ABSTRACT

CAO, ZIHAN. Determination of Particle Size Distribution of Particulate Matter Emitted from a Layer Operation in Southeastern U.S. (Under the direction of Lingjuan Wang.)

This thesis reports a field study on characterizing particle size distribution (PSD) of particulate matter (PM) emitted from a commercial layer operation in the Southeastern U.S. across three seasons from October of 2008 to April of 2009. Six low-volume ($1m^3/h$) total suspended particulate (TSP) samplers were used to collect PM samples in two high-rise layer houses. A laser diffraction particle size analyzer (LS13 320) at North Carolina State University (NCSU), a laser scattering particle size analyzer (LA-300) at University of Illinois at Urbana-Champaign (UIUC) and a Coulter Counter Multisizer 3 (CCM 3) at Texas A&M University (TAMU) or USDA-ARS cotton production & processing unit in Lubbock, TX were used to measure PSD of PM samples collected by the TSP samplers. Particle size distributions measured by these three instruments were compared. Results of the study indicate that TSP concentrations across the three seasons ranged from 888 to 5333 $\mu g/m^3$. Total suspended particulate concentration was affected by season, animal activity, floor, and equivalent air flow rate factor. It was observed that TSP concentration in winter was higher than in spring; concentration on the second floor was higher than that on the first floor; the more active the animals were, the higher the TSP concentration; and the more the fans were on, the lower the concentration. Compared with PM concentration, PSD (characterized by the mass medium diameter, MMD, and geometric standard deviation, GSD) was affected by season, animal activity, floor and equivalent flow rate factor to a lesser extent. Overall MMDs of PM samples collected in fall, winter

and spring, measured by the laser diffraction particle size analyzer were 19.21±1.27 µm, 17.13±0.81µm and 18.44±1.44µm, respectively. Geometric standard deviation (GSD) was relatively constant and not affected by those factors (season, animal activity, floor, and equivalent air flow rate factor). The overall GSD was 2.65±0.08. Significant but constant differences in MMDs and GSDs were detected when comparing PSDs measured by the different instruments. In general, the LA-300 provided the largest MMDs, whereas the CCM 3 gave the smallest MMDs. The LS13 320 provided the largest GSDs, whereas the CCM 3 gave the smallest.

Determination of Particle Size Distribution of Particulate Matter Emitted from a Layer Operation in Southeast U.S.

by
Zihan Cao

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APPROVED BY:

Dr. Lingjuan Wang Committee Chair	Dr. Jeffrey Thompson
Dr. David B. Beasley	Dr. Sanjay B. Shah

BIOGRAPHY

Zihan was born to Guojun Cao and Jieyu Jiang on October 27, 1986 in Jiangxi, China. She obtained her Bachelor's degree in Environmental Engineering from Beijing University of Chemical Technology in 2007. In the fall of 2007, she joined the Air Quality Research Group in the department of Biological and Agricultural Engineering at North Carolina State University. After finishing her Master's degree in BAE, she will pursue another Master's degree in the statistical science department at Cornell University.

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Chapter 1 Introduction

1.1 Background

Particulate matter (PM) consists of small particle of solid or liquid suspended in the air, which was identied as one of six criteria air pollutants in National Ambient Air Quality Standards (NAAQS) by the Environmental Protection Agency (EPA, 1990). In general, PM consists of four major constituents or their combination: sulfates, nitrates, carbon-containing materials including organic carbon (OC) and elemental carbon (EC), and trace elements.

Particulate matter, including feed, feathers, manure and microorganisms, is one of the most important aerial pollutants emitted from animal feeding operations (AFOs) (Harry, 1978; Carpenter, 1986). Particulate matter can be divided into categories by its generation process, primary PM and secondary PM. Primary PM in AFOs is introduced directly into the atmosphere from animal activity, entrainment in ventilation air, etc. (Kondratyev et al., 2006). Secondary PM is formed in the atmosphere by chemical reactions of primary gaseous components.

Particle size is one of the most important parameters for characterizing atmospheric particles in terms of their impacts and fate. Size of particles is directly related to their role in causing health problems since it affects their deposition rate, and thus determing the location of their deposition within the respiratory tract (Mercer, 1978; Janni et al., 1984).

Size distribution of particles is considered as the most important physical parameter guiding their performance (Hinds, 1998).

Environmental protection agency (EPA) regulates PM by size and is concerned about particles smaller than or equal to 10 µm in aerodynamic equivalent diameter (AED), because this fraction of PM can pass through the throat and nose, enter the lungs, and may result in respiratory diseases. These particles are referred as PM_{10}. Particles smaller than or equal to 2.5 µm in AED, referred as $PM_{2.5}$, tend to penetrate deeper into the lungs. It has been reported by Pope et al. (2002) that $PM_{2.5}$ leads to high plaque deposits in arteries, causing vascular inflammation and atherosclerosis.

Furthermore, EPA groups particle pollution into two categories: a) coarse particles with size between 2.5µm and 10µm, such as those found near roadways and dusty industries including power plants and petroleum industries and b) fine particles with size equaling 2.5µm or smaller, such as those found in smoke and haze.

The negative effects of PM on human beings have been demonstrated by a number of incidents, such as the great smog in London in 1952 and smog in Donora in 1948 (EPA, 1996). Many epidemiologic studies show that there is a significant relationship between ambient PM levels and a variety of human health issues, including mortality, eye, and respiratory problems (Edwards et al., 1997; Smyth et al., 2003). The relationship between inhaled PM and respiratory system diseases have been widely studied. The effects of inhaling PM on human health include asthma, lung cancer, cardiovascular issues and premature death. In addition, biological particles, referred as bio-aerosols, may lead to many infectious diseases, such as foot and mouth disease for animals (Wathes, 1995). The large proportion of biological and organic compounds, such as most bacteria, fungal spores, viruses and airborne pollens of PM in animal indoor environment result in more serious health effects than general PM from ambient sources (Tan and Zhang, 2004).

Apart from its impact on the health of human beings, PM can also cause deterioration of the animal indoor environments. When combined with absorbed chemicals and high levels of humidity, PM in animal indoor environments may result in rough surfaces and other building materials, promoting degradation of building structures and equipment installed within buildings (Davis, 1991; Zavaleta, 1976; Rosentrater, 2003).

The adverse effects of PM in animal indoor environments depend on the quantity, composition and physical properties of particles, while the physical properties and fate of PM are strongly related to particle size distribution (PSD). Understanding of PSD is essential in PM related research work.

In the literature, a number of research projects have been conducted on particle size characterization of PM emitted from various AFOs (Capareda et al. 2004, 2005; Sweeten et al. 1998; Heber et al. 1988; Redwine and Lacey, 2001; Lacey et al. 2003). These studies reported on the physical characteristics of PM emitted from swine buildings, feed yards, and broiler houses. Details about these studies will be described in the next chapter. Gaps remain concerning physical characteristics of PM emitted from AFOs, specifically from layer operations.

Validation and comparison of different techniques on characterizing PSD will be of great significance for the assessment of agricultural air quality. It is therefore the aim of the present study to compare different techniques to characterize PSD of PM emitted from layer operations.

1.2 Research objectives

The specific research objectives of this study included:

1. Investigation of factors that might cause variations in PM concentration and PSD in tunnel ventilated high-rise layer houses. These factors include animal activity, ventilation rate, special and seasonal variation.
2. Investigation of different techniques for PSD determination. Differences of PSDs measured by three instruments will be quantified and possible causes of the differences will be addressed.

Chapter 2 Literature Review

2.1 Significance of particle size distribution

Particle size is one of the most significant parameters in determining the physical and chemical properties of atmospheric particles. In addition, the atmospheric deposition rates of particles and light scattering are strongly related to particle size. The scattering pattern of particles is directly related to particle size. Large particles scatter at small angles, while small particles scatter at large particles. Particle size distribution, therefore, has a strong influence on atmospheric visibility (EPA, 1996). Particle size is an important factor that influences how particles deposit in the respiratory tract and affect human health (EPA, 2009). Environmental protection agency (EPA) groups particle pollution into two categories by size: inhalable coarse particles, which are larger than 2.5 μm and smaller than 10 μm in AED, and fine particles, which are 2.5 μm in AED and smaller. Inhalable coarse particles are deposited almost exclusively in the nose and throat, whereas, fine particles generally are able to penetrate deep into the lung. Fine particles are also the major cause of reduced visibility in parts of the United States (EPA, 2009).

2.2 Definition of PSD

Monodisperse aerosols have particles that are all the same size and can be produced in the laboratory for use as test aerosols. Polydisperse aerosols have particles with a wide range of particle sizes, and statistical measures should be used to characterize their particle size. If the particles are mono-dispersed aerosols, the size of these particles can be defined by the particle diameter. However, in the real world, most particles are poly-dispersed aerosols, and the particle dimensions may range from nanometers to millimeters. Particle size of various dimensions are usually characterized by certain statistical distributions or models.

Mean, median and geometric mean are statistical parameters widely used to define a location of the distribution (Hinds, 1999). Mass distribution provides the fraction of the mass of particles in any size range (Hinds, 1999). Mass median diameter (MMD) is the diameter for which half the total mass of particles is larger and half is smaller. Generally, MMD is regarded as one of two statistical parameters for the identification of PSDs in mass. Particle size distributions of PM rarely follow a normal distribution. Instead, a lognormal size distribution is applied to describe PSDs. The density function of the PSD can be described as:

$$df = \frac{1}{\sqrt{2\pi}\ln\sigma_g}\exp\left(-\frac{\left(\ln d_p - \ln\overline{d_p}\right)^2}{2\left(\ln\sigma_g\right)^2}\right)d\ln d_p \qquad (2.1)$$

Where:

d_p = the particle diameter;

$\overline{d_p}$ = the MMD;

σ_g = the geometric standard deviation (GSD).

Equation (2.1) can be rearranged as follow:

$$f\left(d_p, MMD, GSD\right) = \frac{1}{d_p \ln GSD\sqrt{2\pi}} \exp\left[\frac{-\left(\ln d_p - \ln MMD\right)^2}{2\left(\ln GSD\right)^2}\right] \qquad (2.2)$$

As shown in Equation (2.1) and (2.2), GSD is another statistical parameter used to define aerosol PSDs, which follows the lognormal distribution. Geometric standard deviation is mathematically defined by the following equation:

$$GSD = \sigma_g = \left(\frac{d_{84\%}}{d_{16\%}}\right)^{1/2} \qquad (2.3)$$

Where:

d_{84} = the diameter such that particles constituting 84% of the total mass of particles are smaller than this size;

d_{16} = the diameter such that particles constituting 16% of the total mass of particles are smaller than this size.

2.3 Particle shape factor

The most critical problem existing in most available particle size analyzers is that the operation of the analyzers is built upon the assumption of spherical particles (Xu, 2005), such that the distribution obtained from the analyzer is in equivalent spherical distribution (ESD). However, particles can have many different shapes. Some particles have regular geometric shapes while other particles have irregular shapes (Hinds, 1998).

In order to describe particle distribution accurately, a correction factor, called the dynamic shape factor (χ) is used to address the effect of particle shape on particle motion

(Hinds, 1998). By definition, the shape factor (χ) is the ratio of the actual resistance force of the non-spherical particle to the resistance force of a sphere having the same volume and velocity as the non-spherical particle (Hinds, 1998), as shown in Equation (2.4).

$$\chi = \frac{F_D}{3\pi\eta V d_e} \tag{2.4}$$

Where:

F_D = actual resistance force of the non-spherical particle (N);

d_e = equivalent spherical diameter (m) (described later);

V = particle velocity (m/s);

η = dynamic viscosity (Pa·S).

For a spherical particle, the shape factor equals 1. For non-spherical particles, image information can be obtained from microscopy, and the shape factor can only be approximately estimated because of the variety of particle shapes and the complicated calculations. Shape factors of some typical shapes were obtained as reference for estimation. After obtaining the shape information of the particle, the shape factor of the particle can be assumed referring to available shape factors of particles with similar shapes. Generally, the shape factor for non-spherical particles is greater than 1 (Hinds, 1998).

Equation (2.5) and (2.6) relates particle diameter with shape factor, where d_e is equivalent spherical diameter. Stokes' law and terminal settling velocity for irregular particles are:

$$F_D = 3\pi\eta V d_e \chi \tag{2.5}$$

$$V_{TS} = \frac{\rho_p d_e^2 g}{18\eta\chi} \tag{2.6}$$

Where:

d_e = equivalent spherical diameter (m) (described later);

χ = particle shape factor;

ρ_p = particle density (kg/m^3) (described later);

η = dynamic viscosity (Pa·S).

There are two diameters that need to be clearly distinguished, d_e and d_a. d_e is also known as equivalent spherical diameter (ESD). By definition, ESD is the diameter of the sphere having the same density and settling velocity as the particle in question. d_a stands for AED. By definition, AED is the diameter of the sphere having a density of 1000 kg/m^3 and the same settling velocity as the particle in question. An irregular particle and its aerodynamic sphere are shown in Figure 2.1.

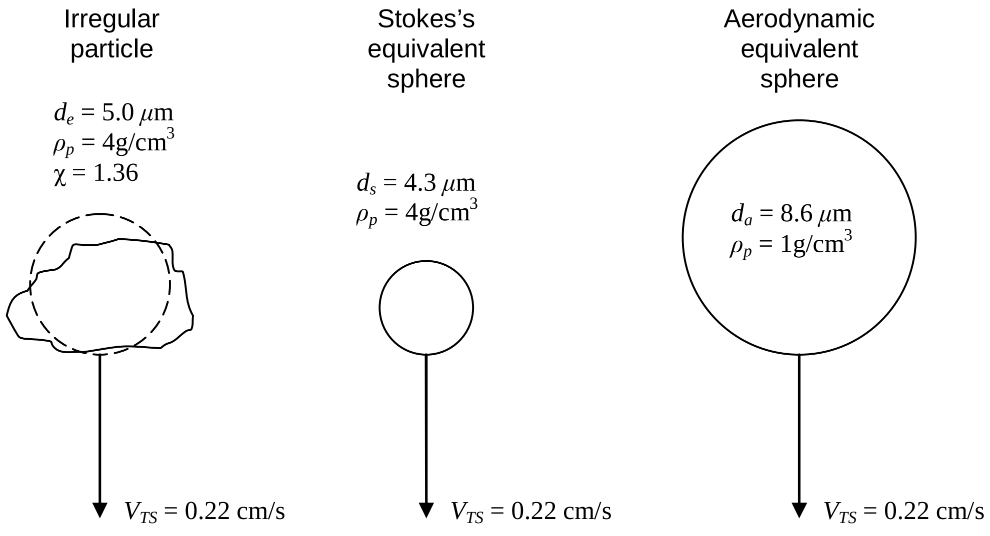

Figure 2.1 An irregular particle and its equivalent spheres (Hinds, 1998)

There are two densities that need to be fully understood, ρ_p and ρ_0. In equations above, ρ_p is particle density, and ρ_0 is the standard particle density at 1000 kg/m^3.

Incorporating AED into Equation (2.6), terminal setting velocity can be presented as follows:

$$V_{TS} = \frac{\rho_p d_e^2 g}{18\eta\chi} = \frac{\rho_0 d_a^2 g}{18\eta} \tag{2.7}$$

Rearranging Equation (2.7) gives:

$$d_a = d_e\left(\frac{\rho_p}{\rho_0\chi}\right)^{\frac{1}{2}} \tag{2.8}$$

Thus, AED can be calculated based upon measurement of ESD, ρp and χ, using following equation:

$$AED = ESD \times \sqrt{\frac{\rho_p}{\rho_0 \chi}} \tag{2.9}$$

Most particle size analyzers, such as the laser diffraction particle size analyzer and coulter counter particle size analyzer, provide PSD in the form of particle volume versus ESD. However, U.S.EPA regulates particle size in terms of AED. Commonly, PM_{10} and $PM_{2.5}$ are referred to particles less than or equal to 10 µm in AED and less than or equal to 2.5 µm in AED, respectively.

2.4 Particle density

Particle density refers to the mass per unit volume of the particle itself, not of the aerosol. Aerosol density, also called mass concentration, refers to the mass of particles in a unit volume of air (Hinds, 1998).

As mentioned in the previous section, there are two types of particle densities, ρ_p and ρ_0. Particle density is always expressed in the unit of kg/m^3 or g/cm^3. ρ_0, referred as standard particle density, equals water density (1000 kg/m^3). As shown in Equation (2.9), particle density needs to be measured in order to convert ESD to AED.

The measurement of particle density includes measurement of particle's mass and volume (Masuda et al., 2006). Mass of particle can be measured by different types of mass balances, usually an electronic balance. Volume of particle can be obtained by pycnometer method, the most representative method (Masuda et al., 2006).

2.5 Technologies for PSD determination

Several techniques have been developed and applied for PSD measurement and determination. These techniques include cascade impaction, optical particle counting, dynamic image analysis, laser diffraction and electrical sensing zone. These techniques are discussed below in detail.

2.5.1 Cascade impactor

Cascade impactors consist of a number of impaction stages connected in series with increasingly smaller cut-off diameters. The cascade impactor is designed to separate PM into different particle size ranges based on different cut-off sizes at different separation stages; thus PSD can be calculated in terms of AED based upon the mass fraction of a given size range. The cascade impactor has the capability of providing onsite PSD determination. Figure 2.2 illustrates the fundamental principle of the cascade impaction system. In this system, an air stream is pulled through the impactor stages. Each stage is assumed to capture all particles reaching it that are larger than its cutoff size (Hinds, 1998).

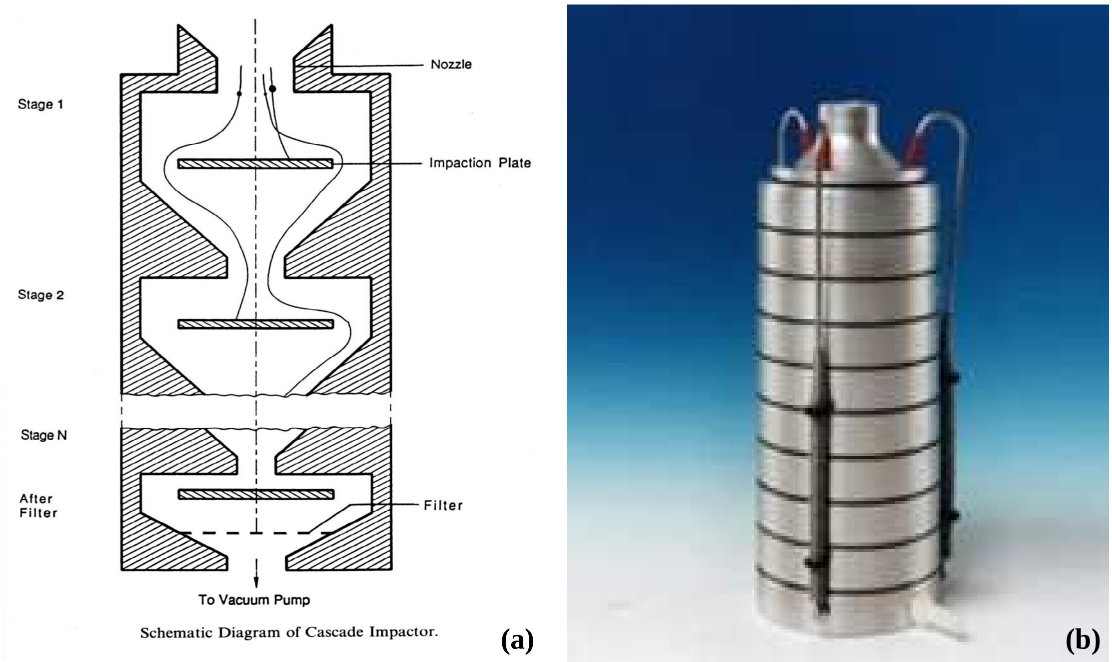

Figure 2.2 Cascade impactor

(a) Schematic diagram. (Hinds, 1998) (b) Eight-stage non-viable cascade impactor (NSE, Roswell, GA)

The cascade impactors have been widely used to measure PSD by mass. In general application, the cascade impactor is located and operated in a testing environment for a given time. The mass of the particles collected on the impaction plates and the filter (Figure 2.2(a)) are determined by weighing the plates and filter before and after sampling. Thus, the mass fractions of particles in different cut-off size ranges can be determined, thus allowing the size distribution of particles.

There are many applications of the cascade impactors in recent research work. Research work on mass size distribution and chemical components has been done at the Pittsburgh supersite during the Pittsburgh Air Quality Study (PAQS) using Micro-orifice uniform deposit impactors (MOUDIs) to collect aerosol samples of PM in different size ranges (Cabada et al., 2004). One MOUDI with aluminum foil was used for analysis of organic materials while two other MOUDIs with Teflon filters were used for determination of inorganic compounds and PM mass. Based on the results, Cabada et al. (2004) concluded that $PM_{0.056}$, $PM_{0.10}$, $PM_{0.18}$, $PM_{0.32}$, $PM_{0.56}$, $PM_{1.0}$, $PM_{1.8}$ and $PM_{2.5}$ were measurable by MOUDIs and the measured aerosol concentrations were higher during the summer than that in winter.

Hughes et al. (1998) conducted measurements on aerosol number concentrations and chemically speciated size distributions using MOUDIs in southern (Pasadena) California in winter. Chung et al. (2001) measured particle number concentrations and chemically speciated size distributions using MOUDIs in central (Bakersfield) California in winter. Both studies concluded that there was no strong correlation between ultrafine number and ultrafine mass concentrations. Carbonaceous material was the largest fraction of the ultrafine mass. Contributions of sulfate, nitrate, ammonium and trace compounds were reported in both studies (Hughes et al., 1998 and Chung et al., 2001).

Watson et al. (1998, 2008) estimated aerosol light scattering based on particle size and chemical measurements using MOUDI at the Fresno supersite as part of the California Regional PM_{10}/$PM_{2.5}$ Air Quality Study. They concluded that aerosol light scattering was underestimated by 25% using chemical size distributions measured with the MOUDI.

Since the 1960s, cascade impactor analysis has been widely accepted as the standard technique for quality control of finished products in pharmaceutical industry (Ziegler and

Wachtel, 2005), and size distribution characterization of aerosol generated by medical aerosol generators (Ranz and Wong, 1952a, b; Hinds, 1998). Many different types of impactors have also been introduced for medical aerosols in the past decades. Marple et al. (2000) developed a new generation impactor and Graseby-Anderson (1985) developed a more complex impactor, the Anderson Mark II, which has more collection stages.

The practical drawbacks and limitations of cascade impactor analysis include: a) this technique is laborious and time consuming; b) classification can only include a small number of size classes; c) inertial removal causes inter-stage loss due to sharp bends in the inter-stage flow path; d) incidences of bounce off from the plates are unavoidable; and e) the formation of a conical mound due to particles collecting on the plate may change flow geometry and cut-point.

2.5.2 Laser diffraction

A laser diffraction particle size analyzer measures size distribution of particles by applying the principles of light scattering. According to Xu (2000), laser diffraction may provide more reliable PSD measurements on account of ease of use and broad size ranges, from nanometers to millimeters. De Boer et al. (2002) and Marriott et al. (2006) reported that the laser diffraction technique was a fast and reliable alternative to the cascade impactor for PSD analysis. It was also reported that compared with cascade impactor, laser diffraction had excellent possibilities for automated recording of data and testing conditions (Cabada et al. 2004).

The laser diffraction technique relies upon the fact that particles will act as scattering centers when exposed to beams of light; thus resultant patterns of scattered light can be produced and analyzed mathematically, with the assumption of spherical particles (ISO 13320-2, 1999; Xu and Guida, 2003). Based on the Mie Theory, the scattering pattern of particles is directly related to particle size. As shown in Figure 2.3, larger particles scatter at smaller angles while smaller particles scatter at larger angles.

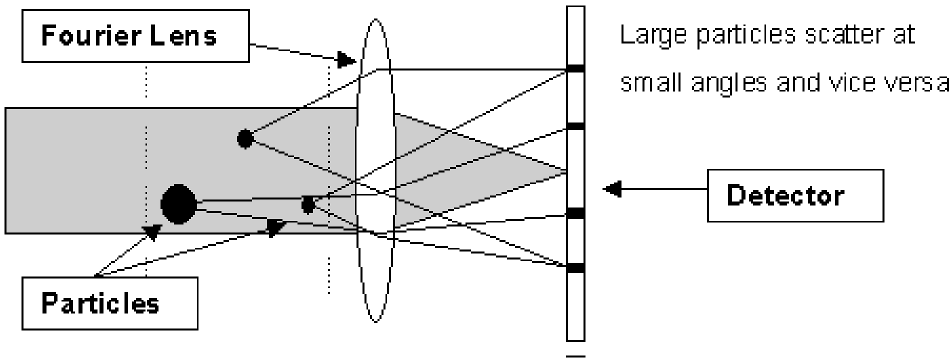

Figure 2.3 Fourier optics (Beckman Coulter, Miami, FL)

In the past three decades, laser diffraction has become a popular and important physical technique for PSD characterization. Clark (1995) and Moren (1981) applied laser diffraction to examine aerosols from nebulizers and pressurized metered dose inhalers, respectively. Little application was found in examining dry powder aerosols in the 1980s (Olsson et al., 1988). When both cascade impactor and laser diffraction techniques were used to test nebulizers (Ziegler and Wachtel, 2005) and pressurized metered dose inhalers (Smyth and Hickey, 2003), the results of aerosol size distributions measured by both of these two techniques correlated with each other very well. However, so far, there has been no study reporting a correlation between the PSD of dry powder aerosol formulations measured by laser diffraction and that obtained by the cascade impactor (Marriott et al., 2006). It was concluded that the laser diffraction technique provided more realistic size distributions for nebulized aqueous drug solutions than cascade impactor technique (Ho et al., 1986). Ranucci (1992) reported that laser diffraction analysis could be an essential technique for analyzing aerosols generated by nebulizers and metered dose inhalers.

Comparisons between the laser diffraction and other PSD technologies have been conducted by several researchers (Xu and Guida, 2003; De Boer et al., 2002; Marriott et al., 2006). Samples of various shapes were characterized using three different techniques (laser diffraction, electrical sensing zone method, and dynamic image analysis) by Xu and Guida (2003). Xu and Guida (2003) reported that laser diffraction provided much larger mean sizes and broader distributions as the shape of the particles deviate more and more from a sphere compared with the other two technologies. It was reported that mean particle size obtained from laser diffraction was much larger than those from sedimentation and ESZ (Xu and Guida, 2003).

An international collaborative study involving 24 laboratories using 21 laser diffraction instruments was performed in 1994 by Merkus et al. (1995). Sedimentation and electrical sensing zone method (ESZ) were used as reference methods in comparing those results (Merkus et al., 1995).

Some practical drawbacks and operational limitations of the laser diffraction technique still exist. In this technique, the difference between the model and the real shapes of the particles is always ignored, with the assumption that all particles are spherical. However, rarely are particles in real world perfectly spherical, which can lead to doubtful results when using the spherical model on real particles. Also, with the laser diffraction technique, AED cannot be measured directly, which can only be obtained by converting ESD to AED with known particle density and shape factor (Equation(2.9)). In addition, size distribution of particles with low concentration may not be measureable by the instrument.

Barreiros et al., (1996) compared the PSD results obtained by a laser diffraction analyzer (Malvern 2600C) and an electric sensing analyzer (Coulter Multisizer II). They discovered that for irregular particles, the Malvern tended to provide larger MMD and broaden PSD. As a result, they reported that particle shape strongly affected performance of the laser diffraction analyzer produced by Malvern Instrument Ltd. This kind of systematic bias is obvious when the results obtained from laser diffraction analyzers were compared with those from other technologies.

2.5.3 Electrical sensing zone (ESZ)

Besides the laser diffraction technique, the ESZ, commonly known as the Coulter Principle, has been widely accepted in the field of medical technology since the 1950s. This technique was originally developed to count blood cells, and it can be also used to count and size any particulate material which can suspend in an electrolyte solution (Xu, 2000). In the ESZ technique, the particles suspended in an electrolyte solution are forced to pass through a small aperture where an electric field is applied. The solution's conductance changes as the particles pass through the aperture. The change in conductance is a function of particle size. When particles pass through the aperture, the particles' individual volumes are directly measured (Xu and Guida, 2003). The advantages of this method are that it measures a single particle's volume and provides high resolution and reproducibility for individual particle assessment. A coulter counter multisizer is one type of electrical particle sizer manufactured by Beckman Coulter, Inc based on the principle of ESZ.

However, this technique has some limitations: a) Particles that can be analyzed are restricted to those that can be dispersed in an electrolyte solution and still retain their original integrity (Lines, 1996); and b) the coulter counter tends to increase the fraction of smaller particles (Xu, 2003).

Xu (2003) compared the ESZ method with other two techniques (laser diffraction and dynamic image analysis) in sizing small particles. He concluded that ESZ produced compatible results with dynamic image analysis (DIA), which are much less affected by particle shape. However, particle size results of non-spherical particles from different instruments are often less consistent with different degrees of deviation when compared with each other. PSD of non-spherical particles is often a function of sample particle shape.

2.5.4 Dynamic image analysis (DIA)

Dynamic image analysis (DIA) is one type of measurement technique providing both size and shape information. Dynamic image analysis (DIA) relies upon the existing microscopic image analysis technique, and makes use of the technology in microelectronics (Xu and Guida, 2003). However, the results depend on the particle's orientation while the images are analyzed independently without any consideration of particle shape. Xu and Guida (2003) concluded that both ESZ and DIA are more appropriate in obtaining equivalent spherical diameters for non-spherical particles. For regularly shaped particles, difference in particle sizes obtained from ESZ and DIA was further investigated.

2.5.5 Summary

In the past decade, various techniques have been developed to characterize PSD for better understanding of particle physical properties. Each technique has its own application strengths and limitations.

The cascade impactor, is the technique recommended by the EPA for characterizing PSD, has been widely used in many fields. It has been used most for characterization of aerosol clouds generated by medical aerosol generators including dry powder inhalers and metered dose inhalers, product development and quality control of the finished products. In addition, there are also some applications of cascade impactor conducted in ambient air quality studies. However, few of those studies were in the area of agriculture air quality.

Laser diffraction, combining modern techniques in lasers, computers, statistics, and automation, has become one of the most popular means for characterizing PM in the pharmaceutical and food industries. The laser diffraction analysis system used in aerosol assessment during the production of medical inhalers and quality control of pharmaceutics and foods, has been evaluated in many studies. Nevertheless, laser diffraction research has been limited in air quality studies, both ambient air quality and animal indoor environment.

Other techniques for PSD analysis include ESZ and DIA, each of which has their own features and are still under development. These techniques have been applied in medical and biological fields. In addition, there are many applications of ESZ in the air quality area.

2.6 Particulate matter and PSD associated with agricultural emissions

Agricultural air emission has become an important topic in air quality studies. As AFOs have become larger, air emissions from AFOs have attracted more and more attention. This may lead to regulations. Animals such as broiler, layer, and swine are raised in confinement buildings. The PM within animal production buildings can cause respiratory system distress and eye irritation in animals and workers, as well as neighbors. In addition, PM can degrade buildings and equipment (Zhang and Tan, 2004). Because of adverse effects of PM in agricultural emission, there is high demand for knowledge of AFO PM characteristics.

As an important characteristic of PM, PSD of PM emitted from agricultural operations has been studied by different researchers. Capareda et al. (2005) conducted PM measurement of cotton gin dust in south Texas. Low volume TSP samplers were used to collect PM. Samplers were placed in the downwind area. Filters in the samplers were changed every 6 h, and samplers collected ambient PM continuously for 24h/d except during the changing of the filters. A total of ten tests were performed and a total of 20 TSP filters were obtained (10 filters per TSP sampler). A coulter counter multisizer 3 was used in that study to measure MMDs and GSDs. Average MMD and GSD of PM collected by the TSP samplers were measured as 11.8 µm and 2.03 µm, respectively. It was also concluded in this study that the lognormal distribution described well the PSDs of cotton gin PM.

Particle size distributions of PM emitted from cattle feed yard and dairy operations were evaluated using coulter counter multisizer by Capareda et al. (2004). They concluded that average MMD and GSD of feed yard PM were 17 µm and 2.8 µm, respectively. Average MMD and GSD of dairy PM were 15 µm and 2.5 µm, respectively (Capareda et al., 2004).

Lacey et al. (2003) conducted a study to characterize PM and ammonia emission factors associated with broiler operation. This study was carried out in four commercial tunnel-ventilated broiler houses. High volume TSP samplers were used to collect PM in the broiler houses, and a coulter counter multisizer was used to analyze PSD of the collected PM samples. The average MMD and GSD of broiler PM were 26.1 µm and 1.6 µm, respectively. Compared with PM from cattle feed yards and swine operations, MMD from broiler houses was larger.

Similarly, Redwine and Lacey (2001) used high volume TSP samplers to collect PM samples in tunnel ventilated broiler houses in summer in Texas. The coulter counter multisizer was used to conduct measurements of PSD. Mass median diameters (MMDs) of broiler PM obtained in this study were between 25.0 and 27.4 µm. It was observed that MMD increased with bird age.

Sweeten et al. (1998) evaluated in PSD of cattle feedlot PM emission. High volume TSP samplers were used to conduct TSP sampling. The average MMD and GSD of PM collected by the high volume TSP samplers were 9.54 µm and 2.8 µm, respectively.

Heber et al. (1988) conducted a study to evaluate the environment inside 11 swine finishing buildings. Particle size distribution, particle identification, and particle morphology were analyzed after PM samples were collected eight times over 9-month period from each swine building (Heber et al, 1988). However, little quantitative data on the morphology, composition, and origin of aerosols were obtained from this study.

As further investigation, Heber et al. (1999) conducted a study on the PM characteristics. Particle size distributions of PM in swine buildings were measured with coulter counter multisizer. In this study, Heber et al. (1999) assumed spherical particles and constant density. Average MMD and GSD obtained from this study were 18.9 µm and 2.58 µm, respectively. It was concluded that size of PM followed a lognormal distribution (Heber et al., 1999).

Chapter 3 Materials and Methods

This chapter reports experimental designs for preliminary studies and studies for the objectives. Optimal parameters to operate the laser diffraction particle size analyzer (LS13 320) were investigated and determined. Liquid medium served as sample solution and appropriate sonication time for sample extraction were chosen through the preliminary studies. Total suspended particulate samples were taken on the research farm for concentration and PSD analyses by different particle size analyzers. Factors that might cause variations in TSP concentration and PSD were investigated. PM_{10} and $PM_{2.5}$ mass fractions obtained from two methods were compared. In this chapter and the chapter 4, LS13 230 represents the laser diffraction particle size analyzer; LA-300 represents the laser scattering particle size analyzer; CCM 3 represents Coulter Counter multisizer 3; and LS230 represents another laser diffraction particle size analyzer.

3.1 Preliminary investigations

The purpose of this preliminary study was to determine optimal operational parameters to run the LS13 320. These included liquid medium selection, pump speed, length of running time and sonication time determination.

3.1.1 Determination of optimal parameters to operate the LS13 320

3.1.1.1 Laser diffraction particle size analyzer with Universal Liquid Module (ULM)

Introduction (LS 13 320 Manual, Beckman Coulter Inc., 2003)

The multi-wave length laser diffraction particle size analyzer (Make: Beckman Coulter, Miami, FL; Model: LS13 320) is one type of laser diffraction particle size analyzer used in this study to measure size distribution of particles emitted from a layer farm. This analyzer consists of an optical bench and a sample handling module, ULM.

The optical bench of this laser diffraction particle size analyzer (LS13 320) consists of a diffraction light source, a sample cell, a Fourier lens and a series of photo detectors. The diagram of the optical system is shown in Figure 3.1. There are two light sources in the optical system. One is polarization intensity differential scattering (PIDS), which can provide information about particles smaller than 0.4 μm. The other one is diffraction light source, which provides information about particles larger than 0.4 μm and smaller than 2000 μm.

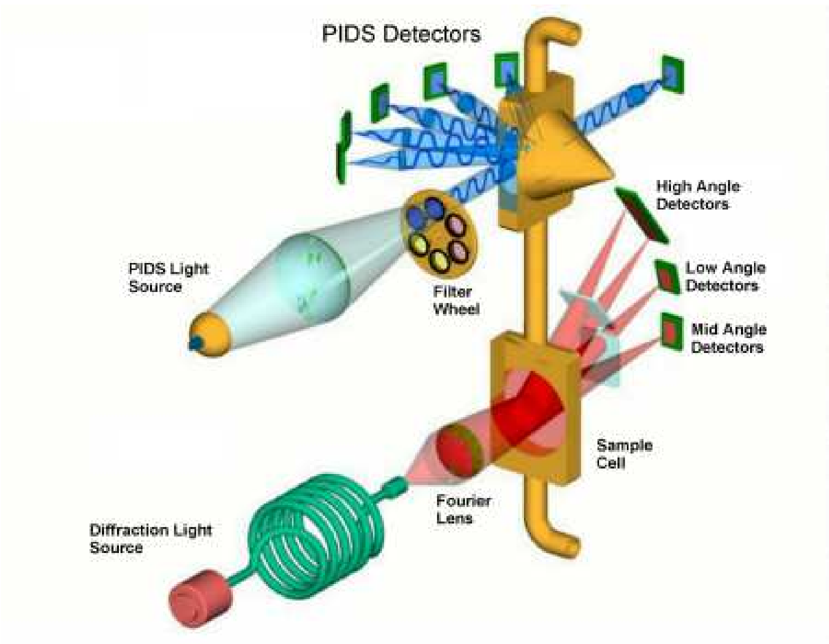

Figure 3.1 Optical system of the LS13 320 (Beckman Coulter, Miami, FL)

The sample handling module is used to deliver particles in the sample to the sensing zone, which is composed of a sample cell and a delivery system. The ULM (Figure 3.2) is intended for use with the Optical Bench. The analyzer is capable of detecting samples in the size range of 0.04µm to 2000µm. It measures the entire sample introduced to the instrument by re-circulating the sample, the amount of which needed depends on concentration of particles.

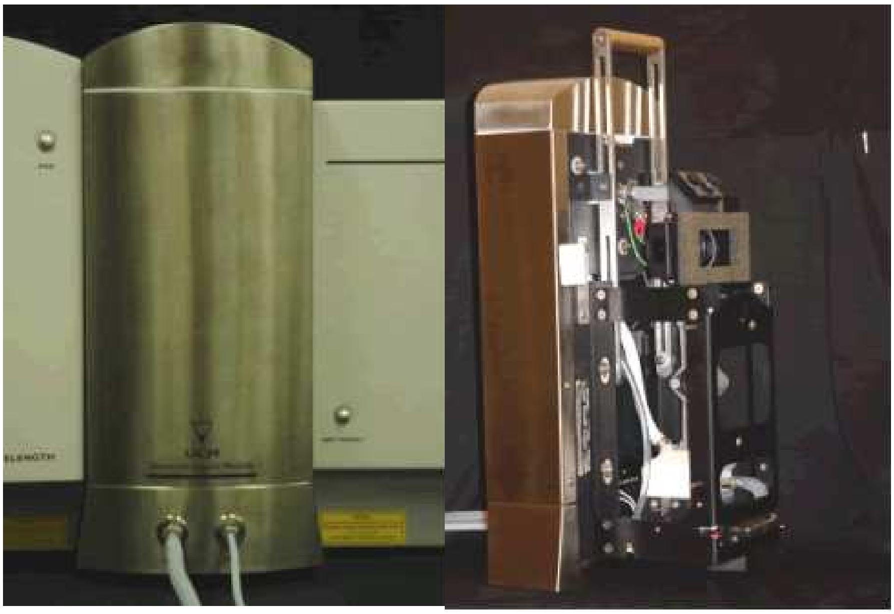

Figure 3.2 The Universal liquid module (Beckman Coulter, Miami, FL)

The laser diffraction particle size analyzer can measure PSD of particles suspended either in a liquid or in dry powder form. For wet-based type, it is only suitable for insoluble particles and the solvent is of critical importance. For dry powder form, there is a high demand for the complete dispersion of particles. In this study, with the application of the ULM, PSD of particles will be measured in a liquid form. Because this instrument could not do onsite measurement, PM samples collected on filters will be used for wet-basis analysis. This instrument calls for a high concentration of particles to analyze. The loading of the sample needs to meet its high mass demand of particles.

3.1.1.2 Experimental design for the determination of optimal parameters for the LS13 320

The objective of this preliminary study was to investigate the effects of different parameter settings on the LS13 320 for PSD measurement of raw dust from broiler houses. Parameters for the particle size analyzer operation include pump speed and length of running time. Instrument operation was conducted following the operation procedure described in Appendix A, and more details can be found in the instrument manual (Beckman Coulter, 2003).

<u>Pump speed</u>

A circulation pump included in the delivery system of the ULM can help better disperse and circulate the particles repeatedly during the process of particle delivery. Speed of the pump may or may not have significant effect on PSD. It was assumed that when measuring large, dense particles, low pump speeds would not be capable of circulating these large particles in the diluent while that high pump speeds may break down large particles. So, it is essential to investigate pump speed impact on measurement results.

Thirty percent (30%), 45% and 60% of full pump speed were selected as three representative pump speeds for the investigation. In order to prevent the formation of air bubbles, it was recommended for samples which required surfactant that the pump speed can be kept as low as 30% (Beckman Coulter, 2003).

Run length

The length of instrument running time may be a factor that affects particle dispersion. It can be set as low as 10s. The run length effect on PSD also needs to be investigated, so that its possible impact on measurement results could be quantified.

Four representative types of instrument running time (30s, 60s, 90s and 120s), where 60s is the time for analysis recommended by the manufacturer (Beckman Coulter Inc., Miami, FL).

For the tests of pump speed and run length, raw dust from a broiler house was used as testing samples. Approximately 25 mg raw dust was introduced and dispersed into 125 mL water. Particulate matter samples in water were then introduced into the analyzer for PSD measurements.

Samples were analyzed by the analyzer when pump speed was set as 30%, 45% and 60%, respectively and running time was set as 30s, 60s, 90s and 120s, respectively. Thus MMDs and GSDs of PM samples under different settings were measured. A 3×4 factorial design was built in terms of these MMDs and GSDs. Two-way analysis of variance (ANOVA) was performed on the data to test whether these two factors had significant impacts on MMDs and GSDs at the 0.05 level of significance (α). For each treatment, there was only one run.

3.1.2 Liquid medium identification

Low concentration dispersants promote dispersion and aid in stabilization of the suspension. When selecting a suitable liquid medium as a diluent or dispersant for aerosol samples, the characteristics and properties of the liquid medium have to be taken into consideration. Aerosol properties such as solubility, reactivity, suspendibility and end use govern the choice of the medium.

Wang et al. (2008) attempted to characterize PSD of PM emitted from a broiler farm. Water was applied as the liquid medium in the LS13 320 for analysis in this study. It was observed that the MMDs and GSDs were significantly greater than those reported in the literature. It was suspected that the water used as the dispersant by the LS13 320 for PSD analysis did not disperse the particles well. Hence, water may not be a good dispersant or diluent.

Particulate matter emitted from AFOs is a complex and highly variable mixture of materials including soil, road dust, feed particles, dander, hair, manure, and bio-aerosols (Auvermann et al., 2006). In general, chemical compositions of PM emitted from AFOs may include carbonaceous materials, sulfates, and nitrates, etc., some of which are soluble in water. As a result, the PSD obtained by the instrument may not be the true PSD for a given PM sample, if water was used as the liquid media.

Based on the finding of the study conducted by Wang et al. (2008) using water as the liquid medium in the LS13 320, non-aqueous media was considered as the dispersant or diluent in this study. Non-aqueous diluents that can be used in the ULM as listed in the LS 13 320 Manual, include ethanol, isopropanol, acetone, mineral oils, hexane, and methanol.

Isopropanol, as a non-aqueous medium, was recommended by the instrument vender as a diluent over water when determining PSD for a sample of fertilizer, which is soluble in water (Beckman Coulter, 2003). However, isopropanol, also called isopropyl alcohol, is a flammable, colorless liquid with an alcohol-like odor. When introduced to heat, or flame, isopropanol becomes unstable. A flammability rating of 3 has been set for isopropanol by the National Fire Protection Association, which denotes a severe fire hazard (Occupational Safety & Health Administration, 2008). Occupational Safety & Health Administration (OSHA) set the permissible exposure limit (PEL) for isopropanol at 400 ppm based on an 8-h time-weighted average concentration. In addition, isopropanol at 400 ppm irritates the eyes, nose and throat even for exposures as short as 3 minutes (Hathaway et al. 1991).

Methanol, known as methyl alcohol, is the simplest alcohol. Methanol is extremely toxic and irritates the skin, eyes, nose and throat (Occupational Safety & Health Administration, 2009).

Ethanol, known as ethyl alcohol, is a volatile, colorless liquid with a strong odor. Ethanol is widely used as a solvent, and it is miscible with water and many organic solvents. Ethanol can be used as an antidote for poisoning by other toxic alcohols.

Compared with methanol and ethanol, isopropanol is less water-compatible. However, similar to methanol, isopropanol is a more toxic and dangerous chemical compared with ethanol. A ventilation system is recommended when handling isopropanol and methanol.

Based on the information collected on methanol, ethanol, and isopropanol; ethanol and isopropanol were chosen as the solvents for further testing to identify the most appropriate liquid medium. Using the same operating procedure, the same type and equal amounts (10 mg each) of samples were put into equal volumes of ethanol and isopropanol. In these tests, raw dust from the broiler houses was used as the PM sample. Approximately two 10 mg raw dust samples were put into two clean 100 mL beakers, respectively. 30 mL ethanol and 30 mL isopropanol were poured into these two beakers, respectively. Measurements of MMDs and GSDs of PM samples were conducted based on the different liquid media (ethanol and isopropanol). A single factor ANOVA test was performed on the data to test whether different liquid media produced significant differences in measured MMDs and GSDs at the 0.05 level of significance (α). There were four or five replicates for each treatment.

3.1.3 Determination of optimal sonication time for PM sample extraction

For PSD measurement of filter-based PM sample, before sample particles are introduced into the LS 13 320, it is necessary to extract PM samples from filters and to break up aggregated particles or large clumps to obtain individual particles. Sonication bath method was used to extract PM samples from filters for PSD analyses. In the mean time, sonication extraction also served as a physical method of liquid dispersion. In the application of sonication for sample extraction and dispersion, the length of sonication time is a concern. If the sonication time is too short, it may not be able to extract the entire PM sample from the filter and break up the aggregated particles, whereas if the sonication time is too long, it may break down individual particles into smaller ones. The FS 15 Sonic Cleaner with power of 50 W, produced by Fisher Scientific was used as the sonication bath for sample extraction at NCSU.

In order to test whether sonication time for PM sample extraction had an impact on PSD measurement, a series of experiments were conducted, the most suitable sonication time was selected based upon the test results.

In the first step of the experiments, six different levels of sonication times were selected. Before the samples were introduced into the analyzer, they were put into the sonication bath for extraction for 0, 5, 10, 15, 20 and 25 minutes, respectively. Raw dust from a broiler house was used as the PM samples in the first step. Ethanol was used as the liquid medium, in which samples dispersed very well.

In the second step, four different levels of sonication times were selected: 0 minute, 5 minutes, 15 minutes and 25 minutes. In this group of tests, 21 filter-based samples were used instead of raw dust. These 21 filter-based samples were TSP samples collected from a layer barn by low-volume TSP samplers in winter 2008-2009. In order to reduce bias produced from different procedure of sample preparation, 21 filter-based samples and 120 mL ethanol were put into a 250 mL clean beaker to form one sample solution. This 120 mL sample solution was then separated into four equal parts. Each part consisting of 30 mL sample solution was put in a 100 mL beaker. The four beakers were labeled as 0, 5, 15 and 25 minutes. The beaker labeled 0 minute was not put into the sonication bath for sample extraction; beakers labeled 5, 15 and 25 minutes were put into the sonication bath for 5, 15 and 25 minutes, respectively. After the extraction procedure, samples in beakers were introduced into the analyzer. Three replicates of sample from each beaker were injected separately into the analyzer for measurements. In total, 12 (3×4) samples were analyzed by the analyzer, thus 12 MMDs and 12 GSDs were measured. A single factor ANOVA test was performed on the data to test whether sonication time affected on MMDs and GSDs at the 0.05 level of significance (α).

In the last step, microscopy was used to investigate whether the sonication time had an effect on particle size of PM qualitatively. Particles in sample solutions with different sonication times (5, 10, 15 and 30 minutes) were examined under microscope and the photos of these solutions with different magnifications (4X, 10X, 20X, 40X, 50X and 100X) were taken. Four filter-based samples collected from a layer barn in fall 2008 and 40 mL of ethanol were put into a 100 mL beaker to form one sample solution. This 40 mL sample solution was then subdivided into four equal volumes. Each part consisting of 10 mL sample solution was put in a 100mL beaker. The beakers labeled 5, 10, 15 and 30 minutes were put into the sonication bath for 5, 10, 15 and 30 minutes, respectively. After the extraction procedure, these four beakers were brought to the microscopy lab for visual particle examinations. Five replicates of sample solutions from each of these four sample solutions were examined at six different magnifications. From the five replicates, one clear and representative sample was photographed.

3.2 Experimental design for objective 1

3.2.1 Particulate matter sampling site – the layer farm

The TSP sampling was conducted on a commercial layer farm located in North Carolina. This farm consists of four tunnel-ventilated high-rise barns, two cross ventilated high-rise barns and three naturally ventilated shallow-pit barns (see Figure 3.3). Among these nine egg production barns, two tunnel-ventilated barns (3 and 4) are being monitored for the baseline emission of PM, NH_3, H_2S, CO_2 and VOCs under the National Air Emission Monitoring Study (NAEMS), which is being overseen by the US EPA (Wang, et al. 2009). Concurrent with the NAEMS study, the TSP sampling was also carried out in these two barns, in which animal activity data, ventilation data, and indoor environmental data are being collected by the NAEMS project team.

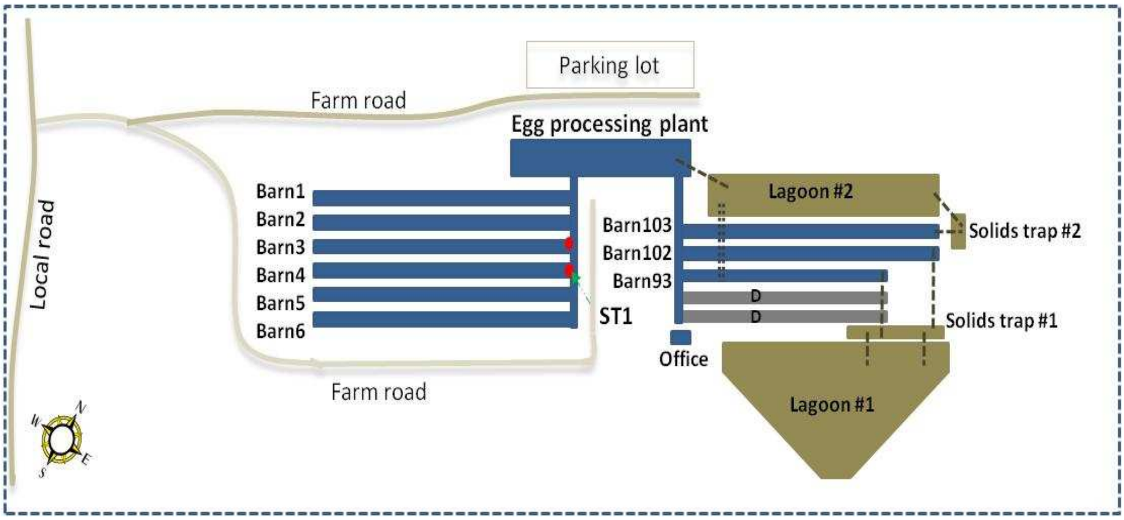

Figure 3.3 Layout of the layer farm and the locations of TSP samplers (round dots) (Wang et al. 2009)

The monitored barns are typical high-rise barns with dimensions of 175 m length × 18 m width, and have approximately 95,000 hens in six rows of 4-tier A-frame/curtain backed cages in the upper floor (referred to as 2nd floor in this study). Manure falls onto the curtain backed and then down into the pit (referred to as 1st floor in this study), where it is stored for up to one year.

3.2.2 Particulate matter samplers

Six low-volume (LV) TSP samplers were used in this study for collecting PM samples from two layer houses on the first floor, or from two floors of one house following two sampling protocols. This LV-TSP sampler was originally designed and manufactured by the research group at Texas A&M University (Wanjura, et.al, 2005). As shown in Figure 3.4, the LV TSP consists of a TSP inlet head, 47 mm filter holder, and a flow control system.

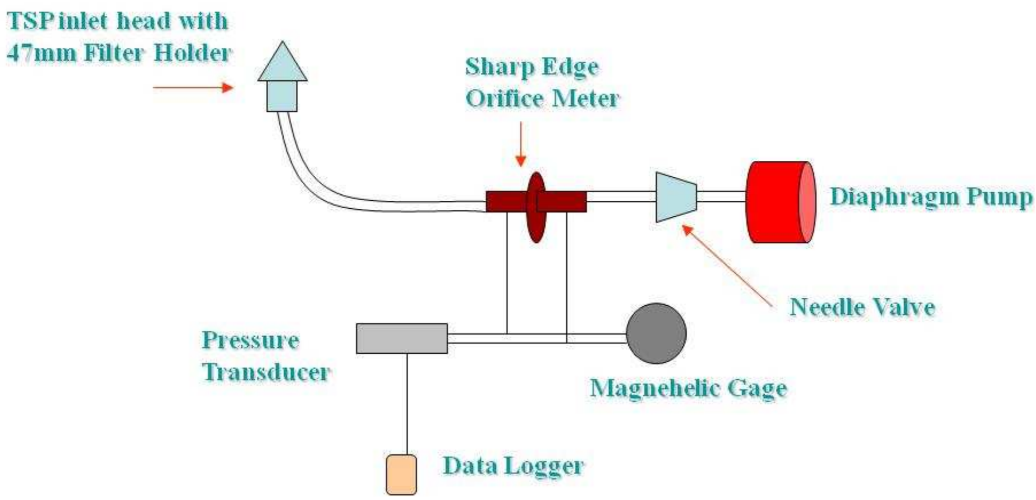

Figure 3.4 Low-volume TSP sampling system (Wanjura, et al., 2005)

In this sampling system, the pump pulls the PM-entrained air through a TSP inlet and then through a 47mm filter. The PM, which enters the TSP inlet, will be captured on the filter, and the filtered air will be discharged. An orifice meter is used to monitor the flow rate at 1 m^3/ h (16.7 l/min) by monitoring the pressure drop across the orifice meter. The following equation was used to set the proper pressure drop across the orifice meter based on the target flow rate:

$$Q = 1.252 * 10^4 * K * D_o^2 * \sqrt{\frac{\Delta P}{\rho_a}} \qquad (3.1)$$

Where:

Q = air flow rate through the orifice meter (m^3h^{-1}),

K = flow coefficient (dimensionless),

D_o = orifice diameter (m),

ΔP = pressure drop cross the orifice (mm H_2O),

ρ_a = air density (kg m^{-3}).

The pressure drop across the orifice meter was measured with a magnehelic differential pressure gauge during the sampling period. A needle valve in the sampling system was used to adjust the flow rate to keep a constant volumetric flow of 1 m^3 /h (16.7 L/min) through the sampler inlet when change in the pressure drop across orifice meter was observed.

3.2.3 Particulate matter filter preparation

Filters used in this study were 47mm Zefluor filters, which provide the least background particle counts for PSD analysis. All the filters were conditioned in an environmentally controlled chamber at 20-23 °C and 30-40 % RH for 48 h before they were weighed for pre- and post- weights.

3.2.4 Determination of TSP concentration

Total suspended particulate concentration was determined using the following equation:

$$Con._{TSP} = \frac{W_f - W_i}{Q * t} \tag{3.2}$$

Where:

$Con._{TSP}$ = TSP concentration (g/m^3),

W_i = TSP filter pre-weight (g),

W_f = TSP filter post-weight (g),

Q = average TSP sampling system air flow rate (m^3/min),

t = TSP sampling duration (min).

The system sampling flow rate (Q) may be calculated using Equation (3.1). The average of the beginning and the ending system flow rates was used for the concentration calculation.

3.2.5 Field sampling protocols

To achieve the specific research objectives, the following two sampling protocols were designed and carried out for this study.

The first set of sample collection events was designed to investigate variations in PSD of PM in two layer houses (barn #3 and barn #4). Sample collection was conducted once per week during fall, 2008 in October and November. Three TSP samplers were co-located in barn #3 and two TSP samplers were co-located in barn #4 at similar locations. All the samplers were placed inside the houses immediately upwind of the exhaust fans. For this set of sample collection, six sampling events were conducted with 5 h daytime duration per sampling event.

The second set of sample collection events was designed to investigate variations in both concentration and PSD of PM in a layer house (barn #3) at different sampling times (daytime and nighttime), on different floor (1^{st} floor and 2^{nd} floor) and across different seasons (winter and spring). This sample collection event was conducted by sampling for four consecutive days (four days and three nights) each season. Three low volume TSP samplers were co-located on the first floor and the other three TSP samplers were co-located on the second floor of the same house immediately upwind of the exhaust fans. This sampling design generated three replicates in TSP samples on each floor for comparison. The placement of the TSP samplers and the views inside the layer house are illustrated in Figure 3.5.

Figure 3.5 Placement of the LV-TSP samplers and inside view of the barn 3

(a) Three low-volume TSP samplers collocated with a TEOM sampler on the 2^{nd} floor of barn 3; (b) Chicken on the 2^{nd} floor of barn 3; (c) Three low-volume TSP samplers on the 1^{st} floor of barn 3; (d) Manure on the 1^{st} floor of barn 3.

To investigate animal activity impact on concentration and PSD of PM in the house, the sample collections were carried out following the schedule for cage lights in the barn. Daytime samples were collected from 3:00am to 8:00pm when all the cage lights were on and the chickens were quite active, whereas nighttime samples were collected from 8:00pm to 3:00am when cage lights were off and chicken activity was at a minimum. The daytime sampling events were conducted with three time intervals from 3:00am to 8:00pm every day and the nighttime sampling was conducted through 7 h duration. A total of thirteen sampling events (ten daytime and three nighttime) were performed and a total of 78 TSP filters were obtained (13 filters per TSP sampler) for each season.

In spring, summer, and fall, the ventilation system was in tunnel mode in which outside air was drawn through the house by the end wall ventilation fans from two large air inlets (36.5 m wide) located mid way of the two side walls (Figure 3.6). During winter, air was pulled by the end wall fans into the second floor through eave and ceiling inlets or diffusers. The ventilation fans were located on the two end walls of each barn and had fan blade diameters of 1.22 m (Figure 3.6 and Figure 3.7). Each end wall had eight fans on the second floor, and nine fans on the first floor.

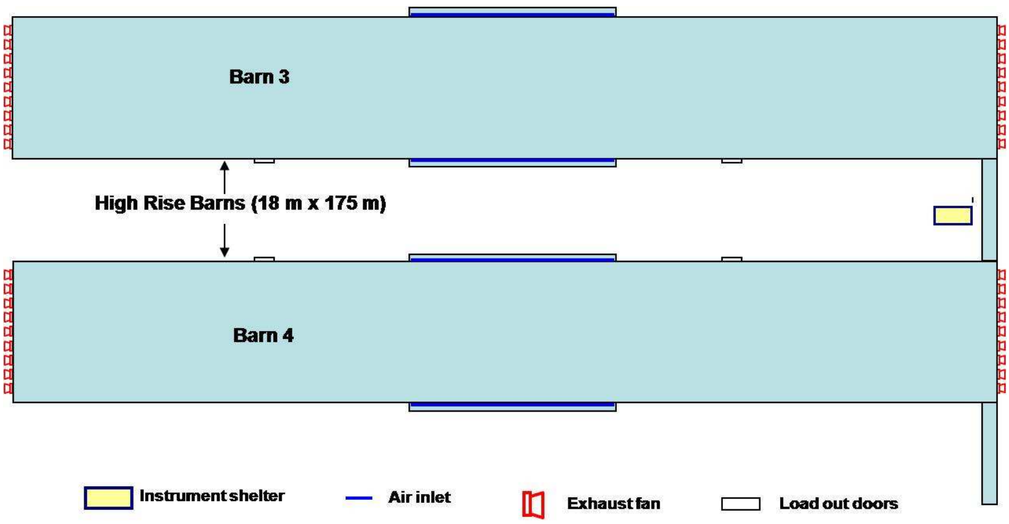

Figure 3.6 Illustration of two tunnel-ventilated barns (barn 3 and barn 4)

Figure 3.7 Outview of ventilation fans on one endwall of the barns

The ventilation system is a major factor influencing PM concentrations in the house. Figure 3.7 illustrates the ventilation fan layout on one end wall of the tunnel ventilated barn. Under the NAEMS, fan on and off status is continuously monitored either by rpm sensors or current switches. In order to keep optimal temperature for the birds in the layer houses, fewer ventilation fans were on when ambient temperatures were low during the winter, while more ventilation fans were on when the ambient temperature increased. To investigate ventilation rate impact on characteristics of PM in the house, fan running times and numbers of running fans during each sampling event were incorporated to develop an equivalent flow rate factor for data analysis. The equivalent flow rate factor is the sum of all fans' running times divided by sampling duration for each sampling event, assuming that all fans had the same flow rate (Equations (3.3) and (3.4)).

$$Equivalent\ \ flow\,rate\ \ factor\left(1st\ \ floor\right)=\frac{t_{F9}+t_{F10}+\cdots+t_{F17}}{t} \tag{3.3}$$

Where:

t_{Fn} = #n fan's running time (s), n=9, 10, $\cdots$17;

t = sampling duration (s).

$$Equivalent\ \ flow\,rate\ \ factor\left(2nd\,floor\right)=\frac{t_{F1}+t_{F2}+\cdots+t_{F8}}{t} \tag{3.4}$$

Where:

t_{Fn} = #n fan's running time (s), n=1, 2, $\cdots$8;

t = sampling duration (s).

3.2.6 Particle size distribution measurement

3.2.6.1 Particulate matter sample extraction

Total suspended particulate samples collected on the filters were used for PSD analysis in the lab. Particulate matter samples were extracted from filters into the liquid medium (ethanol) using the sonication bath method. In the preliminary study, it was observed at NCSU lab that 15 minutes was the time when the most coagulated particles were separated and yet no break-down of the particles was observed. As a result, all the TSP samples were extracted using sonication bath for 15 minutes at NCSU.

3.2.6.2 Particle Size Analyzer

A multi-wave length laser diffraction particle size analyzer (Beckman Coulter Inc., LS13 320, Miami, FL) was used to analyze PSD of PM samples collected by TSP samplers on the farm. Details about the particle size analyzer were described earlier. Particle size distribution of PM tends to follow a lognormal distribution (Stockham and Fochtman, 1977). Mass median diameter and GSD are two parameters characterizing PSD of a lognormal distribution. The MMD for a PSD is defined as the diameter for which half the total mass of particles is larger and half is smaller. The GSD describes how wide the PSD curve spread out. It is mathematically defined in Equation (2.3).

3.2.7 PM_{10} and $PM_{2.5}$ mass fraction analysis

When PSD was measured by the analyzer, the mass fraction of PM_{10} was also measured by the instrument. In the literature, there is another way to obtain mass fraction of PM_{10} and $PM_{2.5}$. This alternative way is to mathematically calculate PM_{10} and $PM_{2.5}$ mass fraction using lognormal distribution equation with measured MMD and GSD. These calculated PM_{10} and $PM_{2.5}$ mass fraction are hereby defined as lognormal fitting values. The lognormal fitting value of PM_{10} is usually what is cited by other researchers if only measured MMD and GSD are reported. It is expected that these two PM_{10} mass fractions (measured & fitting value) are the same, if PSD follows the lognormal distribution well. This research aimed to investigate the difference between these two mass fractions of PM_{10} to address possible errors in results using lognormal fitting method with measured MMD and GSD. Similarly, research was conducted to compare the difference between true $PM_{2.5}$ and $PM_{2.5}$ obtained by lognormal fitting. The relative differences (RD) were calculated using the following equation:

$$RD = \left| \frac{Measured - Lognormal}{Measured} \right| \times 100\% \qquad (3.5)$$

Where:

Measured= mass fraction of PM_{10} or $PM_{2.5}$ measured by the analyzer;

Lognormal=mass fraction of PM_{10} or $PM_{2.5}$ calculated using the lognormal distribution equation and measured MMD and GSD.

If RD is within 5%, it is defined that RD is acceptable.

3.3 Experimental design for objective 2

3.3.1 Comparison of different PSD measurement methods

Three types of instruments were used in this study to investigate the difference in PSD measurements. These three instruments included laser diffraction particle size analyzer (LS13 320) (Beckman Coulter Inc., Miami, FL) owned by Air Quality Research Group in the department of BAE at North Carolina State University (NCSU), laser scattering particle size analyzer (LA-300) (Horiba Instruments Inc., Irvine, CA) owned by Dr. Zhang's Research Group at University of Illinois at Urbana-Champaign (UIUC), Coulter Counter multisizer (CCM3) (Beckman Coulter Inc., Miami, FL) owned by Center for Agricultural Air Quality Engineering and Science (CAAQES) at Texas A & M University (TAMU), Coulter Counter multisizer (CCM 3) and laser diffraction particle size analyzer (LS230) owned by Dr. Mike Buser's group at USDA-ARS Cotton Production and Processing Research Unit in Lubbock, TX, hereby also known as USDA.

Abbreviations (NCSU, UIUC, TAMU and USDA) represent the locations in the following chapters.

3.3.1.1 Laser diffraction particle size analyzer (LS13 320)

The laser diffraction particle size analyzer (LS13 320) was described earlier and was applied to measure PSDs in objective 1. Ethanol was used as the liquid medium to disperse PM samples in this study. A sonication bath time of 15 minutes was used based on the preliminary study result.

3.3.1.2 Laser scattering particle size analyzer (LA-300)

The laser scattering particle size analyzer (LA-300) measures the PSD by angular light scattering techniques (Horiba). Light scattering can be explained by the Mie scattering theory, which is precise, but difficult to use. So, a simple approximate diffraction theory is used, called Fraunhofer diffraction theory. It is considered as Fraunhofer diffraction, when the particle size is greater than λ (wavelength of incident light). Fraunhofer diffraction is described as in Figure 3.8. A 650 nm solid-state, diode laser is focused by an automatic alignment system through the measurement cell. Light is scattered by the particles to a 42-element detector system, including high-angle and back-scatter detectors for a full angular light intensity distribution (LA-300 Overview, Horiba Instruments Inc., Irvine, CA). Larger particles scatter at smaller angles while smaller particles scatter at larger angles. The analyzer is capable of detecting samples in the size range of 0.1µm to 600µm. The LA-300 is portable analyzer, which has a compact size and light weight.

Here, 0.1% NaPMP solution was applied as the dispersant for PM samples, before the PM sample were introduced into the LA-300. The PM sample on filters in 0.1% NaPMP solution were put into a sonication bath for 15 minutes for sample extraction.

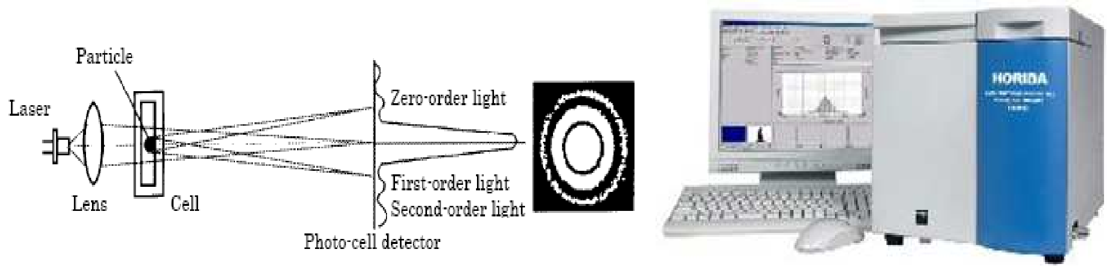

Figure 3.8 Fraunhofer diffraction and LA-300 (Horiba Instruments Inc., Irvine, CA)

3.3.1.3 Coulter Counter multisizer 3

The Coulter Counter multisizer (CCM 3) relies upon the Coulter Counter principle, also known as the Electrical sensing zone (ESZ) method. In this method, a suspension is made to flow through a small aperture separating two electrodes between which an electric current flows (Coulter Multisizer 3 Manual, Beckman Coulter Inc., Miami, FL). As shown in Figure 3.9, as particles pass through the aperture, a current flow is produced and sent into an amplifier, which converts the current into a voltage pulse. The voltage pulse is proportional to the volume of the particle that produces the pulse.

Before PM samples are injected into the analyzer, they are dispersed in an electrically conductive fluid (electrolyte). In order to extract PM samples and to well disperse particles in the electrolyte, sonication is needed. After sonication, the electrolyte with samples was forced to flow through a filtration system (screen) removing all particles larger than the size of the selected aperture, 100 μm in this study. Electrolyte used in this study was 5% lithium chloride in methanol.

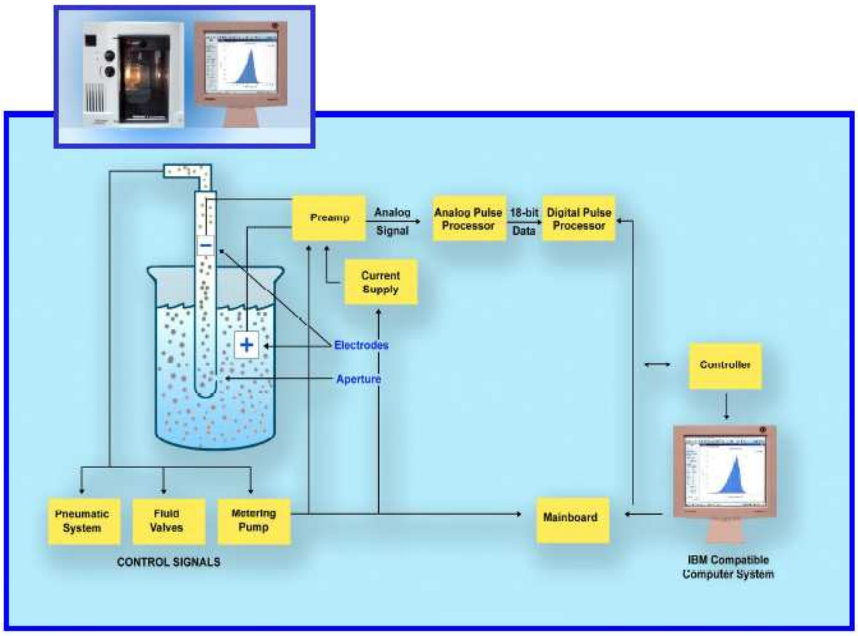

Figure 3.9 Illustration of the Coulter principle in the Multisizer 3 (Beckman Coulter, Miami, FL)

3.3.1.4 Laser diffraction particle size analyzer (LS230)

The laser diffraction particle size analyzer (LS230) is an older version of the LS13 320, and. It also measures particle size in the range of 0.04 -2000μm.

Figure 3.10 LS230 (Beckman Coulter, Miami, FL)

3.3.2 Assignment of PM samples to different locations

As described in the field sampling protocols in the experimental design for objective 1, three low volume TSP samplers were co-located on the first floor upwind of the fans and the other three TSP samplers were co-located on the second floor of the same house immediately upwind of the exhaust fans. This group of sample collection was conducted for four consecutive days (four days and three nights) each season (winter & spring), during which 78 samples were obtained per season. This sampling design generated three replicates in TSP samples on each floor. These three sample replicates on each floor were randomly selected and distributed to three research groups having different instruments at three universities. Thus, for each season, 26 samples were allocated for each of these three instruments. Particulate matter (PM) samples distributed to LA-300 and CCM 3 were shipped to UIUC and TAMU/USDA for analyses, respectively.

3.3.3 Operational procedures for different PSD instruments at different locations

After winter sampling, 26 winter samples each were shipped to TAMU and UIUC for PSD analyses, respectively. One group of 26 winter samples were kept in-house at NCSU for PSD analysis. Once PSD results from these two universities were obtained, they were compared with PSD results measured using the laser diffraction particle size analyzer (LS13 320) at NCSU.

For filter-based PSD analysis, it is required to extract PM samples from the filters and disperse them into the liquid medium for analysis. This sample extraction procedure is of great importance for PSD measurement. For winter sample analysis, sample extraction was conducted following its own operation procedure at three locations. Winter PM samples were extracted in sonication bath for 15 minutes, 15 minutes, and 5 minutes at NCSU, UIUC, and TAMU, respectively.

To reduce the uncertainty produced by different operators at different locations, the same operating procedure for sample preparation were developed and then used by all three groups at NCSU, UIUC and USDA for spring sample analyses. The USDA-ARS lab in Lubbock was also equipped with CCM 3. More details about this operating procedure can be found in Appendix B.

The same sample extraction procedure requires the same strength of sonication among three groups. Sonication baths at three locations are of different specifications (shown in Table 3.1).

Table 3.1 Specifications of the three sonication baths

Comparison of sonicationator information			
	NCSU	UIUC	USDA
Brand	Fisher Scientific	Sterlingtime	Branson
Power	50W	60W	<130W
Sonication frequency	40KHz	42KHz	40KHz
Model	FS-15	CD-4800	3510
Length of sonication time	15mins	12.5mins	15mins at 50W

As shown in this table, sonication frequency can be treated as the same among three sonicators. Thus, length of sonication time was determined according to power differences with the purpose of obtaining the same sonication strength. These lengths of sonication (shown in the Table 3.1) were followed by three groups for spring PM sample analyses.

Ethanol was used as the liquid medium in our lab at NCSU, and methanol was used in UIUC and USDA. As was discussed in the preliminary investigation, ethanol and methanol have almost the same characteristics, so liquid medium used in these three locations can be regarded as the same.

3.3.4 Comparison of PSDs of testing aerosols measured by four different instruments

In order to further investigate the difference in PSDs measured by different instruments, four types of testing aerosols were uesed to reduce the measurement uncertainty possibly caused by sample extraction at three different locations (NCSU, UIUC and USDA). These four types of testing aerosols include limestone, starch, #3 micro aluminum, and #5 micro aluminum. These testing aerosols were prepared using the same operation procedure before they were analyzed by the different analyzers.

3.4 Determination of PSDs in AED

The PSDs provided by laser diffraction particle size analyzer (NCSU), laser scattering particle size analyzer (UIUC) and Coulter Counter Multisizer (TAMU or USDA) are all in the form of ESD. The ESDs obtained from these particle analyzers were converted to the AED using the Equation (2.9).

The particle density was measured by a pycnometer (Micromeritics, AccuPyc 1330 Pycnometer, Norcross, GA) at UIUC. This pycnometer has a sample chamber of 1 cm^3. The AccuPyc uses a gas displacement method to accurately measure volume. Inert gas, helium, is used as the displacement medium. There is a fill cell that contains the sample and an expansion cell in the system. The sample is sealed in the fill cell of known volume; the helium gas is introduced into fill cell, and then released into the expansion cell. Gas molecules rapidly fill the tiniest pores of the sample; only the truly solid phase of the sample displaces the gas. The pressure before and after expansion are measured and used to compute the sample solid phase volume. Dividing this volume into the sample weight gives the gas displacement density (Wang, 2009).

For the particle shape factor investigation, six TSP samples collected in fall, 2008 and one blank filter were examined at RTI International, NC for particle shape image analysis. One representative image is shown in Figure 3.11. From the image, it was observed that most particles smaller than 10 μm appeared spherical. Larger particles were approximately spherical, though some were irregular. The shape factor was assumed to be 1 in this study.

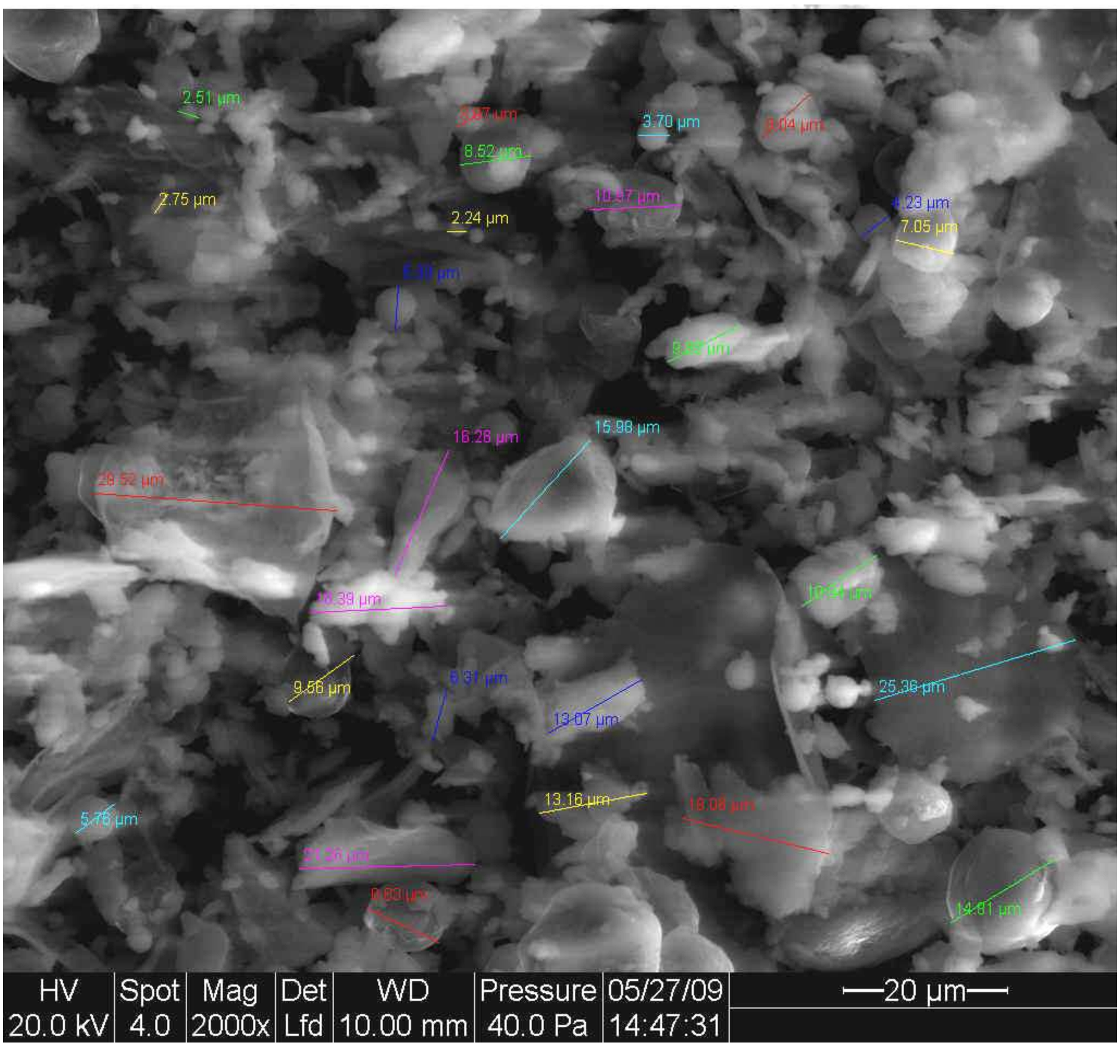

Figure 3.11 Image of particles collected by a Low-Volume TSP sampler in fall 2008, examined by a Environmental Scanning Electron Microscope with an Energy Dispersive X-ray system (E-SEM/EDX)

Chapter 4 Results and Discussions

Results of preliminary study and experiments for different objectives are presented and discussed in this chapter. Factors that might cause variation in TSP concentration and PSD are determined and discussed. PM_{10} and $PM_{2.5}$ mass fractions obtained from two methods were presented and compared.

4.1 Results of the preliminary study

4.1.1 Determination of optimal parameters to operate the LS13 320

Results of MMDs and GSDs measured by the laser diffraction particle size analyzer (LS13 320) are shown in Table 4.1 and Table 4.2. As described in the previous chapter, a 3×4 factorial design was used to test the impacts of pump speed and running time on MMDs and GSDs. Two-way analysis of variance (ANOVA) was performed on the data to test whether these two factors (pump speed & running time) had any impact on MMDs and GSDs. There was only one run for each treatment. Details of these tests are presented in Appendix C.

Table 4.1 Summary of MMDs (μm) with different pump speed and running times

		Running time (s)			
		30	60	90	120
Pump speed (%)	30	34.56	35.31	35.76	34.22
	45	34.18	33.50	34.79	34.85
	60	34.40	32.19	33.59	33.16

Table 4.2 Summary of GSDs with different pump speed and running times

		Running time (s)			
		30	60	90	120
Pump speed (%)	30	2.49	2.45	2.48	2.48
	45	2.51	2.49	2.45	2.44
	60	2.49	2.53	2.48	2.47

Both pump speed ($p=0.058$) and running time ($p=0.442$) had no statistically significant impacts on MMDs of PM samples at the 0.05 level of significance (α). Both pump speed ($p=0.519$) and running time ($p=0.459$) had no statistically significant impact on GSDs of PM samples at the 0.05 level of significance (α). There was no pump speed × running time interaction on MMDs and GSDs.

Since both pump speed and running time had no statistically significant impact on both MMDs and GSDs of PM samples, it was considered to use pump speed and running time recommended by the manufacturer (Beckman Coulter Inc., Miami, FL). The pump speed was 30% and optimal length of running time was 60 s.

4.1.2 Liquid medium identification

As described in the previous chapter, a non-aqueous medium was more suitable served as a diluent for PM than water. Non-aqueous solvents, such as methanol, ethanol and isopropanol were compared in their characteristics and it was concluded that methanol and ethanol had almost the same characteristics. Thus, ethanol and isopropanol were chosen as the solvents for tests to further identify the liquid medium. Mass median diameters and GSDs are summarized in Table 4.3 and Table 4.4. Single factor ANOVA test was performed on the data to test whether different liquid medium lead to significant difference in measurement of MMDs and GSDs. Details of these tests are presented in Appendix D.

Table 4.3 Summary of MMDs of PM dispersed in ethanol and isopropanol

	Ethanol	Isopropanol
	MMD (μm)	
1	27.28	26.54
2	28.24	26.75
3	27.64	29.33
4	27.75	29.07
5	28.33	-

There was no significant difference in MMDs (p=0.917) produced by using ethanol or isopropanol as liquid medium at the 0.05 level of significance (α).

Table 4.4 Summary of GSDs of PM dispersed in ethanol and isopropanol.

	Ethanol	Isopropanol
	GSD	
1	2.87	2.85
2	2.90	2.84
3	2.93	3.04
4	2.93	2.83
5	2.92	-

There was no significant difference in GSDs (p=0.662) produced by using ethanol or isopropanol as liquid medium at the 0.05 level of significance (α).

Since there were no significant differences both in MMDs and GSDs produced by using ethanol or isopropanol as liquid medium, it is better to use ethanol than isopropanol, because isopropanol is toxic and requires a dedicated ventilation system.

4.1.3 Determination of optimal sonication time for PM sample extraction

As described in the previous chapter, before sample particles are introduced into the LS 13 320, it is necessary to extract PM samples from filters and to break up aggregative particles or large clumps to obtain individual particles. In application of sonication for sample extraction and dispersion, the length of sonication time is a concern. A three-step experiment was conducted to determine whether sonication time had any impact on PSD measurement and to determine the optimal sonication time for PM sample extraction.

<u>First step</u>

In the first step, raw dust collected from a broiler house was used as testing aerosol. Six different levels of sonication times were selected, including 0, 5, 10, 15, 20 and 25 minutes. Mass median diameters and GSDs were measured by the analyzer (LS13 320) after PM samples were sonicated under these six different levels of sonication times. There is only one replicate for each treatment. The measurement results of MMDs and GSDs are plotted in Figure 4.1 and Figure 4.2.

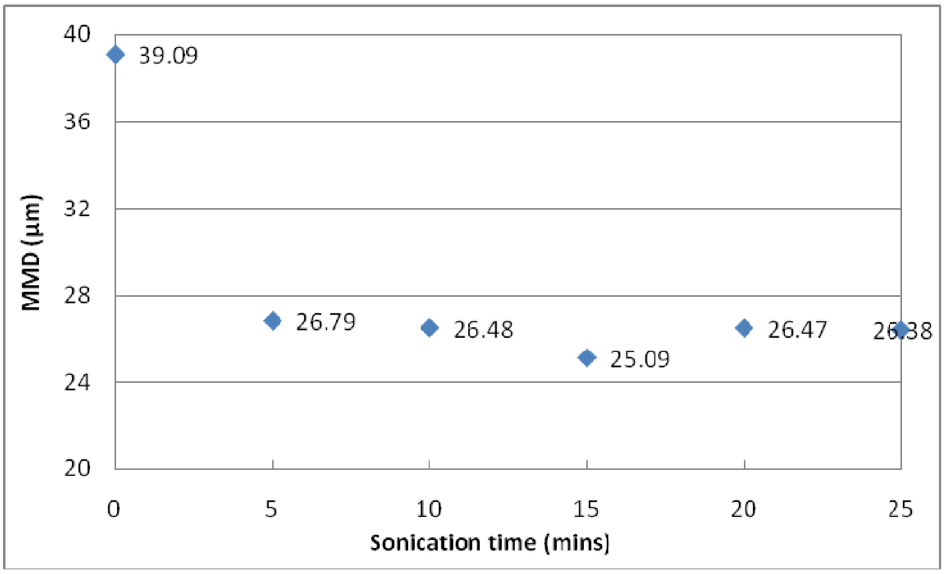

Figure 4.1 Sonication time (mins) vs. MMD (µm)

From Figure 4.1, it was observed that all MMDs were in the range between 25 and 27 µm, except the one with no sonication.

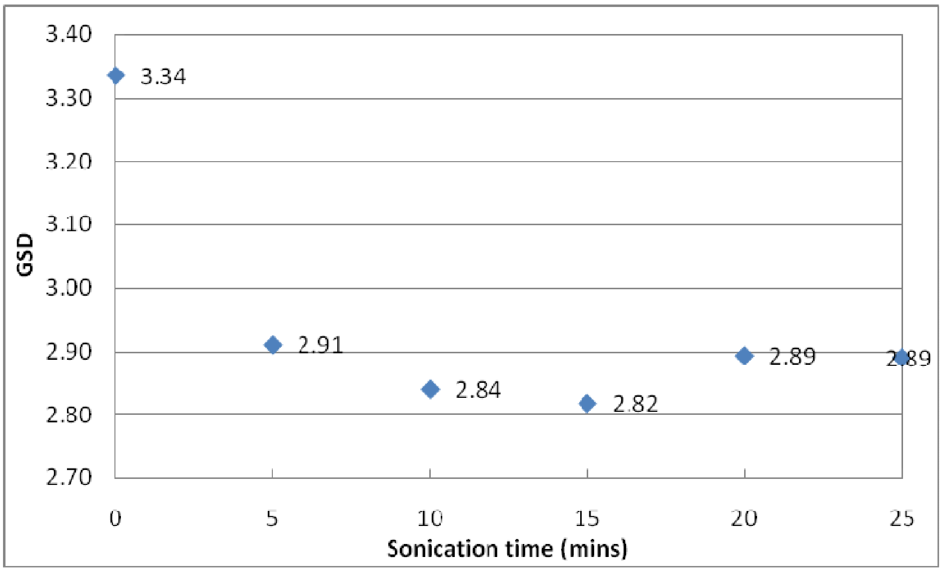

Figure 4.2 Sonication time (mins) vs. GSD

From Figure 4.2, it was observed that all GSDs were in the range between 2.8 and 2.9, except the one with no sonication.

As it is observed, both MMDs and GSDs were relatively constant except the one with no sonication. The observation suggests that sonication has significant impact on MMD and GSD measurements (sonication vs. non-sonication). However, this group of data does not suggest to what extent the sonication time affects the measurements of MMD and GSD. Further investigations were conducted in the following steps.

<u>Second step</u>

In the second step, filter-based PM samples were used as testing aerosol. Four different levels of sonication times were selected, including 0, 5, 15 and 25 minutes. As described in the previous chapter, in total, 12 (3×4) samples were analyzed by the LS13 320 analyzer. Single factor ANOVA test was performed on the data to test whether sonication time affected MMDs and GSDs. MMDs and GSDs are shown in Table 4.5 and Table 4.6. ANOVA test results in details are presented in Appendix E.

Table 4.5 Summary of MMDs (µm) with different sonication times

MMD (µm) comparison					
		Sonication time (mins)			
		0	5	15	25
Replicates	1	18.91	16.73	16.35	16.67
	2	18.37	16.92	16.56	16.85
	3	18.02	17.04	16.80	17.40
	Mean ± SD	18.43±0.45	16.90±0.16	16.57±0.23	16.97±0.38

Table 4.6 Summary of GSDs with different sonication times

		Sonication time (mins)			
		0	5	15	25
Replicates	1	2.67	2.67	2.66	2.65
	2	2.64	2.66	2.64	2.61
	3	2.55	2.66	2.66	2.62
	Mean ± SD	2.62±0.06	2.66±0.01	2.66±0.01	2.63±0.03

From ANOVA test results, it was, once again, observed that there was significant difference in MMDs with or without sonication. However, there was no significant difference in MMDs with different sonication time. This is because the difference was too small to be detected.

<u>Third step</u>

In the last step, microscopy was applied to examine whether the sonication time affected particle size of PM qualitatively. The representative images of particles in PM sample solutions with different magnifications (4X, 10X, 20X, 40X, 50X and 100X) are shown in Figure 4.3-Figure 4.8.

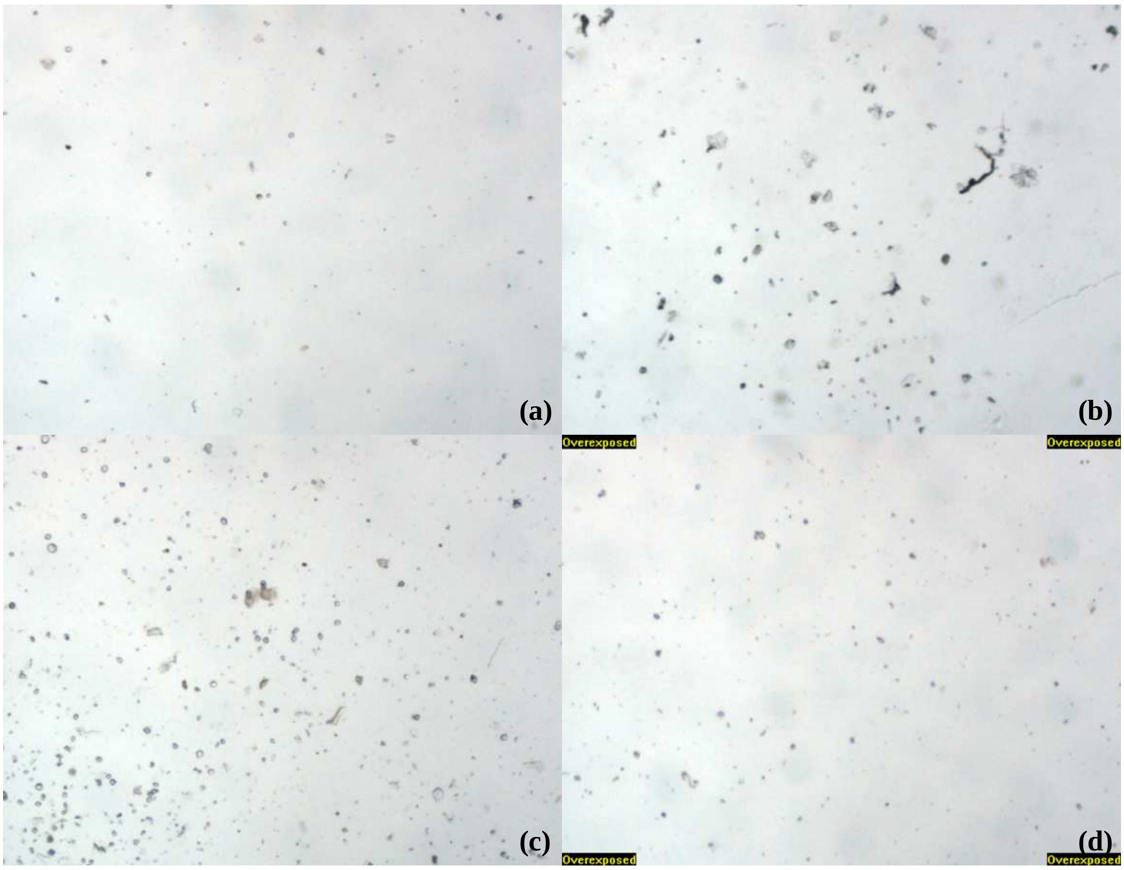

Figure 4.3 Photomicrographs (4X) of TSP sample particles collected in a layer barn in fall 2008 (a) Sonication for 5 minutes. (b) Sonication for 10 minutes. (c) Sonication for 15 minutes. (d) Sonication for 30 minutes.

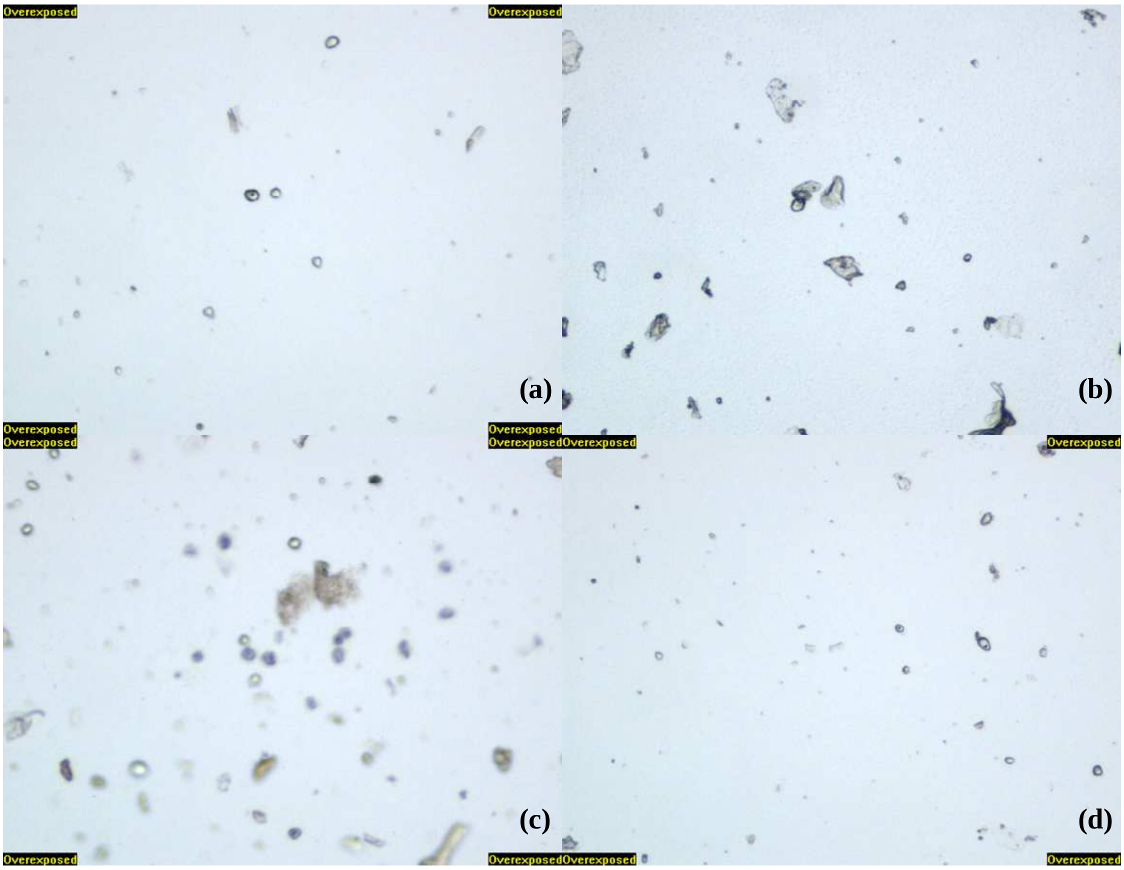

Figure 4.4 Photomicrographs (10X) of TSP sample particles collected in a layer barn in fall 2008 (a) Sonication for 5 minutes. (b) Sonication for 10 minutes. (c) Sonication for 15 minutes. (d) Sonication for 30 minutes.

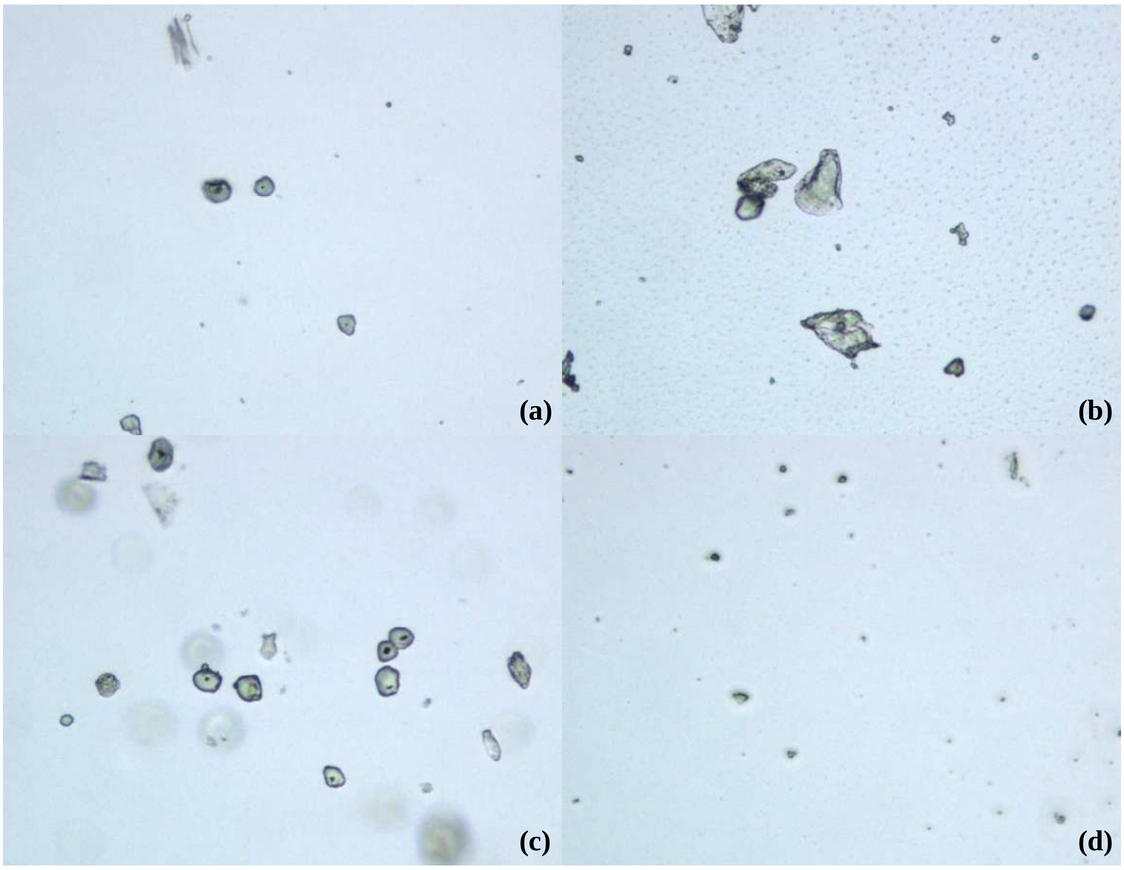

Figure 4.5 Photomicrographs (20X) of TSP sample particles collected in a layer barn in fall 2008 (a) Sonication for 5 minutes. (b) Sonication for 10 minutes. (c) Sonication for 15 minutes. (d) Sonication for 30 minutes.

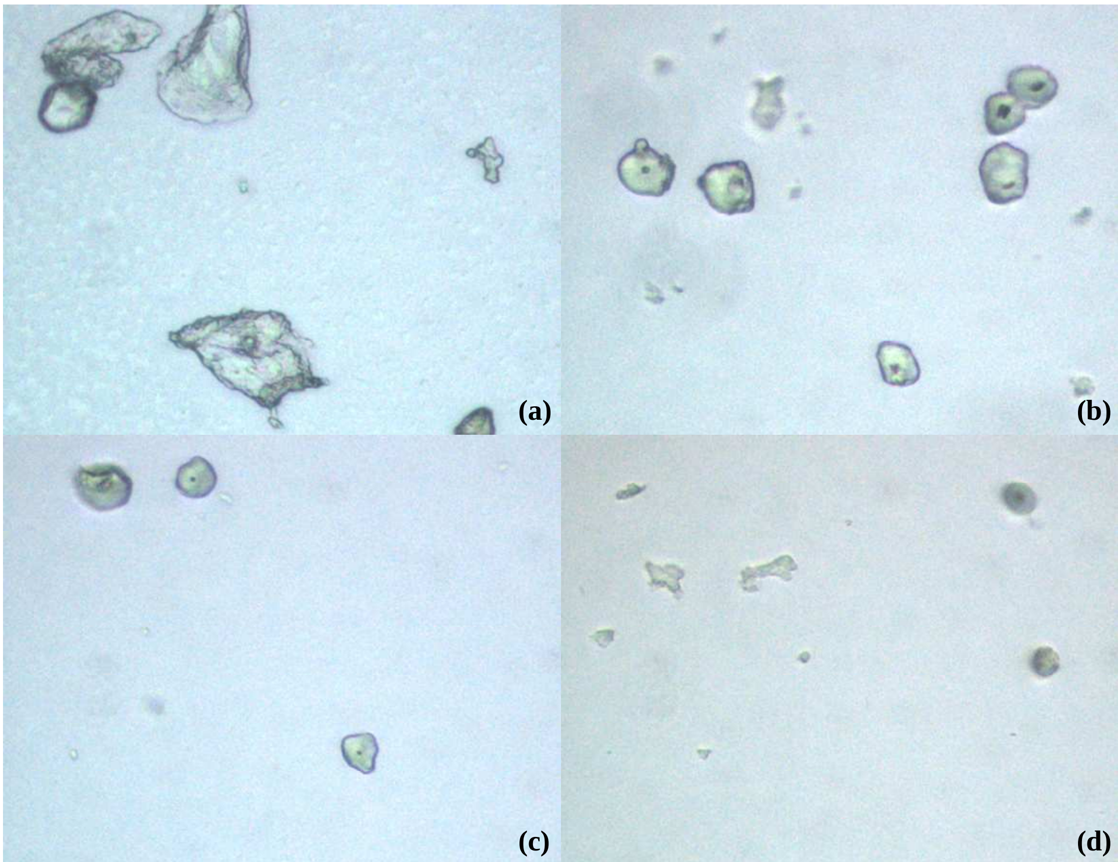

Figure 4.6 Photomicrographs (40X) of TSP sample particles collected in a layer barn in fall 2008 (a) Sonication for 5 minutes. (b) Sonication for 10 minutes. (c) Sonication for 15 minutes. (d) Sonication for 30 minutes.

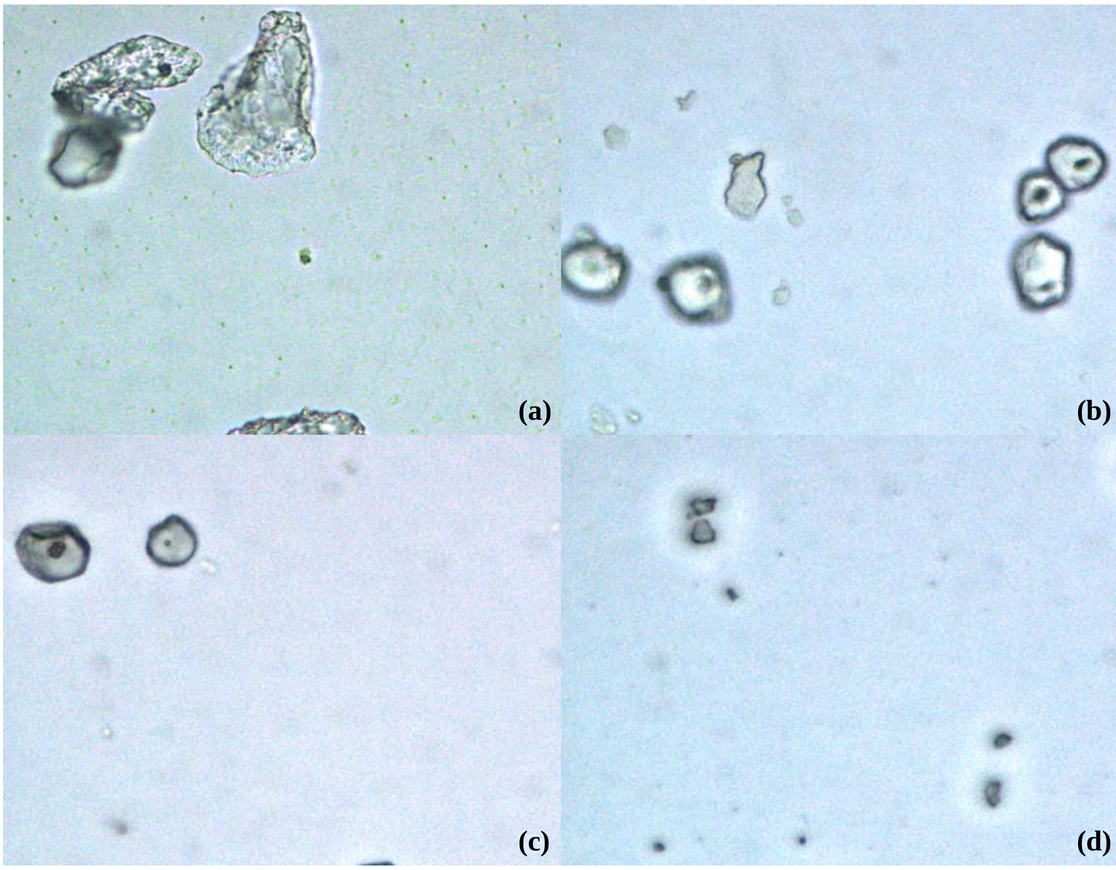

Figure 4.7 Photomicrographs (50X) of TSP sample particles collected in a layer barn in fall 2008 (a) Sonication for 5 minutes. (b) Sonication for 10 minutes. (c) Sonication for 15 minutes. (d) Sonication for 30 minutes.

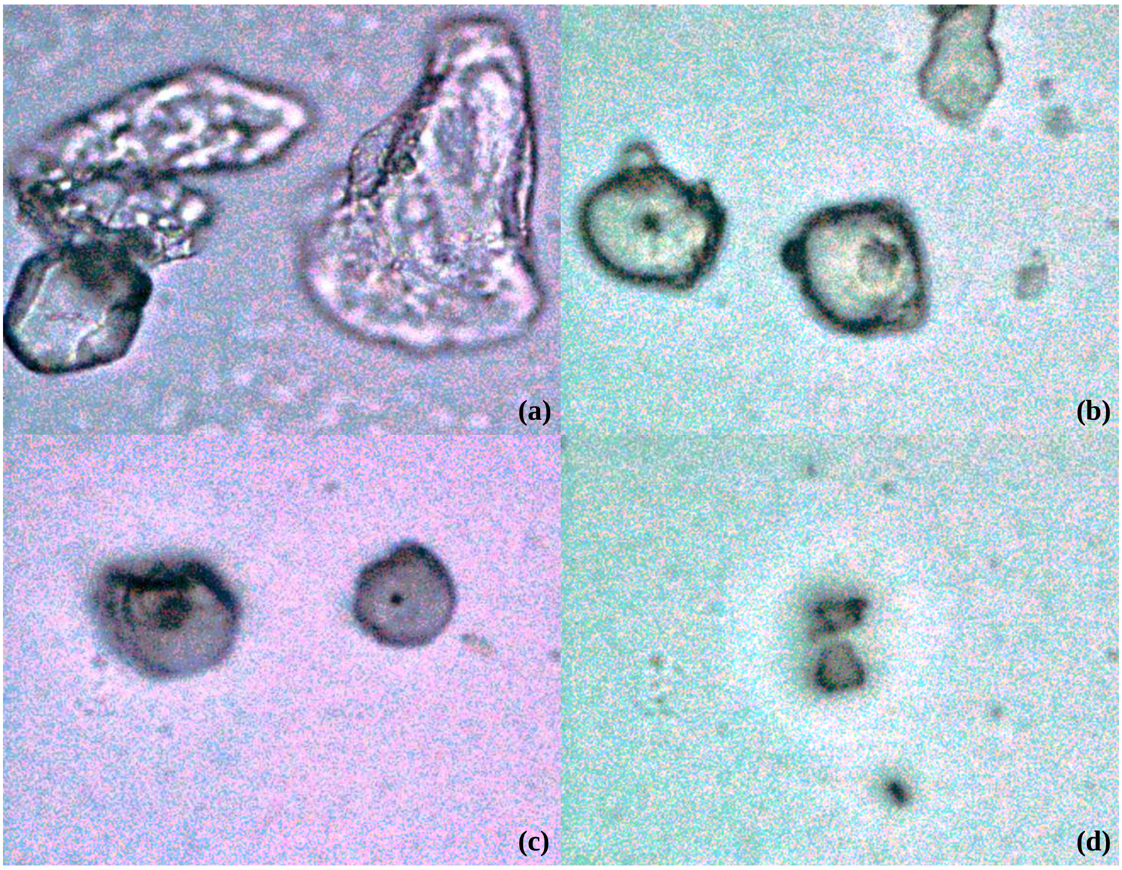

Figure 4.8 Photomicrographs (100X) of TSP sample particles collected in a layer barn in fall 2008

(a) Sonication for 5 minutes. (b) Sonication for 10 minutes. (c) Sonication for 15 minutes. (d) Sonication for 30 minutes.

From Figure 4.3-Figure 4.8, it was noticed that there were aggregative particles or large clumps in photos representing sonication for 5 minutes, whereas there were break down particles in photos representing sonication for 30 minutes. It was observed that in photos representing sonication for 15 minutes, particles dispersed well, while in photos representing sonication for 10 minutes, there were still some aggregative particles, though limited. Hence, in general, it is more suitable for samples to be sonicated for 15 minutes, compared with other sonication times discussed.

4.1.4 Preliminary conclusion

The preliminary investigation results indicate that both length of run time and pump speed had no significant effect on PSDs. Sixty seconds and thirty percent of the full pump speed were optimal operation parameters of the analyzer LS13 320. Ethanol was the most suitable liquid medium for this study. Fifteen minutes was the appropriate sonication time for sample extraction.

4.2 Particle size distribution comparison between two layer houses

The results of the PSDs of PM samples in two layer houses in fall 2008 are shown in Table 4.7 and Table 4.8. Particle size distributions were measured by LS13 320 at NCSU. T-test was performed on the data to determine if there were significant differences between the samples collected from two layer houses with respect to MMDs and GSDs. No significant differences in MMDs ($p=0.9219$) and GSDs ($p=0.4673$) were detected between the two layer houses at the 0.05 level of significance (α). The average MMD in ESD and GSD of PM collected from both two layer houses were 15.80±1.05 µm and 2.60±0.09, respectively.

Table 4.7 Comparison of MMDs in ESD and GSDs of PM in two layer houses (fall 2008)

		House #3		House #4	
Date	Sample	MMD (µm)	GSD	MMD (µm)	GSD
10/16/2008	1	16.15	2.71	14.82	2.74
10/23/2008	2	16.07	2.66	14.73	2.59
10/24/2008	3	17.16	2.55	16.65	2.58
10/30/2008	4	14.87	2.51	14.96	2.80
11/13/2008	5	13.80	2.54	16.71	2.56
11/25/2008	6	16.39	2.68	17.69	2.43
12/4/2008	7	15.49	2.54	15.56	2.61
12/11/2008	8	-[1]	-	15.96	2.58
	Mean ± SD	15.70±1.10	2.60±0.08	15.90±1.07	2.61±0.11

[1] Sample loading was too light and could not be measured by the LS13 320.

Mass median diameters in the form of ESD were converted to AED using Equation (2.6) as described in previous chapter. Results are shown in the Table 4.8. The overall MMD in AED and GSD of PM collected from these two layer houses were 19.21±1.27 µm and 2.60±0.09, respectively.

Table 4.8 Comparison of MMDs in AED and GSDs of PM in two layer houses (fall 2008)

		House #3		House #4	
Date	Sample	MMD (µm)	GSD	MMD (µm)	GSD
10/16/2008	1	19.63	2.71	18.01	2.74
10/23/2008	2	19.53	2.66	17.90	2.59
10/24/2008	3	20.86	2.55	20.24	2.58
10/30/2008	4	18.07	2.51	18.18	2.80
11/13/2008	5	16.78	2.54	20.32	2.56
11/25/2008	6	19.92	2.68	21.50	2.43
12/4/2008	7	18.83	2.54	18.91	2.61
12/11/2008	8	-	-	19.40	2.58
	Mean ± SD	19.09±1.34	2.60±0.08	19.31±1.30	2.61±0.11

[1] MMDs were analyzed using a LS 13320 multi-wave length laser diffraction particle size analyzer in ESD and then were converted to AED using Equation (2.6) with assumed shape factor of 1 and with a particle density of 1.48g/cm^3 measured by an AccuPyc 1330 Pycnometer.

4.3 Total suspended particulate concentration in the layer house

The TSP concentrations in a layer house in winter 2008-2009 and spring 2009 are shown in Table 4.9 and Table 4.10. As indicated in Table 4.9, TSP concentration varied with seasons (winter vs. spring), time of the day (daytime vs. nighttime) and floors (first floor vs. second floor). Paired comparison was first conducted on the concentrations obtained from winter and spring. The winter concentration was significantly higher (P=0.0399) than spring concentration at the 0.05 level of significance (α=0.05). Regression with indicator variables was performed on the data to further investigate the relationship between TSP concentrations and three factors (animal activity, floor and equivalent flow rate factor).

Table 4.9 TSP concentrations ($\mu g/m^3$) in the layer houses

Date	Time	Shift	First Floor[1]	Second Floor
Winter sampling events (House #3)				
01/30/2009	10: 00am-3:00pm	Day [2]	4299[3]	5056
	3:00pm-8:00pm	Day	4031	6104
	8:00pm - 3:00am	Night	935	1665
1/31/2009	3:00am-8:00am	Day	3467	5158
	8:00am-2:00pm	Day	4217	5628
	2:00pm-8:00pm	Day	3815	5557
	8:00pm - 3:00am	Night	914	1658
2/1/2009	3:00am-8:00am	Day	2854	5077
	8:00am-2:00pm	Day	4234	5001
	2:00pm-8:00pm	Day	3833	5507
	8:00pm - 3:00am	Night	815	1397
2/2/2009	3:00am-8:00am	Day	3209	5253
	8:00am-2:00pm	Day	4004	4985
Spring sampling events (House #4)				
Date	Time	Shift	First Floor	Second Floor
4/3/2009	10: 00am-3:00pm	Day	2132	3036
	3:00pm-8:00pm	Day	2127	2560
	8:00pm - 3:00am	Night	1120	1307
4/4/2009	3:00am-8:00am	Day	3942	4155
	8:00am-2:00pm	Day	2747	3052
	2:00pm-8:00pm	Day	2452	2343
	8:00pm - 3:00am	Night	1254	1395
4/5/2009	3:00am-8:00am	Day	3969	4267
	8:00am-2:00pm	Day	2692	4065
	2:00pm-8:00pm	Day	1299	1990
	8:00pm - 3:00am	Night	852	871
4/6/2009	3:00am-8:00am	Day	1315	1452
	8:00am-2:00pm	Day	1869	2417

[1] Samples were collected using six co-located LV-TSP samplers with three placed on the first floor and three placed on the second floor.

[2] The sample collections were carried out following the schedule for cage lights in the barn. Daytime samples were collected from 3:00am to 8:00pm when all the cage lights were on and the chickens were active, whereas nighttime samples were collected from 8:00pm to 3:00am when cage lights were off and chicken activity was at minimum.

[3] Each TSP concentration is the average concentration measured by three co-located TSP samplers.

As summarized in Table 4.10, TSP concentrations ranged from 888 to 5332 μg/m^3.

Table 4.10 Summary of TSP concentrations (μg/m^3) in the layer house

Floor	Winter 08-09		Spring 09	
	Day	Night	Day	Night
1	3796±479[1]	888±64	2454±933	1075±205
2	5332±361	1573±153	2934±967	1191±281

[1] Each TSP concentration is the average concentration measured by three co-located TSP samplers.

A generalized linear model in winter and another generalized linear model in spring were built using SAS 9.1 (SAS Institute Inc., Cary, NC, 2004) to test whether the TSP concentrations were related to these factors. Dummy variables were created to capture effects produced by the absence or presence of some categorical effects, such as animal activities, floor etc. It was defined, if it is daytime (animals were more active), then daytime takes the value 1, nighttime (animal were less active) takes the value 0; if it is first floor, then first floor takes the value 1, second floor takes the value 0. Since animal activity follows the patterns of lighting cycle, which means during the daytime, when the lights are on, animal activity is higher than during the nighttime, when the lights are off.

Equivalent flow rate factor (EFRF) is defined as the sum of all fans' running time divided by sampling duration, assuming that all fans have the same flow rate (Equation (3.3) and (3.4)). With more fans on, the equivalent flow rate factor is higher. Equivalent flow rate factors are summarized in the Table 4.11. Collinearity of factors in these models was ignored. Details about the models are presented in Appendix I.

Table 4.11 Equivalent flow rate factors (EFRFs) in the layer houses

Winter sampling events (House #3)				
Date	Time	Shift	Floor	EFRF
Day 1 01/30/2009	10: 00am-3:00pm	Day	1st	1.39
			2nd	0
	3:00pm-8:00pm	Day	1st	1.27
			2nd	0
	8:00pm - 3:00am	Night	1st	1.01
			2nd	0
Day 2 01/31/2009	3:00am-8:00am	Day	1st	1.15
			2nd	0
	8:00am-2:00pm	Day	1st	1.27
			2nd	0
	2:00pm-8:00pm	Day	1st	1.17
			2nd	0
	8:00pm - 3:00am	Night	1st	1.08
			2nd	0
Day 3 02/01/2009	3:00am-8:00am	Day	1st	1.24
			2nd	0
	8:00am-2:00pm	Day	1st	1.95
			2nd	0
	2:00pm-8:00pm	Day	1st	1.56
			2nd	0
	8:00pm - 3:00am	Night	1st	1.19
			2nd	0
Day 4 02/02/2009	3:00am-8:00am	Day	1st	1.28
			2nd	0
	8:00am-2:00pm	Day	1st	1.92
			2nd	0

Table 4.10 Continued

Spring sampling events (House #4)				
Date	Time	Shift	Floor	EFRF
Day 1 04/03/2009	10: 00am-3:00pm	Day	1st	7.97
			2nd	1.23
	3:00pm-8:00pm	Day	1st	5.36
			2nd	0.38
	8:00pm - 3:00am	Night	1st	1.3
			2nd	0
Day 2 04/04/2009	3:00am-8:00am	Day	1st	1.66
			2nd	0
	8:00am-2:00pm	Day	1st	6.47
			2nd	0.92
	2:00pm-8:00pm	Day	1st	5.27
			2nd	1.54
	8:00pm - 3:00am	Night	1st	1.11
			2nd	0
Day 3 04/05/2009	3:00am-8:00am	Day	1st	1.85
			2nd	0
	8:00am-2:00pm	Day	1st	8.51
			2nd	2.69
	2:00pm-8:00pm	Day	1st	7.92
			2nd	3.79
	8:00pm - 3:00am	Night	1st	3.69
			2nd	0
Day 4 04/06/2009	3:00am-8:00am	Day	1st	7.03
			2nd	0.15
	8:00am-2:00pm	Day	1st	5.84
			2nd	0.13

In winter, the regression model can be described as follow:

$$\widehat{Concentration} = 3333.5 AnimalActivity - 1339.8 Floor + 1900.8 \quad (4.1)$$

Where:

Animal activity=1, if it is the daytime sample; animal activity=0, if it is the nighttime sample;

Floor=1, if the sample was collected on the first floor; floor=0, if the sample was collected on the second floor.

Animal activity ($p<0.0001$) is statistically significant for predicting concentration, given floor is in the model; floor ($p<0.0001$) is statistically significant for predicting concentration, given animal activity is in the model; equivalent flow rate factor ($p=0.58$) is not statistical significance for predicting concentration, given animal activity and floor are in the model.

The regression analysis shows a very good fit ($R^2=0.94$). Animal activity, having an estimate of 3333.5, ranks first in affecting concentration; floor ranks second. In general, the more active the animal were (during the daytime), the higher the concentration on the same floor in winter; concentration on the second floor was higher than that on the first floor during the same period of the day (Table 4.9).

In spring, the regression model can be described as follow:

$$\widehat{Concentration} = 1942.2\,AnimalActivity - 158.7\,EquivalentFlowRateFactor + 1293.5 \tag{4.2}$$

Where:

Equivalent flow rate factor is a numeric variable (see Equation (3.3), (3.4) and Table 4.11);

Other variables in this model are the same as defined in Equation (4.1).

Animal activity ($p<0.0001$) is statistically significant for predicting concentration, given equivalent flow rate factor is in the model; equivalent flow rate ($p=0.0086$) is statistically significant for predicting concentration, given animal activity is in the model; floor ($p=0.1994$) is not statistically significant for predicting concentration, given animal activity and equivalent flow rate are in the model.

The regression analysis shows a good fit ($R^2=0.55$). Animal activity, which has an estimate of 1942.2, ranks first in affecting concentration; equivalent flow rate ranks second. In general, concentration is higher during daytime than during nighttime; the higher the flow rate, the lower the concentration.

4.4. Particle size distribution of PM in the layer house

Table 4.12 lists the results of PSDs of PM samples taken in two seasons. A generalized linear model was also built to test whether MMD is related to those factors (animal activity, floor, season and, equivalent flow rate factor) addressed above. Dummy variables were also created with the same definition as described above. The model ($R^2=0.36$) can be described as follow:

$$M\widehat{M}D = -1.33 Floor + 0.4 EquivalentFlowRateFactor + 17.7 \qquad (4.3)$$

Variables in this model are the same as defined in Equation (4.1) and (4.2). Floor ($p=0.0008$) is statistically significant for predicting MMD, given equivalent flow rate factor is in the model; equivalent flow rate factor ($p<0.0001$) is statistically significant for predicting MMD, given floor is in the model. Both season (p=0.1296) and animal activity (p=0.4919) are not statistically significant in affecting MMD, given floor and equivalent flow rate are in the model. Though R^2 of 0.36 for this model was not good enough, this model was acceptable as an indicator of variables.

The average MMD of winter and spring samples is 17.79±1.33 µm (Table 4.12). MMD affected by floor and equivalent flow rate is within one standard deviation of average MMD. Overall MMD for winter samples is 17.13±0.81 µm. Overall MMD for spring samples is 18.44±1.44 µm.

Similarly, in order to test whether GSD is related to the given factors, a generalized linear model was built. The model (R^2=0.16) can be described as follow:

$$G\hat{S}D = -0.034 Season - 0.021 AnimalActivity + 0.046 Floor + 0.001 EquivalentFlowRate + 2.66 \quad (4.4)$$

Where:

Season=1, if it was in winter; season=0, if it was in spring;

The rest of variables in this model are the same as defined in Equation (4.1) and (4.2).

Season (p=0.2069), animal activity (p=0.4355), floor (p=0.1210) and equivalent flow rate factor (p=0.8602) are all not statistically significant for predicting GSD, given other three factors are in the model. In general, GSD is constant and is not affected by these factors. Overall GSD for both winter and spring is 2.65±0.08. Details about the models are presented in Appendix J.

Table 4.12 Summary of PSDs obtained in two seasons (winter & spring)

Winter sampling events (House #3)						
			First Floor		Second Floor	
Date	Time	Shift	MMD[1] (μm)	GSD	MMD (μm)	GSD
1/30/2009	10: 00am-3:00pm	Day	17.26	2.65	16.7	2.61
	3:00pm-8:00pm	Day	16.82	2.59	17.59	2.61
	8:00pm - 3:00am	Night	18.62	2.72	17.8	2.63
1/31/2009	3:00am-8:00am	Day	15.62	2.7	16.86	2.68
	8:00am-2:00pm	Day	17.21	2.61	17.28	2.64
	2:00pm-8:00pm	Day	16.35	2.63	16.81	2.64
	8:00pm - 3:00am	Night	15.9	2.66	16.8	2.63
2/1/2009	3:00am-8:00am	Day	16.41	2.67	16.41	2.68
	8:00am-2:00pm	Day	17.96	2.61	17.33	2.6
	2:00pm-8:00pm	Day	17.66	2.6	17.23	2.61
	8:00pm - 3:00am	Night	15.91	2.68	17.3	2.58
2/2/2009	3:00am-8:00am	Day	17.03	2.65	17.16	2.61
	8:00am-2:00pm	Day	18.38	2.59	18.97	2.58
Mean ± SD (Winter)			17.13±0.81		2.65±0.08	
Spring sampling events (House #4)						
			First Floor		Second Floor	
Date	Time	Shift	MMD (μm)	GSD	MMD (μm)	GSD
4/3/2009	10: 00am-3:00pm	Day	19.67	2.7	19.41	2.5
	3:00pm-8:00pm	Day	17.36	2.72	18.12	2.65
	8:00pm - 3:00am	Night	17.19	2.72	16.56	2.65
4/4/2009	3:00am-8:00am	Day	17.42	2.75	16.68	2.71
	8:00am-2:00pm	Day	18.69	2.62	18.09	2.61
	2:00pm-8:00pm	Day	17.64	2.61	17.79	2.61
	8:00pm - 3:00am	Night	18.74	2.68	17.66	2.69
4/5/2009	3:00am-8:00am	Day	17.25	2.73	17.75	2.75
	8:00am-2:00pm	Day	18.77	2.57	21.22	2.49
	2:00pm-8:00pm	Day	21.35	3.06	21.98	2.66
	8:00pm - 3:00am	Night	19.14	2.75	20.22	2.61
4/6/2009	3:00am-8:00am	Day	17.3	2.61	17.52	2.66
	8:00am-2:00pm	Day	17.7	2.7	18.23	2.61
Mean ± SD (Spring)			18.44±1.44		2.65±0.08	

Mean ± SD for both winter and spring	17.79±1.33	2.65±0.08

[1] MMDs and GSDs were analyzed using a LS 13320 multi-wave length laser diffraction particle size analyzer in ESD and then were converted to AED using equation 3 with assumption of shape factor of 1 and with a particle density of 1.48 g/cm^3 measured by an AccuPyc 1330 Pycnometer.

4.5 Particle size distributions of PM measured by different instruments

4.5.1 Particle size distributions of PM collected in fall 2008

Particulate matter samples collected in fall 2008 were measured by the laser diffraction particle size analyzer (LS13 320) at NCSU and the laser scattering particle size analyzer (LA-300) at UIUC.

Since no significant differences in PSDs were detected between two layer houses, PM samples from two layer houses were combined to be analyzed. Particulate matter samples were all collected on the first floor of two layer houses in fall, so it was assumed that PM samples measured in NCSU and UIUC were collected under the same environment factors.

The PSDs from NCSU and UIUC are summarized in Table 4.13. T-test was performed on the data to determine if there were significant differences in MMDs ($p=0.018$) and GSDs ($p=0.0214$) measured by LS13 320 and LA-300. Significant differences in MMDs and GSDs measured by these two different instruments were detected at the 0.05 level of significance (α). The overall MMD and GSD obtained from LS13 320 were 19.27±1.27

μm and 2.60±0.09, respectively. The overall MMD and GSD obtained from LA-300 were 24.69±2.44 μm and 2.02±0.18, respectively.

Table 4.13 Summary of PSDs obtained from LS13 320 (NCSU) and LA-300 (UIUC)

	LS13 320		LA-300	
Sample	MMD (μm)	GSD	MMD (μm)	GSD
1	19.63	2.71	23.19	1.81
2	19.53	2.66	23.85	2.07
3	20.86	2.55	23.42	2.12
4	18.07	2.51	23.12	2.18
5	16.78	2.54	22.13	1.80
6	19.92	2.68	19.95	1.75
7	18.83	2.54	25.46	2.09
8	18.01	2.74	24.13	1.96
9	17.90	2.59	20.00	1.86
10	20.24	2.58	24.11	2.05
11	18.18	2.80	27.19	2.08
12	20.32	2.56	27.00	2.41
13	21.50	2.43	26.86	2.06
14	18.91	2.61	26.29	2.07
15	19.40	2.58	27.43	1.81
16	-[1]	-	27.82	2.31
17	-	-	25.41	2.11
18	-	-	27.00	1.86
Mean ± SD	19.21±1.27	2.60±0.09	24.69±2.44	2.02±0.18

[1] Sample loading was too light and could not be measured by the LS13 320.

From Table 4.13, it was noticed that MMDs obtained by LA-300 at UIUC were larger than that obtained by LS13 320 at NCSU. From Table 4.13, it was also noticed that GSDs obtained by LA-300 at UIUC were smaller than that obtained by LS13 320 at NCSU.

4.5.2 Particle size distributions of PM collected in winter 2008-2009

Particulate matter samples collected in winter 08-09 were measured using LS13 320 at NCSU, LA-300 at UIUC and CCM 3 at TAMU. As introduced in the previous chapter, three low volume TSP samplers were co-located on the first floor and the other three TSP samplers were co-located on the second floor of the same house immediately upwind of the exhaust fans. This sampling design generated three replicates in TSP samples on each floor for comparison. Three replicates obtained on each floor were randomly distributed to the three universities for PSD analyses. The PSDs from these three different instruments are presented in Table 4.14 and Table 4.15.

Table 4.14 Summary of MMDs obtained from three instruments (LS13 320, LA-300 & CCM 3)

Date	Time	Shift	Floor	MMD (μm) LS13 320	LA-300	CCM 3
Day 1 01/30/2009	10: 00am-3:00pm	Day	1st floor	17.26	23.38	14.08
			2nd floor	16.70	22.87	14.20
	3:00pm-8:00pm	Day	1st floor	16.82	23.23	14.35
			2nd floor	17.59	22.96	13.88
	8:00pm - 3:00am	Night	1st floor	18.62	20.97	13.71
			2nd floor	17.80	20.14	13.79
Day 2 01/31/2009	3:00am-8:00am	Day	1st floor	15.62	21.72	13.21
			2nd floor	16.86	22.91	13.68
	8:00am-2:00pm	Day	1st floor	17.21	23.60	18.25
			2nd floor	17.28	23.20	13.64
	2:00pm-8:00pm	Day	1st floor	16.35	22.71	14.05
			2nd floor	16.81	22.53	14.03
	8:00pm - 3:00am	Night	1st floor	15.90	20.10	13.57
			2nd floor	16.80	23.51	13.93
Day 3 02/01/2009	3:00am-8:00am	Day	1st floor	16.41	21.65	13.21
			2nd floor	16.41	24.38	13.23
	8:00am-2:00pm	Day	1st floor	17.96	22.58	13.99
			2nd floor	17.33	26.62	14.29
	2:00pm-8:00pm	Day	1st floor	17.66	21.83	14.14
			2nd floor	17.23	22.75	13.95
	8:00pm - 3:00am	Night	1st floor	15.91	21.71	12.48
			2nd floor	17.30	21.00	13.23
Day 4 02/02/2009	3:00am-8:00am	Day	1st floor	17.03	21.98	12.86
			2nd floor	17.16	24.36	14.07
	8:00am-2:00pm	Day	1st floor	18.38	24.70	14.11
			2nd floor	18.97	23.00	14.46
Mean ± SD				17.13±0.81	22.71±1.43	13.94±1.00

From Table 4.14, significant but constant difference in MMDs among three different instruments was observed. In general, LA-300 at UIUC provided the largest MMDs; whereas CCM 3 at TAMU gave the smallest MMDs. The average MMDs in winter obtained by LS13 320, LA-300 and CCM 3 were 17.13±0.81 µm, 22.71±1.43 µm and 13.94±1.00 µm, respectively.

Table 4.15 Summary of GSDs obtained from three instruments (LS13 320, LA-300 & CCM 3)

Date	Time	Shift	Floor	GSD		
				LS13 320	LA-300	CCM 3
Day 1 01/30/2009	10: 00am-3:00pm	Day	1st	2.65	2.05	1.88
			2nd	2.61	2.02	1.87
	3:00pm-8:00pm	Day	1st	2.59	1.96	1.91
			2nd	2.61	1.94	1.92
	8:00pm - 3:00am	Night	1st	2.72	1.96	1.86
			2nd	2.63	1.88	1.84
Day 2 01/31/2009	3:00am-8:00am	Day	1st	2.70	1.96	1.84
			2nd	2.68	1.95	1.87
	8:00am-2:00pm	Day	1st	2.61	2.00	1.84
			2nd	2.64	2.17	1.84
	2:00pm-8:00pm	Day	1st	2.63	2.13	1.80
			2nd	2.64	2.05	1.80
	8:00pm - 3:00am	Night	1st	2.66	2.01	1.87
			2nd	2.63	1.84	1.85
Day 3 02/01/2009	3:00am-8:00am	Day	1st	2.67	2.02	1.85
			2nd	2.68	2.26	1.87
	8:00am-2:00pm	Day	1st	2.61	1.96	1.87
			2nd	2.60	2.37	1.82
	2:00pm-8:00pm	Day	1st	2.60	2.06	1.78
			2nd	2.61	2.01	1.88
	8:00pm - 3:00am	Night	1st	2.68	1.91	1.87
			2nd	2.58	1.91	1.87
Day 4 02/02/2009	3:00am-8:00am	Day	1st	2.65	2.06	1.92
			2nd	2.61	2.08	1.84
	8:00am-2:00pm	Day	1st	2.59	2.05	1.85
			2nd	2.58	1.99	1.80
Mean ± SD				2.63±0.04	2.02±0.11	1.85±0.04

From Table 4.15, significant but constant difference in GSDs among three different universities was observed. In general, LS13 320 provided the largest GSDs; whereas

CCM 3 gave the smallest MMDs. The overall GSDs in winter obtained from LS13 320. LA-300 and CCM 3 were 2.63±0.04, 2.02±0.11 and 1.85±0.04, respectively.

Single factor ANOVA test was performed on the data to test whether PSDs (MMDs & GSDs) obtained by these three instruments are significantly different. It was detected that there was significant difference in MMDs ($p<0.0001$) and GSDs ($p<0.0001$) obtained by these three instruments.

Tukey-Kramer test, one type of multiple comparison procedures (MCPs) was performed on the data to further test whether PSDs (MMDs & GSDs) from these three instruments are significantly different from each other. Significant differences in MMDs ($p<0.0001$) and GSDs ($p<0.0001$) from three instruments were observed. Details about these tests can be found in Appendix F.

4.5.3 Particle size distributions of PM collected in spring 2009

Particulate matter samples collected in spring 09 were measured by LS13 320 at NCSU, LA-300 at UIUC, CCM 3 and LS230 at USDA. The PSDs from LS13 320 at NCSU, LA-300 at UIUC, CCM 3 and LS230 at USDA are summarized in Table 4.16 and Table 4.17.

Table 4.16 Summary of MMDs obtained from four different instruments (LS13 320, LA-300, LS230 and CCM)

Date	Time	Shift	Floor	MMD (μm)			
				LS13 320	LA-300	LS230	CCM 3
Day 1 04/03/2009	10: 00am-3:00pm	Day	1st	19.67	24.50	18.51	13.68
			2nd	19.41	24.41	18.87	14.43
	3:00pm-8:00pm	Day	1st	17.36	21.10	17.47	13.63
			2nd	18.12	24.76	18.15	14.08
	8:00pm - 3:00am	Night	1st	17.19	21.62	18.38	13.08
			2nd	16.56	21.87	17.30	13.70
Day 2 04/04/2009	3:00am-8:00am	Day	1st	17.42	24.01	18.28	-
			2nd	16.68	22.08	17.42	13.93
	8:00am-2:00pm	Day	1st	18.69	23.13	18.11	13.07
			2nd	18.09	23.20	18.40	14.47
	2:00pm-8:00pm	Day	1st	17.64	21.55	18.08	14.53
			2nd	17.79	22.80	17.98	14.21
	8:00pm - 3:00am	Night	1st	18.74	19.67	17.32	13.29
			2nd	17.66	21.78	18.04	12.80
Day 3 04/05/2009	3:00am-8:00am	Day	1st	17.25	22.04	17.32	14.23
			2nd	17.75	22.58	18.18	13.50
	8:00am-2:00pm	Day	1st	18.77	24.70	21.10	14.49
			2nd	21.22	25.01	22.62	15.38
	2:00pm-8:00pm	Day	1st	21.35	27.84	19.59	13.94
			2nd	21.98	25.82	20.91	15.78
	8:00pm - 3:00am	Night	1st	19.14	21.58	17.08	12.91
			2nd	20.22	21.10	19.44	14.50
Day 4 04/06/2009	3:00am-8:00am	Day	1st	17.30	15.92	-	13.86
			2nd	17.52	20.44	-	14.94
	8:00am-2:00pm	Day	1st	17.70	21.85	17.25	13.47
			2nd	18.23	22.77	17.53	13.86
Mean ± SD				18.44±1.44	22.62±2.68	18.47±1.38	13.99±0.74

From Table 4.16, significant but constant difference in MMDs obtained from four different instruments (LS13 320, LA-300, LS230 and CCM) was observed. In general, LA-300 at UIUC provided the largest MMDs (22.62±2.68 μm), whereas CCM at USDA gave the smallest MMDs (13.99±0.74 μm). LS13 320 at NCSU (18.44±1.44 μm) and LS230 at USDA (18.47±1.38 μm) provided very similar results for MMDs.

Table 4.17 Summary of GSDs obtained from four different instruments (LS13 320, LA-300, LS230 and CCM)

Date	Time	Shift	Floor	GSD			
				LS13 320	LA-300	LS230	CCM 3
Day 1 04/03/2009	10: 00am-3:00pm	Day	1st	2.70	1.99	2.58	1.88
			2nd	2.50	2.00	2.50	1.82
	3:00pm-8:00pm	Day	1st	2.72	1.69	2.58	1.82
			2nd	2.65	2.14	2.43	1.82
	8:00pm - 3:00am	Night	1st	2.72	1.99	2.62	1.87
			2nd	2.65	2.07	2.59	1.87
Day 2 04/04/2009	3:00am-8:00am	Day	1st	2.75	2.09	2.72	-
			2nd	2.71	2.02	2.65	1.86
	8:00am-2:00pm	Day	1st	2.62	2.17	2.57	1.84
			2nd	2.61	1.94	2.53	1.78
	2:00pm-8:00pm	Day	1st	2.61	1.90	2.66	1.85
			2nd	2.61	1.98	2.54	1.81
	8:00pm - 3:00am	Night	1st	2.68	1.74	2.71	1.86
			2nd	2.69	1.88	2.73	1.86
Day 3 04/05/2009	3:00am-8:00am	Day	1st	2.73	2.08	2.71	1.84
			2nd	2.75	2.09	2.89	1.81
	8:00am-2:00pm	Day	1st	2.57	2.13	2.33	1.84
			2nd	2.49	2.06	2.47	1.81
	2:00pm-8:00pm	Day	1st	3.06	2.30	3.01	1.94
			2nd	2.66	2.12	3.41	1.81
	8:00pm - 3:00am	Night	1st	2.75	2.01	2.57	1.88
			2nd	2.61	1.78	2.47	1.84
Day 4 04/06/2009	3:00am-8:00am	Day	1st	2.61	1.73	-	1.83
			2nd	2.66	1.90	-	1.73
	8:00am-2:00pm	Day	1st	2.70	2.05	2.76	1.83
			2nd	2.61	1.96	2.62	1.81
Mean ± SD				2.67±0.11	1.99±0.15	2.65±0.22	1.84±0.04

From Table 4.17, significant but constant difference in GSDs obtained from four different instruments (LS13 320, LA-300, LS230 and CCM) was observed. LS13 320 at NCSU (2.67±0.11) and LS230 at USDA (2.65±0.22) provided very similar results for GSDs. In general, LS series instruments (LS13 320 at NCSU and LS230 at USDA) provided the largest GSDs, whereas CCM at USDA gave the smallest GSDs (1.84±0.04).

Single factor ANOVA test was performed on the data to test whether PSDs (MMDs & GSDs) obtained by these four instruments are significantly different. It was detected that there was significant difference in MMDs ($p<0.0001$) and GSDs ($p<0.0001$) obtained by these four instruments.

The T-test was performed on the data to further test whether PSDs (MMDs & GSDs) from these four instruments are significantly different from each other. There was no significant different in MMDs ($p=0.400$) and GSDs ($p=0.297$) between LS13 320 at NCSU and LS230 at USDA. Significant differences in MMDs ($p<0.0001$) and GSDs ($p<0.0001$) from three instruments (LS13 320, LA-300 and CCM) were observed. Details about these tests can be found in Appendix G.

4.5.4 Particle size distributions measurements of testing aerosols

As described in the previous chapter, four types of testing aerosols (limestone, starch, #3 micro aluminum and #5 micro aluminum) were analyzed for PSDs at three different locations (NCSU, UIUC and USDA) using four different instruments. Four types of testing aerosols were measured by two instruments (LS230 and CCM 3) at the USDA. Comparison of PSDs measured by four instruments at these three locations was made.

The PSDs obtained from LS13 320, LA-300, LS230 and CCM 3 are shown in Table 4.18. Comparisons of PSDs are plotted in Figure 4.9 and Figure 4.10. Lines in these two figures are only for trend indication.

Table 4.18 Summary of PSDs of four testing aerosols obtained from four different instruments

	LS13 320		LA-300		LS230		CCM 3	
	MMD (μm)	GSD	MMD (μm)	GSD	MMD (μm)	GSD	MMD (μm)	GSD
Limestone	7.50[1]	3.07	12.29	1.83	8.11	3.15	8.56	1.72
Starch	13.31	1.59	16.78	1.50	14.38	1.55	14.32	1.33
#3 Micro aluminum	5.28	1.98	7.62	1.56	5.37	1.93	5.03	1.42
#5 Micro aluminum	7.09	1.69	8.38	1.49	7.21	1.71	6.31	1.39

[1] Both MMD and GSD presented here are average values of three replicates.

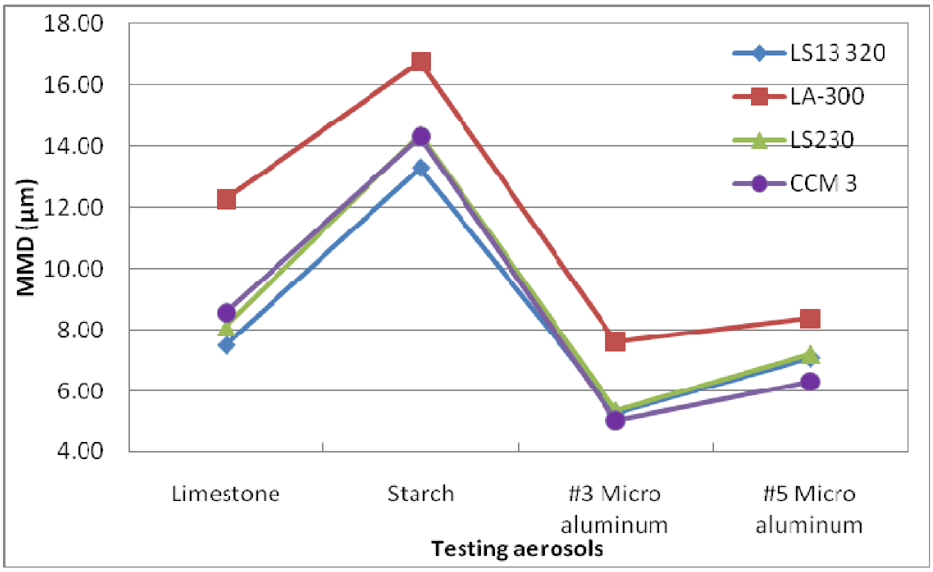

Figure 4.9 Comparison of MMDs of four testing aerosols measured by the four instruments

From Figure 4.9, there was significant but constant difference in MMDs obtained from LA-300 at UIUC and other three instruments (LS13 320, LS230 and CCM). Two laser diffraction type analyzers (LS13 320 at NCSU and LS230 at USDA) provided very similar results for MMDs. In general, LA-300 provided larger MMDs than other three instruments. No significant difference in MMDs among other three instruments (LS13 320, LS230 and CCM) was observed.

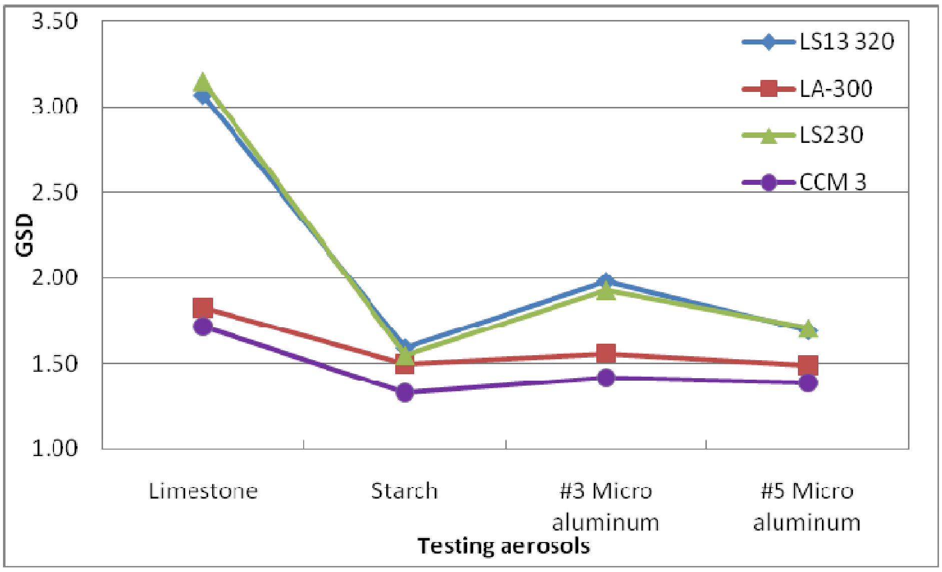

Figure 4.10 Comparison of GSDs of four testing aerosols measured by the four instruments

From Figure 4.10, differences in GSDs obtained from different instruments were observed. In general, laser diffraction series particle size analyzers (LS13 320 at NCSU and LS230 at USDA) provided quite similar GSDs. Small but consistent difference in GSDs was observed between CCM 3 at USDA and LA-300 at UIUC. However, significant differences in GSDs were observed between LS series and CCM 3. In general, LS series provided larger GSDs than CCM 3 and LA-300; LS230 provided larger GSDs than CCM 3.

T-test was performed on the data to further determine if there were significant differences in MMDs and GSDs measured by four different instruments. Significant differences in

MMDs (p=0.014) and GSDs (p=0.079) measured by LS13 320 at NCSU and LA-300 at UIUC were detected at the 0.05 and 0.1 level of significance (α), respectively. No significant differences in PSDs were detected among LS13 320 at NCSU, LS230 at USDA and CCM 3 at USDA. Details about the test are presented in the Appendix H.

The comparative PSD results of testing aerosols measured by different instruments were generally consistent with that of filter-based samples. Differences in PSDs of both filter-based samples and testing aerosols measured by four different instruments at four different locations (LS13 320 at NCSU, LA-300 at UIUC, CCM 3 at TAMU, CCM 3 and LS230 at USDA) are statistically significant but consistent. In general, LA-300 provided the largest MMDs and LS13 320 gave the largest GSDs; the smallest MMDs and GSDs were both provided by CCM 3. LS230 and LS13 320 provided very similar results for both MMDs and GSDs.

LA-300 is a portable laser scattering instrument with much simpler optics and fitting algorithm (Xu, 2009), compared with LS13 320, though both of these two instruments are laser diffraction particle size analyzers. If there are small bumps or peaks in the particle size distribution, LA-300 may not be able to detect. As results indicate, MMDs obtained by LA-300 are larger than that measured by LS13 320, whereas GSDs obtained by LA-300 are smaller than that measured by LS13 320. These differences can be considered as systematic error due to different characteristics of these two laser diffraction particle size analyzers.

Both of LS13 320 and CCM 3 are produced by Beckman Coulter Inc. Xu and Guida (2003) reported that laser diffraction provided much larger mean sizes and broader distributions than electrical sensing zone (ESZ). In this study, LS13 320 provided larger MMDs and GSDs than CCM 3, which is consistent with Xu and Guida (2003) reported. These differences may be systematic error due to different principles that these two instruments rely on.

In operating physical characterization instruments, operators have been shown to be the largest source of uncertainty (Xu, 2009). The instruments were run at different locations by different operators, which may lead to some uncertainty.

LS13 320 and LS230 provided quite similar results for both filter-based samples and testing aerosols (none filter-based samples), though these two instruments were operated by different operators at different locations. This means operators may not be the largest source of uncertainty in this study. Systematic error of instruments because of different characteristics and principles may be the main source that produced the statistically significant differences.

4.6 PM_{10} and $PM_{2.5}$ mass fraction analyses

Mass fractions of PM_{10} and $PM_{2.5}$ were calculated using lognormal distribution function along with MMD and GSD measured by the analyzers. These fractions are often assumed to be the same as measured by the analyzer. This is only true if the PSD follows lognormal distribution. When measuring PSD of a TSP sample, analyzers also measure percentage of PM_{10} and $PM_{2.5}$ in addition to MMD and GSD. Investigation for the differences of PM_{10} and $PM_{2.5}$ fractions between measured and lognormal fitted is a way to test whether a PSD follows a lognormal distribution well. If the relative difference (RD) of PM_{10} or $PM_{2.5}$ mass fraction obtained from two different methods is within 5%, this RD is defined as acceptable error. The RD is defined by Equation (3.5).

4.6.1 Mass fraction analyses of PM_{10} and $PM_{2.5}$ measured by LS13 320 at NCSU

Mass fractions of PM_{10} and $PM_{2.5}$ measured by the laser diffraction particle size analyzer (LS13320) and calculated using lognormal distribution function are presented in Table 4.19 and Table 4.20, respectively.

Table 4.19 Summary of PM_{10} mass fractions obtained from two methods (measured & lognormal fitting values) at NCSU (LS13 320)

Date	Time	Shift	Floor	Winter		
				PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
1/30/2009	10: 00am-3:00pm	Day	1st	37.71	35.93	4.73
			2nd	38.1	37.02	2.83
	3:00pm-8:00pm	Day	1st	37.86	36.65	3.2
			2nd	36.64	34.99	4.5
	8:00pm - 3:00am	Night	1st	35.14	33.49	4.7
			2nd	36.31	34.64	4.6
1/31/2009	3:00am-8:00am	Day	1st	40.9	40.02	2.15
			2nd	38.22	36.97	3.27
	8:00am-2:00pm	Day	1st	37.06	35.83	3.32
			2nd	37.17	35.83	3.61
	2:00pm-8:00pm	Day	1st	39.07	37.96	2.84
			2nd	38.13	36.9	3.23
	8:00pm - 3:00am	Night	1st	40.31	39.16	2.85

			2nd	38.21	36.9	3.43

Table 4.19 Continued

2/1/2009	3:00am-8:00am	Day	1st	39.35	37.95	3.56
			2nd	39.19	38.02	2.99
	8:00am-2:00pm	Day	1st	35.68	34.19	4.18
			2nd	36.81	35.5	3.56
	2:00pm-8:00pm	Day	1st	36.49	34.77	4.71
			2nd	37.27	35.79	3.97
	8:00pm - 3:00am	Night	1st	40.21	39.21	2.49
			2nd	36.82	35.48	3.64
2/2/2009	3:00am-8:00am	Day	1st	38.09	36.44	4.33
			2nd	37.32	35.97	3.62
	8:00am-2:00pm	Day	1st	34.57	33.15	4.11
			2nd	33.65	31.94	5.08
	Mean ± SD (Winter)			37.55±1.73	36.18±1.91	3.67±0.77

				Spring		
Date	Time	Shift	Floor	PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	32.56	31.39	3.59
			2nd	31.47	30.49	3.11
	3:00pm-8:00pm	Day	1st	36.96	36.07	2.41
			2nd	35.27	34.1	3.32
	8:00pm - 3:00am	Night	1st	37.54	36.45	2.9
			2nd	38.37	37.54	2.16
4/4/2009	3:00am-8:00am	Day	1st	37.38	36.09	3.45
			2nd	38.52	37.54	2.54
	8:00am-2:00pm	Day	1st	33.88	32.72	3.42
			2nd	34.54	33.89	1.88

	2:00pm-8:00pm	Day	1st	36.21	34.89	3.65
			2nd	35.82	34.52	3.63
	8:00pm - 3:00am	Night	1st	34.86	33.04	5.22
			2nd	36.6	35.27	3.63

Table 4.19 Continued

4/5/2009	3:00am-8:00am	Day	1st	37.44	36.34	2.94
			2nd	36.65	35.39	3.44
	8:00am-2:00pm	Day	1st	32.85	32.22	1.92
			2nd	27.68	27.06	2.24
	2:00pm-8:00pm	Day	1st	32.27	30.73	4.77
			2nd	28.13	27.23	3.2
	8:00pm - 3:00am	Night	1st	34.12	32.68	4.22
			2nd	31.28	29.76	4.86
4/6/2009	3:00am-8:00am	Day	1st	36.52	35.62	2.46
			2nd	36.36	35.39	2.67
	8:00am-2:00pm	Day	1st	36.51	35.24	3.48
			2nd	34.41	33.62	2.3
Mean ± SD (Spring)				34.78±2.87	33.66±2.85	3.21±0.88
Mean ± SD for both winter and spring				36.16±2.73	34.92±2.71	3.44±0.85

From Table 4.19, overall PM_{10} mass fraction measured by LS13 320 was 36.16±2.73 %, while overall PM_{10} mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 34.92±2.71 %. The relative difference was 3.44±0.85 %, which is acceptable.

Table 4.20 Summary of $PM_{2.5}$ mass fractions obtained from two methods (measured & lognormal fitting values) at NCSU (LS13 320)

Date	Time	Shift	Floor	Winter		
				$PM_{2.5}$ fraction (%) (measured)	$PM_{2.5}$ fraction (%) (lognormal)	RD (%)
1/30/2009	10: 00am-3:00pm	Day	1st	8.42	3.73	55.7
			2nd	8.53	3.8	55.45
	3:00pm-8:00pm	Day	1st	8.3	3.64	56.14
			2nd	8.13	3.37	58.55
	8:00pm - 3:00am	Night	1st	7.89	3.51	55.51
			2nd	8.26	3.38	59.08
1/31/2009	3:00am-8:00am	Day	1st	9.54	4.97	47.9
			2nd	9.05	4.1	54.7
	8:00am-2:00pm	Day	1st	8.17	3.54	56.67
			2nd	8.45	3.67	56.57
	2:00pm-8:00pm	Day	1st	9.03	4.12	54.37
			2nd	8.73	3.9	55.33
	8:00pm - 3:00am	Night	1st	9.09	4.55	49.94
			2nd	8.63	3.88	55.04
2/1/2009	3:00am-8:00am	Day	1st	8.87	4.28	51.75
			2nd	9.19	4.38	52.34
	8:00am-2:00pm	Day	1st	8.07	3.21	60.22
			2nd	8.15	3.42	58.04
	2:00pm-8:00pm	Day	1st	8.05	3.27	59.38
			2nd	8.18	3.54	56.72
	8:00pm - 3:00am	Night	1st	9.8	4.64	52.65
			2nd	8.36	3.35	59.93
2/2/2009	3:00am-8:00am	Day	1st	8.33	3.85	53.78
			2nd	8.21	3.59	56.27

Date	Time	Shift	Floor	PM2.5 fraction (%) (measured)	PM2.5 fraction (%) (lognormal)	RD (%)
	8:00am-2:00pm	Day	1st	7.68	2.92	61.98
			2nd	7.58	2.69	64.51

Table 4.20 Continued

Mean ± SD (Winter)				8.49 ± 0.54	3.74 ± 0.53	56.10 ± 3.63

				Spring		
Date	Time	Shift	Floor	$PM_{2.5}$ fraction (%) (measured)	$PM_{2.5}$ fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	7.64	3.01	60.6
			2nd	7.41	2.17	70.72
	3:00pm-8:00pm	Day	1st	8.89	4.08	54.11
			2nd	8.49	3.36	60.42
	8:00pm - 3:00am	Night	1st	9.1	4.19	53.96
			2nd	8.95	4.11	54.08
4/4/2009	3:00am-8:00am	Day	1st	9.08	4.23	53.41
			2nd	9.5	4.41	53.58
	8:00am-2:00pm	Day	1st	7.87	2.95	62.52
			2nd	8.34	3.14	62.35
	2:00pm-8:00pm	Day	1st	8.09	3.36	58.47
			2nd	8.28	3.26	60.63
	8:00pm - 3:00am	Night	1st	8.15	3.27	59.88
			2nd	8.6	3.77	56.16
4/5/2009	3:00am-8:00am	Day	1st	8.94	4.17	53.36
			2nd	9.11	4.06	55.43
	8:00am-2:00pm	Day	1st	8.04	2.67	66.79
			2nd	7.3	1.66	77.26
	2:00pm-8:00pm	Day	1st	8.48	4.08	51.89
			2nd	6.5	2.16	66.77
	8:00pm - 3:00am	Night	1st	8.63	3.46	59.91
			2nd	7.61	2.4	68.46
4/6/2009	3:00am-8:00am	Day	1st	8.24	3.47	57.89

			2nd	8.5	3.65	57.06
	8:00am-2:00pm	Day	1st	8.56	3.8	55.61
			2nd	8.24	3.1	62.38

Table 4.20 Continued

Mean ± SD (Spring)	8.33±0.67	3.38±0.73	59.76±6.21
Mean ± SD for both winter and spring	8.40±0.61	3.56±0.66	57.90±5.37

From Table 4.20, overall $PM_{2.5}$ mass fraction measured by LS13 320 was 8.41±0.61 %, while overall $PM_{2.5}$ mass fraction obtained from the lognormal distribution using measured MMD and GSDs in fitting was 3.56±0.66 %. The relative difference was 57.90±5.37 %, which is not acceptable.

4.6.2 Mass fraction analyses of PM_{10} and $PM_{2.5}$ measured by LA-300 at UIUC

Mass fractions of PM_{10} and $PM_{2.5}$ measured by the laser scattering particle size analyzer (LA-300) and calculated using lognormal distribution function are presented in Table 4.21 and Table 4.22.

Table 4.21 Summary of PM_{10} mass fractions obtained from two methods (measured & lognormal fitting values) at UIUC (LA-300)

Date	Time	Shift	Floor	Winter		
				PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
1/30/2009	10: 00am-3:00pm	Day	1st	19.86	19.3	2.82
			2nd	20.61	19.66	4.61
	3:00pm-8:00pm	Day	1st	19.53	17.92	8.24
			2nd	19.73	18.13	8.11

	8:00pm - 3:00am	Night	1st	23.12	22.03	4.71
			2nd	23.31	22.2	4.76

Table 4.21 Continued

1/31/2009	3:00am-8:00am	Day	1st	21.68	20.62	4.89
			2nd	19.62	18.3	6.73
	8:00am-2:00pm	Day	1st	19.04	18.04	5.25
			2nd	21.12	21.39	-1.28
	2:00pm-8:00pm	Day	1st	21.63	21.6	0.14
			2nd	21.13	20.67	2.18
	8:00pm - 3:00am	Night	1st	25.18	24.57	2.42
			2nd	16.96	15.1	10.97
2/1/2009	3:00am-8:00am	Day	1st	23.92	21.61	9.66
			2nd	19.87	20.78	-4.58
	8:00am-2:00pm	Day	1st	19.94	18.98	4.81
			2nd	18.11	19.36	-6.9
	2:00pm-8:00pm	Day	1st	22.56	22.03	2.35
			2nd	20.3	19.63	3.3
	8:00pm - 3:00am	Night	1st	20.32	19.57	3.69
			2nd	22.13	21.01	5.06
2/2/2009	3:00am-8:00am	Day	1st	22.48	21.8	3.02
			2nd	19.06	18.39	3.52
	8:00am-2:00pm	Day	1st	18.23	17.31	5.05
			2nd	19.84	18.83	5.09
	Mean ± SD (Winter)			20.74±1.93	19.96±1.99	3.79±3.88

				Spring		
Date	Time	Shift	Floor	PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	15.62	16.7	-6.91

			2nd	17.3	16.93	2.14
	3:00pm-8:00pm	Day	1st	22.85	15.75	31.07
			2nd	18.73	18.74	-0.05
	8:00pm - 3:00am	Night	1st	21.7	21.33	1.71
			2nd	22.26	22.12	0.63

Table 4.21 Continued

4/4/2009	3:00am-8:00am	Day	1st	19.82	18.96	4.34
			2nd	21.63	20.87	3.51
	8:00am-2:00pm	Day	1st	21.41	21.47	-0.28
			2nd	18.32	17.61	3.88
	2:00pm-8:00pm	Day	1st	20.33	19.71	3.05
			2nd	19.72	18.95	3.9
	8:00pm - 3:00am	Night	1st	21.51	20.32	5.53
			2nd	20.02	19.03	4.95
4/5/2009	3:00am-8:00am	Day	1st	22.35	21.94	1.83
			2nd	21.35	21.18	0.8
	8:00am-2:00pm	Day	1st	17.48	18.62	-6.52
			2nd	17.12	17.16	-0.23
	2:00pm-8:00pm	Day	1st	17.52	17.15	2.11
			2nd	17.12	17.04	0.47
	8:00pm - 3:00am	Night	1st	22.35	21.64	3.18
			2nd	19.41	18.14	6.54
4/6/2009	3:00am-8:00am	Day	1st	31.37	31.47	-0.32
			2nd	22.74	21.88	3.78
	8:00am-2:00pm	Day	1st	22.2	21.78	1.89
			2nd	19.44	18.67	3.96
Mean ± SD (Spring)				20.45±3.05	19.81±3.07	2.88±6.54
Mean ± SD for both winter and spring				20.60±2.53	19.88±2.56	3.34±5.34

From Table 4.21, overall PM_{10} mass fraction measured by LA-300 was 20.60±2.53 %, while overall PM_{10} mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 19.88±2.56 %. The relative difference was 3.34±5.34 %, which is acceptable.

Table 4.22 Summary of $PM_{2.5}$ mass fractions obtained from two methods (measured & lognormal fitting values) at UIUC (LA-300)

Date	Time	Shift	Floor	Winter		
				$PM_{2.5}$ fraction (%) (measured)	$PM_{2.5}$ fraction (%) (lognormal)	RD (%)
1/30/2009	10: 00am-3:00pm	Day	1st	5.45	0.26	95.23
			2nd	5.1	0.24	95.29
	3:00pm-8:00pm	Day	1st	4.47	0.15	96.64
			2nd	4.69	0.14	97.01
	8:00pm - 3:00am	Night	1st	5.37	0.24	95.53
			2nd	5.17	0.15	97.1
1/31/2009	3:00am-8:00am	Day	1st	5.14	0.21	95.91
			2nd	4.35	0.15	96.55
	8:00am-2:00pm	Day	1st	4.2	0.18	95.71
			2nd	4.83	0.5	89.65
	2:00pm-8:00pm	Day	1st	4.66	0.45	90.34
			2nd	5.02	0.31	93.82
	8:00pm - 3:00am	Night	1st	5.79	0.38	93.44
			2nd	3.65	0.05	98.63
2/1/2009	3:00am-8:00am	Day	1st	5.53	0.29	94.76
			2nd	4.57	0.6	86.87
	8:00am-2:00pm	Day	1st	4.69	0.17	96.38
			2nd	4.48	0.68	84.82
	2:00pm-8:00pm	Day	1st	5.14	0.37	92.8

			2nd	4.85	0.23	95.26
	8:00pm - 3:00am	Night	1st	4.41	0.14	96.83
			2nd	4.62	0.16	96.54
	3:00am-8:00am	Day	1st	5.77	0.36	93.76
2/2/2009			2nd	5.02	0.27	94.62
	8:00am-2:00pm	Day	1st	4.38	0.2	95.43
			2nd	4.8	0.19	96.04

Table 4.22 Continued

Mean ± SD (Winter)				4.85±0.50	0.27±0.15	94.42±3.22
					Spring	
Date	Time	Shift	Floor	$PM_{2.5}$ fraction (%) (measured)	$PM_{2.5}$ fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	2.41	0.15	93.78
			2nd	3.79	0.16	95.78
	3:00pm-8:00pm	Day	1st	4.51	0.01	99.78
			2nd	4.03	0.35	91.32
	8:00pm - 3:00am	Night	1st	4.33	0.26	94
			2nd	4.94	0.39	92.11
4/4/2009	3:00am-8:00am	Day	1st	4.5	0.29	93.56
			2nd	4.87	0.27	94.46
	8:00am-2:00pm	Day	1st	5.14	0.5	90.27
			2nd	3.82	0.13	96.6
	2:00pm-8:00pm	Day	1st	3.83	0.13	96.61
			2nd	4.56	0.18	96.05
	8:00pm - 3:00am	Night	1st	4.06	0.05	98.77
			2nd	3.97	0.11	97.23
4/5/2009	3:00am-8:00am	Day	1st	5.18	0.39	92.47
			2nd	4.54	0.37	91.85
	8:00am-2:00pm	Day	1st	3.78	0.33	91.27
			2nd	4.1	0.21	94.88

	2:00pm-8:00pm	Day	1st	3.62	0.45	87.57
			2nd	3.66	0.26	92.9
	8:00pm - 3:00am	Night	1st	4.71	0.28	94.06
			2nd	4.01	0.05	98.75

Table 4.22 Continued

4/6/2009	3:00am-8:00am	Day	1st	5.47	0.13	97.62
			2nd	5.08	0.17	96.65
	8:00am-2:00pm	Day	1st	4.39	0.34	92.26
			2nd	4.26	0.16	96.24
Mean ± SD (Spring)				4.29±0.64	0.24±0.13	94.49±2.93
Mean ± SD for both winter and spring				4.57±0.64	0.25±0.14	94.46±3.05

From Table 4.22, overall $PM_{2.5}$ mass fraction measured by LA-300 analyzer was 4.57±0.64 %, while overall $PM_{2.5}$ mass fraction obtained from the lognormal distribution using measured MMD and GSDs in fitting was 0.25±0.14 %. The relative difference was 94.46±3.05 %.

4.6.3 Mass fraction analyses of PM_{10} measured by CCM 3 at TAMU

Mass fractions of PM_{10} measured by the Coulter Counter multisizer 3 (CCM 3) at TAMU for winter samples and calculated using lognormal distribution function are presented in Table 4.23. Mass fractions of $PM_{2.5}$ were not separately measured by the CCM 3 at TAMU, since the lower range of minimum channel was set as 2.95 µm at TAMU, which means all particles less than 2.95 µm were not separated and distributed into the minimum channel.

Table 4.23 Summary of PM_{10} mass fractions obtained from two methods (measured & lognormal fitting values) at TAMU (CCM 3)

Date	Time	Shift	Floor	Winter		
				PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
1/30/2009	10: 00am-3:00pm	Day	1st	41.15	40.03	2.72
			2nd	40.45	39.89	1.38
	3:00pm-8:00pm	Day	1st	40.29	38.84	3.60
			2nd	42.25	41.07	2.79
	8:00pm - 3:00am	Night	1st	42.68	41.70	2.30
			2nd	41.85	41.12	1.74
1/31/2009	3:00am-8:00am	Day	1st	44.75	44.19	1.25
			2nd	42.69	41.92	1.80
	8:00am-2:00pm	Day	1st	27.44	24.15	11.99
			2nd	42.75	42.00	1.75
	2:00pm-8:00pm	Day	1st	40.65	39.62	2.53
			2nd	40.85	39.69	2.84
	8:00pm - 3:00am	Night	1st	43.45	42.41	2.39
			2nd	41.25	40.41	2.04
2/1/2009	3:00am-8:00am	Day	1st	44.85	44.19	1.47
			2nd	44.89	44.07	1.83
	8:00am-2:00pm	Day	1st	41.15	40.34	1.97
			2nd	39.25	38.55	1.78
	2:00pm-8:00pm	Day	1st	39.85	39.13	1.81
			2nd	41.89	40.51	3.29
	8:00pm - 3:00am	Night	1st	48.45	48.22	0.47
			2nd	44.99	44.13	1.91
2/2/2009	3:00am-8:00am	Day	1st	46.75	46.20	1.18
			2nd	40.79	39.85	2.30
	8:00am-2:00pm	Day	1st	40.55	39.66	2.19
			2nd	38.75	37.76	2.55
Mean ± SD				41.72±3.73	40.76±4.20	2.46±2.06

From Table 4.23, overall PM_{10} mass fraction measured by CCM 3 at TAMU was 41.75±3.74 %, while overall PM_{10} mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 40.76±4.19 %. The relative difference was 2.46±2.06 %, which is acceptable.

4.6.4 Mass fraction analyses of PM_{10} and $PM_{2.5}$ measured by CCM 3 at USDA

Mass fractions of PM_{10} and $PM_{2.5}$ measured by the Coulter Counter multisizer 3 (CCM 3) at USDA for spring samples and calculated using lognormal distribution function are presented in Table 4.24 and Table 4.25. Mass fractions of $PM_{2.5}$ were measured by the CCM 3 at USDA, since the lower range of minimum channel was set as 2.00 µm at USDA, which is smaller than 2.5 µm.

Table 4.24 Summary of PM_{10} mass fractions obtained from two methods (measured & lognormal fitting values) at USDA (CCM 3)

Date	Time	Shift	Floor	Spring		
				PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	42.8	42.58	0.51
			2nd	38.6	38.69	-0.24
	3:00pm-8:00pm	Day	1st	43	42.46	1.26
			2nd	40.4	40.28	0.3
	8:00pm - 3:00am	Night	1st	45.9	45.33	1.25
			2nd	42.9	42.41	1.13
4/4/2009	3:00am-8:00am	Day	1st	-	-	-
			2nd	41.9	41.31	1.41
	8:00am-2:00pm	Day	1st	45.3	45.26	0.08
			2nd	38	38.19	-0.5
	2:00pm-8:00pm	Day	1st	39.1	38.64	1.19
			2nd	39.7	39.64	0.15
	8:00pm - 3:00am	Night	1st	44.7	44.3	0.9

			2nd	46.9	46.69	0.45

Table 4.24 Continued

4/5/2009	3:00am-8:00am	Day	1st	41.3	40.55	1.82
			2nd	43.5	42.96	1.24
	8:00am-2:00pm	Day	1st	38.8	38.66	0.37
			2nd	34	34.57	-1.68
	2:00pm-8:00pm	Day	1st	41.7	41.78	-0.19
			2nd	32.7	33.03	-1
	8:00pm - 3:00am	Night	1st	46.3	46.2	0.22
			2nd	38.5	38.56	-0.17
4/6/2009	3:00am-8:00am	Day	1st	41.8	41.45	0.84
			2nd	35.3	35.34	-0.11
	8:00am-2:00pm	Day	1st	43.4	43.25	0.35
			2nd	41.3	41.24	0.13
Mean ± SD (Spring)				41.11±3.68	40.93±3.50	0.39±0.80

From Table 4.24, overall PM_{10} mass fraction measured by CCM 3 at USDA was 41.11±3.68 %, while overall PM_{10} mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 40.93±3.50 %. The relative difference was 0.39±0.80 %, which is excellent.

Table 4.25 Summary of $PM_{2.5}$ mass fractions obtained from two methods (measured & lognormal fitting values) at USDA (CCM 3)

Date	Time	Shift	Floor	Spring $PM_{2.5}$ fraction (%) (measured)	Spring $PM_{2.5}$ fraction (%) (lognormal)	Spring RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	1.57	0.85	45.79
			2nd	1.27	0.45	64.69
	3:00pm-8:00pm	Day	1st	1.32	0.62	53.07
			2nd	1.31	0.51	61.19
	8:00pm - 3:00am	Night	1st	1.81	0.96	47.00
			2nd	1.73	0.80	53.80
4/4/2009	3:00am-8:00am	Day	1st	-	-	-
			2nd	1.51	0.71	52.91
	8:00am-2:00pm	Day	1st	1.63	0.82	49.84
			2nd	1.25	0.35	72.10
	2:00pm-8:00pm	Day	1st	1.45	0.56	61.41
			2nd	1.52	0.47	69.10
	8:00pm - 3:00am	Night	1st	1.80	0.87	51.79
			2nd	1.70	1.04	38.99
4/5/2009	3:00am-8:00am	Day	1st	2.39	0.97	59.49
			2nd	1.31	0.60	54.35
	8:00am-2:00pm	Day	1st	1.24	0.52	57.98
			2nd	1.03	0.31	69.83
	2:00pm-8:00pm	Day	1st	1.40	1.06	24.34
			2nd	1.06	0.28	73.52
	8:00pm - 3:00am	Night	1st	1.62	1.09	32.91
			2nd	1.20	0.50	58.33
4/6/2009	3:00am-8:00am	Day	1st	0.91	0.62	31.68
			2nd	0.63	0.18	71.61
	8:00am-2:00pm	Day	1st	1.44	0.68	52.81
			2nd	1.51	0.52	65.59
Mean ± SD (Spring)				1.43±0.35	0.65±0.26	54.96±13.04

From Table 4.25, overall $PM_{2.5}$ mass fraction measured by CCM 3 at USDA was 1.43±0.35 %, while overall $PM_{2.5}$ mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 0.65±0.26 %. The relative difference was 54.96±13.04 %, which is not acceptable.

4.6.5 Mass fraction analyses of PM_{10} and $PM_{2.5}$ measured by LS230 at USDA

Mass fractions of PM_{10} measured by the laser diffraction particle size analyzer (LS230) at USDA and calculated using lognormal distribution function are presented in Table 4.26 and Table 4.27.

Table 4.26 Summary of PM_{10} mass fractions obtained from two methods (measured & lognormal fitting values) at USDA (LS230)

Date	Time	Shift	Floor	Spring		
				PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	32.41	32.85	-1.36
			2nd	31.46	31.56	-0.32
	3:00pm-8:00pm	Day	1st	35.64	35.11	1.49
			2nd	32.62	32.58	0.12
	8:00pm - 3:00am	Night	1st	34.08	33.39	2.02
			2nd	36.24	35.5	2.04
4/4/2009	3:00am-8:00am	Day	1st	35.11	34.18	2.65
			2nd	36.08	35.57	1.41
	8:00am-2:00pm	Day	1st	33.9	33.65	0.74
			2nd	32.75	32.69	0.18
	2:00pm-8:00pm	Day	1st	34.62	34.26	1.04
			2nd	34.09	33.72	1.09

	8:00pm - 3:00am	Night	1st	36.6	36.11	1.34
			2nd	35.65	34.67	2.75

Table 4.26 Continued

4/5/2009	3:00am-8:00am	Day	1st	36.6	36.11	1.34
			2nd	38.16	35.19	7.78
	8:00am-2:00pm	Day	1st	24.27	25.68	-5.81
			2nd	27.74	24.56	11.46
	2:00pm-8:00pm	Day	1st	32.59	33.2	-1.87
			2nd	28.83	32.91	-14.15
	8:00pm - 3:00am	Night	1st	36.3	35.95	0.96
			2nd	30.58	30.18	1.31
4/6/2009	3:00am-8:00am	Day	1st	- [1]	-	-
			2nd	-	-	-
	8:00am-2:00pm	Day	1st	37.09	36.5	1.59
			2nd	35.59	35.19	1.12
Mean ± SD (Spring)				33.71±3.29	33.39±2.99	0.79±4.45

[1] Sample loading was too light and could not be measured by the analyzer.

From Table 4.26, overall PM_{10} mass fraction measured by LS230 at USDA was 33.71±3.29 %, while overall PM_{10} mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 33.39±2.99 %. The relative difference was 0.79±4.45 %, which is acceptable.

Table 4.27 Summary of $PM_{2.5}$ mass fractions obtained from two methods (measured & lognormal fitting values) at UIUC (LS230)

Date	Time	Shift	Floor	Spring		
				$PM_{2.5}$ fraction (%) (measured)	$PM_{2.5}$ fraction (%) (lognormal)	RD (%)
4/3/2009	10: 00am-3:00pm	Day	1st	7.90	2.84	64.05
			2nd	7.77	2.32	70.14
	3:00pm-8:00pm	Day	1st	8.14	3.27	59.83
			2nd	8.06	2.22	72.46
	8:00pm - 3:00am	Night	1st	7.86	3.11	60.43
			2nd	8.33	3.37	59.54
4/4/2009	3:00am-8:00am	Day	1st	8.82	3.67	58.39
			2nd	8.51	3.64	57.23
	8:00am-2:00pm	Day	1st	8.05	2.95	63.35
			2nd	7.85	2.60	66.88
	2:00pm-8:00pm	Day	1st	8.40	3.45	58.93
			2nd	7.94	2.83	64.36
	8:00pm - 3:00am	Night	1st	9.53	4.06	57.40
			2nd	8.76	3.79	56.74
4/5/2009	3:00am-8:00am	Day	1st	9.53	4.06	57.40
			2nd	9.09	4.60	49.39
	8:00am-2:00pm	Day	1st	5.49	1.09	80.15
			2nd	6.55	1.31	80.00
	2:00pm-8:00pm	Day	1st	8.97	4.52	49.61
			2nd	7.36	5.80	21.20
	8:00pm - 3:00am	Night	1st	8.36	3.41	59.21
			2nd	6.81	2.02	70.34
4/6/2009	3:00am-8:00am	Day	1st	-[1]	-	-
			2nd	-	-	-
	8:00am-2:00pm	Day	1st	9.57	4.38	54.23
			2nd	8.56	3.47	59.46
Mean ± SD (Spring)				8.18±0.96	3.28±1.06	60.44±11.53

[1] Sample loading is too light and could not be measured by the analyzer.

From Table 4.27, overall $PM_{2.5}$ mass fraction measured by LS230 at USDA was 8.18±0.96 %, while overall $PM_{2.5}$ mass fraction obtained from the lognormal distribution fitting using measured MMDs and GSDs in fitting was 3.28±1.06 %. The relative difference was 60.44±11.53 %, which is not acceptable.

The PM_{10} and $PM_{2.5}$ mass fractions obtained from two methods, as well as RD between two methods are summarized in Table 4.28.

Table 4.28 Summary of PM_{10} and $PM_{2.5}$ fractions as well as relative difference between two methods

Location	Model	PM_{10} fraction (%) (measured)	PM_{10} fraction (%) (lognormal)	RD (%)	$PM_{2.5}$ fraction (%) (measured)	$PM_{2.5}$ fraction (%) (lognormal)	RD (%)
NCSU	LS13 320	36.16±2.73	34.92±2.71	3.44±0.85	8.41±0.61	3.56±0.66	57.90±5.37
UIUC	LA-300	20.60±2.53	19.88±2.56	3.34±5.34	4.57±0.64	0.25±0.14	94.46±3.05
TAMU	CCM 3	41.75±3.74	40.76±4.19	2.46±2.06	-	-	-
USDA	CCM 3	41.11±3.68	40.93±3.50	0.39±0.80	1.43±0.35	0.65±0.26	54.96±13.04
	LS230	33.71±3.29	33.39±2.99	0.79±4.45	8.18±0.96	3.28±1.06	60.44±11.53
		RD<5% (PM_{10}), acceptable			RD>>5% ($PM_{2.5}$), not acceptable		

From Table 4.28, it was concluded that all RDs in PM_{10} mass fraction were within 5%, which is acceptable; whereas all RDs in $PM_{2.5}$ mass fraction were much larger than 5%, which is not acceptable.

Since $PM_{2.5}$ mass fractions obtained from two methods (measured by the analyzer & using lognormal distribution equation) did not agree with each other, the fraction of $PM_{2.5}$ obtained by using lognormal distribution equation could not be cited by other researchers based on MMD and GSD.

Chapter 5 Conclusions and Future research

5.1 Conclusions

The major findings of this study were:

1. No significant difference in PSD of PM was observed between the two high-rise layer houses. The average MMD and GSD of PM collected from these two layer houses in fall 2008 was 19.21±1.27 µm and 2.60±0.09, respectively.

2. In-house TSP concentration in winter was higher than in spring. In winter, TSP concentration was affected by both animal activity (daytime vs. nighttime) and floor (1^{st} floor vs. 2^{nd} floor). Animal activity was more significant in affecting TSP concentration than floor. Total suspended particulate concentration was higher during daytime than that at nighttime; TSP concentration was higher on the second floor than that on the first floor. In spring, animal activity was more significant in affecting TSP concentration than equivalent flow rate factor. Total suspended particulate concentration was higher during daytime than that during nighttime; TSP concentration was lower when more fans were on.

3. Compared with PM concentration, PSD (MMD & GSD) was much less affected by the factors of season, animal activity, floor and equivalent flow rate factor. Mass median diameter was affected by floor and equivalent flow rate factor, but the extent of impact was within one standard deviation of MMD. Overall MMD of PM samples collected in winter and spring was 17.13±0.81µm and

18.44±1.44μm, respectively. The difference of MMD among three seasons was within one standard deviation of MMD. Geometric standard deviation was relatively consistent and not affected by those factors. Overall GSD was 2.65±0.08.

4. Significant but consistent differences in MMDs and GSDs of filter-based samples collected in the layer house were detected when comparing PSDs measured with three different particle size analyzers. Uncertainty exists for PSD measurements among different locations by different operators. Different principles and characteristics of these analyzers also led to the differences in PSD measurements. In general, the LA-300 provided the largest MMDs, whereas CCM 3 gave the smallest ones. LS13 320 analyzer provided the largest GSDs, whereas CCM 3 analyzer gave the smallest.
5. The PSD results of testing aerosols measured by different instruments were generally consistent with that of filter-based samples. In general, LA-300 at UIUC provided larger MMDs, whereas LS13 320 at NCSU and LS230 at USDA provided larger GSDs than LA-300 at UIUC and CCM 3 at USDA. It was concluded that the PSD results from two LS series analyzers (LS13 320 and LS230) agreed well with each other.
6. All relative differences in PM_{10} mass fraction obtained by two methods were within 5%, which is acceptable; whereas all RDs in $PM_{2.5}$ mass fraction were much larger than 5%, which is not acceptable.

5.2 Future research

Uncertainties associated with the results of this study are:

1. Particle shape factor (χ) was assumed as 1 in this study. However, real particles emitted from the layer houses are not exactly spherical.

2. Refractive index (RI) set in the laser diffraction particle size analyzer (LS13 320) and laser scattering particle size analyzer (LA-300) are recommended by the manufactures (Beckman Coulter Inc. and Horiba Instrument Ltd.). However, no real refractive index for the agricultural dust is listed in the instrument manuals. What they recommended to use is estimated particle refractive index.

3. Particle density (ρ) was measured as 1.48g/cm^3 using raw dust collected from the blades of fans instead of PM samples in the air for the density measurement. The pycnometer has a sample chamber of 1 cm^3 but the volume of filter-based PM samples could not reach 1 cm^3.

4. Comparison of MMD and GSD obtained from different instruments was used as indicators for PSD comparison. Since PSD does not exactly follow lognormal distribution, uncertainty exists in using MMD and GSD as indicators for PSD. The alternative way for PSD comparison include direct PSD curve fitting (comparison).

Based on the results of this study, the following future research recommendations are:

1. More PM sampling events need to be conducted in summer and fall. Thus, impact of seasonal variation on concentrations and PSDs of PM can be further investigated.
2. Particle shape factor needs to be identified based on the image examination and references.
3. Enough volume of filter-based PM samples needs to be collected for particle density measurement.
4. Further investigation on particle refractive index needs to be conducted.
5. Curve fitting of PSD from different instruments is recommended for further comparison investigation.
6. Since relative differences in $PM_{2.5}$ mass fraction obtained from two methods are all larger than 50%, lognormal distribution equation are not recommended to calculate $PM_{2.5}$ mass fraction. Further research in this issue needs to be explored.

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Appendices

Appendix A Basic operational procedure for the LS13 320

The following procedures present details regarding operating the LS 13 320:

1. Turn on LS 13 320.
2. Check the drain water container and fill water container. Make sure the drain water container is empty and the fill water container is full of clean water.
3. Log in the computer and open ‘LS13 320’ from desktop.
4. After ‘Ready’ appearing on the window, choose Control → Rinse to make sure the instrument is clean before running it.
5. Choose Run → Run Cycle Options to set test conditions as needed. It is recommended to set the following conditions.
6. Choose Run → Run Cycle. If a question mark appears in front of the ‘Measure Background’ box, it means the instrument needs automatic calibration. Click ‘New Sample’ button and check the ‘Enter Run Settings’ box (Both ‘Measure Offsets’ and ‘Align’ boxes will be checked automatically if calibration is needed).
7. Click ‘Sample Information’ button and input all the sample information as needed. Click ‘Run Settings’ button and input all testing conditions as needed.
8. Click ‘Start’ button to start the test.
9. After finishing the background measurement, the software will give out the ‘add sample’ instruction. Pick up the beaker needed when set time is reached. Stir the solution in the beaker with glass rod to ensure particles disperse evenly throughout the solvent. Take some sample solution from the beaker by a dropper.

To guarantee obtaining consistent sample solutions, take sample solutions from the same position within the beaker every time.

10. Inject sample solution into the sample cell slowly, then, you can notice that the obscuration will increase slowly. When the obscuration bar changes color from yellow to green, stop adding sample solution and click 'Start Analysis' button. Notice that the obscuration should be between 8% and 12%.
11. Once testing is done, choose Control → Rinse to make sure the instrument is clean after running it.
12. Close the software 'LS 13 320' and turn off LS.
13. Empty the drain water container and fill the clean water container.

Appendix B Basic operational procedure for sample preparation

1. Put on two latex gloves.
2. Pour water into the sonication bath. Water level must not be higher than ¾ inch (19mm) from top of tank or lower than 2 inches (50mm) from the top.
3. Take the filter-based sample out of the Petri-dish and put it into a 100 milliliters beaker.
4. Obtain 20 milliliters of Ethanol/Methanol and pour in the 100 milliliters beaker.
5. Cover the beaker with a piece of plastic film.
6. Place the beaker containing the sample in the sonication bath.
7. Press “ON/OFF” to start mixing the sample for 15 minutes.
8. After 15 minutes, press “ON/OFF” to stop mixing the sample.
9. The sample is well prepared. Add appropriate sample solution into the cell window of LS 13 320 Laser Diffraction Particle Size Analyzer with a dropper.

Appendix C ANOVA test results for determination of optimal parameters to operate the LS13 320

ANOVA: Two-Factor Without Replication (MMDs)

Source of Variation	SS	df	MS	F	P-value	F crit
Pump speed (%)	5.393686012	2	2.696843006	4.73289364	0.05838989	5.14325285
Running time (s)	1.770667183	3	0.590222394	1.035825893	0.441555681	4.757062664
Error	3.418850975	6	0.569808496			
Total	10.58320417	11				

ANOVA: Two-Factor Without Replication (GSDs)

Source of Variation	SS	df	MS	F	P-value	F crit
Pump speed (%)	0.000954164	2	0.000477082	0.733182114	0.518950859	5.14325285
Running time (s)	0.001928703	3	0.000642901	0.98801395	0.459237015	4.757062664
Error	0.003904201	6	0.0006507			

Appendix D ANOVA results for determination of liquid medium

ANOVA: Single Factor

SUMMARY

Groups	Count	Sum	Average	Variance
Ethanol	5	139.24	27.848	0.19037
Isopropanol	4	111.69	27.9225	2.194625

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between groups	0.012334	1	0.012334	0.011754	0.916708	5.591448
Within groups	7.345355	7	1.049336			
Total	7.357689	8				

ANOVA: Single Factor

SUMMARY

Groups	Count	Sum	Average	Variance
Ethanol	5	14.552	2.9104	0.000759
Isopropanal	4	11.556	2.889	0.010363

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between groups	0.001018	1	0.001018	0.208755	0.661585	5.591448
Within groups	0.034125	7	0.004875			
Total	0.035143	8				

Appendix E ANOVA results for determination of optimal sonication time

ANOVA: Single Factor (MMDs)

SUMMARY

Groups	Count	Sum	Average	Variance
0 (mins)	3	55.3	18.43333333	0.201033333
5 (mins)	3	50.69	16.89666667	0.024433333
15 (mins)	3	49.71	16.57	0.0507
25 (mins)	3	50.92	16.97333333	0.144633333

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	6.180166667	3	2.060055556	19.58227714	0.000482864	4.066180557
Within Groups	0.8416	8	0.1052			
Total	7.021766667	11				

SUMMARY

Groups	Count	Sum	Average	Variance
5 (mins)	3	50.69	16.89666667	0.024433333
15 (mins)	3	49.71	16.57	0.0507
25 (mins)	3	50.92	16.97333333	0.144633333

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	0.275266667	2	0.137633333	1.87881086	0.23249939	5.14325285
Within Groups	0.439533333	6	0.073255556			
Total	0.7148	8				

t-Test: Two-Sample Assuming Equal Variances

	0 (mins)	5 (mins)
Mean	18.43333333	16.89666667
Variance	0.201033333	0.024433333
Observations	3	3
Pooled Variance	0.112733333	
Hypothesized Mean Difference	0	
df	4	
t Stat	5.605303415	
P(T<=t) one-tail	0.00248753	
t Critical one-tail	2.131846782	
P(T<=t) two-tail	0.00497506	
t Critical two-tail	2.776445105	

t-Test: Two-Sample Assuming Equal Variances

	0 (mins)	15 (mins)
Mean	18.43333333	16.57
Variance	0.201033333	0.0507
Observations	3	3
Pooled Variance	0.125866667	
Hypothesized Mean Difference	0	
df	4	
t Stat	6.432515143	
P(T<=t) one-tail	0.001502017	
t Critical one-tail	2.131846782	
P(T<=t) two-tail	0.003004034	
t Critical two-tail	2.776445105	

t-Test: Two-Sample Assuming Equal Variances

	0 (mins)	25 (mins)
Mean	18.43333333	16.97333333
Variance	0.201033333	0.144633333
Observations	3	3
Pooled Variance	0.172833333	
Hypothesized Mean Difference	0	
df	4	

t Stat	4.301151424
P(T<=t) one-tail	0.006317052
t Critical one-tail	2.131846782
P(T<=t) two-tail	0.012634105
t Critical two-tail	2.776445105

ANOVA: Single Factor (GSDs)

SUMMARY

Groups	Count	Sum	Average	Variance
0 (mins)	3	7.857	2.619	0.003463
5 (mins)	3	7.987	2.662333333	0.000102333
15 (mins)	3	7.965	2.655	0.000193
25 (mins)	3	7.879	2.626333333	0.000364333

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	0.004049333	3	0.001349778	1.309616214	0.336724539	4.066180557
Within Groups	0.008245333	8	0.001030667			
Total	0.012294667	11				

Appendix F Comparison of PSDs of PM by three instruments in winter

SAS codes:

```
ods htmL file='MMDs';
data MMDs;
input university$ MMD;
datalines;
NCSU   17.26
NCSU   16.70
NCSU   16.82
NCSU   17.59
NCSU   18.62
NCSU   17.80
NCSU   15.62
NCSU   16.86
NCSU   17.21
NCSU   17.28
NCSU   16.35
NCSU   16.81
NCSU   15.90
NCSU   16.80
NCSU   16.41
NCSU   16.41
NCSU   17.96
NCSU   17.33
NCSU   17.66
NCSU   17.23
NCSU   15.91
NCSU   17.30
NCSU   17.03
NCSU   17.16
NCSU   18.38
NCSU   18.97
UIUC   23.38
UIUC   22.87
UIUC   23.23
UIUC   22.96
UIUC   20.97
UIUC   20.14
UIUC   21.72
UIUC   22.91
UIUC   23.60
UIUC   23.20
UIUC   22.71
UIUC   22.53
UIUC   20.10
UIUC   23.51
UIUC   21.65
```

```
UIUC   24.38
UIUC   22.58
UIUC   26.62
UIUC   21.83
UIUC   22.75
UIUC   21.71
UIUC   21.00
UIUC   21.98
UIUC   24.36
UIUC   24.70
UIUC   23.00
TAMU   14.08
TAMU   14.20
TAMU   14.35
TAMU   13.88
TAMU   13.71
TAMU   13.79
TAMU   13.21
TAMU   13.68
TAMU   18.25
TAMU   13.64
TAMU   14.05
TAMU   14.03
TAMU   13.57
TAMU   13.93
TAMU   13.21
TAMU   13.23
TAMU   13.99
TAMU   14.29
TAMU   14.14
TAMU   13.95
TAMU   12.48
TAMU   13.23
TAMU   12.86
TAMU   14.07
TAMU   14.11
TAMU   14.46
;
proc glm;
      title 'MMD comparison between three universities';
      class university;
      model MMD = university/clparm alpha=0.025;
      estimate 'NCSU vs UIUC' university -1 0 1;
      estimate 'NCSU vs TAMU' university -1 0 1;
      estimate 'UIUC vs TAMU' university -1 0 1;
      /*estimate 'second contrast' university -0.5 -0.5 1;
      means university / tukey;
      model MMD = university;*/
      means university/tukey;
run;
ods htmL end;
```

```
proc glm;
      class university;
      model MMD = university;
      means university / tukey;
run;
proc univariate data=MMDs;
      class university;
run;
title 'paired MMD comparison';
data MMD;
      input NCSU TAMU;
      datalines;
13.99 11.58
13.74 11.68
13.84 11.81
14.47 11.42
15.32 11.28
14.64 11.35
12.85 10.86
13.87 11.26
14.16 15.01
14.22 11.22
13.45 11.56
13.83 11.54
13.08 11.16
13.82 11.46
13.50 10.86
13.50 10.88
14.78 11.51
14.26 11.76
14.53 11.63
14.18 11.48
13.09 10.27
14.23 10.88
14.01 10.58
14.12 11.58
15.12 11.61
15.61 11.89
;
run;
proc ttest;
      paired NCSU*TAMU;
run;
data MMD;
      input NCSU UIUC;
      datalines;
13.99 18.64
13.74 18.25
13.84 18.53
14.47 18.32
15.32 16.82
```

```
14.64 16.20
12.85 17.39
13.87 18.29
14.16 18.80
14.22 18.50
13.45 18.13
13.83 18.00
13.08 16.17
13.82 18.74
13.50 17.34
13.50 19.39
14.78 18.04
14.26 21.08
14.53 17.47
14.18 18.16
13.09 17.38
14.23 16.85
14.01 17.58
14.12 19.37
15.12 19.63
15.61 18.35
;
run;
proc ttest;
      paired NCSU*UIUC;
run;
data MMD;
      input UIUC TAMU;
      datalines;
18.64 11.58
18.25 11.68
18.53 11.81
18.32 11.42
16.82 11.28
16.20 11.35
17.39 10.86
18.29 11.26
18.80 15.01
18.50 11.22
18.13 11.56
18.00 11.54
16.17 11.16
18.74 11.46
17.34 10.86
19.39 10.88
18.04 11.51
21.08 11.76
17.47 11.63
18.16 11.48
17.38 10.27
16.85 10.88
```

```
17.58 10.58
19.37 11.58
19.63 11.61
18.35 11.89
;
run;
proc ttest;
      paired UIUC*TAMU;
run;
ods htmL file='GSDs';
data GSDs;
input university$ GSD;
datalines;
NCSU  2.65
NCSU  2.61
NCSU  2.59
NCSU  2.61
NCSU  2.72
NCSU  2.63
NCSU  2.70
NCSU  2.68
NCSU  2.61
NCSU  2.64
NCSU  2.63
NCSU  2.64
NCSU  2.66
NCSU  2.63
NCSU  2.67
NCSU  2.68
NCSU  2.61
NCSU  2.60
NCSU  2.60
NCSU  2.61
NCSU  2.68
NCSU  2.58
NCSU  2.65
NCSU  2.61
NCSU  2.59
NCSU  2.58
UIUC  2.05
UIUC  2.02
UIUC  1.96
UIUC  1.94
UIUC  1.96
UIUC  1.88
UIUC  1.96
UIUC  1.95
UIUC  2.00
UIUC  2.17
UIUC  2.13
UIUC  2.05
```

```
UIUC  2.01
UIUC  1.84
UIUC  2.02
UIUC  2.26
UIUC  1.96
UIUC  2.37
UIUC  2.06
UIUC  2.01
UIUC  1.91
UIUC  1.91
UIUC  2.06
UIUC  2.08
UIUC  2.05
UIUC  1.99
TAMU  1.88
TAMU  1.87
TAMU  1.91
TAMU  1.92
TAMU  1.86
TAMU  1.84
TAMU  1.84
TAMU  1.87
TAMU  1.84
TAMU  1.84
TAMU  1.80
TAMU  1.80
TAMU  1.87
TAMU  1.85
TAMU  1.85
TAMU  1.87
TAMU  1.87
TAMU  1.82
TAMU  1.78
TAMU  1.88
TAMU  1.87
TAMU  1.87
TAMU  1.92
TAMU  1.84
TAMU  1.85
TAMU  1.80
;
proc glm;
      title 'GSD comparison between three universities';
      class university;
      model GSD = university /clparm alpha=0.05;
      estimate 'NCSU vs UIUC' university -1 0 1;
      estimate 'NCSU vs TAMU' university -1 0 1;
      estimate 'UIUC vs TAMU' university -1 0 1;
      means university / tukey;
run;
ods htmL end;
```

```
proc univariate data=GSDs;
      class university;
run;
title 'paired GSD comparison';
data GSD;
      input NCSU TAMU;
      datalines;
2.63  1.88
2.61  1.87
2.59  1.91
2.61  1.92
2.72  1.86
2.63  1.84
2.70  1.84
2.68  1.87
2.61  1.84
2.64  1.84
2.63  1.80
2.64  1.80
2.66  1.87
2.63  1.85
2.67  1.85
2.68  1.87
2.61  1.87
2.60  1.82
2.60  1.78
2.61  1.88
2.68  1.87
2.58  1.87
2.65  1.92
2.61  1.84
2.59  1.85
2.58  1.80
;
run;
proc ttest;
      paired NCSU*TAMU;
run;
data GSD;
      input NCSU UIUC;
      datalines;
2.63  2.05
2.61  2.02
2.59  1.96
2.61  1.94
2.72  1.96
2.63  1.88
2.70  1.96
2.68  1.95
2.61  2.00
2.64  2.17
```

```
2.63  2.13
2.64  2.05
2.66  2.01
2.63  1.84
2.67  2.02
2.68  2.26
2.61  1.96
2.60  2.37
2.60  2.06
2.61  2.01
2.68  1.91
2.58  1.91
2.65  2.06
2.61  2.08
2.59  2.05
2.58  1.99
;
run;
proc ttest;
      paired NCSU*UIUC;
run;
data GSD;
      input UIUC TAMU;
      datalines;
2.05  1.88
2.02  1.87
1.96  1.91
1.94  1.92
1.96  1.86
1.88  1.84
1.96  1.84
1.95  1.87
2.00  1.84
2.17  1.84
2.13  1.80
2.05  1.80
2.01  1.87
1.84  1.85
2.02  1.85
2.26  1.87
1.96  1.87
2.37  1.82
2.06  1.78
2.01  1.88
1.91  1.87
1.91  1.87
2.06  1.92
2.08  1.84
2.05  1.85
1.99  1.80
;
```

```
run;
proc ttest;
      paired UIUC*TAMU;
run;

ods htmL end;
```

SAS results:

MMD comparison among three universities in winter

The GLM Procedure

Class Level Information		
Class	Levels	Values
university	3	NCSU TAMU UIUC

Number of Observations Read	78
Number of Observations Used	78

Dependent Variable: MMD

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	1024.365138	512.182569	414.4	<.0001
Error	75	92.698012	1.235973		
Corrected Total	77	1117.06315			

R-Square	Coeff Var	Root MSE	MMD Mean
0.917016	6.202	1.111743	17.925

Source	DF	Type I SS	Mean Square	F Value	Pr > F

university	2	1024.365138	512.182569	414.4	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
university	2	1024.365138	512.182569	414.4	<.0001

Parameter	Estimate	Standard Error	t Value	Pr > |t|	97.5% Confidence Limits	
NCSU vs UIUC	5.578	0.30834215	18.09	<.0001	4.8724	6.28296
NCSU vs TAMU	5.578	0.30834215	18.09	<.0001	4.8724	6.28296
UIUC vs TAMU	5.578	0.30834215	18.09	<.0001	4.8724	6.28296

Tukey's Studentized Range (HSD) Test for MMD

Note: This test controls the Type I experimentwise error rate, but it generally has a higher Type II error rate than REGWQ.

Alpha	0.05
Error Degrees of Freedom	75
Error Mean Square	1.236
Critical Value of Studentized Range	3.382
Minimum Significant Difference	0.737

Means with the same letter are not significantly different.			
Tukey Grouping	Mean	N	University
A	22.71	26	UIUC
B	17.13	26	NCSU

C	13.94	26	TAMU

GSD comparison among three universities

The GLM Procedure

Class Level Information		
Class	Levels	Values
university	3	NCSU TAMU UIUC

Number of Observations Read	78
Number of Observations Used	78

Dependent Variable: GSD

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	8.72915641	4.36457821	828.84	<.0001
Error	75	0.39494231	0.0052659		
Corrected Total	77	9.12409872			

R-Square	Coeff Var	Root MSE	GSD Mean
0.956714	3.343881	0.072567	2.170128

Source	DF	Type I SS	Mean Square	F Value	Pr > F
university	2	8.72915641	4.36457821	828.84	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
university	2	8.72915641	4.36457821	828.84	<.0001

Parameter	Estimate	Standard Error	t Value	Pr > |t|	95% Confidence Limits	
NCSU vs UIUC	-0.61	0.02012633	-30.31	<.0001	-0.65009	-0.56991
NCSU vs TAMU	-0.61	0.02012633	-30.31	<.0001	-0.65009	-0.56991

UIUC vs TAMU	-0.61	0.02012633	-30.31	<.0001	-0.65009	-0.56991

Tukey's Studentized Range (HSD) Test for GSD

Note: This test controls the Type I experimentwise error rate, but it generally has a higher Type II error rate than REGWQ.

Alpha	0.05
Error Degrees of Freedom	75
Error Mean Square	0.005266
Critical Value of Studentized Range	3.38156
Minimum Significant Difference	0.0481

Means with the same letter are not significantly different.			
Tukey Grouping	Mean	N	university
A	2.63308	26	NCSU
B	2.02308	26	UIUC
C	1.85423	26	TAMU

Appendix G Comparison of PSDs of PM by four instruments in spring

ANOVA: Single Factor (MMDs)

SUMMARY

Groups	Count	Sum	Average	Variance
LS13 320	23	427.2093539	18.57431973	2.197883219
LA-300	23	527.7705854	22.94654719	3.458783252
LS230	23	425.0468625	18.48029837	1.991206168
CCM 3	23	320.9336593	13.95363736	0.546777996

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	930.2717092	3	310.0905697	151.3624356	1.25462E-34	2.70818651
Within Groups	180.282314	88	2.048662659			
Total	1110.554023	91				

ANOVA: Single Factor (GSDs)

SUMMARY

Groups	Count	Sum	Average	Variance
LS13 320	23	61.397	2.669434783	0.012377348
LA-300	23	46.08	2.003478261	0.019911715
LS230	23	60.93	2.649130435	0.049235573
CCM 3	23	42.32	1.84	0.001113909

ANOVA

Source of Variation	SS	df	MS	F	P-value	F crit
Between Groups	12.82339899	3	4.27446633	206.8993982	9.69809E-40	2.70818651
Within Groups	1.818048	88	0.020659636			
Total	14.64144699	91				

t-Test: Paired Two Sample for Means (MMDs)

	LS13 320	LA-300
Mean	17.12979876	22.7073368

Variance	0.657929945	2.046853711
Observations	26	26
Pearson Correlation	0.140984559	
Hypothesized Mean Difference	0	
df	25	
t Stat	-18.444326	
P(T<=t) one-tail	2.26224E-16	
t Critical one-tail	1.708140745	
P(T<=t) two-tail	4.52449E-16	
t Critical two-tail	2.059538536	

t-Test: Paired Two Sample for Means (GSDs)

	LS13 320	LA-300
Mean	2.633076923	2.022538462
Variance	0.001446154	0.013182738
Observations	26	26
Pearson Correlation	-0.045375873	
Hypothesized Mean Difference	0	
df	25	
t Stat	25.39747027	
P(T<=t) one-tail	1.13056E-19	
t Critical one-tail	1.708140745	
P(T<=t) two-tail	2.26112E-19	
t Critical two-tail	2.059538536	

t-Test: Paired Two Sample for Means (MMDs)

	LS13 320	LS230
Mean	18.57431973	18.48029837
Variance	2.197883219	1.991206168
Observations	23	23
Pearson Correlation	0.739580774	
Hypothesized Mean Difference	0	
df	22	

t Stat	0.4309674	
P(T<=t) one-tail	0.335342654	
t Critical one-tail	1.717144335	
P(T<=t) two-tail	0.670685308	
t Critical two-tail	2.073873058	

t-Test: Paired Two Sample for Means (GSDs)

	LS13 320	LS230
Mean	2.669434783	2.649130435
Variance	0.012377348	0.049235573
Observations	23	23
Pearson Correlation	0.506004633	
Hypothesized Mean Difference	0	
df	22	
t Stat	0.508783071	
P(T<=t) one-tail	0.307984506	
t Critical one-tail	1.717144335	
P(T<=t) two-tail	0.615969012	
t Critical two-tail	2.073873058	

t-Test: Paired Two Sample for Means (MMDs)

	LS13 320	CCM 3
Mean	18.57431973	13.95363736
Variance	2.197883219	0.546777996
Observations	23	23
Pearson Correlation	0.533815886	
Hypothesized Mean Difference	0	
df	22	
t Stat	17.66158911	
P(T<=t) one-tail	8.80464E-15	
t Critical one-tail	1.717144335	
P(T<=t) two-tail	1.76093E-14	
t Critical two-tail	2.073873058	

t-Test: Paired Two Sample for Means (GSDs)

	LS13 320	CCM 3
Mean	2.669434783	1.84
Variance	0.012377348	0.001113909
Observations	23	23
Pearson Correlation	0.713147409	
Hypothesized Mean Difference	0	
df	22	
t Stat	43.94048793	
P(T<=t) one-tail	3.13701E-23	
t Critical one-tail	1.717144335	
P(T<=t) two-tail	6.27402E-23	
t Critical two-tail	2.073873058	

t-Test: Paired Two Sample for Means (MMDs)

	LA-300	CCM 3
Mean	22.94654719	13.95363736
Variance	3.458783252	0.546777996
Observations	23	23
Pearson Correlation	0.500438226	
Hypothesized Mean Difference	0	
df	22	
t Stat	26.59845004	
P(T<=t) one-tail	1.60093E-18	
t Critical one-tail	1.717144335	
P(T<=t) two-tail	3.20186E-18	
t Critical two-tail	2.073873058	

t-Test: Paired Two Sample for Means (GSDs)

	LA-300	CCM 3
Mean	2.003478261	1.84

Variance	0.019911715	0.001113909
Observations	23	23
Pearson Correlation	0.199903383	
Hypothesized Mean Difference	0	
df	22	
t Stat	5.666594757	
P(T<=t) one-tail	5.33306E-06	
t Critical one-tail	1.717144335	
P(T<=t) two-tail	1.06661E-05	
t Critical two-tail	2.073873058	

Appendix H Comparison of PSDs of testing aerosols by four instruments

t-Test: Paired Two Sample for Means (MMDs)

	LS13 320	LS230
Mean	8.295	8.7675
Variance	12.10816667	15.300425
Observations	4	4
Pearson Correlation	0.998936269	
Hypothesized Mean Difference	0	
df	3	
t Stat	-2.035713086	
P(T<=t) one-tail	0.067300504	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.134601007	
t Critical two-tail	3.182446305	

t-Test: Paired Two Sample for Means (MMDs)

	LS13 320	LA-300
Mean	8.295	11.2675
Variance	12.10816667	17.69169167
Observations	4	4
Pearson Correlation	0.940803719	
Hypothesized Mean Difference	0	
df	3	
t Stat	-3.954066779	
P(T<=t) one-tail	0.014433399	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.028866798	
t Critical two-tail	3.182446305	

t-Test: Paired Two Sample for Means (MMDs)

	LS13 320	CCM 3
Mean	8.295	8.555
Variance	12.10816667	16.9003

Observations	4	4
Pearson Correlation	0.984288524	
Hypothesized Mean Difference	0	
df	3	
t Stat	-0.56466172	
P(T<=t) one-tail	0.305884811	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.611769622	
t Critical two-tail	3.182446305	

t-Test: Paired Two Sample for Means (MMDs)

	LA-300	CCM 3
Mean	11.2675	8.555
Variance	17.69169167	16.9003
Observations	4	4
Pearson Correlation	0.985544766	
Hypothesized Mean Difference	0	
df	3	
t Stat	7.604289244	
P(T<=t) one-tail	0.002359756	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.004719512	
t Critical two-tail	3.182446305	

t-Test: Paired Two Sample for Means (GSDs)

	LS13 320	LS230
Mean	2.0825	2.085
Variance	0.460758333	0.528366667
Observations	4	4
Pearson Correlation	0.998670745	
Hypothesized Mean Difference	0	
df	3	
t Stat	-0.08304548	
P(T<=t) one-tail	0.469523101	
t Critical one-tail	2.353363435	

P(T<=t) two-tail	0.939046202
t Critical two-tail	3.182446305

t-Test: Paired Two Sample for Means (GSDs)

	LS13 320	LA-300
Mean	2.0825	1.595
Variance	0.460758333	0.0255
Observations	4	4
Pearson Correlation	0.995285477	
Hypothesized Mean Difference	0	
df	3	
t Stat	1.874682265	
P(T<=t) one-tail	0.078760786	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.157521572	
t Critical two-tail	3.182446305	

t-Test: Paired Two Sample for Means (GSDs)

	LS13 320	CCM 3
Mean	2.0825	1.465
Variance	0.460758333	0.0303
Observations	4	4
Pearson Correlation	0.994019965	
Hypothesized Mean Difference	0	
df	3	
t Stat	2.440128589	
P(T<=t) one-tail	0.046244814	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.092489628	
t Critical two-tail	3.182446305	

t-Test: Paired Two Sample for Means (GSDs)

	LA-300	CCM 3
Mean	1.595	1.465
Variance	0.0255	0.0303
Observations	4	4
Pearson Correlation	0.985732688	
Hypothesized Mean Difference	0	
df	3	
t Stat	8.221921916	
P(T<=t) one-tail	0.001883055	
t Critical one-tail	2.353363435	
P(T<=t) two-tail	0.00376611	
t Critical two-tail	3.182446305	

Appendix I Statistical investigation for factors impacting TSP concentration in the layer houses

SAS codes:

```
ods htmL file='Zihan1';
Data Zihan1;
      input concentration season$ time$ floor$ fan;
      if season="winter" then winter=1; else winter=0;
      if time="day" then day=1; else day=0;
      if floor="first" then first=1; else first =0;
      datalines;
4299.34 winter day first 1.39
5055.57 winter day second 0.00
4031.44 winter day first 1.27
6104.29 winter day second 0.00
935.16 winter night first 1.01
1665.10 winter night second 0.00
3466.89 winter day first 1.15
5157.55 winter day second 0.00
4216.89 winter day first 1.27
5627.72 winter day second 0.00
3815.31 winter day first 1.17
5556.83 winter day second 0.00
914.21 winter night first 1.08
1658.19 winter night second 0.00
2854.45 winter day first 1.24
5077.11 winter day second 0.00
4233.76 winter day first 1.95
5000.95 winter day second 0.00
3832.53 winter day first 1.56
5507.21 winter day second 0.00
815.46 winter night first 1.19
1397.14 winter night second 0.00
3208.67 winter day first 1.28
5252.84 winter day second 0.00
4003.69 winter day first 1.92
4984.72 winter day second 0.00
2131.50 spring day first 7.97
3036.42 spring day second 1.23
2126.74 spring day first 5.36
2559.80 spring day second 0.38
1120.05 spring night first 1.30
1307.01 spring night second 0.00
3941.61 spring day first 1.66
4155.28 spring day second 0.00
2746.65 spring day first 6.47
```

```
3051.51 spring day second 0.92
2451.92 spring day first 5.27
2343.30 spring day second 1.54
1253.97 spring night first 1.11
1395.06 spring night second 0.00
3968.71 spring day first 1.85
4267.43 spring day second 0.00
2692.13 spring day first 8.51
4065.28 spring day second 2.69
1298.97 spring day first 7.92
1990.40 spring day second 3.79
851.89 spring night first 3.69
871.14 spring night second 0.00
1314.85 spring day first 7.03
1451.93 spring day second 0.15
1868.83 spring day first 5.84
2416.70 spring day second 0.13
;

proc glm data=Zihan1;
      model concentration=winter day first fan;
run;

proc glm data=Zihan1;
      model concentration=winter day fan;
      output out=zihanout r=r p=p;
run;

/*proc gplot;
title2 "diagnostic plot, residuals vs predicted";
title3 " ";
plot r*p;
symbol v=dot height=.2;
run; quit;

proc gplot;
title2 "diagnostic plot, residuals vs fan";
title3 " ";
plot r*fan;
symbol v=dot height=.2;
run; quit;*/

proc univariate plots data = zihanout;
var r;
run;
proc univariate data=;
      var r p;
      qqplot;
proc univariate data=zihanout;
      qqplot r p;
run;
```

```
proc univariate data=zihanout;
      qqplot r / cframe=ligr;
run;
proc univariate data=zihanout;
      qqplot r / normal (mu=est sigma=est);
run;
proc univariate data=Zihan1;
      qqplot concentration / normal (mu=est sigma=est);
run;
ods htmL end;
```

```
ods htmL file='Zihan1';
Data Zihan1;
      input concentration season$ time$ floor$ fan;
      /*if season="winter" then winter=1; else winter=0;*/
      if time="day" then day=1; else day=0;
      if floor="first" then first=1; else first =0;
      datalines;
4299.34 winter day first 1.39
5055.57 winter day second 0.00
4031.44 winter day first 1.27
6104.29 winter day second 0.00
935.16 winter night first 1.01
1665.10 winter night second 0.00
3466.89 winter day first 1.15
5157.55 winter day second 0.00
4216.89 winter day first 1.27
5627.72 winter day second 0.00
3815.31 winter day first 1.17
5556.83 winter day second 0.00
914.21 winter night first 1.08
1658.19 winter night second 0.00
2854.45 winter day first 1.24
5077.11 winter day second 0.00
4233.76 winter day first 1.95
5000.95 winter day second 0.00
3832.53 winter day first 1.56
5507.21 winter day second 0.00
815.46 winter night first 1.19
1397.14 winter night second 0.00
3208.67 winter day first 1.28
5252.84 winter day second 0.00
4003.69 winter day first 1.92
4984.72 winter day second 0.00

;
```

```
proc glm data=Zihan1;
      model concentration=/*winter*/ day first fan;
run;

proc glm data=Zihan1;
      model concentration=/*winter*/ day first;
      output out=zihanout r=r p=p;
run;


ods htmL end;


ods htmL file='Zihan1';
Data Zihan1;
      input concentration season$ time$ floor$ fan;
      /*if season="winter" then winter=1; else winter=0;*/
      if time="day" then day=1; else day=0;
      if floor="first" then first=1; else first =0;
      datalines;

2131.50 spring day first 7.97
3036.42 spring day second 1.23
2126.74 spring day first 5.36
2559.80 spring day second 0.38
1120.05 spring night first 1.30
1307.01 spring night second 0.00
3941.61 spring day first 1.66
4155.28 spring day second 0.00
2746.65 spring day first 6.47
3051.51 spring day second 0.92
2451.92 spring day first 5.27
2343.30 spring day second 1.54
1253.97 spring night first 1.11
1395.06 spring night second 0.00
3968.71 spring day first 1.85
4267.43 spring day second 0.00
2692.13 spring day first 8.51
4065.28 spring day second 2.69
1298.97 spring day first 7.92
1990.40 spring day second 3.79
851.89 spring night first 3.69
871.14 spring night second 0.00
1314.85 spring day first 7.03
1451.93 spring day second 0.15
1868.83 spring day first 5.84
2416.70 spring day second 0.13
;

proc glm data=Zihan1;
      model concentration=/*winter*/ day first fan;
```

```
run;

proc glm data=Zihan1;
      model concentration=/*winter*/ day fan;
      output out=zihanout r=r p=p;
run;
ods htmL end;
```

SAS results:

The SAS System

The GLM Procedure

Number of Observations Read	52
Number of Observations Used	52

Dependent Variable: concentration

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	96150373.6	24037593.4	40.94	<.0001
Error	47	27598762.2	587207.7		
Corrected Total	51	123749136			

R-Square	Coeff Var	Root MSE	Concentration Mean
0.776978	25.00584	766.2948	3064.463

Source	DF	Type I SS	Mean Square	F Value	Pr > F
winter	1	27760374.6	27760374.6	47.28	<.0001
day	1	55279371.6	55279371.6	94.14	<.0001
first	1	9788315.46	9788315.46	16.67	2E-04
fan	1	3322311.95	3322311.95	5.66	0.022

Source	DF	Type III SS	Mean Square	F Value	Pr > F
winter	1	9964506.99	9964506.99	16.97	2E-04
day	1	57910219.8	57910219.8	98.62	<.0001
first	1	1184636.22	1184636.22	2.02	0.162

fan	1	3322311.95	3322311.95	5.66	0.022

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	974.2367	270.034702	3.61	0.0007
winter	1088.164	264.157192	4.12	0.0002
day	2665.645	268.423515	9.93	<.0001
first	-408.019	287.265841	-1.42	0.1621
fan	-169.225	71.1443052	-2.38	0.0215

The SAS System

The GLM Procedure

Number of Observations Read	52
Number of Observations Used	52

Dependent Variable: concentration

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	94965737.4	31655245.8	52.79	<.0001
Error	48	28783398.4	599654.1		
Corrected Total	51	123749136			

R-Square	Coeff Var	Root MSE	concentration Mean
0.767405	25.26946	774.3734	3064.463

Source	DF	Type I SS	Mean Square	F Value	Pr > F
winter	1	27760374.6	27760374.6	46.29	<.0001
day	1	55279371.6	55279371.6	92.19	<.0001
fan	1	11925991.2	11925991.2	19.89	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F

winter	1	8815401.18	8815401.18	14.7	4E-04
day	1	65244804	65244804	108.8	<.0001
fan	1	11925991.2	11925991.2	19.89	<.0001

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	898.32084	267.482762	3.36	0.0015
winter	938.25887	244.710189	3.83	0.0004
day	2753.4183	263.967006	10.43	<.0001
fan	-237.20931	53.1906007	-4.46	<.0001

The SAS System

The UNIVARIATE Procedure

Variable: r

Moments			
N	52	Sum Weights	52
Mean	0	Sum Observations	0
Std Deviation	751.252528	Variance	564380.36
Skewness	-0.4277642	Kurtosis	0.0102575
Uncorrected SS	28783398.4	Corrected SS	28783398
Coeff Variation	.	Std Error Mean	104.17998

Basic Statistical Measures			
Location		Variability	
Mean	0	Std Deviation	751.25253
Median	5.941037	Variance	564380
Mode	.	Range	3679
		Interquartile Range	1077

Tests for Location: Mu0=0				
Test	Statistic		p Value	
Student's t	t	0	Pr > |t|	1
Sign	M	0	Pr >= |M|	1
Signed Rank	S	29	Pr >= |S|	0.7946

Quantiles (Definition 5)	
Quantile	Estimate
100% Max	1514.29198
99%	1514.29198
95%	1051.63388
90%	917.21198
75% Q3	591.62141
50% Median	5.94104
25% Q1	-485.6123
10%	-943.13682
5%	-1204.20194
1%	-2164.22776
0% Min	-2164.22776

Extreme Observations			
Lowest		Highest	
Value	Obs	Value	Obs
-2164.23	50	966.832	12
-1441.41	15	1037.722	10
-1204.2	52	1051.634	44
-1077.7	23	1059.042	43
-1001.8	30	1514.292	4

The UNIVARIATE Procedure

Variable: r

```
                        Stem Leaf                     #
Boxplot
                          14 1                        1
|
                          12
|
                          10 456                      3
|
                           8 327                      3
|
                           6 223686                   6
|
                           4 11790037                 8             +-
----+
                           2 79                       2             |
|
                           0 451                      3             *-
-+--*
                          -0 87373                    5             |
|
                          -2 98265                    5             |
|
                          -4 0740                     4             +-
----+
                          -6 64776                    5
|
                          -8 45                       2
|
                         -10 80                       2
|
                         -12 0                        1
|
                         -14 4                        1
|
                         -16
                         -18
                         -20 6                        1
0
                             ----+----+----+----+
                         Multiply Stem.Leaf by 10**+2
```

```
                                         Normal Probability Plot
                      1500+
+++ *
```

```
                            |
+++
                            |                                                +**
*
                            |                                             ***
                            |                                         *****
                            |                                     ****+
                            |                                    *+++
                            |                                  **+
                            |                                ***
                       -300+                              ***
                            |                           **
                            |                       ****
                            |                     +**
                            |                   *+*
                            |                ++*
                            |             ++*
                            |   ++
                            |++
                      -2100+  *
                             +----+----+----+----+----+----+----+----+-
---+----+
                                 -2        -1         0         +1
+2
```

The SAS System

The UNIVARIATE Procedure
Variable: concentration

Moments			
N	52	Sum Weights	52
Mean	3064.4635	Sum Observations	159352.1
Std Deviation	1557.7078	Variance	2426453.6
Skewness	0.1702761	Kurtosis	-1.2270217
Uncorrected SS	612077824	Corrected SS	123749136
Coeff Variation	50.831339	Std Error Mean	216.01521

Basic Statistical Measures

Location		Variability	
Mean	3064.463	Std Deviation	1558
Median	2945.435	Variance	2426454
Mode	.	Range	5289
		Interquartile Range	2670

Tests for Location: Mu0=0				
Test	Statistic		p Value	
Student's t	t	14.18633	Pr > |t|	<.0001
Sign	M	26	Pr >= |M|	<.0001
Signed Rank	S	689	Pr >= |S|	<.0001

Quantiles (Definition 5)	
Quantile	Estimate
100% Max	6104.29
99%	6104.29
95%	5556.83
90%	5157.55
75% Q3	4225.33
50% Median	2945.44
25% Q1	1555.06
10%	1120.05
5%	871.14
1%	815.46
0% Min	815.46

Extreme Observations			
Lowest		Highest	
Value	Obs	Value	Obs
815.46	21	5252.84	24
851.89	47	5507.21	20
871.14	48	5556.83	12
914.21	13	5627.72	10
935.16	5	6104.29	4

The SAS System (Winter model)

The GLM Procedure

Number of Observations Read	26
Number of Observations Used	26

Dependent Variable: concentration

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	63013315.9	21004438.7	113.59	<.0001
Error	22	4068174.57	184917.03		
Corrected Total	25	67081490.5			

R-Square	Coeff Var	Root MSE	concentration Mean
0.939355	11.33087	430.0198	3795.116

Source	DF	Type I SS	Mean Square	F Value	Pr > F
day	1	51287528.4	51287528.4	277.35	<.0001
first	1	11667943.1	11667943.1	63.1	<.0001
fan	1	57844.54	57844.54	0.31	0.5816

Source	DF	Type III SS	Mean Square	F Value	Pr > F
day	1	44150205.2	44150205.2	238.76	<.0001
first	1	1330656.53	1330656.53	7.2	0.0136
fan	1	57844.54	57844.54	0.31	0.5816

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	1932.22369	202.713839	9.53	<.0001
day	3292.631203	213.091034	15.45	<.0001
first	-1676.34063	624.910378	-2.68	0.0136
fan	250.28651	447.501732	0.56	0.5816

Number of Observations Read	26
Number of Observations Used	26

Dependent Variable: concentration

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	62955471.4	31477735.7	175.47	<.0001
Error	23	4126019.11	179392.14		
Corrected Total	25	67081490.5			

R-Square	Coeff Var	Root MSE	concentration Mean
0.938492	11.16032	423.5471	3795.116

Source	DF	Type I SS	Mean Square	F Value	Pr > F
day	1	51287528.4	51287528.4	285.9	<.0001
first	1	11667943.1	11667943.1	65.04	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
day	1	51287528.4	51287528.4	285.9	<.0001
first	1	11667943.1	11667943.1	65.04	<.0001

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	1900.777436	191.829056	9.91	<.0001
day	3333.511333	197.15044	16.91	<.0001
first	-1339.801538	166.128836	-8.06	<.0001

The SAS System (Spring model)

The GLM Procedure

Number of Observations Read	26

Number of Observations Used	26

Dependent Variable: concentration

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	3	16864711.5	5621570.5	10.27	0.0002
Error	22	12042559.2	547389.06		
Corrected Total	25	28907270.7			

R-Square	Coeff Var	Root MSE	concentration Mean
0.583407	31.70169	739.8575	2333.811

Source	DF	Type I SS	Mean Square	F Value	Pr > F
day	1	11243686.2	11243686.2	20.54	0.0002
first	1	1017499.04	1017499.04	1.86	0.1866
fan	1	4603526.21	4603526.21	8.41	0.0083

Source	DF	Type III SS	Mean Square	F Value	Pr > F
day	1	15822072.8	15822072.8	28.9	<.0001
first	1	958292.08	958292.08	1.75	0.1994
fan	1	4603526.21	4603526.21	8.41	0.0083

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	1082.77619	345.849721	3.13	0.0049
day	2145.14404	398.999935	5.38	<.0001
first	592.026016	447.445246	1.32	0.1994
fan	-241.57626	83.302301	-2.9	0.0083

Number of Observations Read	26
Number of Observations Used	26

Dependent Variable: concentration

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	15906419.4	7953209.7	14.07	0.0001
Error	23	13000851.3	565254.4		
Corrected Total	25	28907270.7			

R-Square	Coeff Var	Root MSE	concentration Mean
0.550257	32.21487	751.834	2333.811

Source	DF	Type I SS	Mean Square	F Value	Pr > F
day	1	11243686.2	11243686.2	19.89	0.0002
fan	1	4662733.17	4662733.17	8.25	0.0086

Source	DF	Type III SS	Mean Square	F Value	Pr > F
day	1	15218783.3	15218783.3	26.92	<.0001
fan	1	4662733.17	4662733.17	8.25	0.0086

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	1293.49618	311.96879	4.15	0.0004
day	1942.21657	374.308479	5.19	<.0001
fan	-157.681486	54.9012711	-2.87	0.0086

Appendix J Statistical investigation for factors impacting PSDs of PM in the layer houses

SAS codes:

```
ods htmL file='Zihan2';
Data Zihan2;
      input MMD season$ time$ floor$ fan;
      if season="winter" then winter=1; else winter=0;
      if time="day" then day=1; else day=0;
      if floor="first" then first=1; else first =0;
      datalines;
17.26 winter      day   first 1.39
16.70 winter      day   second      0.00
16.82 winter      day   first 1.27
17.59 winter      day   second      0.00
18.62 winter      night first 1.01
17.80 winter      night second      0.00
15.62 winter      day   first 1.15
16.86 winter      day   second      0.00
17.21 winter      day   first 1.27
17.28 winter      day   second      0.00
16.35 winter      day   first 1.17
16.81 winter      day   second      0.00
15.90 winter      night first 1.08
16.80 winter      night second      0.00
16.41 winter      day   first 1.24
16.41 winter      day   second      0.00
17.96 winter      day   first 1.95
17.33 winter      day   second      0.00
17.66 winter      day   first 1.56
17.23 winter      day   second      0.00
15.91 winter      night first 1.19
17.30 winter      night second      0.00
17.03 winter      day   first 1.28
17.16 winter      day   second      0.00
18.38 winter      day   first 1.92
18.97 winter      day   second      0.00
19.67 spring      day   first 7.97
19.41 spring      day   second      1.23
17.36 spring      day   first 5.36
18.12 spring      day   second      0.38
17.19 spring      night first 1.30
16.56 spring      night second      0.00
17.42 spring      day   first 1.66
16.68 spring      day   second      0.00
18.69 spring      day   first 6.47
```

```
18.09 spring       day   second       0.92
17.64 spring       day   first 5.27
17.79 spring       day   second       1.54
18.74 spring       night first 1.11
17.66 spring       night second       0.00
17.25 spring       day   first 1.85
17.75 spring       day   second       0.00
18.77 spring       day   first 8.51
21.22 spring       day   second       2.69
21.35 spring       day   first 7.92
21.98 spring       day   second       3.79
19.14 spring       night first 3.69
20.22 spring       night second       0.00
17.30 spring       day   first 7.03
17.52 spring       day   second       0.15
17.70 spring       day   first 5.84
18.23 spring       day   second       0.13
;
proc glm data=Zihan2;
      model MMD=winter day first fan;
run;
proc glm data=Zihan2;
      model MMD=first fan;
run;
proc univariate data=Zihan2;
      qqplot MMD / normal (mu=est sigma=est);
run;
ods htmL end;


ods htmL file='Zihan3';
Data Zihan3;
      input GSD season$ time$ floor$ fan;
      if season="winter" then winter=1; else winter=0;
      if time="day" then day=1; else day=0;
      if floor="first" then first=1; else first =0;
      datalines;
2.65 winter day first 1.39
2.61 winter day second 0.00
2.59 winter day first 1.27
2.61 winter day second 0.00
2.72 winter night first 1.01
2.63 winter night second 0.00
2.70 winter day first 1.15
2.68 winter day second 0.00
2.61 winter day first 1.27
2.64 winter day second 0.00
2.63 winter day first 1.17
2.64 winter day second 0.00
2.66 winter night first 1.08
2.63 winter night second 0.00
```

```
2.67 winter day first 1.24
2.68 winter day second 0.00
2.61 winter day first 1.95
2.60 winter day second 0.00
2.60 winter day first 1.56
2.61 winter day second 0.00
2.68 winter night first 1.19
2.58 winter night second 0.00
2.65 winter day first 1.28
2.61 winter day second 0.00
2.59 winter day first 1.92
2.58 winter day second 0.00
2.70 spring day first 7.97
2.50 spring day second 1.23
2.72 spring day first 5.36
2.65 spring day second 0.38
2.72 spring night first 1.30
2.65 spring night second 0.00
2.75 spring day first 1.66
2.71 spring day second 0.00
2.62 spring day first 6.47
2.61 spring day second 0.92
2.61 spring day first 5.27
2.61 spring day second 1.54
2.68 spring night first 1.11
2.69 spring night second 0.00
2.73 spring day first 1.85
2.75 spring day second 0.00
2.57 spring day first 8.51
2.49 spring day second 2.69
3.06 spring day first 7.92
2.66 spring day second 3.79
2.75 spring night first 3.69
2.61 spring night second 0.00
2.61 spring day first 7.03
2.66 spring day second 0.15
2.70 spring day first 5.84
2.61 spring day second 0.13
;
proc glm data=Zihan3;
      model GSD=winter day first fan;
run;
proc glm data=Zihan3;
      model GSD=winter /*day*/ first;
run;
proc univariate data=Zihan3;
      qqplot GSD / normal (mu=est sigma=est);
run;
ods htmL end;
```

SAS results:

The SAS System

The GLM Procedure

Number of Observations Read	52
Number of Observations Used	52

Dependent Variable: MMD

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	36.3354549	9.08386373	7.84	<.0001
Error	47	54.4546451	1.15860947		
Corrected Total	51	90.7901			

R-Square	Coeff Var	Root MSE	MMD Mean
0.400214	6.052219	1.076387	17.785

Source	DF	Type I SS	Mean Square	F Value	Pr > F
winter	1	22.3355077	22.3355077	19.28	<.0001
day	1	0.27044333	0.27044333	0.23	0.631
first	1	0.72027692	0.72027692	0.62	0.434
fan	1	13.009227	13.009227	11.23	0.002

Source	DF	Type III SS	Mean Square	F Value	Pr > F
winter	1	2.75710414	2.75710414	2.38	0.13
day	1	0.55591009	0.55591009	0.48	0.492
first	1	9.32995081	9.32995081	8.05	0.007
fan	1	13.009227	13.009227	11.23	0.002

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	18.25031	0.37930821	48.11	<.0001

winter	-0.572391	0.37105228	-1.54	0.1296
day	-0.261172	0.37704503	-0.69	0.4919
first	-1.145059	0.40351218	-2.84	0.0067
fan	0.334865	0.09993389	3.35	0.0016

Number of Observations Read	52
Number of Observations Used	52

Dependent Variable: MMD

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	32.3548792	16.1774396	13.57	<.0001
Error	49	58.4352208	1.19255553		
Corrected Total	51	90.7901			

R-Square	Coeff Var	Root MSE	MMD Mean
0.35637	6.140241	1.092042	17.785

Source	DF	Type I SS	Mean Square	F Value	Pr > F
first	1	0.72027692	0.72027692	0.6	0.441
fan	1	31.6346023	31.6346023	26.53	<.0001

Source	DF	Type III SS	Mean Square	F Value	Pr > F
first	1	15.4176546	15.4176546	12.93	8E-04
fan	1	31.6346023	31.6346023	26.53	<.0001

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	17.73475	0.21663508	81.86	<.0001
first	-1.330645	0.37007726	-3.6	0.0008
fan	0.403182	0.0782816	5.15	<.0001

The SAS System

The GLM Procedure

Number of Observations Read	52
Number of Observations Used	52

Dependent Variable: GSD

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	4	0.0529886	0.0132472	2.22	0.082
Error	47	0.2810883	0.0059806		
Corrected Total	51	0.3340769			

R-Square	Coeff Var	Root MSE	GSD Mean
0.158612	2.916584	0.077334	2.651538

Source	DF	Type I SS	Mean Square	F Value	Pr > F
winter	1	0.0177231	0.0177231	2.96	0.092
day	1	0.0035703	0.0035703	0.6	0.444
first	1	0.0315077	0.0315077	5.27	0.026
fan	1	0.0001876	0.0001876	0.03	0.86

Source	DF	Type III SS	Mean Square	F Value	Pr > F
winter	1	0.0097965	0.0097965	1.64	0.207
day	1	0.0037004	0.0037004	0.62	0.436
first	1	0.0149112	0.0149112	2.49	0.121
fan	1	0.0001876	0.0001876	0.03	0.86

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	2.659844142	0.0272519	97.6	<.0001

winter	-0.034119422	0.0266587	-1.28	0.2069
day	-0.021308278	0.0270893	-0.79	0.4355
first	0.045776693	0.0289908	1.58	0.121
fan	0.001271499	0.0071799	0.18	0.8602

Number of Observations Read	52
Number of Observations Used	52

Dependent Variable:
GSD

Source	DF	Sum of Squares	Mean Square	F Value	Pr > F
Model	2	0.0492308	0.0246154	4.23	0.02
Error	49	0.2848462	0.0058132		
Corrected Total	51	0.3340769			

R-Square	Coeff Var	Root MSE	GSD Mean
0.147364	2.875472	0.076244	2.651538

Source	DF	Type I SS	Mean Square	F Value	Pr > F
winter	1	0.0177231	0.0177231	3.05	0.087
first	1	0.0315077	0.0315077	5.42	0.024

Source	DF	Type III SS	Mean Square	F Value	Pr > F
winter	1	0.0177231	0.0177231	3.05	0.087
first	1	0.0315077	0.0315077	5.42	0.024

Parameter	Estimate	Standard Error	t Value	Pr > |t|
Intercept	2.645384615	0.0183133	144.45	<.0001
winter	-0.036923077	0.0211464	-1.75	0.0871

first	0.049230769	0.0211464	2.33	0.0241

Appendix K Particle density data

AccuPyc 1330					
Density and Volume Report					
Sample ID:	Dust		Started:	5/18/2009 13:06	
Sample Weight:	0.2986g		Completed:	5/18/2009 13:27	
Temperature:	22.7 °C				
Number of Purges:	10		Equilibration Rate:	0.0050 psig/min	
Cell Volume:	3.6503 cm3		Expansion Volume:	9.3619 cm3	
Chamber Insert:	1.0 cm3		Calibration Factor:	1.001444	
Run#	Volume cm^3	Deviation cm^3	Density g/cm^3	Deviation g/cm^3	Elapsed Time (h:m:s)
1	0.2023	0.0002	1.4762	-0.0015	0:06:10
2	0.2020	-0.0001	1.4780	0.0004	0:07:42
3	0.2017	-0.0004	1.4804	0.0027	0:09:22
4	0.2021	0.0000	1.4778	0.0002	0:11:02
5	0.2015	-0.0006	1.4818	0.0042	0:12:57
6	0.2027	0.0006	1.4734	-0.0043	0:14:35
7	0.2024	0.0003	1.4755	-0.0021	0:16:05
8	0.2023	0.0002	1.4764	-0.0013	0:17:36
9	0.2019	-0.0001	1.4787	0.0010	0:19:09
10	0.2020	-0.0001	1.4782	0.0006	0:20:53
Average Volume:	0.2021 cm3		Standard Deviation:	0.003 cm3	
Average Density:	1.4776 g/cm3		Standard Deviation:	0.0024 g/cm3	