

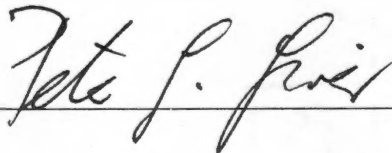
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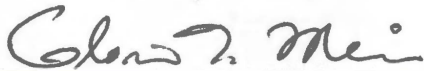
I am submitting herewith a thesis written by Brett Allan Sumner entitled "Measurement of the Median Particle Size Diameter of Airborne Particles Collected Within Duratek's Metal Melt Facility." I have examined the final paper copy of this thesis for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Nuclear Engineering.

Larry Miller, Major Professor

We have read this thesis and
recommend its acceptance:







Acceptance for the Council:



Vice Provost and Dean of
Graduate Studies

DEDICATION

*Thesis
2002
.587*

This thesis is dedicated to my wife, Marlene, and my son, Allan. The joy of finishing the degree requirements is only surpassed by our love for each other. Allan, our desire and prayer for you is that you better yourself through higher education.



**MEASUREMENT OF THE MEDIAN PARTICLE SIZE DIAMETER
OF AIRBORNE PARTICLES
COLLECTED WITHIN DURATEK'S METAL MELT FACILITY**

**A Thesis
Presented for the
Master of Science
Degree
The University of Tennessee, Knoxville**

**Brett Sumner
December 2002**

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ABSTRACT

Concerns related to the occupational health of workers employed in the nuclear industry continue unabated. Often the concern for workers involves the potential absorbed dose to critical organs, and the associated potential health risk that results from inhaling airborne radioactive particulate material. The ability to determine the particle size in the workplace provides useful information to this end. Measurement techniques used to measure and quantify particle size diameters must be capable of providing data on the contaminant within the size range reaching the critical organs.

Representative large area air samples were collected on four days over an eight-month period, atop the furnace deck located inside the contamination zone (CZ) process area in Duratek's Metal Melt facility (MMF). These samples were collected as various client nuclear power customers' waste metals were processed. Following the sample collection process, the samples were weighed and analyzed using two different techniques to determine the subsequent particle size distribution.

The sampling and analysis techniques used for this work provide a reasonable estimate for the typical particle size distribution of airborne particulate material found within Duratek's MMF. Completion of this work has resulted in the determination of the median projected area diameter (MPAD), and the associated activity median aerodynamic diameter (AMAD) of airborne particulate material in the MMF during routine operation. The MPAD finding will be used to determine the effectiveness of current procedural and engineering controls in use to protect workers from the inhalation

of airborne particulate material. In addition, the AMAD value can be used in conjunction with Duratek's bioassay program to correct intake estimates.

Results from this analysis yielded an MPAD value of 2.5 μm , having a geometric standard deviation of 2.6. An MPAD value of 2.5 μm correlates to an AMAD value of 5 μm , assuming a particle physical density of 4 g/cm^3 and is spherical in shape. This value is 5 times the size of the 1 μm value currently used in the International Commission Radiological Protection (ICRP) 30 lung deposition model. However, it should be noted that ICRP 66 lists 5 μm AMAD values for possible use in internal dosimetry calculations.

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LIST OF ABBREVIATIONS/ACRONYMS

Activity Median Aerodynamic Diameter.....	AMAD
Centimeter.....	cm
Contamination Zone.....	CZ
Cubic feet per minute.....	cfm
Dust Deposition Model.....	DDM
Feet	ft
International Commission of Radiological Protection.....	ICRP
Kilowatt.....	kW
Liters per minute.....	lpm
Low-Level Radioactive Waste.....	LLRW
Mass Median Aerodynamic Diameter	MMAD
Metal Melt Facility	MMF
Median Projected Area Diameter.....	MPAD
micron	μm
millimeters	mm
Nasopharyngeal.....	N-P
Nuclear Regulatory Commission.....	NRC
Occupational Safety and Health Administration	OSHA
Pulmonary Lymph Node.....	L
Pulmonary.....	P
Reactor	Rx
Tracheo-bronchial.....	T-B

CHAPTER 1

INTRODUCTION

1.1 Background

The potential effects and possible adverse health implications associated with the inhalation of soluble and insoluble particulate materials are dependent on numerous factors. The potential site of deposition is one of the most important factors. Inside the respiratory tract, there exists a network of connecting structures that vary in terms of their function and size. These variables make the regional deposition site and its accompanying elimination pathway noteworthy.

It is widely accepted among health professionals that the two most significant properties associated with aerosol particles are their size and chemical composition. Once inhaled into the lungs, the subsequent health effects vary according to physical, chemical, and biological properties of the particulate material. Of these properties, the physical property of particle size and the selective-size characteristics of the human respiratory tract are perhaps the most significant. Prior to the 1940's, the effects these characteristics had on the respiratory tract had largely been ignored. While all the properties mentioned above influence the interaction, deposition, and clearance mode within the respiratory tract, the physical property of particulate size is the primary focus in this study.

During the early 1950's, an increasing number of scientists began studies dealing with size-selective health hazard sampling. The scientific community drafted several helpful documents that aided their collective efforts. In 1952, the British Medical

Research Council adopted a definition for “respirable dust” as dust reaching the alveoli region of the lungs. Their definition is in terms of percent respirable versus diameter for spheres of unit density [1]. This gave the researchers a standard definition on which to base their work. In 1959, the ICRP adopted the respiratory tract deposition and retention models, which are valuable for performing internal dosimetric calculations [2]. Both of these actions helped steer the course toward better understanding the associated risks of particulate material inhalation.

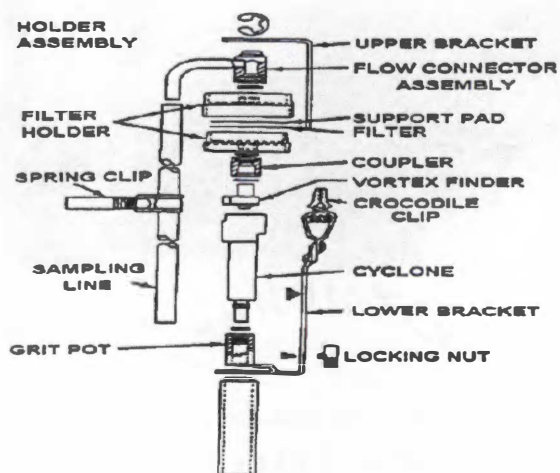
Air sampling techniques included a variety of sampling instruments. Before elaborate sampling equipment became common, a single-stage collection device like the one shown in Figure 1.1 was commonly used. The primary goal for using a single stage collector is determining a sample’s total concentration. There is generally no interest in determining the actual particle size. Consequently, a single-stage collection device will most often use a glass fiber filter media. As a result, these simple sampling devices are not the best choices for studying size distributions. More advanced sampling techniques that incorporate multiple filtering stages are better suited for collecting these data.

Multiple-stage instruments, like the cyclone shown in Figure 1.2, have an advantage over single-stage devices since they allow the direct fractionation of airborne dust based on the size of the dust particles. Some of the first multiple-stage instruments developed are two-stage samplers like the cyclone, and horizontal elutriation sampling devices. These instruments became popular because they are easy to use and directly provide particle size information. Later, sampling instruments advanced to contain still more stages. These advancements combined to save valuable sampling time by acquiring samples over multiple size ranges at one time.



Figure 1.1 **Single-Stage Collection Device.**
The device that is shown is a high volume air-sampler fitted with a glass-fiber filter paper.

Source: Daniel A. Gollnick, *Basic Radiation Protection Technology*, Pacific Radiation Corporation, p. 344, 1988.



Cyclone Part Diagram

Figure 1.2. Multiple-Stage Cyclone Collection Device.

Source: Millipore Products Catalog, Millipore Corporation, Copyright 1998.

In any multiple-stage sampling system, the first stage of an instrument functions as the pre-collector where high collection efficiency for large particles is needed. The first stage is followed by later stages, which collect particles down to some critical size. These later stages collect those particles that make it past the pre-collector, but are too large to escape collection by the succeeding stage.

In these studies, the particle size distribution obtained from sample data collected by the final filter may be equally, more, or even less significant than the findings from the preceding filter sections. It is a matter of defining the intended purpose for the data beforehand so that the intended direction of the study is defined before beginning the sampling process. This will ensure the most relevant data will be collected as the study goes forward.

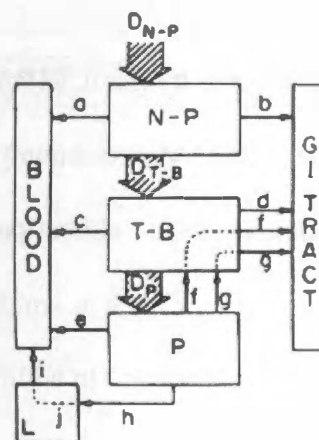
Primarily, multi-stage samplers were designed to simulate deposition of particulate material in the lower divisions of the respiratory tract. What followed was a method to assign a given particle size to a definite deposition site within the respiratory system. Knowing the particular deposition site, and its specific clearance mode, helps scientists calculate the dose equivalent resulting from the uptake of radioactive material.

In 1959, the ICRP published a report detailing the equations and methods of calculation used to derive maximum permissible body burdens to the critical organs, and the maximum permissible concentrations in both air and water. This report was one of the first descriptions of respiratory tract deposition and clearance models, providing the basis for radiation protection for internal emitters for decades in the United States [3]. The respiratory tract clearance model, known as ICRP Publication 2, greatly enhanced a scientist's ability to perform reproducible internal dosimetric calculations. The

respiratory tract clearance model shown in Figure 1.3 outlines the clearance mechanism, and transport, within the following regions: nasopharyngeal (N-P), tracheo-brochial (T-B), pulmonary (P), and pulmonary lymph nodes (L). The regions are specified in the far-left column in Figure 1.3. Here it shows that for every breath taken in by the lungs, 30% of inhaled dust is deposited in the N-P region, 8% in the T-B region, 25% in the P region, and the remaining 37% is exhaled. These deposition fractions are based on inhalation and deposition of an aerosol whose Activity Median Aerodynamic Diameter (AMAD) is 1 μm and whose geometric standard deviation is 4.

The ICRP Publication 2 model has been revised and subsequently enhanced over the years. In 1965, the "Committee II" set up a special task group that proposed changes to both the deposition and retention models based on nose breathing. The next model came from the work published by the Task Group on Lung Dynamics (1966), and was subsequently incorporated into ICRP publication 30. In July 1978, the ICRP adopted Publication 30, Limits for Intake of Radionuclides by Workers, Part 1 [4]. This publication is currently recognized by the State of Tennessee's Department of Radiological Health and Environmental Services. It is used at all Duratek production facilities in the state. The ICRP 30 model, as it is known, does not provide the ability to select specific inhalation materials, or the ability to consider other modifying factors related to inhalation, deposition, or clearance modes.

REGION	CLASS	CLASS					
		D		W		Y	
		T	F	T	F	T	F
N-P ($D_{N-P}=0.30$)	a	0.01	0.5	0.01	0.1	0.01	0.01
	b	0.01	0.5	0.40	0.9	0.40	0.99
T-B ($D_{T-B}=0.08$)	c	0.01	0.95	0.01	0.5	0.01	0.01
	d	0.2	0.05	0.2	0.5	0.2	0.99
P ($D_P=0.25$)	e	0.5	0.8	50	0.15	500	0.05
	f	n.a.	n.a.	1.0	0.4	1.0	0.4
	g	n.a.	n.a.	50	0.4	500	0.4
	h	0.5	0.2	50	0.05	500	0.15
L	i	0.5	1.0	50	1.0	1000	0.9
	j	n.a.	n.a.	n.a.	n.a.	∞	0.1



Respiratory tract clearance model used in calculating dose from inhaled radioactivity. (From S. B. Watson and M. R. Ford. A User's Manual to the ICRP Code: A Series of Computer Programs to Perform Dosimetric Calculations for the ICRP Committee 2 Report, ORNL/TM-6980, Feb., 1980.) The values for the removal half-times, T_{a-j} , and compartmental fractions, F_{a-j} , are given in the tabular portion of the figure for each of the three classes of retained materials. The values given for D_{N-P} , D_{T-B} , and D_P (left column) are the regional depositions based on an aerosol with an AMAD of $1\mu\text{m}$. The schematic drawing identifies the various clearance pathways in the model, a - j, in relation to the depositions D_{N-P} , D_{T-B} , D_P and the three respiratory regions, N-P, T-B and P. The entry n.a. indicates not applicable.

Figure 1.3. Respiratory Tract Clearance Model (ICRP Publication 2).

Source: Herman Cember, *Introduction to Health Physics*, Pergamon Press, New York, p. 211, 1985.

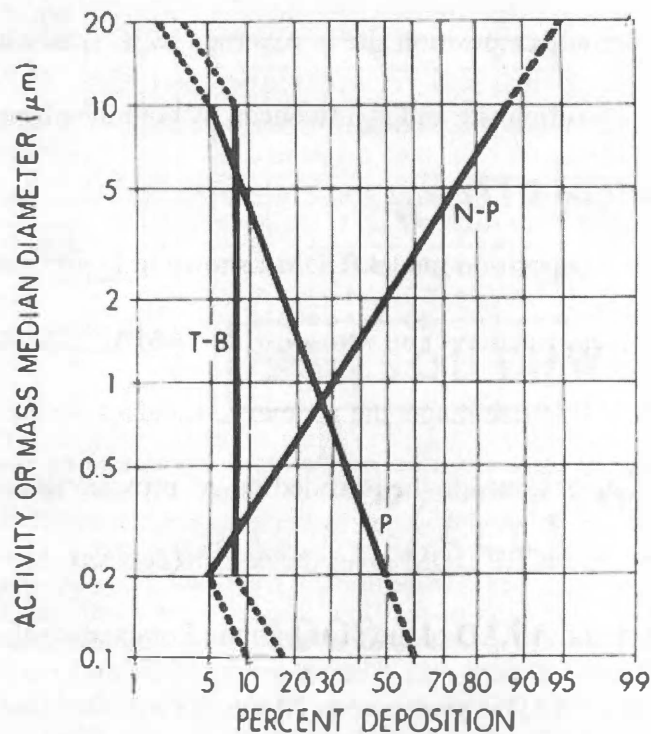
These enhancements came in the ICRP Task Group's 1966 model. This latest model was adopted by the ICRP in 1994, and issued as the new "Human Respiratory Tract Model" (HRTM), commonly referred to as ICRP 66 [5]. This model has numerous enhancements over the Publication 30 lung model, like the ability to consider the effects of modifying factors, such as smoking, pollution and disease on lung deposition, clearance, and inhalation. In addition, the ICRP 66 model is based on the assumption that individuals have two pathways for inspiration of air, nasal and buccal [5]. Additionally, unlike its predecessors, this model addresses the different fractions of air inspired through the nasal and buccal pathways as a function of breathing rate for adult males and females [5]. This model reflects the current advanced state of knowledge for the behavior of inhaled particles.

The ICRP Publication 2 lung clearance model is shown in Figure 1.3 as a reference. For its day (cir.1959) it is quite detailed, giving removal half times, T_{a-j} , and compartment transfer fractions, F_{a-j} , for each of the three classes of retained particles. The class distinction accounts for the impracticality of determining the clearance route and respective clearance rate for each element or compound deposited. Consequently, a broad interpretation is made by assigning the retention periods of day (D), week (W), and year (Y) of retained materials. Similar to this model's retention periods, the ICRP 30 model also uses the (D), (W), and (Y) time distinctions. Unlike the two earlier lung models, the ICRP 66 model uses slow (S), moderate (M), and fast (F) for the period distinctions. The ICRP 2 model gives the clearance pathways, $a - j$, in relation to the deposition sites D (N-P), D (T-B), D (P), within the three respiratory regions, N-P, T-B, and P. The HRTM from ICRP 66 divides the lung regionally as Extrathoracic airway,

Thoracic airway, and Lymphatic tissue. All the lung clearance models discussed take into account that a particle disposition site is governed by its size, while its clearance rate is governed by the deposition site and is influenced by both the physical and chemical properties of the particle.

The ICRP dust deposition model (DDM), shown in Figure 1.4, is a helpful tool for determining the mass fraction of an aerosol whose AMAD size range is between $0.2\text{ }\mu\text{m}$ and $10\text{ }\mu\text{m}$. In this size range, the geometric standard deviation is less than 4.5. For deposition percentages outside the intended range, provisional estimates are indicated by the dashed lines. The ICRP DDM gives the percent deposition in the N-P, T-B, and P regions, in relation to the AMAD of the distribution. For excessively large distributions where the AMAD or MMAD is greater than $20\text{ }\mu\text{m}$, it's assumed that 100% N-P deposition occurs. In addition, when the AMAD or Mass Median Aerodynamic Diameter (MMAD) size distributions are smaller than $0.1\text{ }\mu\text{m}$, the model no longer applies [3]. The acronym MMAD is defined as the diameter where 50% of the mass (or volume) of the aerosol includes particles less than the given diameter.

The regions that make up the respiratory tract differ greatly from one another in terms of their retention time, the subsequent elimination pathway, and the elimination method used to remove contaminants. It is for this reason that determining the particle size of nuisance dust in the workplace is useful and a concern to health professionals. In addition, when there is the potential for the dust being radioactive, this adds still another reason for concern.



ICRP dust deposition model. The radioactive or mass fraction of an aerosol which is deposited in the N-P, T-B, and P regions is given in relation to the Activity or Mass Median Aerodynamic Diameter (AMAD or MMAD) of the aerosol distribution. The model is intended for use with aerosol distributions having an AMAD or MMAD between 0.2 and 10 μm and whose geometric standard deviations are less than 4.5. Provisional deposition estimates further extending the size range are given by the dashed lines. For the unusual distribution having an AMAD or MMAD greater than 20 μm , complete N-P deposition can be assumed. The model does not apply to aerosols with AMAD or MMAD below 0.1 μm .

Figure 1.4. ICRP Dust Deposition Model

Source: Herman Cember, *Introduction to Health Physics*, Pergamon Press, New York, p. 212, 1985.

1.2 Description of metal processing at Duratek

Duratek's Bear Creek facility, shown in Figure 1.5, is the largest processor of radioactive waste in the United States and it has the capacity to process up to 50 million pounds of radioactive waste annually. One of the aspects of Duratek's business is processing radioactive metals. The Nuclear Regulatory Commission (NRC) estimates metals will be removed from decommissioned nuclear power plants and government facilities at maximum rates of up to 71,000 tons per year in the coming years [6].

Throughout the United States and abroad, atomic transformation of fissile material inevitably generates radioactive waste. The type of wastes may include any number of items and waste forms, from tiny cloth cotton liners to sections of wood scaffold planking, with many items falling somewhere in between. Cotton liners, for example, are a form of protective clothing worn on the hands, underneath radiation workers' cloth coveralls to prevent hand contamination. The question is often asked, "Why does wood planking find its way into a nuclear power facility?" Wood scaffold planks provide a work platform above the floor in areas where ladders cannot be used. These are just two examples of material that becomes radioactive waste once its usefulness has ended. In addition to these items, a fair amount of metal becomes contaminated. Waste metals take on a variety of forms depending on the method of use, and on the place of origin within the nuclear facility.

For instance, a (Rx) building, which houses vital components associated with the Rx, contains hundreds of miles of cable in cable trays. These cable trays help keep the many wiring systems separated and out of the way. This minimizes their chance of being unintentionally damaged. During scheduled



Our Bear Creek facility in Oak Ridge, Tennessee has the only commercial incinerators, the largest supercompactor, and the largest metal melting operation for radioactively contaminated materials.

Figure 1.5. Duratek's Bear Creek Facility.
The MMF is the building located bottom right in the photograph.

maintenance, operations workers take apart various components by breaching contaminated piping systems. At times, these cable trays and other nonessential items become contaminated. Once the plant modifications are complete, the removed materials that have no further use are often discarded as radioactive waste. This is a common occurrence and the fate of many replaced pieces of equipment that are no longer needed. The majority of removed system components are small and easily handled. Some contaminated components are difficult-to-handle due to their sheer size and bulkiness. Large contaminated components like steam generators, pressurizers, and primary system circulation piping are examples of difficult to handle components.

In nuclear power generating facilities, reactor cooling water piping has to be inspected routinely and replaced when structural degradation through corrosion has occurred. These cooling systems contain an elaborate series of piping, valves, and monitoring equipment used to circulate water through the Rx core. Within pressurized water reactor systems, the primary cooling system is pressurized to more than 2000 lb/in². This prevents the cooling water from turning to steam as it moves through the nuclear core. In this system, steam generators transfer heated primary coolant to a secondary coolant operated at a lower pressure where boiling can occur. This process is the first step toward removing the heat energy used to create electricity.

A common source of contaminated piping occurs when Rx cooling water is drained from the cooling system. Once the piping system is drained and allowed to dry, radioactive residue in the form of crud and other impurities is left behind. Crud is comprised mainly of corrosion products and, combined with other impurities, is troublesome. Crud deposits can foul components and monitoring equipment when left

unchecked. This type of surface contamination occupies only the top layer of the metal surface. This is true whether the water passed through a pipe or a valve. Surface contamination can usually be removed with soap and water, but sometimes mild chemicals or abrasives may be needed.

Alternately, another way that metal components become radioactive is through neutron activation. This process occurs in any nuclear facility where fissile material emits neutrons while undergoing nuclear transformation. The activation process involves the absorption of thermal neutrons by metal atoms in close proximity to fissile material undergoing nuclear transformation. This type of radioactive metal is not easily decontaminated. A method used to remove the radioactive constituents caused by both activated and surface-contaminated metals is through the smelting process. This process is both effective and economical, and is the chosen method to volumetrically reduce waste metals at Duratek's MMF.

At this facility, Duratek utilizes a metal processing program that provides its customers a cost-effective solution for treating radioactively contaminated metals. Duratek operates the largest electric-induction furnace used exclusively for melting metals contaminated with radioactive material in the United States. The furnace holds up to 20 tons and operates at a power rating of 7200 kW electric while processing various types of both ferrous and non-ferrous contaminated metals.

During the smelting process, the majority of the contaminated impurities are removed as slag from the top of the molten bath as the process goes forward (see Figure 1.6). The molten metal is then poured into a cast mold, as shown in Figure 1.7, hardening into a block-shaped ingot that's "slightly" radioactive. The final product is a block-



Figure 1.6. Molten Metal Bath.

The thermocouple in the picture is used to measure the temperature of the molten metal bath. The thermocouple is the cylindrical shaped object extending down from the top left corner of the picture.



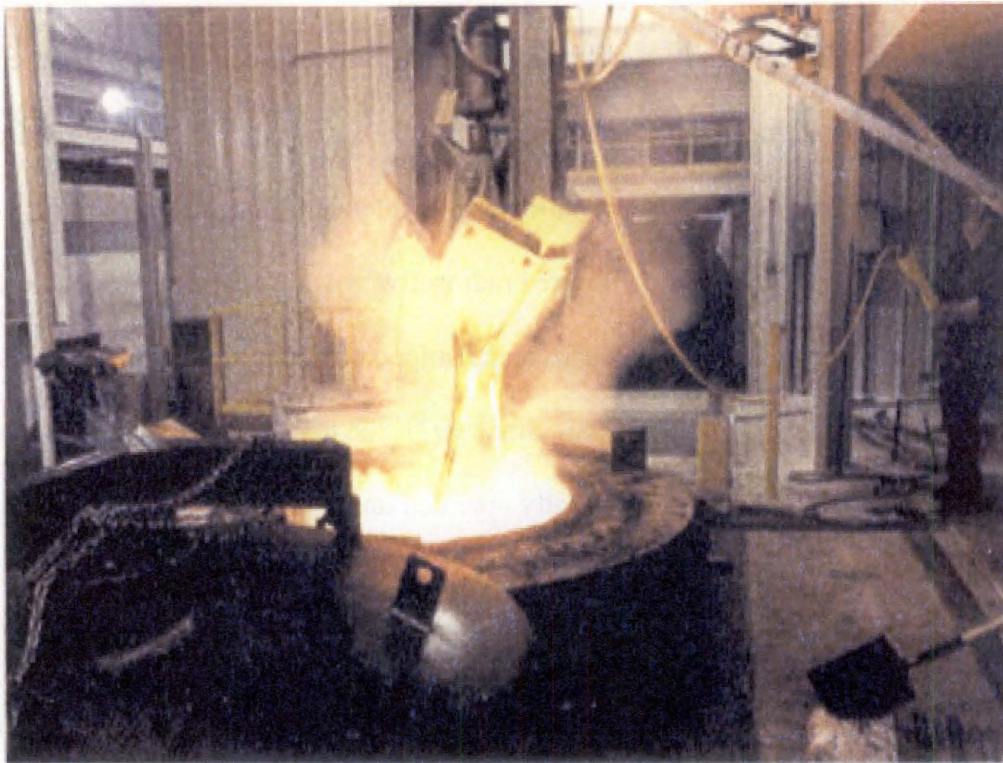
Figure 1.7. Block Shape Ingot. The block ingot shown is called an S-2 and weighs approximately 10 tons. This ingot has cooled and is painted "Duratek blue." When the blue paint is dry, the top and all four sides will be stenciled "caution radioactive material."

shaped ingot which is painted to inhibit rust, stenciled with the words “Caution Radioactive Material,” and number stamped for quality assurance purposes. These ingots are suitable for providing shielding around structures like particle accelerators and reactor buildings where some background radiation can be tolerated. The unusable "slag" portion, shown in Figure 1.8, is considered radioactive waste since it no longer has a useful purpose. This metal is containerized and shipped for long-term storage outside the rural town of Barnwell, South Carolina. Since 1971, the low-level radioactive waste (LLRW) burial site has been commercially operated under a 99-year lease agreement with the state of South Carolina.

The Barnwell LLRW burial site consists of approximately 300 acres and is located near the eastern boundary of the Department of Energy's Savannah River Site (DOE SRS), (Figure 1.9). The DOE SRS occupies an area of approximately 310 square miles and is located 12 miles south of Aiken, South Carolina.

The State of South Carolina licenses the operation of the LLRW burial site through the State of South Carolina's Department of Health and Environmental Control under authority delegated to it by the NRC. This protocol for managing radioactive licenses is typical in those states described as “agreement state.” NRC regulations covering the transfer of authority to Agreement States are found in the Title 10, Part 150 of the Code of Federal Regulations.

The State of Tennessee is also an “agreement state.” As such, all day-to-day activities involving the safe handling of radioactive materials are managed by the State of Tennessee's Department of Radiological Health and Environmental Services. The State of Tennessee has regulatory authority granted by the NRC to pursue, prosecute, and levy



Duratek has the capacity to recycle over 25 million pounds of metal each year.

Figure 1.8. Removing Slag Metal from the Furnace using Metal Tongs. Notice in the foreground the furnace cover in the removed position. This picture was taken during test runs with clean metal. (i.e., prior to processing radioactive material). Of particular interest, notice the operator in the top right portion of the picture. Absent is all the protective clothing worn by furnace tenders today. Protective clothing includes a full set of fire-resistant, anti-contamination clothing; full face respirator; aluminum apron; and gloves.

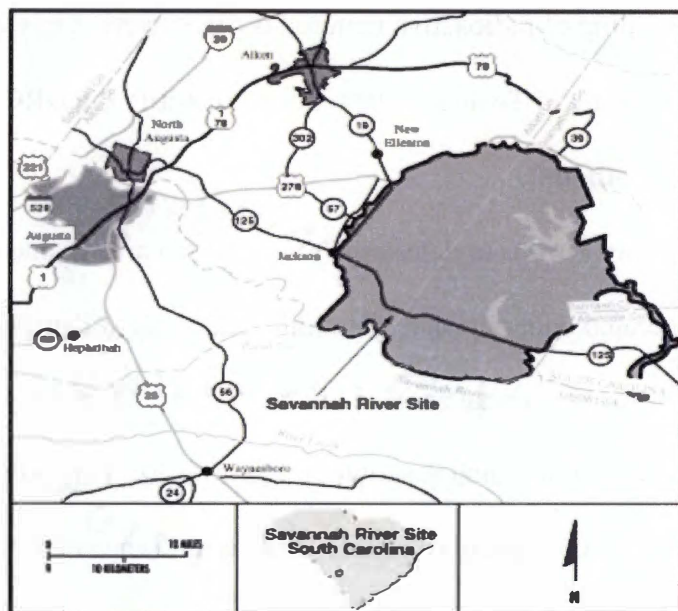


Figure 1.9. Department of Energy Savannah River Site

Source: World Wide Website globalsecurity.org/wmd/facility/savannah_river.htm

finest when negligent handling of radioactive material is discovered. Only during rare “unusual events” will the State of Tennessee be required to notify the NRC.

1.3 Objectives and methods

The objective of this work is to determine the projected area diameter of airborne particulate material generated under normal operating conditions in Duratek's MMF. In this facility, pieces of radioactive metal are sized into 2 foot by 2-foot sections using a variety of cutting methods one of which is shown in Figure 1.10. This process, along with the operation of the 20-ton induction furnace, creates airborne smoke, fumes, and dust illustrated in Figure 1.11. Determining the projected area diameter is important for a variety of reasons. If the measured particle size diameter is found to be significantly different from the typical 1 μm AMAD value, correcting intake estimates for particle size differences is an option. Additionally, these measurements will be useful in determining the effectiveness of engineering and procedural controls currently being implemented in the MMF.

One engineering control currently used throughout the MMF is the overhead ventilation system shown in the top portion of Figure 1.12. This system works to remove those airborne contaminants that become suspended in air during processing operations. Even under ideal operating conditions, not all the suspended particulate material is collected by the ventilation system. In addition to already suspended contaminants, resuspension of loose surface particulate material is a common occurrence. Common causes for resuspension include moving air currents, air disturbances caused by power tools, and fork truck exhaust. For this reason, additional procedural controls are used to protect employees.



Figure 1.10. Cutting Technique-using an Oxygen Lance.
Notice at the bottom right of the photograph, a fire-watch attendant with a fire extinguisher at their side.



Figure 1.11. Induction Furnace.

Notice the opening of the furnace is uncovered for charging purposes. The red “ember like” projectiles are pieces of molten metal. Notice also the ventilation hood above the furnace that captures the rising smoke plume. This viewing angle is taken from the ground looking upward toward the furnace deck.



Figure 1.12. Charging the 20 ton Induction Furnace.
2 foot by 2 foot pieces of metal are dropped into the furnace by a remotely operated charging cart traveling on a rail. The viewing angle is across the furnace deck toward the control room (obscured from view by the ventilation hood at the top of the photograph).

An example of how Duratek uses procedures to protect its workers is through its As Low As Reasonably Achievable (ALARA) program. The ALARA concept is widely used and commonplace in the nuclear industry. General radiation protection requirements that apply to all NRC licensees are contained in Title 10, Part 20 of the Code of Federal Regulations. These standards are based in part upon the recommendations of the ICRP and the National Council on Radiation Protection and Measurements (NCRP). Part 20 applies to occupational workers who receive exposure to radiation on the job. This federal regulation also has provisions to protect individuals in the general public from unreasonable exposure to radiation.

Like other nuclear programs in this country, Duratek's goal is to maintain its workers' exposure ALARA. Consideration is given to reducing both the internal and external exposure of workers to radioactive material. An example of the ALARA concept at work is Duratek's respiratory protection program. A policy found in the ALARA program is the desire to minimize all employees' exposure to airborne radioactive material. Consequently, while working in the MMF contamination zone (CZ), all radiation workers wear respiratory protection devices.

These personal protection devices filter both lead and suspended radioactive particulate material out of the air. Consequently, the results of this work will provide useful information for prescribing the proper respiratory protection devices necessary for working in this facility. In the unlikely event of unprotected smoke inhalation, knowing the median size of airborne particles within the MMF would be useful when performing internal dosimetric calculations. This information, used in conjunction with the ICRP 66 lung model, provides a method for determining the biological elimination pathway.

Collectively, this work will undoubtedly improve the accuracy of internal dosimetric calculations, should they become necessary.

1.4 Technical basis for methodology

In 1965, R. F. Hounam and R. J. Sherwood published a paper describing how they constructed a cascade centripeter that consisted of three inertial separation stages. Their device removed larger particles in the early stages, while allowing the smaller particles to be collected in the terminal filters stages [7]. In 1957, N. Zurlo developed a sampler with eight inertial separation stages. Through a series of 90-degree turns, the device projected oversized particles into individual collection cups, while the final backup filter collected the sought after sized particulate [8]. Like Hounam's and Sherwood's instrument, Zurlo's sampling device contained multiple stages which were then followed by a backup filter. Here both sampling methods represent multiple stage samplers that project large particles into a still air volume for collection as a method of sampling. Other multiple stage samplers have been designed and used by other scientists in these studies as well.

The most widely used type of multi-stage sampler is the cascade impactor. These instruments are readily available commercially in a variety of designs. A major limitation to their application is that only limited sample masses can be collected without re-entrapment. In addition, further limitations common with other multi-stage instruments like the cascade centripeter are deposit losses along the wall and between collection stages, and the increased number of samples that are required [8]. A simplified approach to separate particles by size is to use a two-stage sampler.

Some of the most commonly used first stage devices that comprise a two-stage sampler include both cyclones and horizontal elutriation devices. Of the two, cyclones

are perhaps the most commonly used respirable mass sampler. When using a cyclone, a sample of air entering the cyclone pre-collector is depleted of larger particles based on the cutoff characteristic of the device. Smaller particles will pass through the axial exhaust tube of the cyclone being deposited directly onto a backup filter that captures them. Morton Lippmann, a pioneer in the field of selective size particle studies, often incorporated cyclone pre-collectors in his sampling devices. In these studies, each two-stage sampler consisted of some type of cyclone followed by an efficient backup filter used to capture the final particulate material.

The collection characteristics of cyclone samplers is flow-rate dependent. Sometimes Dr. Lippmann used a single sampling device with one cyclone pre-collector. Other times, he uses a parallel array of cyclones with all pre-collectors drawing suction from the same sampling tube, each operated at varying flow-rates. This creates a series of size cuts, since the ratio between the cyclone collection and the backup filter collection is dependent upon the sampling rate. In this arrangement, each cyclone has a different size-cut characteristic allowing the overall size-mass distribution to be determined from analyses of the backup filters. The data is normalized for the varying flow-rates and plotted against the characteristic cutoff data for that stage directly on log-probability paper [9].

The major advantage in using cyclones is that performance is not significantly affected by the amount of sample collected. Other practical advantages include minimal particle bounce and re-entrainment, large capacity for loading, compact size, and insensitivity to orientation [10]. In addition, due to the partitioning ability of the device through size cut-off classifications, cyclones make particle size measurements readily

available. Moreover, these devices have been used to directly simulate particulate deposition in the N-P and T-B regions of the respiratory tract. "Cyclone separators are frequently used since they tend to have performance characteristics with shapes similar to airway penetration curves." [11] The objective here is to collect particles that are deposited in the N-P and T-B regions in approximately the same proportion which otherwise would be deposited in lungs. The particles that are then collected on the filter, having penetrated past the inertial separator pre-collector, provide the size fraction representing the particles available within the gas exchange region of the lung [12]. Consequently, once in this region, contaminants then pass directly into the blood stream allowing the subsequent effects to be more widespread throughout the body.

Horizontal elutriation devices have a predictable performance based upon gravitational settling of the particles once they've successfully passed between the horizontal collection plates. These devices are usually coupled in front of backup filters, but have been used with impingers as well. These instruments have been operated at high sampling rates, with rates upwards of 40 cubic feet per minute (cfm) being common. Despite the many benefits of their use, some drawbacks are well documented.

Both cyclone and horizontal elutriator pre-collection devices have been used extensively but not without some difficulty. One common problem with elutriator pre-collectors is the difficulty in recovering all the collected material, making it nearly impossible to minimize contamination of the second-stage collection by re-entrained dust. Another limitation is orientation concerns, since the elutriator must be held in the horizontal position while sampling. This is also a concern shared when using water-filled pre-impingers. Furthermore, water-filled pre-impinging devices tend to be particularly

bulky and would be hard to miniaturize [13]. Besides the orientation limitations, both horizontal elutriators and cyclones share another common element – a relatively high cost.

Cyclone pre-collectors have several drawbacks as well. One in particular involves an important performance factor. Sampling very high dust concentrations may cause particle agglomeration and increased collection efficiency within the cyclone [14]. This was a major consideration because frequently Duratek's MMF has high airborne dust concentrations. Nevertheless, despite their drawbacks both horizontal elutriations and cyclones have been the most widely used first-stage collectors, while filters have been used as the back-up filter stage in most two-stage samplers [15]. If cost were not an issue in this study, cyclone pre-collectors would have been the sampling devices of choice for this work.

Numerous types of filters have been used in both the first and second stages of size-selective particle sampling devices. J. F. Roesler's work titled *Application of Polyurethane Foam Filters for Respirable Dust Separation* illustrates using filters in the first-stage collection with favorable results. He states, "The most economical approach to selective size sampling by the National Air Sampling Network (NASN) would be use of a prefilter" [16]. Polyurethane foam filters appeared feasible and had been investigated as a size-separating device by K. T. Whitby [16]. Additionally, scientists Thomas A. Cahill and Lowell L. Ashbaugh in their paper titled *Analysis of Respirable Fractions in Atmospheric Particulates via Sequential Filtration* describe the use of a stacked filter sampler. Their instrument collects particles on 47-mm Nuclepore filters mounted in a commercially available sequential filter holder. Nuclepore filters were used

because “... of their low blank weight and low hygroscopicity making them suitable for gravimetric and elemental analyses” [17]. Consequently, following a review of these past works that utilized filters as first-stage collectors, it was decided to incorporate them in this work as well. Additionally, in this work, filters will be used in both the first and second stages of collection.

CHAPTER 2

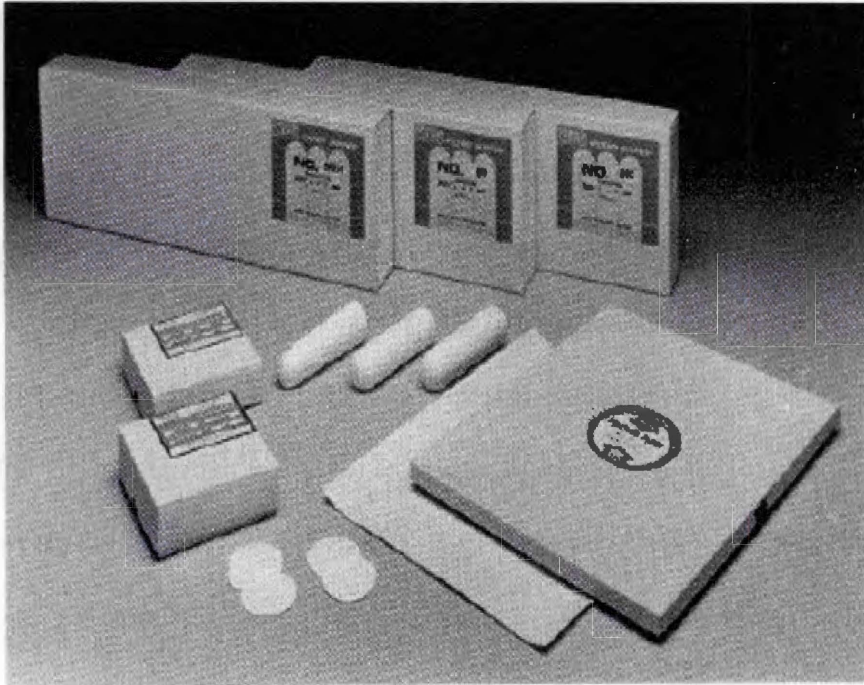
DESCRIPTION OF SAMPLING METHODS AND EQUIPMENT

2.1 Description of various air sampling methods

Particulate airborne material may be sampled using five common methods: sedimentation, electrostatic precipitation, thermal precipitation, impaction, and filtration [3]. All of these sampling methods have been used in a variety of industries, including the Health Physics field, with satisfactory results. However, one in particular is the most direct approach for this work. Another major consideration in sampling selection was the associated cost. Cascade impactor, or cyclone pre-collector sampling instruments alone would cost thousands of dollars.

First, the four methods of sampling that were not chosen for the work will be discussed. This will be followed by an in-depth explanation of filtration methodology, the method of sampling selected.

The sedimentation method uses an adhesive-coated filter paper of known size, shown in Figure 2.1, to collect particulate material suspended in the air by natural forces. Over a given sampling period, particulate material falls onto and adheres to the paper surface. From the resultant sample, the corresponding radioactive content per unit area per unit time is calculated. The time allotted for sampling varies as a function of the airborne contaminants concentration. Therefore, areas of low contaminant concentration require a longer sampling period than areas of higher suspended contaminant concentrations. This is a common method used to measure and calculate atmospheric radioactive particulate fallout.



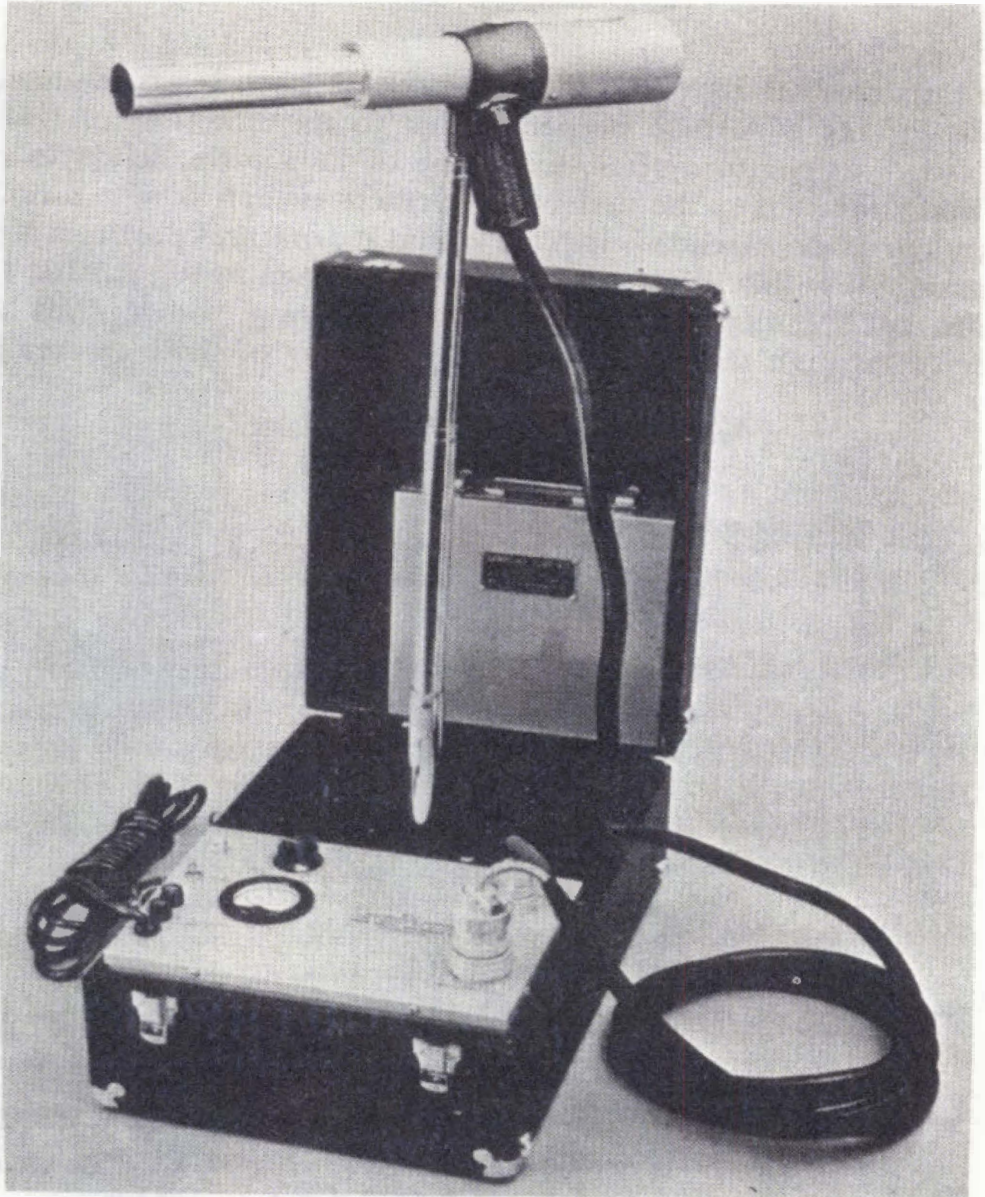
Four grades of fibrous filter media are available in sheets, discs and thimble form for air pollution monitoring.

Figure 2.1. Adhesive-Coated Filter Papers.
Made from fibrous material and in several forms depending on their intended use.

Source: Millipore Products Catalog, Millipore Corporation, Copyright 1998.

Another method, which uses a device shown in Figure 2.2, is an electrostatic precipitator. Electrostatic precipitators utilize a large charge, which is placed across two electrodes while keeping the collecting anode at ground potential. The applied voltage causes the central electrode to create a corona discharge around a 1 millimeter (mm) diameter wire. This causes the surrounding dust particles to become negatively charged. The negatively charged dust particles are then collected by the outer cylindrical-shaped anode. The anode is easily removed and rinsed off to collect the sample. Samples are then dried in thermally controlled ovens to acquire the sample. The multi-step sample collection and preparation makes this method time consuming and cumbersome. This method is frequently used by industrial hygienists for sampling particles in the size range of 0.2 to 5 μm .

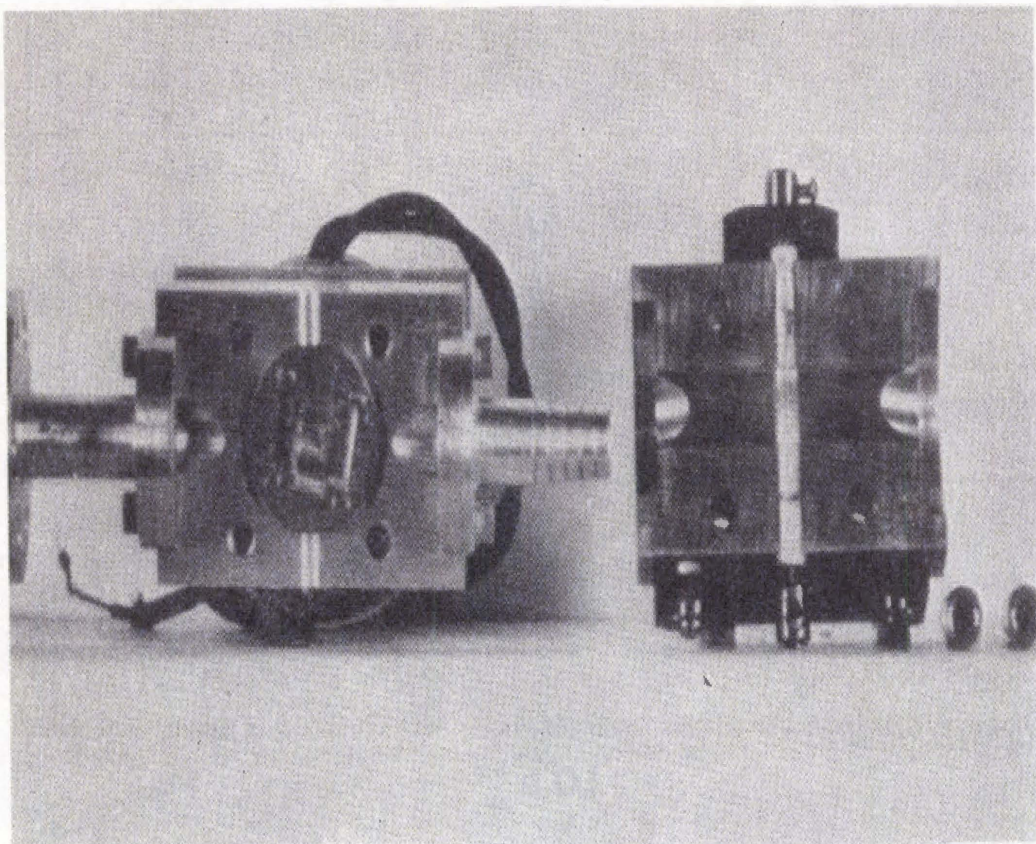
A seldom-used sampling technique utilizes thermal precipitators, shown in Figure 2.3. One reason for their limited use can be attributed to this method's slow sampling rate, on the order of several milliliters per minute [3]. This sampling technique uses a heated wire to create a thermal gradient between the wire and a cold collecting surface. The collection surface is often comprised of a glass cover slip, or glass microscope slide. Particles that enter the sampler's inlet diffuse away from the wire due to the thermal gradient produced by heating the central wire. This causes the particulate material to be collected on the cold collection surface. The thermal gradient that is created is proportional to the amount of heat energy that is supplied. The collection slide is then removed and may be viewed under a microscope, or counted with suitable equipment to determine the corresponding activity.



Electrostatic precipitator for collecting samples of atmospheric dust, fumes, and smoke at a rate of 3 ft³/min. (Courtesy Mine Safety Appliances Co.)

Figure 2.2. Electrostatic Precipitator.

Source: Herman Cember, *Introduction to Health Physics*, Pergamon Press, New York, pp. 401, 1985.



Thermal precipitator. The part on the right has an electrically heated wire across the channel through which the dust-laden air is drawn. The cold collecting plate, a square glass cover slip used by microscopists, is placed into the part on the left. The two parts are then assembled, with the cover slip sandwiched between them. The cover slip is rotated at 1 r.p.m. in order to obtain a uniform size distribution of the collected particles.

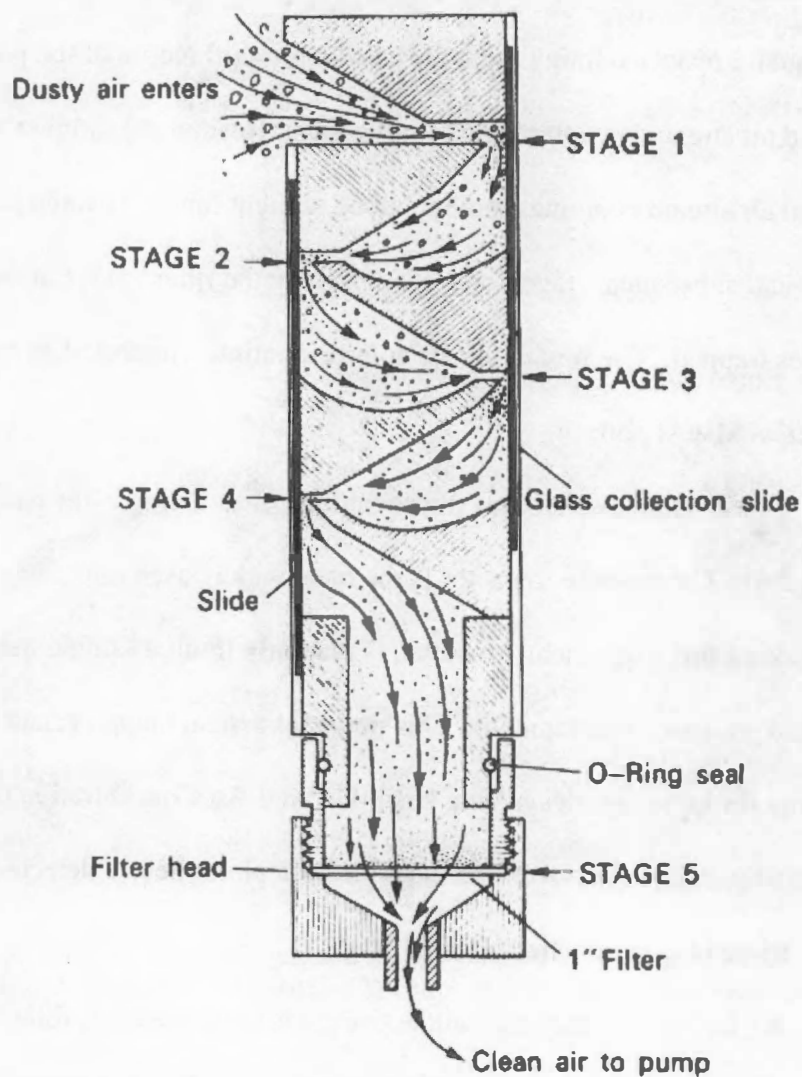
Figure 2.3. Thermal Precipitator.

Source: Herman Cember, *Introduction to Health Physics*, Pergamon Press, New York, pp. 399, 1985.

The last sampling method considered for this study utilizes the impaction technique. Impact sampling methods use the forward inertia of the particles within the sampled air stream to capture and remove them. Beyond the sampler's inlet orifice, the sampled air stream continues moving along straight lines. At some point, within the second and subsequent stages, and upon striking the filter matrix, a sampled contaminant becomes trapped. The remaining air volume continues unabated as exhaust at the sampler's exhaust port.

One instrument that uses this technique of sampling is the cascade impactor (see Figure 2.4). The cascade impactor is the most widely used multi-stage air sampler. One limitation of this instrument, however, is that only limited sample masses can be collected without re-entrainment. This may make them inappropriate to use when sampling for radionuclides having small Derived Air Concentration (DAC) values, since other dust particles may overload the collection plates before detectable levels of the radionuclide in question are collected [18].

By far, when sampling radioactive particulate material, filtration is the most commonly used collection method [3]. Filtration media are manufactured from many different types of material, and they come in numerous shapes and sizes. The choices include many different types of filter papers that will be discussed later. For now, the mechanism that filter papers use to collect particulate material will be examined. Additionally, after explaining their mechanism of operation, some pros and cons for two common filtration methods involving filter papers will be examined.



Cascade impactor. The dust particles are collected on two standard 1×3 in glass microscope slides (stages 1, 2, 3, and 4) and on a membrane filter (stage 5). (Courtesy Union Industrial Equipment Co.)

Figure 2.4. Cascade Impactor.

Source: Herman Cember, *Introduction to Health Physics*, Pergamon Press, New York, pp. 402, 1985.

Filter papers incorporate two main mechanisms of operation. The first mechanism to be discussed uses impaction theory. This method traps particulate material throughout the filter matrix regardless of the filter's pore size. This occurs as particles succumb to inertial forces that propel them along in straight lines, even as the air bends and traverses through the filter matrix. If a particle strikes a filter's pore opening that's smaller than its own diameter, very likely it will become trapped. For this reason, the filtration efficiency of most filters increase over the sampling period because the availability of pore openings is diminished. Another possible scenario is for the particle to bounce off the filter media. In this case, other factors determine whether it will be recaptured and ultimately collected on the filter paper. Factors such as sampling rates, particle charge, and type of filter media may have an impact on the collection results. An advantage of this filtering method is an increased particle load capacity. This is especially true for filter papers made of paper products or glass fiber.

Additionally, since the collected particles are retained on the surface and throughout the filter's matrix, they are the best choice for high flow rates. Filters that operate by this principle are called depth filters, because filtration is achieved within the "depths" of the filter. Some common disadvantages arise from not having a clearly defined pore size. Consequently, the randomly constructed matrix places no definite upper limit on the size of particles that will pass through the filter's material matrix.

The second mechanism of filter paper operation is called sieving. The sieving method captures particles larger than the filter paper pore size on the surface of the filter paper. Another term used to describe this process is screening since just the top surface of the filter paper is exposed to the particulate material. Collecting particles only on the

surface of the filter allows particle size measurements because of the established upper size boundary imposed during manufacturing of these filters. This is an advantage that sieving has over the depth filtration method. A possible shortcoming of this method of filtration is that these filters have a low particle load capacity. In addition, the filters used in this method can clog rapidly when sampling air containing high particle concentrations.

Commercially, there is an extensive list of materials used to make filter paper media. Some choices of materials include paper products, glass fiber, and membrane varieties that may comprise a host of chemical compounds. For this study, care was taken to select the best type of filter paper media. Glass fiber and paper filters were not chosen because they lack a clearly defined pore size distinction. These types of filters fall in the category of depth filters, trapping and removing particles within its matrix. Since they lack a pore size distinction inherent to depth filters, they cannot be used to measure particle size. A membrane filter was selected for this study because its method of filtration is that of a screen filter.

Membrane filters, which have a pore opening that is certified and rigidly controlled during the manufacturing process, are ideal for applications requiring quantitative retention of particles, such as air monitoring of particles [19]. Their construction is extremely thin, retaining a minimal amount of moisture due to their non-hygroscopic nature. Membrane filters are commonly manufactured from materials like polycarbonate, polypropylene, polyvinylchloride, and cellulose ester compounds, to name just a few. The membrane filter chosen for this study was Millipore's Isopore™

Polycarbonate Track-Etched Membrane filter shown in Figure 2.5. More specifics on these filters will be provided in the description of equipment section.

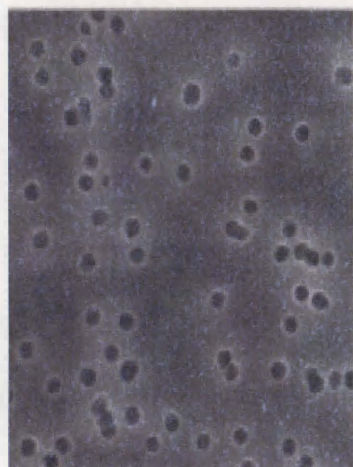
This sampling technique was chosen over others for a variety of reasons. First, and quite compellingly, because of the relative ease of acquiring sample data. Secondly, it was important this project achieve its intended purpose of providing useful sample data, while keeping an eye toward curtailing costs. The methodology for this work is supplemented by my experience using filtration sampling at Duratek. This work will combine the widely used second-stage filtration technique with a practical, cost-effective method of removing larger particles in the first stage.

2.2 Description of air sampler

The mechanical suction device used to collect representative samples was the low-volume air sampler shown in Figure 2.6. It's the model LV-14M, manufactured by F & J Specialty Products, Inc., of Ocala, Florida. This air sampler is oil-less, and it contains a carbon vane and vacuum pump with a constant flow regulator. This feature allows the air sampler to maintain a constant flow even during extended periods of sampling. In this study, the airflow regulator was adjusted to maintain 20 liters per minute (lpm) for the duration of the sampling period. This unit also incorporates an adjustable aluminum gooseneck in its design, which allows the sampling inlet to be placed at heights of 122 centimeters (cm), or 4 feet (ft.) to 213 cm (7 ft.) above the floor surface. The sample holder positioning for this type of study should be within 30 cm (~1 ft.) from the breathing zone of the typical radiation worker's mouth. The typical radiation worker's height is 175 cm (5 ft. 8.9 in.)[20]. The sample holder is adjusted to the breathing zone height above the floor surface of 165 cm (5 ft. 5 in.).



Isopore Polycarbonate Membrane Filters



Material

Polycarbonate film

Wettability

Hydrophilic [wetting agent: polyvinylpyrrolidone (PVP)]

Max. Temperature, °C

Autoclave: 121

Filtration: 140

Strength

Tensile: 200 kg/cm²

Burst: 1 bar min – unsupported membrane

Optical Properties

Translucent with slight green tinge

Weight

Ash: 0.6 µg/cm² – incinerates at 550–600 °C

Tare: 1 mg/cm² consistent – no need for matched-weight filters

Fluid Adsorption

Non-hygroscopic – not more than 0.24% increase > 24 hr in water.

For additional specifications, see the Filter Specifications Chart and Chemical Compatibility Chart.

Figure 2.5. Isopore™ Polycarbonate Track-Etched Membrane Filter.

Source: Millipore Products Catalog, Millipore Corporation, Copyright 1998.



Figure 2.6. F&J Specialty Products Low-Volume Air Sampler.

2.3 Description of filter paper

The filter paper chosen for this work was Millipore's IsoporeTM Polycarbonate Track-Etched Membrane filters. This type of filter is often called a screen filter since it retains particles only on its surface. Screen filter's method of filtration differs from depth filters, which achieves filtration within the depth of the filter material. Depth filters have an irregular, poorly defined pore structure, unlike the membrane filter that uses a unique straight-through pore, which allows only the penetration of particles smaller than its pore size.

For this reason, a membrane filter pore type was chosen because the pore opening size and uniformity is definable and strictly controlled during manufacturing, making it possible to predict the size of the largest passable particle.

Another consideration was potential adverse effects that humidity may have on the sample data. The technical specifications that accompanied the filters eased this concern. The technical description sheet states water vapor equilibrium is not necessary. Left unpackaged, these filters are non-hygroscopic and will gain no more than 0.24 percent water weight, even after 24 hours in water [19]. These filters are manufactured with several different outside diameter measurements. For this study, a standard 47 mm size diameter filter paper was selected.

2.4 Description of filter holder

The filter holder, shown in Figure 2.7, is comprised of two polished aluminum segments that are threaded for easy assembly. Each of the two segments has an indented surface that holds the 47 mm diameter filter in place. The outer filter paper is held in



Figure 2.7. Polished Aluminum Filter Holder. This filter holder was built exclusively for this thesis work. The filter holder shown here uses a standard width 47 mm outer diameter filter paper. The filter holder secures two filter papers held in series approximately two inches apart.

place with a retaining ring, while the inner filter paper is sandwiched between the two following segments. These filter holders are commonly made of plastic material, and are widely used in the Health Physics industry. Plastic materials may create static fields of charge around the filter holder that would negatively influence experimental results. For instance, a negatively charged filter holder would attract positively charged particulate matter but would repel the negatively charged ones. Of course, the converse would also hold true. As a result, polished aluminum filter holders were used to eliminate the possibility of static charge formation during the sample collection process.

2.5 Description of electronic balance

The Mettler AE200 electronic balance, shown in Figure 2.8, has a readout sensitivity of 0.1 milligram (mg), and a linearity of ± 0.3 mg. During weighing, the typical stabilization period is 5 seconds, and when stability is achieved, a green dot indicator light in the display goes out. With this level of sensitivity, room wind currents can influence readings. To help minimize errant reading, this scale contains a sliding glass door, which eliminates wind effects.

2.6 Description of Venturi air-flow calibrator

Digital Venturi airflow calibrators are controlled by a microprocessor and incorporate a digital display for easy readout. The unit is shown in Figure 2.9. Its LCD display will readout in units of Standard cubic feet per minute (SCFM), or Standard liters per minute (SLPM). The calibrator displays a flow value corrected to the standard temperature of 70 degrees Fahrenheit and a barometric pressure of 29.92 inches of



Figure 2.8. Mettler AE 200 Electronic Balance



Figure 2.9. Digital Venture Airflow Calibrator

mercury. The differential pressure versus flow relationship for the specific venturi flow sensor was calibrated by relating the differential pressure of the venturi flow sensor to the flow rate of a National Institute of Standard Instrument (NIST) traceable reference flow sensor. This was performed at a minimum of 10 different volumetric levels and was certified by the manufacturer. A factory calibration should be completed annually to verify the unit is within tolerance.

CHAPTER 3

EXPERIMENTAL METHOD

3.1 General description of sampling location and method

Having selected filtration as the sampling method of choice, the low-volume air sampler shown in Figure 2.6 was used to draw a sample of air through two filter papers held in series (see Figure 2.7). The low-volume air sampler was calibrated to maintain a continuous sampling flow rate of 20 lpm. Using this set-up, representative air samples were collected atop the MMF furnace deck shown in Figures 1.8 and 1.11. Each of the air samples was collected for 30 minutes to ensure adequate sample volume. This location was chosen because personnel routinely monitor the molten metal bath from this area. This location, because of the presence of the furnace, is a major contributor of airborne particulate radioactive material throughout the MMF. As a result, this area provides a suitable location to sample airborne radioactive material for the purpose of estimating the subsequent risk of exposure.

At the end of the 30-minute sampling period, the air sample filter papers were removed from the holder. Each filter paper was weighed twice prior to sampling and twice again while containing their representative sample. From the resultant sample weights, a mathematical analysis was completed using an Excel™ spreadsheet and the results were plotted using the computer software Sigma Plot™. Using this method, the MPAD value corresponds directly to the fifty percent probability found on the best-fit line containing the plotted data points. Additionally, a least squares method was used to calculate the projected area diameter from a determinate set of 14 mass equations

containing 9 unknown values developed from the sample data. This method uses matrix algebra, and is solved by calculating the least squares fit of the data discussed further in Chapter 4.

3.2 Sample collection difficulties

There are several collection difficulties encountered while performing this work. The Isopore™ Polycarbonate Track-Etched Membrane filters are extremely thin and flimsy. This made it difficult to handle them during collection and especially during the weighing process. During each weighing, care was taken not to excessively bump the weighing pan.

Additionally, the Mettler AE200 electronic balance with a sensitivity of 0.1 mg lacked the level of sensitivity that was needed to more effectively separate the sample weights. An electronic balance with a sensitivity of 0.01 mg or greater would be more appropriate.

Problems arise with the calibration of sampling devices for efficiency of dust removal versus the particulate size. This requires the use of particles that have a known size. Often this is very difficult to verify because of numerous underlying factors. Consequently, the reference literature at times made inferences that commercially available sampling devices may not meet the specifications of the sampling instruments used in the published results. Having made that statement, working with particles of known size would have been beneficial for calibration and for methods validation. This information would have allowed efficiency determinations to be made in this work for comparison.

3.3 Methods for determining particle size

One method of measuring particle size is to use a microscope to visually classify the particles. This approach is more easily defined for spherical particles, with the norm being a measurement of the particle diameter. For non-spherical particles, it is a different matter entirely. Definitions like Martin's diameter, Feret's diameter, or the projected area diameter are used to describe their sizes. These techniques are commonly used in powder technology, but their usefulness and value have diminished with the advancement of more generalized approaches. As a consequence, there is great variety in the instruments and methods available for particle measurement.

The more generalized approach is to classify particle size by a common term. The most widely used term to define particle size is to measure its aerodynamic equivalent diameter. This term is defined as the diameter of a unit density sphere that has the same settling velocity as the particle being studied. This term is defined based upon an objective performance measurement and not solely on the appearance of the particle [21]. That makes this approach more desirable because the size analysis is less subjective.

Methods that have been derived from other sizing techniques include the terminal settling velocity (in air), optical equivalent diameter, and the measurement of particle mass and volume using a laser. Some definitions emerged rapidly with technological advances of equipment like the optical particle counter. The optical equivalent diameter is defined as the diameter of a polystyrene latex particle that will scatter the same amount of light as the particle in question. Unfortunately, this technique relies to a large extent on the design of the particular optical counter [21]. None of these methods were

seriously considered due to the sizable cost associated with purchasing the necessary equipment.

An alternate approach to determine the particle size distribution is through construction of a size mass distribution using cascade impactor data. The scientific result is achieved by plotting the cumulative sampled mass (i.e., both the masses on the collection stage together with the masses on the succeeding stages versus a characteristic size for that stage). This is similar to what is accomplished through parallel cyclone sampling [22]. Again, this method is offered as an alternate approach for completeness, but was not included because of the cost considerations.

3.4 Calibration of low-volume air sampler

The model LV-14 air sampler was calibrated to a flow rate of 20 lpm. This flow rate was chosen to compliment the sample collection process and minimize the likelihood of filter loading. In addition, this flow rate was chosen to minimize particle bounce that could be followed by re-entrapment. The flow rate was monitored throughout the experiment to ensure the calibration adjustment remained constant.

3.5 Filter paper-weighing protocol

1. Check the calibration date on the electronic balance. Make sure its calibration is current. Make sure the instrument has been warmed up properly. This process takes 2 to 3 minutes according to the manufacturer.
2. Handle all filter papers with tweezers to minimize oily residue contamination caused when handling by hand.
3. Zero the balance prior to use. After each sample weighing, re-check the zero adjustment to make sure it has not been altered.

4. Weigh all filter papers at least twice prior to use. Ensure the precision of the two subsequent readings. If there is a greater than 0.1 mg difference between the two readings, determine the reason for this difference. Document the results.

5. Re-weigh at least twice all filter papers containing a sample to ensure consistent results. If there is a difference greater than 0.1 mg, determine the reason. Document the results.

CHAPTER 4

AIR-SAMPLING AND METHODOLOGY DISCUSSION

4.1 Air-sampling discussion

The membrane filters used in this work were manufactured by the Millipore Corporation, which has been in the business of making filter media for over 40 years. An area of specialty for them is the manufacture of several varieties of membrane filters. The specific type used in this study was Isopore™ Polycarbonate Track-Etched Membrane Filters. These filters are formed from a polycarbonate compound material, with each filter containing a uniform, cylindrical, straight-through pore design [19].

Nine different filter pore sizes, ranging in size from 0.05 μm to 5.0 μm , were chosen for use in this work. Different grouping combinations were chosen and each grouping used the following filter arrangement, namely, that each sample consists of two different sized filters placed in series within the aluminum filter holder. The aluminum filter holder secures the filter papers two inches apart with the filter containing the larger sized pores located in the outermost position, the assumption being that all particles larger than the pore size of the outer filter are captured on the outer filter's surface through the screening process. This absolute retention on the outer smooth surface of the pore prevents oversized particles from passing through to the inner filter. This process effectively sizes particles into two groups.

The first group contains particles whose size is greater than the outer filter's pore size. In this case, the particles have a diameter size greater than the outer filter pore size that effectively removes them at the surface by the screening process. Collectively, this

group of particles is defined as C_o , representing the mass of particulate collected on the “outer” filter. Any particles with a diameter smaller than those in the C_o group will pass through to the inner filter or beyond depending on their size.

The second group contains particles whose diameter size is larger than the pores of the inner filter, but smaller than those found in the outer filter. Accordingly, the size of particles found in this group lie somewhere between the outer and inner filter pore sizes. This group makes up the second constituent of the sample data. Collectively, this group of particles is defined as C_i , representing the mass of particulate collected on the “inner” filter.

A third group of particles exists which contains those particles with a smaller diameter size than both the outer and inner filter pore sizes. These particles pass directly through uncollected by either filter papers and are removed through the air sampler’s exhaust port.

4.2 Methodology discussion

The sample collection process requires repetitive samples for each of the filter paper pairings to be collected. A total of 50 samples are collected using 14 different filter paper grouping combinations. From the acquired sample data, two different approaches are used to determine the projected area diameter. The MPAD is the diameter where 50% of the mass (or volume) of the aerosol includes particles less than a given diameter. One method uses a least squares analysis with 14 mass equations that contain 9 unknown quantities, C_1 through C_9 , which represent the unknown particle diameter sizes found on the CDF. These 14 equations are set up in matrix form (see Figure 4.2) and were solved using both MathLab™ and Excel™ software, but each attempt provided inclusive results.

An alternate approach takes two groups of sample data and using an Excel™ spreadsheet examines the recursive nature of the sample mass measurements. This approach utilizes the repetitive sampling measurements and calculates an average ratio (mass collected on the inner filter relative to the mass collected on both filters) value for each of the filter grouping combinations. The average ratio is represented by the mass of particulate on the inner filter divided by the sum of particulate material mass on both the inner and outer filters. Calculating an average value for the grouping combination was done in order to minimize the effects of measurement errors. The average ratio values are then used in a series of iterative calculations to determine their corresponding values on the Cumulative Distribution Function (CDF). These average ratio values were gathered over a sampling interval that is continuous from 0.05 to 5.0 μm. The computer software program SigmaPlot™ is used to graph the calculated CDF values that corresponded to the inner filter diameters forming the continuous interval over the chosen sampling range.

Initially, some commonly used statistical terms used to describe the data will be defined. The sample population is represented by all the sampled masses collected through the experimental process. Specifically, the sample population is represented by the samples that are collected. Inherent to every experimental process involving data collection is sample randomness. A common way of presenting this phenomenon uses histograms sometimes referred to as a frequency diagram, or frequency distribution to better visualize the data. A frequency diagram (Figure 4.1) is often constructed to illustrate the relative frequency $f_i = n_i / N$ along the y-axis, while the sample observations

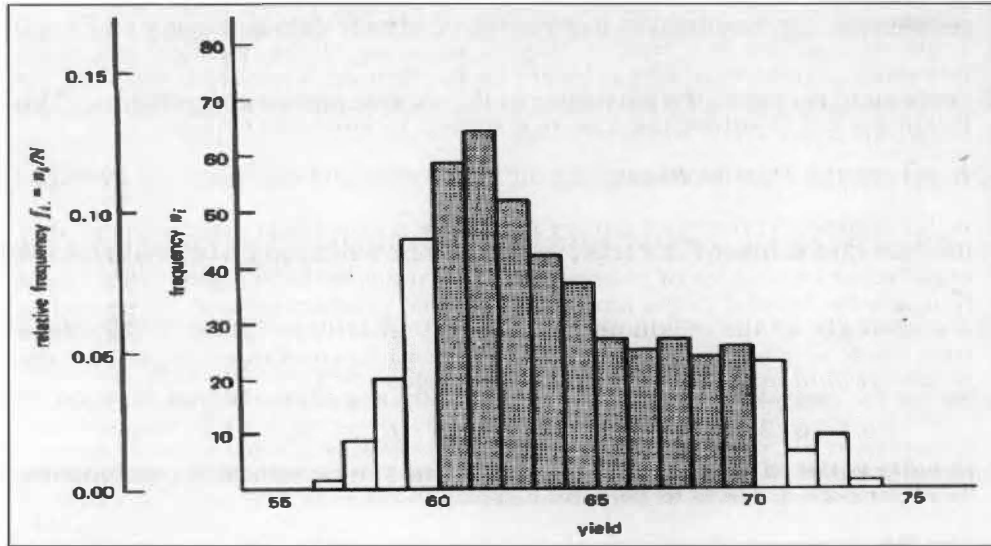


Figure 4.1 Bar graph Illustration of a Frequency Diagram.

are illustrated along the x-axis. The value of n_i is the number of observations in the i^{th} interval. The sample population represented by N can then be divided into i^{th} intervals, each represented by a rectangle, and collectively used to create a histogram. If the area of each rectangle constructed on the i^{th} interval were equal to the relative frequency of values in that interval, then the area under the entire histogram is equal to unity [23].

Over the grouping interval, the relative frequency estimates the probability of a randomly chosen particle being collected. Consequently, it's also called a probability distribution and the ordinate values along the vertical axis $p(x)$ may then be defined as the probability density. The probability density for a given interval is determined by dividing its probability by the length of the interval. As a result, the area under a probability distribution will represent its probability and the area under the whole distribution is equal to one. The ordinate values (i.e., probability densities) become a probability when they are multiplied by a given interval width. Hence, probability = probability density

multiplied by the interval size [23]. In this work, the pore size-grouping interval of 0.05 to 5.0 μm is the interval over which data were collected to obtain the probability distribution. We estimate the fraction of particles which are smaller than 0.05 μm , since no data was collected over the 0 to 0.05 μm interval. Accordingly, the expression $p(x)$ will be used to define the density function that represents the particle size distribution. The mass on the outer filter paper is given by:

$$M_{\text{outer filter}} = k \int_{S_0}^{\infty} p(x) dx \quad 4.1$$

Where the particle sizes collected vary from the outer filter size, S_0 , to infinity, and k is the normalization constant. The mass on the inner filter paper is given by:

$$M_{\text{inner filter}} = k \int_{S_i}^{S_0} p(x) dx \quad 4.2$$

where, the particle size varies from inner filter, S_i , to outer filter, S_0

In addition, the following equality holds true,

$$\int_{S_0}^{\infty} p(x) dx = 1 - C_0 \quad 4.3$$

and,

$$\int_{S_i}^{S_0} p(x) dx = C_{\text{outer filter}} - C_{\text{inner filter}} = C_0 - C_i \quad 4.4$$

In particular, if we form the ratio of the mass on the inner filter to the total mass on both filters, we can obtain a recursive relationship. The C_0 represents the mass of particulate collected on the outer filter, while the C_i represents the mass of particulate on the inner filter.

$$M_{\text{inner}} / (M_{\text{inner}} + M_{\text{outer}}) = (C_o - C_i) / [(C_o - C_i) + (1 - C_o)] = (C_o - C_i) / (1 - C_i) \quad 4.5$$

The “o” and “i” subscripts on the “C” values denote the filter placement as either “outer” or “inner,” respectively. Note: the outer filter paper is always the larger of the two pore sizes. The filter placement is varied in each of the 14 different filter paper grouping combinations such that repetitive samples are collected over each of the 14 different pore size intervals. Table 1 is a five-column table that contains the filter paper grouping combination designation, outer and inner filter sizes, each grouping’s average mass ratio, and the CDF equations.

The following example demonstrates the method of subscript substitution used to calculate the ratio of the sample mass for each of the filter paper grouping combinations. This particular example is for the “A” set of measurements. Table 2 lists the subscript designation for each of the filter pore sizes approximated along the CDF.

$$M_{\text{inner}} / (M_{\text{inner}} + M_{\text{outer}}) = (C_o - C_i) / (1 - C_i) = (C_2 - C_1) / (1 - C_1) = R_A$$

In the numerator, $C_2 - C_1$ represents the particles whose diameter size is between 0.2 and 0.05 μm . In the denominator, the $1 - C_1$, represents the particles with diameters greater than 0.05. The calculated average ratio values for each of the intervals are multiplied by the value $(1 - \text{CDF}_i)$ in an Excel™ spreadsheet. We chose a starting point on the CDF for the fraction of particles with diameters less than 0.05 μm , and recursively solved for the other points on the CDF. The product of the first calculation will be added to the starting point on the CDF, namely 0.02. (Numerous starting points were tried, but 0.02 was selected because it results in the best log normal distribution for the CDF. The best fit determination is based on a visual assessment of the goodness of fit on a probability plot and on symmetry in the geometric standard deviation.) This iterative

calculation is continued until each of the subsequent CDF values for the selected filter paper groupings is calculated.

By way of example, when making the first calculation, the value of $R(1-CDF_i)$ is chosen to be equal to 0.02. This value is the direct result of our estimation, since no data is collected for the 0 to 0.05 μm interval. This value corresponds to a 2 % chance of finding particles in this size interval. The first average mass ratio value, in this case either b-avg. or a-avg. shown in column 2 in Table 6, is multiplied by (1-0.02). The product will then be added to 0.02 and becomes the second value on the CDF. This CDF value is then subtracted from 1 and multiplied by the next average mass ratio value becoming the second $R(1-CDF_i)$ value shown in column 4 in Table 5. This iterative calculation is continued until each of the approximated CDF values in the sequence is calculated. Once all the probability values have been recursively solved, the cumulative distribution is plotted using SigmaPlot™ computer software. The graphed CDF is evaluated at 50 % probability to determine the MPAD value for the sampled particles.

The location of each filter paper pore size on the CDF is shown in Table 2 and the mass equations which contain the average ratio values are provided in Tables 3, 4, and 5. Table 3, shows the complete list of all 14 mass equations which make up the entire sampled mass population. Tables 4 and 5 show the selected mass equations that are used in SigmaPlot™ to graph probability Plots 1 and 2, Figures 5.1 and 5.2, respectively.

Table 1. Filter Paper Grouping Combinations				
Grouping Combination	S _o (outer size) Microns	S _i (inner size) Microns	Average Mass Ratio	CDF Equations
A	0.2	0.05	0.205	$(C_2-C_1)/(1-C_1)$
B	0.4	0.05	0.015	$(C_3-C_1)/(1-C_1)$
C	0.6	0.05	0.081	$(C_4-C_1)/(1-C_1)$
D	0.6	0.2	0.092	$(C_4-C_2)/(1-C_2)$
E	0.6	0.4	0.132	$(C_4-C_3)/(1-C_3)$
F	1.2	0.4	0.112	$(C_6-C_3)/(1-C_3)$
G	1.2	0.8	0.092	$(C_6-C_5)/(1-C_5)$
H	2.0	0.8	0.227	$(C_7-C_5)/(1-C_5)$
I	3.0	1.2	0.321	$(C_8-C_6)/(1-C_6)$
J	3.0	2.0	0.131	$(C_8-C_7)/(1-C_7)$
K	5.0	3.0	0.180	$(C_9-C_8)/(1-C_8)$
EA	0.8	0.05	0.137	$(C_5-C_1)/(1-C_1)$
EB	0.8	0.4	0.132	$(C_5-C_3)/(1-C_3)$
EC	0.8	0.6	0.128	$(C_5-C_4)/(1-C_4)$

Table 2. CDF Location	
Filter Size (μm)	CDF
0.05	C ₁
0.2	C ₂
0.4	C ₃
0.6	C ₄
0.8	C ₅
1.2	C ₆
2.0	C ₇
3.0	C ₈
5.0	C ₉

Table 3. All Mass Equations			
GROUPING COMBINATION	EQUATION	GROUPING COMBINATION	EQUATION
A	$C_2 = C_1 + R_A(1 - C_1)$	H	$C_7 = C_5 + R_H(1 - C_5)$
B	$C_3 = C_1 + R_B(1 - C_1)$	I	$C_8 = C_6 + R_I(1 - C_6)$
C	$C_4 = C_1 + R_C(1 - C_1)$	J	$C_8 = C_7 + R_J(1 - C_7)$
D	$C_4 = C_2 + R_D(1 - C_2)$	K	$C_9 = C_8 + R_K(1 - C_8)$
E	$C_4 = C_3 + R_E(1 - C_3)$	EA	$C_5 = C_1 + R_{EA}(1 - C_1)$
F	$C_6 = C_3 + R_F(1 - C_3)$	EB	$C_5 = C_3 + R_{EB}(1 - C_3)$
G	$C_6 = C_5 + R_G(1 - C_5)$	EC	$C_5 = C_4 + R_{EC}(1 - C_4)$

Table 4. Plot 1 Mass Equations	
GROUPING COMBINATION	EQUATION
R_A	$C_2 = C_1 + R_A(1 - C_1)$
R_D	$C_4 = C_2 + R_D(1 - C_2)$
R_{EC}	$C_5 = C_4 + R_{EC}(1 - C_4)$
R_G	$C_6 = C_5 + R_G(1 - C_5)$
R_I	$C_8 = C_6 + R_I(1 - C_6)$
R_K	$C_9 = C_8 + R_K(1 - C_8)$

Table 5. Plot 2 Mass Equations	
GROUPING COMBINATION	EQUATION
R_B	$C_3 = C_1 + R_B(1 - C_1)$
R_E	$C_4 = C_3 + R_E(1 - C_3)$
R_{EC}	$C_5 = C_4 + R_{EC}(1 - C_4)$
R_G	$C_6 = C_5 + R_G(1 - C_5)$
R_I	$C_8 = C_6 + R_I(1 - C_6)$
R_K	$C_9 = C_8 + R_K(1 - C_8)$

The equations for each mass measurement are given in Figure 4.2 below.

R_A-1	1	0	0	0	0	0	0	0	$\begin{bmatrix} C1 \\ C2 \\ C3 \\ C4 \\ C5 \\ C6 \\ C7 \\ C8 \\ C9 \end{bmatrix} = \begin{bmatrix} R_A \\ R_B \\ R_C \\ R_D \\ R_E \\ R_F \\ R_G \\ R_H \\ R_I \\ R_J \\ R_K \\ R_{EA} \\ R_{EB} \\ R_{EC} \end{bmatrix}$
R_B-1	0	1	0	0	0	0	0	0	
R_C-1	0	0	1	0	0	0	0	0	
0	R_D-1	0	1	0	0	0	0	0	
0	0	R_E-1	1	0	0	0	0	0	
0	0	R_F-1	0	0	1	0	0	0	
0	0	0	0	R_G-1	1	0	0	0	
0	0	0	0	R_H-1	0	1	0	0	
0	0	0	0	0	R_I-1	0	1	0	
0	0	0	0	0	0	R_J-1	1	0	
0	0	0	0	0	0	0	R_K-1	1	
$R_{EA}-1$	0	0	0	1	0	0	0	0	
0	0	$R_{EB}-1$	1	0	0	0	0	0	
0	0	0	$R_{EC}-1$	1	0	0	0	0	

Figure 4.2 Matrix Containing 14 Mass Equations.

The 14 x 9 matrix (Figure 4.2) is defined as A. The 14 x 1 vector of scalars is defined as R. The vector of unknown values that represents the CDF is defined as c.

Following substitution, the equations for the least squares estimate become:

$$Ax = y \quad 4.6$$

$$Ac = R \quad 4.7$$

$$A^T A c = A^T R \quad 4.8$$

$$c = (A^T A)^{-1} A^T R \quad 4.9$$

The values C_1 through C_9 representing a vector whose points are located along the CDF were calculated using MathLab™ and Excel™ computer software. Due to numerical uncertainties, the calculated solutions were inconclusive, yielding a solution of 1 for each unknown values. This approach uses all the sample data and is offered as an alternative method for calculating the projected area diameter of the sampled particles.

4.3 MMAD to AMAD Conversion

Regardless of the approach, what is being measured in this work is the particle's physical diameter. Consequently, the MPAD measured in this work must be corrected and expressed in terms of the aerodynamic diameter so it can be compared to AMAD values specified in regulatory guides.

The physical diameter for non-spherical particles may be defined by several types of diameters, like the Stokes diameter (d_{st}) and the aerodynamic (equivalent) diameter (d_{ae}). The aerodynamic equivalent diameter is the diameter of a unit density sphere that has the same terminal settling velocity as the particle being studied [26]. In most cases, an aerosol contains particles having a wide range of sizes that may also include a wide variety of shapes. If we assume the particles to be spherical in shape, the geometric diameter is the Stokes' diameter and its relationship to the aerodynamic diameter is as follows:

$$d_{ae} = d_p \sqrt{\rho C(d_p) / \sqrt{\rho^* C(d_{ae})}} \quad 4.10$$

where the value d_p is the projected area (or geometric) diameter, ρ is the particle density, $C(d_p)$ is the slip correction factor based on the diameter d_p , d_{ae} is the aerodynamic diameter, $C(d_{ae})$ is the slip correction factor based on the diameter d_{ae} , and the ρ^* is unit density (1 gm/cm^3) [26]. The slip correction factor based on diameter d_p and d_{ae} is:

$$C(d_p) = 1 + (\lambda / d_p) \{ 2.514 + 0.800 \exp[-0.55(d_p / \lambda)] \} \quad 4.11$$

$$C(d_{ae}) = 1 + (1 / P(\text{cm Hg})) [13.571 + 4.312 \exp(-0.1022 P d_{ae})] \quad 4.12$$

In the equations for both $C(d_p)$ and $C(d_{ae})$, the exponential term in particular, along with the other values in the parentheses, add little in their summation with unity. Essentially, both quantities, $C(d_p)$ and $C(d_{ae})$, when calculated are each nearly equal to unity. What remains after this simplification is the ratio of the square root of the particle densities multiplied by the geometric diameter d_p . If we assume the particulate is spherical-shaped metal with a density of 4 gm/cm^3 , and that the activity is distributed through the particles' volume. The AMAD of the particles sampled in this work equals $d_{ae} \cong 2.5 \sqrt{4} / \sqrt{1} = 5 \text{ }\mu\text{m}$.

Since almost all aerosols contain particles with a wide range of sizes, they are likely to be described by some type of size distribution function. Aerosols that are produced by a single mechanism are often easily represented by a log-normal distribution. The aerosols sampled in the MMF are largely created by one mechanism, the oxidation of metals by either the furnace or cutting operations. As a result, the CDF is expected to be log-normally distributed and to contain particles of varying shapes and sizes. "For homogeneously radioactive aerosols, the MMAD will be equal to the AMAD and, in this case, either MMAD or AMAD will be suitable to describe them"[26].

The CDF that describes the sampled particles in this work is graphed on logarithmic-probability scales. If the plotted data lie on a straight line, a log-normal distribution is a good approximation for the particle size distribution. Other possibilities exist like the mixing of two dusts from entirely different sources; this distribution may, or may not, be log-normally distributed. This type of distribution is termed bimodal.

CHAPTER 5

RESULTS

The Excel™ spreadsheet shown below (Table 6) is used to perform the necessary iterative calculations. In the spreadsheet, Column 5 represents the CDF values that are plotted by SigmaPlot™ along the Y axis. Column 6 represents the outer filter diameter values that are plotted along the X axis. The mass equations found in TABLE 3, are calculated below in column 2 under the heading AvgRatio. The method used to calculate Columns 4 and 5 is discussed in CHAPTER 4, Section 4.2.

Probability plots are made using different starting values for the CDF (see Figures 5.1 through 5.12). The probability plots and the calculated CDF values generated using 0.02 as the beginning CDF value yielded the best fit to a log-normal distribution. Drawing reference from probability Plots 1 and 2 (Figures 5.1 and 5.2), the MPAD of particles for the set of sample data shown in Table 6 is approximately 2.5 μm , having a geometric standard deviation (GSD) of 2.6. The GSD is taken from the upper portion of the plots, at the 84 % probability, where there is less scatter in the data. What occurred in the lower portion of the plots is not fully understood.

The spreadsheet in Table 6 is used to create probability plots 1 and 2, shown in Figures 5.1 and 5.2.

Table 6. Microsoft Excel™ Calculation Spreadsheet

1	2	3	4	5	6
PLOT 1					
	Avg.Ratio	C_i to C_o	$R(1-CDF_i)$	CDF	Inner filter diameter μm
		0 to 0.05	0.02	0.02	0.05
a-avg	0.205	0.05 to 0.2	0.2009	0.2209	0.2
d-avg	0.091887	0.2 to 0.6	0.0716	0.2925	0.6
ec-avg	0.127717	0.6 to 0.8	0.0904	0.3829	0.8
g-avg	0.092434	0.8 to 1.2	0.0570	0.4399	1.2
l-avg	0.180133	1.2 to 3.0	0.1009	0.5408	3
k-avg	0.320996	3.0 to 5.0	0.1474	0.6882	5
PLOT 2					
	Avg.Ratio	C_i to C_o	$R(1-CDF_i)$	CDF	Inner filter diameter μm
		0 to 0.05	0.02	0.02	0.05
b-avg	0.014554	0.05 to 0.4	0.0143	0.0343	0.4
e-avg	0.132499	0.4 to 0.6	0.1280	0.1622	0.6
ec-avg	0.127717	0.6 to 0.8	0.1070	0.2692	0.8
g-avg	0.092434	0.8 to 1.2	0.0675	0.3368	1.2
l-avg	0.180133	1.2 to 3.0	0.1195	0.4562	3
k-avg	0.320996	3.0 to 5.0	0.1745	0.6308	5

Probability Plot Versus Inner Filter Diameter

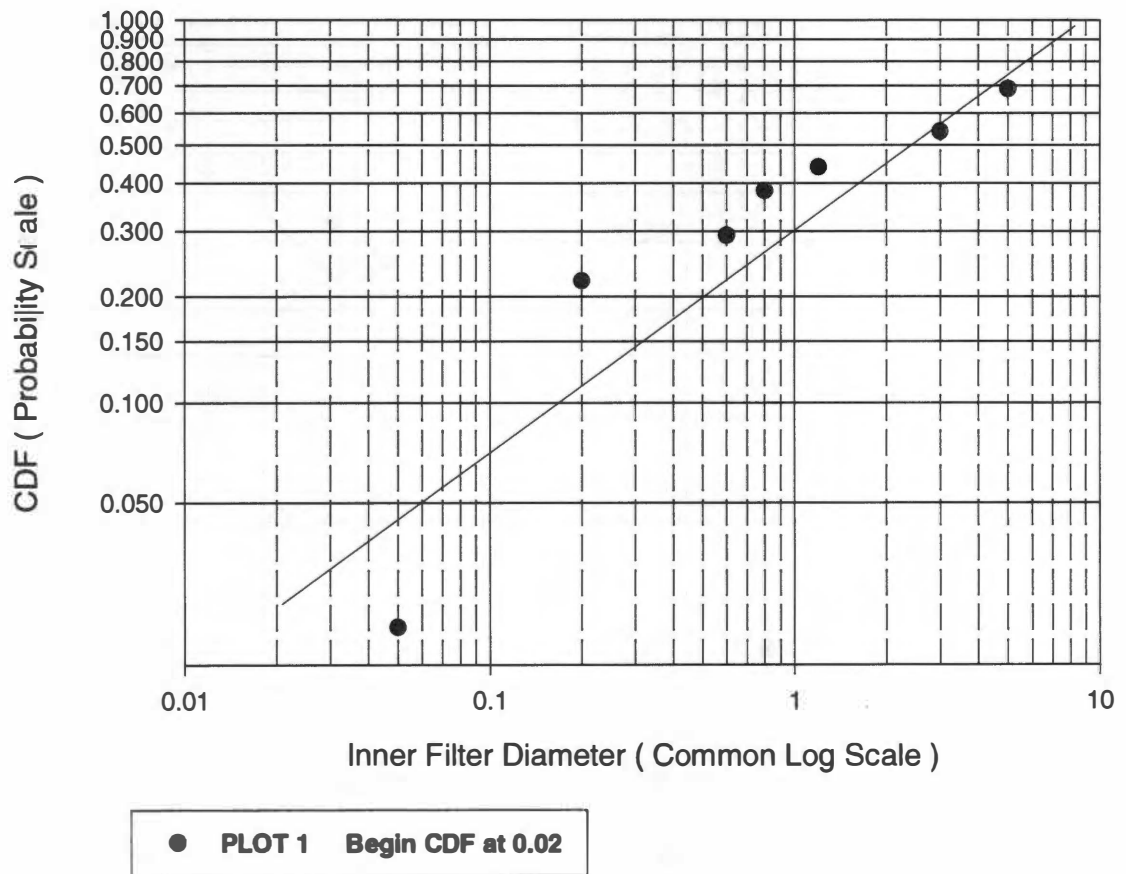


Figure 5.1 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

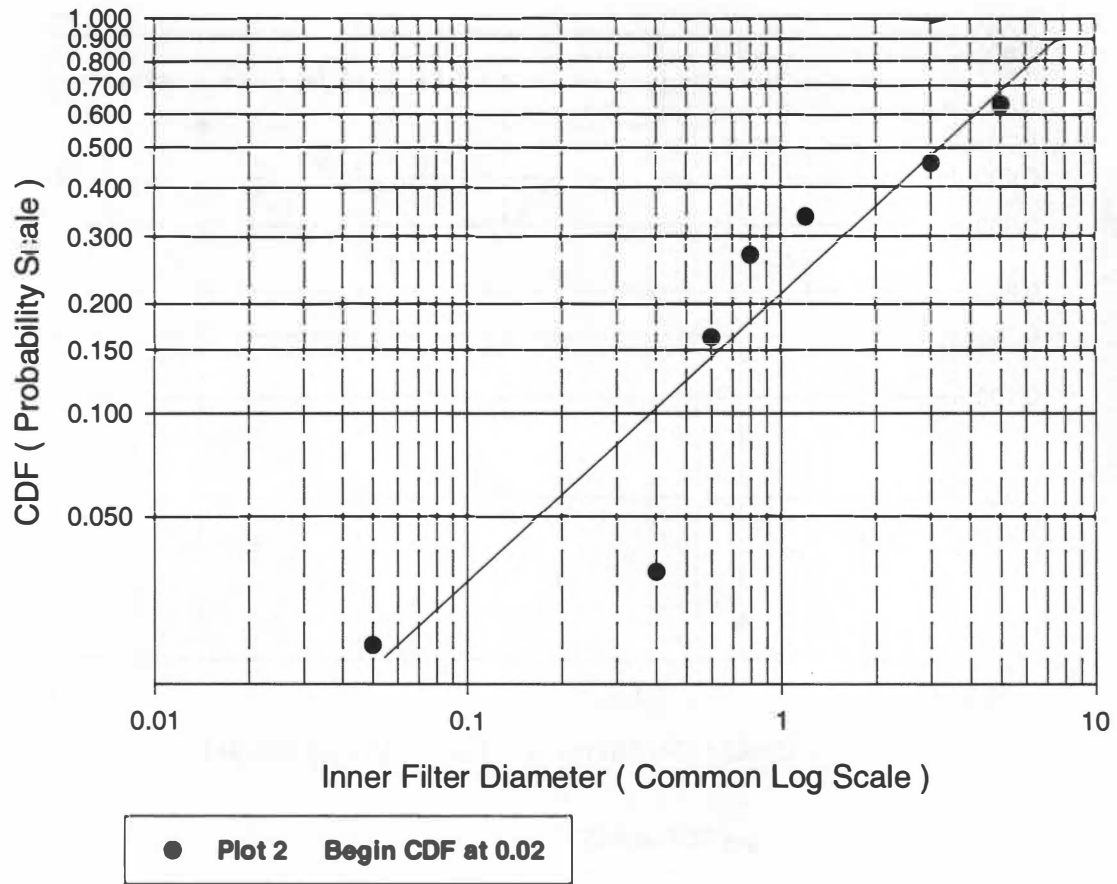


Figure 5.2 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

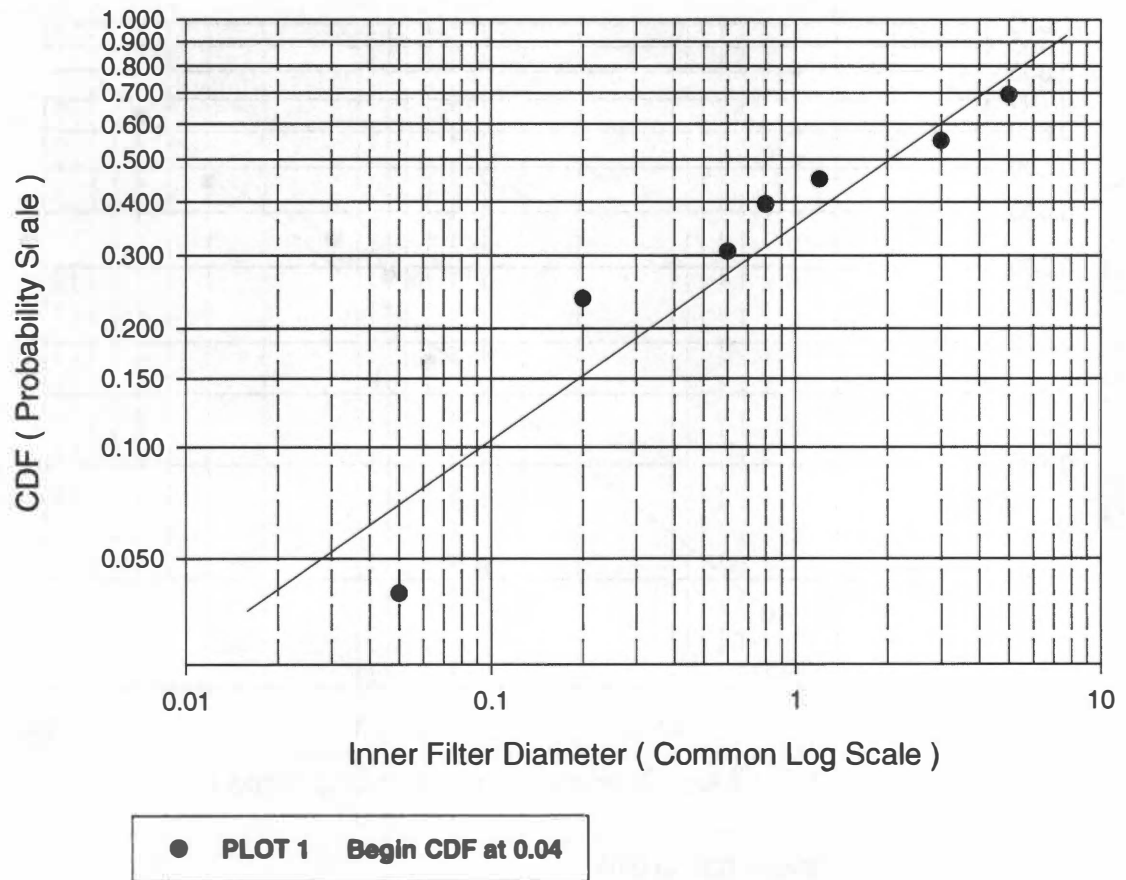


Figure 5.3 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

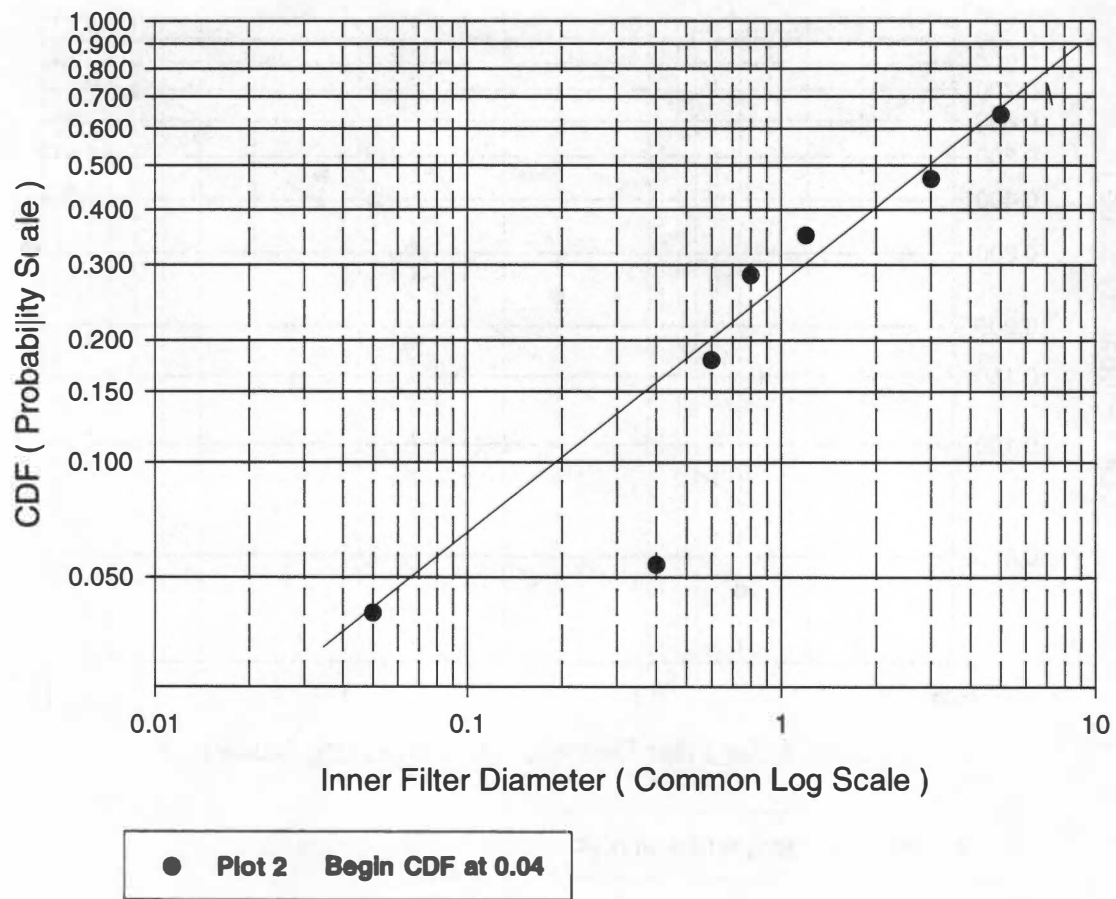


Figure 5.4 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

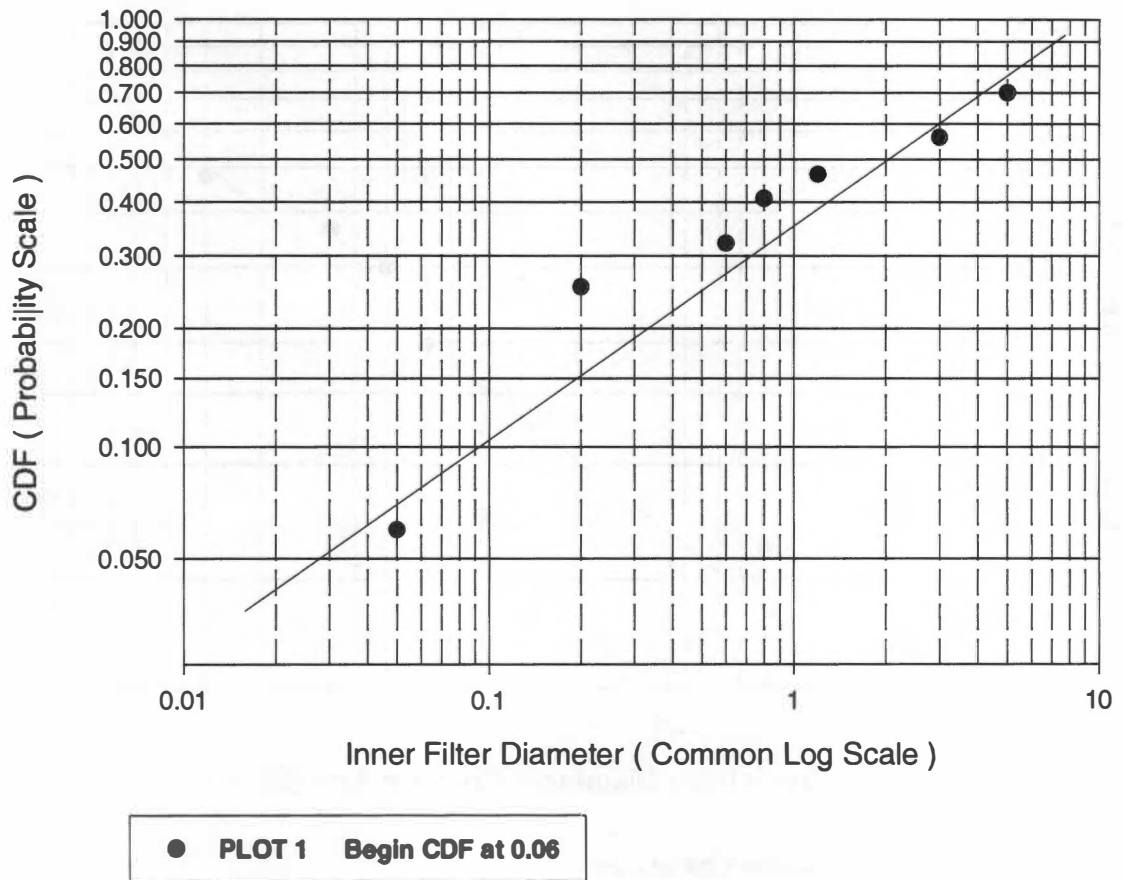


Figure 5.5 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

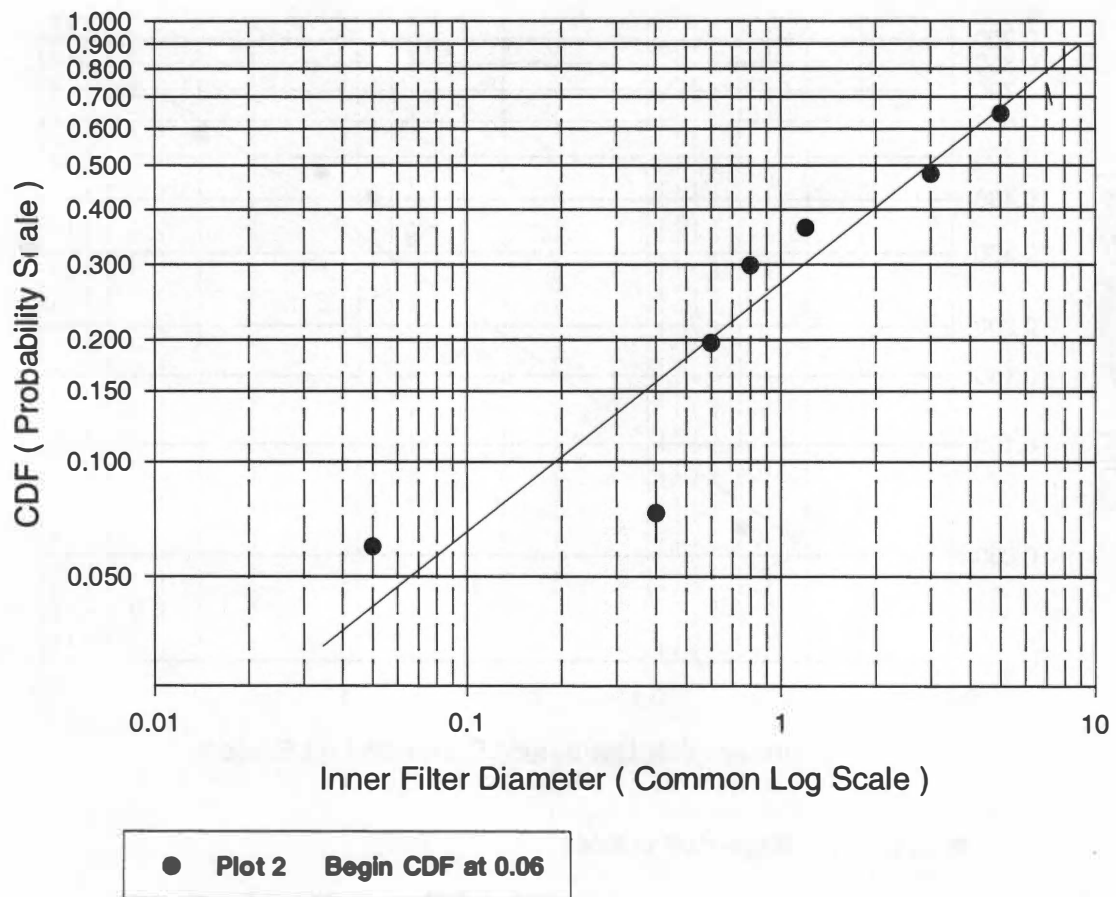


Figure 5.6 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

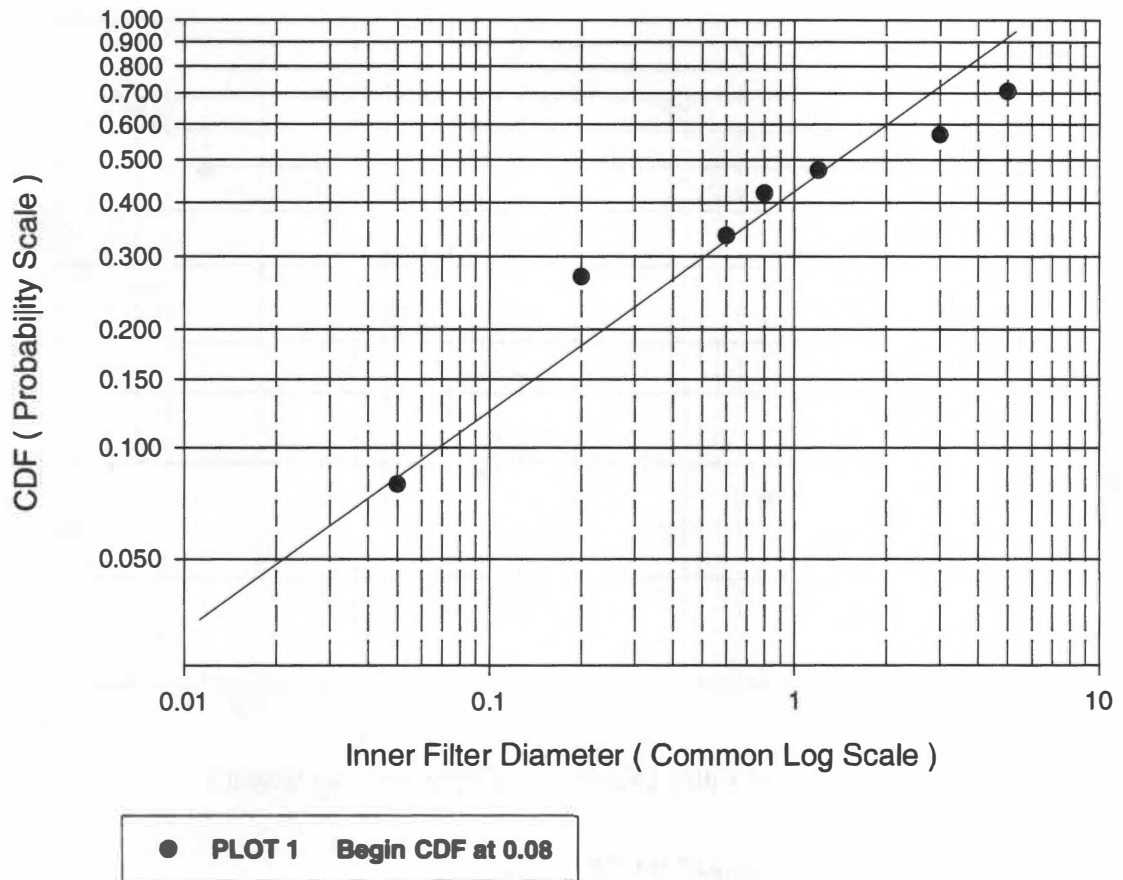


Figure 5.7 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

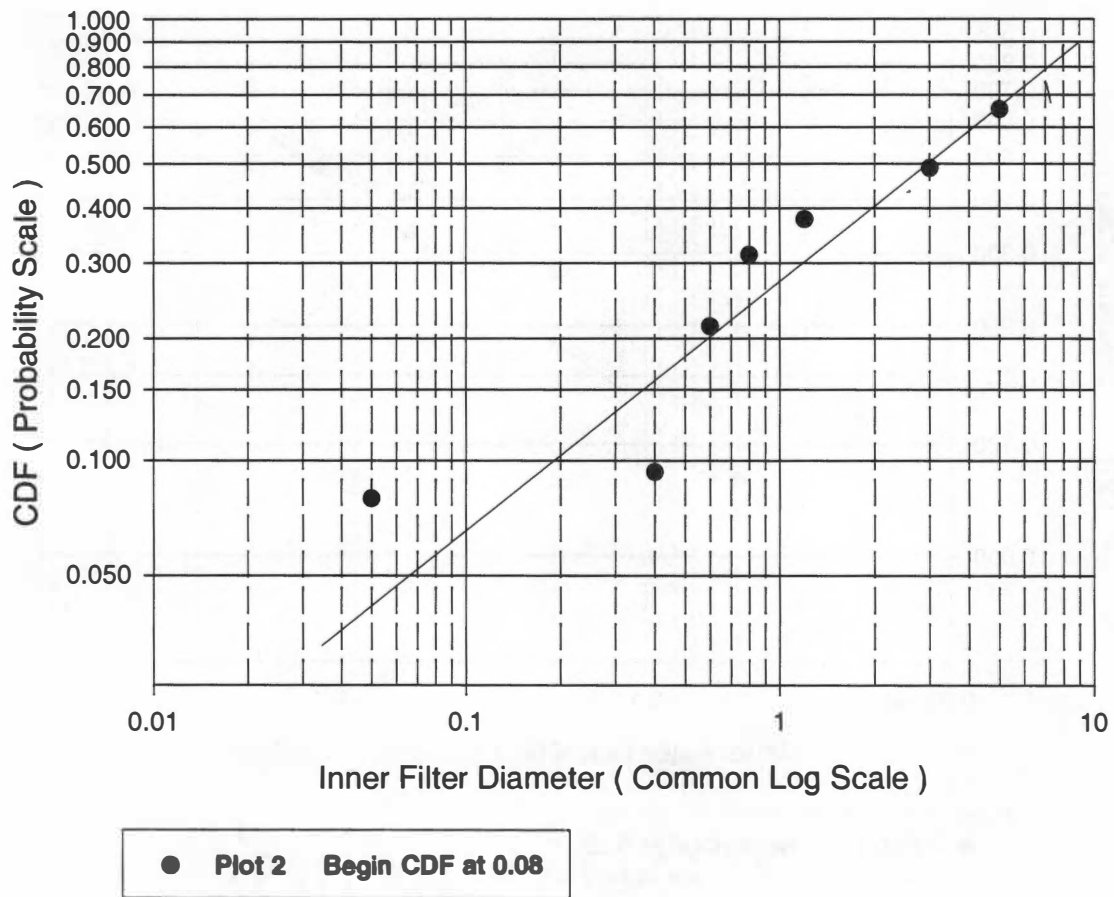


Figure 5.8 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

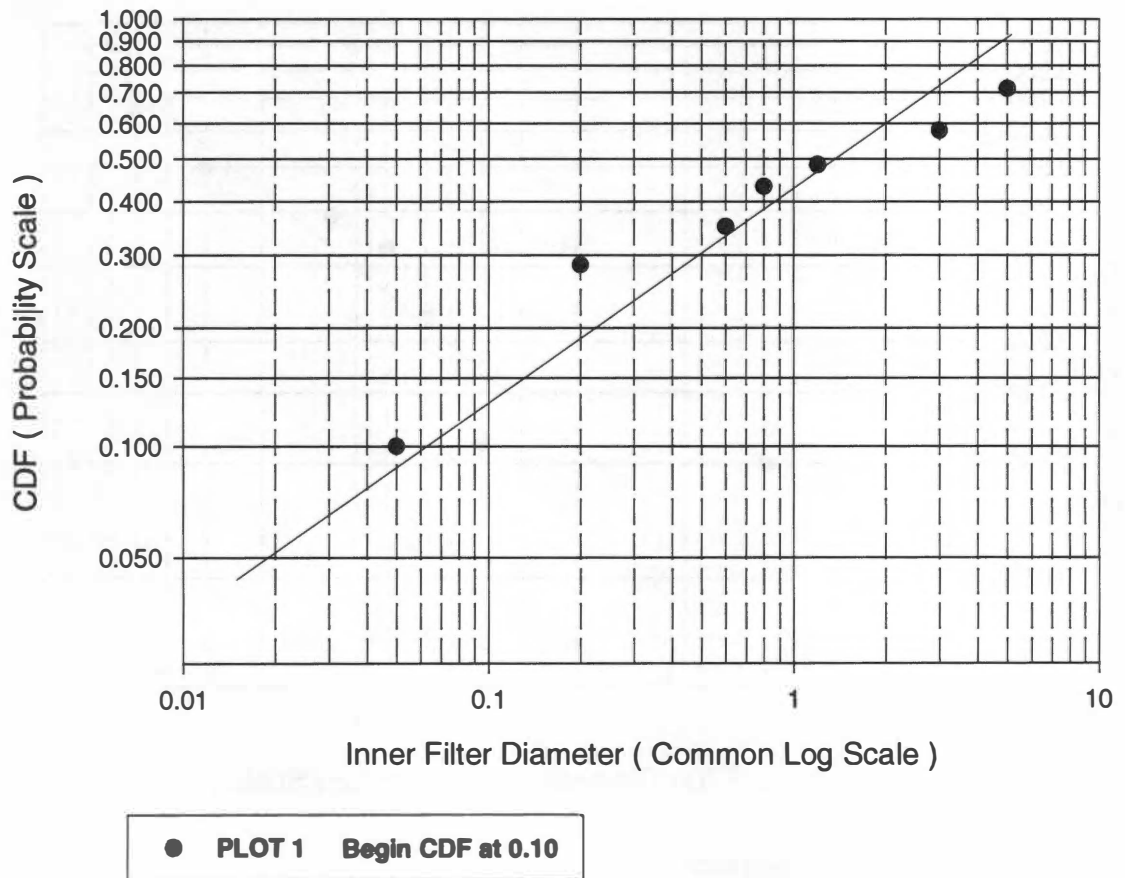


Figure 5.9 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

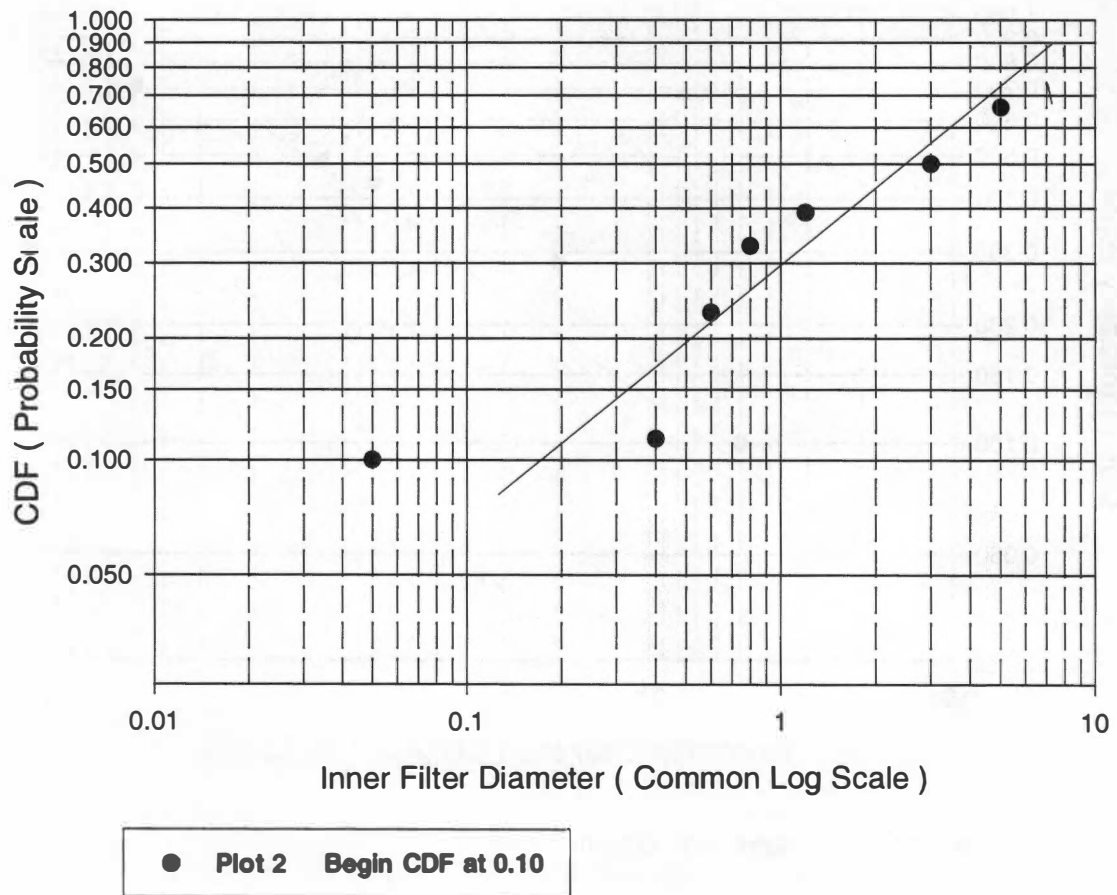


Figure 5.10 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Inner Filter Diameter

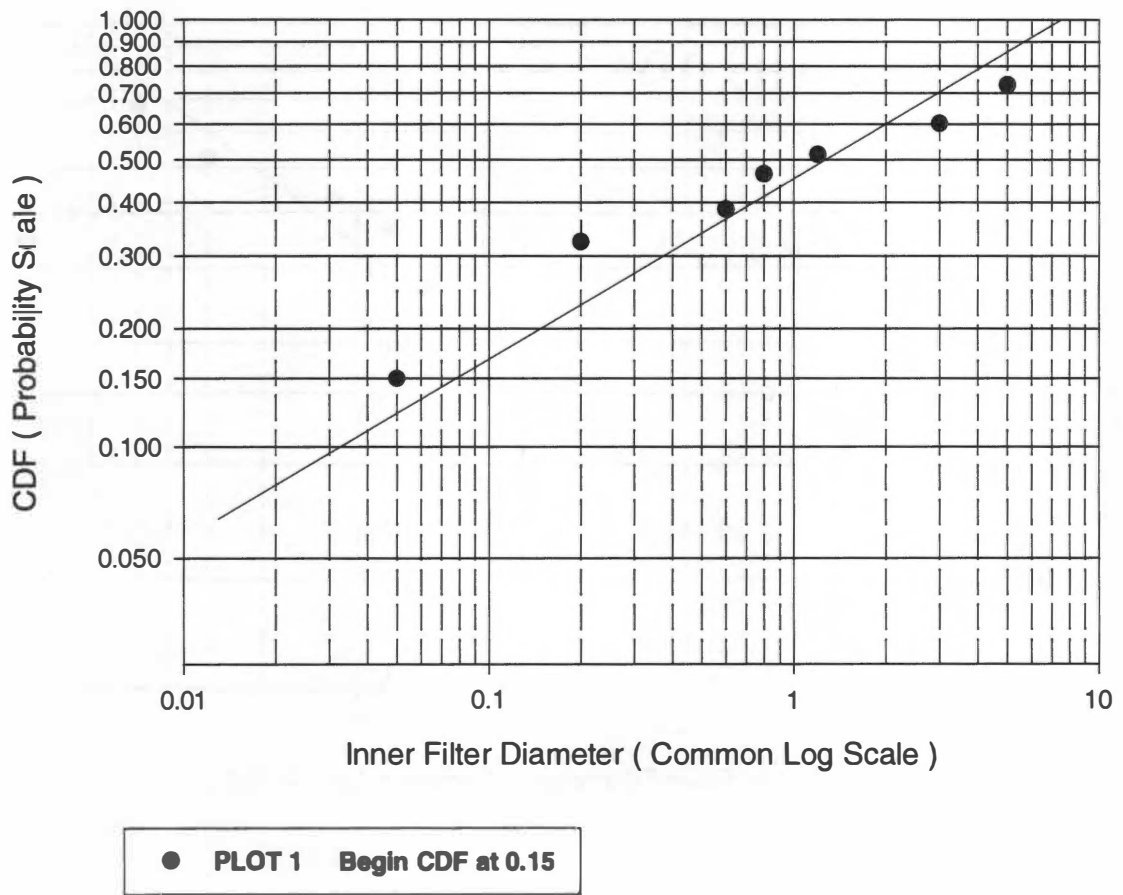


Figure 5.11 Probability Plot versus Inner Filter Diameter.

Probability Plot Versus Outer Filter Diameter

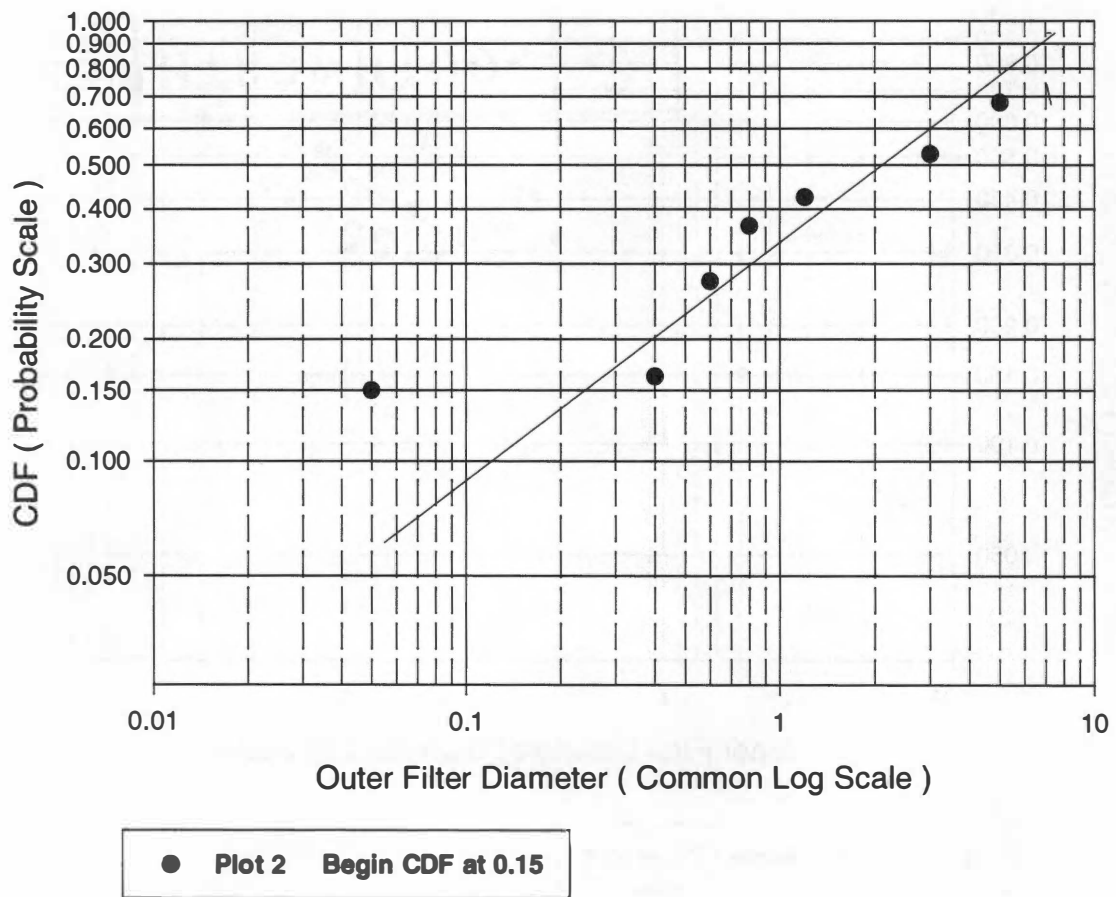


Figure 5.12 Probability Plot versus Inner Filter Diameter.

The objective of the work is to determine the median particle size of particulate material generated under normal operating conditions at Duratek's MMF. The value that is measured in this work is the MPAD, defined as the diameter where 50% of the mass (or volume) of the aerosol includes particles less than the given diameter. The MPAD can be corrected and realized in terms of the AMAD, or "the diameter in an aerodynamic particle size distribution for which the total activity above and below this size are equal. Usually, in so doing, a log-normal distribution of particle sizes is assumed"[26]. The result yields an MPAD value of approximately 2.5 μm , having a GSD of 2.6. An MPAD value of 2.5 μm correlates to an AMAD of 5 μm , which assumes the airborne material is spherical metal particulate having a density of 4 gm/cm^3 , and the activity is distributed through the particles' volume.

Based on this result, it can be stated that Duratek's requirement for workers to wear respirators while tending the furnace effectively provides a level of protection from airborne particulate material. Approximately 85 percent of all particles sampled have a diameter size greater than 0.3 μm . All of Duratek's respiratory protection devices provide a minimum protection standard of being 99.97 percent efficient against particulate aerosols larger than 0.3 μm .

The particle sizes are distributed mostly as log-normal. For the particle sizes to be considered log-normally distributed, the data points when plotted on logarithmic-probability scales should fall approximately on a straight line. This was the case for all the data points except for one. This result is illustrated in Figures 5.1 through 5.12. The divergence from log-normality in Plot 2 resulted from the unusually small ratio values using the 0.4 and 0.05 μm (outer & inner) pore size filter arrangement. This may indicate

that the outer filter's pores ($0.4\ \mu\text{m}$) became clogged and prevented the smaller particles from reaching the inner filter paper. Another explanation might be attributed to sample mass measurement inaccuracies. An Excel™ spreadsheet is included in Appendix B, which illustrates what each of the individual calculated ratio values were before becoming the AvgRatio value used in Chapter 5.

Additionally, following the analysis of sample data, it is evident that the overhead ventilation system provides a degree of protection against the median size of particulate material sampled during this work. Even under the most ideal conditions only a portion of the suspended particulate material is ever collected. The ventilation system provides a degree of protection by filtering air through three levels of filtration. The prefilters and intermediate filters are immediately followed by a high efficiency particulate air (HEPA) filter. Similar to the respirators worn by personnel in the MMF, the ventilation system's HEPA filters remove from the air at least 99.9% or more of $0.3\ \mu\text{m}$ diameter size particles.

Finally, the AMAD particle size of $5\ \mu\text{m}$ does differ from the $1\ \mu\text{m}$ value assumed by the current compartment deposition and clearance models. However, "Rarely (if ever) will the actual distribution of airborne particulates be completely characteristic of $1\ \mu\text{m}$ AMAD particles. Evaluating different particle size distributions can assist in explaining retention and excretion rates that are different than would be expected, based on standard modeling"[25]. Correcting intake estimates for particle size differences is an acceptable approach to evaluate what effect the size differences had on lung deposition and transfer models. If, for instance, the measured AMAD value was $20\ \mu\text{m}$ or larger, it would explain why these larger particles deposited preferentially in the

N-P region and have a fast clearance rate. The particles will have the short biological retention more typical of an ingestion intake than an inhalation intake because these particles will be cleared from the sinuses by the throat and swallowed.

CHAPTER 6

CONCLUSION

The procedural requirement for wearing respiratory protection devices in the MMF is supported by the results which find the particles are small enough to be respirable. In general, particles of concern as a health hazard generally are considered to be smaller than 10 μm in diameter. These particles are small enough to reach the alveoli (pulmonary) region of the lungs. The epithelium cells in this region are non-ciliated; consequently, any intrusive insoluble particle deposited in this region will be removed at a very slow rate.

The respiratory protection currently in use in the MMF provides a degree of protection from airborne particulate material. All the respirators currently in use are fitted with filters that remove 99.97% of particles greater than 0.3 μm . Approximately 85% of all particles sampled have a diameter size that is greater than 0.3 μm . Therefore, respiratory protection is required for all entries into the metal processing contaminated areas.

This work found that the MMF engineering controls provide a degree of protection against the median diameter size particulate material sampled. The overhead ventilation systems located above the furnace and in the cutting/sizing areas remove airborne particulate material. These systems pull air through a series of 3 filters the last one being a HEPA filter which removes 99.97% of particles greater than 0.3 μm . The ventilation system operates 24 hours a day, 7 days a week—not just during processing hours.

The particle sizes were distributed log-normally, as demonstrated by the data points when plotted on logarithmic-probability scales, falling approximately on a straight line. This would indicate that the particles are generated by a single source, representing a monomodal distribution. If this had been the mixing of two dusts from entirely different sources, a multimodal distribution would result. Most likely, this distribution would not be log-normal.

The mathematical analysis of the sample data yielded an MPAD value of 2.5 μm . This value correlates to an AMAD value of 5 μm , which assumes the particles are spherical, having a physical density of 4 g/cm^3 with the activity distributed throughout the particles' volume. This AMAD value will compare favorably to the 5 μm value listed in ICRP 66, which lists compartmental deposition percentages for both 1 and 5 μm diameter values.

Finally, the AMAD measured in this work is a factor of 5 larger than the default value of 1 μm currently used in ICRP 30. This result means there is no direct comparison between the aerodynamic diameter measured to that which the model is based upon. Derived guides for other AMAD values can be computed from information in ICRP 30. For any definitive action to be taken to alter Duratek's current internal dosimetry procedures much further study would be required.

CHAPTER 7

SUGGESTED FUTURE STUDY

A costly but straight forward approach for measuring a particle size distribution is to use a cascade impactor. Using this instrument saves time by partitioning the distribution of particle sizes each time a sample is collected. As a result, this method reduces the total number of samples that are needed. The acquired sample(s) is(are) then weighed for an accurate mass analysis in each size fraction. Consequently, this method reduces the amount of mathematical interpolation necessary to determine the particle size distribution.

Another instrument that can be used is a cyclone collection device. This instrument has an upper size cutoff, regulating the size of the particle that is allowed to pass. A predetermined fraction of each particle size penetrates to the second-stage collection device which retains all particles passing through the cyclone. The mass of particles for each cutoff size is plotted and combined to determine the total particle size distribution. As with the last method, the MMAD value is readily available once the size distribution is known.

In the event a continuation of the current work is initiated, the following suggestions should be considered. In the current work, the electronic balance used to weigh the mass samples had a readout sensitivity of 0.1 milligrams. A microbalance with a higher degree of sensitivity would improve the ability to separate the sample mass measurements. This increased separation provides a higher degree of precision, thereby making the sample plots more useful.

The current work uses a sampling device with an unknown collection efficiency. An improvement would be to complete a collection efficiency study for each membrane filter size using the current sampling method and device. This is performed using a particle generator that produces particles of known size.

The air sample data collected for this work was obtained on 4 days over an eight month period. To more accurately quantify the AMAD of airborne particulate material within the MMF, a more comprehensive study is needed. More detail should be given to determine how different processing operations affect facility air movement patterns. An emphasis should be placed on studying how metal ore composition and production throughput affects airborne particle concentration. Finally, due to the seasonal changes inherent in this area, a study on what affect climate variability has would also be useful.

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APPENDICES

APPENDIX A

Sample Data

Filter Paper Pore Size Grouping			Outer filter	Inner filter	Collection Date
			0.2 micron	0.05 micron	
A1	Prewrite (mg)		30.5	37.7	11/12/99
	Postweight (mg)		30.9	37.8	
	Difference		0.4	0.1	
A2	Prewrite (mg)		30.2	37.8	6/14/99
	Postweight (mg)		31	38.1	
	Difference		0.8	0.3	
A3	Prewrite (mg)		30.5	37.3	3/17/99
	Postweight (mg)		32.2	37.8	
	Difference		1.7	0.5	
A4	Prewrite (mg)		30.9	37.4	11/18/99
	Postweight (mg)		33.1	37.7	
	Difference		2.2	0.3	
			0.4 micron	0.05 micron	
B1	Prewrite (mg)		15.3	37.5	11/12/99
	Postweight (mg)		20.7	37.6	
	Difference		5.4	0.1	
B2	Prewrite (mg)		15.1	37.9	11/18/99
	Postweight (mg)		22.8	38	
	Difference		7.7	0.1	
B3	Prewrite (mg)		15.1	37.4	6/14/99
	Postweight (mg)		22.9	37.5	
	Difference		7.8	0.1	
			0.6 micron	0.05 micron	
C1	Prewrite (mg)		31.7	37.4	11/18/99
	Postweight (mg)		45.9	38.1	
	Difference		14.2	0.7	
C2	Prewrite (mg)		31.5	37.3	11/12/99
	Postweight (mg)		40.3	38.5	
	Difference		8.8	1.2	
C3	Prewrite (mg)		31.5	37.4	3/17/99
	Postweight (mg)		41.2	38.2	
	Difference		9.7	0.8	
			0.6 micron	0.2 micron	
D1	Prewrite (mg)		32.7	31	11/18/99
	Postweight (mg)		45.5	31.8	
	Difference		12.8	0.8	

Filter Paper Pore Size Grouping		Outer filter	Inner filter	Collection Date
D2	Prewrite (mg)	31.4	30.8	11/12/99
	Postweight (mg)	40.3	31.7	
	Difference	8.9	0.9	
D3	Prewrite (mg)	32.7	30.4	3/17/99
	Postweight (mg)	41.8	31.7	
	Difference	9.1	1.3	
		0.6 micron	0.4 micron	
E1	Prewrite (mg)	31.5	14.9	3/17/99
	Postweight (mg)	33.4	15.3	
	Difference	1.9	0.4	
E2	Prewrite (mg)	31.8	14.9	11/12/99
	Postweight (mg)	34.8	15.5	
	Difference	3	0.6	
E3	Prewrite (mg)	31.5	15.2	11/18/99
	Postweight (mg)	38	16.1	
	Difference	6.5	0.9	
E4	Prewrite (mg)	31.5	15	6/14/99
	Postweight (mg)	37	15.4	
	Difference	5.5	0.4	
		1.2 micron	0.4 micron	
F1	Prewrite (mg)	27.3	15.2	11/12/99
	Postweight (mg)	34.1	15.9	
	Difference	6.8	0.7	
F2	Prewrite (mg)	27.4	15.1	11/18/99
	Postweight (mg)	34.9	15.7	
	Difference	7.5	0.6	
F3	Prewrite (mg)	26.8	15.1	3/17/99
	Postweight (mg)	31.2	16	
	Difference	4.4	0.9	
		1.2 micron	0.8 micron	
G1	Prewrite (mg)	27.2	30.5	11/18/99
	Postweight (mg)	34.9	30.9	
	Difference	7.7	0.4	
G2	Prewrite (mg)	26.7	30	3/17/99
	Postweight (mg)	30.7	30.5	
	Difference	4	0.5	

Filter Paper Pore Size Grouping		Outer filter	Inner filter	Collection Date
G3	Preweight (mg)	26.3	30.4	11/12/99
	Postweight (mg)	32.5	30.9	
	Difference	6.2	0.5	
G4	Preweight (mg)	26.8	30.4	6/14/99
	Postweight (mg)	31.3	31.1	
	Difference	4.5	0.7	
		2.0 micron	0.8 micron	
H1	Preweight (mg)	31.3	29.9	11/18/99
	Postweight (mg)	32.4	30.2	
	Difference	1.1	0.3	
H2	Preweight (mg)	31	30.1	6/14/99
	Postweight (mg)	31.6	30.4	
	Difference	0.6	0.3	
H3	Preweight (mg)	30.7	30	6/14/99
	Postweight (mg)	32.8	30.4	
	Difference	2.1	0.4	
H4	Preweight (mg)	28.4	26.9	3/17/99
	Postweight (mg)	28.8	27	
	Difference	0.4	0.1	
		3.0 micron	1.2 micron	
I 1	Preweight (mg)	28.6	26.7	11/12/99
	Postweight (mg)	30.5	27	
	Difference	1.9	0.3	
I 2	Preweight (mg)	27.9	27.6	11/12/99
	Postweight (mg)	28.1	27.9	
	Difference	0.2	0.3	
I 3	Preweight (mg)	27.8	27.2	6/14/99
	Postweight (mg)	28	27.3	
	Difference	0.2	0.1	
I 4	Preweight (mg)	28.3	27	3/17/99
	Postweight (mg)	29.4	27.3	
	Difference	1.1	0.3	
		3.0 micron	2.0 micron	
J1	Preweight (mg)	27.8	31.3	11/18/99
	Postweight (mg)	38.3	31.9	
	Difference	10.5	0.6	

Filter Paper Pore Size Grouping		Outer filter	Inner filter	Collection Date
J2	Preweight (mg)	28.2	30.7	3/17/99
	Postweight (mg)	30.5	31.4	
	Difference	2.3	0.7	
J3	Preweight (mg)	28.7	31.2	11/12/99
	Postweight (mg)	34.2	31.8	
	Difference	5.5	0.6	
J4	Preweight (mg)	28.3	30.9	6/14/99
	Postweight (mg)	34	31.8	
	Difference	5.7	0.9	
		5.0 micron	3.0 micron	
K1	Preweight (mg)	28.5	28.8	3/17/99
	Postweight (mg)	28.7	29	
	Difference	0.2	0.2	
K2	Preweight (mg)	28.7	28.5	6/14/99
	Postweight (mg)	31.1	28.8	
	Difference	2.4	0.3	
K3	Preweight (mg)	28.8	28.6	11/18/99
	Postweight (mg)	35.1	28.8	
	Difference	6.3	0.2	
K4	Preweight (mg)	28.7	28.7	11/12/99
	Postweight (mg)	36.9	29.4	
	Difference	8.2	0.7	
		0.2 micron	0.05 micron	
L1	Preweight (mg)	30.5	37.6	6/14/99
	Postweight (mg)	30.8	37.7	
	Difference	0.3	0.1	
L2	Preweight (mg)	30.4	37.7	11/12/99
	Postweight (mg)	30.6	37.9	
	Difference	0.2	0.2	
L3	Preweight (mg)	30.5	38.0	11/12/99
	Postweight (mg)	30.8	38.2	
	Difference	0.3	0.2	
		0.8 micron	0.05 micron	
EA1	Preweight (mg)	30.5	38.1	3/17/99
	Postweight (mg)	31.6	38.3	
	Difference	1.1	0.2	

Filter Paper Pore Size Grouping		Outer filter	Inner filter	Collection Date
EA2	Preweight (mg)	30.4	38.2	6/14/99
	Postweight (mg)	32.7	38.5	
	Difference	2.3	0.3	
EA3	Preweight (mg)	30.5	38.1	11/12/99
	Postweight (mg)	31.7	38.3	
	Difference	1.2	0.2	
		0.8 micron	0.4 micron	
EB1	Preweight (mg)	30.4	15.2	6/14/99
	Postweight (mg)	34.8	15.5	
	Difference	4.4	0.3	
EB2	Preweight (mg)	30.4	15.2	11/18/99
	Postweight (mg)	30.8	15.3	
	Difference	0.4	0.1	
		0.8 micron	0.6 micron	
EC1	Preweight (mg)	30.4	30.5	11/18/99
	Postweight (mg)	32.5	30.8	
	Difference	2.1	0.3	
EC2	Preweight (mg)	30.5	31.7	11/18/99
	Postweight (mg)	32.5	32.0	
	Difference	2.0	0.3	

APPENDIX B
Excel Calculation Worksheet

Measurement			Avg. Ratios	Filter Paper Sizes	Filter Size (microns) Pore Sizes	CFD CDF Pt	Integral
a1	0.2	a-avg	0.205	0.05 to 0.2	0.05	1	0.1
a2	0.272727	b-avg	0.014554	0.05 to 0.4	0.2	2	0.1845
a3	0.227273	c-avg	0.081057	0.05 to 0.6	0.4	3	0.013098
a4	0.12	d-avg	0.091887	0.2 to 0.6	0.6	4	0.072951
a-avg	0.205	e-avg	0.132499	0.4 to 0.6	0.8	5	0.123626
b1	0.018182	f-avg	0.112406	0.4 to 1.2	1.2	6	0.110934
b2	0.012821	g-avg	0.092434	0.8 to 1.2	2	7	0.198853
b3	0.012658	h-avg	0.226905	0.8 to 2.0	3	8	0.285387
b-avg	0.014554	i-avg	0.320996	1.2 to 3.0	5	9	0.128725
c1	0.04698	j-avg	0.130528	2.0 to 3.0			
c3	0.12	k-avg	0.180133	3.0 to 5.0			
c3	0.07619	Ea-avg	0.137363	0.05 to 0.8			
c-avg	0.081057	Eb-avg	0.131915	0.4 to 0.8			
d1	0.058824	Ec-avg	0.127717	0.6 to 0.8			
d2	0.091837						
d3	0.125						
d-avg	0.091887	b-avg	0.014554	0.05 to 0.4			
e1	0.173913	e-avg	0.132499	0.4 to 0.6			
e2	0.166667	Ec-avg	0.127717	0.6 to 0.8			
e3	0.121622	g-avg	0.092434	0.8 to 1.2			
e4	0.067797	i-avg	0.320996	1.2 to 3.0			
e-avg	0.132499						
ea-avg	0.137363						
eb-avg	0.131915						
ec-avg	0.127717						
f1	0.093333						
f2	0.074074						
f3	0.169811						
f-avg	0.112406						
g1	0.049383						
g2	0.111111						
g3	0.074627						
g4	0.134615						
g-avg	0.092434						
h1	0.214286						
h2	0.333333						
h3	0.16						
h4	0.2						
h-avg	0.226905						
i1	0.136364						
i2	0.6						
i3	0.333333						
i4	0.214286						
i-avg	0.320996						
j1	0.054054						
j2	0.233333						
j3	0.098361						
j4	0.136364						
j-avg	0.130528						
K1	0.5						
K2	0.111111						
K3	0.030769						
K4	0.078652						
k-avg	0.180133						

Data used to create Sigmaplot™ graphs

VITA

Mr. Brett Sumner has been working in the Health Physics field for 16 years. He earned an Associate Degree from Central Florida Community College in 1985 and his Bachelor of Science from Frances Marion University in 1991. Mr. Sumner has been employed by Duratek, Inc. for the past 10 years. His current duties include providing readiness reviews for high radiation projects, and performing dose-to-curie shielding analysis using Microshield™ dose modeling software to support ongoing projects.