pollution from railways and ports also contribute.

Thus, it is important to assess potential changes in exposure to air pollutants over time. Additionally, to develop adequate and cost-effective monitoring strategies for particulate matter (PM), it is useful to determine the representativeness of a short-term (e.g., single-season) sampling, which, in turns, requires characterizing the seasonal variation of health-relevant aerosol concentrations (e.g., $PM_{2.5}$) and elemental composition.

While meteorological data (e.g., daily wind speed) are often recorded during sampling, there have been few studies incorporating this information when analyzing aerosol concentration and composition data obtained from 24-hour ambient air samples. This issue is important for those living up or down wind from the source. Also, few studies simultaneously examine spatial and seasonal variability of traffic-related aerosol concentrations. The present investigation addresses this gap.

$PM_{2.5}$ and its elemental composition, including organic carbon (OC) and elemental carbon (EC), have been used to assess a broad spectrum of air pollutants, including those originated by traffic. While OC is produced by all combustion sources, EC is primarily generated by traffic sources, largely by diesel-powered vehicles (Birch and Cary, 1996). As such, EC is frequently used as a surrogate for traffic generated aerosols (Holguin, 2008; Ryan *et al*., 2009; Hochstetler *et al*., 2011). The EC concentration and its contribution to $PM_{2.5}$ are greater in areas near highways and both increase with increased truck traffic (Kinney *et al*., 2000; Lena *et al*., 2002; Martuzevicius *et al*., 2004). The ratio of elemental carbon to organic carbon (EC/OC) provides an estimate of the overall percentage of the total carbon (EC + OC) attributable to combustion of diesel. Where traffic exhaust is the primary source of the ambient aerosol diesel, this ratio represents the fraction of the total carbon attributable to diesel-powered vehicles (Maykut *et al*., 2003).

The main objective of this investigation is to examine changes in the $PM_{2.5}$ concentration and elemental composition between two monitoring campaigns that were carried out over the past decade within the Cincinnati metropolitan area, which has high volume North-South diesel truck traffic. The first campaign was conducted between 2002 and 2005 (when the primary portion of the CCAAPS air monitoring was done); the second campaign was carried out in 2010–2011. The data obtained in the second campaign were also analyzed for spatial and seasonal variability. Additionally, the role of wind in the comparative analysis was examined. The $PM_{2.5}$ samples were characterized with respect to elemental carbon (EC), organic carbon (OC), and other relevant elements.

METHODS

Sampling Sites

The four study sampling sites were selected to represent the metropolitan area with differing levels of traffic-related pollutions. The selection was based on our previous ambient aerosol measurements (Martuzevicius *et al*., 2004, 2005), modeling (Hu *et al*., 2006) and DEP exposure assessment (Ryan *et al*., 2007). The characteristics of these sites are presented in Table 1. The first site, referred to as DOWNTOWN, is located in a residential area of downtown Cincinnati and was previously identified to have high levels of EC attributable to traffic and EC attributable to diesel (Martuzevicius *et al*., 2004; Hu *et al*., 2006; Sahu *et al*., 2011). The second and third sites, GROOMS and NEWPORT, are respectively located on a road in a northern suburb and in Newport city, Kentucky, south of Ohio River. These two sites represent areas with mid-level traffic pollution; while close to major highways, these two sites are located upwind from the traffic and the corresponding highway stretches are characterized by a moderate traffic density with natural mitigating factors such as trees and terrain/bridge. The forth site, MONT, is located in a low-polluted north-eastern suburb of Cincinnati (Montgomery), far from highways.

Sampling Campaigns and Seasons

The above four sites were used in both campaigns of ambient air monitoring supported by the CCAAPS. Due to the CCAAPS original design, the earlier campaign (2002–2005) featured different number of samples (n) collected at each site and subjected to different types of analysis; no attempt was made to stratify by season or account for daily wind variables. A total of n = 38 carbon samples were

Table 1. Characteristics of sampling sites.

Site	Distance to nearest highway (m)	Upwind or downwind*	Level of traffic air pollution**
DOWNTOWN	180	downwind	high
GROOMS	270	upwind	medium
NEWPORT	75	upwind	medium
MONT	> 2,000	downwind	low

* From a nearest highway relative to the primary wind direction.

** Based on Martuzevicius *et al*. (2004, 2005), Hu *et al*. (2006) and Ryan *et al*. (2007).

originally collected at DOWNTOWN, n = 15 at GROOMS, n = 4 at NEWPORT, and n=16 at MONT. The corresponding numbers of samples collected for $PM_{2.5}$ and elemental analysis were n = 96, 71, 22, and 59. In the 2010–2011 campaign, we established a protocol to collect twelve daily carbon samples per site during three seasons – Summer, Fall, and Winter. Since Spring and Fall are similar in terms of the air temperature, humidity range and wind characteristics no data were collected in Spring. On each of the twelve sampling days per season, the carbon samples were collected at the four sites simultaneously. This generated a total of 36 carbon samples per site in the 2010–2011 campaign. Additionally, $PM_{2.5}$ samples collected in downtown on each monitoring day (in parallel to the carbon samples) were subjected to elemental analysis. Due to quality assurance considerations 37 samples were collected at the DOWNTOWN where the highest level of traffic pollution was determined. Additionally, two blank samples were collected for every set of measurements in each of the two campaigns.

Collection and Analytical Procedures for Air Samples

For both campaigns, the 24-hour air samples were collected on weekdays starting at 9 AM. The sampling occurred when precipitation was below ½ inch and was not conducted when non-ordinary particle emitting activities (e.g., major road construction) took place. At each site, a Harvard-type $PM_{2.5}$ impactor (MS&T Area Sampler, Air Diagnostics and Engineering, Inc., Harrison, ME, USA) with a 2.5 mm cut-off size nozzles operated at a flow rate of 20 L/min. The pumps were calibrated with a flowmeter (DryCal DC-Lite, BIOS International Corporation, Butler, NJ, USA) before each 24-h measurement period and the flow rate was checked after each measurement period. The impactor was mounted on a tripod under rain-protective covers. A meteorological station (Vantage Pro, Davis, CA, USA) was mounted on the same tripod. Each impactor was equipped with a 37-mm quartz filter (Whatman Inc., Clifton, NJ, USA). The polyester ''Drain disk'' pads (Whatman Inc., Clifton, NJ, USA) were deployed with the filters to maintain integrity. Quartz filters were pre-baked at 550°C for at least 24 h before sampling (Schauer and Cass, 2000; Bae *et al*., 2004). Quartz filters were analyzed for carbon (EC and OC) using the Thermal-Optical Transmittance (TOT) technique (NIOSH-5040 method, Sunset Laboratory Inc., Hillsborough, NC, USA). This enabled the quantification of ambient mass concentration of EC and OC for each sampling day and determined a ratio of EC/OC as a surrogate of the aerosol diesel emission.

The additional Harvard-type $PM_{2.5}$ impactor deployed at the DOWNTOWN location was equipped with a 37-mm Teflon membrane filter (pore size = 1 μm) and a support ring (Pall Corporation, Ann Arbor, MI, USA) to generate the mass concentration data on $PM_{2.5}$ and its constituents. Before weighing, Teflon filters were conditioned for at least 24 h in a humidity chamber at a relative humidity of 30–40% and a temperature of 22–24°C (McDonald, 2003). Once the $PM_{2.5}$ mass concentration was quantified gravimetrically, Teflon filters were sent for X-Ray Fluorescence (XRF) analysis (Chester Labnet, Tigard, OR, USA) for a total of 38 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) that included traffic emission related elements as well as those representative of the regional ambient aerosol background. The ambient mass concentrations of these elements were determined and used for comparison between the sampling campaigns.

Wind Index

Wind speed and direction were recorded for each day. Daily averages of the available hourly values were determined for each 24-hour sample collection period. A wind index was calculated for each site as follows:

$$\text{Wind Index} = \frac{1-\cos(\theta - x)}{2} \quad (1)$$

where θ = the Euclidian angle of the site to the nearest highway and x = Euclidian angle of the wind vector (Fig. 1) (Ryan *et al*., 2008; Sparks *et al*., 2013). The wind index is a rescaling of the angle between two vectors: from the nearest major traffic source to the site and predominant wind direction to a scale of zero to one. This index is a continuous variable. Sites directly upwind of the traffic source have a wind index equal to zero, sites directly downwind have a wind index of 1, and those perpendicular have an index of 0.5. The data analyses were performed with and without incorporating the wind index into the model.

Statistical Analysis

The objectives of this analysis were to (1) compare the geometric mean values of EC, OC and EC/OC measured during the first and second campaigns at each site, (2) examine the variability of EC, OC and EC/OC across sampling sites and seasons within the second campaign, and (3) examine the seasonal variability of $PM_{2.5}$ and its elemental constituents at one site during the second campaign. Given the unbalanced data, a general linear model (GLM) was used for each analysis. In order to address the first objective (a comparison of the first and second campaign at the four sites) a GLM model was used to compare the sampling period (historic or present) by site (DOWNTOWN, GROOMS, MONT, or NEWPORT). A second GLM model

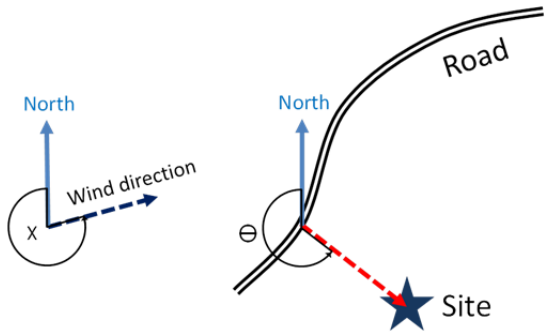

Fig. 1. Wind direction relative to the highway configuration and site location.

was used to compare 2010–2011 concentrations of EC, OC, and EC/OC across sampling sites and seasons. Finally, a third GLM model compared XRF elemental concentrations values at the single DOWNTOWN site across the three seasons. All analyses were performed with and without including the wind index in the model. A Bonferroni correction for multiple comparisons was utilized in cases when conducting pairwise *post hoc* comparisons (e.g., testing differences across sites and seasons). No correction was made for multiple comparisons when testing *a priori* hypotheses (e.g., comparing 2001–2005 to 2010–2011 sampling campaigns).

RESULTS AND DISCUSSION

EC, OC and EC/OC Obtained at Both Campaigns for Four Sites

Fig. 2(a) presents the EC data obtained at the four test sites during the two campaigns. The geometric means of EC obtained in the earlier campaign varied from 0.40 μg/m^3 at MONT to 1.12 μg/m^3 at DOWNTOWN while those measured in the 2010–2011 campaign ranged from 0.29 μg/m^3 at MONT to 0.82 μg/m^3 at DOWNTOWN. Although the EC-values showed a decline from the first to the second campaign at all sites, this decrease was statistically significant only in DOWNTOWN ($p = 0.02$ without accounting for wind and $p < 0.01$ with the wind data incorporated). Both the wind speed and direction were found to be significant factors in the model.

Fig. 2(b) shows the OC data presented in the similar fashion. The OC level dropped between the two campaigns (except GROOMS that showed a slight, but not statistically significant increase). The decrease was significant only at one site, MONT. We could not clearly identify the reason why a more sizable OC reduction occurred at that site. Similar to the EC comparison, the significance level of differences in OC was the same regardless whether or not wind was in the model.

Fig. 2(c) presents the integrated EC/OC data, which are particularly important for tracing a contribution of the heavy-duty vehicle diesel emission to the $PM_{2.5}$ aerosol fraction. The EC/OC geometric mean values obtained in the 2002–2005 campaign varied from 0.11 at MONT to 0.26 at DOWNTOWN while those measured in the 2010–2011 campaign ranged from 0.10 at MONT to 0.19 at DOWNTOWN. Strong statistically significant difference in EC/OC between the two campaigns was observed in the downtown area ($p = 0.001$ without accounting for wind and $p < 0.001$ with the wind data incorporated). Additionally, the GROOMS site also showed a significant decreasing trend ($p = 0.035$ with and without accounting for wind). The historical and recent EC/OC levels at the other two sites were not statistically different from one another.

In conclusion, the only site, which showed consistent and statistically significant decrease of both diesel/traffic markers, EC and EC/OC was the DOWNTOWN site. The GROOMS exhibited no significant change in EC; the EC/OC change at that site was primarily driven by an unexpected increase in OC, which we were unable to interpret. Thus, overall,

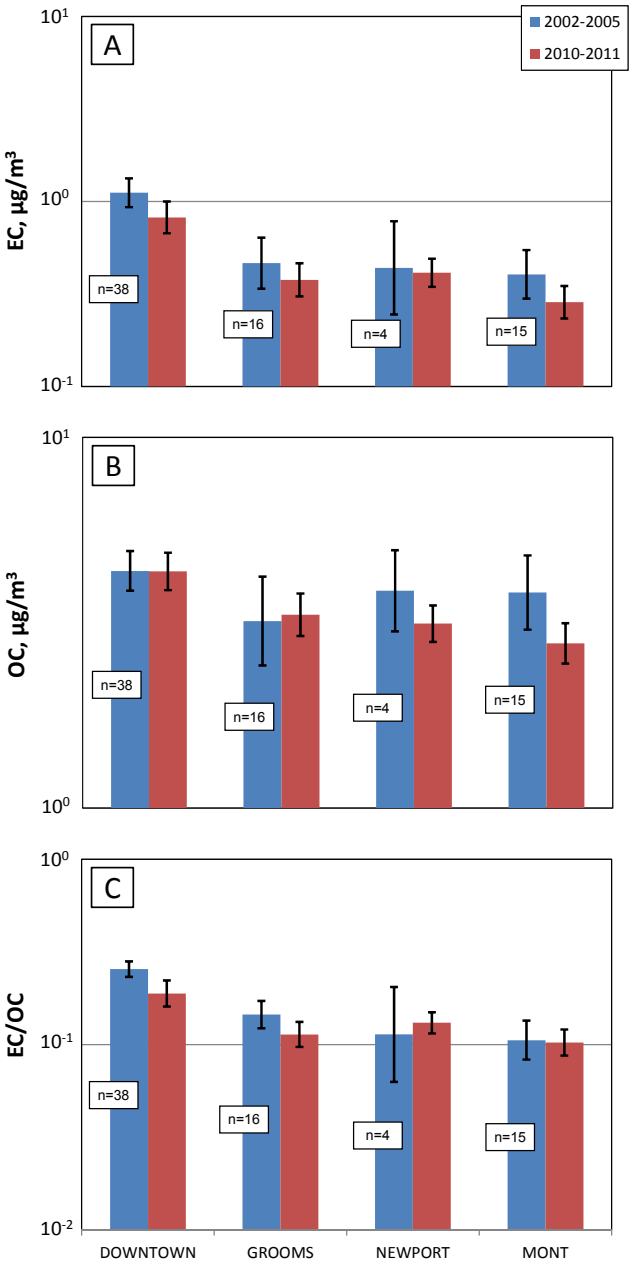

Fig. 2. The values of EC (A), OC (B) and EC/OC (C) obtained at four test sites during two monitoring campaigns. The number of measurements, n, is specified for each bar corresponding to the 2002–2005 campaign; in the 2010–2011 campaign, the same number of measurements (n = 36) was performed at each site. Each bar represents a geometric mean with error bars representing a 95% CI.

while the traffic/diesel relevant factors such as EC and EC/OC showed some decreasing trend, this decrease was not significant across the entire Cincinnati area.

$PM_{2.5}$ and Elemental Concentrations Obtained in the Two DOWNTOWN Campaigns

Fig. 3 shows the mass concentrations of $PM_{2.5}$ and ten elements determined in the two campaigns in the DOWNTOWN sampling station. This comparison is made

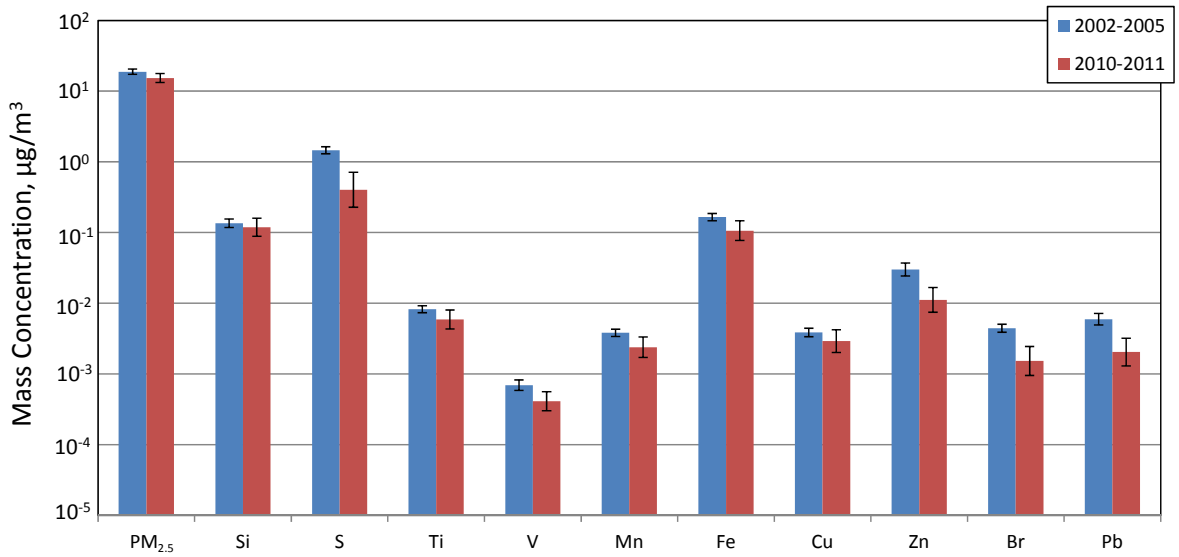

Fig. 3. The concentrations of $PM_{2.5}$ and ten elements obtained in the DOWNTOWN sampling station during the two monitoring campaigns. Each bar represents a geometric mean with error bars representing a 95% CI.

specifically for the site in which EC/OC exhibited the greatest change from 2002–2005 to 2010–2011. The ten elements were chosen to be: (a) representative of either diesel exhaust emission or the regional ambient aerosol background and (b) above the limit of detection established in the XRF analysis. The geometric mean of the $PM_{2.5}$ concentration for the first and second campaigns was approximately 18.8 µg/m^3 and 15.4 µg/m^3, respectively. These levels were consistent with the $PM_{2.5}$ data reported in the literature for metropolitan areas with similar aerosol pollution sources and traffic intensity (Martuzevicius *et al*., 2004; Health Effects Institute, 2013). In both campaigns, sulfur demonstrated the highest concentration among all elements analyzed by XRF. This element has been recognized as a substantial contributor to the regional background aerosol in the Greater Cincinnati area as it is likely related to 14 coal power plants located along the Ohio River valley. Sulfur may also be associated with diesel sources. Crustal elements, including Si, were also essential contributors to $PM_{2.5}$. A contribution of traffic-related trace elements (especially Fe and Zn) to the $PM_{2.5}$ fraction measured in downtown was noticeable, especially in the earlier campaign. This is consistent with relatively high contribution of EC to $PM_{2.5}$ (approximately 5–6% in both campaigns) recorded at this site.

The data obtained in the two campaigns at DOWNTOWN were compared using GLM, based on the total of 37 data points from the recent campaign versus 96 from the earlier time period. For the $PM_{2.5}$ fraction and each of the analyzed elements, the recent geometric mean values were lower than "historic" ones. This difference was statistically significant for $PM_{2.5}$, S, Ti, V, Mn, Fe, Zn, Br, and Pb (all p-values < 0.05); only Si and Cu were not significantly different between the two campaigns. Thus, most of traffic/diesel relevant elements recorded at the downtown location showed statistically significant difference between the two campaigns regardless whether the wind speed and direction were included.

Spatial and Seasonal Variations Obtained in the 2010–2011 Campaign

Fig. 4 presents the seasonal mean values of the mass concentrations of EC, OC and EC/OC measured in three seasons at the four tested sites during the recent campaign. Spatial and temporal variability was examined, with and without accounting for wind data.

Elemental Carbon (EC)

Individual comparisons across sampling sites revealed significant ($p < 0.01$) between-site differences in overall average EC levels with the exception of GROOMS vs. NEWPORT ($p = 0.07$). Further analyses examined spatial variability across sampling sites within sampling season. Similar between-site differences were obtained for Summer. In Fall, significantly higher EC concentrations were observed in downtown as compared to the other three sites. In Winter, we found no significant difference between NEWPORT and GROOMS and between GROOMS and MONT; the downtown area had significantly higher levels of EC than all other sites. The above findings on spatial variability are consistent with the EC sources in the area and with the previously measured EC levels at those sites (Martuzevicius *et al*., 2004). Seasonal EC levels across all sites were not significantly different between Summer and Fall; however, the Winter levels were significantly lower. Generally, similar results were obtained when comparing seasonal EC levels within each site. Incorporating wind data in the statistical analysis did not substantially change the results. Some ambient air monitoring studies performed in other metropolitan areas also revealed lower EC levels in Winter (e.g., Peltier *et al*., 2011).

Organic Carbon (OC)

OC showed a consistent pattern within sites and across sites with DOWNTOWN the highest and MONT the lowest (although the difference between MONT and NEWPORT

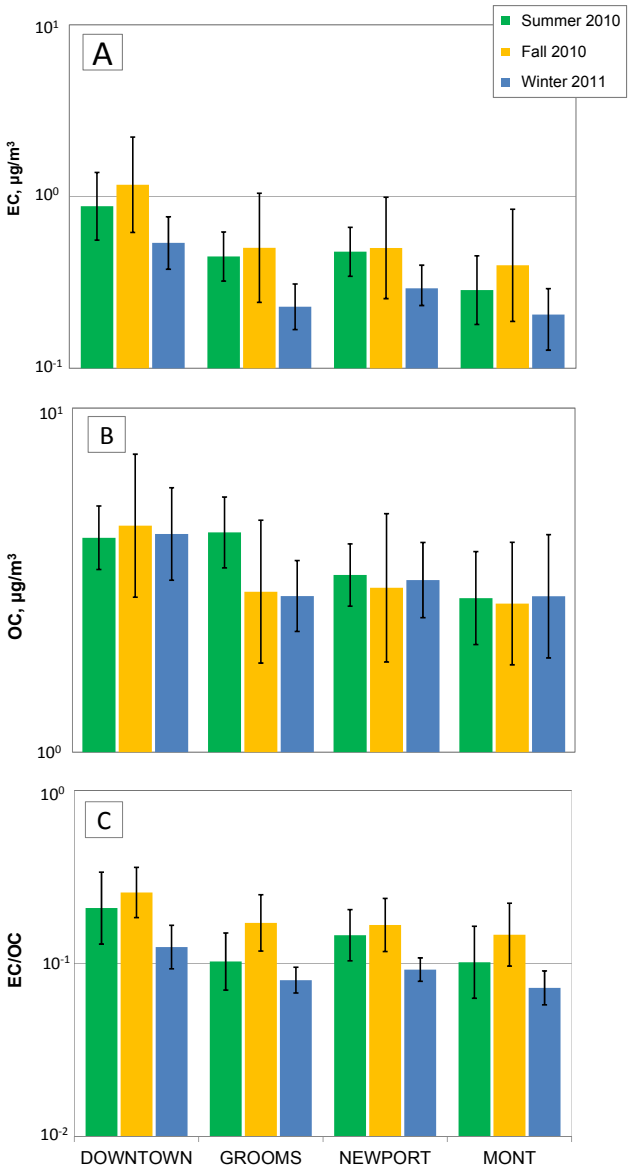

Fig. 4. The seasonal values of EC (A), OC (B) and EC/OC (C) obtained at the four tested sites in Summer, Fall and Winter of the 2010–2011 monitoring campaign. Each bar represents a geometric mean with error bars representing a 95% CI.

was not significant). In Summer, OC measured at the DOWNTOWN site was significantly ($p < 0.001$) higher than at NEWPORT and MONT. In Fall and Winter, the downtown OC level was significantly greater than at all the other sites, while no significant differences were observed among the latter. The overall seasonal average OC concentrations across all sites were not significantly different, which was consistent with the seasonal variability of OC within each site, except for GROOMS that revealed the Summer level significantly ($p < 0.001$) higher than either the Fall or Winter ones. We have not identified any OC-emitting activities in the GROOMS site during the Summer monitoring days to which the above difference could be attributed. Accounting for wind variables did not produce any changes in the conclusions drawn for OC. This is consistent with the fact that OC is a background component of the regional aerosol.

EC-to-OC Ratio

Analysis of the spatial variability of the season-averaged EC/OC values without adjusting for wind showed significant differences except between GROOMS and MONT. The DOWNTOWN site was found to exhibit significantly ($p < 0.001$) higher EC/OC than any other site, while the NEWPORT site had higher EC/OC than either GROOMS or MONT (with a weaker significance level though). These findings are consistent with the values of EC attributable to traffic and EC attributable to diesel that were previously measured in these locations (Martuzevicius *et al*., 2004; Ryan *et al*. 2007, Sahu *et al*., 2011). The EC/OC spatial variability analysis within each season revealed essentially the same patterns as the across-season analysis with the downtown area exhibiting significantly higher EC/OC than any other of the tested. The seasonal variability of EC/OC averaged across all sites showed no significant differences between the Summer and Fall data. The Winter EC/OC, however, were significantly ($p < 0.001$) lower than either Summer or Fall levels. The same pattern was observed within each site. After incorporating the wind data in the analysis, the findings were generally similar to those observed without adjusting for wind.

$PM_{2.5}$ and Elemental Concentrations

Fig. 5 presents the seasonally-integrated values of the mass concentrations of $PM_{2.5}$ and ten elements determined in the recent campaign in the downtown location. The $PM_{2.5}$ concentrations showed no significant difference due to season regardless whether adjusted for wind direction. Almost all elements exhibited a significant difference between the Fall (higher) and Winter (lower) concentration levels. The exceptions are V (when modeling with no wind accounted for) and Ti and Fe (when accounting for wind data). The differences between the traffic-related $PM_{2.5}$ constituents measured in Summer versus Winter were mostly insignificant regardless whether the wind data were incorporated. The findings are generally consistent with the seasonal analysis of EC/OC.

CONCLUSIONS

We observed a slight decrease in EC, a surrogate of traffic PM, over the previous decade at all four tested sampling sites; however, the decrease was significant only at the DOWNTOWN site nearest to the traffic sources, which has earlier been identified as a “hot” spot for traffic pollution. The same site also showed a significant decrease of EC/OC, a surrogate of diesel exhaust, as well as several traffic/diesel-relevant elements. Additionally, we observed some significant seasonal variability in traffic-related pollutants with concentrations generally highest in the Fall and lowest in the Winter months. At the same time, the differences between Fall and Summer and between Fall and Winter were not statistically significant for most of the elements.

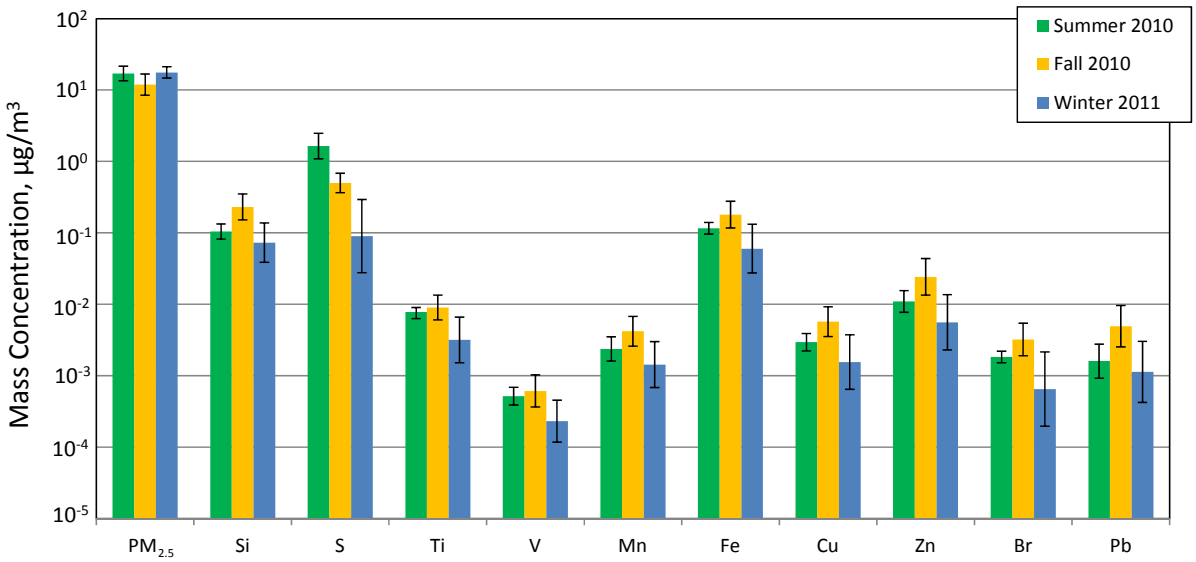

Fig. 5. The concentrations of $PM_{2.5}$ and ten elements obtained in the DOWNTOWN sampling station in Summer, Fall and Winter of the 2010–2011 monitoring campaign. Each bar represents a geometric mean with error bars representing a 95% CI.

Changes in diesel fuel composition and vehicle emissions, which occurred in the USA during the first decade of this century due to implementation of environmental protection policies, may have resulted in improved air quality near major roadways. However, the data collected in this study showed no evidence of a significant decrease of the traffic/diesel air pollution levels consistently across the Cincinnati area. The ambient air monitoring should be continued in the area, perhaps involving a greater number of sites with various traffic characteristics in order to quantify the traffic exposure evolution and determine if the decreases in exposures may become sustainable over a longer time.

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