Multi-Site Evaluations of Candidate Methodologies for Determining Coarse Particulate Matter (PMc) Concentrations

Paper # 10

Robert W. Vanderpool, Thomas G. Ellestad, Mary K. Harmon, Timothy D. Hanley, Richard D. Scheffe, and Elizabeth T. Hunike
United States Environmental Protection Agency, 109 T.W. Alexander Drive, Research Triangle Park, NC 27711

Paul A. Solomon
United States Environmental Protection Agency, P.O. Box 93478, Las Vegas, NV 89193

Robert W. Murdoch, Sanjay Natarajan, and Christopher A. Noble
Research Triangle Institute, 3040 Cornwallis Road, Research Triangle Park, NC 27709

Jeffrey L. Ambs
Rupprecht and Patashnick, 26 Tech Valley Road, East Greenbush, NY 12061

Gilmore J. Sem
TSI Inc., 500 Cardigan Road, Shoreview, MN 55126

John Tisch
Tisch Environmental Inc., 145 S. Miami Avenue, Cleves, OH 45002

ABSTRACT

Comprehensive field studies were conducted to evaluate the performance of sampling methods for measuring the coarse fraction of PM_{10} in ambient air. Five separate sampling approaches were evaluated at each of three sampling sites. As the primary basis of comparison, a discrete difference method was used which employs two designated FRM samplers, one to measure $PM_{2.5}$ and the other PM_{10}. The numerical difference of these reference method concentrations (PM_{10}-$PM_{2.5}$) represented an estimate of PMc. A second sampling approach involved a sequential dichotomous sampler, which provided both $PM_{2.5}$ and PMc measurements. In both of these filter-based, time-integrated measurement approaches, the collected aerosol mass was analyzed gravimetrically in the laboratory under controlled conditions. Three continuous coarse particle samplers that measure PMc directly with a time resolution of 1 hour or less were also evaluated. One such sampler was a commercially available system based on beta attenuation, the second was based on TEOM technology. Both of these measurement approaches used dichotomous virtual impactors for separating fine and coarse particles. The third real-time sampler evaluated was an aerodynamic particle sizer (APS) that measures the aerodynamic

diameter of individual particles, calculates the mass of the particle based on an assumed particle density, then sums the mass within the size range of interest to estimate the PMc mass concentration.

Sampling sites and timing of the studies were selected to provide diverse challenges to the samplers with respect to aerosol concentration, aerosol particle size distribution, and aerosol composition. Results from performance evaluations of the candidate PMc samplers at Gary, IN, Phoenix, AZ, and Riverside, CA will be presented.

INTRODUCTION

In response to increasing evidence of the adverse health effects associated with exposure to ambient fine particles, the United States Environmental Protection Agency (EPA) promulgated in 1997 a national ambient air quality standard (NAAQS) for $PM_{2.5}$[1]. Accompanying the standard were strict design and performance requirements which candidate $PM_{2.5}$ samplers must meet in order to be approved by EPA for use in making compliance measurements[2]. The 1997 regulations retained the existing annual PM_{10} standard and made only slight modifications to the statistical basis upon which to assess compliance with the 24-hour PM_{10} standard.

Based on subsequent litigation, the U.S. Court of Appeals for the District of Columbia reviewed the 1997 regulations and upheld EPA's promulgation of the $PM_{2.5}$ standard. While acknowledging the need to regulate coarse particles, the Court vacated the 1997 PM_{10} standard after concluding that PM_{10} is a "poorly matched indicator for coarse particulate pollution" because PM_{10} includes the $PM_{2.5}$ fraction. EPA did not appeal this ruling and now intends to promulgate a new NAAQS for PMc (i.e. the coarse fraction of PM_{10}).

Inherent to any new NAAQS is the need for sampling and analysis methods capable of measuring the new metric with known quality. In support of this goal, the purpose of this field study series was to conduct a survey of available instrumentation designed to measure the coarse fraction of PM_{10}, and to conduct a multi-site performance evaluation of these instruments. Sampling sites were selected in order to evaluate the instruments under a wide variety of environmental conditions, particle concentrations, particle size distributions, and particle compositions. At three separate cities (Gary, IN, Phoenix, AZ, and Riverside, CA) thirty daily, 22- hour tests were conducted. In addition to filter-based samplers which provide integrated test results, near real-time PMc monitors were evaluated which possess time resolutions of one hour or less. Multiple monitors of each type were used in order to determine the inherent precision of each sampler's design.

This report provides a description of the instruments evaluated in this study, outlines the sampling and analysis procedures used to conduct the performance evaluations, describes the characteristics of each of the three sampling sites, and provides a summary of test results. Because chemical analysis of archived filters has not yet been completed, this report will focus solely on mass concentration results reported by the various PMc samplers.

DESCRIPTION OF PMc SAMPLERS

Selection of the samplers to be involved in the field comparison study was based on the

following criteria. First, all samplers must be designed to provide a measurement of the mass concentration of PMc aerosols based on aerodynamic diameter. Selected filter-based samplers must be capable of providing integrated samples at least every 24 hours and use the $PM_{2.5}$ FRM's standard cassette and Teflon afterfilter. Selected continuous and semi-continuous instruments must be capable of providing PMc mass measurements at least every 1 hour. All samplers must be capable of automated operation over a period of 24 hours with active control of flow rates. Last, all selected samplers must be either commercially available or in the final prototype stage of their design.

Based on these criteria, five separate PMc measurement approaches were selected for evaluation in this study. Table 1 lists each sampler used in this study, its manufacturer, and the number of samplers used at each sampling site. For the filter-based samplers, the filter composition is listed along with the species to be determined during the filter's post-sampling gravimetric and/or chemical analysis. Due to funding constraints, not all the collected filters could be chemically analyzed. Instead, a representative subset of archived filters from each site was selected for chemical analysis based on the review of the comparative mass concentration results.

Table 1. Inventory of samplers used in the performance evaluation.

Measurement Method	PM Metric	Sampler Manufacturer(s)	Number Used	Filter Composition	Species Analyzed
Integrated FRM	PM_{10}	BGI, R&P, AND	3	Teflon	Mass, sulfate, nitrate, metals
Integrated FRM	PM_{10}	BGI	1	Quartz	EC, OC
Integrated FRM	$PM_{2.5}$	BGI, R&P, AND	3	Teflon	Mass, sulfate, nitrate, metals
Integrated FRM	$PM_{2.5}$	AND	1	Quartz	EC, OC
Integrated Dichot, sequential	$PM_{2.5}$, PMc	R&P	3	Teflon	Mass, sulfate, nitrate, metals
Integrated Dichot, sequential	$PM_{2.5}$, PMc	R&P	1	Quartz	EC, OC
TEOM	PMc	R&P	3	none	none
Beta Attenuation	$PM_{2.5}$, PMc	Tisch	3	none	none
Time of Flight (APS)	PMc	TSI	2	none	none
			Total = 20		

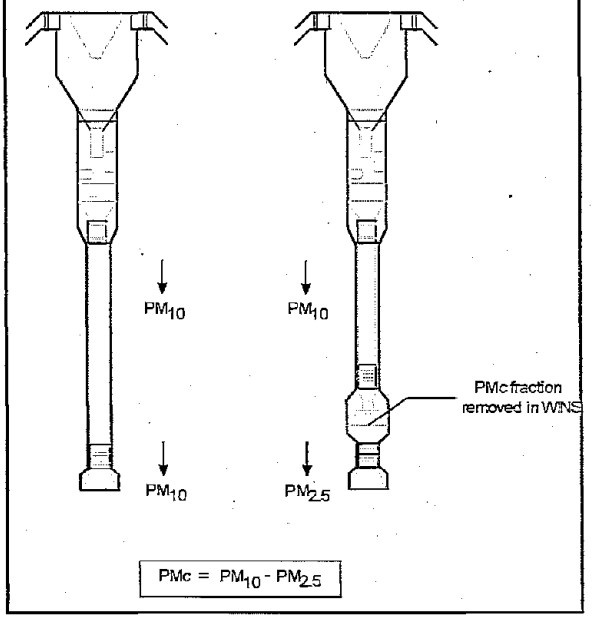
The voluntary participation and involvement of the PMc sampler manufacturers during this study was a critical component of the study's success. With the exception of the $PM_{2.5}$ and PM_{10} FRM samplers which were supplied by EPA, all field samplers in this study were supplied by their respective manufacturers. The supplied samplers all represented the latest models of each design

and were equipped with the most current hardware, firmware, and software. All manufacturers supervised installation and calibration of their respective samplers during the initial shakedown tests conducted in Research Triangle Park (RTP), NC and provided technical reviews of SOPs written for the instrument's setup, calibration, and operation. Each manufacturer was also provided the opportunity to visit each field site during site setup in order to verify the working condition of their samplers. At the completion of sampling at each field site, each manufacturer was supplied their respective field data in order to ensure that their sampler data was being properly retrieved from the instrument, correctly analyzed, and correctly interpreted.

Collocated $PM_{2.5}$ and PM_{10} FRM Samplers

In the first PMc measurement approach, commonly referred to as the "difference method", a designated $PM_{2.5}$ FRM sampler is collocated with a designated PM_{10} FRM sampler. For accurate determination of PMc concentrations, the PM_{10} sampler is simply a designated $PM_{2.5}$ FRM with its WINS fractionator replaced by a straight downtube (Figure 1). Both samplers are installed, calibrated, operated, and analyzed using standard $PM_{2.5}$ protocols. The two samplers thus have identical inlet aspiration characteristics, produce identical PM_{10} fractions, and collect aerosol at the same face velocity through the same filter media. At the completion of concurrent sampling periods, the PMc concentration is calculated as the numerical difference between the measured PM_{10} concentration and the measured $PM_{2.5}$ concentration. Due to its fundamental measurement principle, the difference method was used as the basis of comparison upon which to evaluate the performance of the other PMc samplers in the study. For purposes of this paper, data collected using this method is termed "PMc FRM" data.

Figure 1. Schematic diagram of the FRM samplers used in the PMc difference method.



In this study, a designated PM_{10}-$PM_{2.5}$ FRM pair was used from each of three separate sampler manufacturers: Thermo-Andersen (AND), BGI, and Rupprecht and Patashnick (R&P). Each of these six FRM samplers were operated with preweighed Teflon filters for subsequent gravimetric and ion chromatography (IC) or x-ray fluorescence (XRF) analysis. A fourth set of $PM_{2.5}$ and PM_{10} FRM samplers was used and both sampler's were equipped with a quartz filter to enable subsequent thermal optical measurement of the aerosol's elemental carbon (EC) and organic carbon (OC) constituents. In this study, the prefired quartz filters were not analyzed gravimetrically but were archived under cold conditions for subsequent EC/OC analysis.

R&P Model 2025 Sequential Dichotomous (Dichot) Sampler

The Model 2025 dichot was designed to provide integrated measurement of both fine and coarse fractions of a PM_{10} aerosol. The sampler actively provides volumetric flow control through a standard 16.7 actual liters per minute (alpm) PM_{10} inlet. Following the aspirated aerosol's fractionation in the inlet's internal fractionator, the resulting PM_{10} aerosol enters a virtual impactor where the aerosol is then split into major and minor flow streams. Ideally, the major flow (maintained at 15 lpm) is intended to collect only the $PM_{2.5}$ fraction of the PM_{10} aerosol while the minor flow (maintained at 1.7 alpm) is intended to collect only the PMc fraction of the PM_{10} aerosol. In practice, however, this size fractionation is never ideal and 10% of the $PM_{2.5}$ mass theoretically deposits onto the PMc filter. The presence of these fine particles is numerically accounted for during subsequent calculation of the PMc concentration. Assuming that particle losses within the instrument are negligible, the sum of the measured $PM_{2.5}$ and PMc concentrations provide a measure of the ambient aerosol's PM_{10} concentration.

The Model 2025 sequential dichot allows unattended, multi-day operation through the use of a filter exchange mechanism for transferring filter cassettes from a supply tube to the sampling position, then conducting a post-sampling transfer of the cassettes to a storage tube. During this study, however, the multi-day capability of the Model 2025 was not utilized and supply tubes were manually loaded with only one cassette shortly before each test and the post-sampling cassette was manually retrieved from the storage tube shortly after each test. Procedures for gravimetric and chemical analysis of the Model 2025's filters were identical to those of the FRM's filters.

Four separate R&P sequential dichotomous samplers were used during this study, three of which were equipped with Teflon filters while the fourth was equipped with prefired quartz filters to enable determination of elemental and organic carbon components of the ambient aerosol.

Tisch Inc. Model SPM-613D Dichotomous Beta Gauge

Manufactured by Kimoto Electric Co., LTD., the Tisch SPM-613D dichot beta gauge is designed to provide near real-time measurement of both the fine and coarse fractions of the PM_{10} aerosol. Similar to the R&P Model 2025 dichot, the SPM-613D aspirates the ambient aerosol through a standard 16.7 lpm inlet and introduces the fractionated PM_{10} aerosol into a custom designed virtual impactor. The virtual impactor in the SPM-613D has different dimensions than that of the R&P design and operates its major and minor flow rates at slightly different flow rates, 15.2 lpm and 1.5 lpm, respectively. Flow control in the two SPM-613D channels is monitored using separate mass flow sensors. The system's flow control system, however, is designed to maintain the calibrated mass flow rate and thus does not maintain true volumetric flow rates through the inlet at actual ambient temperature and pressure conditions. By conducting flow rate calibrations at the sampler's inlet under actual temperature and pressure conditions, however, the effect of this lack of volumetric flow control is minimal if ambient conditions do not differ substantially from those existing during the flow calibration.

Downstream of the SPM-613D's virtual impactor, the separate fine and coarse flow streams are continuously collected on a paper roll composed of low hygroscopicity polyfon.

Following each hour of aerosol collection, the attenuation of ^{147}Pm beta rays by each channel's aerosol deposit is quantified using two separate sets of beta sources and detectors. Based on previous span calibrations performed by the user, the theoretical relationship between beta attenuation and collected aerosol mass is used to estimate the mass of each separate aerosol deposit. Because beta rays are also attenuated by condensed water, an external heater is located downstream of the sampler's inlet and maintains the temperature of the aspirated airstream above 25 °C. As in the R&P 2025 dichot, numerical corrections are made to account for the theoretical mass of fine particulates contained within the SPM-613D's coarse channel filter. Three identical SPM-613D beta gauges were used during the course of the study at all three sampling sites.

R&P Continuous Coarse TEOM Monitor

As designed by Misra, et al.[3] and licensed to R&P, the coarse TEOM was designed to provide a near real-time measurement of PMc concentrations. The instrument aspirates ambient aerosol at 50 lpm through a custom size-selective inlet, which was made by modifying a standard 16.7 lpm size-selective PM_{10} inlet by adjusting the internal dimensions in an effort to provide a 10 µm cutpoint. Downstream of the inlet, the PM_{10} fraction then enters a custom virtual impactor whose major and minor flow rates are 48 lpm and 2 lpm, respectively. In this design, the fine fraction (major flow) is collected in a replaceable total filter and the collected fine aerosol mass is not subsequently quantified. Downstream of the virtual impactor, coarse aerosols in the minor flow stream are first heated to 50 °C to minimize interferences from particle bound water and are then deposited in a standard R&P 1400a Tapered Element Oscillating Microbalance (TEOM). The mass of the deposited aerosol is then estimated based on the observed change in vibrational frequency of the TEOM filter during the collection period. Due to the high flow rate ratio between the total and minor flows (25 to 1), no correction is made for the mass of fine particles on the coarse filter in this design. The PMc mass concentration is then calculated as the measured coarse mass divided by the volume of ambient air aspirated during the sampling event. Three replicate R&P coarse TEOMs were used during this field study in order to determine the inherent measurement precision of the samplers.

TSI Inc. Model 3321 Aerodynamic Particle Sizer (APS)

The final measurement approach used in the field study involved the TSI Inc. Model 3321 APS to estimate the mass of ambient coarse particles based on their aerodynamic properties in an accelerating flow stream. To adapt the 5 lpm APS to field use, a standard 16.7 lpm PM_{10} inlet was used in conjunction with a custom designed flow splitter located downstream of the inlet. In the splitter, a sharp-edged, isokinetic nozzle extracts a representative sample of the PM_{10} aerosol for measurement in the APS. The remaining 11.7 lpm portion of the PM_{10} aerosol was drawn through a total filter using a volumetrically controlled vacuum source. The mass of the aerosol collected on the total filter was not quantified.

The 5 lpm representative aerosol sample is then introduced into the APS and the aerodynamic diameter of individual particles estimated using time of flight technology. The volume of each particle is then calculated based on its measured aerodynamic diameter and a

particle density specified by the user. For purposes of this field study, a particle density of 2 g/cm^3 was assumed as representative for the coarse fraction of PM_{10} aerosols. The mass concentration of PMc aerosols is then calculated as the sum of the mass of all particles penetrating the PM_{10} inlet whose aerodynamic diameters were greater than 2.5 micrometers. Because the APS is only capable of resolving particles larger than approximately 0.7 micrometers aerodynamic diameter, the system is not applicable for measurement of either $PM_{2.5}$ or PM_{10} ambient concentrations because particulate mass less than 0.7 micrometers is not quantified.

It should be noted that the primary purpose of incorporating the two APS units into the field study was to provide ambient aerosol size distribution information at each site. Because the APS's measurement method has legitimate potential for providing continuous PMc concentration measurements, it was evaluated in this study in the same manner as the other PMc samplers.

SITE SETUP AND OPERATING PROCEDURES

All field and laboratory activities associated with this study were conducted by Research Triangle Institute (RTI) under EPA contract 68-D-00-206. Prior to conducting the study, RTI developed a Quality Assurance Project Plan (QAPP) which encompassed all aspects of the study's field and laboratory activities. The QAPP was subsequently reviewed and approved by QA personnel within EPA's Office of Research and Development (ORD) prior to initiation of the study. All field and laboratory operations of the study were also reviewed and approved during a comprehensive Systems Audit conducted by ORD prior to the field tests.

The multi-site performance evaluations of the 20 separate field samplers presented a unique logistical challenge. With the exception of the FRM samplers and the R&P dichots, none of the other samplers have weather enclosures and must thus be protected from the elements during sampling. To enable efficient transportation of all field equipment and to house the field samplers, a 25 foot long motor home was adapted for use in this study. The twelve FRM and R&P dichot samplers were installed either on the roof of the motor home or on a 10' by 10' auxiliary platform positioned immediately adjacent to the motor home. The remaining eight PMc samplers were installed inside the motor home with their downtubes extending through the roof of the motor home and attached to their respective inlets. The motor home's environmental controls maintained the interior temperature at 23 °C ± 3 °C during all field tests. Per compliance testing requirements, the inlets of all samplers were installed 2 m ± 0.2 m above the sampling platform and all samplers were spaced horizontally at least 1 m apart from each other. At each site, the motor home and auxiliary platform were free of nearby obstructions which might adversely influence the spatial uniformity of PMc concentrations.

Prior to each field test, all samplers were cleaned and leak-checked. Each sampler was then calibrated for volumetric flow rate, ambient temperature, and ambient pressure measurement using a calibrated transfer standard (BGI DeltaCal). For calibration of the 50 lpm of the R&P coarse TEOM, a BGI TriCal was equipped with a 55 lpm capacity flow module which had been specifically designed for this purpose. Following the calibration of each instrument, a performance audit was conducted using a separate audit device and any necessary adjustments were made to the instruments. In addition to the initial audit conducted at each field site, performance audits were also conducted following Run 15 and Run 30. Field blank tests of the filter-based samplers were conducted at the same frequency as that of the performance audits.

At each sampling site, 30 daily, 22-hour tests were conducted from 11 am (local time) to 9 am of the following morning. The two hour interval between successive tests enabled the site operator sufficient time for sample changeover, data recording, and minor maintenance while still allowing for daily sampling. Typically, a 45 day test period was required to complete site setup, 30 days of sampling, and site shutdown.

Gravimetric analysis of the filter-based samplers' teflon filters was conducted both in the EPA weighing facility in RTP, NC and at each sampling site. In RTP, presampling filters were equilibrated and preweighed in an environmentally controlled chamber whose temperature and relative humidity setpoints were 22 °C and 35%, respectively. All filter weighings were conducted using a Cahn C-44 microbalance which had a readability of 1 μg and a capacity of 5 g. The analytical balance was tared and calibrated prior to each weighing session and Class 1 calibration weights were used during each session to verify the balance's internal calibration. In order to increase the confidence in the gravimetric analysis, 100% replicate weighings (with a 5 μg reweigh threshold) were used for each filter during all preweighing and postweighing operations. Quality control also included the use of three laboratory blank filters during each weighing session. At the completion of the preweighing in RTP, the filters were loaded in sampling cassettes, the cassettes sealed with metal endcaps, and the cassettes placed in Thermo-Andersen cassette canisters. The canisters were then shipped to the field site in coolers designed to maintain postsampling filters at temperatures below 2 °C.

Upon receipt of the preweighed filter from RTP, field personnel would then unpack and equilibrate the filters in a weighing facility setup within a hotel room. Through careful monitoring of the room's conditions and through use of an automated dehumidifier, site personnel were able to maintain the site's weighing conditions within allowable temperature and relative humidity limits. Presampling and postsampling site weighings were conducted using a Sartorius MC5 microbalance with the same capability as the Cahn microbalance used for the RTP weighings. Identical weighing protocols were used at all field sites and at the RTP weighing facility. Once postsampling filters were weighed at the site, they would be shipped to RTP for final postweighing and subsequent archiving under cold conditions. Conducting filter weighing at the field site enabled faster determination of test results than could be obtained if samples were shipped back to RTP. Conducting filter weighing at the site and at RTP also enabled measurement of particle losses which might occur during shipping. Last, site weighing provided valid test results in the event that a cooler might be inadvertently damaged or lost during its shipment back to the RTP weighing facility.

SITE CHARACTERISTICS

Following the initial installation and evaluation of the PMc samplers in RTP, NC to verify the proper operating condition of the samplers and to finalize operating protocols, successive field tests were conducted in Gary, IN, Phoenix, AZ, and Riverside, CA. The following section will provide a description of these three sites along with the meteorological conditions and aerosol characteristics encountered during each site's field tests.

Gary, IN

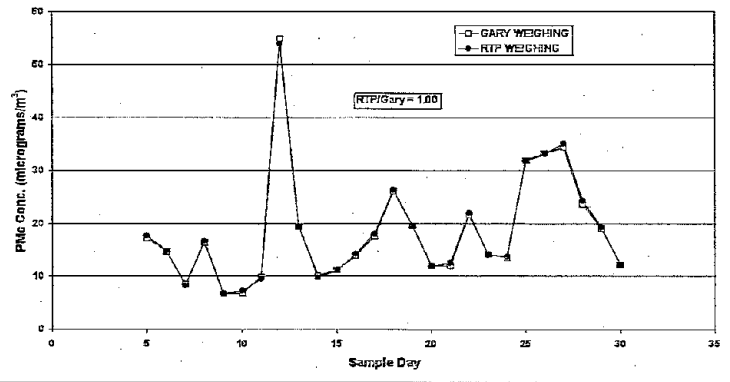
The Gary, IN site was selected as representing a midwest industrial city where primary PMc aerosols are predominantly generated by industrial activity rather than by wind blown soils. Selection and setup of the Gary, IN sampling site was made in cooperation with personnel from the Indiana Department of Environmental Management. This State and Local Air Monitoring (SLAM) site (AIRS # 18-089-0022) is located approximately 2 km south of Lake Michigan and is immediately adjacent to the property line of a steel mill. Nearby sources of emissions include the steel mill which was located approximately 0.7 km northwest of the site and a 0.5 km long open coal pile which was located approximately 0.5 km northeast of the site.

The 30 days of testing at the Gary site were conducted from March 6 to April 7, 2003. Weather at the site was typically cloudy, windy, and cold and only one rain event occurred during the study period. Temperatures at the site ranged from -15.1 °C to 27.8 °C and a mean daily site temperature of 4.6 °C was recorded.

As measured by the three collocated FRM samplers, $PM_{2.5}$ concentrations measured at the Gary site during the tests ranged from 10.3 $\mu g/m^3$ to 46.9 $\mu g/m^3$ with a measured mean of 22.8 $\mu g/m^3$. Excellent inter-manufacturer agreement was observed among the filter-based $PM_{2.5}$ FRM samplers as expressed by the coefficient of variation (CV) of 1.5%. As expressed by a coefficient of variation of 2.4%, excellent inter-manufacturer agreement was also observed for the PM_{10} FRM measurements. PM_{10} concentrations measured during the tests ranged from 22.6 $\mu g/m^3$ to 85.0 $\mu g/m^3$ with a measured mean of 42.6 $\mu g/m^3$. PMc concentrations (expressed as the numerical difference between collocated PM_{10} and $PM_{2.5}$ FRM measurements), ranged from 4.5 $\mu g/m^3$ to 58.1 $\mu g/m^3$ with a measured mean of 19.8 $\mu g/m^3$. Inter-manufacturer precision of PMc concentrations was determined to be 5.7% CV. As indicated by a mean $PM_{2.5}$ /PM_{10} ratio of 0.55 during the 30 sampling events, slightly more than one-half of the site's PM_{10} aerosol was associated with $PM_{2.5}$ aerosols. $PM_{2.5}$ /PM_{10} ratios ranged from 0.32 to 0.83 during the 30 days of testing indicating that the size distribution of ambient aerosols was quite variable during the month-long field tests. Predominant winds from the direction of the nearby steel mill typically contributed to $PM_{2.5}$ concentrations at the site while winds predominating from the direction of the open coal piles resulted in measurement of high PMc site concentrations.

Filter weighing at the Gary site began with Run 5 filters. As indicated in Figure 2, excellent agreement was seen between PMc concentrations based on the site weighings versus the RTP weighings during Runs 5 through 30. The filter shipping and handling protocols designed for the study, therefore, appeared to result in negligible $PM_{2.5}$ or PM_{10} particle loss from the FRM filters during their transport from the field site to the RTP weighing facility.

Figure 2. Site versus RTP, NC weighing results of PMc FRM concentrations in Gary, IN.



Phoenix, AZ

Tests were conducted in Phoenix, AZ during early summer

of 2003 in order to challenge the coarse particle samplers with high concentrations of dry, wind blown crustal materials. Through cooperation with personnel at the Air Quality Division of the Maricopa County Environmental Services Department, the county-operated Durango Complex sampling site (AIRS # 04-013-9812) in the southwestern portion of Phoenix was selected as an appropriate field site. The site is impacted from the east and north by nearby commercial districts and two main interstate highways. With the predominant wind direction being from the west and southwest, however, the site is primarily impacted by large windblown soils originating from nearby earthmoving equipment and non-vegetated, open fields.

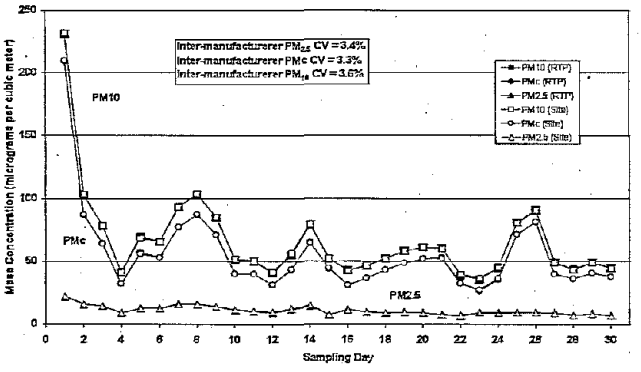
The month-long field tests at the Phoenix sampling site were conducted from May 14 to June 15, 2003. Weather at the site was typically clear, windy, and very hot and no rain events occurred during the 30 day study period. Temperatures at the sampling site ranged from 17.1 °C to 43.5 °C and a mean daily site temperature of 32.3 °C was recorded.

$PM_{2.5}$ concentrations measured during the Phoenix tests ranged from 6.4 $\mu g/m^3$ to 22.0 $\mu g/m^3$ with a measured mean of 11.0 $\mu g/m^3$. As observed during the Gary tests, excellent inter-manufacturer agreement was achieved among the filter-based FRM samplers. As expressed by the coefficient of variation, mean inter-manufacturer precision for $PM_{2.5}$ was determined to be 3.4%. As expressed by a coefficient of variation of 3.3%, excellent inter-manufacturer agreement was also observed for the PM_{10} FRM measurements. PM_{10} concentrations measured during the tests ranged from 35.6 $\mu g/m^3$ to 230.9 $\mu g/m^3$ with a measured mean of 66.6 $\mu g/m^3$. PMc concentrations (expressed as the numerical difference between collocated PM_{10} and $PM_{2.5}$ FRM measurements), ranged from 26.5 $\mu g/m^3$ to 209.0 $\mu g/m^3$ with a measured mean of 55.6 $\mu g/m^3$. Inter-manufacturer precision of PMc concentrations measured by the three FRM pairs was determined to be 3.6% CV. As indicated by the mean $PM_{2.5}/PM_{10}$ ratio of 0.18 during the 30 sampling events, a large fraction of the site's PM_{10} concentration was associated with PMc aerosols. $PM_{2.5}/PM_{10}$ ratios ranged from 0.10 to 0.28 which indicated that coarse particle mass dominated the PM_{10} concentrations during each day of the Phoenix tests. Figure 3 depicts the daily dominance of the coarse particles in the Phoenix area and also shows the strong agreement obtained between the FRM filter weighings conducted at the sampling site versus those conducted at the RTP weighing facility.

Riverside, CA

The Riverside, CA sampling site was selected as a west coast site where significant secondary fine mode aerosols might be present in conjunction with primary coarse aerosols. Selection and setup of the Riverside site was made through cooperation with the University of California-Riverside (UCR). The monitoring site is located on the grounds of UCR's Agricultural

Figure 3. Site versus RTP, NC weighing of $PM_{2.5}$, PMc, and PM_{10} concentrations at the Phoenix site.



Operations Center and is operated by the South Coast Air Quality Management District (California ARB Site # 33162). Local sources of ambient aerosols include those from agricultural research activities as well as from mobile source emissions in the area.

Field tests were conducted at the Riverside sampling site from July 23 to August 24, 2003. Weather at the sampling site during the 30 daily tests was typically warm with clear or partly cloudy skies. No rain events occurred during the Riverside field tests although morning fog was occasionally observed at the site. Temperatures at the site ranged from 15.4 °C to 40.4 °C and a mean daily site temperature of 25.9 °C was recorded.

As had been experienced during the Gary and Phoenix sites, excellent inter-manufacturer agreement was observed among the filter-based FRM samplers. As expressed by the coefficient of variation, mean inter-manufacturer precision for $PM_{2.5}$ was determined to be 3.1%. Daily $PM_{2.5}$ concentrations measured during the tests ranged from 9.9 $\mu g/m^3$ to 32.7 $\mu g/m^3$ with a measured mean of 17.7 $\mu g/m^3$. As expressed by a coefficient of variation of 2.9%, excellent inter-manufacturer agreement was also observed for the PM_{10} FRM samplers. PM_{10} concentrations measured during the tests ranged from 27.0 $\mu g/m^3$ to 69.3 $\mu g/m^3$ with a measured mean of 48.0 $\mu g/m^3$. PMc concentrations (expressed as the numerical difference between collocated PM_{10} and $PM_{2.5}$ FRM measurements), ranged from 16.2 $\mu g/m^3$ to 46.1 $\mu g/m^3$ with a measured mean of 30.4 $\mu g/m^3$. Inter-manufacturer precision of PMc FRM measurements was determined to be 4.1% CV. As indicated by the mean $PM_{2.5}/PM_{10}$ ratio of 0.37 during the 30 sampling events, approximately two-thirds of the sites PM_{10} concentration was associated with PMc aerosols. $PM_{2.5}/PM_{10}$ ratios ranged from 0.25 to 0.50 during the 30 days of testing at the Riverside site indicating that coarse particles dominated the PM_{10} aerosol during all tests.

TEST RESULTS

Federal Reference Method Samplers

As previously described, field tests involved the use of four sets of $PM_{2.5}$ and PM_{10} samplers from BGI, Andersen, and R&P. Because there exist no absolute standards for ambient particulate matter, the absolute accuracy of these devices cannot be determined from these tests. However, the performance of the three separate manufacturers' samplers with respect to each other can be calculated. As summarized in Table 2, the inter-manufacturer precision of the samplers was considered to be excellent for all three metrics ($PM_{2.5}$, PMc, and PM_{10}) at all three sampling sites. Calculating the PMc concentration as the numerical difference between collocated designated PM_{10} and $PM_{2.5}$ FRMs did not produce any zero or negative PMc concentrations.

Table 2. Inter-manufacturer precision of the collocated FRM samplers.

Metric	Gary, IN	Phoenix, AZ	Riverside, CA
$PM_{2.5}$	1.5%	3.4%	3.1%
PMc	5.7%	3.6%	4.1%
PM_{10}	2.4%	3.3%	2.9%

With the exception of a pump failure and a faulty ambient temperature sensor connection, few functional problems were experienced with the eight FRM samplers despite the wide range of environmental conditions experienced during the study. The three performance audits conducted at each sampling site revealed that the FRMs generally maintained their flow rate, temperature, and pressure calibrations within the required specifications. Overall data capture rate for the FRM samplers during the three site study was determined to be 99%.

R&P Dichotomous Samplers

Only two operational problems were experienced with the four R&P Model 2025 sequential dichotomous samplers during the study. In Gary, a faulty cassette seal in one of the dichots' coarse channels caused the majority of the coarse aerosol to bypass the collection filter. As a result, the coarse particle mass concentration measured by this instrument was significantly less than that measured by the other collocated dichots. The data for this sampler's coarse channel was thus invalidated. The second problem experienced with the Model 2025 dichots occurred during the latter half of the Phoenix tests where significantly low $PM_{2.5}$ and PM_{10} measurements were obtained by one of the dichots. At the completion of the Phoenix tests, this behavior was explained by the discovery of a dense spider web in the dichot's size selective inlet. The $PM_{2.5}$ and PM_{10} measurements for this instrument were thus invalidated for 17 of the 30 sampling events. At all sites, invalid data were not used to calculate daily aerosol mass concentrations nor used to estimate intrasampler precision. Discounting the invalid data obtained due to the presence of the spider web, overall data capture rate of the dichots during the study was 98%. Performance audits of the Model 2025 dichots indicated that they maintained their flow rate, temperature, and pressure calibrations within the required specifications.

Table 3 summarizes the field performance of the Model 2025 dichots at all three sites in comparison to the collocated FRM samplers. As the table indicates, excellent intrasampler precision was observed for the R&P dichots at all three sites for all three metrics. As an example, the precision (expressed as the coefficient of variation) in Gary for $PM_{2.5}$, PMc, and PM_{10} concentrations was determined to be 3.8%, 3.2%, and 1.9%, respectively. The largest coefficient of variation (4.2%) was observed in Phoenix for measurement of PMc aerosols.

Table 3. Performance of the R&P 2025 Dichot versus the FRM.

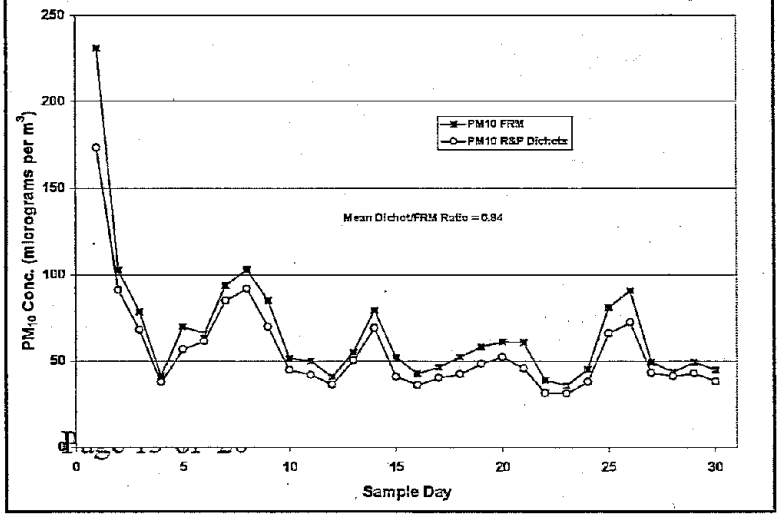
Metric	Performance Criteria	Gary, IN	Phoenix, AZ	Riverside, CA
$PM_{2.5}$	Dichot CV	3.8%	2.3%	1.3%
	Regression Equation (Dichot vs. FRM)	Dichot = 0.99*FRM + 0.0	Dichot = 1.24*FRM - 1.6	Dichot = 0.998*FRM + 0.0
	Coefficient of determination (R^2)	0.998	0.97	0.995
	Mean Dichot/FRM Ratio	1.00	1.09	1.00

Metric	Performance Criteria	Gary, IN	Phoenix, AZ	Riverside, CA
PMc	Dichot CV	3.2%	4.2%	1.7%
	Regression Equation (Dichot vs. FRM)	Dichot = 0.87*FRM + 0.39	Dichot = 0.70*FRM + 5.0	Dichot = 0.95*FRM + 0.25
	Coefficient of determination (R^2)	0.969	0.98	0.98
	Mean Dichot/FRM Ratio	0.90	0.79	0.96
PM_{10}	Dichot CV	1.9%	3.0%	1.2%
	Regression Equation (Dichot vs. FRM)	Dichot = 0.95*FRM - 0.47	Dichot = 0.75*FRM + 5.9	Dichot = 1.00*FRM - 1.21
	Coefficient of determination (R^2)	0.981	0.98	0.99
	Mean Dichot/FRM Ratio	0.94	0.84	0.97

In Gary and Riverside, the $PM_{2.5}$ concentrations measured by the R&P dichots agreed almost exactly with concentrations measured by the collocated FRM samplers. In Phoenix, however, the dichots consistently over-predicted the $PM_{2.5}$ concentration by about 9%. This over-measurement is hypothesized to be due to the inadvertent intrusion of coarse mode aerosols into the fine channel, which has been known to occur in virtual impactors[4].

The Model 2025 dichots consistently under-measured PMc concentrations at all three sites although results were highly correlated (mean R^2 equaled 0.976). A high coefficient of determination at a site indicates that the sampler's behavior, with respect to the collocated FRMs, was very consistent during the 30 days of testing. For PMc, mean sampler to FRM ratios at Gary, Phoenix, and Riverside were determined to be 0.90, 0.79, and 0.96, respectively. Summing the dichot's measured $PM_{2.5}$ and PMc concentration to estimate the PM_{10} concentration, it was observed that mean sampler to FRM ratios for PM_{10} in Gary, Phoenix, and Riverside were 0.94, 0.84, and 0.97, respectively. For Phoenix, therefore, 16% of the aspirated PM_{10} aerosol mass cannot be accounted for when compared to the collocated PM_{10} FRM samplers. The consistency of this behavior in Phoenix is illustrated in Figure 4. Although results are preliminary, recent follow-up testing in Phoenix has suggested that this particle loss may occur during postsampling transfer of the dichot's coarse filter cassette from the sampling position to the storage position. R&P is currently investigating engineering solutions to address

Figure 4. Timeline of R&P dichot versus FRM PM_{10} concentrations in Phoenix.



the observed measurement bias.

R&P Coarse TEOM Samplers

Few operational problems were experienced with the three R&P coarse TEOM monitors during the three site study. The exception occurred during the Riverside testing where the third coarse TEOM monitor consistently measured about 17% higher than the other two coarse TEOM units, which agreed extremely well with each other. The exact reason for the consistent difference between the third unit and the other two units is not known but may have been an operational problem associated with the TEOM control unit itself. In two successive tests, exchanging the inlets and virtual impactors between units three and two did not appear to correct the noted discrepancy. For purposes of calculating instrument precision at Riverside, therefore, data from TEOM unit three was not used.

Table 4 summarizes the field performance of the R&P coarse TEOM monitors at all three sampling sites in comparison to the collocated FRM samplers. Considering that these are automated samplers which provide both sampling and mass analysis, excellent intra-manufacturer precision was observed for the three coarse TEOM monitors at all three sites. The coarse TEOM precision at Gary, Phoenix, and Riverside was observed to be 4.4%, 6.6%, and 1.7%, respectively.

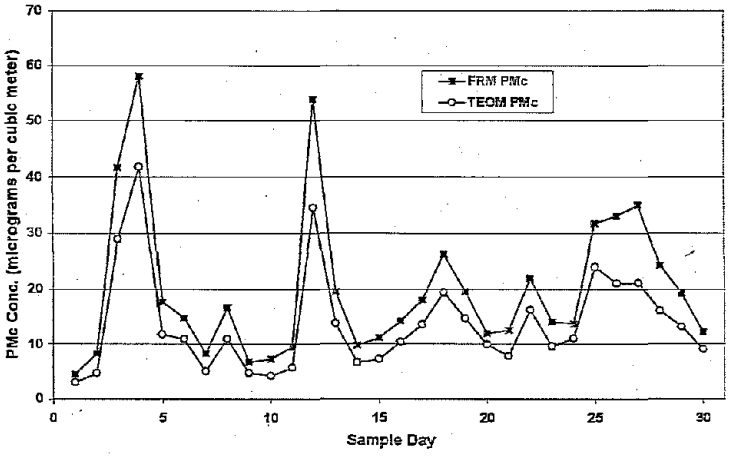
Table 4. Performance of the R&P Coarse TEOM versus the FRM.

Metric	Performance Criteria	Gary, IN	Phoenix, AZ	Riverside, CA
PMc	TEOM CV	4.4%	6.6%	1.7%
	Regression Equation (TEOM vs. FRM)	TEOM = 0.68*FRM + 0.2	TEOM = 0.79*FRM + 12.8	TEOM = 0.74*FRM - 0.64
	Coefficient of determination (R^2)	0.982	0.951	0.948
	Mean TEOM/FRM Ratio	0.69	1.05	0.76

At the Gary and Riverside field sites, the coarse TEOMs produced PMc values which were consistently lower than those measured by the collocated FRMs. On average, the coarse TEOMs provided PMc measurements that were 31% and 24% lower than the FRMs in Gary and Riverside, respectively. This underestimation may be partly due to the fact that the sampler's inlet reportedly provides an internal cutpoint closer to 9 μm than its 10 μm design cutpoint[5]. Note from the table that the data is strongly correlated for Gary and Riverside and that near zero intercepts were observed for regressions of the coarse TEOMs versus the collocated FRMs. The timeline presented in Figure 5 illustrates that the coarse TEOM monitors track the FRMs well but consistently provide an under-measurement of PMc concentrations. Based on the high coefficient of determination in Gary of 0.982, this behavior was very consistent as a function of concentration during the 30 day test period.

Better agreement between the coarse TEOMs and the FRM was observed during the May to June 2003 tests conducted in Phoenix. For these tests, the coarse TEOMs provided PMc concentrations that averaged 5% higher than those measured by the collocated FRM samplers. As depicted in Table 4, however, the slope and intercept for the TEOM versus FRM regression deviated significantly from one and zero, respectively.

Figure 5. R&P coarse TEOM versus FRM PMc concentrations in Gary, IN.



Tisch SPM-613D Dichotomous Beta Gauge Monitors

No significant operational problems were encountered during field operation of the Tisch SPM-613D samplers at the three sampling sites. Overall data capture rate was near 100% at all three sites.

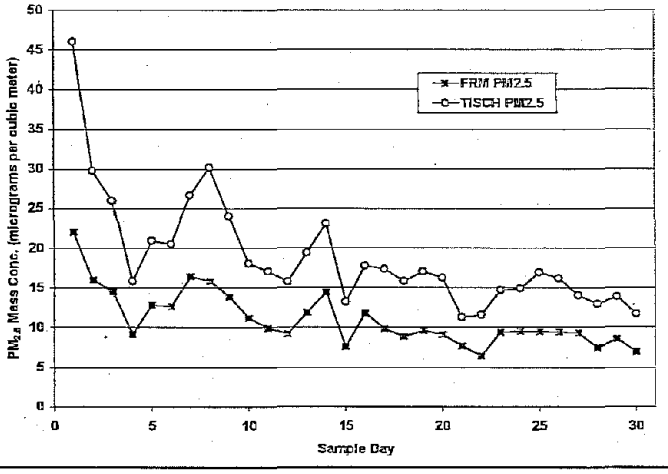
Table 5 summarizes the performance of the three Tisch units in comparison with the collocated FRM samplers. Inspection of the table reveals that precision of the samplers was generally good for all three metrics at all three sampling sites. In general, higher intra-sampler CV values (i.e. less precision) were observed during measurement of PMc concentrations than during measurement of $PM_{2.5}$ concentrations.

Table 5. Performance of the Tisch SPM-613D Beta Gauge Dichot versus the FRM.

Metric	Performance Criteria	Gary, IN	Phoenix, AZ	Riverside, CA
$PM_{2.5}$	Tisch CV	7.1%	5.9%	4.1%
	Regression Equation (Tisch vs. FRM)	Tisch = 1.17*FRM + 1.6	Tisch = 2.03*FRM - 3.4	Tisch = 2.07*FRM + 6.9
	Coefficient of determination (R^2)	0.945	0.946	0.904
	Mean Tisch/FRM Ratio	1.26	1.70	1.65
PMc	Tisch CV	10.5%	9.5%	5.8%
	Regression Equation (Tisch vs. FRM)	Tisch = 0.885*FRM + 0.34	Tisch = 0.92*FRM + 6.0	Tisch = 1.17*FRM - 2.7
	Coefficient of determination (R^2)	0.978	0.995	0.957
	Mean Tisch/FRM Ratio	0.91	1.04	1.08
PM_{10}	Tisch CV	4.3%	7.4%	3.5%
	Regression Equation (Tisch vs. FRM)	Tisch = 1.02*FRM + 2.5	Tisch = 1.02*FRM + 7.8	Tisch = 1.53*FRM - 10.6
	Coefficient of determination (R^2)	0.987	0.996	0.88
	Mean Tisch/FRM Ratio	1.09	1.16	1.29

At all three sites, the Tisch SPM-613D units tended to significantly over-estimate the $PM_{2.5}$ concentrations when compared to the collocated $PM_{2.5}$ samplers. For $PM_{2.5}$ measurements, the mean sampler to FRM ratio at Gary, Phoenix, and Riverside was calculated as 1.26, 1.70, and 1.65, respectively. This over-estimation was quite consistent as illustrated in Figure 6 which plots the performance of the Tisch's $PM_{2.5}$ concentrations versus those of the collocated $PM_{2.5}$ FRM samplers. As was the case for the R&P dichot, it is hypothesized that this over-estimation might be due, in part, to the inadvertent intrusion of coarse mode particles into the sampler's fine mode channel. This hypothesis is supported by the fact that larger overestimations occur at

Figure 6. Tisch SPM-613D versus FRM $PM_{2.5}$ concentrations in Phoenix, AZ.



sites with the lowest mean $PM_{2.5}/PM_{10}$ ratios. The fact that the Tisch sampler typically provides PM_{10} concentrations higher than the collocated PM_{10} FRM samplers, however, may indicate that other measurement uncertainties may be responsible for the observed $PM_{2.5}$ measurement bias.

The Tisch SPM-613D units provide more accurate measurements of ambient PMc concentrations than $PM_{2.5}$ concentrations. For PMc measurements, the mean sampler to FRM ratio at Gary, Phoenix, and Riverside was calculated as 0.91, 1.04, and 1.08, respectively. Consistency of this performance during the month-long sampling at each site is demonstrated by the high coefficient of determination (0.978, 0.995, and 0.957, respectively) obtained during sampler versus FRM regressions.

TSI Inc. Model 3321 APS

Few problems were experienced with the two TSI Model 3321 APS units during the course of the field tests. The exception occurred approximately halfway through the field sampling in Phoenix when the response of APS Unit 2 began to deviate substantially from that of Unit 1. During the units' subsequent return to the manufacturer for cleaning, a circuit board within Unit 2 was diagnosed as faulty and was replaced. Data from this unit during the second half of the Phoenix tests, therefore, were not used in comparing the performance of the APS units to that of the collocated FRM samplers. Overall data capture rate for the APS units during the three city study was 85%.

A summary of the performance of the APS units during this study is provided in Table 6. With the exception of results obtained in Gary during which a few day's results tended to skew precision calculations, the precision between the two APS units was generally good. This general level of agreement is illustrated in Figure 7.

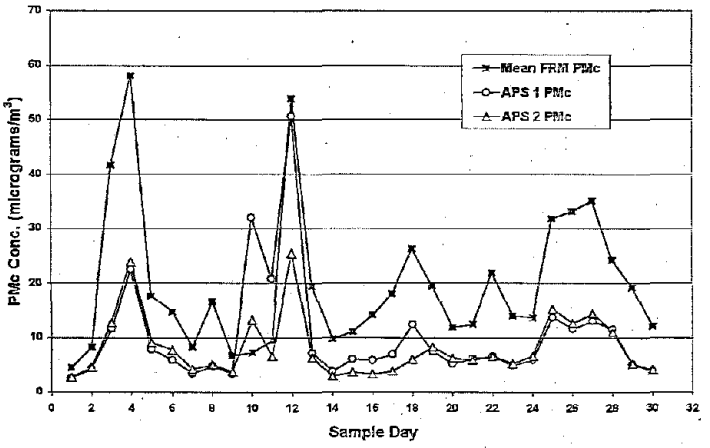
Table 6. Performance of the TSI APS versus the FRM.

Metric	Performance Criteria	Gary, IN	Phoenix, AZ	Riverside, CA
PMc	Sampler CV	16.8%	2.2%	8.5%
	Regression Equation (Sampler vs. FRM)	APS = 0.42*FRM + 0.48	APS = 0.56*FRM - 0.2	APS = 0.66*FRM - 2.3
	Coefficient of determination (R^2)	0.80	0.99	0.82
	Mean Sampler/FRM Ratio	0.42	0.55	0.58

The APS units tended to under-predict the PMc concentration when compared to measurements provided by the FRM samplers. Mean sampler to FRM PMc ratios for Gary, Phoenix, and Riverside were determined to be 0.42, 0.55, and 0.58, respectively. As a rule, the APS units tended to track the FRMs on a daily basis but under-measure PMc concentrations by about a factor of two. This behavior appeared to be relatively independent of sampling site or sampling day. Regressions of APS performance versus the collocated FRMs indicated that regression intercepts were close to zero at all three sampling sites. Although the exact reason for

the under-measurement is not known, the field behavior of these units is consistent with laboratory tests of the Model 3321 APS conducted by Peters, et al.[6].

Figure 7. APS versus FRM PMc concentrations in Gary, IN.



SUMMARY

1. Through coordination with state and local air monitoring agencies, the study sites selected met the study's siting objectives well and challenged the candidate samplers with a wide range of aerosol size distributions, aerosol concentrations, and meteorological conditions. Relatively few operational problems were experienced with the sampling equipment and the overall data capture rate for the study exceeded 95%. Prestudy, midstudy, and poststudy performance audits conducted at each sampling site revealed that the samplers typically held their calibrations well during the month-long field tests. The involvement and cooperation of the various sampler manufacturers was a key factor in the study's ability to successfully determine the inherent performance of the samplers.

2. The filter-based, integrated samplers involved in the study provided precise test results at all three sampling sites. For the FRM samplers, the mean inter-manufacturer coefficient of variation for $PM_{2.5}$, PMc, and PM_{10} was 2.7%, 4.5%, and 2.9%, respectively. Intra-manufacturer precision of the three R&P Model 2025 dichotomous samplers for $PM_{2.5}$, PMc, and PM_{10} measurements was 2.5%, 3.0%, and 2.0%, respectively. Effective shipping protocols resulted in negligible particle loss during transport of collected aerosol samples from each sampling site to the RTP weighing facility.

3. With the exception of Phoenix where coarse particles may have intruded into the samplers' fine channel, the R&P dichots typically provided $PM_{2.5}$ measurements which agreed closely with the collocated $PM_{2.5}$ FRM samplers. In regressions versus the collocated FRMs, all R&P dichot test results were highly correlated. The R&P dichots, however, underestimated PMc concentrations at all sampling sites and had a 21% under-measurement recorded at the Phoenix site. Mass balance calculations revealed that 16% of the aspirated PM_{10} mass in Phoenix is not accounted for during subsequent gravimetric measurement of fine and coarse channel filters. Recent tests have indicated that loss of coarse mode aerosols during the sampler's automated, post-sampling movement of the coarse particle cassette to the sample storage position may account for the observed bias.

4. With the exception of the problem noted during the Riverside tests, excellent inter-manufacturer precision of the R&P coarse TEOM samplers was observed at all three sampling sites and no operational problems were encountered with the samplers. However, with the exception of the Phoenix tests, the coarse TEOM tended to underestimate the PMc concentration by as much as 30%. The high correlation between the coarse TEOMs' response versus the collocated FRMs indicated that this performance was very consistent from one sampling event to another.

5. The Tisch SPM-613D samplers provided precise, highly correlated test results at all three sites for $PM_{2.5}$, PMc, and PM_{10} measurements. Although performance varied by site, the Tisch units generally provided PMc measurements within 10% of that of the collocated FRM samplers. However, the SPM-613D units consistently provided $PM_{2.5}$ concentrations significantly higher than the collocated $PM_{2.5}$ FRM samplers. As an example, the mean overestimation in $PM_{2.5}$ concentrations at the Phoenix site was 70%. Similar to the behavior of the R&P dichot, intrusion of coarse particles into the Tisch unit's fine channel may account for the majority of this observed behavior.

6. With the exception of a single electronics failure, the two TSI Model 3321 units appeared to function well and provided acceptable levels of precision. Although the APS units were observed to track the PMc FRM concentrations well, they typically underestimated PMc mass concentrations by a factor of two at all sampling sites. This field behavior is consistent with previous laboratory tests of the Model 3321 conducted under controlled conditions.

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1. US EPA, RTP, NC
2. Research Triangle Institute, RTP, NC
3. Rupprecht & Pattashnick Co., Inc., Albany, NY
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16. Abstract Comprehensive field studies were conducted to evaluate the performance of sampling methods for measuring the coarse fraction of PM_{10} in ambient air. Five separate sampling approaches were evaluated at each of three sampling sites. As the primary basis of comparison, a discrete difference method was used which employs two designated FRM samplers, one to measure $PM_{2.5}$ and the other PM_{10}. The numerical difference of these reference method concentrations (PM_{10}-$PM_{2.5}$) represented an estimate of PMc. A second sampling approach involved a sequential dichotomous sampler, which provided both $PM_{2.5}$ and PMc measurements. In both of these filter-based, time-integrated measurement approaches, the collected aerosol mass was analyzed gravimetrically in the laboratory under controlled conditions. Three continuous coarse particle samplers that measure PMc directly with a time resolution of 1 hour or less were also evaluated. One such sampler was a commercially available system based on beta attenuation, the second was based on TEOM technology. Both of these measurement approaches used dichotomous virtual impactors for separating fine and coarse particles. The third real-time sampler evaluated was an aerodynamic particle sizer (APS) that measures the aerodynamic diameter of individual particles, calculates the mass of the particle based on an assumed particle density, then sums the mass within the size range of interest to estimate the PMc mass concentration.

Sampling sites and timing of the studies were selected to provide diverse challenges to the samplers with respect to aerosol concentration, aerosol particle size distribution, and aerosol composition. Results from performance evaluations of the candidate PMc samplers at Gary, IN, Phoenix, AZ, and Riverside, CA were presented.

17. KEY WORDS AND DOCUMENT ANALYSIS

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