PARTICLE SIZE DISTRIBUTION OF PARTICULATE MATTER EMITTED BY AGRICULTURAL OPERATIONS: IMPACTS ON FRM PM_{10} AND $PM_{2.5}$ CONCENTRATION MEASUREMENTS

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Abstract

The EPA approved federal reference methods (FRM) for measuring particulate matter concentrations of PM_{10} and $PM_{2.5}$ are affected by the particle size distribution (PSD) of the particulate matter (PM) present in ambient air. The PSD of PM in the ambient air emitted by agricultural operations (rural areas) is significantly larger than that of PM present in urban areas. Typically, urban PM will have a mass median diameter (MMD) less than 10 μm aerodynamic equivalent diameter (AED) whereas, agricultural PM will have an MMD larger than 10 μm AED. An MMD of as high as 24 μm AED has been reported for PM emitted through agricultural operations. The EPA- approved FRM PM_{10} and $PM_{2.5}$ samplers have been shown to exhibit over-sampling problems for particles having MMDs of greater than 10 μm such as agricultural dust. As a consequence, agricultural operations are not being regulated fairly. This work presents some procedures that can be done to correct this over sampling problem.

Introduction

Particulate matter (PM) in ambient air of less than 10 microns (PM_{10}) and 5 microns ($PM_{2.5}$) in size are currently being regulated by the National Ambient Air Quality Standards (Cooper and Alley, 2002). The concentrations of PMs in ambient air are measured using the EPA-approved Federal Reference Method (FRM) samplers. Size selective PM concentration measurements are made using a two-stage process. The first stage consists of a pre-separator designed to remove the larger particles. The second stage consists of a filter used to accumulate the PM mass that penetrated the first stage. For a PM_{10} sampler, for example, the first stage theoretically removes particles larger than 10 micrometers (μm) aerodynamic equivalent diameter (AED). The net mass of PM collected on the filter from the second stage divided by the volume of air sampled provides a measure of the concentration in units of mass per unit volume (ug/m^3). If the mass of PM on the filter is more than the mass of PM_{10} present in the ambient air, the concentration measurement over-sampled the ambient air concentrations in terms of PM_{10}.

For the FRM PM_{10} sampler, an ideal pre-separator (virtual cut) would separate all PM larger than 10 μm, allowing all PM less than 10 μm to be captured by the filter. It is not possible to engineer a pre-separator with a virtual cut at 10 μm. The engineering description of the performance of a pre-separator is the fractional efficiency curve (FEC). This is a mathematical description of the percent mass captured versus particle size (see Figure 2). *Two parameters define a pre-separator FEC: cut-point (d_{50}) and slope of the penetration curve ($d_{84.1}/d_{50}$).* These parameters are typically assumed to be constant and the curve is most commonly represented by a lognormal distribution. The cut-point is the particle size where 50% of the targeted sized PM is captured by the filter and 50% are not. The slope is the ratio of the 84.1% and 50% particle sizes ($d_{84.1}/d_{50}$) or the ratio of the 50% and 15.9 % particle sizes ($d_{50}/d_{15..9}$) from the FEC (Hinds, 1982). If the slope of the fractional efficiency curve is greater than 1.0 for any PM_{10} sampler, a fraction of the PM larger than 10 μm is removed by the pre-separator and a fraction is captured by the filter. This condition is likewise true for particles smaller than 10 um. The 50% efficiency means that the fraction of smaller particles captured is equal to the portion that were not captured and the error cancels each other to give the true PM_{10} reading. Research results have shown that the performance of EPA-approved FRM samplers are affected by the particle size distribution (PSD) of the PM present in ambient air**.**

The FRM $PM_{2.5}$ sampler on the other hand is assumed to have a d_{50} of 2.5 μm and a slope of 1.18 (EPA, 2001). Studies by Buch (1999) and Pargman, et al. (2001) showed that there was a shift in the cut-point for FRM $PM_{2.5}$ to 2.7$\pm$0.41 μm and a slope of 1.32$\pm$.03 μm. This shift in the cut-point also creates over sampling problems.

No particle sizing of the captured dust by both the PM_{10} and $PM_{2.5}$ samplers is required by the EPA. Whatever is collected by the sampler is assumed to be PM_{10} or $PM_{2.5}$. However, as will be shown in the following discussion, when the dust particles have MMD greater than 10 μm, over-sampling is likely. For the $PM_{2.5}$ samplers, because of the shift in the cut point, even for particles with an MMD of 5 µm and a GSD (geometric standard deviation) of 2.0, over-sampling may occur.

PSD of Agricultural Dusts

The PSD of any dust can be characterized by its mass median diameter (MMD) and geometric standard deviation (GSD). Table 1 gives a summary of the oversampling errors observed for different types of agricultural dust, namely cotton gin dust, cornstarch, broiler dust, feedyard dust and dairy dust. The MMD of these dusts ranged from 15-24 and the GSD ranged from 1.4 to 2.8. Figure 1 shows plots of the actual PSD of the five agricultural dusts as determined using the Coulter Counter Multisizer (Shaw, et al., 2002). The particles less than 10 µm comprised only a very small fraction of the total PSD for all the samples. The PSD of these agricultural dusts are shown to follow a lognormal distribution (Buser, et al., 2002).

Performance of FRM PM_{10} and $PM_{2.5}$ Sampler

The FEC of a PM_{10} sampler with a cut-point of 10 $\pm$ 0.5 µm and a slope of 1.5 $\pm$ 0.1 µm is shown in Figure 2. The lower limit of its performance is shown on the curve on the left and the upper limit, on the right. The middle portion is for the cut-point of 10 µm and a slope of 1.5 µm. Superimposed in this figure (hard black line) is the virtual cut-point of 10 µm and a slope of 1. This line would show that all particles less than 10 µm are captured by the filter and those larger than 10 µm are removed by the pre-separator. An actual field sampler could not have such an accurate performance. A thorough discussion on the performance of FRM PM_{10} and $PM_{2.5}$ sampler has been presented by Parnell, et al. (2000). It will be illustrated in this document how this particular sampler will perform with various dust types.

Illustrative Examples for FRM PM Concentration Measurements and Over-sampling Problems

To illustrate the performance of an FRM PM sampler on a variety of dust samples, simulated runs were made for particles having an MMD of from 5.0 to 25 µm with a GSD of 2.0. In the first set of runs, the concentration of the total suspended particles (TSP) of ambient air was assumed to be 1000 **µ**g/m^3, the sampler cut-point was 10 **µ**m and slope was 1.5 **µ**m. The results of performance tests of the PM_{10} sampler are shown in Table 2. As indicated in the table, the PM_{10} sampler collected 500 µg/m^3 (the true concentration) of PM_{10} for particles with an MMD of 10 and a GSD of 2.0. If the particles have an MMD of 5.0 and a GSD of 2.0, the true PM_{10} would have been 840 µg/m^3 but the sampler was only able to capture 800 µg/m^3 of PM_{10} which is a 40 µg/m^3 under-sampling. Likewise for particles with an MMD of 20 and a GSD of 2.0, the true PM_{10} would be 160 µg/m^3 but the sampler was able to collect 190 µg/m^3 PM_{10}, a clear over sampling by more than 30 µg/m^3. Thus, over sampling increases as the MMD of the particle gets larger. Figure 3 shows the results of the performance evaluation of the PM_{10} sampler for dust with 1000 ug/m^3 TSP. Shown in the figure are the ranges of PM_{10} concentrations a sampler would report if its performance is based on a cut point of 10 $\pm$ 0.5 µm and a slope of 1.5 $\pm$ 0.1.

Another set of runs made use of the assumption that the ambient air dust sample has a true PM_{10} of 140 µg/m^3, which is below the standard maximum of 150 µg/m^3. Figure 4 shows the concentrations measured by the PM_{10} sampler compared with the true PM_{10} for the differently-sized particles. Correct sampling is expected for particles with MMD of 10 m. For dust particles with an MMD of 5 µm, there would be an under sampling of 6 µg/m^3. Over sampling would be obtained when the dust particles have MMD of ≥15 µm. With MMD of 15 m, over sampling gives a PM_{10} concentration of 154 mg/m^3, which is already above the maximum allowed by current regulation. The results clearly indicate that even for dust samples with actual PM_{10} within the acceptable level, concentration readings from samplers may indicate values way above the regulation standard. Shown in Figure 5 is the over sampling calculated for the five agricultural dusts. Although there was no over sampling for dairy and feedyard dusts, cotton gin, corn and broiler dust had an over large sampling of 78 µg/m^3, 529 µg/m^3 and 215 µg/m^3, respectively. The upper and lower limits of PM_{10} dust concentrations collected by a PM_{10} sampler for dusts of different MMDs are shown in Figure 6.

The results of similar simulated runs done for the $PM_{2.5}$ sampler are illustrated in Figures 7 and 8. The performance of the $PM_{2.5}$ sampler was based on the results of work conducted by Buch (1999) with the sampler having a cut point of 2.7$\pm$0.41 µm and a slope of 1.32$\pm$0.03 µm. For figures 7 and 8, it was assumed that the dust samples have a true $PM_{2.5}$ concentration of 60 µg/m^3, which is again below the max of 65 µg/m^3 NAAQS standard. For all the dust types of sizes 5-25 m MMD over-sampling is observed. The expected $PM_{2.5}$ concentration for the smallest particle size of 5.0 m (GSD 2.0) is above the limit for the $PM_{2.5}$ concentrations by about 20 µg/m^3, already a violation of the NAAQS standard. Figure 8 shows the ranges of $PM_{2.5}$ dust concentrations collected by the sampler from various agricultural dusts. Over sampling is again evident. In all instances, the measured $PM_{2.5}$ is way above the true $PM_{2.5}$ concentration, in violation of the standards, despite the fact that the true $PM_{2.5}$ is supposedly wtihin the standard.

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Summary of Issues and Problems

We have shown that inherent problems are expected for PM_{10} and $PM_{2.5}$ samplers when operating on agricultural dusts or dust particles with an MMD of greater than 10 um. Agricultural dusts have PSDs much larger than urban dusts for which the criteria was originally based. This difference between the PSD of many ambient dusts in rural areas with that of urban dust has been overlooked. The MMD of urban dust is normally below 10 µm (EPA, 2001). For the $PM_{2.5}$ sampler, the error was compounded by the shift in its cut-point. This shift in the cut-point showed that even for dust particles with an MMD of 5 µg/m^3, over-sampling is likely.

It is not possible to design a sampler with a virtual cut for a specific particle size. Thus, a procedure has to be developed to correct the biases without resorting to redesigning the thousands of EPA-approved samplers already in place in most parts of the country. There are several ways of ensuring a correct estimate of PM_{10} in ambient air. Foremost is the determination of PSDs of all ambient dust types by fractionation according to size and determining the weight of sizes less than 2.5 or 10 µm. This will require the use of equipment such as the Coulter Counter Multisizer, or some other devices such as the WINs Impactor and the like. Unfortunately, these devices are expensive and only limited research facilities have them. Once the PSD is determined, the PM_{10} or $PM_{2.5}$ concentrations are readily calculated as a mass fraction. One other option is to establish the MMD and GSD of several dust types under ideal conditions and publish their PM_{10} or $PM_{2.5}$ concentration. Using a log normal distribution, the amount of PM_{10} or $PM_{2.5}$ may be easily determined. In this procedure, one has to refer to published values of MMD and GSD and run a log-normal distribution using a spreadsheet software or MATHCAD. Another way of correcting over-sampling is to use the ratio of PM_{10}/TSP readings bya PM_{10} and a TSP sampler for various dust types. Our past research has shown that when this ratio is established, the measured PM_{10} could be corrected (Parnell, et al., 2003). The procedure is briefly discussed below.

Correcting for Over-Sampling

Figure 9 shows the measured feedyard PM concentrations after a rain event. It can be observed that the ratio of PM_{10} to TSP becomes linear as the fugitive dust gets drier. The collocating of PM_{10} and TSP samplers has been a practice in our work with agricultural dusts to allow a means of double checking the concentration of PM_{10} using the TSP sampler. The accuracy of the PM_{10} sampling test can thus be validated. When the PSD of the captured dust on the filter from the TSP sampler is determined using our Coulter Counter Multi-sizer, the fraction of PM_{10} in the collected dust can be determined. Over the years of testing, both PM_{10} concentrations in the PM_{10} sampler and the PM_{10} concentrations from the TSP sampler have been recorded using the actual PSD. These data from sampling tests have been used to establish a relationship between PM_{10} concentrations and TSP concentrations.

The 4th to 6th columns of Table 1 list the results of an iterative process to derive the true PSD of ambient PM by using PM_{10} and TSP collocated measurements for five agricultural dusts. Figure 10 illustrates a graph showing the measured PM_{10}/TSP ratio and corrected PM_{10}/TSP ratio. The curve is approximated by the following equation:

$$CR = 1.1443 * MR - 0.0746 \quad \ldots\ldots(1)$$

Where,

CR = corrected ratio of PM_{10}/TSP

MR = measured ratio of PM_{10}/TSP

The measured PM_{10}/TSP ratio is calculated by dividing the PM_{10} concentrations by the TSP concentrations from collocating both samplers. The corrected PM_{10}/TSP is obtained from the curve by projecting the point to the y-axis However, this graph is applicable only for a specific dust particle with a given MMD and GSD. A more detailed illustration of this procedure has been presented by Wang, et. al., (2004).

Conclusion

The performance of EPA approved PM_{10} and $PM_{2.5}$ samplers are affected by the PSD of PM in ambient air. The PM_{10} sampler was meant to operate with a cut-point of 10 um and a slope of 1.5 and applicable for dust with an MMD of about 10 um. Likewise the $PM_{2.5}$ sampler is supposed to have a cut-point of 2.5 um and a slope of 1.18. It is impossible to design a PM_{10} sampler that provides perfectly sharp cuts at 10 µm or a $PM_{2.5}$ sampler with a virtual cut-point of 2.5 um. There will be particles larger than 10 µm that will be captured by the PM_{10} sampler and particles smaller than 10 µm that are not collected and those two values may not be equal for dust particles with MMD other than 10 µm. The particle size distribution of PM in the ambient air emitted by agricultural operations is significantly larger than PM present in urban areas. When PM_{10} samplers are operated under agricultural dust types, over sampling has been shown to occur. As a consequence, agricultural operations are not properly regulated. Agricultural PM's have MMD greater than 10 µm and over sampling may be as much as 810% on some actual agricultural dust samples. For the EPA approved $PM_{2.5}$ sampler, the predicted cut-point was 2.7 µm with a slope of 1.32. Evaluation of its performance for the different types of dust samples showed that over sampling may occur for dust particles with an MMD of as low as 5 µm. Thus, dust particles having an MMD of greater than 10 um may be inappropriately regulated for PM_{10} concentrations and those with an MMD of 5 µm would not be properly regulated for $PM_{2.5.}$

Some procedures to correct for these over sampling problems were presented. One method is to determination the PSDs of all dust samples to estimate an accurate amount of PM_{10} but this would require very expensive equipment like particle counters and impactors. The MMD and GSD of a given dust particle may also be established and a log normal distribution may be used to determine the exact amount of PM_{10} in a given sample. Another procedure presented in this work is the use of concentration ratios of PM_{10} to TSP sampler by collocating those samplers. Our studies have shown that a linear relationship exists between PM_{10} and TSP sampler readings for a given dust type. When this relationship is established, the correct PM_{10} may be estimated through an iterative process.

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Table 1. Summary of PM_{10} over-sampling error for agricultural dust.

Agricultural Dust Type	PSD of Agricultural Dust		Sampler concentration/True concentration		
			d50=9.5	d50=10	d50=10.5
	MMD	GSD	slope=1.4	slope=1.5	slope=1.6
Cotton Gin Dust (Wang, et. al. 2002)	23	1.8	120%	150%	181%
Cornstarch (Wang, et.al. 2002)	20	1.4	343%	565%	810%
Broiler Dust (Redwine, et. al., 2002)	24	1.6	159%	225%	297%
Feedyard Dust	17	2.8	92%	104%	105%
Dairy Dust	15	2.5	92%	104%	106%

Table 2. Amount of PM_{10} captured and over sampling by the PM_{10} sampler for different dust particles having different MMD's and GSD's and with TSP concentration of 1000 μg/m^3.

			True PM10	Amt Captured	Over sampling/(Under sampling)
Dust Type	MMD	GSD	ug/m3	ug/m3	ug/m3
Simulated Dust	5.0	2.0	841	806	(35)
Simulated Dust	10.0	2.0	500	500	0
Simulated Dust	15.0	2.0	280	307	27
Simulated Dust	20.0	2.0	160	194	34
Simulated Dust	25	2.0	93	127	34
Dairy Dust	15	2.5	330	343	13
Feedyard Dust	17	2.8	303	316	13
Broiler Dust	24	1.6	31	79	48
Cornstarch	20	1.4	20	94	74
Cotton Gin Dust	23	1.8	78	122	44
Almond Dust	17	2.1	237	265	28

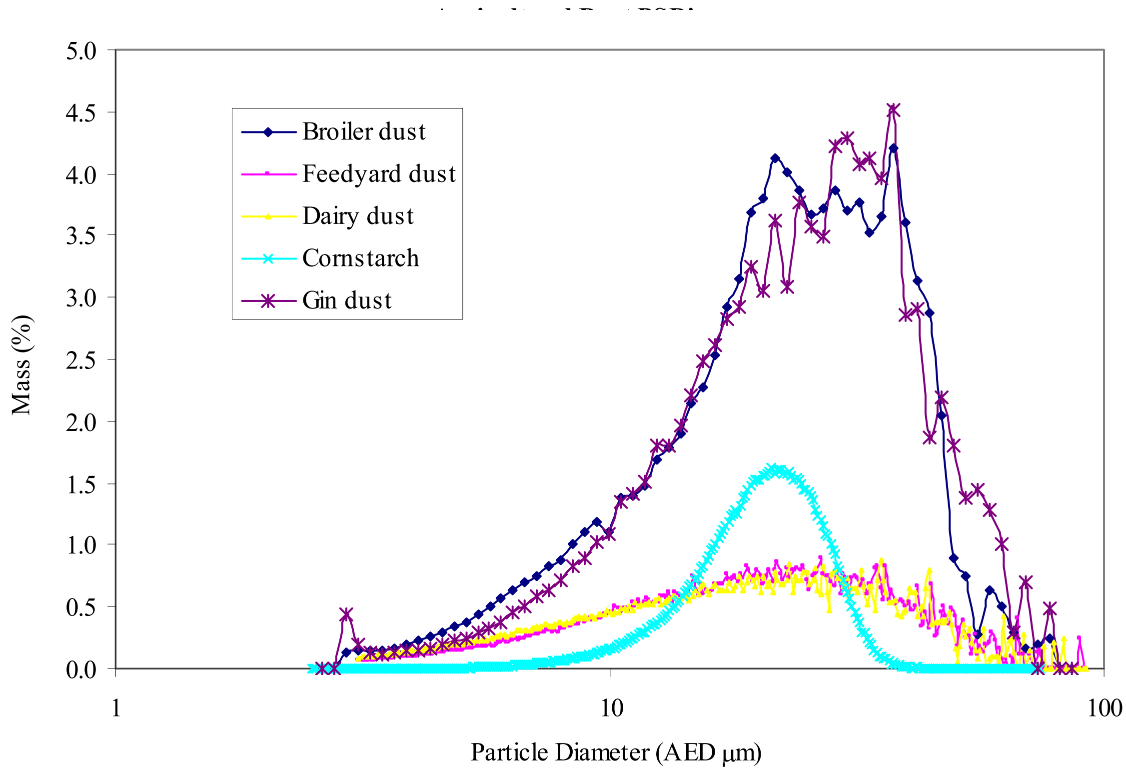

Figure 1. PSD of agricultural dusts using the Coulter Counter Multisizer.

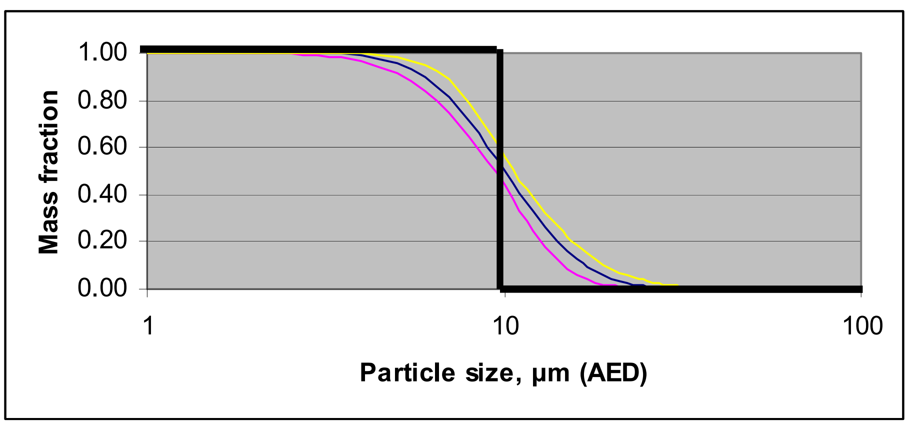

Figure 2. Fractional efficiency curve (FEC) for a PM10 sampler showing the lower, middle and upper limit of operation and the virtual cut line (dark line).

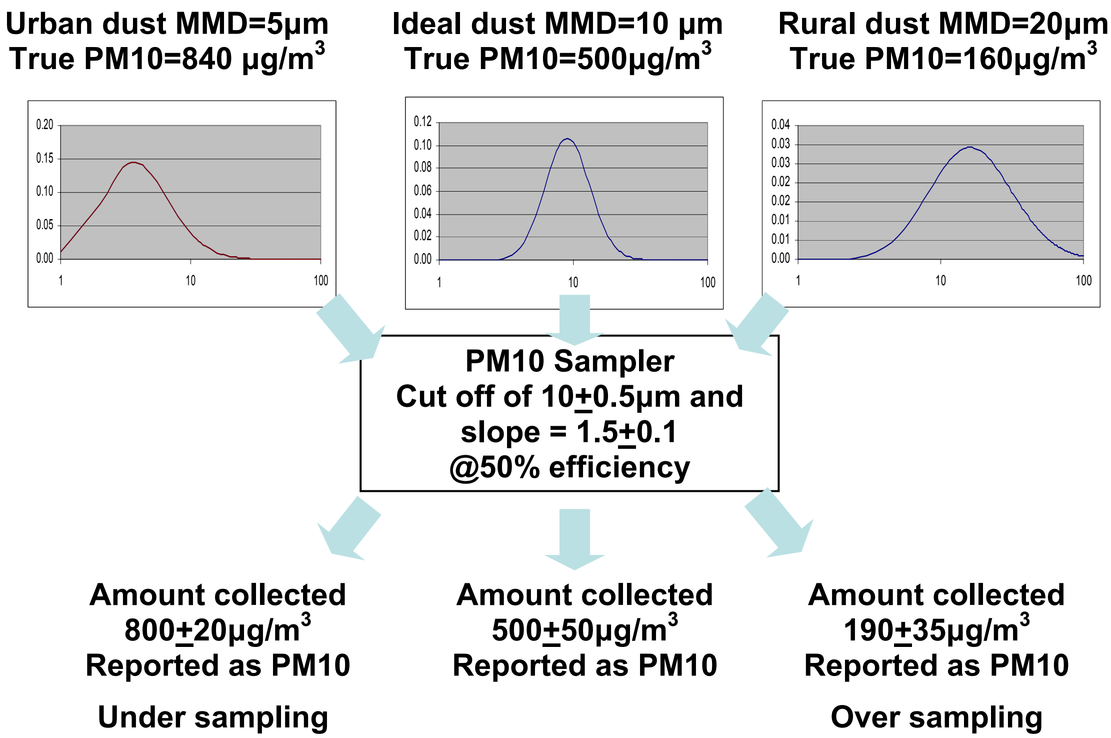

Figure 3. Performance of the PM_{10} sampler for dust with 1000 µg/m^3 TSP.

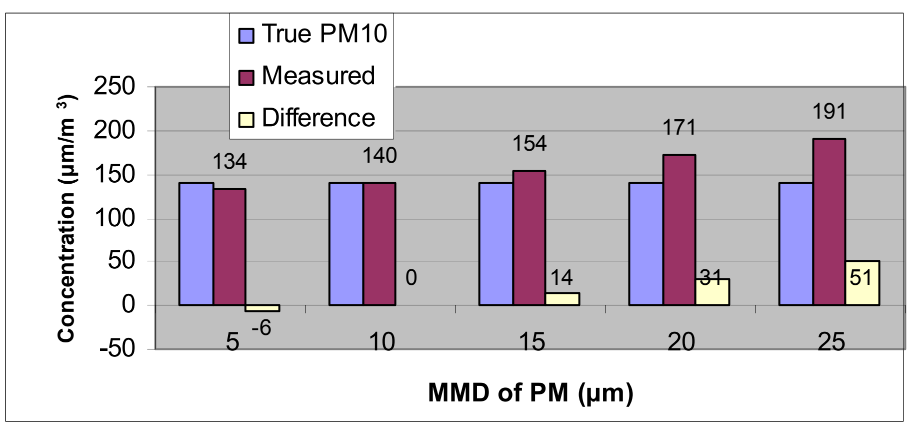

Figure 4. Amount of PM_{10} captured by an FRM PM_{10} sampler for dusts of varying MMD showing under and over sampling. (True PM_{10} = 140 µg/m^3)

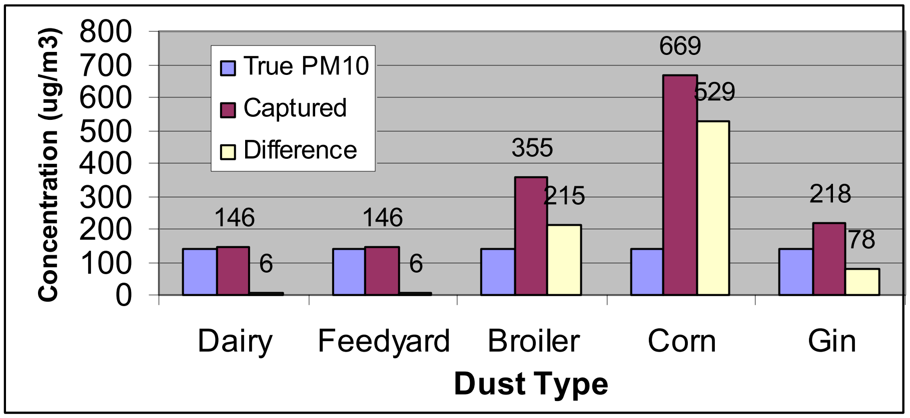

Figure 5. Amount of PM_{10} captured by an FRM PM_{10} sampler for different agricultural dust types showing over sampling (True PM_{10} = 140 μg/m^3).

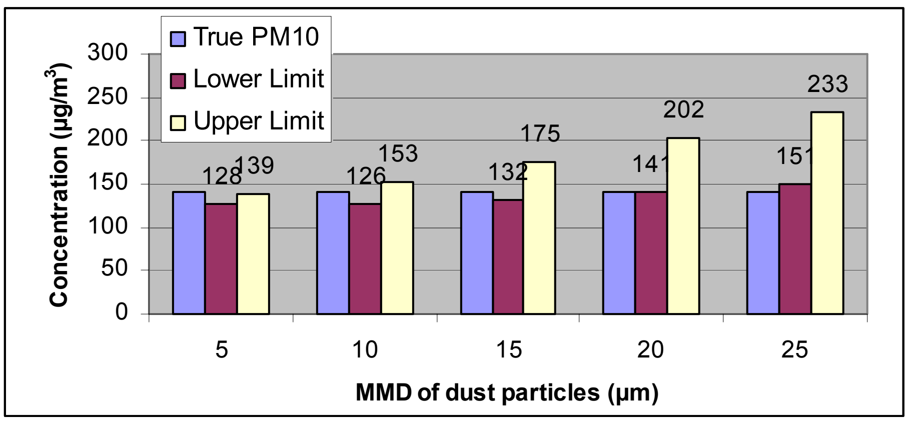

Figure 6. Possible range of concentrations reported by a PM_{10} sampler (cut-point of 10 $\pm$ 0.5 um and slope of 1.5 $\pm$ 0.1) for dusts with different MMD's (True PM_{10} is 140 μg/m^3).

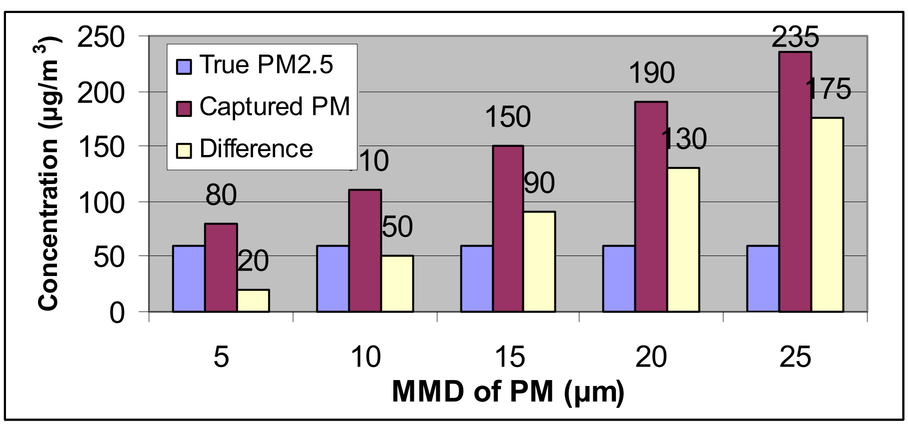

Figure 7. Amount of $PM_{2.5}$ captured by an FRM $PM_{2.5}$ sampler for dust types (given MMD) with a true $PM_{2.5}$ concentration of 60 μg/m^3 showing over sampling for all dust types.

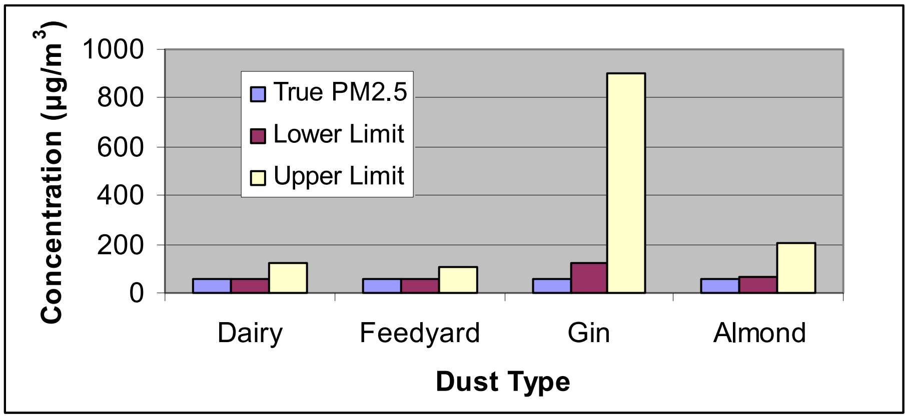

Figure 8. . Possible range of concentrations reported by a $PM_{2.5}$ sampler (cut-point of 2.7 $\pm$ 0.41 um and slope of 1.32 $\pm$ 0.03) for agricultural dusts (True $PM_{2.5}$ is 60 µg/m³).

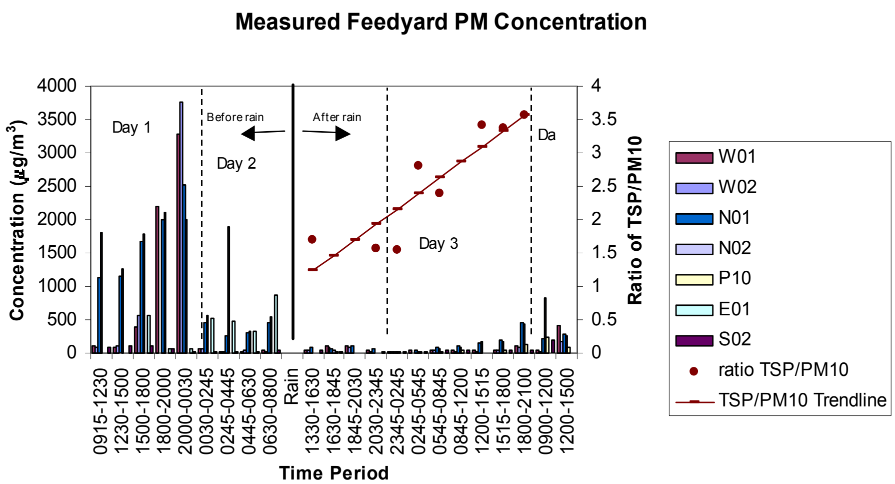

Figure 9. PM concentrations in a feedyard before and after a rain event showing the relationship between PM_{10} and TSP concentration measurements.

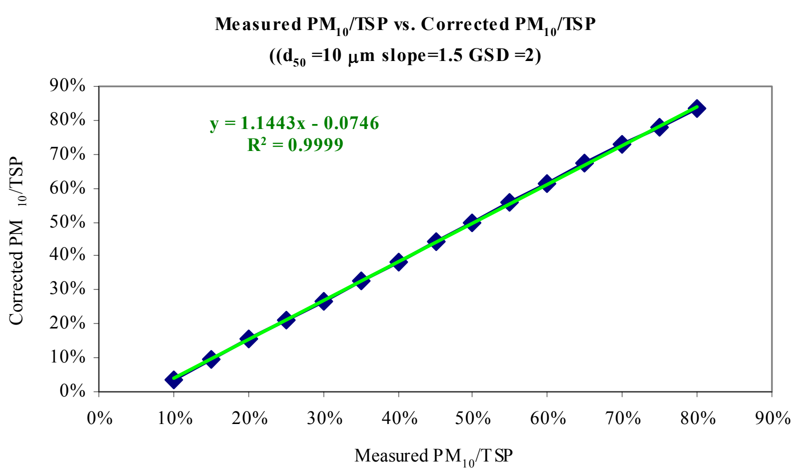

Figure 10. Graph used for correcting PM_{10} concentrations using collocated PM_{10} and $PM_{2.5}$ sampler.