2	Field Evaluation of Portable and Central Site PM
3	Samplers Emphasizing Additive and Differential Mass
4	Concentration Estimates
5	Fu-Lin Chen ^{a,*} , Robert Vanderpool ^a , Ronald Williams ^a , Fred Dimmick ^a ,
6	Brett D. Grover ^a , Russell Long ^a , and Robert Murdoch ^b
7	^a U.S. Environmental Protection Agency, Office of Research and Development, RTP, NC
8	27711, USA
9	^b RTI International, Research Triangle Park, North Carolina 27709
10	
11	Abstract
12	The US Environmental Protection Agency (EPA) published a National Ambient Air Quality
13	Standard (NAAQS) and the accompanying Federal Reference Method (FRM) for PM ₁₀ in 1987.
14	The EPA revised the particle standards and FRM in 1997 to include PM _{2.5} . In 2005, EPA
15	proposed revisions to this NAAQS to include $PM_{10\text{-}2.5}$ but only finalized revisions with a $PM_{2.5}$
16	FRM and the development of a national monitoring network in 2006. Presently, no EPA
17	designated reference or equivalent method sampler has the ability to directly measure the mass
18	concentrations of PM_{10} , $PM_{10-2.5}$, and $PM_{2.5}$ simultaneously. An additive approach has been used
19	for samplers like the dichotomous monitors to calculate PM_{10} mass concentrations from
20	independent measures of $PM_{10-2.5}$ and $PM_{2.5}$ (i.e. $PM_{10} = PM_{10-2.5} + PM_{2.5}$). A differential approach
21	has been used to calculate $PM_{10-2.5}$ from identical collocated PM_{10} and $PM_{2.5}$ samplers

22 (i.e. $PM_{10-2.5} = PM_{10} - PM_{2.5}$). Since these two approaches have been used widely for PM

measurements, it is informative to evaluate their precision and comparability.

24

26

27

28

30

31

32

33

34

35

23

25 EPA performed collocated tests of five different particle samplers in the Research Triangle Park

area of North Carolina to evaluate the comparability and to characterize the additive and

differential approaches used to determine particle mass concentrations. The intra-sampler

precision of MiniVol, Omni, and dichotomous samplers was less than 8.4%. The precision of

 PM_{10} measurements using the additive approach with dichotomous samplers was less than 3.5%.

The poorest precision of the various PM_{10-2.5} differential approaches was less than 15.1%. No

zero or negative PM_{10-2.5} concentrations were calculated using the differential approach. A

coefficient of determination of 0.81 or higher was obtained for all paired comparison of PM_{10-2.5}.

The reported test results show that concentrations calculated from both the additive and

differential approaches generally agree among the portable samplers, the more established

dichotomous sampler and the Federal Reference Methods.

36

37

Keywords: PM₁₀, PM_{10-2.5}, PM_{2.5}, additive and differential approaches

38

39

1. Introduction

- 40 The Clean Air Act requires EPA to regulate particulate matter (PM) concentrations nationwide.
- 41 PM is a complex mixture of small solid particles and liquid droplets. The size of particles has
- been directly linked to their potential for causing health problems (Lippmann et al., 2003). In
- 43 general, EPA is more focused upon inhalable particles, PM₁₀, that are 10 micrometers (μm) in

diameter or smaller because those particles can affect the heart and lungs and cause serious health effects if inhaled (EPA, 2006a; Zanobetti et al., 2009). In 2006, EPA proposed revisions to the NAAQS for ambient PM (EPA, 2006b). These revisions were made in response to the increasing evidence of adverse health effects associated with human exposure to PM. In recognition that these health effects vary as a function of particle size, the proposed revisions included separate standards for fine particles (PM_{2.5}) and coarse particles (PM_{10-2.5}), but the latter was not promulgated. PM_{2.5} is the particle size fraction $\leq 2.5 \,\mu m$ in diameter while the PM_{10-2.5} size fraction represents those particles between 2.5 and 10 µm in diameter. A number of studies have identified adverse health effects associated with PM₁₀ and PM_{2.5} (Schwartz et al., 1996; Anderson et al., 2005). Recent studies indicate that short-term exposure to ambient $PM_{10-2.5}$ is associated with increased mortality risk and hospital admissions, primarily due to cardiovascular and respiratory effects (Metzger et al., 2004; Houthuijs et al., 2001). Table 1 lists a chronology of EPA's particle pollution standards (http://www.epa.gov/pm/standards.html). Wilson et al. (1997) reported that the aerodynamic particle diameter for 50 percent penetration into the thoracic airways and lower respiratory tract is 10 µm. PM₁₀ is the diameter of particles smaller than 10 µm and divided into PM_{2.5} and PM_{10-2.5} fractions. The two size fractions have different predominant sources and formation mechanisms and consequently different chemical compositions (Brunekeef et al., 2005). PM_{2.5} and PM_{10-2.5} are viewed as separate classes of pollutants and have been measured separately in research and epidemiologic studies. The primary sources of PM_{2.5} are anthropogenic, such as combustion and products of gas to particle conversion. The primary formation mechanisms of PM_{10-2.5} are typically comprised of dust, sand, crushing,

44

45

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

64

grinding and non-exhaust vehicle emissions like tire rubber. PM_{10-2.5} can also consist of biological substances such as pollen and fungi (EPA, 2004; EPA, 2005).

Three means of determining PM size fractions of interest exist. These include the direct approach where the monitor's design allows for particles of a given size to be captured independent of others. An additive approach is used when one single monitor collect two independent size fractions and then mathematically added together to estimate new fraction. The differential approach involves independent collection of two size fractions which then mathematically subtracted to yield estimates of a third.

The EPA, state, and local monitoring agencies have maintained a national network to collect and measure PM₁₀ since the 1980's and PM_{2.5} since 2000 (EPA, 2000). Dichotomous samplers (dichot) have been used to directly measure the PM_{2.5} and PM_{10-2.5} concentrations since 1980. Using dichotomous data, the additive approach could also be used to estimate the PM₁₀ mass concentrations. The differential approach, therefore, could be used to estimate PM_{10-2.5} mass concentrations from collocated PM₁₀ and PM_{2.5} data. Several studies have reported on comparisons of PM_{10-2.5} estimates using direct and indirect measurements (Vanderpool et al., 2005; Allen et al., 1999). Findings from these reveal that the additive and differential approaches of mass estimation can produce comparable results to direct measurement for the PM₁₀ and PM_{10-2.5} size fractions.

Differential and additive approaches have been used in PM spatial variation studies. In general,

the FRM (PM_{2.5} or PM₁₀) and dichot (PM_{2.5} and PM_{10-2.5}) are used at central monitoring sites with more portable samplers (PM_{2.5} or PM₁₀) used at satellite sites. Comparing the mass concentrations collected from a central monitor and dispersed portable samplers allows one to assess PM spatial variation across an area. Knowledge of the spatial variation of PM is critical for characterizing potential exposures to particle pollution which can vary greatly from one area to another. PM_{2.5} mass is often seen as homogeneous across many urban areas although it varies temporally (Williams et al., 2003). PM_{10-2.5} exhibits more spatial variability and varies substantially across time because it settles by gravity relatively quickly and the sources are not uniformly distributed across an urban area (Ott et al., 2008). Several studies have indicated that PM_{10-2.5} measurements at a single central monitoring site do not provide an adequate measure of the average daily concentration of PM_{10-2.5} across the community compared to that for PM_{2.5} measurement (Moon et al., 2001; Wilson et al., 2005). Chen et al. (2007) and Williams et al. (2000) reported poor spatial agreement for PM_{10-2.5} mass concentrations between a central monitor (dichotomous sampler) and residential outdoor monitoring stations (portable samplers) in the same geographical area. Williams et al. (2008) indicates that the ability of community-based monitors to act as a surrogate for PM_{10-2.5} mass might be problematic. Many factors might have influenced the poor spatial agreement in PM_{10-2.5} measurements reported above. These factors include the disperse nature of PM_{10-2.5} sources, rapid particle settlement, and differences in sampler specifications associated with their deployment (i.e. particle separation technique, flow rate, and sampling height). Any single or combination of these effects could have a major effect on PM mass concentration determinations. There was a lack of information existing as to the comparability of various samplers often used to assess air quality, especially that focusing on the $PM_{10-2.5}$ size fraction.

88

89

90

91

92

93

94

95

96

97

98

99

100

101

102

103

104

105

106

107

108

Therefore, we undertook this study to evaluate and compare these two calculation approaches (additive and differential) for both research and FRM type monitors.

The purpose of this study was to compare two portable samplers (Omni and MiniVol) to the FRM and the dichot and then evaluate the additive and differential approaches for estimating ambient PM. PM₁₀ mass concentration calculated from the additive approach using the dichot and PM_{10-2.5} mass concentration calculated from the differential approach for the Omni, Minivol, and FRM samplers were compared with direct mass measurements. Precision and comparability of the various instruments to estimate PM₁₀, PM_{10-2.5}, and PM_{2.5} were determined and are reported.

2. Materials and Methods

121 2.1. Study design

To evaluate the performance of the PM samplers, collocated tests were conducted in Research Triangle Park (RTP), North Carolina in 2007. The RTP area was chosen because it served as a convenient (local) method testing site for protocols being developed for future Birmingham (Alabama) studies. The Birmingham studies were designed to determine the spatial and temporal distribution of coarse mode particles within an urban airshed with elevated PM_{10-2.5} levels. The RTP test was designed primarily to ensure functionality of equipment, development of standard operating procedures, and provide operator training for the Birmingham study. The RTP area does not typically experience the levels of PM_{10-2.5} expected for Birmingham and other areas with elevated PM_{10-2.5}. Thus, it may not have been an ideal location for the inter-comparison of these samplers and for evaluating the calculation approaches for estimating PM_{10-2.5} levels. Nevertheless,

it did provide an opportunity to gather relevant data for comparing these samplers and assessing the implications of the additive and differential approaches on PM estimates.

Figure 1 shows the sampler placement on the platform with all inlets at the same 5 m elevation above ground level and spaced one meter apart to minimize interference. The 3.3 by 3.3 m auxiliary sampling platform adjacent to the 8 m long motor laboratory has been used for previous EPA studies and provided a safe and secure deployment of the instrumentation (Vanderpool et al., 2005). Three pairs of portable MiniVol (Airmetric, Eugene, Oregon) and Omni (BGI Inc. Waltham, MA) samplers were installed on the monitoring platform. Two Rupprecht and Patashnick dichotomous air samplers (R&P Model 2025 Sampler, Albany, NY), a pair of Graseby-Andersen FRM (Andersen FRM, RAAS-100, Smyrna, GA) and a pair of Rupprecht and Patashnick FRM (R&P FRM, Partisol-FRM 2000, Albany, NY) samplers were also installed on the platform. Each pair of samplers included a PM₁₀ and a PM_{2.5} sampler.

Table 2 lists the specifications of each sampler. Prior to each field test, all samplers were cleaned and passed a leak test. Each sampler was calibrated for volumetric flow rate using a calibrated BGI TriCal. All of the samplers in this study were filter-based employing a 2 µm pore size and 46.2 mm Diameters PTFE filter (Polytetrafluoroethylene, 7592-304, Whatman inc., Florham, NJ). Field blank tests of the samplers were conducted in every sampling event for performance audits. Field blank values were not used to correct the measured PM concentrations but served as means to detect any dramatic filter mass change that would have indicated improper filter handling and transport. No such events occurred. Blank correction is also not permitted per data treatment

procedures outlined in the Federal Reference Method for PM_{2.5} (Winter et al., 2004).

155

156

157

158

159

160

161

162

163

164

165

166

167

168

169

170

171

172

173

174

154

The study consisted of twenty, 45 hour sampling periods. A 45-hour integrated sampling period was used for convenience and to ensure adequate filter mass loading to examine the study hypothesis. Earlier US EPA research (Vanderpool et al., 2006) focusing on determining candidate methods for PM measurement, had identified a range of PM mass concentrations across various U.S. geographical areas. Table 3 now reports summary mass concentration finding from the Vanderpool et al. (2006) effort in comparison to that associated with the current (RTP) data collections. This provides information as to the variability of PM size fraction concentrations observed in North America as it relates to the RTP site. In general, mean PM_{2.5} RTP mass concentrations were very similar those from Gary, Phoenix and Riverside. However, PM_{10-2.5} and PM₁₀ mass concentrations were significantly lower in the RTP area in comparison to those aforementioned cities. The RTP area had a sufficiently low PM_{10-2.5} and PM₁₀ average daily mass concentration that selection of a 24-hour sampling protocol in establishing the current study design might have significantly impacted our ability to fully test the two approaches. Therefore, the selection of an extended 45-hour sampling period was ultimately ensured sufficient data quality to test the study hypothesis. Deviation from the standard 24-hour FRM protocol was not expected to negatively impact sampler operation or the resulting data quality of the size fractionation relative to overall sampler performance. The use of an extended 45-hour sampling period in the current study was viewed as crucial to meet critical data demands (gravimetric analysis) in evaluating the two mass determination approaches, especially those associated with low flow samplers.

176 2.2. Sampler Description

177 2.2.1 Federal Reference Method (FRM)

The FRM is a filter-based, 24 hour sampling period technique with pre- and post- conditioning and weighing of the filter under standardized laboratory conditions. The Andersen FRM and R&P FRM are stand-alone sampling systems meeting national ambient air quality regulations for single day sampling of $PM_{2.5}$ or PM_{10} . The $PM_{2.5}$ FRM samples ambient air through a PM_{10} inlet with a 50% cutpoint of 10 μ m. The air stream is then directed via the downtubes to the Well Impactor Ninety-Six (WINS) stage with a 50% cutpoint of 2.5 μ m (Peters et al., 2001a). For the PM_{10} FRM, the WINS impactor is replaced with an equivalent length of tubing with no size selective characteristics. The nominal flow rate through these FRM systems is 16.67 liter per minute (lpm).

2.2.2 Dichotomous Sampler

PM₁₀ aerosol. Ambient aerosol is sampled through a PM₁₀ size selective inlet and subsequently divided by a virtual impactor into separate PM_{10-2.5} and PM_{2.5} fractions. These samplers operate at a flow rate of 16.67 lpm with the fine particle flow rate at 15 lpm and coarse particle flow rate at 1.67 lpm. Particles in the two resulting flow streams are collected on separate filters for subsequent gravimetric analysis. However, commercially available dichots are inconvenient or impossible to use in remote areas because of their size and power requirements.

Dichots are designed to provide direct measurements of both PM_{2.5} and PM_{10-2.5} fractions of a

196 2.2.3 Portable Sampler – Omni and MiniVol Sampler

In 1980, the EPA encouraged state and local air monitoring groups to conduct short-term multi-site

pollutant monitoring studies using relatively inexpensive and small portable samplers (Jones et al., 1998). The concept was to "saturate" an area with easily deployable, inexpensive filter-based samplers using battery operation and low flow rates to assess air quality in areas with high concentrations of pollutants or at reclamation sites. Saturation monitoring has also been conducted to characterize the spatial distribution of pollutant concentration or to evaluate source contributions in support of receptor modeling. The Omni and MiniVol samplers are saturation samplers which have been widely used in remote areas lacking AC power. The AC power option was used for all samplers in this study to minimize potential data loss.

The BGI Omni samplers are lightweight, weatherproof and portable saturation samplers which can operate for up to 48 hours on battery power. A constant volumetric flow rate of 5 lpm was maintained at the system's inlet using calibrated temperature, pressure, and flow sensors. A PM_{10} inlet was used to collect ambient aerosol less than 10 μ m on the PTFE filter. A BGI very sharp cut cyclone with a cutpoint of 2.5 μ m was installed inline following the PM_{10} inlet for collection of $PM_{2.5}$ mass on the PTFE sampling filter.

The Airmetric MiniVol sampler is a compact, lightweight, battery-operated monitor, constructed from durable PVC and self-shrouded from weather effects. The MiniVol sampler consists of a size-selective inlet, a flow control unit, a programmable timer, and a rechargeable battery. The PM₁₀ inlet was equipped with a PM₁₀ impactor. The PM_{2.5} inlet was equipped with a PM₁₀ scalping impactor followed by an in-line PM_{2.5} impactor for final size range designation. The standard electrical system for these units was modified to be AC capable for this study. The flow rate of the

sampler was 5 lpm. Glisseal high vacuum grease (Lot 130475, borer Chemie, CH-4528 Zuchwil) was used to grease impactor surface on all inlets to eliminate particle bouncing (Baldauf et al., 2001; Tropp et al., 1998; Hill et al., 1999).

223

224

225

226

227

228

229

230

231

232

233

234

235

236

237

238

220

221

222

2.3. Quality Control

MiniVol samplers were examined each sampling day to confirm proper operation. Daily quality control reviews before and after sampling included checks on flow rate, volume of air sampled, elapsed time, air leaks, and unusual filter conditions. The Omni, FRM, and dichot samplers required little maintenance. They performed automated calibrations for volumetric flow rate involving ambient temperature, filter temperature, and ambient pressure measurements. All samplers were audited for nominal flow rates prior to and immediately after sampling using a BGI TriCal Volumetric Air Flow Calibrator. Samplers were expected to maintain nominal flow rates ± 5% of initial settings to be considered valid. All filter-based sampling was performed using a single lot of filters respective to size and media type. The MiniVol sampler's impactor was cleaned and recoated after every two sampling periods (90 hours). The impaction well of the FRM WINS was replaced after every two sampling periods. Although the impactors was recoated and the WINS replaced after every 90 hours of sampling, they had minimal observable build-up upon inspection which would have potentially increased particle rebound effects and resulting decreases in sampler precision. Consequently, the selection of the extended 45-hour sampling period had no obvious negative impact on sampler performance.

240

241

239

Gravimetric mass analysis of filters prior to and following exposure was conducted in an

environmentally controlled balance room. This balance room was an enclosed chamber where temperature and relative humidity (RH) were made constant. The normal set-points were $21.5 \pm 0.5^{\circ}$ C and $35 \pm 5\%$ RH, in conformity with current Federal requirements for PM filter mass determination. The filters were placed in the balance chamber for a minimum of 24 hour undisturbed to allow for filter equilibration. Gravimetric analysis was then performed at least twice for each filter. In order to verify the gravimetric analysis, 100% replicate weighing (with a $\pm 5~\mu g$ reweigh threshold) was used for each filter during all pre- and post- weighing operations. The average mass was then used to calculate concentrations.

3. Results and Discussion

252 3.1. Precision

The precision of each sampler was based upon comparisons of collocated duplicate samplers. Precision of dichot sampler was defined as the (A-B)*100/(A+B) when N=2. The "A" and "B" represent the observed mass concentration of the sampler replicates. Precision of the MiniVol and Omni were defined as the average of abs((A-m)/m) and abs((B-m)/m) when N=3 where m is the median value. Table 4 shows the average precision and general statistics calculated from the equations above associated with 20 sampling events for the dichot, MiniVol, and Omni samplers. The results indicate that the precision error of direct measurements of PM mass in three size fractions was less than 8.4% from all samplers. The precision of dichot PM₁₀ measurements using the additive approach was less than 3.5%. The precision of PM_{10-2.5} measurements determined by the differential approach was less than 15.1%.

The precision of the FRMs cannot be reported because only one PM_{2.5} and one PM₁₀ FRM were operated in this study. But in previous studies, Peters et al. (2001B) reported precision error of PM_{2.5} FRMs to be 2 to 6%. Wilson et al. (2002) indicated that PM_{2.5} and PM₁₀ FRMs provide relatively precise (±10%) methods for determining the mass of material remaining on a Teflon filter after equilibration. Chow et al. (2008) indicated that well prepared collocated samplers could have precision of approximately 10% for a variety of PM_{2.5} FRM samplers.

Wilson et al. (2002) indicated that the PM_{10-2.5} precision error ranged from a few percent to as much as 40%. Allen et al. (1999) measured PM_{10-2.5} precision of 15% for the Harvard Impactor using the difference between independent PM₁₀ and PM_{2.5} monitors. They also indicated that the precision for PM_{10-2.5} measurements may be three or four times worse than the associated PM_{2.5} or PM₁₀ data, depending on the measurement approach, PM_{2.5} to PM₁₀ ratio, and PM_{10-2.5} concentrations.

- 278 3.2. Mass Concentration Comparison in Three Size Fractions
- Table 5 shows the average mass concentrations (AMC) measured from 20 sampling measurements.
- Data reporting mass concentrations associated with each of the various monitors are reported. The
- use of the additive or differential approach is indicated as appropriate. Discussion about unique
- 282 findings for the various combinations are reported below.

- 284 3.2.1 PM_{10} (all direct except dichot being additive)
- 285 The comparison of AMC determined by indirect measurement to direct measurement was very

similar. The AMC of two portables sampler was the same as the Andersen FRM but about 4% less than the R&P FRM. The AMC of the dichot calculated using the additive approach was 1% higher than the AMC of the two FRMs. The PM_{10} dichot concentration calculated from the additive approach was 3% higher than the concentrations measured directly from the MiniVol and Omni samplers.

- 3.2.2 PM_{2.5} (all direct measurement)
- The AMC of the MiniVol and Omni portable samplers were 9% higher than the AMC of the dichot and FRMs samplers. The AMC of the dichot was the same as the Andersen FRM but 2% higher than the R&P FRM. The AMC of the MiniVol was 4% higher than the Omni samplers.

- 297 3.2.3 PM_{10-2.5} (all but dichot being differential)
 - The results show significantly more variability in the resulting PM_{10-2.5} mass concentrations as compared to the PM₁₀ or PM_{2.5} estimates. The poorest PM_{10-2.5} AMC agreement observed was that from the two portable samplers which represented only 72% of that provided by the dichot. The AMC collected from the two FRMs deviated by approximately 10% in comparison to the dichot. Lower flow rate samplers (5 lpm) had an AMC for this size fraction that was less than the higher flow rate samplers (16.67 lpm) regardless of the approach involved. Even so, samplers operating at the same flow rate were sometimes observed to yield AMC differences greater than 10%. In general, the direct measurement approach routinely provided higher AMCs as compared to those produced by the differential approach.

Even for the direct measurement of PM_{10-2.5} from the dichot sampler, there still exists some uncertainty because of its complicated analysis of the PM_{10-2.5} concentration. Allen et al. (1999) reported that the dichots have substantial PM_{10-2.5} error due to a decrease in PM_{10-2.5} measurement precision when PM_{2.5} to PM_{10-2.5} ratios are high, changes in sampler characteristics as particles deposit on inside surfaces, or the loss of coarse particles from filters during transport. Some particle losses within the dichot have been reported (Marple et al., 1990).

The Coefficient of Determination (R²) of three PM size fractions had been reported in Tables 6 – 8. R² is used to indicate the strength of a relationship between two inter-samplers. R² values of at least 0.95 for all the PM₁₀ and PM_{2.5} comparisons were obtained. R² of PM_{10-2.5} between and among samplers having the same flow rate were also investigated. Between the two portable (i.e. MiniVol and Omni) samplers, the R² value was 0.87. For the more stationary samplers, the R² value of PM_{10-2.5} between dichot vs. Andersen FRM, dichot vs. R&P FRM, and Andersen FRM vs. R&P FRM were 0.82, 0.87 and 0.86, respectively. The R² values between portable and stationary samplers ranged from 0.81 to 0.92. An R² of 0.92 was obtained for the best paired comparison of PM_{10-2.5} in this study (R&P FRM vs MiniVol or Omni).

4. Conclusion

This study provided the means to directly test the comparability of vastly different approaches (direct, additive, differential) in estimating various PM size fractions mass concentrations. Such testing was needed to determine the comparability of data being produced in studies carried out throughout the world involving the different approaches and often involving highly different

sampling instrumentation. All of the various approaches or instrumentation available to researchers have their own specific advantages and disadvantages that might influence the scientific community-at-large in decisions to deploy them in future research studies. The findings presented here represent a first attempt in defining some of the issues that need to be considered when researchers select any combination of these various mass estimation instrumentation approaches.

Reproducible (< 15.1% precision) mass concentrations for the PM_{10-2.5} size fraction would appear to be capable of being generated in environmental monitoring studies regardless of the selection of any approach or instrumentation type evaluated as part of this study. Even so, precision error of this particular size fraction was often two to five times poorer than that established for the other two size fractions even when sampling periods double that of the normal daily monitoring periods were evaluated. Shorter time periods (e.g., 24-hour) would be suspected of having even higher precision errors due to lower mass loadings negatively impacting the resulting gravimetric analyses. It is suspected that numerous factors associated with the individual mass estimation and instrumentation type selections evaluated here resulted in the higher cumulative PM_{10-2.5} precision errors.

Comparability of mass concentrations within the $PM_{2.5}$ or PM_{10} size fractions were extremely well correlated ($R^2 > 0.95$) regardless of the approach or instrumentation selected. Conversely, the $PM_{10-2.5}$ size fraction exhibited slightly poorer mass concentration agreement ($R^2 > 0.81$) under various combinations of test conditions. Both the additive and differential approaches would appear to yield highly similar mass estimates for the $PM_{10-2.5}$ size fraction regardless of

instrumentation type. Even so, low flow rate monitors (MiniVol and Omni) would however, appear to yield mass concentration estimates slightly lower for this size fraction in comparison to those like the higher flow rate FRMs. Therefore, the state-of-the science at this point would indicate researchers can be fairly confident that mass estimates they obtain regardless of technique selection are generally representative of the environment being monitored. However, enough variability was exhibited between the various test combinations that would indicate common instrument deployment (same monitor type and selection of mass estimation procedures) across all spatial settings would be advantageous in obtaining PM_{10-2.5} mass estimates of the highest degree of uniformity.

Acknowledgements

The U.S. Environmental Protection Agency through its Office of Research and Development funded and managed the research described here. It has been subjected to Agency review and approved for publication. The authors thank field support staff of the RTI International for their assistance in the collection of data presented here under Alion Science and Technology contract EP-05-D-065.

Figure 1. Samplers set up on the platform at field site (RTP, NC, 2007)



Table 1. Particle pollution standards, (http://www.epa.gov/pm/standards.html)

Date	National Ambient Air Quality Standards	24 h average (μg/m³)	Annual (μg/m³)
1971	Total Suspended Particles (TSP)	260	75
1987	PM ₁₀ , particles size smaller than 10 micrometers (replaced TSP)	150	50
1997	Added PM _{2.5} , particles size smaller than 2.5 micrometers, standards	35	15
2006	Proposed PM _{10-2.5} standards	70	N/A

Table 2. Specifications of tested Samplers

Sampler	Fraction C	ollected	Additive or Differential approach	Flowrate, lpm
R&P Dichot	PM _{10-2.5} ,	PM _{2.5}	$PM_{10}=PM_{10-2.5}+PM_{2.5}$	16.67
Anderson FRM	PM ₁₀ ,	PM _{2.5}	$PM_{10-2.5} = PM_{10}-PM_{2.5}$	16.67
R&P FRM	PM ₁₀ ,	PM _{2.5}	$PM_{10-2.5} = PM_{10}-PM_{2.5}$	16.67
Airmetric MiniVol	PM ₁₀ ,	PM _{2.5}	$PM_{10-2.5} = PM_{10}-PM_{2.5}$	5.0
BGI Omni	PM ₁₀ ,	PM _{2.5}	$PM_{10-2.5} = PM_{10}-PM_{2.5}$	5.0

Table 3. Mass concentration range and mean ($\mu g/m^3$) from the Vanderpool et al. (2006) locations and this study.

	PM _{2.5}	PM _{10-2.5}	PM ₁₀
Gary, IN. 2003	10.3 – 46.9	4.5 – 58.1	22.6 - 85.0
March - April	Mean = 22.8	Mean = 19.9	Mean = 42.6
Phoenix, AZ. 2003	6.4 – 22.0	26.5 – 209.0	37.1 – 230.9
May - June	Mean = 11.0	Mean = 55.7	Mean = 66.7
Riverside, CA. 2003	9.9 – 32.7	16.2 – 46.1	27 – 69.3
July - August	Mean = 17.7	Mean = 30.4	Mean = 48.0
Phoenix, AZ. 2004	6 – 22.4	7.7 – 95.1	14.8 – 177.5
January - February	Mean = 13.2	Mean = 39.5	Mean = 52.8
Phoenix, AZ. 2005	4.9 – 16.6	23.4 – 122.8	30.1 – 134.6
April - May	Mean = 12.8	Mean = 46.2	Mean = 56.0
RTP, NC. 2007	5.2 – 22.4	3.1 – 14.6	10.7 – 30.8
April - May	Mean = 12.8	Mean = 8.0	Mean = 20.8

Table 4. General statistics of the Intra-Sampler Precision (%) of MiniVol, Omni and Dichot from 20 sampling events.

Size Fraction		MiniVol, N=3	Omni, N=3	Dichot, N=2
PM ₁₀	Mean	2.3	6.2	3.5*
PM ₁₀	Max.	8.6	18.1	12.6
PM ₁₀	Min.	0.6	1.1	0.3
PM _{2.5}	Mean	4.6	5.7	3.6
PM _{2.5}	Max.	11.5	12.9	20.0
PM _{2.5}	Min.	0.5	1.7	0.0
PM _{10-2.5}	Mean	13.2**	15.1**	8.4
PM _{10-2.5}	Max.	42.1	28.0	21.0
PM _{10-2.5}	Min.	1.3	5.3	· 1.2

^{*} additive approach

^{**} differential approach

Table 5. General mass concentration (µg/m³) statistics (N=20) of the collocated samplers.

		Dichot	Andersen FRM	R&P FRM	MiniVol	Omni
PM ₁₀ ,	Mean	21.2 ± 5.5*	20.5 ± 5.8	21.4 ± 5.9	20.5 ± 6.1	20.6 ± 5.5
PM ₁₀ ,	Max.	30.8*	29.3	30.8	30.7	32.2
PM ₁₀ ,	Min.	12.4*	10.7	11.7	12.2	11.9
PM _{2.5} ,	Mean	12.5 ± 4.2	12.5 ± 4.5	12.2 ± 4.4	13.8 ± 4.7	13.2 ± 4.2
PM _{2.5} ,	Max.	20.2	20.5	19.9	22.4	21.6
PM _{2.5,}	Min.	5.5	5.2	5.2	7.1	6.3
PM _{10-2.5} ,	Mean	8.7 ± 3.2	7.9 ± 3.1**	9.2 ± 3.4**	6.6 ± 2.7**	7.4 ± 2.7**
PM _{10-2.5} ,	Max.	14.0**	14.3**	14.6**	11.6**	14.3**
PM _{10-2.5} ,	Min.	4.9**	3.3**	5.4**	3.1**	4.2**

^{*} additive approach

Table 6. Coefficient of Determination, R^2 , for PM_{10} Fraction Comparison All P-values < 0.0001

	Omni	Dichot (Additive)	Andersen FRM	R&P FRM
MiniVol	0.97	0.97	0.98	0.97
Omni		0.95	0.98	0.98
Dichot (Additive)			0.96	0.95
Andersen FRM				0.96

^{**} differential approach

Table 7. Coefficient of Determinations, R^2 , for $PM_{2.5}$ Fraction Comparisons All P-values < 0.0001

	Omni	Dichot	Andersen FRM	R&P FRM
MiniVol	0.97	0.98	0.98	0.99
Omni		0.96	0.96	0.98
Dichot			0.97	0.98
Andersen FRM				0.98

Table 8. Coefficient of Determinations, R^2 , for $PM_{10-2.5}$ Fraction Comparisons

All P-values < 0.0001

	Omni	Dichot	Andersen FRM	R&P FRM
MiniVol	0.87	0.87	0.81	0.92
Omni		0.83	0.86	0.92
Dichot	1	*	0.82	0.87
Andersen FRM				0.86

References

Allen, G., Oh, J., Koutrakis, P., Sioutas, C., 1999. Techniques for high-quality coarse particle mass measurements, Journal of the Air & Waste Management Association 49, 133-141.

Anderson, R., Atkinson, W., Peacock, L., Sweeting, J., Marston, M., 2005. Ambient particulate matter and health effects - publications bias in studies of short-term associations, Epidemiology 16, 155-163.

Baldauf, R., Lane, D., Marotz, G., Wiener, R., 2001. Performance evaluation of the portable MiniVol particulate matter sampler, Atmospheric Environment 35, 6087-6091.

Brunekeef, B., Forsberg, B., 2005. Epidemiological evidence of effects of coarse airborne particles on health, Eur Respir. J. 26, 309-318.

Chen, F., Williams, R., Svendsen, E., Yeatts, K., Creason, K., Scott, J., Terrell, D., Case, M., 2007. Coarse particulate matter concentrations from residential outdoor sites associated with the North Carolina Asthma and Children's Environment Studies (NC-ACES), Atmospheric Environment 41, 1200-1208.

Chow, J., Doraiswamy, P., Watson, J., Chen, L., Ho, S., Sodeman, D., 2008. Advances in integrated and continuous measurements for particle mass and chemical composition, Journal of the Air & Waste Management Association 58, 141-163.

EPA, 2000, Aeromatric Information Retrieval System (AIRS), Research Triangle Park, NC: Office of Air Quality Planning and Standards. (http://www.epa.gov/tta/airs/airsaqs)

EPA, 2004. Air Quality for Particulate Matter. Research Triangle Park, NC: Office of Research and Development; report # EPA/600/P-99/002a,bF.

EPA, 2005. Review of the National Ambient Air Quality Standards for Particulate Matter: Policy Assessment of Scientific and Technical Information, Research Triangle Park, NC: Office of Air Quality Planning and Standards; Report # EPA-452/R-05-005a.

EPA, 2006a, National Ambient Air Quality Standards for Particulate Matter; Final Rule, 40 CFR Part 50, October 17 (http://www.epa.gov/ttn/naqps/standards/pm/data/ft20061017.pdf)

EPA, 2006b, Federal register: National Ambient Air Quality Standards for Particulate Matter.

Revision to Ambient Air Monitoring Regulations; Proposed Rule, 40 CFR Part 50, 53, and 58,

January 17 (http://www.epa.gov/air/particles/fs20051-20pm.html)

Hill, J., Patel, P., Turner, J., 1999. Performance characterization of the MiniVol PM_{2.5} sampler, Air & Waste Management Association's 92nd meeting, June 20-24. (Air Quality Laboratory, Report # WUAQL-0399-01).

Houthuijs, D., Breugelmans, O., Hoek, G., Vaskovi, E., 2001. PM₁₀ and PM_{2.5} concentrations in

Central and Eastern Europe: results from the Cesar study, Atmospheric Environment 35, 2757-2771.

Jones, K., Berg, N., Sleva, S., Murchie, S., 1998. EPA saturation monitor repository 98-TA31A.04, Presentation at the Air & Waste Management Association's 91st Annual Meeting.

Lippmann, M., Ito, K., Nadas, A., Burnett, R., 2003. Association of particulate matter components with daily mortality and morbidity in urban populations. Res Rep Health Eff Inst 2003; 5-82.

Marple, V., Liu, B., Burton, R., 1990. High-Volume Impactor for sampling fine and coarse particles, Journal of the Air & Waste Management Association 40, 762-767.

Metzger, B., Tolbert, E., Klein, M., 2004. Ambient air pollution and cardiovascular emergency department visits. Epidemiology 15, 46-56.

Moon, C., 2001. Exposure assessment of air pollutants: a review on spatial heterogeneity and indoor/outdoor/personal exposure to suspended particulate matter, nitrogen dioxide and ozone, Atmospheric Environment 35, 1-32.

Ott, D., Kumar, N., Peters, T., 2008. Passive sampling to capture spatial variability in PM_{10-2.5}, Atmospheric Environment 42, 746-756.

Peters, T., Vanderpool, R., Wiener, R., 2001a. Design and calibration of the EPA $PM_{2.5}$ Well

Impactor Ninety-Six (WINS), Aerosol Science and Technology 34, 389-397.

Peters, T., Norris, G., Vanderpool, R., Gemmill, D., Wiener, R., Murdoch, R., McElroy, F., Pitchford, M., 2001b. Field performance of PM_{2.5} Federal Reference Method samplers. Aerosol Science and Technology 34, 433-447.

Schwartz, J., Dockery, W., Neas, M., 1996. Is daily mortality associated specifically with fine particles? Journal of the Air Waste Management Association 46, 927-939.

Tropp, R., Jones, K., Kuhn, G., Berg, N., 1998. Comparison of PM_{2.5} saturation samplers with prototype PM_{2.5} Federal Reference Method samplers, Air & Waste Management Association's 91st Annual meeting, June 14-18.

(http://www.airmetrics.com/products/studies/1998eparepository.html).

Vanderpool, R., Hanley, T., Dimmick, F., Hunkie, E., Solomom, P., McElroy, F., Murdoch, R., Natarajan, S., 2005. Multi-Site evaluations of candidate methodologies for determining coarse particulate matter (PM_{10-2.5}) concentrations: Updated report regarding second-concentration and new PM_{10-2.5} samplers, EPA/600/R-06/026, 2006.

Vanderpool, R., Hanley, T., Dimmick, F., Hunkie, E., Solomom, P., McElroy, F., Murdoch, R., Natarajan, S., 2006. Multi-Site evaluations of candidate methodologies for determining coarse particulate matter (PM_{10-2.5}) concentrations: August 2005 updated report regarding second-concentration and new PM_{10-2.5} samplers – attachment 2. EPA/PB#2006-115765.

Williams, R., Suggs, J., Zweidinger, R., Evans, G., Creason, J., Kwok, R., Rodes, C., Lawless, P., Sheldon, L., 2000. The 1998 Baltimore particulate matter epidemiology – exposure study: Part 1. Comparison of ambient, resident outdoor, indoor and apartment particulate matter monitoring, Journal of Exposure Analysis and Environmental Epidemiology 10, 518-532.

Williams, R., Suggs, J., Rea, A., Leovic, K., Vette, A., Croghan, C., Sheldon, L., Rodes, C., Thornburg, J., Ejire, A., Herbst, M., Sanders, W., 2003. The Research Triangle Park particulate matter panel study: PM mass concentrations relationships, Atmospheric Environment 37, 5349-5363.

Williams, R., Case, M., Yeatts, K., Chen, F., Scott, J., Svendsen, E., Devlin, R., 2008. Personal coarse particulate matter exposures in an adult cohort, Atmospheric Environment 42, 6743-6748.

Wilson, J., Kingham, S., Pearce, J., Sturman, A., 2005. A review of intraurban variations in particulate air pollution: Implications for epidemiological research, Atmospheric Environment 39, 6444-6462.

Wilson, W., Suh, H., 1997. Fine particles and coarse particles: concentration relationships relevant to epidemiologic studies, Journal of the Air Waste Management Association 47, 1238-1249.

Wilson, W., Chow, J., Claiborn, C., Fusheng, W., Engeibrecht, J., Watson, J., 2002. Monitoring of particulate matter outdoors. Chemosphere 49, 1009-1043.

Winter, S., Bilonick, R., Withum, J., Conrad, V., Statnick, R., Ebelt, S., Suh, H., Koutrakis, P.,

2004. A field evaluation to compare the performance of personal exposure multi-pollutant samplers vs. Federal Method monitors at a central ambient air monitoring station in Steubenville, Ohio, Air Quality IV, September 22-23, 2004, Arlington, VA.

Zanobetti, A., Schwarz J., 2009. The effect of fine and coarse particulate air pollution on mortality: A national analysis. Environmental Health Perspective 117, 898-903.