2004 BELTWIDE COTTON CONFERENCES, SAN ANTONIO, TX – JANUARY 5-9

APPLICATION OF THE NATIONAL AMBIENT AIR QUALITY STANDARDS (NAAQS) IN URBAN VERSUS RURAL ENVIRONMENTS

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Abstract

The US Environmental Protection Agency is in the process of assessing the need for a National Ambient Air Quality Standard for the coarse fraction of particulate material (PM_{CF}). Specifically, the PM indicator being considered is particulate matter between 2.5 and 10 µm in aerodynamic equivalent diameter (AED). EPA is primarily relying on the available epidemiological studies that examine the possible health effects of PM_{CF} to reach a decision about developing a coarse particulate matter standard. These epidemiological studies utilize data from size selective PM samplers to estimate the study population's exposure to each PM indicator as defined by EPA (PM_{10}, $PM_{2.5}$, and PM_{CF}). Epidemiological studies typically focus on urban populations in order to obtain sufficient sample size and to increase statistical certainty associated with study findings. Because of this focus on the urban environment, there has been a lack of studies that evaluate the effect of coarse particulate matter in rural environments on human health.

There are a number of key differences between the urban and rural environments in the United States that can lead to critical mistakes in applying data from urban studies to the rural environment. These include differences in particle sources, which affect particle size distribution and composition, differences in the concentration of gaseous co-pollutants, and differences in PM sampler performance in the two environments. It is our contention that these differences between the urban and rural environment are significant and that the epidemiological studies cited by EPA rely on data that are not representative of the rural environment. These factors raise serious concerns that the implementation of a PM_{CF} standard in the rural environment will impose an unfair and unwarranted regulatory burden on the businesses and citizens in the rural areas.

Introduction

Measurement Issues in Determination of PM Fractions

Measurement of particulate matter suspended in the atmosphere is a particularly challenging problem. Several excellent reviews (Chow 1995; McMurry 2000; Wilson *et al.* 2002) have discussed in detail many of the issues related to ambient sampling of the various PM fractions. For this report, the issues that are most directly affected by the differences between rural and urban environments will be highlighted. In the following pages, the characteristics of the samplers used in the EPA referenced studies will be briefly discussed followed by a description of some of the sources of error in obtaining measurements of PM in ambient environments.

All size-selective particulate matter samplers rely on a pre-separator inlet to allow particles of the desired size to be captured on a filter and to prevent non-desired particles from reaching the filter. A sampler's pre-separator collection efficiency curve is most commonly represented by a lognormal distribution, characterized by a d_{50} (also referred to as cut-point) and a slope, which indicates how close to ideal the sampler performs. An ideal sampler would have a slope of 1.0. The cut-point is the particle size where 50% of the PM is captured by the pre-separator and 50% of the PM penetrates to the filter (Hinds 1999). The slope is the ratio of the particle sizes corresponding to cumulative collection efficiencies of 84.1% and 50% ($d_{84.1}/d_{50}$), 50% and 15.9% ($d_{50}/d_{15.9}$), or the square root of 84.1% and 15.9% ($\sqrt{d_{84.1}/_{15.9}}$) (Hinds 1999). Collection efficiency curves are usually assumed as constant and independent of particle size; in other words, it is assumed that a significant loading of large particles does not affect the pre-separators collection efficiency for smaller particles. This assumption has been shown to be in error for certain samplers under heavy loading (Ono *et al.* 2000). Concentration data used to generate a sampler's pre-separator collection efficiency curve are typically determined by conducting an array of tests over several mono-disperse particle sizes using known ambient concentrations. An example of the ideal $PM_{2.5}$ sampler efficiency curve is shown in Figure 1.

The most current list of EPA Designated Reference and Equivalent Methods (DREM) (EPA 2003) lists those samplers that have been approved as conforming to the Federal Reference Method (FRM) and are designated as reference samplers and those samplers that perform equivalently to the FRM but rely on different operating principles and are designated as equivalent samplers. The reference method for sampling $PM_{2.5}$ is specified in Title 40, Part 50 Appendix L of the [U.S.] Code of Federal Regulations (40 CFR Part 50) and the reference method for PM_{10} is specified in 40 CFR Part 50 Appendix M. Samplers are approved for addition to the DREM in accordance with Title 40, Part 53 of the Code of Federal Regulations (40 CFR Part 53). The performance criteria specified by EPA for PM_{10} and $PM_{2.5}$ samplers are summarized in Table 1.

$PM_{2.5}$ samplers can be approved in one of three classes; a *Class I equivalent method* means a method for PM2.5 based on a sampler that is very similar to that specified as the FRM in 40 CFR Part 53, a *Class II equivalent method* means a method for PM2.5 in which an integrated PM2.5 sample is obtained from the atmosphere by filtration and is subjected to a subsequent filter conditioning process followed by a gravimetric mass determination, but which is not a Class I equivalent method because of substantial deviations from the FRM design specifications, and a *Class III equivalent method* means a method for PM2.5 that has been determined by EPA not to be a Class I or Class II equivalent method. This method includes samplers and continuous analyzers based on designs and measurement principles different from those specified for reference methods as determined by EPA.

Sampling Artifacts Caused by Semi-Volatile Compounds. Semivolatile PM exists almost entirely in the fine ($PM_{2.5}$) fraction of the sample. The effects of semivolatile PM are particularly vexing because they may result in either an under-estimation of the $PM_{2.5}$ fraction (negative artifact) or an over-estimation of the $PM_{2.5}$ fraction (positive artifact). For example, particulate nitrates have been shown to incur loses as large as 50% of the total nitrate mass during sampling (Lipfert 1994; Tsai and Huang 1995; Shuang-Neng *et al.* 1996; Yu-Hsiang and Chuen-Jinn 1996; Eatough *et al.* 1999; Hering and Cass 1999; Chang *et al.* 2001; Pang *et al.* 2002). Semivolatile organic compounds (SVOC) have been shown to have losses as high as 50% of the SVOC (Ding *et al.* 2002; Pang *et al.* 2002; Eatough *et al.* 2003). On the other hand, sulfates have been shown to result in positive sampling artifacts on quartz filters (Shuang-Neng *et al.* 1996). A study in Riverside, California has shown that taking all artifacts into account, the $PM_{2.5}$ FRM sampler underestimated by an average of 34% (Pang *et al.* 2002).

Particle Distributions in the Rural Environment. It has generally been believed that fine particles are more strongly associated with mortality and morbidity than larger particles (EPA 1996). Fine particles typically originate as products of combustion or are formed from gases whereas coarse particles tend to be generated mechanically (Lundgren and Burton 1995). Data collected over the past 10 years at Texas A&M University do not indicate a significant quantity of fine particles in the rural environments sampled. Representative values of these data are shown in Tables 2 and 3.

Sampler Performance - Over and Under Estimation of PM Fractions. Engineering choices made in designing a PM sampler may result in systematic errors in the measurements. One example of this has been documented based solely on the published sampler performance characteristics and the distribution of PM in the atmosphere (Buser *et al.* 2003; Buser *et al.* 2003). The essence of the analysis is based on the fact that when size selective PM samplers are operated in the environment for which they are designed (i.e. an urban environment) then the measured concentrations of PM_{10} and $PM_{2.5}$ are very close to the true values and the derived PM_{CF} values are also close to true. However, when operated in an environment that is not representative of the one the sampler is designed for then the values of PM_{10} and $PM_{2.5}$ are over or under estimated and the calculated value for PM_{CF} can be in error.

The following scenarios have been developed to demonstrate how over and under estimation of the PM_{10} and $PM_{2.5}$ fractions can lead to errors in the values for PM_{CF}. These cases assume that the particle size is distributed according to a log normal distribution. The log normal distribution is described by two parameters; the mass median diameter (MMD) and the geometric standard deviation (GSD). Different distribution parameters of PM_{10} and $PM_{2.5}$ are used depending on the ambient environment. For these examples, $PM_{CF} = PM_{10} - PM_{2.5}$.

Three scenarios for urban PM were evaluated. The Urban 1 scenario was chosen to evaluate sampler performance with low GSD for both coarse and fine modes. Urban 2 represents the MMD and GSD characteristics identified as typical by Hinds (1982). Urban 3 represents the idealized size distributions for a reference PM_{10} measurement method (Chow 1995).

Four scenarios for rural distributions were evaluated. These correspond to measured particle size distributions associated with cattle feedyards, poultry broiler production, dairy, and cotton gin operations. These particle size distributions are described in Table 3.

Sampler performance was evaluated for each of these scenarios based on upper and lower ideal limits from EPA's range of acceptable performance characteristics published in 40 CFR parts 50 and 53. The sampler separation efficiency was applied mathematically to each size distribution to determine the concentration that would be measured if the sampler performs according to the upper or lower ideal limits. (Buser et al. 2003a). These measured concentrations as well as the "true" concentrations for $PM_{2.5}$, PM_{10}, and PM_{CF} are reported in Tables 4 – 6. Other sampling errors discussed in this report add to the uncertainty associated with each sampler's performance.

When exposed to PM dominated by large diameter particles as is typical of rural environments, especially near agricultural sources of PM, the samplers typically over estimate the concentration of $PM_{2.5}$, PM_{10}, and PM_{CF}. In urban conditions evaluated, the samplers may over or under estimate values for each PM fraction depending on the specific sampler and particle size distribution.

Measurement Uncertainty in Gravimetric Sampling. The determination of the concentration of particulate matter in air relies on a calculation of two values; the differential weight accumulated on a filter and the volume of air pulled through that filter during a given time. Dividing the weight by the volume yields the average concentration of PM in the atmosphere during that time. Even the TEOM, which yields a near-continuous estimate of PM concentration, relies on this principle. The TEOM utilizes the frequency shift in the tapered element oscillations as an indicator of accumulated mass and the mass of air as reported by a mass flow controller converted to volume to arrive at this estimate. Regardless of the instrument, these measurements, mass and volume, have associated with them some uncertainty (a.k.a. error) which is reflected in a final uncertainty in the reported concentration value.

Using a Taylor Series approximation, the total uncertainty surrounding determination of TSP concentration was determined for several gravimetric samplers (Price and Lacey 2003). Evaluation of the FRM TSP sampler indicated that the uncertainty in the final measurement ranged from 8 to 12% of the value, depending on the flow rate through the sampler, although all flow rates were within EPA guidelines. The measurement of the total volume of air through the filter accounted for 98% of the total uncertainty and the differential pressure measurement across the orifice meter accounted for 60% - 80% of the total uncertainty.

The FRM PM_{10} sampler operates under identical flow conditions, separating PM_{10} prior to measurement on a TSP filter, and the uncertainty in these measurements would be expected to be the same. It is possible that the uncertainty would be greater since the PM_{10} sampler must run for longer times and draw more air through the filter, in the same environment to collect sufficient PM for weighing.

Conclusions

There are a number of key differences between the urban and rural environments in the United States that can lead to critical mistakes in applying data from urban studies to the rural environment. These include differences in particle sources, which affect particle size distribution and composition, differences in the concentration of gaseous co-pollutants, and differences in PM sampler performance in the two environments. It is our contention that these differences between the urban and rural environment are significant and that the epidemiological studies cited by EPA rely on data that is not representative of the rural environment. These factors raise serious concerns that the implementation of a PM_{CF} standard in the rural environment will impose an unfair and unwarranted regulatory burden on the businesses and citizens in the rural areas.

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Table 1. Performance criteria specified by EPA in 40 CFR Parts 50 and 53.

Fraction	Lower Limit µg/m^3	Minimum Upper Limit µg/m^3	Sampler Cutpoint µm	Sampler Performance Slope	Specified Precision
PM_{10}	Depends on weighing equipment	300	10 ± 0.5	1.6	5 µg/m^3 for concentrations below 80 µg/m^3 and 7% for concentrations above 80 µg/m^3
$PM_{2.5}$	2	200	Class II - 2.5 ± 0.2	1.186	Data precision of 10% coefficient of variation

Table 2. PM characteristics of rural environments.

PM Source	MMD (µm)	GSD (µm)
Gin Dust (Wang *et al.* 2002)	23	1.8
Cornstarch (Wang *et al.* 2003)	20	1.4
Broiler Dust (Buser *et al.* 2003)	24	1.6
Feedyard Dust	20	2.2
Dairy Dust	19	2.1

Table 3. PM size distributions used in sampler performance evaluation.

		MMD		TSP	PM_{10}	PM2.5	PMcf
Scenario	Mode	µm	GSD	µg/m^3	µg/m^3	µg/m^3	µg/m^3
Urban 1	Coarse	10	1.6	124.29	62.14	0.20	61.95
	Fine	1	1.2	82.86	82.86	82.85	0.01
	TOTAL			207.14	145.00	83.04	61.96
Urban 2	Coarse	8	2.3	154.43	93.52	12.55	80.97
	Fine	0.3	2.05	51.48	51.48	51.40	0.08
	TOTAL			205.91	145.00	63.95	81.05
Urban 3	Coarse	14	2	225.00	70.58	1.46	69.12
	Fine	0.5	2	75.00	74.99	74.24	0.75
	TOTAL			300.00	145.57	75.70	69.87
Feedyard	Coarse	20	2.2	764.49	145.00	3.19	141.81
	Fine	0	0	0.00	0.00	0.00	0.00
	TOTAL			764.49	145.00	3.19	141.81
Broiler	Coarse	24	1.6	4639.51	145.00	0.00	145.00
	Fine	0	0	0.00	0.00	0.00	0.00
	TOTAL			4639.51	145.00	0.00	145.00
Dairy	Coarse	19	2.1	749.39	145.00	2.35	142.65
	Fine	0	0	0.00	0.00	0.00	0.00
	TOTAL			749.39	145.00	2.35	142.65
Cotton Gin	Coarse	23	1.8	1853.33	145.00	0.15	144.85
	Fine	0	0	0.00	0.00	0.00	0.00
	TOTAL			1853.33	145.00	0.15	144.85

Table 4. $PM_{2.5}$ sampler performance for the scenarios evaluated[1].

Scenario	True $PM_{2.5}$ µg/m^3	Ideal Lower $PM_{2.5}$ µg/m^3	Ideal Upper $PM_{2.5}$ µg/m^3
Urban 1	83.04	83.02	83.40
Urban 2	63.95	62.34	66.95
Urban 3	75.70	75.06	76.70
Feedyard	3.19	2.80	4.99
Broiler	0.00	0.01	0.03
Dairy	2.35	2.08	3.89
Cotton Gin	0.15	0.16	0.43

[1] Ideal Lower and Ideal Upper refer to performance characteristics from 40 CFR parts 50 and 53.

Table 5. PM_{10} sampler performance for the scenarios evaluated[1].

Scenario	True PM_{10} µg/m^3	Ideal Lower PM_{10} µg/m^3	Ideal Upper PM_{10} µg/m^3
Urban 1	145.00	141.17	148.65
Urban 2	145.00	139.70	145.98
Urban 3	145.57	147.36	157.25
Feedyard	145.00	159.54	184.51
Broiler	145.00	378.65	495.51
Dairy	145.00	161.12	187.17
Cotton Gin	145.00	222.44	275.65

1 Ideal Lower and Ideal Upper refer to performance characteristics from 40 CFR parts 50 and 53.

Table 6. PM_{CF} measurement range for the samplers and scenarios evaluated.

Scenario	True PM_{cf} µg/m^3	Lower PM_{cf} µg/m^3	Upper PM_{cf} µg/m^3
Urban 1	61.96	57.76	65.63
Urban 2	81.05	72.75	83.64
Urban 3	69.87	70.66	82.19
Feedyard	141.81	154.55	181.71
Broiler	145.00	378.62	495.5
Dairy	142.65	137.7	183.01
Cotton Gin	144.85	222.01	275.49

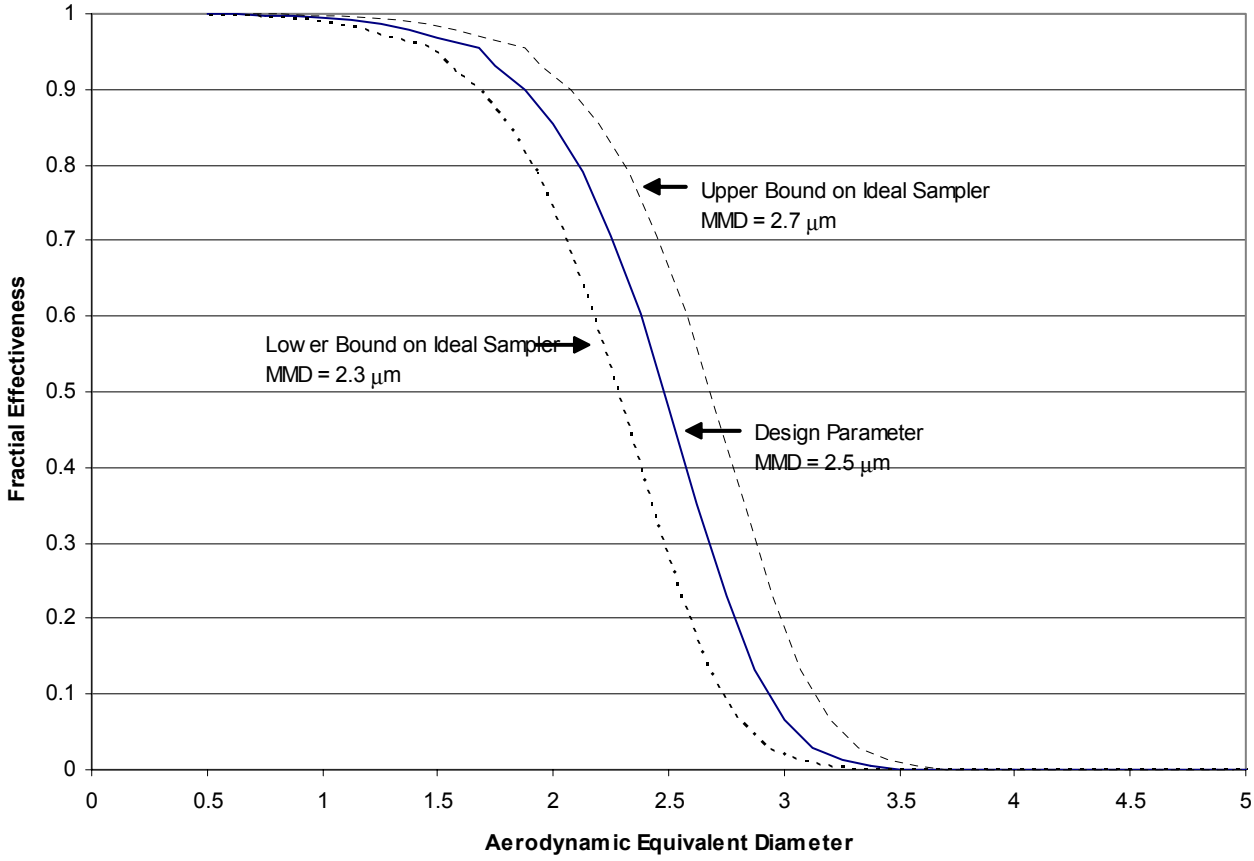

Figure 1. $PM_{2.5}$ sampler efficiency curve for the FRM guidelines (40 CFR Part 53).