JEFFREY DAVID YANOSKY

Fine Particle Exposure of Prescribed Fire Workers in the Southeastern United States and a Comparison of Several Particulate Matter Sampling Methods

(Under the direction of DAVID MACINTOSH)

Personal exposure concentrations of particles with aerodynamic diameter <2.5 μm ($PM_{2.5}$) of prescribed fire workers were measured at two locations in the southeastern United States. Non-impacted ambient concentrations were measured as an estimate of background concentrations during burn activities. Four sampling method comparison studies were designed and performed to compare the FRM with 1) other gravimetric $PM_{2.5}$ sampling methods in ambient air, 2) optical $PM_{2.5}$ sampling methods in indoor air, 3) an optical sampling method (Grimm) for particles with aerodynamic diameter <10 μm (PM_{10}) in ambient air, and 4) a gravimetric $PM_{2.5}$ sampling method downwind of prescribed fires. The gravimetric $PM_{2.5}$ sampling methods agreed well in ambient air (R^2>0.96 for all) except for the MiniVol, the optical $PM_{2.5}$ sampling methods agree less well in indoor air,(R^2>0.592), the Grimm optical PM_{10} method agrees well in ambient air(R^2>0.944 for all), and the personal method agrees well (n=9, R^2=0.994) downwind of prescribed fires.

INDEX WORDS: Particulate matter, Prescribed fire, Wood smoke, Ambient air, $PM_{2.5}$, PM_{10}, Indoor air, Gravimetric, Direct-reading, Optical, FRM, KTL, Harvard impactor, MiniVol, DustTrak, APS, Grimm

FINE PARTICLE EXPOSURE OF PRESCRIBED FIRE WORKERS IN THE SOUTHEASTERN UNITED STATES AND A COMPARISON OF SEVERAL PARTICULATE MATTER SAMPLING METHODS

by

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CHAPTER 1

INTRODUCTION

Chapter 1 details information contained in each following chapter. Chapter 2 presents a selected review of key relevant literature and comprises two parts; the first concerning the comparison of several different $PM_{2.5}$ sampling methods, the second part concerning previous studies on forest firefighter exposure to fine particles and other pollutants. Chapter 3 describes the fine particle exposure assessment conducted on prescribed fire workers at two locations in the southeastern United States. The form that is referred to in the text as the prescribed fire worker log sheet is shown in Figure A-1 in the Appendix. The form used in the calibration of the FRM $PM_{2.5}$/PM_{10} samplers is shown in Figure A-2 in the Appendix. Chapter 4 contains a manuscript to be published in the June 2001 issue of the Journal of the Air and Waste Management Association describing the comparison of four gravimetric fine particle sampling methods in ambient air. Scatter plots of the ambient $PM_{2.5}$ sampler comparisons are presented in Figures A-3 through A-8 in the Appendix. Chapter 5 contains a manuscript submitted for publication in Atmospheric Environment in April 2001 describing a comparison of two direct-reading fine particle sampling methods with the Federal Reference Method for $PM_{2.5}$ in indoor air. Chapter 6 contains a report prepared for Grimm Technologies, Inc. describing a comparison of the Grimm 1.107 Environmental Dust Monitor and the Federal Reference Method for PM_{10} in ambient air. Chapter 7 contains conclusions and recommendations based on findings in the previous chapters.

CHAPTER 2

SELECTED LITERATURE REVIEW OF FINE PARTICLE SAMPLING METHODOLOGY AND FINDINGS

I. Selected fine particle sampling method literature review

Exposure to fine particulate matter with aerodynamic diameter <2.5 μm ($PM_{2.5}$) has been associated with changes in morbidity and mortality rates, with changes in respiratory function, and with changes in cardiovascular hospital admissions in numerous epidemiological studies. One such study by Schwartz et al. (1996) examining the strength of associations between mortality and different size fractions of ambient particulate matter in six cities in the eastern U.S. found higher associations between $PM_{2.5}$ levels than for particles with aerodynamic diameter <10 μm (PM_{10}) or with particles with aerodynamic diameter between 2.5 μm and 10 μm (coarse particles). The effects of weather and time of year were controlled for and were shown to have little influence on the exposure-effect relationship. Populations with chronic obstructive pulmonary disease and ischemic heart disease were found to have higher associations between mortality rates and $PM_{2.5}$ levels. Also, a review of the toxicology and current hypotheses regarding the mechanism of ambient particulate matter are presented. It is suggested that the mechanism of toxicity involves pulmonary inflammation by the production of metallic sulfates formed on the surface of acid-coated metallic particles, and that these particles may result in oxidant generation in pulmonary tissue or may produce an immune response causing the release of cytokines.

Many other studies have evaluated the relationship between $PM_{2.5}$ levels several cardio-pulmonary health effects in different areas at different times (Neas et al., 1999; Korrick et al., 1998; Peters, 2000). Considerable attention has focused on the interpretation of variation in results of such studies (Levy et al., 2000) and the

relationship between ambient and personal exposure to fine particulate matter (Bahadori et al., 1999). Several methods for measuring $PM_{2.5}$ are available and have been employed in past research. Continued research into these issues warrants investigation into the comparability of methods of measuring different $PM_{2.5}$ exposure concentrations (e.g., ambient, indoor, personal).

U.S. EPA designated Federal Reference Method $PM_{2.5}$ samplers (U.S. EPA, 1999) are now in widespread use for regulatory compliance monitoring nationwide and are anticipated to be a source of exposure data for many future $PM_{2.5}$ source-to-exposure and exposure-effect studies. Alternatives to a Federal Reference Method $PM_{2.5}$ sampler (FRM), such as the Airmetrics MiniVol, are often used to measure $PM_{2.5}$ in ambient air because of their reduced size, weight, and cost. For the same reasons and others, personal and indoor $PM_{2.5}$ sampling methods such as the BGI, Inc. GK2.05(KTL) Respirable/Thoracic Cyclone (KTL) and the Harvard-Marple Impactor (HI) are used to measure personal and indoor air fine particle levels, respectively.

A U.S. EPA document entitled "Prototype $PM_{2.5}$ Federal Reference Method Field Studies Report" reported a comparison of twenty collocated $PM_{2.5}$ samples from a then prototype FRM sampler (the Anderson RAAS) and the MiniVol in Phoenix, AZ (U.S. EPA, 1997). The results from the EPA comparison show a mean difference between the FRM sampler and the MiniVol of 1.70 $\mu g/m^3$, a collocated precision (standard deviation of between sampler differences) of 2.43 $\mu g/m^3$, and, from weighted least squares regression, a slope of 0.90, intercept of 3.24, and R^2=0.91. The estimates of the regression slope for the FRM versus the MiniVol are similar between the above

comparison and the present study, further supporting the observed tendency of the MiniVol to underestimate the FRM as concentrations increase.

The HI is a small, lightweight $PM_{2.5}$ sampler that has been widely used for ambient and indoor air sampling and its performance well characterized in many studies (Suh et al., 1997; Allen et al., 1999; Turner et al., 2000). One such study compared the HI and the FRM during 81 individual 24-hr sampling periods at Bakersfield, CA, and Riverside, CA, and found results consistent with those presented in Chapter 4. Within sampler type comparisons of the HI agreed well in both studies; the precision as measured by the Root-Mean-Square Error (RMSE) of collocated HI samplers reported by Babich et al. (2000) was 1.39 $\mu g\ m^{-3}$ and was 1.52 $\mu g\ m^{-3}$ for the present study. For comparisons between the FRM and HI, regression statistics showed similar results, though a higher R^2 was reported by Babich et al. Combined, these comparisons show that the HI compares well in measuring aerosols of differing composition across different regions of the United States.

In the present research presented in Chapter 4, simultaneous 24 hr-average $PM_{2.5}$ measurements in ambient air made by the BGI, Inc. PQ200 Federal Reference Method $PM_{2.5}$ sampler (FRM), the HI, the KTL, and the MiniVol are compared. This method comparison study augments previous evaluations of these non-FRM devices by comparing each to an FRM sampler over the range of 5-35 $\mu g/m^3$, applying corrections for the buoyant effects of air on gravimetric measurements, and reporting on the correlation, precision, and systematic and proportional bias within and between the sampling methods. If these methods are demonstrated to be comparable, then artifacts

of the sampling method can be dismissed as accounting for differences between ambient, indoor, and personal $PM_{2.5}$ measurements made using these devices that are greater than reported measurement error.

Recent advances in aerosol instrument technology have made it possible to measure and log aerosol concentrations in real time, that is, to collect time-resolved information on both the number and mass concentration as well as the size distribution of an aerosol. Such instruments offer insight into particulate levels during time series of short intervals (e.g., 5-min); this type of information cannot be obtained by gravimetric methods using current technology. Real-time measurements can also aid in identification of activities that contribute significantly to indoor particulate exposure (Abt et al., 2000). However, techniques used by real time aerosol monitors differ substantially from traditional gravimetric methods and therefore use of these new methods warrants investigation into the comparability of the methods in measuring $PM_{2.5}$ levels.

Lehocky and Williams reported an R^2 of 0.94 between DustTrak levels and gravimetric methods for respirable coal dust, though in that study regression techniques were used that force the intercept through zero (1996). The R^2 of 0.859 between FRM and DustTrak levels in the present study is similar to these findings. However, the slope in the present study for the FRM and DustTrak (β_1=2.57) is substantially different than found by Lehocky and Williams (β_1=0.73). Differences in the performance of the DustTrak in the present study may be attributed to lower concentration range, different

aerosol composition, and use of the internal impactor for $PM_{2.5}$ (versus an external 10-mm Dorr-Oliver cyclone with a 3.5 μm 50% cut-point).

II. Selected literature review of personal sampling for suspended particulate matter in forest smoke literature review

Prescribed fire has been an accepted forest management practice in the southeastern United States for a number of years. Prescribed fire activities are a source of numerous pollutants to the atmosphere and have been found to have a significant impact on air quality of a region (Sandberg et al., 1990). Prescribed fire activities are an essential part of an effective forest management program for many reasons including the prevention of uncontrolled wildfires (Glitzenstien et al., 1995). Fire workers engaged in prescribed burning practices are likely to be exposed to high levels of $PM_{2.5}$ as they often encounter smoky conditions. Wood smoke is known to contain numerous chemicals, in gaseous, liquid, and solid form. The primary health threats in wood smoke from prescribed burning identified by Reinhardt et al. (2000) are carbon monoxide, acrolein, formaldehyde, and respirable particulate matter. Other pollutants of concern include numerous other organic compounds, polynuclear aromatic hydrocarbons, nitrogen and sulfur oxides, and perhaps pesticide and herbicide residues, crystalline silica, and, at the Savannah River Site (SRS), even radioactive substances.

Fire worker exposure to these some of these pollutants during prescribed burning has been investigated by Reinhardt et al. in the Pacific Northwest region of the United States (2000). Results from that study showed that approximately 10% of the prescribed fire workers monitored had exposures above the American Conference of

Governmental Industrial Hygienists Threshold Limit Value (ACGIH TLV) of 3 mg m^{-3} for respirable particulate matter and about 5% had exposures above the U.S. Occupational Safety and Health Administration Permissible Exposure Limit (OSHA PEL) of 5 mg m^{-3}. Another study conducted in 1988 at two locations in Georgia found that prescribed fire workers were exposed to between 0.2 mg m^{-3} and 3.7 mg m^{-3} of particulate matter with aerodynamic diameter <2.3 μm throughout a work shift (McMahon and Bush, 1992). Fire worker exposure to these some of these pollutants during wildland fire has also been investigated in the Pacific Northwest region of the United States, but exposure levels have been shown to be lower overall and to have different determinants (Reinhardt et al., 2000).

Long-term epidemiological data relating occupational smoke exposure and health effects is unavailable for forest fire workers (Reinhardt et al., 2000). Ambient $PM_{2.5}$ levels have been implicated in numerous studies as being associated with increased incidence of mortality and morbidity in numerous populations, as discussed above. Little is known, however regarding the short-term and long-term effects on pulmonary function of exposure to pollutants in wood smoke, including suspended particles. Future research focuses include relating personal levels of fine particulate matter ($PM_{2.5}$) to various short-term and long-term health effects in individuals.

The purpose of the research addressed in Chapter 3 was to investigate $PM_{2.5}$ exposure concentrations of prescribed fire workers at the Savannah River Site (SRS) and Oconee National Forest (ONF) and to identify work activities or other factors most likely to contribute to increased exposure to $PM_{2.5}$. This information can be used in the

design of future studies that address exposure to $PM_{2.5}$ and other pollutants with short-term and long- term measures of respiratory health, such as spirometric indices or development of respiratory disease. The personal exposure concentrations of prescribed fire workers were also compared to ambient/background levels measured throughout the burning period; a comparison of the sampling methods used to collect the ambient/background and personal samples is also presented.

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CHAPTER 3

FINE PARTICLE EXPOSURE OF PRESCRIBED FIRE WORKERS IN THE SOUTHEASTERN UNITED STATES[1]

[1]Yanosky, J.D. and MacIntosh, D.L. To be submitted to (to be determined).

Abstract

Prescribed fire is an established practice in forestry management and its benefits widely recognized. Ambient levels of particulate matter in air, especially the fine particle component ($PM_{2.5}$; defined as particles in air with aerodynamic diameter <2.5μm), have been associated with increases in mortality and morbidity in numerous studies. Workers conducting prescribed burning activities often encounter high concentrations of wood smoke and as a result concern has arisen over levels of occupational exposure to pollutants that comprise wood smoke. Personal $PM_{2.5}$ exposure concentrations and work activities of prescribed fire workers were measured at the Savannah River Site (SRS) and the Oconee National Forest (ONF). Non-impacted ambient concentrations were also measured with a U.S. EPA Federal Reference Method (FRM) $PM_{2.5}$ sampler as an estimate of background concentrations during burn activities. The personal $PM_{2.5}$ concentrations measured during prescribed burning activities ranged from 64.3 μg m^{-3} to 5354.9 μg m^{-3} with mean and standard deviation 1156.1 μg m^{-3} and 1091.1 μg m^{-3}, respectively. The background/ambient $PM_{2.5}$ concentrations on days when prescribed burns were performed ranged from 5.0 μg m^{-3} to 18.5 μg m^{-3} with mean and standard deviation 10.4 μg m^{-3} and 4.4 μg m^{-3}, respectively. Significant relationships between personal $PM_{2.5}$ concentrations and worker activities were not found for any specific prescribed fire worker activities. A comparison of the sampling methods used to collect the personal and ambient/background samples is also presented; no significant systematic or proportional

bias was found between the FRM and KTL and the two methods agreed well (R^2=0.994).

Introduction

Prescribed fire has been an accepted forest management practice in the southeastern United States for a number of years. Prescribed fire activities are a source of numerous pollutants to the atmosphere and can have a significant impact on air quality of a region (Sandberg et al., 1990). Prescribed fire activities are an essential part of an effective forest management program for many reasons including the prevention of uncontrolled wildfires (Glitzenstien et al., 1995).

Fire workers engaged in prescribed burning practices are likely to be exposed to high levels of $PM_{2.5}$ as they often encounter smoky conditions. Wood smoke is known to contain numerous chemicals, in gaseous, liquid, and solid form. The primary health threats in wood smoke from prescribed burning identified by Reinhardt et al. (2000) are carbon monoxide, acrolein, formaldehyde, and respirable particulate matter. Other pollutants of concern include numerous other organic compounds, fractions of particulate matter of smaller aerodynamic size ($PM_{2.5}$, PM_1, etc.), polynuclear aromatic hydrocarbons, nitrogen and sulfur oxides, and possibly pesticide and herbicide residues and crystalline silica. Ambient fine particulate matter (particles with aerodynamic diameter <2.5 μm) levels have been implicated in numerous studies as being associated with increased incidence of mortality and morbidity in numerous populations (Schwartz et al., 1996; Neas et al., 1999; Korrick et al., 1998; Peters, 2000). Future research focuses include relating personal levels of fine particulate matter ($PM_{2.5}$) to various

short-term and long-term health effects in individuals. Investigation of $PM_{2.5}$ exposure-effect relationships in highly exposed populations such as prescribed fire workers may be an efficient means of obtaining improved knowledge about this occupational and public health issue.

A standardized method of measuring $PM_{2.5}$, the U.S. EPA Federal Reference Method (FRM) for $PM_{2.5}$, has been developed (U.S. EPA, 1997c) and is in widespread use across the United States. The FRM samplers are designed for use in ambient air and have undergone extensive comparison to other methods (U.S. EPA, 1997b; Babich et al, 2000; Yanosky and MacIntosh, 2001).

The purpose of the present research is to investigate exposure concentrations of prescribed fire workers and to identify work activities most likely to contribute to increased exposure to $PM_{2.5}$. We used the BGI, Inc. GK2.05(KTL) Respirable/Thoracic cyclone (KTL) to obtain time-integrated personal air $PM_{2.5}$ measures. These data will provide preliminary information on occupational exposure levels of $PM_{2.5}$ to prescribed fire workers necessary for a long-term prescribed fire worker health study.

The KTL is a relatively new sampling inlet designed for use in a study of non-occupational pollutant exposure of adults in Europe: the EXPOLIS study (Koistinen et al., 1999). We are not aware that its performance has been evaluated at the high (e.g., 1 mg m^{-3}) exposure levels anticipated for this population. Due to these high concentrations of $PM_{2.5}$, the different composition of wood smoke from ambient air (likely including greater relative abundance of organic components), and the lack of information regarding the comparability of these different sampling methods under

these conditions, a secondary objective of this work was to investigate the comparability of the sampling methods to assure that the results from the personal samplers are not biased compared to FRM measurements.

Methods

Sample collection

Ambient and personal levels of $PM_{2.5}$ were measured during prescribed burning activities at the U.S. Department of Energy Savannah River Site (SRS) near Aiken, SC and at the U.S. Department of Agriculture Oconee National Forest (ONF) near Macon, GA during the burn season (January 1 through March 31) of 2001. The BGI, Inc. GK2.05(KTL) Respirable/Thoracic cyclone (KTL; BGI, Inc. Waltham, MA, USA) was used to measure personal $PM_{2.5}$ exposure concentrations in the breathing zone of prescribed fire workers. A U.S. EPA designated Federal Reference Method (FRM) $PM_{2.5}$ sampler, the BGI, Inc. PQ200, was used to measure ambient/background $PM_{2.5}$ levels an upwind or remote location (usually 16-20 km from the burn area). Upon retrieval of the personal samplers at the end of the work shift, prescribed fire workers were asked to complete a form (referred to as fire worker log sheets) providing information on work activities for that day, including the amount of time spent during each of several classes of prescribed burning activities. These activities were holding, lighting, direct attack, mop-up, and burn boss. The fire worker log sheets also contained a few short questions regarding personal cigarette smoking, use of respiratory protection, and, for a subset of workers, amount of wood smoke encountered during the work shift.

Samplers were also collocated downwind of the prescribed burn to compare the performance of different sampler types at high smoke concentrations. Sampling inlets of the collocated KTL and FRM samplers were placed approximately 2.5 meters apart at a height of 2.0 meters from the ground. The KTL inlet was not shielded to protect from rain or to prevent wind effects at these comparison sites to better represent operational conditions when worn by a person during personal sampling. A size-resolved optical sampling method, the Grimm 1.108 Environmental Dust Monitor, was available during two of the prescribed burns and was collocated with the other samplers. The Grimm monitor was operated according to instructions from the manufacturer.

Mass concentrations for the KTL were obtained from gravimetric analysis on the filters and the sample volume calculated as the product of the sample duration and the mean of pre- and post-sampling flow rates. Mass concentrations for the FRM sampler were obtained from gravimetric analysis on the filters and the sample volume, which is logged by the FRM sampler throughout the sampling period. BGI AFC-400S personal sampling pumps were used with the KTL; internal pumps are integral to the FRM sampler. The KTL used 37 mm Zeflour filters (Gelman R2PJ047) and the FRM sampler used 47 mm Teflo filters (Gelman R2PJ047). Both filter types have a PTFE membrane and a 2.0 μm pore size; the Teflo filters also have a PMP support ring. Sample filters were loaded into filter cassettes and were transported in clean, anti-static plastic bags, and, after sampling, were transported to the analytical laboratory in a cooler containing blue ice. The maximum temperature in the cooler was logged and

was required to be less than 10 degrees Celsius for the samples to be considered valid. No samples were voided due to uncontrolled temperature during transportation.

The KTL sampler flow rate of 4.0 l min^{-1} was measured using either a calibrated Matheson FM-1051 rotameter (Matheson Instruments, Montgomeryville, PA, USA) or a Bios DryCal DC-Lite Model DCL20K (Bios International, Butler, NJ, USA). The FRM samplers were calibrated in accordance with standard procedures for the State of Georgia's Fine Particle Monitoring Network and U.S. EPA protocols (U.S. EPA, 1999). The FRM sampler flow rate of 16.67 l min^{-1}, temperature, and barometric pressure readings were calibrated and verified using a BGI, Inc. DeltaCal volumetric flow, temperature, and barometric pressure calibrator, respectively.

Gravimetric analysis

Filters were stored under controlled temperature and relative humidity conditions for at least two weeks prior to initial preweighing and for at least 48 hours prior to initial postweighing. Filters were weighed at least twice prior to sampling (preweights) and at least twice after sampling (postweights) using a Cahn C-32 Microbalance with a sensitivity of +/-1 μg following guidelines in EPA's Quality Assurance Handbook, Vol. II, Part II (1997). The tolerance for reweighing of filters was agreement to within +/-5 μg; approximately 51% and 28% of all 37 mm and 47 mm filters required reweighing (more than two preweights or postweights), respectively. Of these, postweights accounted for 85% of all filters (both 37 mm and 47 mm) that required reweighing. In the event of a reweighing, the mean of all preweights or postweights was used as the best estimate of the actual filter mass before and after

sampling, respectively. Once four separate measurements of the mass of a filter either before or after sampling were made, regardless of the precision of the measurements, no additional reweighing was performed. One 37 mm sample filter was designated as a 'loaded' (used for sampling) lab blank; its mass was observed to decrease by about 190 μg between initial postweighing (measured within 9 days of sampling) and subsequent postweighings (measured up to 60 days after sampling), suggesting loss of collected material due to volatilization at a rate of about 4 μg day^{-1}. To avoid this type of bias, filters that required replicate postweighing were postweighed within 10 days of sample collection or the initial postweighing; this was accomplished for all but three filters. Two 37 mm and two 47 mm filters were kept in the dessicator and weighed repetitively as 'clean' (not used for sampling) lab blanks. The pooled standard deviation of the two 37 mm and two 47 mm lab blank filter masses were 6.3 μg (n=84) and 2.7 μg (n=18), respectively. The mean net mass of the appropriate field blanks was subtracted from the net mass of the sample filters and were 16.5 μg and 3.4 μg for 37 mm and 47 mm filters, respectively. The limits of detection in concentration units for the KTL and FRM sampling methods were calculated as the three times the standard deviation in the appropriate field blanks and were 12.9 μg m^{-3} and 1.3 μg m^{-3}, respectively. The air density during the weighing session, nominal density of calibration masses, and density of each filter type were used to correct the balance readings for the buoyant effect of air following Koistinen et al. (1999) and Yanosky and MacIntosh (2001).

Data analysis

Data from the gravimetric analysis of filters and from the prescribed fire activity log sheets were input into Microsoft Excel 97. Quality control measures on data entry included verification of 50% of all values by a third party. Ordinary least squares regression statistics and descriptive statistics were generated using SAS 6.12 for Windows (SAS Institute, Cary, NC). Means reported are arithmetic means unless otherwise noted. Geometric means were calculated as the inverse natural log of the mean of the natural log-transformed concentrations. A significance level of 0.05 was used for all statistical tests. If more than one sample was obtained from a worker during a work shift, the average exposure concentration on a burn day for each worker was calculated as the time-weighted average of the individual sample concentrations.

The correlation of activity and exposure data from the fire worker log sheets and the personal sample concentrations was determined by testing for significant relationships between time spent during an activity and the average exposure concentration throughout the work shift. To correct for differences between the reported total time and sample duration, the reported time in each activity was adjusted by the ratio of the total time reported on the prescribed fire worker log sheet to the duration of the personal exposure (ratio referred to as the adjustment factor) to standardize the time activity values. Samples where the adjustment factor was less than 0.7 or greater than 2 were excluded from further analysis, either because the sampler malfunctioned and did not operate during the entire work shift or because of errors in reporting the time-activity information. Spearman correlation was used to test for a

significant relationship between the ranks of the work-activity and time-weighted average concentration variables and Pearson correlation was used to tests for a significant linear relationship between the variables. Additionally, the generalized F-test was used to tests for a linear relationship between the variables.

Results

Prescribed burn $PM_{2.5}$ sampling was performed on 9 burn days between January 25, 2001 and March 14, 2001 at SRS and ONF. On 8 of the days when samples were collected, prescribed burning was performed; on the remaining one day samples were collected though no burning was performed. Two types of ignition techniques were used to set the burns, rapid ignition and hand lighting. A total of 48 personal and 9 ambient background samples were obtained from 27 individual prescribed fire workers and at an upwind and/or unaffected site, respectively. Work activity questionnaires were collected from prescribed fire workers after collection of personal $PM_{2.5}$ sampling equipment for all but one of the personal samples. Additionally, 9 sets of collocated KTL and FRM samples were collected at an impacted location downwind of the prescribed fire.

On days when prescribed burning activities were performed (8 of the 9 days when monitoring was performed; 41 samples), the personal $PM_{2.5}$ concentrations ranged from 64.3 $\mu g\ m^{-3}$ to 5354.9 $\mu g\ m^{-3}$ with mean and standard deviation 1156.1 $\mu g\ m^{-3}$ and 1091.1 $\mu g\ m^{-3}$, respectively. See Figure 3-1 for cumulative distribution function of the personal $PM_{2.5}$ concentrations on burn days. The duration of the personal samples on days when burns were conducted ranged from 99 min to 539 min with mean and

standard deviation 368 min and 119 min, respectively. For the burn day personal samples, 93% of the average flow rates were within +/-20% of the nominal value. However, the remaining 7% of the personal burn-day samples had post-sampling flow rates lower than the lower limit of the above tolerance. Flow rates below the design flow of 4 l min^{-1} result in an increase in the 50% cut-point of particle size selection efficiency of the KTL cyclone. Thus, low flow rate could only overestimate the actual $PM_{2.5}$ component. Consistent with the findings of others (McMahon, 1983), size-resolved samples from a collocated Grimm 1.108 taken downwind of prescribed burns during the present study indicated that suspended particles in the sampled wood smoke consisted almost entirely of $PM_{2.5}$, most of this (95%) with an aerodynamic diameter <1 μm. No samples were voided due to low flow rates as they were unlikely to be biased substantially. The personal $PM_{2.5}$ concentrations from each individual were tested for differences among individuals using an analysis-of-variance least significant difference comparison (n=41); three workers were observed to have significantly higher exposure levels than the other prescribed fire workers though no unique work activities were identified by these individuals on days when sampling was performed. On days when prescribed burning activities were performed, the background/ambient $PM_{2.5}$ concentrations ranged from 5.0 μg m^{-3} to 18.5 μg m^{-3} with mean and standard deviation 10.4 μg m^{-3} and 4.4 μg m^{-3}, respectively. The duration of the background/ambient samples were greater than for the personal samples as the background/ambient samples were started before and ended after the work shift. The duration of the

background/ambient samples ranged from 360 min to 1442 min with mean and standard deviation 630 min and 343 min, respectively.

On days when prescribed burning activities were not performed (the remaining day out of 9 days when monitoring was performed), four personal samples were collected; the personal $PM_{2.5}$ concentrations ranged from 14.3 μg m^{-3} to 448.7 μg m^{-3}. Two of the samples resemble the background/ambient concentration (14.3 μg m^{-3} and 24.4 μg m^{-3}), though the remaining two were substantially elevated (81.4 μg m^{-3} and 448.7 μg m^{-3}). The duration of the personal samples collected on the non-burn day ranged from 440 min to 460 min with a mean of 451 min. For the four non-burn day personal samples, all of the average flow rates were within +/-20% of the nominal value. On the one day when prescribed burning activities were not performed, the background/ambient $PM_{2.5}$ concentration was 10.8 μg m^{-3} and was collected over a period of 512 min.

The personal and ambient/background sampling method comparisons show good agreement between the two methods. The range of FRM $PM_{2.5}$ concentrations measured at sites downwind of prescribed burns (n=9) was 11.4 μg m^{-3} to 713.0 μg m^{-3} with mean and standard deviation 227.0 μg m^{-3} and 285.9 μg m^{-3}, respectively. None of the collocated FRM or KTL samples at downwind sites were voided because of failure to meet quality control criteria. Ordinary least-squares regression on the collocated FRM and KTL $PM_{2.5}$ levels showed no statistically significant systematic or proportional bias (intercept and slope not significantly different from one and zero,

respectively). See Figure 3-2 for regression statistics and a scatter plot of the collocated FRM and KTL $PM_{2.5}$ concentrations.

Values for the Pearson correlation coefficient between reported time spent during a specific prescribed burning activity and time-integrated personal $PM_{2.5}$ concentrations were not statistically significant (H_0:R=0) for any activity identified on the prescribed fire worker log sheets ($p>0.3497$ for all). Values for the Spearman rank correlation coefficient were not statistically significant (H_0:R=0) for any activity identified on the prescribed fire worker log sheets ($p>0.0938$ for all). No statistically significant linear relationships between time spent in an activity and exposure concentration throughout the work shift were found using the general F-test for regression relationship ($p>0.3215$ for all). A subset of prescribed fire workers were asked to qualitatively describe the amount of smoke that they encountered during the work day (i.e., "How much wood smoke do you feel you encountered today?"). The most statistically significant linear relationship ($p=0.0164$) was found using a generalized linear model between the response to this question and personal $PM_{2.5}$ concentrations ($n=18$, $R^2=0.648$).

Discussion and Conclusions

The results in the present study are similar to those found for prescribed fire workers in other regions of the United States. In over 300 samples of prescribed fire worker exposure to respirable particulate matter taken between 1991 and 1995 at various forests in the Pacific Northwest, approximately 10% of exposures were above the TLV of 3 mg m^{-3}, about 3% were above the PEL of 5 mg m^{-3}, and the geometric

mean exposure level was 1 mg m^{-3} (Reinhardt et al., 2000). The geometric mean $PM_{2.5}$ exposure level for the burn-day personal samples in the present study was 756.4 μg m^{-3}. The results are also similar to a study by McMahon and Bush (1992) that measured prescribed fire worker exposure to particles with aerodynamic diameter <2.3 μm ($PM_{2.3}$) at two locations in Georgia. Though the range of personal exposure concentrations was larger for the present study (0.2 mg m^{-3} to 3.7 mg m^{-3} for $PM_{2.3}$ for McMahon and Bush versus 0.0643 mg m^{-3} to 5.3549 mg m^{-3} for $PM_{2.5}$ in the present study), average exposure levels were similar (median of 1.3 mg m^{-3} for McMahon and Bush versus mean of 1.1561 mg m^{-3} for the present study).

Personal exposure levels were substantially higher than background/ambient samples for all burn days and for the one non-burn day in the present study. Elevated personal levels on the non-burn day are presumed to be accurate, but could be due to non-compliance with sampling protocols (e.g., contact of the sampling inlet on ground or other dusty surface) as this type of contact was observed briefly by an investigator (JDY) for two of the personal samples. Tobacco smoke in the vicinity of the worker could have contributed to personal $PM_{2.5}$ exposure, though only one of the workers sampled reported smoking during the work-shift. In future work, data on proximity to other smokers should be obtained in addition to data on the individual's smoking habits. Finally, diesel exhaust or resuspended road dust during travelling by truck could have contributed to personal exposure concentrations, though these are contributions from these are unlikely to be large (Whitaker et al., 1999).

No specific prescribed fire activities were identified as contributing significantly to exposure, perhaps because of reporting error of the workers or lack of statistical power in the tests applied to available data (n=38 for the samples with valid adjustment factors). Though the analyses of the correlation between types of prescribed burning activities and personal $PM_{2.5}$ concentration in the present study do not reveal any particular activity as being likely to result in increased $PM_{2.5}$ exposure, other factors may be identified in the future. For example, the information provided by asking prescribed fire workers the question "How much wood smoke do you feel you encountered today?" may be useful in the future as a simple, cost and labor effective means of providing an exposure estimate. Five categories were provided as answers in the present study, ranging from "None to very little" to "A high level." With only this information, a generalized linear model was able to explain 65% of the variance in personal $PM_{2.5}$ concentrations (R^2=0.649).

The results suggest that comparing total time spent in an activity over the entire work shift and the time-integrated exposure over that work shift is not a sensitive enough method to identify specific activities that result in increased exposure, at least for sample sizes less than 40. Reinhardt et al. (2000) collected respirable particulate samples from prescribed fire workers over intervals during which only one activity was performed. Average levels across several types of activities were shown to differ by about a factor of two (from about 1 mg m^{-3} to about 2 mg m^{-3}), but no statistical evidence was presented regarding the significance of these differences by activity. The use of a time-resolved sampling method and more accurate and highly-resolved

information on specific time intervals when activities were performed would aid the identification of activities related to increased exposure, would help identify sampling errors, and would also provide information on peak concentrations during specific activities.

As expected, the mean personal exposure level is much greater than the U.S. EPA National Ambient Air Quality Standards for $PM_{2.5}$ in ambient air of 65 μg m^{-3} for a 24-hr period (U.S. EPA, 1997a), even when averaged with zero exposure for time not sampled. Though the $PM_{2.5}$ samples taken in the present study are not directly analogous to occupational standards for respirable particulate matter (aerodynamic diameter <3.5 μm for respirable particulate matter), comparison to these standards can provide limited information on the relative magnitude of the exposure concentrations, though the mass contribution from particles of aerodynamic diameter between 2.5 μm and 3.5 μm is unaccounted for. The American Conference of Governmental Industrial Hygienists Threshold Limit Value (TLV) of 3.0 mg m^{-3} for respirable dust(ACGIH, 2001) as the time-weighted average over an 8-hr work shift was exceeded for only two of the personal $PM_{2.5}$ samples (4.9% of the 41 samples; see Figure 3-1). Similarly, the U.S. Occupational Safety and Health Administration Permissible Exposure Limit (PEL) of 5.0 mg m^{-3} for respirable dust (Federal Register, 1997) was exceeded for only one (2.4%) of the personal samples (see Figure 3-1). However, these limits are intended mainly for nuisance dusts and not specifically for wood smoke. The OSHA PEL of 2.4 mg m^{-3} for respirable coal dust (Federal Register, 1997) may be more applicable to the smoke exposure in the present study because of the higher relative contribution of

organic compounds in coal dust; four of the 41 personal samples (9.8%) in the present study were above this limit (see Figure 3-1).

The sampling methods used for the personal and ambient/background $PM_{2.5}$ sampling agreed well at collocated comparison sites (R^2=0.994). These samplers have been shown to perform well when used in several indoor and ambient air comparisons (Koistinen et al., 1999; Yanosky and MacIntosh, 2001). The performance of the KTL and FRM samplers in the present study confirms that they can be used to accurately measure the $PM_{2.5}$ component of wood smoke even at highly elevated concentrations downwind of prescribed fires.

Clearly, the composition of the suspended particles and levels of other pollutants must be included in a comprehensive assessment of prescribed fire worker exposure to air pollutants in wood smoke. However, these data allow characterization of the levels of $PM_{2.5}$ in the breathing zone of prescribed fire workers that can be expected during similar prescribed burning activities in the southeastern U.S. Though none of the activities investigated in the present study appear to provide accurate and precise information on the determinants of the level of $PM_{2.5}$ exposure, significant relationships may be found for other variables such as emission factors (including terms for type, wetness, and amount of fuel), area-specific meteorological parameters, or more detailed questions regarding worker perception of smoke encountered. This study identifies prescribed fire workers in the southeastern United States as reasonable prospective participants in a long-term health study relating exposure levels to various pollutants in wood smoke (of which $PM_{2.5}$ is an important component) and health effects. A small

number of firefighters in the western United States have been studied and short-term differences in spirometric indices of lung function were found (Betchley et al.,1997). Little information is available regarding long-term respiratory health effects with concurrent measures of exposure to pollutants in wood smoke. The information in the present study can serve to aid in the design of a long-term study of a larger number of prescribed fire workers in the southeastern U.S. where actual exposure measurements and respiratory parameters (e.g., spirometric measurements, tracking of respiratory disease, etc.) are measured and detailed information on meteorological and forest fuel parameters is collected.

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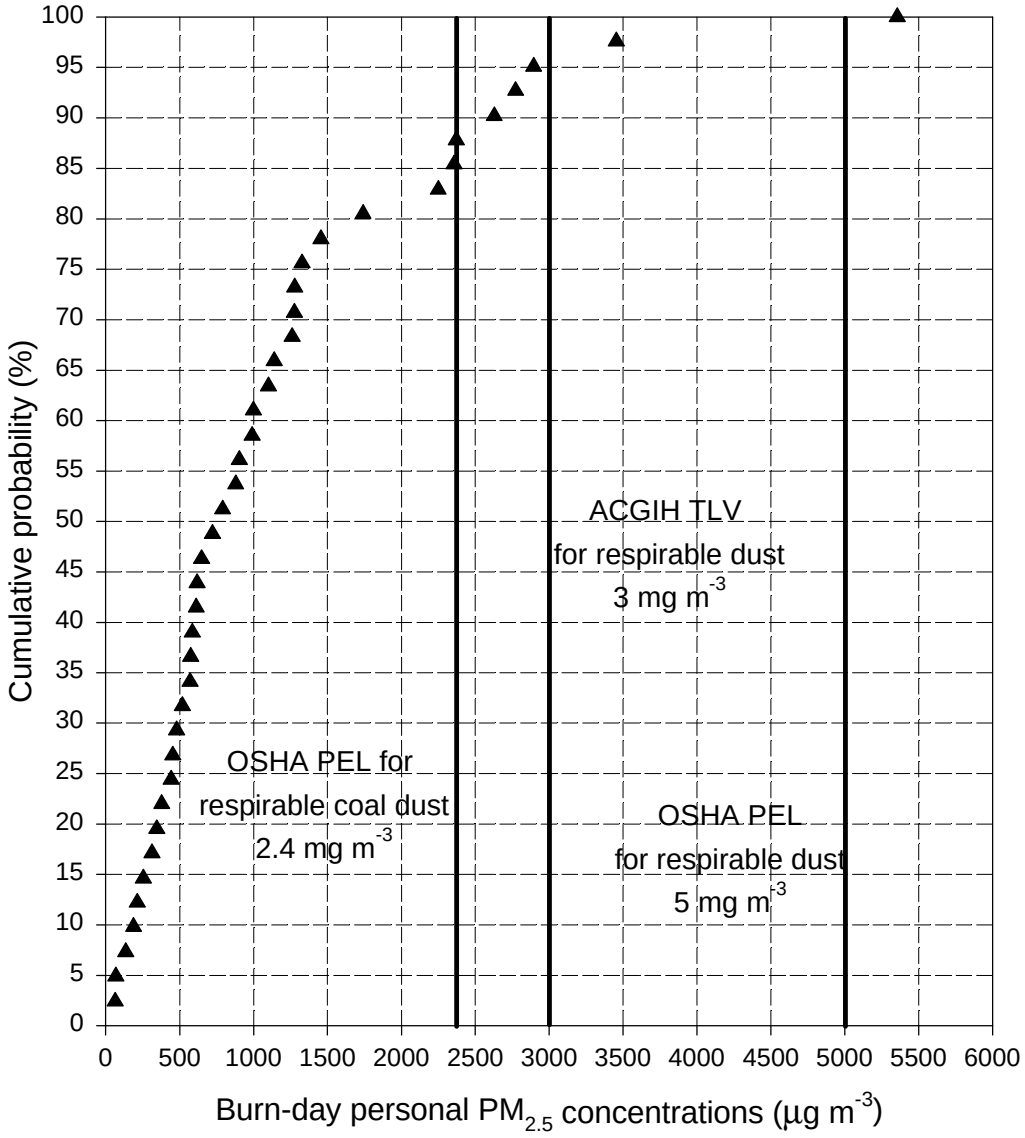

Figure 3-1. Cumulative distribution of burn-day personal $PM_{2.5}$ concentrations
Savannah River Site, Aiken, SC, and Oconee National Forest , Macon, GA
January 25, 2001 to March 14, 2001

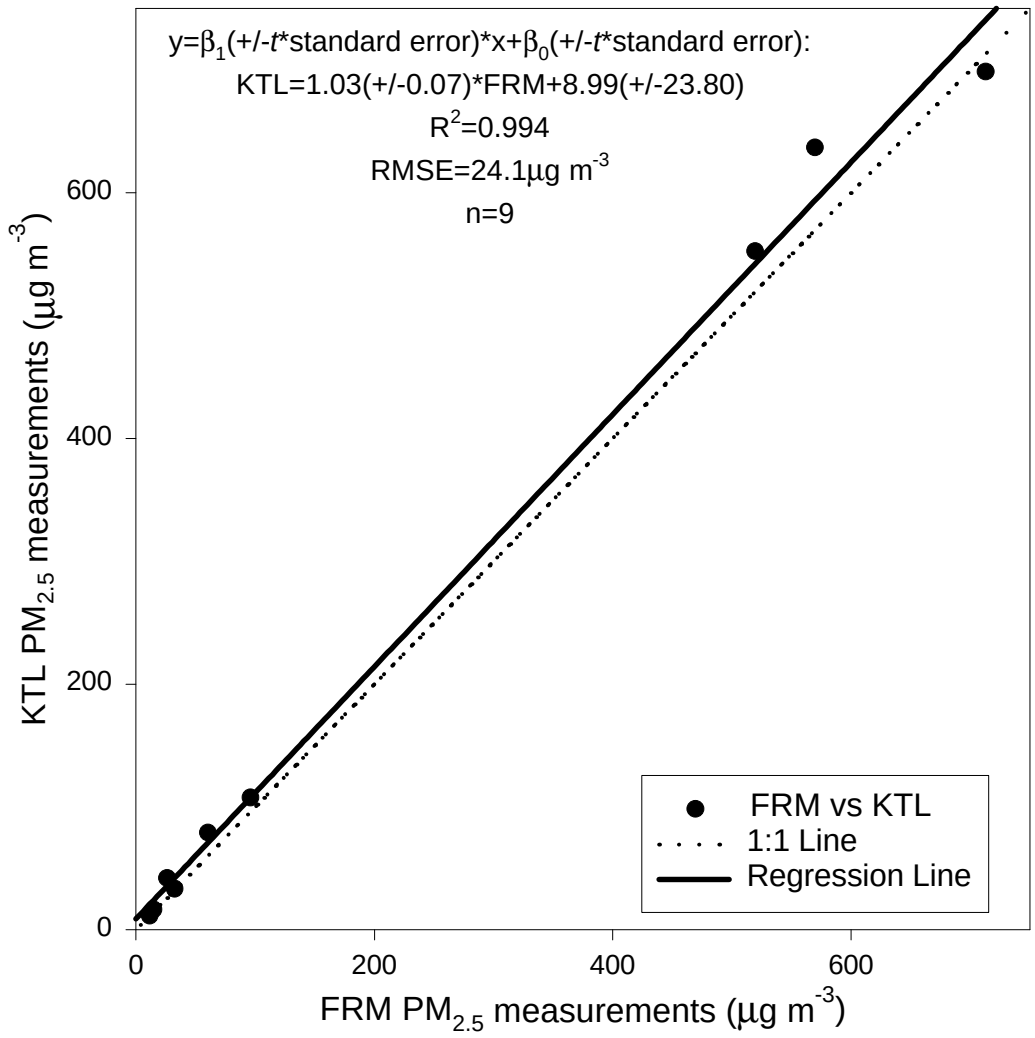

Figure 3-2. Scatter plot of FRM vs. KTL $PM_{2.5}$ measurements
at downwind burn-impacted sites
January 27, 2001 through March 14, 2001
Savannah River Site, Aiken, SC, and Oconee National Forest, Macon, GA

CHAPTER 4

A COMPARISON OF FOUR GRAVIMETRIC FINE PARTICLE SAMPLING METHODS[1]

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[1]Yanosky, J.D. and MacIntosh D.L. 2001. Accepted by the Journal of the Air and Waste Management Association. Reprinted here with permission of publisher.

ABSTRACT

A study was conducted to compare four gravimetric methods of measuring fine particle ($PM_{2.5}$) concentrations in air: the BGI, Inc. PQ200 Federal Reference Method $PM_{2.5}$ (FRM) sampler, the Harvard-Marple Impactor (HI), the BGI, Inc. GK2.05 KTL Respirable/Thoracic Cyclone (KTL), and the AirMetrics MiniVol (MiniVol). Pairs of FRM, HI, and KTL samplers and one MiniVol sampler were collocated and 24-hr integrated $PM_{2.5}$ samples were collected on 21 days from January 6, 2000 through April 9, 2000. The mean and standard deviation of $PM_{2.5}$ levels from the FRM samplers were 13.6 $\mu g/m^3$ and 6.8 $\mu g/m^3$, respectively. Significant systematic bias was found between mean concentrations from the FRM and the MiniVol (1.14 $\mu g/m^3$, p=0.0007), the HI and the MiniVol (0.85 $\mu g/m^3$, p=0.0048), and the KTL and the MiniVol (1.23 $\mu g/m^3$, p=0.0078) according to paired t-test analyses. Linear regression on all pairwise combinations of the sampler types was used to evaluate measurements made by the samplers. None of the regression intercepts was significantly different from zero and only two of the regression slopes were significantly different from one, that for the FRM and the MiniVol (β_1=0.91, 95%CI (0.83-0.99)) and the KTL and the MiniVol (β_1=0.88, 95%CI(0.78-0.98)). Regression R^2 terms were 0.96 or greater between all pairs of samplers and regression Root Mean Square Error (RMSE) terms were 1.65 $\mu g/m^3$ or less. These results suggest that the MiniVol will underestimate measurements made by the FRM, the HI, and the KTL by an amount proportional to $PM_{2.5}$ concentration. Nonetheless, these results indicate that all of the sampler types are comparable if ~10% variation on the mean levels and on individual measurement levels

is considered acceptable and the actual concentration is within the range of this study (5-35 $\mu g/m^3$).

IMPLICATIONS

Bias in measurements of personal, indoor, and ambient air fine particle levels ($PM_{2.5}$) due to the sampling method may obscure relationships among personal, indoor, and ambient concentrations. The results from this inter-comparison study show that the FRM (an ambient sampler), the HI (an indoor and ambient sampler), and the KTL (a personal sampler) provide comparable measures of $PM_{2.5}$ and can be used to evaluate ambient concentrations without substantial concern over bias in the results. The results indicate some concern about proportional bias in the measurements from the MiniVol. In some situations, the errors-in-variables problem with linear regression observed in this study may have important implications for determining regulatory equivalence of these $PM_{2.5}$ samplers.

INTRODUCTION

Exposure to fine particulate matter with aerodynamic diameter<2.5 μm ($PM_{2.5}$) has been associated with changes in morbidity and mortality rates, with changes in respiratory function, and with changes in cardiovascular hospital admissions in numerous epidemiological studies.[1,2,3,4] Considerable attention has focused on the interpretation of variation in results of such studies[5] and the relationship between ambient and personal exposure to fine particulates.[6] Several methods for measuring $PM_{2.5}$ are available and have been employed in past research. Continued research into

these issues warrants investigation into the comparability of methods of measuring different $PM_{2.5}$ exposure concentrations (e.g., ambient, indoor, personal).

EPA designated Federal Reference Method $PM_{2.5}$ samplers[7] are now in widespread use for regulatory compliance monitoring nationwide and are anticipated to be a source of exposure data for many future $PM_{2.5}$ source-to-exposure and exposure-effect studies. Alternatives to a Federal Reference Method $PM_{2.5}$ sampler, such as the Airmetrics MiniVol, are often used to measure $PM_{2.5}$ in ambient air because of their reduced size, weight, and cost. For the same reasons and others, personal and indoor $PM_{2.5}$ sampling methods such as the BGI, Inc. GK2.05(KTL) Respirable/Thoracic Cyclone (KTL) and the Harvard-Marple Impactor (HI) are used to measure personal and indoor air fine particle levels, respectively.

The Airmetrics MiniVol $PM_{2.5}$ sampler (MiniVol) has been evaluated by the U.S. EPA as part of preliminary field testing of prototype Federal Reference Method samplers for $PM_{2.5}$[8] and continues to be used.[9] The HI is a small, lightweight $PM_{2.5}$ sampler that has been widely used for ambient and indoor air sampling and its performance well characterized.[10,11,12,13] The KTL cyclone is a relatively new device intended for use as a personal monitor of $PM_{2.5}$ exposure using small, low-flow, quiet, lightweight, and portable sampling pumps.[14]

In this study, we compare simultaneous 24 hr average $PM_{2.5}$ measurements made by the BGI, Inc. PQ200 Federal Reference Method $PM_{2.5}$ sampler (FRM), the HI, the KTL, and the MiniVol. This method comparison study augments previous evaluations of these non-FRM devices by comparing each to an FRM sampler over the

range of 5-35 μg/m^3, applying corrections for the buoyant effects of air on gravimetric measurements, and reporting on the correlation, precision, and systematic and proportional bias within and between the sampling methods. If these methods are demonstrated to be comparable, then artifacts of the sampling method can be dismissed as accounting for differences between ambient, indoor, and personal $PM_{2.5}$ measurements made using these devices that are greater than reported measurement error.

METHODS

Experimental Design

$PM_{2.5}$ concentrations in ambient air were measured using four types of collocated $PM_{2.5}$ samplers. Samplers were operated on 21 days from midnight to midnight (12 A.M.-12 A.M.) from January 6, 2000 through April 9, 2000. Samplers were placed outdoors on the roof (the surface of the roof is approximately 4.5 m above ground level, sampler inlets were approximately 1.5 m above the surface) of the Environmental Health Science Building on the University of Georgia campus in Athens, Georgia. The samplers were configured along a line and approximately equally spaced over a distance of 12 m. A large tub made of high-density polyethylene was inverted and placed over the HI and KTL devices to shield them from rain and to minimize potential for wind-dependent aspiration effects. Duplicate samples were collected during each sampling period using the FRM, the HI (in the 4 L/min configuration), and the KTL samplers to allow for comparisons between samplers of the same type (within sampler comparisons) and between samplers of different type (between sampler comparisons). The sample

size was selected to detect 10% differences between mean concentrations from each sampler type as significant (α=0.05) with relatively high statistical power (1-β=0.8). Based on preliminary estimates of the RMSE between collocated samplers, the required sample size was calculated to be 16 sample periods.[15] Approximately 15% of all samples were collected as field blanks, defined as filters loaded into a sampler, transported to the sample site, returned to the laboratory following set-up of the primary samplers, stored until breakdown of the primary samples, and transported to the site and then from the site with primary samplers. The limit of detection in concentration units for each sampler type was calculated as three times the standard deviation in the net mass of the field blanks divided by the appropriate nominal sample volume.

Gravimetric Analysis

Filters were weighed twice prior to sampling and twice after sampling using a Cahn C-32 Microbalance with a sensitivity of +/-1 μg following guidelines in U.S. EPA's Quality Assurance Handbook, Vol. II, Part II.[16] The tolerance for reweighing of filters was agreement to within +/- 10 μg. Following methods described in the literature,[17] the mean net mass of the field blanks for each sampler type was subtracted from the net mass of the sample filters, though due to the minimal change in the field blank masses, this had little effect on the sample concentrations. The air density during the weighing session, nominal density of the calibration masses, and the measured density of each filter type were used to correct the balance readings of all filters (including blanks) for the buoyant effect of air following Koistinen, Kousa et al.[17] The density of each filter type was estimated by immersion in fluids of known density, using mixtures of

deionized water, ethanol and 85% phosphoric acid (phosphoric acid was used because of its greater density than water, it had no discernable effect on the filters). Filter densities were determined to be 840 kg/m^3 for 47 mm and 41 mm Teflo filters (Gelman R2PJ047 and R2PJ041), 1380 kg/m^3 for 47 mm Zeflour filters (Gelman P5PJ047), and 1180 kg/m^3 for 37 mm Zeflour filters (Gelman P5PJ037).

Data Collection

The FRM samplers and the MiniVol used 47 mm Teflo and Zeflour filters. The HI used 41 mm Teflo filters, 37 mm drain disks (Whatman 230800), 37 mm backing pads (Milipore AP10), and otherwise were operated following methods described elsewhere.[12] All valid samples collected using the KTL used 37 mm Zeflour filters (37 mm MCE filters were used on a trial basis, however no results were reported using MCE filters as they had poor gravimetric reproducibility). All four filter types have a PTFE membrane and a 2.0 μm pore size. The Teflo filters also use a PMP support ring. Before each sampling period, the impaction surfaces of the KTL, HI, and MiniVol were cleaned, and Dow 704 diffusion pump oil and Dow high vacuum grease was applied to the impaction surfaces of the HI and the MiniVol, respectively.

For the FRM samplers, sample volume is logged by the instrument throughout the sample period. For the other sampler types, sample volume was calculated as the product of the sample duration and the mean of pre and post sampling flow rates measured using an electronic flow meter (either The Gilibrator, Gilian Instrument Corp., West Caldwell, New Jersey, or a DryCal DC-Lite, Bios International, Butler, New Jersey). The FRM and MiniVol nominal flow rates of 16.67 l min^{-1} and 5.0

l min^{-1}, respectively, were obtained by internal pumps integral to the samplers. The HI and KTL nominal flow rate of 4.0 l min^{-1} was obtained by SKC, Inc. Model 224-PCXR8 pumps.

Statistical Analysis

The data were analyzed to detect both systematic and proportional bias and to assess the correlation and precision of $PM_{2.5}$ levels within and between each sampler type. A significance level of 0.05 was used for all statistical tests. For between sampler type comparisons, the mean collocated concentration per day for each sampler type was calculated as the mean of the duplicate samples (excluding the MiniVol). Paired t-tests were performed on the mass concentrations to detect systematic bias within and between each sampler type. The paired t-test evaluates the mean difference in the concurrently measured 24 hr average concentrations, so that the daily fluctuation of $PM_{2.5}$ levels is eliminated.

Ordinary least squares linear regression was performed on the $PM_{2.5}$ levels within and between each sampler type. For within sampler type comparisons, one sampler of a pair was arbitrarily designated as the independent variable and the other as the dependent variable in the regression model. For between sampler comparisons, the concentration from a single sampler was used if the result from one sampler of a pair was voided; thus the precision for some points in these comparisons is slightly lower than for the other data points. Measurement error in the independent and dependent variables of regression models can bias estimates of regression slopes[18,19]. To evaluate the effect of differential measurement error between sampler types on regression model

results, two models were fit to each pair of sampler types, one where sampler type *i* was the independent variable and sampler type *j* was the dependent variable, and a second model where sampler type *j* was the independent variable and sampler type *i* was the dependent variable. For all models, regression intercepts significantly different from zero were considered to indicate systematic bias of $PM_{2.5}$ levels between samplers. Regression slopes significantly different from one were considered to indicate proportional bias of $PM_{2.5}$ levels between samplers (bias that varies relative to concentration). The coefficient of determination (R^2) was used to describe the strength of association of measured levels between samplers. The Root Mean Square Error (RMSE) is given in units of $\mu g/m^3$ and was used to describe the precision of the sampling methods.

Additional measures of sampler precision were generated to afford comparison of our results with data from other evaluations of these samplers.[8,17] The collocated precision was calculated as the standard deviation in the differences between collocated samplers. Also, the mean and standard deviation of the absolute differences in the concentrations from different sampler types were calculated from the absolute value of the differences in the mass concentrations of collocated samplers.

RESULTS

Twenty-one batches of samples were collected every third day in cycles of approximately three weeks on and one week off from January 6, 2000 through April 9, 2000. A batch consisted of samples from two FRM samplers (duplicates), two KTL cyclones, two HI samplers, and one MiniVol. A total of 132 valid samples were

collected out of 147 attempted. The mean and standard deviation (expressed as mean (SD)) of the differences in the replicate filter weights by filter type were 2.0 (4.0), 1.6 (4.9), 1.0 (3.8), and 1.0 (3.7) μg for 47 mm Teflo, 47 mm Zeflour, 37 mm Zeflour, and 41 mm Teflo filters, respectively. Fifteen samples were voided as a result of power interruptions (3), inadequate gravimetric reproducibility of mixed cellulose ester (MCE) filters used on a trial basis with the KTL cyclones (10), and sampler malfunctions (2). The mean field blank values were 8.7, 8.1, 15.4, 11.1 μg for the FRM, KTL, HI, and MiniVol, respectively. Limits of detection were 0.4, 5.6, 5.6, and 3.9 $\mu g/m^3$ for the FRM, the HI, the KTL, and the MiniVol, respectively. Three samples were marginally below the appropriate limit of detection, but were included in the results presented here as their omission did not substantively alter our findings.

The range of the 24 hr average $PM_{2.5}$ concentrations measured by the FRM was 5.5 $\mu g/m^3$ to 33.3 $\mu g/m^3$. Other simple statistics on the mass concentrations are listed in Table 4-1. The top section includes data collected from all days when the FRM, HI, and MiniVol were operated; the lower section includes data from only the last sixteen days of sampling when the KTL cyclone was used with the other samplers.

See Table 4-2 for a summary of the within sampler comparisons. The values listed under 'Mean Difference' are the mean differences in the sampler type in the 'X' column minus the sampler type in the 'Y' column. None of the within sampler mean differences were significantly different from zero using the paired t-test. None of the within sampler intercept and slope estimates were significantly different from zero and

one, respectively. The lowest R^2 among the within sampler comparisons was 0.96 and the largest RMSE was between the KTL cyclones at 1.65 $\mu g/m^3$.

See Table 4-3 for a summary of the between sampler comparisons. The variables in the "X" and "Y" columns listed as "Mean" are the mean collocated concentrations per day for each sampler type for which there were duplicate samples. If one of the duplicate samples was voided, the valid sample was used in place of the mean collocated concentration per day. The top section includes comparisons for evaluating regulatory compliance between each of the non-FRM samplers to the FRM sampler where the former is specified as the dependent variable in the regression model and the latter is the independent variable. The middle section includes comparisons shown in the top section although the designation of independent and dependent variable in the regression model is reversed. These regressions are appropriate for use in predicting FRM concentrations from data obtained using the other sampler types. The bottom section includes comparisons between the non-FRM sampler types. A significant difference was found using the paired t-test between the mean collocated FRM and the MiniVol ($p=0.0007$), between the mean collocated HI and the MiniVol ($p=0.0048$), and between the mean collocated KTL and the MiniVol ($p=0.0078$). In each comparison, the mean from the MiniVol was less than the mean from the other sampler types, suggesting that the MiniVol measurements are systematically biased low. None of the between sampler intercepts were significantly different from zero ($H_o:\beta_0=0$). A between sampler slope significantly different from one ($H_o:\beta_1=1$) was found for the mean collocated FRM (independent variable) versus the MiniVol

(dependent variable) at 0.91 (95%CI (0.83-0.99), p=0.0331), but was not significant when the sampler types were reversed in the regression model. Also, a between sampler slope significantly different from one (H_o:β_1=1) was found for the mean collocated KTL versus the MiniVol at 0.88 (95%CI (0.78-0.98), p=0.0348), but again the slope was not significant when the sampler types were reversed in the regression model. The lowest R^2 among the between sampler comparisons was between several of the sampler types at 0.96. The highest between sampler RMSE was between the mean collocated HI and mean collocated KTL at 1.60 $\mu g/m^3$.

DISCUSSION AND SUMMARY

Substantial bias in $PM_{2.5}$ measurements due only to the type of sampler used might erroneously influence exposure estimates and thereby obscure source-to-exposure or exposure-effect relationships examined in future studies. In the present study, all of the within sampler comparisons show good agreement (i.e., no substantial systematic or proportional bias, and high correlation and precision). From Table 4-2, the two FRM samplers were the most highly correlated (R^2>0.99), and the pairs of HI and KTL samplers were equally well correlated (R^2=0.96). The higher correlation in the FRM methods may be due in part to increased precision on the gravimetric determinations of the net mass of the FRM filters relative to the precision of the microbalance as the FRM samplers have a higher flow rate (16.67 l min^{-1}), to more efficient particle size selection characteristics of the Well Impactor Ninety-Six (WINS) impactor used by the FRM samplers, or to constant volumetric flow control used by the FRM pump in contrast to the other pumps.

The mean concentration from the MiniVol across all sampling days was significantly lower than means from the other sampler types using the paired t-test. The only regression slopes significantly different from one were given by the mean collocated FRM versus the MiniVol and the mean collocated KTL versus the MiniVol, but only when the MiniVol was considered the dependent variable in each comparison (i.e., Y=MiniVol for both). These results suggest that some proportional bias exists in the MiniVol measurements (i.e., that the difference between the concentrations given by these devices increases as concentration increases). However, when the MiniVol was identified as the independent variable (i.e., X=MiniVol), the regression slopes between it and any of the other sampler types were no longer significant. Thus, in these cases, the selection of which variable is considered the independent and dependent variable influences the outcome of statistical tests on the regression slopes. This outcome likely results from different degrees of measurement error between sampler types, a situation in regression theory commonly referred to as the error-in-variables problem, which has been shown to bias the regression slope low[18,19,20,].

A U.S. EPA document entitled "Prototype $PM_{2.5}$ Federal Reference Method Field Studies Report"[8] reported a comparison of twenty collocated $PM_{2.5}$ samples from a then prototype FRM sampler (the Anderson RAAS) and the MiniVol in Phoenix, AZ. The results from the EPA comparison show a mean difference between the FRM sampler and the MiniVol of 1.70 $\mu g/m^3$, a collocated precision (standard deviation of between sampler differences) of 2.43 $\mu g/m^3$, and, from weighted least squares regression, a slope of 0.90, intercept of 3.24, and R^2=0.91. In the present study, we

found a mean difference between the FRM sampler and the MiniVol of 1.14 μg/m^3, a standard deviation of the difference of 1.30 μg/m^3, a slope of 0.91, an intercept of 0.07, and an R^2=0.97 for twenty-one collocated sample pairs, although ordinary least squares regression was used. The estimates of the regression slope for the FRM versus the MiniVol are similar between the two studies, further supporting the observed tendency of the MiniVol to underestimate the FRM as concentrations increase.

The results from the KTL cyclones agree well with those from the FRM samplers. However, the KTL cyclone had the worst precision (RMSE=1.65 μg/m^3 in Table 4-2) of any of the sampler types in this study, though the intercept and slope estimates from the mean collocated FRM versus the mean collocated KTL were not significantly different from zero and one, respectively. Koistinen et al.[17] reported that the mean concentration using the KTL was 4% lower than that using the EPA WINS impactor for eleven sampling periods (ten 24 hr periods and one 46 hr period), and that the mean absolute difference for eighteen collocated KTL sampler pairs was 2.1 μg/m^3 with a standard deviation of 2.0 μg/m^3. In the present study, we found that the mean KTL concentration (15.1 μg/m^3) across the sixteen days when the KTL cyclones were used agreed to within <1% of the mean FRM concentration (15.0 μg/m^3) for the same period, and that the mean absolute difference for fifteen collocated KTL sampler pairs was 1.2 μg/m^3 with a standard deviation of 1.08 μg/m^3. Considered together, these results suggest that on average the KTL cyclone provides comparable results to the

FRM, although there may be more error in any one measurement from the KTL cyclone than from the FRM.

The results from the HI are also comparable to those from the FRM as no systematic bias was indicated by the paired t-test and the intercepts and regression slopes were not significantly different from 0.0 $\mu g/m^3$ and 1.0 $\mu g/m^3$, respectively. In addition, the correlation between the HI and the FRM is as great as that between the two HI samplers (R^2=0.96 for both), and the precision is comparable between the mean collocated FRM versus the mean collocated HI (RMSE=1.32 $\mu g/m^3$ from Table 4-3) and one HI sampler versus the other (RMSE=1.44 $\mu g/m^3$ from Table 4-2). The results of the HI to FRM comparison found in the present study are comparable to those reported elsewhere for monitoring performed at two locations in California[21], though a slightly better correlation was reported there than in the present study. Allen et al.[12] reported that the standard deviation in the difference between thirty-eight collocated HI $PM_{2.5}$ sample pairs collected in Chicago between October and November, 1997, was 0.33 $\mu g/m^3$ at a mean concentration of 17.6 $\mu g/m^3$ with R^2>0.99. In this study, we found that the standard deviation in the difference between twenty-one collocated HI $PM_{2.5}$ sample pairs was 1.50 $\mu g/m^3$ at a mean concentration of 13.3 $\mu g/m^3$ with an R^2=0.96. The decrease in overall performance of the HI that we observed may be due to the lower flow rate of the impactors used in our study (4.0 l min^{-1} versus 10 l min^{-1}), which resulted in less mass collected on the filters and therefore less gravimetric precision relative to the precision of the microbalance.

The U.S. EPA has established that $PM_{2.5}$ sampling methods that have demonstrated both comparability and similar measurement precision to the Federal Reference Method for $PM_{2.5}$ may attain Federal Equivalent Method (FEM) status. The performance requirements for FEM status given by linear regression as listed in Table C-4 to Subpart C of 40CFR53[7] are a regression slope of 1+/-0.05, intercept of 0+/-1, a correlation coefficient (equal to the square root of R^2) greater than or equal to 0.97, and a precision in the replicate FRM measurements of less than 2 $\mu g/m^3$ or 5%. The regulations do not specify whether the FRM or candidate FEM samplers must be considered the independent or dependent variables in the regression model, though it is intuitive to consider the FRM the independent variable. Although this study was not designed to assess FEM status as defined in 40CFR Part 53, it is interesting to consider the results from this comparability study to the FEM criteria. The HI data in this study meet all of the above requirements except for the regression slope of 0.94 when the mean collocated FRM and the mean collocated HI are regressed as the independent and dependent variables, respectively. However, when the mean collocated HI is considered the independent variable, the slope of 1.03 is within the tolerance specified in Table C-4 to Subpart C of 40CFR53[7]. Thus, the selection of which sampler type is considered the independent and dependent variable in the regression model may influence the determination of equivalency status due to the error-in-variables problem noted above.

The KTL data in this study meet the FEM requirements as the regression estimates and correlation coefficient are within the specified tolerances. The results

from the MiniVol in this study meet all of the above requirements except for the regression slope (0.91 when the mean collocated FRM and the MiniVol are regressed as the independent and dependent variables, respectively, and 1.06 when the model variables are reversed). The FEM requirements for the precision of the FRM samplers were met, as the standard deviation in the differences between collocated FRM samplers was 0.38 $\mu g/m^3$. We reiterate that comparison of our results to FEM criteria is presented to provide another method of evaluating the performance of the non-FRM samplers. The FEM status of those samplers should not be inferred from these data as some requirements for demonstration of FEM status were not met by our study design, for example, triplicate measurements from each sampler type per day, or a minimum number of sampling days within a specified concentration range.[7] In addition, data from other locations and times of year would be valuable for assessing comparability of these fine particle collection devices.

CONCLUSIONS

This study presents additional information on the comparability of $PM_{2.5}$ measurements made by the FRM, the HI, the KTL and the MiniVol. The results suggest that the MiniVol may underestimate the concentration given by the other sampler types as concentrations increase. However, mean concentrations agree to within 9% between each sampler type in the range of 5-35 $\mu g/m^3$. In addition, the results indicate that a single measurement from one of the non-FRM devices is likely to be within 10% (largest RMSE for the between sampler comparisons with the FRM of 1.32 $\mu g/m^3$ divided by mean FRM concentration of 13.6 $\mu g/m^3$) of a corresponding FRM

measurement. Thus, from these results, all of the sampler types are comparable if ~10% variation on the mean levels and on individual measurement levels is considered acceptable and the actual concentration measured is within this range.

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Table 4-1. Simple statistics on 24-hr average $PM_{2.5}$ mass concentrations from the four collocated sampler types

Sampler type	n	Mean (mg/m^3)	SD (mg/m3)	Interquartile Range (mg/m3)	Min. (mg/m3)	Max. (mg/m3)
Statistics generated using data from all 21 days						
FRM	21	13.6	6.8	5.5	5.5	33.3
HI	21	13.3	6.5	6.1	5.7	30.2
MiniVol	21	12.4	6.3	5.6	4.5	28.2
Statistics generated using data from the last 16 days when valid data were collected from all four sampler types						
FRM	16	15.0	7.2	7.7	5.5	33.3
HI	16	14.6	6.9	7.5	5.7	30.2
KTL	16	15.1	7.3	7.9	6.1	33.8
MiniVol	16	13.9	6.6	8.1	4.5	28.2

Abbreviations: n=number of samples in comparison, SD=standard deviation, Min.=minimum value, Max.=maximum value

Table 4-2. Summary of within sampler type comparisons

Sampler Types			Mean Difference	SD Difference	Regression Model			
X[a]	Y[b]	n	(mg/m^3)	(mg/m^3)	Intercept (b_0) (+/- 2.0SE)	Slope (b_1) (+/- 2.0SE)	R^2	RMSE (mg/m^3)
FRM 1	**FRM 2**	19	0.02	0.38	-0.11 (+/- 0.41)	1.01 (+/- 0.03)	>0.99	0.39
HI 1	**HI 2**	19	-0.14	1.50	-0.34 (+/- 1.60)	1.04 (+/- 0.11)	0.96	1.52
KTL 1	**KTL 2**	15	0.23	1.59	-0.42 (+/- 2.00)	1.01 (+/- 0.12)	0.96	1.65

[a]: Corresponds to X in the model $Y=\beta_0+\beta_1(X)$

[b]: Corresponds to Y in the model $Y=\beta_0+\beta_1(X)$

Abbreviations: n=number of samples in comparison, SD=standard deviation, SE=standard error, R^2=coefficient of determination, RMSE=root mean square error

Note: Within sampler type comparisons were not possible for the MiniVol as only one sample was collected per day.

Table 4-3. Summary of between sampler comparisons

Sampler Types			Mean Difference	SD Difference	Regression Model			
					Intercept (b_0)	Slope (b_1)		RMSE
X[a]	**Y**[b]	**n**	**(µg/m3)**	**(µg/m^3)**	**(+/- 2.0SE)**	**(+/- 2.0SE)**	**R^2**	**(µg/m3)**
Comparisons for regulatory equivalence using FRM as independent variable (i.e., "X=mean FRM")								
Mean FRM	**Mean HI**	21	0.29	1.36	0.58 (+/- 1.31)	0.94 (+/- 0.09)	0.96	1.32
Mean FRM	**Mean KTL**	16	-0.13	0.82	-0.01 (+/- 1.00)	1.01 (+/- 0.06)	0.99	0.84
Mean FRM	**MiniVol**	21	1.14[c]	1.30	0.07(+/- 1.17)	0.91 (+/- 0.08)[d]	0.97	1.18
Comparisons for predicting FRM concentrations from data from other sampler types (i.e., "Y=mean FRM")								
Mean HI	**Mean FRM**	21	-0.29	1.36	-0.06 (+/- 1.40)	1.03 (+/- 0.10)	0.96	1.38
Mean KTL	**Mean FRM**	16	0.13	0.82	0.20 (+/- 0.98)	0.98 (+/- 0.06)	0.99	0.83
MiniVol	**Mean FRM**	21	-1.14[c]	1.30	0.38 (+/- 1.25)	1.06 (+/- 0.09)	0.97	1.27
Other comparisons								
Mean HI	**Mean KTL**	16	-0.47	1.56	-0.08 (+/- 1.93)	1.04 (+/- 0.12)	0.96	1.60
Mean KTL	**Mean HI**	16	0.47	1.56	0.73 (+/- 1.78)	0.92 (+/- 0.11)	0.96	1.51
Mean HI	**MiniVol**	21	0.85[c]	1.23	-0.23 (+/- 1.24)	0.95 (+/- 0.08)	0.96	1.23
MiniVol	**Mean HI**	21	-0.85[c]	1.23	0.70 (+/- 1.24)	1.01 (+/- 0.09)	0.96	1.26
Mean KTL	**MiniVol**	16	1.23[c]	1.61	0.53 (+/- 1.67)	0.88 (+/- 0.10)[d]	0.96	1.41
MiniVol	**Mean KTL**	16	-1.23[c]	1.61	0.07 (+/- 1.87)	1.08 (+/- 0.12)	0.96	1.57

[a]: Corresponds to X in the model $Y=\beta_0+\beta_1(X)$

[b]: Corresponds to Y in the model $Y=\beta_0+\beta_1(X)$

[c]: Significantly different from zero ($\alpha=0.05$)

[d]: Slope estimates significantly different from one ($\alpha=0.05$)

Abbreviations: n=number of samples in comparison, SD=standard deviation, SE=standard error, R^2=coefficient of determination, RMSE=root mean square error

CHAPTER 5

A COMPARISON OF TWO DIRECT-READING AEROSOL MONITORS WITH THE FEDERAL REFERENCE METHOD FOR $PM_{2.5}$[1]

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[1]Yanosky, J.D., Williams, P.L., and MacIntosh, D.L. 2001. Submitted to Atmospheric Environment, 04/09/01.

Abstract

Two types of direct-reading aerosol monitoring devices, the TSI, Inc. Model 3320 Aerodynamic Particle Sizer (APS), and the TSI, Inc. Model 8520 DustTrak Aerosol Monitor (DustTrak), were collocated indoors with a U.S. EPA designated Federal Reference Method (FRM) $PM_{2.5}$ sampler, the BGI, Inc. PQ200, to assess the comparability of the sampling methods. Simultaneous 24-hour samples were collected from two APS instruments, one DustTrak and one FRM sampler for 20 sample periods. The 30-minute average concentrations during the 24-hour sample periods were also logged and compared for the APS and DustTrak. Statistical analysis on the mass concentrations obtained from each sampler type included paired t-tests and linear regression. The 24-hour average $PM_{2.5}$ levels from the FRM samplers were approximately normally distributed and ranged from 5.0 $\mu g\ m^{-3}$ to 20.4 $\mu g\ m^{-3}$ with mean and standard deviation 11.4 $\mu g\ m^{-3}$ and 4.0 $\mu g\ m^{-3}$, respectively. The 24-hour average DustTrak levels are well correlated with FRM levels (R^2=0.859) but show significant proportional bias (β_1=2.57, $p<0.0001$). The 24-hour average mean collocated APS levels are less highly correlated with the FRM (R^2=0.592) and do not show statistically significant proportional bias. The 30-minute average levels between the two APS instruments show a high correlation (R^2=0.979) but significant proportional bias (β_1=1.31, $p<0.0001$). The results suggest that though the DustTrak provides precise measurements of $PM_{2.5}$, the accuracy of the measurements compared to the FRM can be improved through statistical adjustment. In contrast, APS $PM_{2.5}$

measurements are less precise and less accurate compared to the FRM and therefore results from the APS should be interpreted with caution.

1. Introduction

Exposure to fine particulate matter with aerodynamic diameter <2.5 μm ($PM_{2.5}$) in ambient air has been associated with changes in morbidity and mortality rates, with changes in respiratory function, and with changes in cardiovascular hospital admissions in numerous epidemiological studies (Schwartz, Dockery, and Neas, 1996; Neas, L.M., 1999; Korrick, SA., 1998; Peters, A., 2000). Outdoor particles (especially the finer size fractions such as $PM_{2.5}$) have been shown to readily penetrate indoors, with resulting indoor $PM_{2.5}$ levels resembling outdoor levels (Brauer et al., 1989). Recent advances in aerosol instrument technology have made it possible to measure and log aerosol concentrations in real time, that is, to collect time-resolved information on both the number and mass concentration as well as the size distribution of an aerosol. Such instruments offer insight into particulate levels during time series of short intervals (e.g., 5-min); this information cannot be obtained by gravimetric methods using current technology. Real-time measurements can also aid in identification of activities that contribute significantly to indoor particulate exposure (Abt et al., 2000). However, techniques used by real time aerosol monitors differ substantially from traditional gravimetric methods and therefore use of these new methods warrants investigation into the comparability of the methods in measuring $PM_{2.5}$ levels.

U.S. EPA designated Federal Reference Method (FRM) $PM_{2.5}$ samplers are now in widespread use for regulatory compliance ambient air monitoring nationwide and are

anticipated to be a source of exposure data for many future $PM_{2.5}$ source-to-exposure and exposure-effect studies. The TSI, Inc. Model 3320 Aerodynamic Particle Sizer (APS) is intended for indoor use and provides time-resolved mass concentrations as well as information on the size distribution of the sampled aerosol. The TSI, Inc. Model 8520 DustTrak Aerosol Monitor (DustTrak) is a small and portable direct-reading aerosol monitor that is intended for indoor or outdoor use (an enclosure is available from the manufacturer for monitoring ambient air; this enclosure was not used as part of the present study) as a survey instrument or as an area sampler.

In the present study, we compare (1) simultaneous 24-hour average $PM_{2.5}$ measurements made by the FRM, the DustTrak, and the APS, and (2) simultaneous 30-minute average measurements made by the DustTrak and the APS. The present study augments existing information concerning the performance of the non-FRM devices by comparing each to an FRM sampler over the range of 5.0-20.4 $\mu g\ m^{-3}$, applying corrections for the buoyant effects of air on gravimetric measurements, and reporting on the correlation, precision, and systematic and proportional bias within and between the sampling methods.

2. Methods

2.1 Experimental design

$PM_{2.5}$ concentrations in indoor air were measured using three types of collocated $PM_{2.5}$ samplers, the BGI, Inc. PQ200 FRM $PM_{2.5}$ sampler (BGI, Inc. Waltham, MA, USA), APS, and DustTrak (TSI, Inc. St. Paul, MN, USA). Samplers were operated on 20 days from midnight to midnight (12 A.M.-12 A.M.) from January 6, 2000 through

April 9, 2000. Samplers were placed indoors in the Environmental Health Science Building on the University of Georgia campus in Athens, Georgia, USA. Two APS samplers were available for the present study and the pair was used to obtain duplicate measurements from this type of sampling method. The set of duplicate data generated from the two APS samplers afforded analysis of within sampler type variability. The 24-hour average concentrations were obtained from the FRM, the DustTrak, and the two APS samplers. Also, the 30-minute average concentrations during the 24-hour sample periods were obtained from the DustTrak and the two APS samplers. Approximately 20% of all FRM samples were collected as field blanks. The limit of detection in concentration units of 0.4 μg m^{-3} for the FRM sampler was calculated as three times the standard deviation in the net mass of the field blanks divided by the nominal FRM sample volume of 24 m^3. None of the FRM samples were below the limit of detection.

2.2 Gravimetric analysis

Filters for the FRM sampler were weighed twice prior to sampling and twice after sampling using a Cahn C-32 Microbalance (Thermo Orion, Beverly, MA, USA) with a sensitivity of +/-1 μg following guidelines in the U. S. EPA Quality Assurance Handbook, Vol. II, Part II (1997). The tolerance for reweighing of filters was agreement to within +/-10 μg; no replicate measurements of filter mass were outside this tolerance. The mean and standard deviation of the replicate filter masses for all filters were 0.5 μg and 4.4 μg, respectively. The mean net mass of the field blanks (8.7 μg) was subtracted from the net mass of the sample filters. The air density during the

weighing session, nominal density of calibration masses, and density of each filter type were used to correct the balance readings for the buoyant effect of air following Koistinen et al.(1999) and Yanosky and MacIntosh (2001).

2.3 Data collection

Mass concentrations for the FRM sampler were obtained from gravimetric analysis on the filters and the sample volume which is logged by the sampler throughout the sampling period. The FRM sampler used 47 mm Teflo and Zeflour filters (Gelman R2PJ047 and P5PJ047). Both filter types have a PTFE membrane and a 2.0 μm pore size; the Teflo filters also have a PMP support ring. The FRM and DustTrak sampler flow rates were calibrated using an electronic flow meter (The Gilibrator, Gilian Instrument Corp., West Caldwell, New Jersey). The Coefficient-of-Variation (CV) logged by the FRM sampler was required to be less than 4% and the DustTrak flow required to be within

+/-0.1 l min^{-1} of the nominal value after sampling for the FRM and DustTrak samples to be considered valid, respectively. The FRM Well-Impactor Ninety-Six (WINS) was replaced after every third 24-hour sampling period. The APS mass concentrations were calculated by the instrument using a density factor of 1 g ml^{-1} (unit density); the DustTrak mass concentrations were calculated using the factory default calibration factor of 1.0.

The APS is a time-of-flight spectrometer that arrives at mass concentrations by optically counting the number of particles able to be sized into categories (52 channels, 0.3-20 μm size range; mass cumulative concentration for PM<2.458 μm used for

$PM_{2.5}$). No external calibration of the internal flow sensor or of the particle size classification of the APS was performed after calibration by the manufacturer using polystyrene latex spheres of known density in August 1999. No impactor or inlet accessories were used with the APS as part of the present study.

The DustTrak uses 90-degree light scattering to measure the mass concentration of particles in an air stream that passes through an impactor assembly. The DustTrak was calibrated by the manufacturer using Arizona road dust (particle size range 0.1μm to 10 μm) in March 1999. The DustTrak inlet designed by the manufacturer for $PM_{2.5}$ monitoring was used for this test. Before each 24-hour sampling period, the DustTrak impaction plate was cleaned and greased and the DustTrak zero point was verified and/or reset by attaching a HEPA filter according to the manufacturer's instructions. The FRM, APS, and DustTrak nominal flow rates of 16.67 l min^{-1}, 5.0 l min^{-1}, and 1.7 l min^{-1}, respectively, were obtained by internal pumps integral to the samplers.

2.4 Statistical analysis

The data were analyzed to detect both systematic and proportional bias and to assess the correlation and precision of $PM_{2.5}$ levels within and between each sampler type. A significance level of 0.05 was used for all statistical tests. The mean collocated APS levels were calculated as the mean of the duplicate APS levels per day. Paired t-tests were performed on the mass concentrations to detect systematic bias between each sampler type and to detect within sampler type systematic bias between the APS instruments. Ordinary least squares linear regression was performed on the $PM_{2.5}$ levels, both within and between each sampler type. Regression intercepts significantly

different from zero were considered to indicate systematic bias of $PM_{2.5}$ levels between samplers. Regression slopes significantly different from one were considered to indicate proportional bias of $PM_{2.5}$ levels between samplers (bias that varies relative to concentration). The coefficient of determination (R^2) was used to describe the correlation of measured levels between samplers and the Root Mean Square Error (RMSE) was used to describe the precision of the sampling methods. Structural linear models that account for measurement error in the independent variable were also fit to data from the present study and the results were not substantially different from those using ordinary least squares regression.

3. Results

Twenty batches of 24-hour samples were collected every third day in cycles of approximately three weeks on and one week off from January 6, 2000 through April 9, 2000. A batch consisted of 24-hour samples from two APS samplers (duplicates), one DustTrak, one FRM sampler. A total of 71 valid 24-hour samples were collected out of 80 attempted. Nine samples were voided due to computer errors while transferring data (6), power interruptions (2), and errors programming the sample times. The distribution of the 24-hour average FRM $PM_{2.5}$ levels was not significantly different from normal using the Shapiro-Wilk test (n=19, p=0.3539). The range of the 24-hour average FRM $PM_{2.5}$ levels was from 5.0 $\mu g\ m^{-3}$ to 20.4 $\mu g\ m^{-3}$ with mean and standard deviation 11.4 $\mu g\ m^{-3}$ and 4.0 $\mu g\ m^{-3}$, respectively. Figures 5-1 through 5-4 contain regression statistics and scatter plots of the mass concentrations from each sampler type for both the 24-hour average and 30-minute average concentrations. The regression

equations are presented as $y=\beta_1(+/-t*SE)*x + \beta_0(+/-t*SE)$ where x is the dependent variable, y is the independent variable, and t*SE is the product of the appropriate t-statistic and the standard error of the parameter estimate.

For the within sampler type APS comparison (one APS vs. the other APS) of the 30-minute average levels (n=816; see Figure 5-1), the intercept was significantly different from zero (β_0=0.6, p<0.0001) and the slope was significantly different from one (β_1=1.3, p<0.0001). The R^2 between the two APS samplers was 0.979 with an RMSE of 0.92 μg m^{-3}. Significant systematic bias was found between 30-minute average levels from the two APS samplers using the paired t-test (mean difference=-2.68 μg m^{-3}, p<0.0001).

For the between sampler type comparisons of the 24-hour average levels, significant systematic bias was found between FRM and DustTrak levels (mean difference=-16.8 μg m^{-3}, p<0.0001) and between FRM and mean collocated APS levels (mean difference=4.24 μg m^{-3}, p<0.0001) using the paired t-test. However, neither of the intercepts were significantly different from zero (see Figures 5-2 and 5-3). For the FRM versus the DustTrak 24-hour average levels (see Figure 5-3), the slope is significantly different from one (n=17; β_1=2.57, p<0.0001). The correlation between the FRM and DustTrak is higher than between the FRM and the mean collocated APS (R^2=0.859 and R^2=0.592, respectively).

For the between sampler type comparison of the 30-minute average levels (data available for the APS and DustTrak only; see Figure 5-4), the intercept and slope were

statistically significantly different from zero and one, respectively (n=720; β_0=12.16, p<0.0001; β_1=2.18, p<0.0001). Significant systematic bias was found between the 30-minute average levels from the APS and DustTrak using the paired t-test (mean difference=-22.15 μg m^{-3}, p<0.0001). The correlation and precision between the APS and DustTrak 30-minute average levels (R^2=0.460, RMSE=13.65 μg m^{-3}) is lower and worse, respectively, than for any of the 24-hour average between sampler comparisons.

4. Discussion and Summary

Substantial bias in $PM_{2.5}$ measurements due only to the type of sampler used might erroneously influence exposure estimates and thereby obscure source-to-exposure or exposure-effect relationships examined in future studies. The within sampler type comparison of the two APS samplers shows a high correlation and good precision (R^2=0.979 and RMSE=0.92 μg m^{-3}). However, significant systematic and proportional bias is present according to the paired t-test analyses and statistical tests on the intercept and slope. The results suggest that a substantial amount of error resulting in bias may be involved in the calibration of the APS units by the manufacturer. However, only two APS units were available for evaluation in the present study. The bias observed between the two APS instruments from the within sampler type comparison may be due to inaccurate flow sensor calibration, inaccurate size channel calibration, or differences in the sensitivity of the optical components. The comparability of individual APS units should be assessed before they are used in any measurement situation.

The DustTrak measurements are of lower precision than those made by the APS (RMSE=4.31 μg m^{-3} for FRM vs. DustTrak and RMSE=2.80 μg m^{-3} for FRM vs. mean

collocated APS). The strong correlation of the 24-hour average DustTrak levels with the FRM (R^2=0.859), however, suggests that the proportional bias in the DustTrak levels can be corrected using the slope and intercept from regression of the DustTrak versus FRM levels (DustTrak as the independent variable, reverse of what is shown in Figure 5-3). The correction equation was as follows for the present study:

$$y=0.33*x + 2.25$$

where y equals the corrected value for the DustTrak and x equals the uncorrected DustTrak concentration. Applying the above equation to the DustTrak levels in the present study reduces the root-mean squared difference from 18.3 μg m^{-3} between the FRM and uncorrected DustTrak $PM_{2.5}$ levels to 1.5 μg m^{-3} between the FRM and corrected DustTrak $PM_{2.5}$ levels. Lehocky and Williams reported an R^2 of 0.94 between DustTrak levels and gravimetric methods for respirable coal dust, though in that study regression techniques were used that force the intercept through zero (1996). The R^2 of 0.859 between FRM and DustTrak levels in the present study is similar to these findings. However, the slope in the present study for the FRM and DustTrak (β_1=2.57, p<0.0001) is substantially different than found by Lehocky and Williams (β_1=0.73). Differences in the performance of the DustTrak in the present study may be attributed to lower concentration range, different aerosol composition, and use of the internal impactor for $PM_{2.5}$ (versus an external 10-mm Dorr-Oliver cyclone with a 3.5 μm 50% cut-point).

The regression equation for the within sampler type comparison of the two APS samplers can be used to correct $PM_{2.5}$ measurements from these samplers in the future,

provided the sampled aerosol is similar to that for the present study. The same type of correction using the regression equation for the APS can be used to improve the accuracy of the APS levels relative to those from the FRM, but does not account for all of the variation in the APS levels. This suggests some other source of discrepancy in the APS measurements, such as detection of components of the aerosol that are not detected in the FRM measurements (e.g., volatile components) and/or differential response to particles with different optical properties, or a significant contribution to the FRM levels from particles with aerodynamic diameter <0.3μm (not detected by the APS) or aerodynamic diameter >2.458 μm (due to less than 100% particle size selection efficiency of the WINS impactor at 2.5 μm). Chen et al. examined the performance of the TSI, Inc. 3310 APS, an instrument similar to the APS used in the present study, with regard to the aspiration and penetration efficiency of the inlet (1998). Aspiration effects are not expected to account for large changes in the APS measurements as samplers were collocated indoors (aspiration efficiency is high at low wind speeds; Chen, 1998). The penetration efficiency of the APS 3310 for $PM_{2.5}$ according to Chen et al. is 88.7% (1998). Thus, some of the systematic bias observed in the present study may be explained by imperfect penetration efficiency; adjustment of the APS levels using the Chen et al. correction factor reduces the mean difference between FRM and APS levels by 27%, from 4.24 μg m^{-3} to 3.08 μg m^{-3}. The adjusted mean difference between the FRM and APS levels, however, remains statistically significant (p=0.0001).

This study presents additional information on comparability of $PM_{2.5}$ measurements made by the FRM, APS, and DustTrak. The advantages of the APS for

monitoring indoor and ambient air are that it is small, portable, should require little periodic maintenance, and that it can collect time-resolved information on the mass and size distribution of a sampled aerosol. The disadvantages of the APS are that data logging requires connection to a computer, the accuracy and precision of measurements are lower than for other methods, and that it requires protection from the elements if used outdoors. The low correlation (R^2=0.592) between FRM and APS levels warrants further investigation into sources of error in APS measurements. The APS can collect light-scattering data (not collected for the present study) and future work should focus on the ability of the light-scattering data to identify and distinguish between aerosols of differing composition as well as on determining if the APS mass concentrations in the range of 2.458 μm to approximately 3.523 μm (3.523 μm being the closest channel on the APS that corresponds to an estimated 0% particle size selective efficiency of the WINS; Turner et al., 2000) are related to the difference in filter-based and optical measurements.

The DustTrak offers similar advantages for monitoring $PM_{2.5}$ in indoor and ambient air in that it provides time-resolved data and is small and portable, though it does require minor periodic maintenance (e.g., cleaning and greasing the impactor, verifying the zero point). The disadvantages of the DustTrak involve the precision of the measurements relative to measurements from traditional gravimetric methods, its inability to measure different size fractions of the sampled aerosol simultaneously, and that it requires protection from the elements when used outdoors. The results from the present study suggest that the DustTrak measurements are reasonably precise

(R^2=0.859) compared to the FRM and that valuable and reliable information on $PM_{2.5}$ levels in indoor air can be obtained using the DustTrak; however, the DustTrak measurements should be validated using a collocated, well-characterized method to determine a correction equation to avoid bias in the results.

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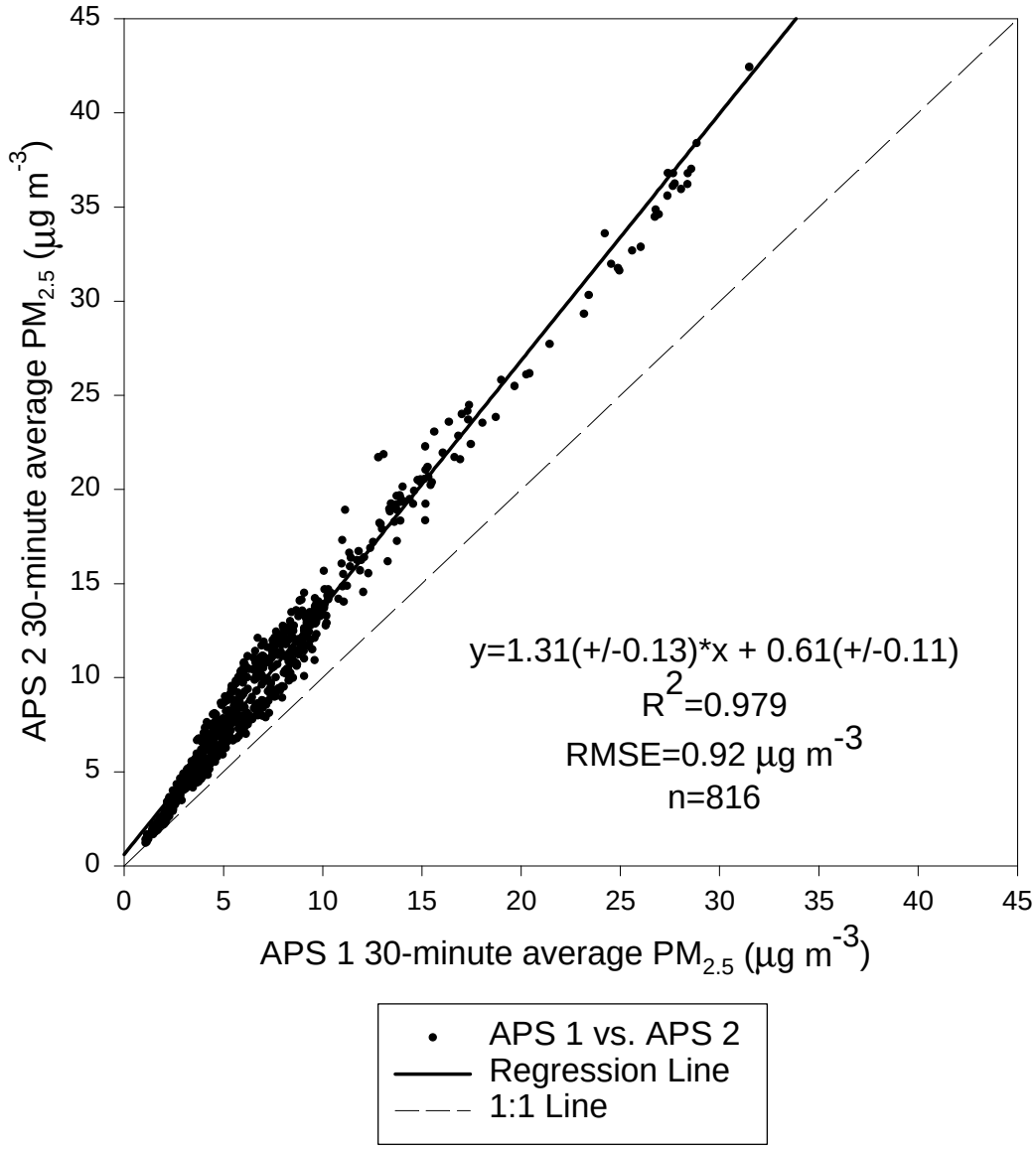

Figure 5-1. Scatter plot of APS 1 vs. APS 2 30-minute average indoor $PM_{2.5}$ measurements in Athens, GA

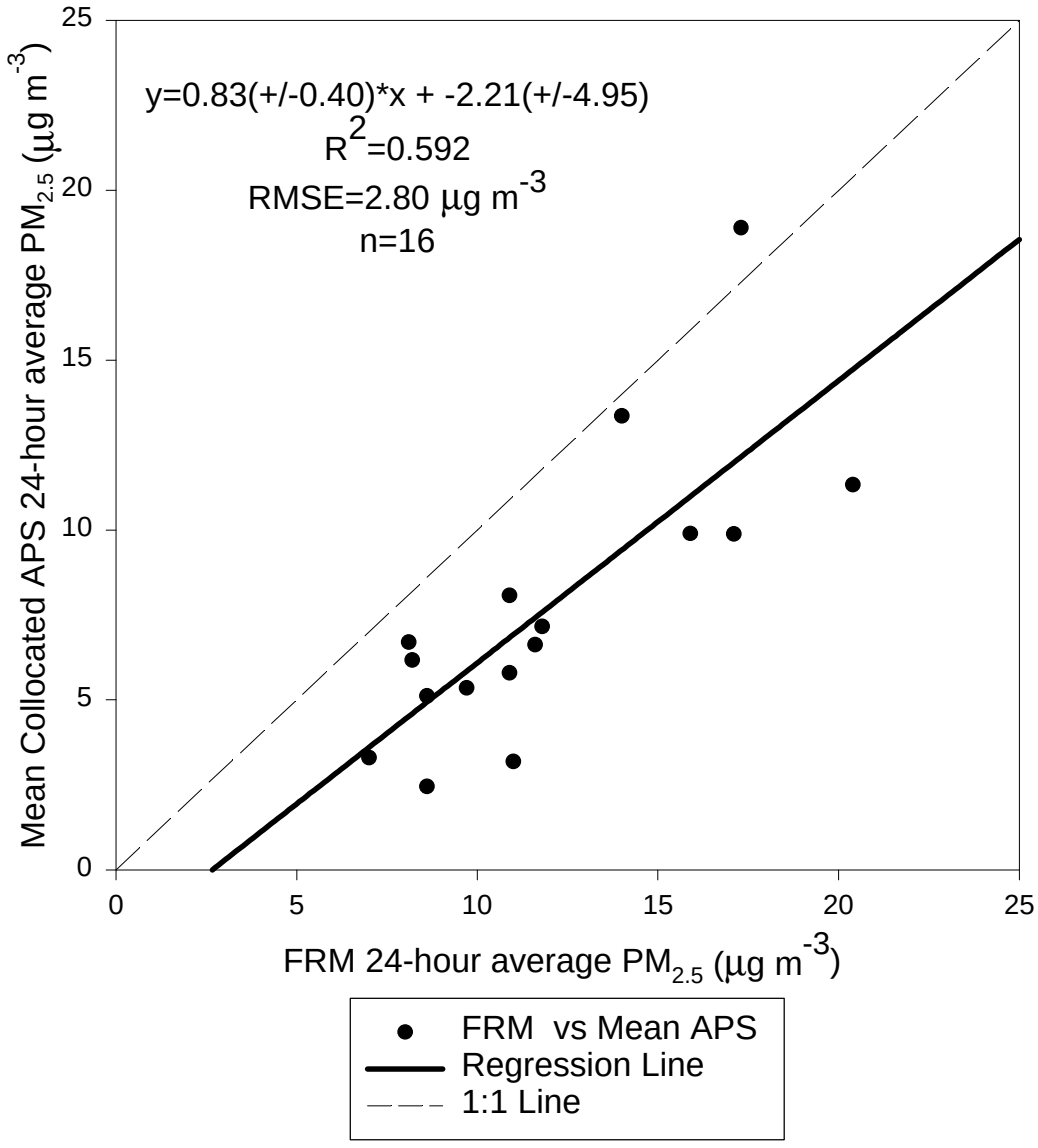

Figure 5-2. Scatter plot of FRM vs. mean collocated APS 24-hour average indoor $PM_{2.5}$ measurements in Athens, GA

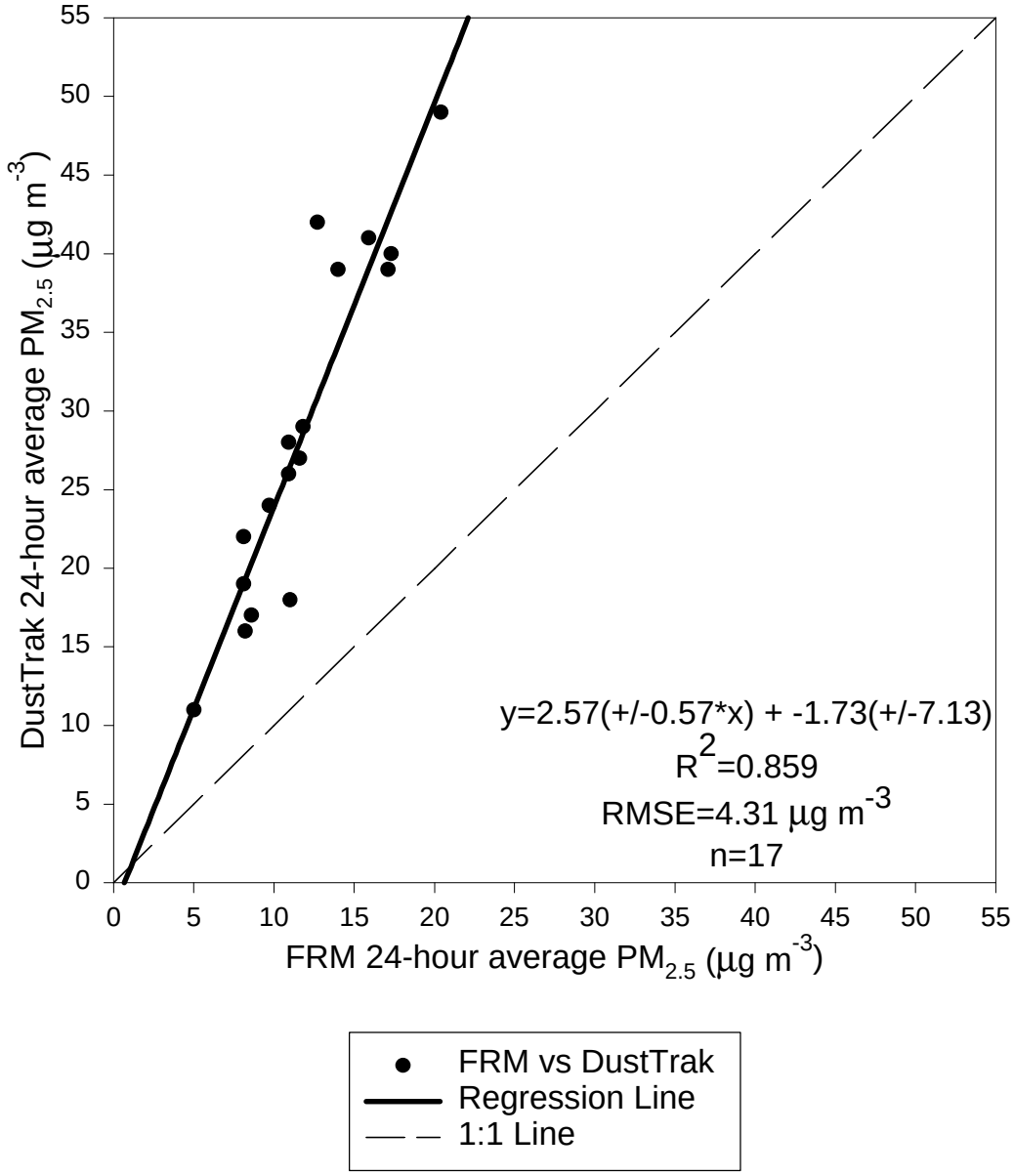

Figure 5-3. Scatter plot of FRM vs. DustTrak 24-hour average indoor $PM_{2.5}$ measurements in Athens, GA

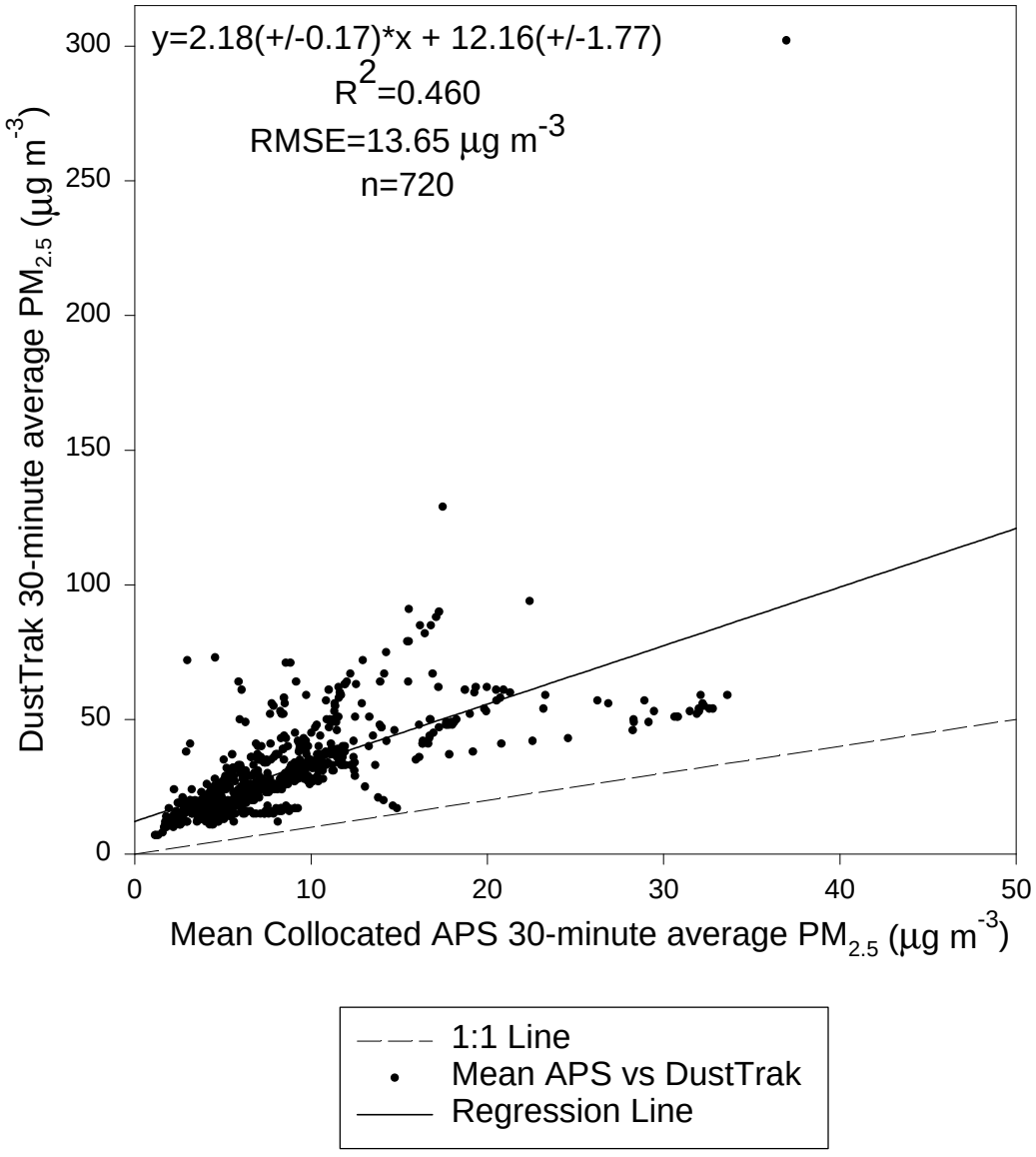

Figure 5-4. Scatter plot of mean collocated APS vs. DustTrak 30-minute average indoor $PM_{2.5}$ measurements in Athens, GA

Note: Axes not to scale

CHAPTER 6

COMPARISON OF THE GRIMM 1.107 ENVIRONMENTAL DUST MONITOR WITH THE FEDERAL REFERENCE METHOD FOR PM_{10}[1]

[1]Report prepared for Grimm Technologies, Inc. for submission to the U.S. Environmental Protection Agency to evaluate performance of the Grimm 1.107 Environmental Dust Monitor compared to the Federal Reference Method for PM_{10}.

I. Objective

The objective of this study was to determine comparability between the Grimm Environmental Dust Monitor, Model 1.107 (Grimm monitors) and a Federal Reference Method (FRM) PM_{10} sampler, the BGI, Inc. PQ200. Comparability was assessed according to the procedures outlined in Title 40 Code of Federal Regulations Part 53 Subpart D (1999b).

II. Summary

A total of twenty-nine (29) valid data sets meeting acceptability criteria specified in the study protocol were collected. Based on a statistical analysis of the data collected:

1) The linear regression parameters (slope, intercept and correlation coefficient) of the relationship between the Grimm monitor data set and the FRM data set are within allowable tolerances for the regression guidelines specified in Title 40 Code of Federal Regulations Part 53 Table C-4 to Subpart C (1999c).
2) The range of values for the precision of the replicate reference method measurements was from 0.0 $\mu g/m^3$ to 1.8 $\mu g/m^3$ with mean of 0.5 $\mu g/m^3$. The maximum of 1.8 $\mu g/m^3$ was within the applicable tolerance of 5 $\mu g/m^3$ listed in Table C-4 (1999c).
3) The percent bias between the mean of triplicate FRM and Grimm measurements per day for the twenty-nine (29) valid data sets collected ranged from 0.0% to (–)25.0% with a mean of –0.9%.

III. Introduction

The University of Georgia's Environmental Health Science Department (EHS) conducted a comparison test of two sampling methods (an optical particle counter and the filter-based reference method) that measure mass concentrations of airborne particulate matter with aerodynamic diameter less than 10 micrometers (PM_{10}). Simultaneous measurements from each type of method (FRM and Grimm) were collected in triplicate for each sampling period. The duration of the nominal sampling period for this test was 24 hours.

This report details the procedure and presents the results of the comparison test including scatter plots, regression parameters, tabulated raw data, and FRM sampler calibration and flow verification reports.

IV. Background

The United States Environmental Protection Agency (EPA) adopted standards for PM_{10} levels in ambient air in 1987. The PM_{10} standard was adopted by EPA to protect the public from the health effects of short and long-term exposure to ambient PM_{10} (24-hour and annual standards, respectively). The PM_{10} standards are expressed as a mass of PM_{10} per volume of air. The EPA's 24-hour PM_{10} standard is 50 $\mu g/m^3$. EPA's annual PM_{10} standard, calculated as the annual geometric mean of the 24-hour concentrations, is 30 $\mu g/m^3$.

The EHS PM_{10} monitoring program used BGI, Inc. PQ200 Federal Reference Method PM_{10} samplers to collect PM_{10} on 47 mm Teflon filters (Gelman R2PJ047).

This method is designated by EPA a Federal Reference Method for the measurement of PM_{10} under designation number RFPS-1298-125.

V. Description of Grimm Environmental Dust Monitor, Model 1.107 Sampler

The Grimm Environmental Dust Monitor, Model 1.107 is an optical instrument that performs particulate measurements using the principle of Mie scattering. Mie scattering is the scattering of light by particulate matter of sizes comparable to the wavelength of light. A laser emits constant light at a specific wavelength into a measurement cell. As the sample enters the measurement cell, it passes through a flat laser beam produced by a low power laser diode. Particulate matter or dust in the sample stream scatters the laser signal in relation to the particle size and surface. The scattered signals are detected by a photo-diode with a 15-channel pulse height analyzer for size classification. The counts from the sized pulses are simultaneously converted to mass concentrations in size categories of PM_{10}, $PM_{2.5}$ and PM_1.

The monitor has a sample flow control system assuring a flow rate of 1.2 liters per minute (LPM) at the inlet. The monitor's sample pump, which is internally monitored, operates at a constant 2 LPM. The remaining 0.8 LPM of flow is routed through an internal zero air filter. This filtered air, referred to as sheath air, is used as jet stream along the edges of the sample stream to keep the optics clean and minimizes the need for maintenance. The moisture content of the sample is controlled by means of a desiccant dryer. When the monitor's relative humidity sensor records a value above a user defined set point (80% for this test), the monitor's microprocessor directs one-half of the sample flow through a zero air filter, and then through a desiccant dryer. This

clean, dry air is reintroduced into the sample stream before the optics, removing excess moisture from the sample, obviating the need for a heated sample inlet. All measurements are stored in real time on a memory card and were downloaded via a laptop computer through the RS-232 port using the Grimm 1.174 software.

VI. Procedure

The operating procedures used in this comparison study were developed using the Grimm monitor operating manual and 40 CFR Part 53 Subpart D. The following section details the test operating procedures.

Study Period:

The study was conducted between August 15, 2000 and October 30, 2000. Triplicate PM_{10} samples from each type of sampling method (FRM and Grimm) were collected simultaneously for 24-hour periods. Each 24-hour period began on a rotating schedule, either one hour after the previous 24-hour period ended or beginning the next morning.

Study Location:

Samples were collected on the roof of the Environmental Health Science Building, University of Georgia, Athens, Georgia. The EHS site was selected because of its security and relatively urban nature, because of the diversity of particle compositions and magnitude of particle concentrations in the area, and for its convenience.

Grimm Operation:

Three Grimm monitors were positioned on the EHS rooftop in between the FRM samplers so that the sampling inlets of all six samplers (three FRM and three Grimm) were approximately three (3) meters apart. The Grimm monitors were initially calibrated by the manufacturer and are designed to perform a self-diagnostic check at the start of each sample. No additional calibration procedures were performed prior to the initial sampling period of this test. Prior to each 24-hour sampling period, the mean mass and total sample volume values of each monitor were reset to zero per the Grimm operating manual instructions. The Grimm samplers were also turned off to ensure that the samplers had been reset properly. At the end of each sampling period, the sampling information on the display of the Grimm monitors was recorded on the field data sheets. PM_{10} mass concentrations used in the calculation of the 24-hour averages were obtained every five minutes from the Grimm monitors and stored on the internal data logger cards. The data cards were downloaded periodically (usually every day) throughout the study using the Grimm 1.174 software program provided to EHS during the study period. The temperature and relative humidity sensors on the Grimm instruments require the input of calibration factors into the Grimm 1.174 software to make necessary temperature and relative humidity measurements. These values were provided by Grimm Technologies, Inc.

FRM Operation:

Three FRM samplers were positioned on the EHS rooftop in between the Grimm monitors so that the sampling inlets of all six samplers (three FRM and three Grimm)

were approximately three (3) meters apart. Prior to the start of the study, all FRM samplers were calibrated according to the BGI, Inc. PQ200 instruction manual in conjunction with guidelines used by the State of Georgia's Environmental Protection Division for U.S. EPA reference method particulate monitoring. At the beginning of each 24-hour sampling period, the pre-weighed FRM filters were installed, the samplers were programmed, and the setup portion of the sample reports for each filter were completed. At the end of each 24-hour sampling period, the FRM filters were collected, the sample log sheets were completed, and the filters were transported to the analytical lab in the EHS building. The FRM samplers log the volumetric flow rate every five (5) minutes. The flow rate and sample duration information was downloaded from each FRM sampler after each sampling period. Periodically throughout the study period, a flow rate verification procedure was performed on the FRM samplers by comparison to an external flow standard. Also, subsequent to any maintenance activities (e.g., replacing a pump), the samplers were also again calibrated. After all samples had been collected, the calibration of the FRM samplers was again verified using the aforementioned procedures. Calibration and flow rate verification forms are attached at the end of this document. Gravimetric, flow rate, and sample duration data were used to generate mass concentrations of PM_{10} in $\mu g/m^3$.

Pre- and post-sampling gravimetric analysis of the FRM filters was performed in accordance with EPA's the Section 2.12, of EPA's Quality Assurance Handbook, Vol. II, Part II (1997). Filters were weighed at least twice prior to sampling and twice after sampling using a Cahn C-32 Microbalance with a sensitivity of +/-1 μg. The tolerance

for reweighing filters was agreement to within +/- 10 μg. The mean of all replicate pre- or post-weighings were used to calculate the mass difference of the filters. Approximately 10% of all FRM samples were collected as field blanks, defined as filters that were transported along with sample filters to the EHS rooftop, loaded into the FRM sampler, and transported back to the analytical lab during the sampling period. Field blank filters were again transported to the sampling site and back to the analytical lab during retrieval of the sample filters. The mean net mass of the field blanks was 4 μg with a standard deviation of 7 μg. The air density during the weighing session, nominal density of the calibration masses, and the measured density of each filter type were used to correct the balance readings of all filters (including blanks) for the buoyant effect of air following Koistinen, Kousa et al (1999a).

In accordance with Title 40 CFR Subpart D (1999b), the precision of the replicate FRM measurements was calculated as the standard deviation of the FRM measurements for each day where the mean of the FRM measurements for that day was below 80 $\mu g/m^3$. All of the mean FRM values for each day were below 80 $\mu g/m^3$ for this test. Values for the precision of the FRM measurements are expressed in $\mu g/m^3$ and are listed in Table 6-1.

The percent difference between the mean FRM and mean Grimm measurements per day is also presented as an indicator of the accuracy of the Grimm measurements compared to the FRM measurements for each day and was calculated according to the formula:

Percent difference=((X-Y)/X)*100%

where,

X=mean of triplicate FRM mass concentrations on day *j*,

Y=mean Grimm mass concentrations on day *j*,

and *j*=day.

VII. Results

The 24-hour average PM_{10} mass concentrations for both the FRM and the Grimm monitors are summarized in Table 6-1. The results from the Grimm monitors represent the data values obtained using the Grimm 1.174 software package. The mean of the triplicate FRM measurements for each day is listed in the column labeled "Mean FRM/day". The mean of the triplicate Grimm measurements on each day is listed in the column labeled "Mean Grimm/day". Also listed is the precision of the FRM samplers in $\mu g/m^3$ (standard deviation of the triplicate measurements on each day, in accordance with Title 40 CFR Subpart D, 1999b) and the percent difference between the FRM and Grimm samplers for each day (formula shown above). For sample days where all instruments collected valid 24-hour PM_{10} samples, the precision of the FRM samplers ranged from <0.0 to 1.8 $\mu g/m^3$, the 24-hour average PM_{10} mass concentrations from the FRM samplers ranged from 8.5 $\mu g/m^3$ to 50.7 $\mu g/m^3$, and the 24-hour average PM_{10} concentrations from the Grimm monitors ranged from 10.3 to 56.7 $\mu g/m^3$.

Data capture:

A total of (226) valid 24-hour samples out of a possible two hundred forty (240) were collected representing a data capture rate for the 24-hour samples of 94%. Of the fourteen (14) void samples, ten (10) were voided due to malfunctions downloading data from the Grimm instruments and of these, nine (9) were due to a defective data card (not an integral part of the instrument, only a data storage device) and one (1) due to operator error in the downloading process. The other two (2) Grimm samples that were voided were also due to operator error (the samplers were not turned off at the correct time). The only FRM sample that was voided was due to a pump failure on one of the FRM samplers. The pump was replaced and the sampler calibrated before the next sampling period.

A total of twenty-nine (29) days out of a possible forty (40) sampling days had valid samples for all six individual samplers (three (3) FRM and three (3) Grimm). The data capture rate for valid sampling days (six (6) valid samples per day) was 72%.

Linear regression and precision of reference measurements:

Ordinary least squares regression was performed on the FRM and Grimm measurements. The requirements for adequate comparability between a reference and candidate method PM_{10} sampler listed in Title 40 CFR 53 Table C-4 to Subpart C (1999c) are that the linear regression parameters (slope, intercept and correlation coefficient) meet the following values:

Slope of regression relationship..... 1 ± 0.1

Intercept of regression relationship, $\mu g/m^3$..... 0 ± 5

Correlation of reference method and candidate

method measurements..... ≥ 0.97

The relationship between measurements made by the FRM samplers and Grimm monitors for all valid data sets is as follows:

Slope of regression relationship.....1.03

Intercept of regression relationship, $\mu g/m^3$..... -0.79

Correlation of reference method and candidate

method measurements..... 0.972

The regression parameters for this test are within the tolerances listed above. Figure 6-1 shows a scatter plot of the FRM and Grimm measurements as well as plots of the regression line and the 1:1 line. Additionally, the precision of the replicate reference measurements, expressed as the standard deviation of the triplicate FRM measurements on each day, must be less than 5.0 $\mu g/m^3$ for each day. The maximum value for the precision of the replicate reference method measurements in the present study is 1.8 $\mu g/m^3$ with a mean of 0.5 $\mu g/m^3$. The precision of the replicate reference method measurements made as part of this test are thus also within the allowable tolerance listed in Table C-4 (1999c).

Grimm monitor density factor

The 47 mm Teflon filters from the Grimm monitors were not weighed as part of this test. Fresh filters were installed before the initial sample period to ensure that the Grimm monitor pumps were not subjected to undue stress caused by excessive loading of the filters. These filters were not changed during the test period. The default value

of 1 g/ml was used as the density factor for the Grimm monitors. No gravimetric analysis was required to generate the Grimm measurements used in this test.

VIII. Conclusions

A total of twenty nine (29) sets of 24-hour PM_{10} samples (six (6) samples per set, three (3) FRM and three (3) Grimm) were collected during this study. Linear regression of the FRM and Grimm data gives values for the slope, intercept, and correlation coefficient (R) of 1.03, -0.79, and 0.972, respectively. Based on these data, the Grimm instruments appear to agree well with the FRM in the range of concentrations of this test and for the type of ambient aerosol typical in Athens, GA in the fall. No adjustments were made using the density factor on the Grimm instruments. Atmospheric effects such as wind speed and relative humidity were not evaluated for their contribution to the influence they have on the Grimm measurements.

XI. References

Koistinen, K.J., Kousa, A., Tenhola, V., Hanninen, H., Jantunen, M. J., Oglseby, L., Kuenzli, N., and Georgoulis, L. Fine particle (PM2.5) measurement methodology, quality assurance procedures, and pilot results of the EXPOLIS study. *J. Air & Waste Manage. Assoc*. Vol. 49, Issue 10, pp. 1212-20. 1999a.

Title 40 Code of Federal Regulations Part 53 Subpart D-Procedures for Testing Performance Characteristics of Methods for PM_{10}. 07/01/99 Edition. 1999b.

Title 40 Code of Federal Regulations Part 53 Subpart C Table C-4 Section 53.34. 07/01/99 Edition. 1999c.

United States Environmental Protection Agency. *Quality Assurance Handbook*, Vol. 2, Part 2. Section 2.12. 1997.

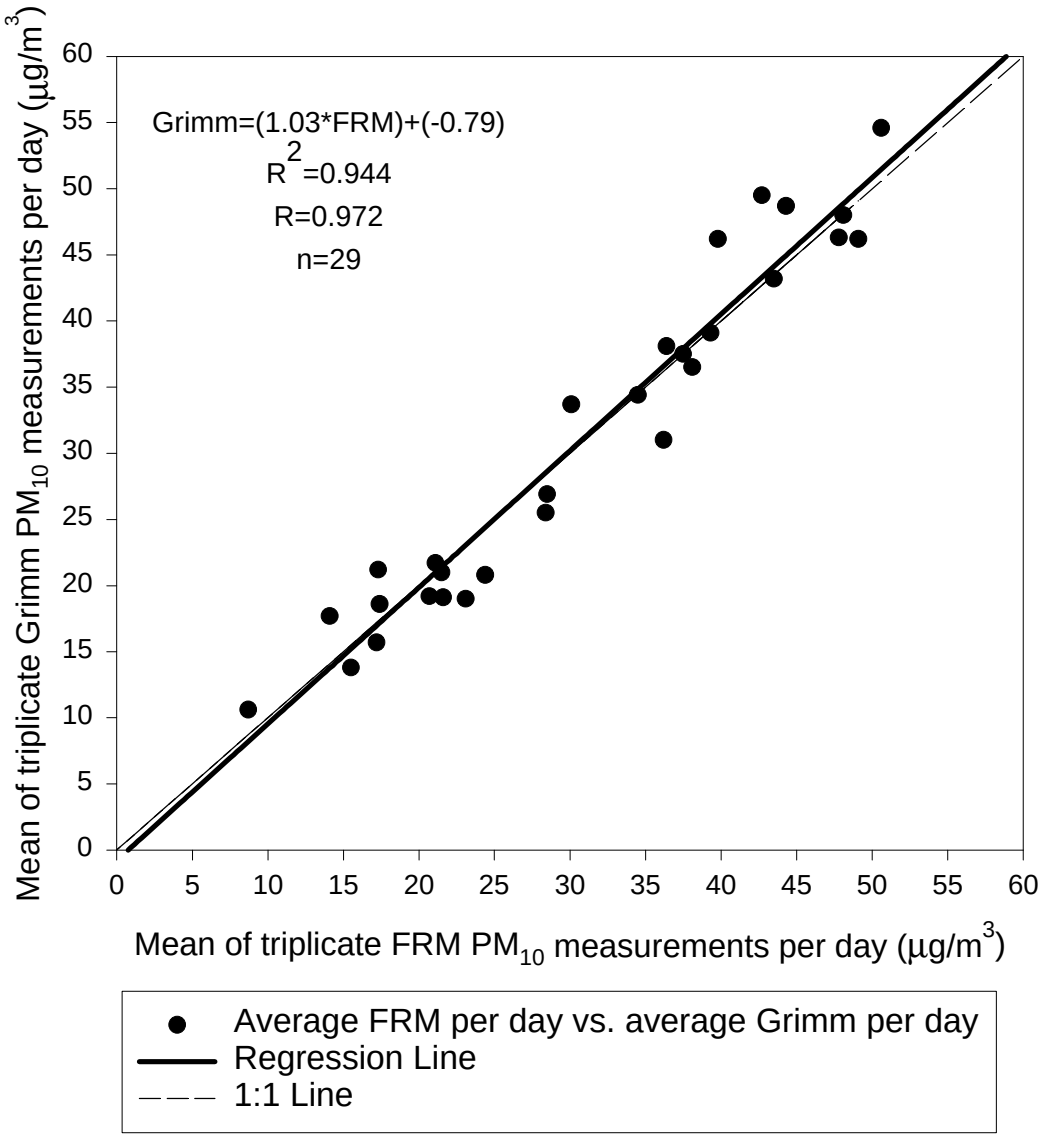

Figure 6-1. Scatter plot of mean triplicate FRM vs. mean triplicate Grimm PM_{10}
24-hour average measurements
August 15, 2000 through October 30, 2000
Ambient air in Athens, GA

Table 6-1. Tabulated raw data on FRM and Grimm PM_{10} measurements

Start date	Day	FRM concentration (mg/m3)		Grimm concentration (mg/m3)		Avg. FRM/day (mg/m3)	Avg. Grimm/day (mg/m3)	Precision of FRM (mg/m3)	FEM acceptable FRM	FEM acceptable Grimm	Percent difference (%)
08/22/00	1	FRM 1	37.9	Grimm 1	36.8	38.1	36.5	0.2	38.1	36.5	4.3
		FRM 2	38.2	Grimm 2	37.2						
		FRM 3	38.2	Grimm 3	35.4						
08/23/00	2	FRM 1	47.2	Grimm 1	46.2	49.1	46.2	1.8	49.1	46.2	6.0
		FRM 2	49.4	Grimm 2	48.4						
		FRM 3	50.7	Grimm 3	43.9						
08/24/00	3	FRM 1	46.2	Grimm 1	46.9	48.1	48.0	1.8	48.1	48.0	0.3
		FRM 2	48.5	Grimm 2	51.0						
		FRM 3	49.7	Grimm 3	46.1						
08/25/00	4	FRM 1	25.4	Grimm 1	27.6	25.6		0.1			
		FRM 2	25.6	Grimm 2	28.7						
		FRM 3	25.7	Grimm 3	VOID						
08/26/00	5	FRM 1	33.7	Grimm 1	29.1	32.7		1.0			
		FRM 2	31.7	Grimm 2	29.7						
		FRM 3	32.6	Grimm 3	VOID						
08/29/00	6	FRM 1	34.4	Grimm 1	32.0	34.3		0.2			
		FRM 2	34.4	Grimm 2	31.5						
		FRM 3	34.1	Grimm 3	VOID						
08/30/00	7	FRM 1	20.0	Grimm 1	18.4	20.5		0.5			
		FRM 2	20.5	Grimm 2	18.6						
		FRM 3	20.9	Grimm 3	VOID						
08/31/00	8	FRM 1	29.9	Grimm 1	29.6	30.5		0.8			
		FRM 2	30.3	Grimm 2	29.7						
		FRM 3	31.4	Grimm 3	VOID						
09/02/00	9	FRM 1	28.2	Grimm 1	26.4	28.3		0.2			
		FRM 2	28.4	Grimm 2	26.7						
		FRM 3	28.5	Grimm 3	VOID						
09/03/00	10	FRM 1	22.2	Grimm 1	23.2	22.2		0.5			
		FRM 2	21.7	Grimm 2	23.3						
		FRM 3	22.7	Grimm 3	VOID						
09/04/00	11	FRM 1	25.9	Grimm 1	24.9	25.9		0.4			
		FRM 2	26.3	Grimm 2	25.7						
		FRM 3	25.5	Grimm 3	VOID						

Table 6-1: Page i of iv

Table 6-1. Tabulated raw data on FRM and Grimm PM_{10} measurements

Start date	Day	FRM concentration (mg/m3)		Grimm concentration (mg/m3)		Avg. FRM/day (mg/m3)	Avg. Grimm/day (mg/m3)	Precision of FRM (mg/m3)	FEM acceptable FRM	FEM acceptable Grimm	Percent difference (%)
09/05/00	12	FRM 1	26.6	Grimm 1	23.6	26.7		0.5			
		FRM 2	27.2	Grimm 2	24.5						
		FRM 3	26.3	Grimm 3	VOID						
09/09/00	13	FRM 1	25.4	Grimm 1	21.1	24.4	20.8	0.9			15.0
		FRM 2	24.3	Grimm 2	21.4						
		FRM 3	23.6	Grimm 3	19.8						
09/10/00	14	FRM 1	22.4	Grimm 1	19.4	21.6	19.1	0.8			11.5
		FRM 2	21.7	Grimm 2	19.5						
		FRM 3	20.7	Grimm 3	18.4						
09/11/00	15	FRM 1	28.2	Grimm 1	27.5	28.5	26.9	0.3			5.8
		FRM 2	28.6	Grimm 2	27.3						
		FRM 3	28.7	Grimm 3	25.8						
09/12/00	16	FRM 1	34.5	Grimm 1	34.6	34.5	34.4	0.2	34.5	34.4	0.4
		FRM 2	34.7	Grimm 2	35.4						
		FRM 3	34.3	Grimm 3	33.2						
09/13/00	17	FRM 1	42.9	Grimm 1	42.9	43.5	43.2	0.6	43.5	43.2	0.8
		FRM 2	43.9	Grimm 2	44.6						
		FRM 3	43.7	Grimm 3	42.0						
09/14/00	18	FRM 1	40.0	Grimm 1	38.9	39.3	39.1	0.7	39.3	39.1	0.3
		FRM 2	38.8	Grimm 2	40.2						
		FRM 3	38.9	Grimm 3	38.3						
09/15/00	19	FRM 1	17.2	Grimm 1	15.4	17.2	15.7	0.2			8.7
		FRM 2	17.5	Grimm 2	16.0						
		FRM 3	17.0	Grimm 3	15.7						
09/16/00	20	FRM 1	16.0	Grimm 1	13.4	15.5	13.8	0.5			11.0
		FRM 2	15.2	Grimm 2	14.1						
		FRM 3	15.2	Grimm 3	13.8						
09/18/00	21	FRM 1	20.8	Grimm 1	19.2	20.7	19.2	0.1			7.4
		FRM 2	20.8	Grimm 2	19.8						
		FRM 3	20.5	Grimm 3	18.5						
09/19/00	22	FRM 1	28.7	Grimm 1	25.9	28.4	25.5	0.2			10.3
		FRM 2	28.2	Grimm 2	26.4						
		FRM 3	28.3	Grimm 3	24.1						

Table 6-1: Page ii of iv

Table 6-1. Tabulated raw data on FRM and Grimm PM_{10} measurements

Start date	Day	FRM concentration (mg/m3)		Grimm concentration (mg/m3)		Avg. FRM/day (mg/m3)	Avg. Grimm/day (mg/m3)	Precision of FRM (mg/m3)	FEM acceptable FRM	FEM acceptable Grimm	Percent difference (%)
09/20/00	23	FRM 1	17.4	Grimm 1	18.3	17.4	18.6	0.3			-6.8
		FRM 2	17.7	Grimm 2	18.7						
		FRM 3	17.0	Grimm 3	18.7						
09/21/00	24	FRM 1	14.4	Grimm 1	17.6	14.1	17.7	0.2			-25.0
		FRM 2	14.0	Grimm 2	17.7						
		FRM 3	14.0	Grimm 3	17.7						
09/24/00	25	FRM 1	20.9	Grimm 1	20.6	21.5	21.0	0.5			2.1
		FRM 2	21.9	Grimm 2	21.9						
		FRM 3	21.6	Grimm 3	20.5						
09/25/00	26	FRM 1	8.8	Grimm 1	10.3	8.7	10.6	0.2			-21.1
		FRM 2	8.8	Grimm 2	11.1						
		FRM 3	8.5	Grimm 3	10.3						
09/27/00	27	FRM 1	21.2	Grimm 1	21.2	21.1	21.7	0.3			-2.6
		FRM 2	21.3	Grimm 2	22.1						
		FRM 3	20.8	Grimm 3	VOID						
09/28/00	28	FRM 1	23.6	Grimm 1	19.1	23.1	19.0	0.5			17.8
		FRM 2	22.5	Grimm 2	19.5						
		FRM 3	23.3	Grimm 3	18.4						
10/04/00	29	FRM 1	36.6	Grimm 1	37.8	36.4	38.1	0.3	36.4	38.1	-4.6
		FRM 2	36.6	Grimm 2	39.3						
		FRM 3	36.1	Grimm 3	37.2						
10/05/00	30	FRM 1	36.6	Grimm 1	31.6	36.2	31.0	0.6	36.2	31.0	14.3
		FRM 2	36.5	Grimm 2	31.9						
		FRM 3	35.5	Grimm 3	29.6						
10/06/00	31	FRM 1	17.5	Grimm 1	21.0	17.3	21.2	0.2			-22.8
		FRM 2	17.2	Grimm 2	21.8						
		FRM 3	17.1	Grimm 3	20.9						
10/16/00	32	FRM 1	37.7	Grimm 1	37.4	37.5	37.5	0.6	37.5	37.5	0.0
		FRM 2	38.0	Grimm 2	38.6						
		FRM 3	36.9	Grimm 3	36.6						
10/17/00	33	FRM 1	45.3	Grimm 1	48.7	44.3	48.7	1.0	44.3	48.7	-10.0
		FRM 2	43.9	Grimm 2	49.6						
		FRM 3	43.5	Grimm 3	47.8						

Table 6-1: Page iii of iv

Table 6-1. Tabulated raw data on FRM and Grimm PM_{10} measurements

Start date	Day	FRM concentration (mg/m3)		Grimm concentration (mg/m3)		Avg. FRM/day (mg/m3)	Avg. Grimm/day (mg/m3)	Precision of FRM (mg/m3)	FEM acceptable FRM	FEM acceptable Grimm	Percent difference (%)
10/18/00	34	FRM 1	30.6	Grimm 1	32.4	30.1	33.7	0.5	30.1	33.7	-12.0
		FRM 2	29.9	Grimm 2	34.6						
		FRM 3	29.7	Grimm 3	34.1						
10/19/00	35	FRM 1	VOID	Grimm 1	47.5		49.9				
		FRM 2	45.5	Grimm 2	51.7						
		FRM 3	45.1	Grimm 3	50.6						
10/20/00	36	FRM 1	42.6	Grimm 1	48.8	42.7	49.5	0.0	42.7	49.5	-16.1
		FRM 2	42.7	Grimm 2	50.8						
		FRM 3	42.7	Grimm 3	49.0						
10/21/00	37	FRM 1	40.3	Grimm 1	45.3	39.8	46.2	0.4	39.8	46.2	-15.9
		FRM 2	39.7	Grimm 2	47.5						
		FRM 3	39.6	Grimm 3	45.7						
10/22/00	38	FRM 1	50.6	Grimm 1	53.1	50.6	54.6	0.0	50.6	54.6	-7.9
		FRM 2	50.5	Grimm 2	56.7						
		FRM 3	50.6	Grimm 3	53.9						
10/24/00	39	FRM 1	48.4	Grimm 1	45.1	47.8	46.3	0.7	47.8	46.3	3.3
		FRM 2	47.9	Grimm 2	48.4						
		FRM 3	47.1	Grimm 3	45.3						
				Mean-		42.2	46.7	0.3	42.2	46.0	-9.7
				Count (n)-		5	6	5	5.0	5.0	5
				Overall FRM CV (%)-				0.01			

Explantion of VOID samples:
08/23/00-09/05/00: Grimm 3 data card malfunction-data lost
09/27/00: Grimm 3 download error
10/19/00: FRM 1 had a pump failure. Pump replaced and sampler recalibrated.

Table 6-1: Page iv of iv

CHAPTER 7

CONCLUSIONS AND RECOMMENDATIONS

I. Fine particle exposure assessment of prescribed fire workers

This document presents information on the levels of $PM_{2.5}$ exposure to prescribed fire workers in the southeastern United States as well as comparisons of several particulate matter sampling methods in ambient air, in indoor air, and at locations downwind of prescribed fires.

Fire worker $PM_{2.5}$ exposure was shown to be substantially greater than background levels. In comparison to occupational standards for respirable particulate matter, however, only a small fraction of prescribe fire workers are expected to be overexposed to this pollutant. Activities that contributed significantly to $PM_{2.5}$ exposure were not able to be identified. Results suggest that a more sensitive method and more specific information regarding physical characteristics of the prescribed burn are needed to identify specific activities that result in increased $PM_{2.5}$ exposure, such as the use of a time-resolved sampling method and more accurate and highly-resolved information on specific time intervals when activities were performed. This would aid the identification of activities related to increased exposure, would help identify sampling errors, and would also provide information on peak concentrations during specific activities. Though none of the activities investigated in the present study appear to provide accurate and precise information on the determinants of the level of $PM_{2.5}$ exposure, significant relationships may be found for other variables such as emission factors (including terms for type, wetness, and amount of fuel), area- or person-specific meteorological parameters (relative humidity, wind speed, etc), or more detailed questions regarding worker perception of smoke encountered.

Future research should focus on the components of collected particulate matter from the personal, ambient/background, and downwind site $PM_{2.5}$ samples. Analysis of these filters for content of silica, elemental and organic carbon, carbon[12] and carbon [14], volatile organics, polynuclear aromatic hydrocarbons, and other components may provide additional insight into the toxicological significance of suspended particles from wood smoke. Sample filters may also be used to determine the toxicological potency of collected particulate matter, or its components, to human cells (e.g., alveolar macrophages) or other indicator organisms.

Future refinements of the sampling methods should include investigation of the use of different types of 37 mm filters with the KTL cyclone. Gelman Zeflour filters were used for the present study and worked well for sampling at ambient levels, but significant pressure drop was observed at the high concentrations during prescribed burning. Quartz-fiber filters are a potential candidate for use; however, they are expected to have increased field blank variability due to the nature of the matrix of the filter material. A bench trial to determine the reduction in pressure drop upon loading of the filter versus the increased field blank variability should be undertaken. Also, sampling at lower flow rate should be considered because this would afford reduced loading of the filters. The use of a cyclone or size selective inlet that is equipped with a shrouded inlet (e.g., the BGI Triplex cyclone) is expected to reduce sampling errors caused by contact with the ground or other foreign objects. Finally, exposure to other pollutants in wood smoke need to be addressed with regards to their effects on respiratory health effects in prescribed fire workers.

II. Particulate matter sampling method comparisons

The sampling methods used for the personal and ambient/background $PM_{2.5}$ sampling during prescribed burning agreed well at collocated comparison sites (R^2=0.994). The performance of the KTL and FRM samplers in the present study confirms that they can be used to reliably and accurately measure the $PM_{2.5}$ component of wood smoke even at highly elevated concentrations downwind of prescribed fires.

The results from the comparisons of gravimetric samplers in ambient air suggest that the MiniVol may underestimate the concentration given by the other gravimetric sampler types as concentrations increase. However, mean concentrations agree to within 9% between each gravimetric sampler type in the range of 5-35 $\mu g/m^3$ in ambient air.

This study also presents additional information on comparability of $PM_{2.5}$ measurements made by the FRM, APS, and DustTrak. The results suggest that though the DustTrak provides precise measurements of $PM_{2.5}$, the accuracy of the measurements compared to the FRM can be improved through statistical adjustment. In contrast, APS $PM_{2.5}$ measurements are less accurate and precise compared to the FRM and therefore results from the APS should be interpreted with caution. The advantages of the APS for monitoring indoor and ambient air are that it is small, portable, should require little periodic maintenance, and that it can collect time-resolved information on the mass and size distribution of a sampled aerosol. The disadvantages of the APS are that data logging requires connection to a computer, the accuracy and precision of measurements are lower than for other methods, and that it requires protection from the

elements if used outdoors. The low correlation (R^2=0.592) between FRM and APS levels warrants further investigation into sources of error in APS measurements. The APS can collect light-scattering data (not collected for the present study) and future work should focus on the ability of the light-scattering data to identify and distinguish between aerosols of differing composition as well as on determining if the APS mass concentrations in the range of 2.458 μm to approximately 3.523 μm (3.523 μm being the closest channel on the APS that corresponds to an estimated 0% particle size selective efficiency of the WINS; Turner et al., 2000) are related to the difference in filter-based and optical measurements.

The DustTrak offers similar advantages for monitoring $PM_{2.5}$ in indoor and ambient air in that it provides time-resolved data and is small and portable, though it does require minor periodic maintenance (e.g., cleaning and greasing the impactor, verifying the zero point). The disadvantages of the DustTrak involve the precision of the measurements relative to measurements from traditional gravimetric methods, its inability to measure different size fractions of the sampled aerosol simultaneously, and that it requires protection from the elements when used outdoors. The results from the present study suggest that the DustTrak measurements are reasonably precise (R^2=0.859) compared to the FRM and that valuable and reliable information on $PM_{2.5}$ levels in indoor air can be obtained using the DustTrak; however, the DustTrak measurements should be validated using a collocated, well-characterized method to determine a correction equation to avoid bias in the results.

The Grimm 1.107 monitor evaluated as part of the present study is small and portable, can provide accurate and precise time- and size-resolved information on the sampled aerosol. Atmospheric effects such as wind speed and relative humidity were not evaluated for their contribution to the influence they have on the Grimm measurements. Limited information collected at sites downwind of prescribed burns suggests that the Grimm monitors may detect compounds not detected by the FRM, possibly water droplets, steam, or volatile components in wood smoke. Several techniques are available from the manufacturer to correct, reduce, or at least describe these types of errors and evaluation of their effectiveness in the field remains of interest.

In summary, prescribed fire workers in the southeast are exposed to elevated levels of $PM_{2.5}$ that can not be easily predicted from information on their work activities. The gravimetric $PM_{2.5}$ sampling methods agreed well in ambient air (R^2>0.96 for all) except for the MiniVol, the optical $PM_{2.5}$ sampling methods agree less well in indoor air,(R^2>0.592), the Grimm optical PM_{10} method agrees well in ambient air(R^2>0.944 for all), and the personal method agrees well with the FRM (n=9, R^2=0.994) downwind of prescribed fires.

APPENDICES

Prescribed Fire Log

UGA-USFS Smoke Study

Burn #

Date: _____ Name: _____

Fire type: Site Prep Fuels Reduction

Number of new fires worked today: _____

Activity	**Total time spent on each activity** (to within 15 minutes; for example, 3 1/2 hr, 2 1/4 hr, etc.)
Holding	
On-foot	
Mule	
4-wheeler	
Vehicle	
Dozer	
Supervisor	
Direct Attack	
Lighting	
Drip-torch	
ATV	
Mop-Up	
On-foot	
Mule	
4-wheeler	
Vehicle	
Dozer	
Supervisor	
Burn Boss	
TOTAL (hrs)	

Did you use any type of respiratory protection today?

Yes No

If yes, what type? _____

Did you smoke cigarettes/cigars during the work day?

Yes No

If yes, how many? _____

How much wood smoke do you feel you encountered today?

None to very little | A low level | A medium level | A medium to high level | A high level

Please describe any problems with the sampling equipment today (hose disconnected, pump light not blinking, cyclone fell off, battery disconnected, etc.,)-

Figure A-1. Example fire worker log sheet

Calibration Form for BGI, Inc. PQ200 FRM PM2.5/PM10 Sampler							
Date-				Press. Std Make/Model-			
Sampler S/N-				Press. Std S/N-			
Location-				Press. Std. Cert. Date-			
Temp. Std Make/Model-				Flow Std Make/Model-			
Temp. Std S/N-				Flow Std S/N-			
Temp. Std. Cert. Date-				Flow Std. Cert. Date-			
Sampler Date Correct?-				Sampler Time (EST) Correct?-			
Ambient Temperature (Ta) Sensor Verification					EST Time:		
Sampler Indicated Ta-		deg C	(A)	Sampler Indicated Tf-			
Temp. Std. Indicated Ta-		deg C	(B)	Temp. Std. Indicated Tf-			
Ta Error=(A)-(B)=		deg C		Tf Error=			
Tolerance: Ta Error </= +/- 2 deg C							
Sampler Ta within tolerance?-							
Ambient Pressure (Pa) Sensor Verification					EST Time:		
Sampler Indicated Pa-		mmHg	(C)				
Press. Std. Indicated Pa-		mmHg	(D)				
Pa Error=(C)-(D)=		mmHg					
Tolerance: Pa Error </= +/- 10 mmHg							
Sampler Pa within tolerance?-							
Leak Check Results					EST Time:		
External Leak Check (adapter closed on downtube)							
Leak Check Passed?-							
Internal Leak Check (impermeable membrane in cassette)							
Leak Check Passed?-							
Flow Rate Calibration/Verification-					EST Time:		
Calibration ("Select and Calibrate a Flow Rate"):							
Target: 15.0	L/min	Target:	16.7	L/min	Target:	18.3	L/min
Corrected Q-		Corrected Q-			Corrected Q-		
Verification ("Verify Flow Calibration" or "Continue w/ Current Run"):							
Target:		15.0	L/min	16.7	L/min	18.3	L/min
Flow Std. Ind. Q (>29)-			lpm(>)		lpm(>) (E)		lpm(>)
Sampler Indicated Avg.Q-			lpm		lpm (F)		lpm
Sampler Flow % Error=((E-F)/E)*100%-							
Tolerance: Sampler Flow % Error </= +/- 4%							
Sampler Flow % Error within tolerance?-							
Sampler Flow Absolute Error=((16.7-F)/16.7)*100%=							
Tolerance: Sampler Flow Absolute Error </= +/- 5%							
Sampler Flow Absolute Error within tolerance?-							
Sampler Flow set to 16.7 L/min?-							
Comments:							
Technician signature:							

Figure A-2. Example FRM calibration form for BGI, Inc. PQ200 $PM_{2.5}$/PM_{10} sampler

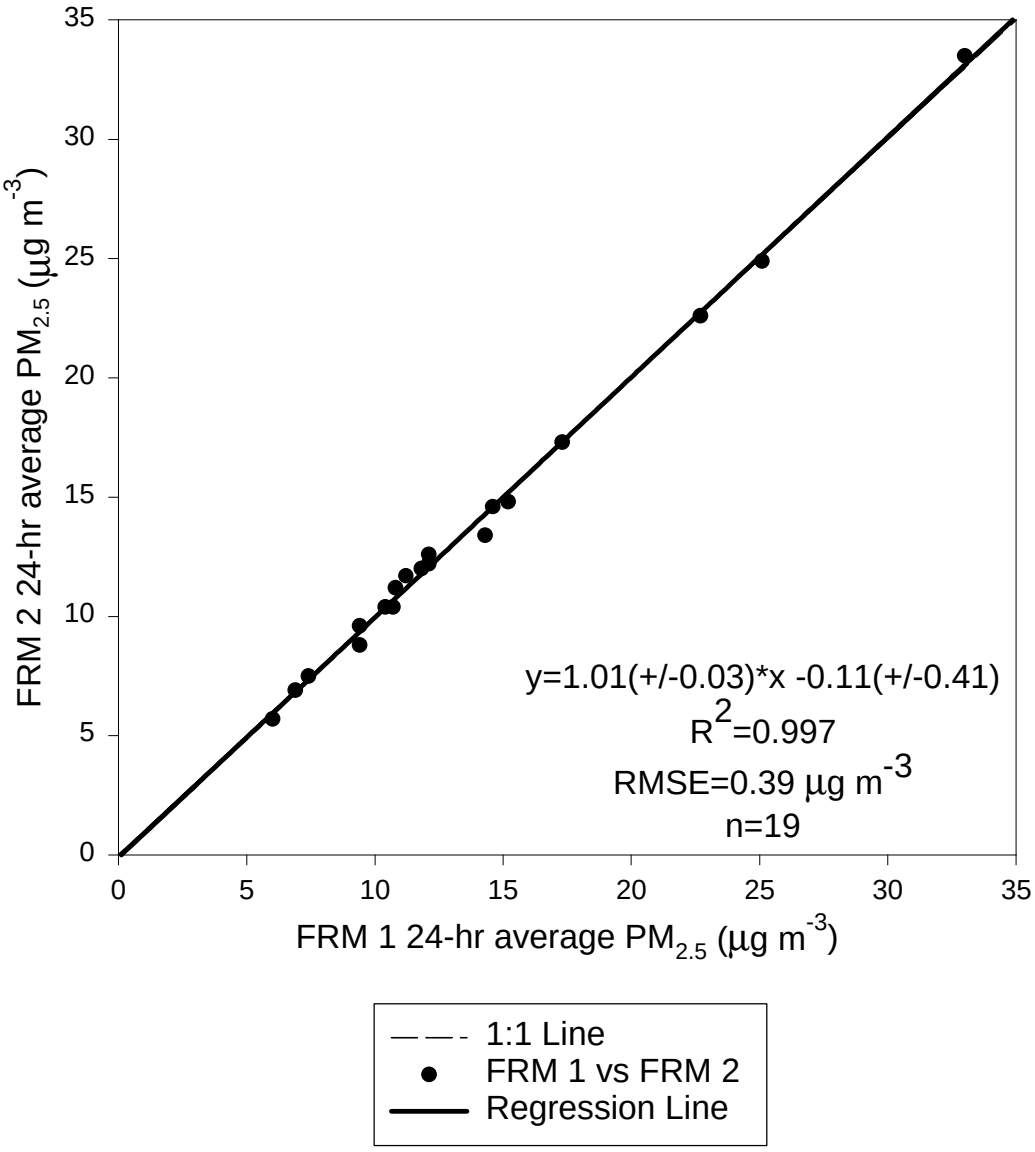

Figure A-3. Scatter plot of FRM 1 vs. FRM 2 24-hr average $PM_{2.5}$ measurements

January 6, 2000 through April 9, 2000
Ambient air in Athens, GA

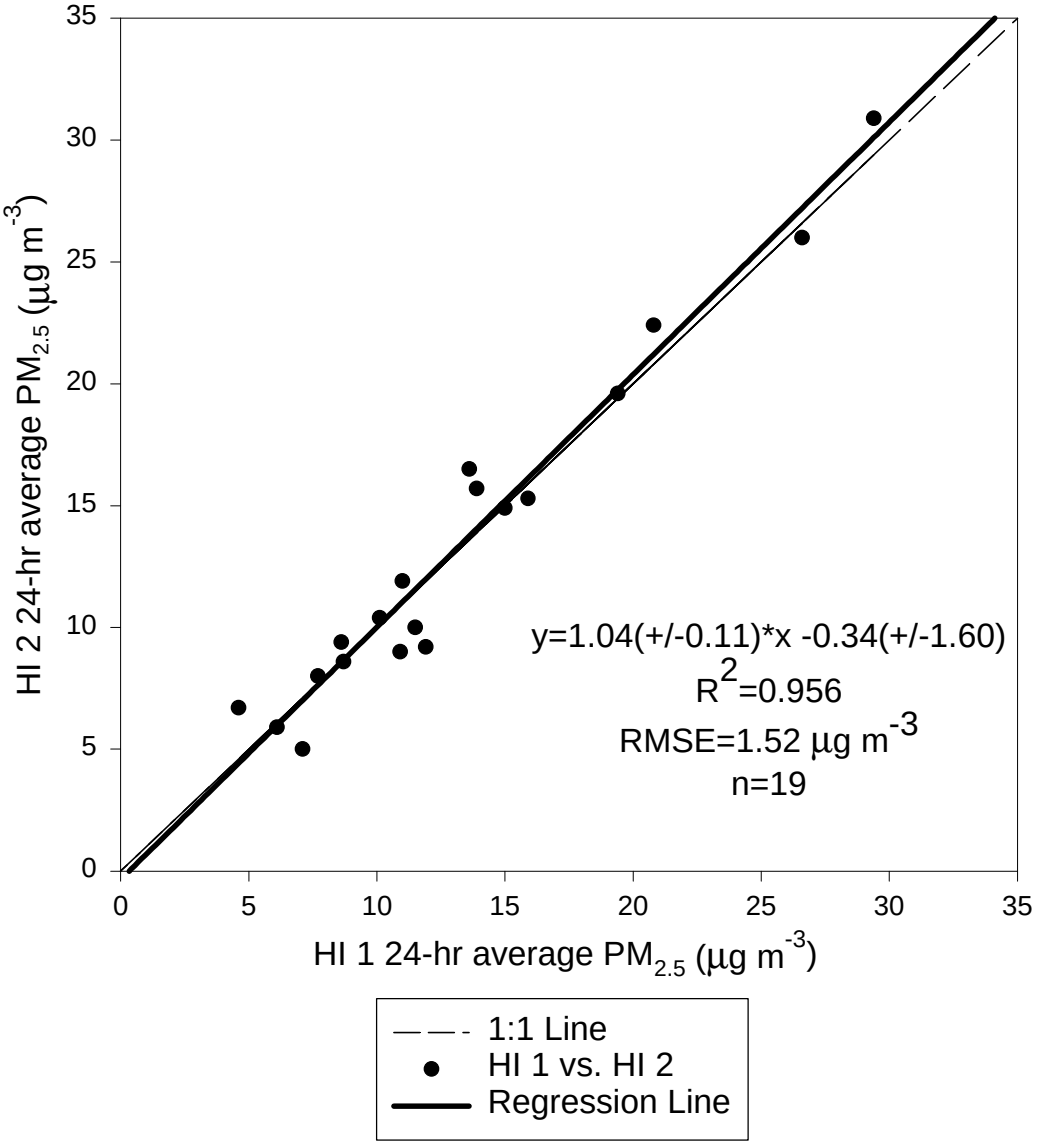

Figure A-4. Scatter plot of HI 1 vs. HI 2 24-hr average $PM_{2.5}$ measurements

January 6, 2000 through April 9, 2000
Ambient air in Athens, GA

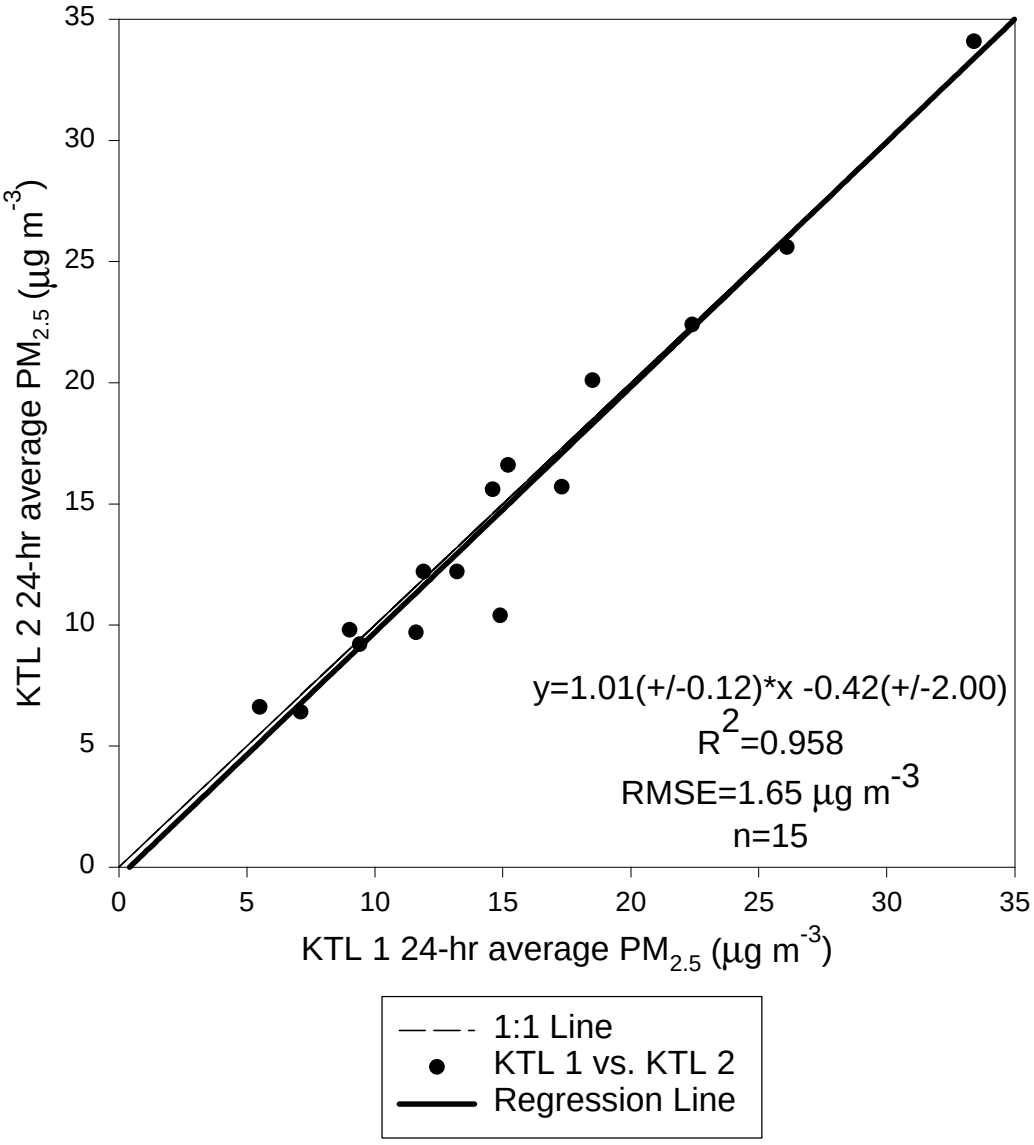

Figure A-5. Scatter plot of KTL 1 vs. KTL 2 24-hr average $PM_{2.5}$ measurements

January 6, 2000 through April 9, 2000
Ambient air in Athens, GA

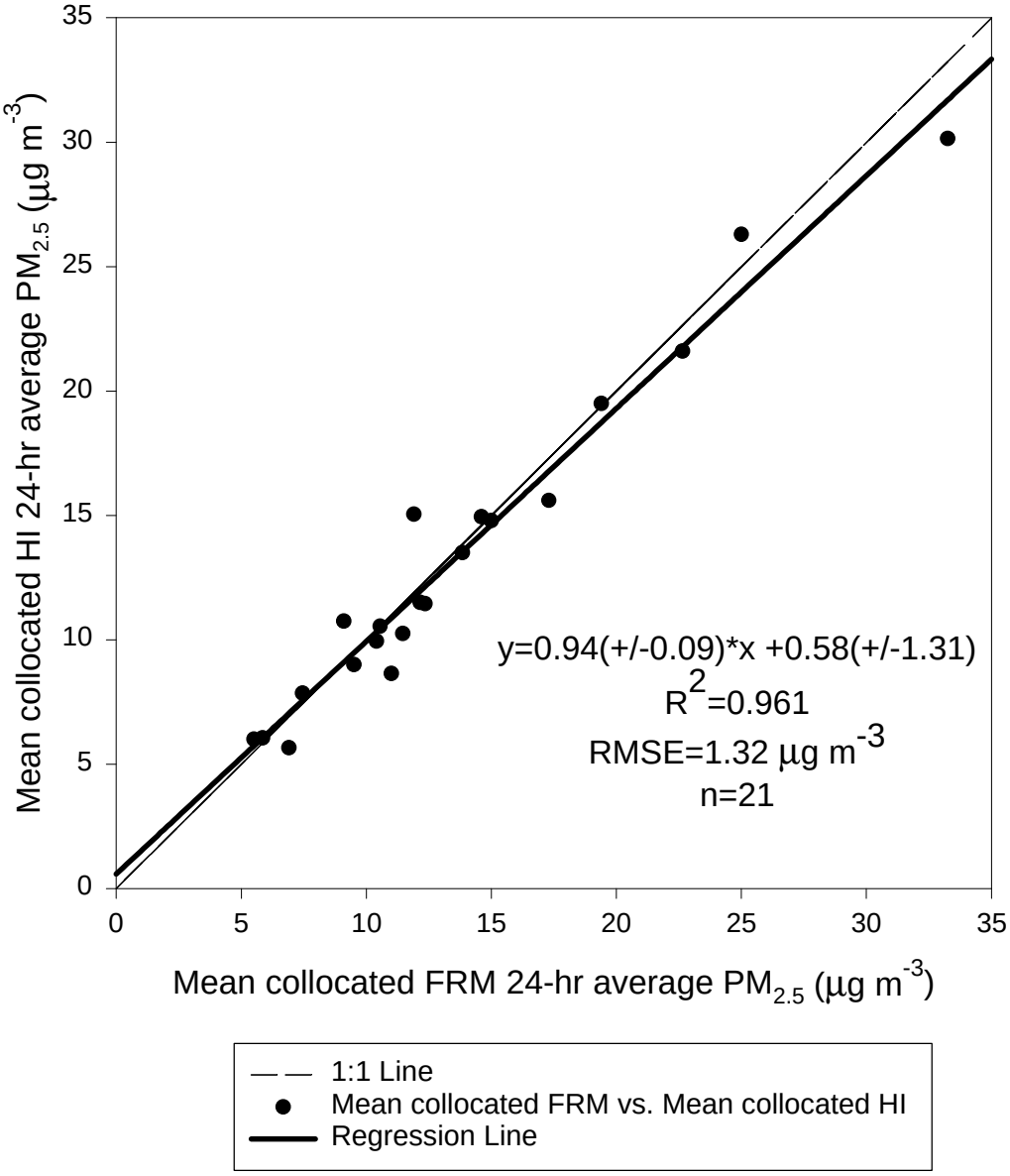

Figure A-6. Scatter plot of mean collocated FRM vs. mean collocated HI 24-hr average $PM_{2.5}$ measurements

January 6, 2000 through April 9, 2000
Ambient air in Athens, GA

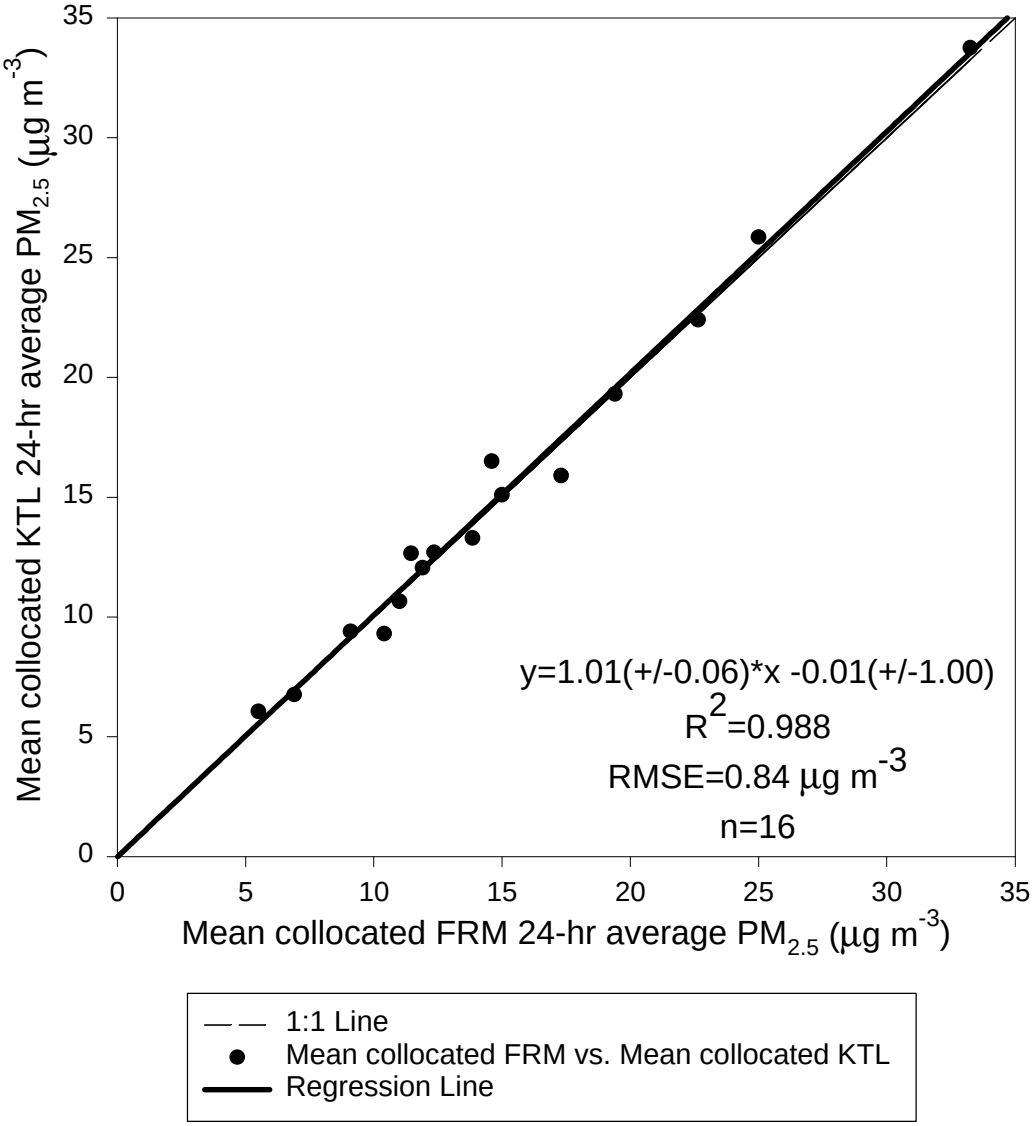

Figure A-7. Scatter plot of mean collocated FRM vs. mean collocated KTL 24-hr average $PM_{2.5}$ measurements

January 6, 2000 through April 9, 2000
Ambient air in Athens, GA

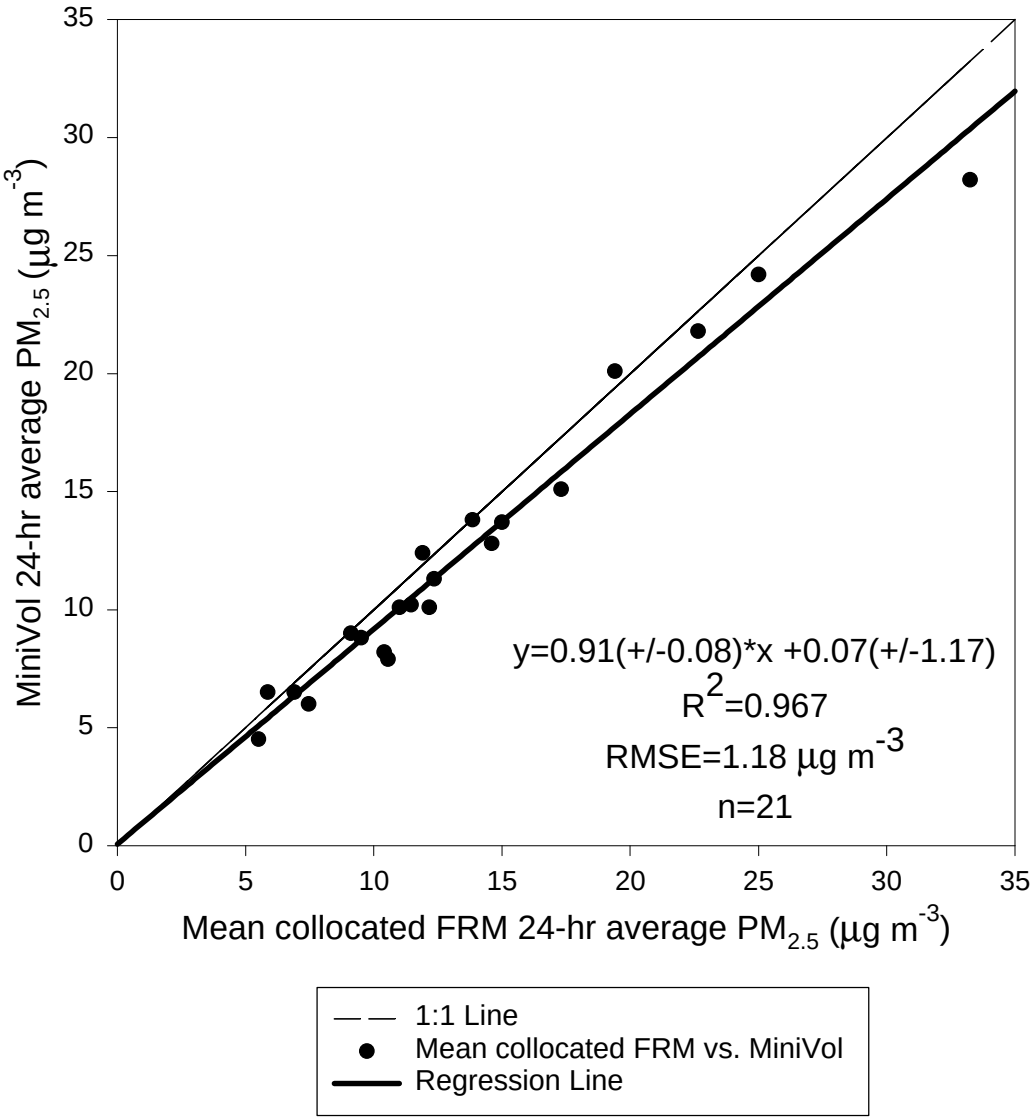

Figure A-8. Scatter plot of mean collocated FRM vs. MiniVol 24-hr average $PM_{2.5}$ measurements

January 6, 2000 through April 9, 2000
Ambient air in Athens, GA