



1 **Abstract**

2           The Detroit Exposure and Aerosol Research Study (DEARS) provided data to compare outdoor  
3 residential coarse particulate matter (PM<sub>10-2.5</sub>) concentrations in six different areas of Detroit with data  
4 from a central monitoring site. Daily and seasonal influences on the spatial distribution of PM<sub>10-2.5</sub> during  
5 Summer 2006 and Winter 2007 were investigated using data collected with the newly developed coarse  
6 particle exposure monitor (CPEM). These data allowed the representativeness of the community  
7 monitoring site to be assessed for the greater Detroit metro area. Multiple CPEMs collocated with a  
8 dichotomous sampler determined the precision and accuracy of the CPEM PM<sub>10-2.5</sub> and PM<sub>2.5</sub> data.

9           CPEM PM<sub>2.5</sub> concentrations agreed well with the dichotomous sampler data. The slope was 0.97  
10 and the R<sup>2</sup> was 0.91. CPEM concentrations had an average 23% negative bias and R<sup>2</sup> of 0.81. The  
11 directional nature of the CPEM sampling efficiency due to bluff body effects probably caused the negative  
12 CPEM concentration bias.

13           PM<sub>10-2.5</sub> was observed to vary spatially and temporally across Detroit, reflecting the seasonal  
14 impact of local sources. Summer PM<sub>10-2.5</sub> was 5 µg/m<sup>3</sup> higher in the two industrial areas near downtown  
15 than the average concentrations in other areas of Detroit. An area impacted by vehicular traffic had  
16 concentrations 8 µg/m<sup>3</sup> higher than the average concentrations in other parts of Detroit in the winter due  
17 to the suspected suspension of road salt. PM<sub>10-2.5</sub> Pearson Correlation Coefficients between monitoring  
18 locations varied from 0.03 to 0.76. All summer PM<sub>10-2.5</sub> correlations were greater than 0.28 and  
19 statistically significant (p-value < 0.05). Winter PM<sub>10-2.5</sub> correlations greater than 0.33 were statistically  
20 significant (p-value < 0.05). The PM<sub>10-2.5</sub> correlations found to be insignificant were associated with the  
21 area impacted by mobile sources during the winter. The suspected suspension of road salt from the  
22 Southfield Freeway, combined with a very stable atmosphere, caused concentrations to be greater in this  
23 area compared to other areas of Detroit. These findings indicated that PM<sub>10-2.5</sub>, although correlated in  
24 some instances, varies sufficiently across a complex urban airshed that that a central monitoring site may  
25 not adequately represent the population's exposure to PM<sub>10-2.5</sub>.

26

1 **Introduction**

2 Epidemiological evidence indicates fine (PM<sub>2.5</sub>) and coarse (PM<sub>10-2.5</sub>) particulate matter have an  
3 acute effect on morbidity and mortality (Dockery et al., 1993; Brunekreef and Forsberg, 2005). These  
4 epidemiology studies frequently use data from one or a few central monitoring stations as a proxy for  
5 personal exposure. Ito et al. (2001) noted central monitoring sites are frequently sited for convenience,  
6 and not specifically to support epidemiology studies. Therefore, the particulate matter concentration data  
7 may not be representative of the exposure of the general population. Williams et al. (2008a) found  
8 personal PM<sub>10-2.5</sub> were not correlated with concentrations measured at a centrally located outdoor site.  
9 Wilson et al. (2005) concluded the degree of intra-urban spatial heterogeneity in particulate matter  
10 concentrations should be determined before using central monitoring site data as an estimate of personal  
11 exposures. This spatial analysis will minimize exposure misclassification and reduce uncertainty in  
12 relative risk estimates in longitudinal cohort studies.

13 Recent personal exposure studies have focused on the effects of PM<sub>10-2.5</sub> because of persistent  
14 concerns about the toxicity of this size fraction combined with a paucity of exposure data compared with  
15 the PM<sub>2.5</sub> fraction. Yeatts et al. (2007) showed that PM<sub>10-2.5</sub> increments of as little as 1 µg/m<sup>3</sup> can alter  
16 heart rate variability in adults with asthma. This finding suggests that PM<sub>10-2.5</sub> spatial and temporal  
17 variability need to be fully understood if concentration data from a central monitoring site are to be used to  
18 represent the exposure of the general population.

19 Source apportionment methodologies also are being used to link ambient PM<sub>2.5</sub> concentrations to  
20 personal exposure to specific sources (Hopke et al., 2001) and specific species (Landis et al., 2001;Zhao  
21 et al., 2006). Again, these techniques rely upon data from one or two central monitoring sites to provide  
22 the requisite speciation data. Extension of the methods to PM<sub>10-2.5</sub> exposure also will require a better  
23 understanding of the concentration spatial and temporal variability.

24 Spatial and temporal variability of particulate matter concentrations have been previously  
25 investigated to understand the influence of meteorology and geographical location of the sample  
26 collection instruments with respect to local sources. These studies focused on PM<sub>2.5</sub> and PM<sub>10</sub>, where  
27 PM<sub>10-2.5</sub> were calculated by difference. Burton et al. (1996) found population density to be a surrogate for  
28 local sources. They also found that PM<sub>10-2.5</sub> spatial variability was determined by wind direction, but not

1 wind speed. Guerra et al. (2006) found a similar influence of wind direction on  $PM_{10}$  and  $PM_{2.5}$   
2 concentrations in southeast Kansas. Suh et al. (1997) found one site in Washington D.C. with elevated  
3  $PM_{10-2.5}$  compared to the other sites. However, Wilson and Suh (1997) concluded that their analysis was  
4 limited by the low measurement precision resulting from calculating low coarse particle concentrations by  
5 the difference between  $PM_{10}$  and  $PM_{2.5}$  concentrations. Chen et al. (2007) used low volume  $PM_{10}$  and  
6  $PM_{2.5}$  samplers to assess  $PM_{10-2.5}$  spatial variability and concluded that local sources affected the strength  
7 of the correlation with a centrally located monitor.

8 This study used a new coarse particulate matter sampler (CPEM) to provide direct measurement  
9 of  $PM_{10-2.5}$  and  $PM_{2.5}$  concentrations. The CPEM sample flow and size were designed for personal  
10 exposure assessment, but the instrument was also designed to be sufficiently rugged for monitoring at  
11 outdoor and indoor locations. Data collected with the CPEM characterized the  $PM_{10-2.5}$  spatial and  
12 temporal variability across the Detroit metropolitan airshed during the Detroit Exposure and Aerosol  
13 Research Study (DEARS). Specifically, this portion of the study compared outdoor residential  
14 concentrations with those from a community monitoring site. Daily and seasonal influences on the spatial  
15 distribution of the  $PM_{10-2.5}$  were examined. Also, the  $PM_{10-2.5}$  and  $PM_{2.5}$  concentration data collected at the  
16 community monitoring site allowed the CPEM performance to be evaluated against a referee  
17 dichotomous sampler.

## 19 **Methods**

### 20 *CPEM*

21 The CPEM simultaneously provides  $PM_{10-2.5}$  and  $PM_{2.5}$  concentrations from one compact  
22 instrument. The CPEM is a series of three separation stages designed to be inserted into the MSP Model  
23 200  $PM_{10}$  PEM (MSP Corp, Shoreview, MN) (Figure 2). The CPEM operates on a battery powered pump  
24 to collect  $PM_{10-2.5}$  by impaction on two, sequential 25 mm Teflo filters (Pall Corp., East Hills, NY). The  
25 final stage collects the  $PM_{2.5}$  on a 37 mm Teflo filter (Pall Corp., East Hills, NY). Teflo filters allow  
26 speciation analyses to be conducted on both size fractions. The CPEM's small dimensions (4.7 cm high,  
27 4.2 cm wide), light weight (110 g), low flow (2 Lpm), yet rugged construction allow the system to be  
28 deployed as a personal exposure monitor as well as a stationary indoor or outdoor monitor. Flow

1 measurements measured with a Drycal DC-Lite (BIOS International, Butler NJ ) at CPEM deployment and  
2 retrieval were averaged to calculate the sample volume. If necessary, pump flow was adjusted at  
3 deployment to achieve 2 Lpm

#### 4 *DEARS Study*

5 A primary goal of the DEARS was to evaluate the uncertainty associated with using community-  
6 based monitoring as a surrogate for human exposures to pollution in a metropolitan area impacted by  
7 multiple types of sources. A second DEARS research objective evaluated new exposure measurement  
8 technologies. Williams et al. (2008b) and Williams (2005) describe the DEARS objectives and study  
9 design in detail. CPEMs were deployed during the final two (of six) sampling seasons to assess the  
10 sampler performance capabilities to measure PM<sub>10-2.5</sub> and PM<sub>2.5</sub> concentrations and evaluate spatial and  
11 temporal variability. Summer season samples were collected between July 11 and August 26, 2006,  
12 while winter samples were collected from January 23 to February 24, 2007.

13 Sample collection focused on six Enumeration Measurement Areas (EMAs) around Detroit  
14 (Figure 1) selected to highlight specific air pollution source categories in each area. The distance  
15 between EMAs varied from 2 to 48 km. EMAs 1 and 5, located immediately southwest of downtown  
16 Detroit, examined the impact of industrial sources. Residences in EMA 6 were within 300 m of Highway  
17 M39-Southfield Freeway, a major north-south route through the residential neighborhoods in the western  
18 portion of Detroit. EMA 4 was a residential neighborhood on the northeast side of Detroit and downwind  
19 of the industrial and mobile sources. EMA 7, located west of Detroit in Belleville, had no identified source  
20 categories and represented the regional background aerosol. EMA 0, the central monitoring site, was  
21 located at the Michigan Department of Environmental Quality (MDEQ) site in Allen Park, MI. MDEQ and  
22 U.S. EPA provided meteorological data. MDEQ provided hourly wind speed and direction data at EMA 0.  
23 U.S. EPA provided daily average Monin-Obukhov lengths as a measure of atmospheric stability.

#### 24 *Experimental Design*

25 Simultaneous measurements with the CPEM and an Andersen Model SA-244 Dichotomous  
26 sampler (Andersen, Smyrna, GA) collocated at EMA 0 assessed the CPEM comparability against an EPA  
27 referee sampler. Accuracy (n = 72) was calculated as the difference between the CPEM and referee  
28 concentrations divided by the referee concentration, where a value of 0 indicated no difference.

1 Collocated duplicate CPEM samples (n = 12) deployed at EMA 0 assessed precision, quantified as the  
2 coefficient of variation.

3 CPEMs were deployed daily on a Tuesday through Sunday schedule in each EMA to investigate  
4 PM<sub>10-2.5</sub> spatial and temporal variability. Summer had 35 days of sample collection in each EMA. Winter  
5 had 25 days of samples, except in EMA 6 which had 20 due to a lack of participants. The sample  
6 collection interval was 24 hours, nominally between 9:00 AM to 9:00 AM. CPEMs located outdoors were  
7 mounted on a bluff body about 2 m above the ground (Rodes and Thornburg, 2006). CPEMs deployed in  
8 EMA 0 faced northeast due to space limitations on the southwest side of the bluff body. CPEMs deployed  
9 in other EMAs were deployed in residential backyards and deliberately oriented to face away from  
10 structures or other obstacles.

11 EMAs 1, 4, 5, and 6 had multiple days each season when two or three outdoor residential  
12 locations were sampled simultaneously within the EMA (Table 1). The number of intra-EMA duplicate  
13 comparisons varied from a minimum of 9 (summer, EMA 1) to a maximum of 20 (summer, EMA 4). This  
14 replication allowed spatial variability on scales less than 1 km to be studied. The residences sampled  
15 during these days were labeled as A, B, or C. Residence A was the closest to the Southfield Freeway  
16 (EMA 6) or closest to the geographical center of all other EMAs. Residence B was the next closest and  
17 Residence C was the furthest away. The average distance between residences was  $0.8 \pm 0.2$  km.

### 18 *Analysis*

19 Gravimetric analysis of PM<sub>10-2.5</sub> and PM<sub>2.5</sub> filters was conducted according to procedures  
20 described in Lawless and Rodes (1999). Field and laboratory blanks provided a blank correction factor  
21 applied to all mass concentrations.

22 SAS version 9 (SAS Inc., Cary, NC) was used to perform all statistical analyses using an alpha of  
23 0.05. Concentrations were log-transformed for statistical analyses comparing EMAs to satisfy normal  
24 distribution assumptions. The Tukey-Kramer least squares analysis procedure was used to determine if  
25 concentration differences between EMAs, within EMAs, and seasonal variations were statistically  
26 significant. The Coefficient of Divergence (COD) between EMAs assessed PM<sub>10-2.5</sub> spatial homogeneity  
27 (Pinto et al., 2004). A COD of 0 indicates complete homogeneity and a value of 1 indicates maximum  
28 differences. The Pearson Correlation Coefficients (r) assessed the temporal homogeneity in PM<sub>10-2.5</sub>

1 between EMAs. Spatial-temporal modeling was conducted with a non-linear, autoregressive analysis  
2 procedure to assess the concentration differences within and between EMAs. The autoregressive  
3 structure controlled for the daily variability in meteorology conditions such that comparisons between the  
4 independent variables could be made. The model included distance between EMAs, weekday/weekend,  
5 wind direction, wind speed, and atmospheric stability.

## 7 **Results**

8 The 2 Lpm CPEM performed well compared to the 16.7 Lpm dichotomous sampler for 24 hour  
9 integrated samples (Figure 2). The  $R^2$  from the linear regression was 0.91 for  $PM_{2.5}$  and 0.81 for  $PM_{10-2.5}$ .  
10 For  $PM_{2.5}$ , the statistically significant linear regression slope was 0.97 ( $p$ -value  $< 0.0001$ ) and not different  
11 from unity (95% C.I.: 0.90 to 1.05). The  $1 \mu\text{g}/\text{m}^3$  intercept was statistically insignificant ( $p$ -value = 0.14).  
12 The  $PM_{10-2.5}$  linear regression slope of 0.77 was also statistically significant ( $p$ -value  $< 0.0001$ ) but was  
13 different from unity (95% C.I.: 0.68 to 0.86). Again, the  $1 \mu\text{g}/\text{m}^3$  intercept was statistically insignificant ( $p$ -  
14 value = 0.13). Accuracy (Figure 3) and precision (Figure 4) of the CPEM  $PM_{10-2.5}$  and  $PM_{2.5}$  data typically  
15 became poorer as the concentration decreased below  $10 \mu\text{g}/\text{m}^3$ . However, there were exceptions to this  
16 trend.

17 Figure 5 shows the  $PM_{10-2.5}$  geometric mean and standard deviation for each EMA as a function  
18 of season.  $PM_{10-2.5}$  spatial variability between EMAs was evident. EMAs 1 and 5, with average summer  
19 concentrations greater than  $12 \mu\text{g}/\text{m}^3$ , had significantly greater  $PM_{10-2.5}$  than the other EMAs ( $p$ -value  $<$   
20  $0.01$ ). Summer  $PM_{10-2.5}$  in EMAs 0, 4, 6, and 7 were statistically similar, with average concentrations  
21 between  $6.3$  and  $7.5 \mu\text{g}/\text{m}^3$ . Winter 2007  $PM_{10-2.5}$  spatial variability between EMAs showed a different  
22 trend. EMA 7 concentrations were lower ( $p$ -value  $< 0.05$ ) than concentrations in EMAs 0, 1, 5, and 6.  
23 EMA 4 concentrations were lower ( $p$ -value  $< 0.05$ ) than concentrations in EMAs 0 and 6. Winter  $PM_{10-2.5}$   
24 in EMAs 0, 1, 5, and 6 were similar and varied from  $9.4$  to  $11.0 \mu\text{g}/\text{m}^3$ .

25 Figure 5 also shows a seasonal influence on the  $PM_{10-2.5}$  spatial distribution. EMAs 1, 5, and 7  
26 had significantly greater  $PM_{10-2.5}$  in summer than in winter ( $p$ -value  $< 0.05$ ). Winter  $PM_{10-2.5}$  was greater  
27 than summer  $PM_{10-2.5}$  in EMAs 0 and 6 ( $p$ -value  $< 0.05$ ). Seasonal  $PM_{10-2.5}$  variations in EMA 4 were  
28 small and statistically insignificant.

1            Table 2 presents r and COD values between EMAs for each season. All summer r values were  
2 statistically significant (p-value < 0.02) and ranged from 0.28 to 0.63. Winter r values ranged from 0.03 to  
3 0.76. Winter EMA 6 PM<sub>10-2.5</sub> was not correlated temporally (p-values > 0.4) with the PM<sub>10-2.5</sub> other EMAs.  
4 COD values also showed a seasonal pattern. Summer CODs ranged from 0.17 to 0.41. Winter CODs  
5 spanned 0.26 to 0.50. For each season, the paired r and COD values were logically divided or  
6 “clustered” into 3 distinct groups. In summer, the first cluster was the singular comparison of EMAs 1 and  
7 5, which had the lowest COD. CODs between EMAs 0, 4, 6, and 7, varying from 0.21 to 0.26, comprised  
8 the second summer cluster. The third summer cluster, with CODs generally greater than 0.33, compared  
9 EMAs 1 and 5 to EMAs 0, 4, 6, and 7. The first winter group contained EMA 6 comparisons to the other  
10 EMAs because of the statistically insignificant r values. The second winter group, with COD values  
11 greater than 0.3 and statistically significant r values compared EMAs 0, 1, 5. EMA 4 and 7 comparisons  
12 with the other EMAs formed the third winter cluster.

13            The DEARS sample scheme planned for duplicate participants within certain EMAs to be  
14 sampled weekly. This scheme allowed a comparison of the intra-EMA variability in the particulate matter  
15 concentrations accounting for daily variability in the concentrations (Table 1). PM<sub>10-2.5</sub> daily variability was  
16 evident for both summer and winter, as shown by the statistically significant p-values for the “Day”  
17 variable. The statistically insignificant p-values for the “Location” variable indicate PM<sub>10-2.5</sub> was spatially  
18 similar within most EMAs. Only winter PM<sub>10-2.5</sub> within EMA 6 varied spatially (p-value < 0.05). The trend  
19 indicated PM<sub>10-2.5</sub> decreased with increasing distance from the Southfield Freeway.

20            Spatial-temporal modeling determined atmospheric stability was the only significant variable (p-  
21 value < 0.01) affecting summer and winter PM<sub>10-2.5</sub> concentrations. In summer, greater atmospheric  
22 instability, as indicated by small Monin-Obukhov lengths, promoted PM<sub>10-2.5</sub> concentration homogeneity  
23 between EMAs. Conversely, atmospheric stability during the winter produced PM<sub>10-2.5</sub> concentrations  
24 differences between EMA 6 and the other EMAs. Distance between EMAs, weekday/weekend, wind  
25 speed, and wind direction did not influence summer or winter PM<sub>10-2.5</sub> concentrations across Detroit .

26

## 27 **Discussion**

### 28 *CPEM Performance*

1           The easy maintenance, assembly, and deployment of the CPEM made the sampler ideal for field  
2 use. All samples were processed in a field office without the need for impactor greasing or special filter  
3 handling. Trained technicians unloaded returned CPEMs and prepared new CPEMs at a rate of 12 per  
4 hour. Use of Teflo filters for sample collection will allow chemical speciation analyses to be conducted in  
5 the future. CPEM sample completeness exceeded the DEARS minimum data quality objective of 90%.  
6 Summer 2006 and winter 2007 had 97% and 96% valid samples. DEARS data quality objectives for  
7 CPEM PM<sub>2.5</sub> and PM<sub>10-2.5</sub> performance were ± 20% accuracy and ± 20% precision (Williams et al., 2000).  
8 CPEM PM<sub>2.5</sub> data achieved the accuracy and quality objectives. PM<sub>10-2.5</sub> performance achieved the  
9 precision data quality objective of ± 20% precision, but the cumulative accuracy was 24%, slightly higher  
10 than the target.

11           CPEM PM<sub>10-2.5</sub> measurement performance is very similar to that of other low flow particulate  
12 matter samplers used for saturation studies. Chen et al. (2007) used PM<sub>10</sub> and PM<sub>2.5</sub> MiniVol samplers  
13 (Airmetrics, Eugene, OR) to calculate PM<sub>10-2.5</sub> by difference. The performance of the MiniVol was nearly  
14 identical to the CPEM performance during DEARS. Their study showed the MiniVol had a negative bias  
15 of 20% compared to the reference method and an R<sup>2</sup> value of 0.78. Chen et al. attributed the differences  
16 to differences in face-velocity through the filter, aspiration efficiencies into the inlet, or particle bounce in  
17 the dichotomous sampler. Ott et al. (2008) recently reported PM<sub>10-2.5</sub> measured with the Wagner and  
18 Leith passive sampler (Wagner and Leith, 2001) during a field study in Iowa. Their results showed  
19 excellent precision, yet a 29% positive bias compared to the dichotomous sampler was observed. This  
20 systematic bias was attributed to assumptions made in calculating mass concentration from the SEM  
21 images, and not with the coarse particle collection efficiency. The Personal Respirable Particulate  
22 Sampler (PRPS) is the most similar to the CPEM in operation in that PM<sub>10-2.5</sub> is measured directly,  
23 although the unit sampled at 5 Lpm and collected PM<sub>10-2.5</sub> on a PUF substrate (Demokritou et al., 2003).  
24 Case et al. (2008) collocated a modified version of the PRPS with an Andersen dichotomous sampler.  
25 Their analysis showed a 10% positive bias, a statistically significant R<sup>2</sup> of 0.87, and coefficient of variation  
26 generally less than 2.

27           The negative bias in the CPEM PM<sub>10-2.5</sub> may have been caused by the orientation of the sampler  
28 when mounted on the bluff body. The CPEM uses a MSP Model 200 inlet cap that is inherently

1 directional when used outdoors, as opposed to the omni-directional dichotomous sampler inlet. In  
2 addition, the bluff body system is designed to simulate personal inhalation exposures to particulate  
3 matter, where the presence of the human body is known to alter particle flow trajectories (Kenny et al.,  
4 1997). In studies such as the DEARS where comparisons are made between personal and outdoor  
5 sampling technologies, the use of a bluff body to simulate the human form should provide similar flow  
6 profiles to bias both personal and outdoor measurements in the same manner. As a result, the bluff body  
7 is not an omni-directional platform. The bluff body mounted CPEM faced northeast at EMA 0, when the  
8 prevailing wind direction during both seasons covered an arc from the south to northwest. That meant  
9 the bluff body was a barrier that could have prevented complete collection of the coarse particles under  
10 the prevailing wind direction. With the CPEM on the downwind side, the airflow around the bluff body  
11 would have formed eddy vortices that could have carried the particulate matter away from the bluff body  
12 and decreased the coarse particle concentration in proximity to the CPEM inlet. Both wind speed and  
13 direction would then be expected to influence the effect of the eddy vortices on the sampled  
14 concentration. In wind tunnel experiments, Kenny et al. (1997) demonstrated that personal aerosol  
15 samplers located on the back, downstream side of a manikin sampled 80% to 90% of 6  $\mu\text{m}$  diameter  
16 particles. This study was not designed to characterize this effect, and insufficient data were available to  
17 develop a statistically valid concentration correction factor accounting for wind speed and wind direction.  
18 However, the analysis did suggest the magnitude of the bluff body effect decreased the measured CPEM  
19  $\text{PM}_{10-2.5}$  by approximately 2 to 5  $\mu\text{g}/\text{m}^3$ , equivalent to the -20% bias seen in the data.

20 Instances of poor CPEM accuracy and precision at concentrations less than 10  $\mu\text{g}/\text{m}^3$  can be  
21 explained by a propagation of error analysis. Potential sources of error in the calculation of the  
22 particulate matter concentration were the gravimetric mass, sample flow, and sample time  
23 measurements. The error associated with digital flowmeters and a synchronized clock was minimal,  
24 especially when the variability in the pump flow and sample collection period was negligible. However,  
25 the error associated with gravimetric measurement of the mass collected on the filter can be significant.  
26 Across both seasons, the error in the  $\text{PM}_{10-2.5}$  gravimetric analysis, calculated from a combination of field  
27 and laboratory blanks, was 2.8  $\mu\text{g}$ . When converted to a concentration, the error in the CPEM and  
28 dichotomous sampler measurements was 1  $\mu\text{g}/\text{m}^3$  and 0.17  $\mu\text{g}/\text{m}^3$ , respectively. Considering that almost

1 70% of the  $PM_{10-2.5}$  measured during DEARS were less than  $10 \mu\text{g}/\text{m}^3$ , the impact of the low sample  
2 volume on the accuracy and precision of the CPEM concentration measurements was magnified  
3 compared to the dichotomous sampler measurements.

4 Based on the preceding analysis, we concluded the CPEM data were suitable for investigating  
5  $PM_{10-2.5}$  spatial and temporal variability across Detroit.

#### 6 *Comparison with FEM Requirements*

7 The CPEM comparability data were compared against the EPA federal acceptance criteria for  
8 federal equivalent methods (FEM). The federal acceptance criteria for  $PM_{10-2.5}$  Class II instruments  
9 specify a regression slope of  $1 \pm 0.1$ , intercept of  $-3.8$  to  $3.8 \mu\text{g}/\text{m}^3$ ,  $R^2$  greater than 0.9, and precision of  
10 15% (U.S. EPA, 2007). Class II  $PM_{2.5}$  FEM acceptance criteria are the same except the intercept range  
11 is  $-1.5$  to  $1.5 \mu\text{g}/\text{m}^3$ . CPEM performance for  $PM_{2.5}$  met all the EPA acceptance criteria. However, the  
12 CPEM  $PM_{10-2.5}$  performance only satisfied the criteria for the y-intercept. The linear regression slope  
13 showed the CPEM underestimated  $PM_{10-2.5}$  by an average of 23%, 11% more than the minimum  
14 acceptable level. The  $R^2$  of 0.81 was slightly less than the target value. The aggregate CPEM  $PM_{10-2.5}$   
15 precision, calculated as the root mean square of all comparisons, was 18%. Although all FEM criteria  
16 were not achieved, Chow and Watson (2008) recently noted that a portable, battery powered sampler like  
17 the CPEM can sacrifice FEM requirements to achieve study objectives. Research studies like the  
18 DEARS require inexpensive, portable, easy to use instrumentation, like the CPEM, to provide  
19 representative spatial and temporal  $PM_{10-2.5}$  mass and chemical composition distributions at a reasonable  
20 cost.

#### 21 *Summer $PM_{10-2.5}$ Concentration Gradients*

22  $PM_{10-2.5}$  during DEARS showed spatial differences between EMAs during the summer.  $PM_{10-2.5}$  in  
23 EMAs 1 and 5 was more than  $5 \mu\text{g}/\text{m}^3$  higher than the other EMAs. Corresponding CODs between EMAs  
24 1 or 5 with the other EMAs were greater than 0.33, also indicating spatial variability. The summer spatial  
25  $PM_{10-2.5}$  gradients possibly were due to localized industrial sources in those areas and higher population  
26 density (Phillips et al., 2008). Burton et al. (1996) reported statistically significant differences of up to 3  
27  $\mu\text{g}/\text{m}^3$  across different portions of Philadelphia due to the presence of local sources in more densely

1 populated areas. Higher  $PM_{10-2.5}$  is expected in industrial, urban areas, although periods do occur when  
2  $PM_{10-2.5}$  in rural areas is greater than urban areas (Querol et al., 2008).

3 Even though spatial concentration gradients existed, summer  $PM_{10-2.5}$  was temporally correlated  
4 across all EMAs. The highly unstable atmosphere induced by daytime heating promoted mixing and  
5 dispersion of  $PM_{10-2.5}$  during the summer to yield statistically significant  $r$  values, ranging from 0.28 to  
6 0.63. As a result of the atmospheric instability, a large portion of the  $PM_{10-2.5}$  in the summer may have  
7 originated from a common source that impacted all of Detroit.

8 Although a detailed  $PM_{10-2.5}$  size distribution characterization was not part of the DEARS study  
9 design, we hypothesize that these findings suggest the  $PM_{10-2.5}$  in Detroit during the summer is comprised  
10 of two components. One component, most likely consists of particles smaller than  $5\ \mu\text{m}$  that can be  
11 transported over substantial distances, especially in unstable atmospheres, to create an urban  
12 background concentration.  $PM_{10-2.5}$  in EMAs influenced the most by the urban background, and probably  
13 minimally impacted by local sources, consistently exhibited the lowest concentrations, statistically  
14 significant correlations, low CODs, and large distances between them. EMAs 0, 4, 6, 7 during the  
15 summer fit this category. Alternatively, EMAs with high  $PM_{10-2.5}$ , high correlations and low CODs signified  
16 areas impacted by the same, local source(s). Therefore, the other  $PM_{10-2.5}$  component consists of larger  
17 particles from local sources to generate areas of elevated  $PM_{10-2.5}$ . Reid et al. (2003) suggested these  
18 larger particles are between 5 to  $12\ \mu\text{m}$ . Summer  $PM_{10-2.5}$  in EMAs 1 and 5 fit this profile.

#### 19 *Winter $PM_{10-2.5}$ Concentration Gradients*

20 Spatial differences also existed in the winter.  $PM_{10-2.5}$  in EMAs 7 and 4 were as much as  $8\ \mu\text{g}/\text{m}^3$   
21 lower than EMAs 0 and 6. CODs for these four comparisons were also the highest ( $>0.43$ ). Furthermore,  
22 the temporal variability indicated by the statistically insignificant  $r$  values corresponding to EMA 6  
23 suggested the presence of a unique source limited to that EMA. The spatial and temporal variability  
24 possibly was caused by the presence of resuspended road salt from the Southfield Freeway (EMA 6) and  
25 Interstate 75 (near EMA 0). These two freeways average more than 100,000 vehicles per day, and  
26 frequent snow events necessitated recurrent application of salt to the roads. Whereas other EMAs were  
27 residential areas with few major roads an insignificant daily traffic counts to suspend the road salt that

1 was applied. In addition, the highly stable atmosphere inhibited dispersion and mixing of  $PM_{10-2.5}$   
2 resuspended from the roads.

3 The proximity of the EMA 6 sampling locations to the highway and the high concentration of  
4 resuspended road salt probably caused the statistically insignificant r values. The more stable  
5 atmosphere in the winter, identified as a statistically significant variable by the spatial-temporal modeling,  
6 probably minimized the mixing and dilution of the resuspended road salt. As a result, elevated  $PM_{10-2.5}$   
7 concentrations were not correlated with the other EMAs. Within EMA 6, the decrease in  $PM_{10-2.5}$  over a  
8 lateral distance of 300 m from the Southfield Freeway indicated the atmospheric stability limited  
9 dispersion and dilution of the suspended road salt over short distances.

10 The size of the resuspended coarse particles also probably influenced the insignificant r values  
11 between EMA 6 and other EMAs. A detailed assessment of the  $PM_{10-2.5}$  particle size distribution was not  
12 part of the DEARS study design. However, Eleftheriadis and Colbeck (2001) reported the size  
13 distributions for a number of crustal elements in the coarse fraction. Their urban data had a modal size  
14 generally greater than 5  $\mu m$ . Assuming their findings are applicable to Detroit, the particles sampled in  
15 EMA 6 probably were greater than 5  $\mu m$ . Therefore, the increased mass of these “larger” particles  
16 minimizes their long range transport. Because detailed  $PM_{10-2.5}$  size distribution measurements were not  
17 obtained using multi-stage impactors or real-time instruments, this hypothesis cannot be confirmed until  
18 size distribution and composition of the collected particles is measured using electron microscopy.

### 19 *Central Site Representativeness*

20 The central monitoring site in EMA 0 is also the MDEQ SLAMS site for Wayne County. Based on  
21 the DEARS data for summer 2006 and winter 2007, the applicability of this site for monitoring  $PM_{10-2.5}$  for  
22 the entire Detroit metropolitan area was evaluated. Monn (2001) noted the spatial variation of  $PM_{10-2.5}$  in  
23 an urban environment makes the collection of representative samples critical for accurate exposure  
24 assessment. When applied to ambient monitoring, representative sample collection refers to placement  
25 of the monitor in a location that characterizes the largest percentage of the population. Factors to  
26 consider include micro-meteorology, population density, types of sources (point vs. line vs. area), number  
27 of sources per square kilometer, and source emission rates.

1           PM<sub>10-2.5</sub> at EMA 0 were not as representative for the entire Detroit metropolitan area as PM<sub>2.5</sub>  
2 (Rodes et al., 2008). PM<sub>10-2.5</sub> in EMA 0 differed by -6 to + 6 µg/m<sup>3</sup> from the other EMAs during both  
3 seasons. EMA 0 exhibited spatial differences in PM<sub>10-2.5</sub> with EMAs 1 and 5 during the summer, and  
4 EMAs 4 and 7 during the winter. Temporal differences were noted between EMAs 0 and 6 during the  
5 winter.

6           Multiple mechanisms possibly can influence the representativeness of EMA 0 as a central  
7 monitoring site. Turbulent mixing near the source and atmospheric instability are the two mechanisms  
8 that disperse PM<sub>10-2.5</sub> in the atmosphere. Turbulent mixing probably suspends particles to a sufficient  
9 height where the thermally induced atmospheric instability creates the buoyancy to carry PM<sub>10-2.5</sub> to a  
10 height that favors long range transport (Hasegawa et al., 2007). Deposition rates are controlled by  
11 Stokes settling (Noll and Aluko, 2006), and therefore are strongly proportional to particle diameter. The  
12 tree canopy (Freiman et al., 2006; McDonald et al., 2007) and possibly building density also promote  
13 deposition and hinder long range transport, especially as particle diameter increases to 10 µm. Another  
14 consideration is the effect of topography on coarse particle transport. Intervening ridges may concentrate  
15 coarse particles in one valley and minimize their transport up the slope to the top of the ridge and into  
16 neighboring valleys. Although this phenomenon is not applicable in Detroit, cities like Birmingham, AL  
17 and Pittsburgh, PA may be affected. Understanding the impact of these mechanisms on the size  
18 distribution and composition of the PM<sub>10-2.5</sub> at multiple locations in an urban airshed is necessary to  
19 develop the criteria for representative monitoring of PM<sub>10-2.5</sub>. Alternatively, this type of detailed data can  
20 be used to develop models (Georgopoulos et al., 2005) and other exposure surrogates for  
21 epidemiological studies and risk assessments.

## 22

### 23 **Conclusions**

24           The evaluation of the new CPEM sampler during DEARS effort demonstrated its versatility and  
25 compactness to allow its application in future studies as a stationary or personal sampler. The accuracy  
26 and precision of the CPEM PM<sub>10-2.5</sub> and PM<sub>2.5</sub> concentrations, compared to a dichotomous sampler, were  
27 similar to other portable instruments.

1           PM<sub>10-2.5</sub> was measured during summer 2006 and winter 2007 of the DEARS. Summer PM<sub>10-2.5</sub>  
2 exhibited spatial variability across the Detroit airshed, whereas winter PM<sub>10-2.5</sub> showed spatial and  
3 temporal variability. The monitoring locations suggested the summer PM<sub>10-2.5</sub> spatial gradients were  
4 caused by localized industrial sources. Temporal consistency of summer PM<sub>10-2.5</sub> concentrations caused  
5 by atmospheric instability suggested a potential regional PM<sub>10-2.5</sub> background concentration consisting of  
6 particles between 2.5 to 5  $\mu\text{m}$ . Spatial and temporal gradients between an area most likely impacted by  
7 suspended road salt and the other areas were found in the winter. A stable atmosphere limited mixing  
8 and dispersion of the PM<sub>10-2.5</sub> particles from the near roadway area, except within 300 m of the monitored  
9 freeway. Wind speed and direction, distance, and day of week were variables that did not influence the  
10 spatial and temporal concentration gradients between areas. The size distribution of the PM<sub>10-2.5</sub> particles  
11 most likely was also a key factor, and should be measured in future studies. The tree canopy, building  
12 density, and topography are other factors to be considered when characterizing PM<sub>10-2.5</sub> spatial and  
13 temporal variability.

14           PM<sub>10-2.5</sub> speciation data are needed to definitively identify the sources and components that  
15 contribute to the spatial and temporal variability. Besides the standard inorganic and organic chemical  
16 speciation analyses, the possibly large microbiological contribution to the PM<sub>10-2.5</sub> mass should be  
17 quantified. This speciation data then could identify a marker, like sulfate for PM<sub>2.5</sub>, for penetration of  
18 PM<sub>10-2.5</sub> of ambient origin into buildings.

19           This research provided additional information for the development of National Ambient Air Quality  
20 Standard compliance monitoring requirements for PM<sub>10-2.5</sub>. These findings indicated PM<sub>10-2.5</sub>, although  
21 highly correlated in some instances, vary in magnitude sufficiently that a central monitoring site may not  
22 adequately represent the population's exposure in a complex urban airshed. Factors that should be  
23 considered when developing compliance monitor placement criteria are micro-meteorology, population  
24 density, types of sources (point vs. line vs. area), number of sources per square kilometer, and source  
25 emission rates.

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11

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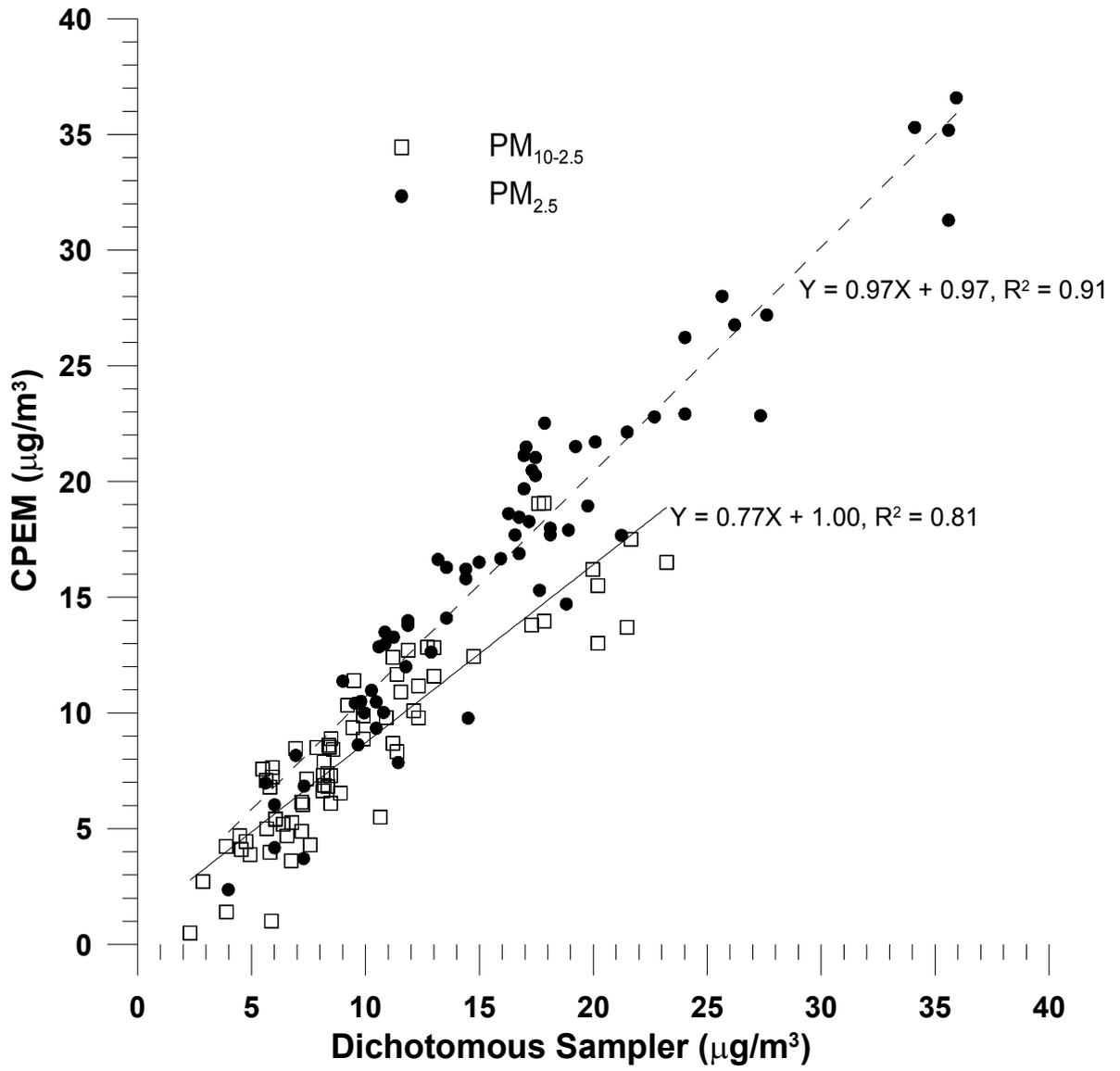
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1 **Figure 1. Locations of the central monitoring site (EMA 0) in relation to the Enumeration**  
2 **Measurement areas (EMAs) where outdoor residential  $PM_{10-2.5}$  and  $PM_{2.5}$  measurements were**  
3 **conducted. EMA 7 is located approximately 23 km west of the central site, along Interstate 94.**  
4  
5 **Figure 2. Scatterplot of  $PM_{10-2.5}$  and  $PM_{2.5}$  concentrations measured by the CPEM and Andersen**  
6 **Dichotomous sampler at the central monitoring site (EMA 0)**  
7  
8 **Figure 3. Accuracy of the CPEM compared to the Andersen Dichotomous sampler.  $PM_{10-2.5}$  and**  
9  **$PM_{2.5}$  sample sizes were 72 and 66. A fractional difference of 0 indicates perfect agreement.**  
10  
11 **Figure 4. Precision of the CPEM based on collocated duplicate samples. Precision presented as**  
12 **the coefficient of variation in the measurements.  $PM_{10-2.5}$  and  $PM_{2.5}$  sample sizes were 72 and 66.**  
13  
14 **Figure 5. Seasonal and spatial  $PM_{10-2.5}$  variability in each EMA. Error bars show 1 standard**  
15 **deviation of the geometric mean concentration. EMAs marked with asterisks had statistically**  
16 **significant differences between seasons ( $p < 0.05$ ). Concentration bars with number symbols (#)**  
17 **or carets (^) indicate statistically significant spatial differences in summer or winter, respectively.**  
18



1 Figure 2.

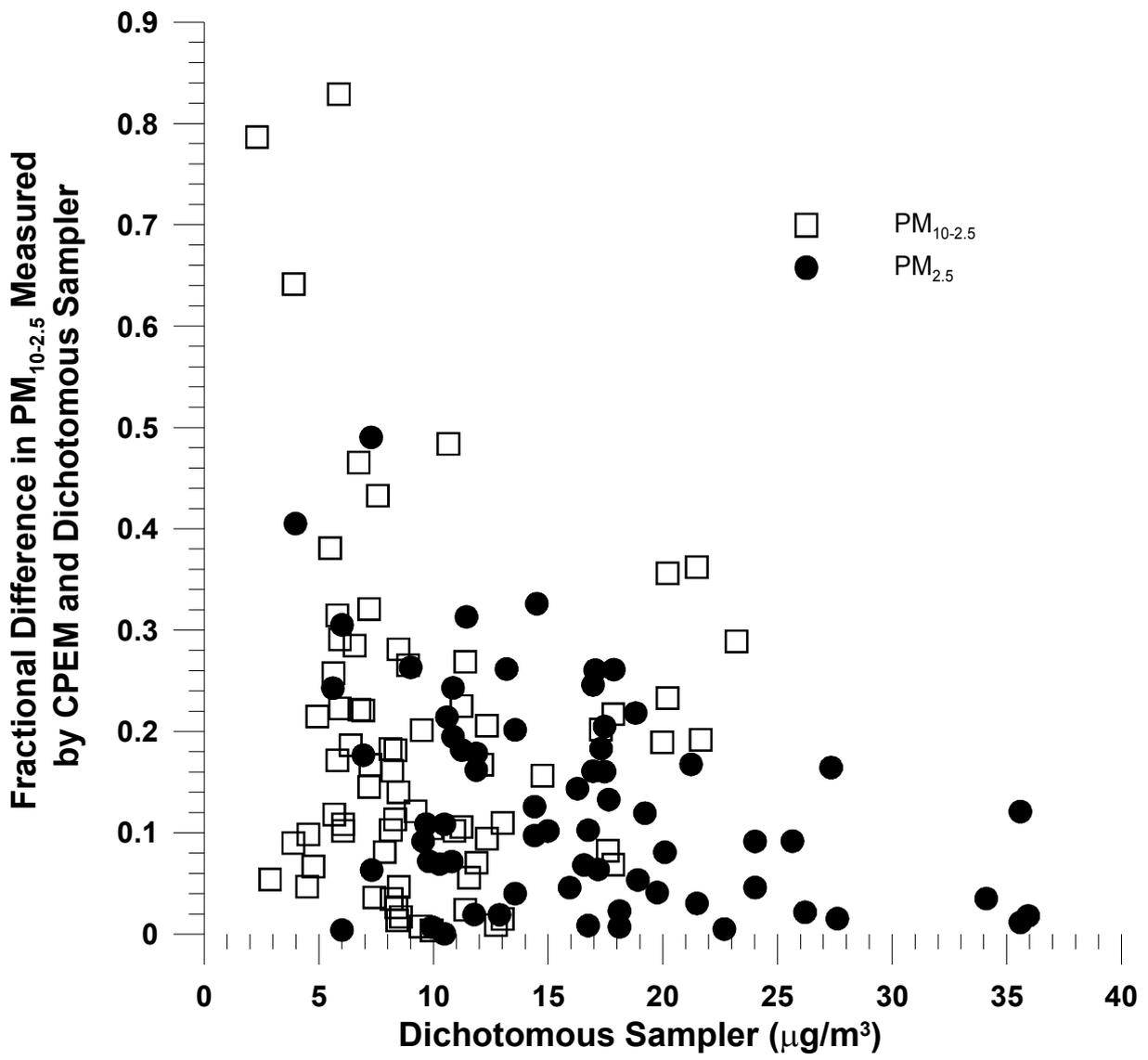
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1 Figure 3.

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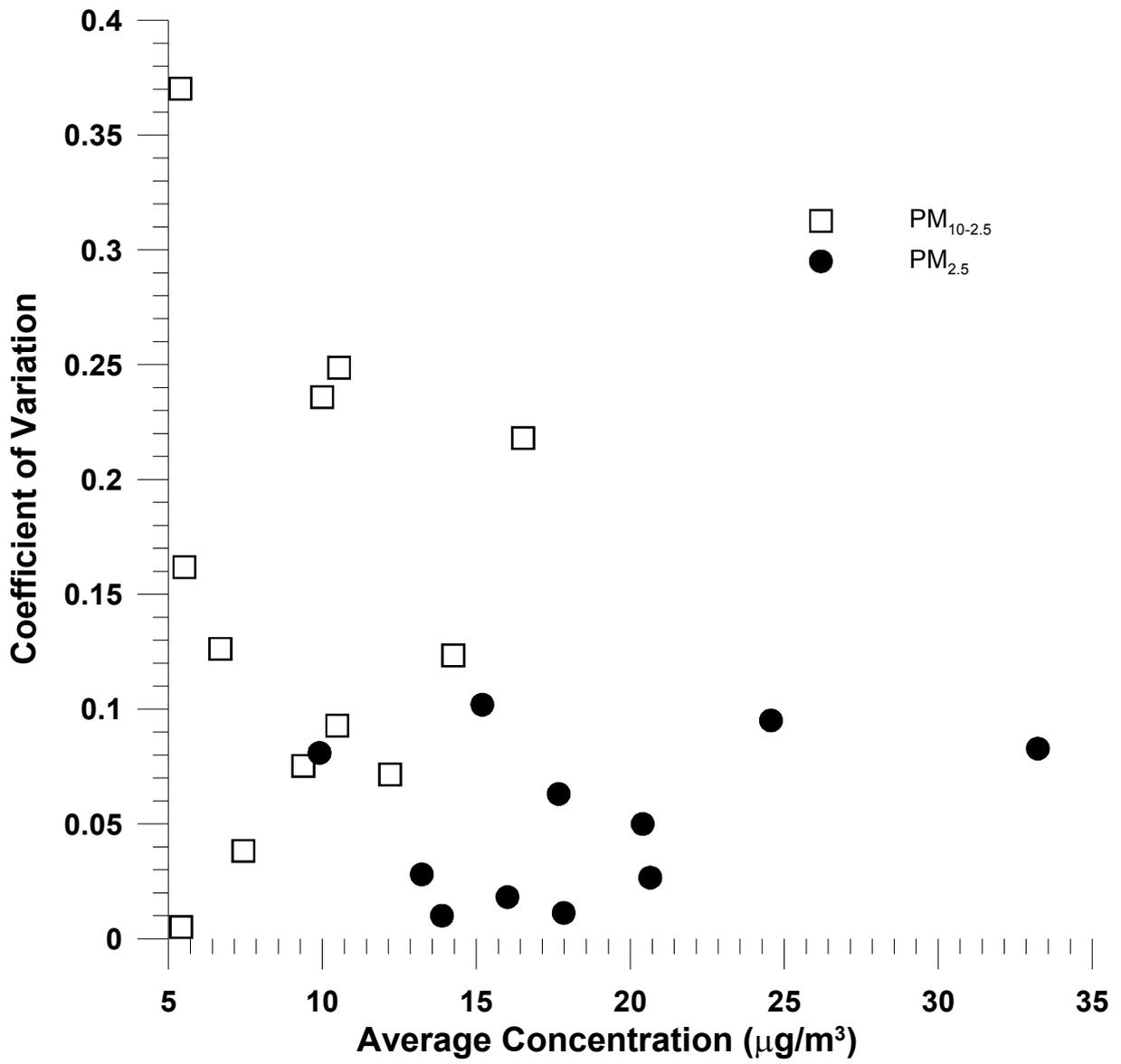
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1 Figure 4

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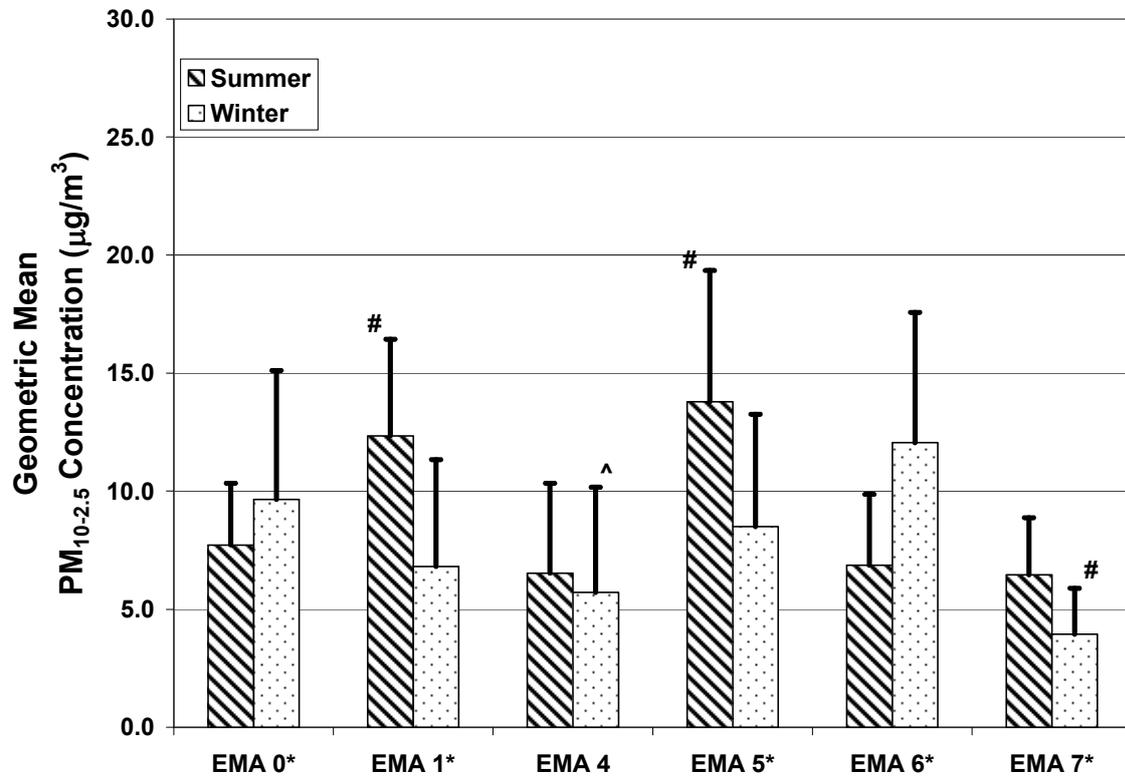
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1 Figure 5.



2

1 **Table 1. Statistical analysis of the PM<sub>10-2.5</sub> spatial variability (Location) within an EMA**  
2 **accounting for daily variations in PM<sub>10-2.5</sub> (Day)**

3

4 **Table 2. Coefficients of divergence (COD) and Pearson correlation coefficients (r) between**  
5 **PM<sub>10-2.5</sub> measured at the EMAs. Correlations in bold text are statistically insignificant (p-**  
6 **value > 0.05) to highlight temporal differences between EMAs.**

**Table 1.**  
**Summer**

	EMA 1 (n = 9)		EMA 4 (n = 20)		EMA 5 (n = 10)		EMA 6 (n = 19)	
	df	p-value	df	p-value	df	p-value	df	p-value
Location	1	0.26	1	0.41	1	0.68	1	0.48
Day	8	< 0.001	19	0.002	9	0.003	18	0.001
Error	8		19		9		18	
Group A mean	12.6 ± 4.3		6.5 ± 3.3		13.0 ± 6.5		6.9 ± 2.6	
Group B mean	13.2 ± 4.4		7.3 ± 5.1		13.5 ± 4.6		6.5 ± 3.2	

**Winter**

	EMA 1 (n = 15)		EMA 4 (n = 15)		EMA 6 (n = 15)	
	df	p-value	df	p-value	df	p-value
Location	2	0.47	1	0.89	1	< 0.001
Day	9	< 0.001	14	0.07	14	< 0.001
Error	13		14		14	
Group A mean	4.9 ± 3.7		6.1 ± 5.5		14.5 ± 5.2	
Group B mean	5.8 ± 4.4		6.3 ± 4.3		11.1 ± 5.5	
Group C mean	3.0 ± 2.6		-		-	

Note: EMA 5 did not have duplicate participants in the same week in Winter.

**Table 2.**

Summer					Winter				
Cluster	EMAs	Distance (km)	COD	r	Cluster	EMAs	Distance (km)	COD	r
1	1 to 5	2	0.17	0.49	1a	5 to 6	16	0.29	<b>0.13</b>
2	4 to 6	21	0.23	0.57		1 to 6	16	0.29	<b>0.09</b>
	0 to 7	23	0.21	0.60		0 to 6	24	0.26	<b>0.31</b>
3	0 to 6	24	0.23	0.52	1b	4 to 6	21	0.43	<b>0.03</b>
	0 to 4	31	0.23	0.54		6 to 7	34	0.50	<b>0.10</b>
	6 to 7	34	0.24	0.56	2	1 to 5	2	0.31	0.70
	4 to 7	48	0.26	0.32		0 to 5	11	0.30	0.63
	0 to 5	11	0.33	0.41		0 to 1	12	0.31	0.76
	0 to 1	12	0.28	0.53	3	1 to 4	18	0.40	0.50
	5 to 6	16	0.40	0.45		4 to 5	19	0.41	0.47
	1 to 6	16	0.33	0.63		0 to 7	23	0.46	0.61
	1 to 4	18	0.37	0.59		5 to 7	31	0.42	0.67
	4 to 5	19	0.41	0.28		0 to 4	31	0.47	0.54
5 to 7	31	0.41	0.37		1 to 7	32	0.40	0.62	
1 to 7	32	0.33	0.54		4 to 7	48	0.40	0.64	