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COMPARISON OF PARTICLE SIZING METHODS FOR MEASUREMENT OF AIR EMISSIONS FROM AGRICULTURAL OPERATIONS

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Abstract

The faculty at the Center for Agricultural Air Quality Engineering and Science over the past few decades developed a method of accurately determining particulate matter concentrations from industries that emit air pollution. The CCM method of characterizing the size distribution of aerosols has shown that the EPA approved PM_{10} and $PM_{2.5}$ samplers over sample agricultural dusts. New methods of determining particulate matter size distributions are continuously sought out for possible use. This study compares two new methods – A and B – to the CCM method.

All three methods have distinct advantages and disadvantages for use in agricultural settings. Method A's disadvantages were found to outweigh its advantages and therefore will not be investigated further. Method B shows promise as an instrument for use in agricultural applications. However, differences between the sizes reported compared to the CCM for two separate dusts indicate that more testing should be performed on Method B with a different distribution system.

Introduction

The Environmental Protection Agency (EPA) requires that the primary pollutant PM_{10} (particulate matter equal to and less than 10 microns) be sampled with an approved PM_{10} sampling device. Likewise, EPA requires the primary pollutant, $PM_{2.5}$, to be sampled with an approved $PM_{2.5}$ sampling device. Sampling devices are defined in the EPA's publication 40 CFR Part 53. When sampling urban dusts, these samplers typically provide an accurate sample of the particulate size in question, meeting the definition of the respective sampler. However, when such devices sample particulate matter from typical agricultural operations, the amount of PM_{10} and $PM_{2.5}$ is over predicted. Both Pargmann, et. al. (2001) and Buser et. al. (2002) determined that this over prediction is due to (1) sampler uncertainties, (2) environmental conditions and (3) characteristics of the particulate matter being sampled

To circumvent over prediction, the Center for Agricultural Air Quality Engineering & Science (CAAQES) operating out of the Biological and Agricultural Engineering Department of Texas A&M University, employs a system whereby PM_{10} and $PM_{2.5}$ concentrations are more accurately determined than with the EPA approved samplers. This system, referred as CCM, consists first of capturing particulate with a TSP (total suspended particulate) sampler onto a filter. TSP samplers are designed to capture particles 45 microns and less, thereby capturing PM_{10} and, consequently, $PM_{2.5}$. Secondly, the staff members of CAAQES determine the TSP concentration of the particulate matter. Thirdly, a sampler from the filter is run through a Coulter Counter Multisizer III (Coulter), to determine the particle size distribution (PSD) of the particulate matter captured by the TSP. The PSD analyses compare percent mass versus particle size. Finally, the mass fraction of PM_{10} or $PM_{2.5}$ of all particulate matter captured by the TSP is determined and multiplied by the TSP concentration to give PM_{10} or $PM_{2.5}$ concentrations. The assumption is made that particle density does not vary between the different sizes of the particulate matter sampled. The CCM method of determining PM concentrations has been used at Texas A&M University since the 1980's.

The CCM method is a reliable method for determining PM_{10} and $PM_{2.5}$ concentrations. With advances in sizing technology and greater knowledge of particle behavior, alternative methods of determining PSD have been made commercially available. Many instruments now available only provide the user with particle count versus particle size. However, CAAQES requires the determination of percent mass or volume versus particle size in order to calculate PM_{10} and $PM_{2.5}$ concentrations from TSP concentrations. Two such alternative instruments are discussed herein and evaluated based on application, advantages and possible disadvantages.

Background

When studying air quality and air pollution engineering it is often necessary to obtain the PSD of particulate matter entrained in the air or the gas of interest. This PSD gives us substantial information about how that particulate matter will behave when transported in an aerosol.

There are many aspects of particle size that are of interest to researchers and industry. Some people may be interested in the greatest length across a particle while others may be interested in the volume of the particle. For our purposes, we are most interested in the aerodynamic diameter (d_a). The aerodynamic diameter is defined as the diameter of a unit density sphere that has the same settling velocity as that of the particle in question (Hinds, 1999). This diameter is the best description of

how a particle will behave in an aerosol. It can be used to predict whether a particle will be captured by an abatement device or filter, or how far it can penetrate into the respiratory system. This measure is useful because it is independent of particle shape, material or density.

The primary problem with using aerodynamic diameter is in trying to measure it. By definition, the measurement would need to be made by some sort of sedimentation method in still air. While instruments do exist that perform this method (Timbrell, 1972), the times involved are generally too long for regular analysis of large numbers of samples. Therefore, other methods of determining aerodynamic diameter have been developed that use measurements of other particle properties as surrogates of aerodynamic diameter.

Three of the more commonplace methods are the electrical sensing zone method, the time-of-flight method and the light scatter method. The electrical sensing zone (ESZ) method, by which the Coulter operates, uses a device that measures the impedance caused by a particle passing through an electrical sensing zone. This impedance is related to the particle's volume. The volume is then converted into an equivalent spherical diameter by the formula:

$$d_e = \left(\frac{6V}{\pi}\right)^{1/3} \tag{1}$$

where V is the particle volume obtained from the ESZ instrument and d_e is the equivalent spherical diameter, which is not the same as the aerodynamic diameter. To convert from d_e to d_a, the following formula is used

$$d_a = d_e\left(\frac{\rho_p}{\rho_o}\right)^{1/2} \tag{2}$$

where ρ_p is the particle density and ρ_o is unit density (1 g/cm^3). This method works well for spherical or nearly spherical particles but can perform poorly for non-spherical and irregularly shaped particles (Tomb and Corn, 1973, Chung and Thompson, 1989). When dealing with agricultural dusts we rarely see spherical shapes. We are often times dealing with soil particles that have very irregular shapes. Because of these irregular shapes, the above formula can yield incorrect aerodynamic diameters. One method of addressing this potential error is the use of a dynamic shape factor, χ. The dynamic shape factor is a ratio of the actual drag force to that of a sphere of the same size (Hinds, 1999) and is determined experimentally. When this factor is used, equation 2 becomes

$$d_a = d_e\left(\frac{\rho_p}{\rho_o\chi}\right)^{1/2} \tag{3}$$

Another instrument used to estimate aerodynamic particle size will be referred to as Method A. Method A uses the time-of-flight method. This method operates by pulling a sample directly from the aerosol of interest and accelerating it through a nozzle. As the air stream passes through the nozzle it crosses two LASER beams. These LASER beams measure the time it takes for a particle to pass between them. This time reflects the velocity of the particle, which is related to the drag force on and density of the particle. The instrument is calibrated such that the velocity for each aerodynamic size is known (Miller and Lines, 1988).

A third instrument referred to as Method B uses a type of light scatter method, referred to as LASER diffraction. This method is comprised of a LASER beam focused and pointing through the center of a detector array and onto a second detector. When a particle entrained from a sample enters the LASER beam, it will block the beam from the second detector and scatter light onto the detector array. The angle of light scatter produced by the particle blocking the beam is inversely proportional to the particle size. Small particles will produce large angles of scatter while large particles will produce smaller angles of scatter. Method B normally reports particle diameter as the equivalent spherical diameter, d_e, therefore equation 2 is used to convert to aerodynamic diameter, d_a.

Discussion

The three methods, CCM, Method A and Method B each have advantages and disadvantages to providing concentrations when applied to agricultural sampling of dusts. These advantages and disadvantages will be discussed as they relate to each other and the EPA approved sampling devices.

The EPA approved PM_{10} and $PM_{2.5}$ samplers both allow for ease in determining respective concentrations in $\mu g/m^3$ by (1) pre- and post-weighing a filter used for collecting particulate matter with the sampler, (2) taking the mass difference to determine the weight of particulate collected (assumed to be $PM_{10}/PM_{2.5}$) and (3) dividing by the volume of air flow through the filter. All three methods under consideration require added laboratory work and/or calculations to determine $PM_{10}/PM_{2.5}$ concentrations in $\mu g/m^3$.

An advantage to employing the CCM method and Method B is that a TSP sampler can be used in the field to capture the particulate matter for subsequent laboratory particle size analysis as opposed to a PM_{10} sampler that has been shown to over predict concentrations. Moreover, Method A requires that samples be taken directly from the aerosol of interest in order to determine settling velocity; so, one disadvantage of Method A is that a $39,000 piece of equipment must be taken to the field for use. In agricultural settings, any number of forces could cause damage to the equipment, for example, cattle at a feed lot could easily knock the equipment over.

While the CCM method and Method B equipment remain in the laboratory and perform testing on sample filters taken from the field, each method comes with an expense. The CCM method instrument, Coulter Counter Multisizer III, costs approximately $40,000, and the equipment used for determining the PSD in Method B costs approximately $65,000.

One may ask why samples from the TSP filters could not be used to run particle size distributions using Method A, just like is done with the CCM method and Method B. As mentioned before, Method A requires samples be taken directly from the aerosol of interest. The developers of Method A have no process or chamber into which a sample obtained off of a filter can be entrained into the equipment.

Therefore, the CCM method and Method B have a great advantage over Method A. If there is a question about an outlying concentration data point, the instruments used in determining concentrations from the CCM method and Method B can retest the TSP filter sample corresponding to the PM concentration in question. Using Method A, no retest would be possible because field conditions could not be recreated exactly.

One excellent advantage to Method A is in obtaining real-time aerodynamic diameter size distributions of spherical particles. Real time data could indicate fluctuations in concentration due to wind directional and speed changes, processing variations, animal behavior and more. However, Method A can report values with significant error for both irregularly shaped particles, having a higher drag force than spherical particles, and highly dense particles when compared to values reported via sedimentation methods (Marshall and Mitchell, 1990, Marshall, Mitchell and Griffiths, 1990).

CAAQES faculty suspect an inaccuracy with Method A's theory behind calculation of the drag force exerted by the fluid on the particle, that leads to calculation of aerodynamic diameter, d_a. The developers of Method A have assumed laminar flow where Reynolds numbers are less than 1.0 – a flow regime known as the Stokes regime. However, in air pollution we very rarely have laminar flow. As Reynolds numbers increase above 1.0, the formation of eddy currents occurs downstream of the particle, causing turbulent flow (Cooper and Alley, 2002). Stokes regime is not valid for turbulent flow; therefore, other calculations than what are used in Method A are required for calculating the drag force leading to the determination of d_a.

A second inaccuracy Method A exhibits, documented by Stein, Beck and Gabrio (2000), is that it detects "large" particles that do not actually exist. The reason for these particles being miss-sized is unknown, but the theory is that either the particles escaped the aerosol flow or they recirculate back into the air stream. The miss-sized particles are known as phantom counts. These particles usually account for a very small percentage of the total number of particles, around 0.07%, but when converted to a mass or volume basis can alter the distribution significantly because of their relatively large volume. One way to determine the existence of these phantom counts, is to collect both time-of-flight and light scattering data simultaneously with Method A. Larger particles will scatter more light than smaller particles. By examining the light scattering intensity versus the particle size as determined by time-of-flight then the miss-sized particles with low light scattering for their particle size can be determined and removed from the distribution. One method of doing this is by using a "mask" that will remove the phantom counts and give a corrected particle size distribution and corresponding statistics. This mask is based on the minimum threshold of light scattered for each size particle.

Method B instrumentation allows for sizing of particles in smaller and larger size ranges compared to the Coulter. Method B equipment sizes particles from 0.010 μm to 10,000 μm, while the Coulter sizes particles from 2 μm to 100 μm. For purposes of agricultural air pollution, however, the particle size range of interest is less than 100 μm, therefore, any advantage Method B equipment may have provided by reporting more particle sizes is superfluous.

In the operation of Method B instrumentation, CAAQES faculty have observed a possible problem in size reporting due to the orientation of irregular shaped particles as they pass through the LASER beam. A sphere will "look" exactly the same to the LASER beam no matter what its orientation while being measured. A cylinder particle will result in different particle sizes depending on what axis it travels on during measurement. The cylinder could pass through with a perceived surface

area of a circle, a rectangle or a rhombus. One would expect that orientation would be random so that measurement would not be skewed towards the length of the short circle or the long rectangle, but give the average particle size. This may not be the case. Discussion about particle orientation is ongoing with the manufacturer of the instrument used in Method B.

A Comparison of CCM to Method A

Chung and Thompson (1989) compared Method A with the Coulter, for several particle shapes. They showed that for regularly shaped latex spheres and silicon carbide test dust, there was very good correlation between the two methods. They showed that for irregularly shaped particles such as fibers and plates, the two methods were not consistent with each other due to inherent inaccuracies for these shapes, such as tumbling of the plates and the orientation of the fiber in the air stream. For roughly spherical hollow particles, the two methods were also not consistent because the CCM is not able to fully detect holes in the particles.

In 2002, our earlier work (Shaw, McClure and Parnell) compared Method A with the Coulter for irregular shaped particles as would be found in agricultural air quality research. A dust chamber design, sampling protocol and filter analysis were developed for this comparison. The data analysis consisted of a comparison of particle size distributions for two Arizona road dusts, one with a mass median diameter of approximately 2.5 μm, and one with a mass median diameter of approximately 10 μm. The mass median diameter (MMD) and geometric standard deviation (GSD) are reported in Table 1. It was found that the Coulter analyses resulted in consistently larger diameters compared to the analyses by Method A with a mask applied to remove the phantom counts.

One reason for the differences in the MMDs is the fact that we are analyzing irregular shaped particles. Both methods are accurate for determining the MMD of a spherical particle. The differences surface when testing is performed on irregular particles. The Coulter instrument used for analysis in the CCM method accounts for the volumes and densities of the particles but not for the shape of the particles. Therefore, it is more likely for the CCM to report higher diameters than actually exist since it cannot account for increased drag forces due to shape irregularities. Method B, on the other hand, has not been proven to perform well for extremely irregular shaped particles as well. The miss-sizing of some particles by Method B results in a tendency to undersize irregular shaped particles. These two inherent actions of each instrument result in opposite trends, thereby giving the differences observed.

A shape factor similar to in equation 3, should be developed to scale and compare data collected by the Coulter and Method A. In order to achieve this comparison with an accurate shape factor, more testing would be required.

A Comparison of CCM to Method B

For comparison of CCM to Method B four test dusts were used to run preliminary analyses to compare particle size distributions. Corn starch, aluminum oxide, fly ash and gin trash were analyzed. The MMD and GSD for each dust and method are displayed in Table 1. Results of the corn starch and aluminum oxide are similar for the two methods. However, the results for fly ash and gin trash are different. Method B resulted in larger diameters and deviations than the Coulter. Differences may be attributed to two possibilities.

First, in running the preliminary analyses using Method A, the delivery mechanism used, entrained a dry sample of each dust. Many materials have a tendency to agglomerate when dry. The fly ash and gin trash materials may have this tendency, therefore resulting in higher diameters compared to the Coulter. The Coulter employs a delivery system using a dispersing liquid that prevents agglomeration. The manufacturer of Method A also has available a delivery mechanism whereby the dust is dispersed in a liquid. Further testing comparing Method A to the Coulter should be performed with the liquid delivery mechanism for both instruments.

Second, gin trash consists of irregular shaped dust particles and long fibrous particles from cotton seed coat fibers. Method B may not be accurately sizing the long fibers. The LASER diffraction due to the fiber being oriented along its length and passing through the LASER beam would result in a much larger diameter than if the fiber were oriented on its end "looking" more like a circle passing through the LASER beam. More information from the manufacturer of Method B will be sought in order to determine the effects of this phenomenon and how comparison to the Coulter should be made.

Conclusions

In conclusion, the CCM method is the method of choice by the Center for Agricultural Air Quality Engineering & Science at this time for determining PM_{10} and $PM_{2.5}$ concentrations. One step of the CCM method is determining particle size distributions based on mass versus diameter of particles using the Coulter instrument.

Analyses of other instruments to provide particle size distributions based on mass versus diameter of particles is of interest to CAAQES. By analysis of Method A, it has been determined that Method A equipment will not be feasible for our agricultural field applications. Subjecting an expensive instrument to forces in field sampling such as unpredictable cattle and acts of nature beyond our control would be too great a risk. Also, samples from a filter can not be used to run subsequent PSD's using Method A. Moreover, inappropriate calculations for laminar flow when turbulent flow dominates in sampling at field operations render Method A useless for our applications. Further testing of Method A is not recommended.

Preliminary analysis of Method B, indicates that Method B equipment should be analyzed further in order to determine its applicability to our needs. Testing of dusts using the liquid dispersing mechanism should be performed. Information about particle orientation as it passes through the LASER beam should be sought from the manufacturer. Further testing of Method B is recommended.

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Table 1. Mass median diameters (MMD) µm and geometric standard deviations (GSD) reported from the Coulter and Method A instrument analyses of two dust types.

	10 µm Road Dust		2.5 µm Road Dust	
Method	**MMD**	**GSD**	**MMD**	**GSD**
CCM	9.6	1.3	2.6	1.4
Method A w/o "mask"	8.8	1.4	3.7	2.6
Method A with "mask"	8.6	1.4	2.6	1.7

Table 2. Mass median diameters (MMD) μm and geometric standard deviations (GSD) reported from the Coulter and Method B instrument analyses of four dust types.

	Corn Starch		Aluminum Oxide		Fly Ash		Gin Trash	
Method	**MMD**	**GSD**	**MMD**	**GSD**	**MMD**	**GSD**	**MMD**	**GSD**
CCM	19	1.3	9	1.4	11	2.0	21	1.9
Method B	17	1.4	10	1.7	20	4.3	56	3.2