A COMPARISON OF METHODS FOR MEASURING AND MONITORING PARTICULATE MATTER DOWNSTREAM OF HEPA FILTERS

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ABSTRACT

High Efficiency Particulate (HEPA) filters are defined to have a removal efficiency of at least 99.97% for particles 0.3 micrometers or larger. Filtering efficiency is routinely determined with an aerosol photometer using DOP (dioctyl phthalate) smoke as the challenge agent. This testing does not, however, directly answer the question of how much particulate matter penetrates a HEPA filter while the filter is actually in use. To answer this question, one would require the ability to measure both particle size distribution and number density downstream of the HEPA filter. Variability in the size distribution, chemical matrix of the particles, and particle density lead to significant uncertainty in such measurements. In spite of these considerable difficulties, increasing public concern over possible emissions and increasing regulatory scrutiny may eventually require that operators continuously monitor the particulate matter (PM) concentration downstream of HEPA filters.

A number of standards may be applicable to these measurements, including those issued by US-EPA, US-DOE, ASME, IEST, and ASTM. Because the expected particle concentration downstream of a properly functioning HEPA filter is so low, the statistics of particle counting and the application of uncertainty analysis are important considerations.

DIAL has undertaken a project to measure the performance of HEPA filters, using challenge aerosols other than DOP. We have employed four different techniques for these measurements:

1) EPA Reference Method 5i – gravimetric.
2) Electrical Low-Pressure Impactor (ELPI) – determination of PM mass concentration, particle size distribution and number density concentration.
3) Scanning Mobility Particle Sizer (SMPS) - electric mobility determination of PM mass concentration, particle size distribution and number density concentration.
4) Diffusion Battery and Condensation Particle Counter (CPC) - determination of PM mass concentration, particle size distribution and number density concentration.

In addition, we have made measurements of gross particle number density, without size discrimination, using a condensation particle counter.

We compare these various instruments and techniques, with particular emphasis on two applications: measuring emissions and monitoring HEPA filters for failure. Results include comparisons among different instruments while sampling the same aerosol source. Both operational considerations and performance of the instrumentation will be discussed.

INTRODUCTION

Public concern regarding possible emissions from facilities employing HEPA filtration systems and increasing regulatory scrutiny of such facilities has caused many to question the viability of monitoring particulate matter concentration downstream of such HEPA filters. The draft hazardous waste combustor (HWC) MACT issued on April 19, 1996 (61 FR 17358) indicated EPA's intent to require facilities to use

continuous emission monitoring systems for particulate matter as soon as technology became available. In conjunction with this intent, EPA initiated a study of the current technology in particulate matter continuous emission monitoring systems (PM CEMS). This consisted of a written report published as EPA Report 454/R-00-039 "Current Knowledge of Particulate Matter (PM) Continuous Emission Monitoring" in September, 2000 and at least seven demonstration projects involving head-to-head evaluation of current models at existing incinerators.[1] EPA head-to-head demonstrations were conducted at: (1) a mixed solid and liquid hazardous waste incinerator in Bridgeport, NJ; (2) a hazardous waste cement kiln in Fredonia, KS; (3) the DuPont Experimental Station's hazardous waste incinerator in Wilmington, DE; (4) a coal-fired boiler in Battleboro, NC; and (5) the TSCA incinerator at the DOE Oak Ridge facility. Two industry demonstrations were conducted: one at Georgia Power Company's Plant Yates coal-fired boiler in Newnan, GA by the Electric Power Research Institute (EPRI); the other at a liquid hazardous waste incinerator at the Eli Lilly Clinton Lab in Clinton, IN by Eli Lilly, the Chemical Manufacturer's Association and the Coalition for Responsible Waste Incineration.

Information gathered during EPA's evaluation of PM CEM technology indicates that it is feasible to accomplish the intended monitoring activities with today's technology. However, it should be noted that this is contingent upon two assumptions: (1) a regulatory threshold of 32 mg/m^3 and (2) a particle size distribution of the emitted particulate matter that would allow detection using optical methods. It should also be noted that mass emission rates of particulate matter downstream of HEPA filters will be orders of magnitude lower than the MACT and the count median diameter of these particles will be in the range of 140 nm. Such low emission rates and small particle sizes make monitoring by current technologies or measurement using EPA's Reference Method 5i (RM5i) virtually impossible.

In its "Current Knowledge of Particulate Matter Continuous Emission Monitoring", EPA summarizes the commercial PM CEMS available at that time. These instruments are based on several different principles: beta attenuation, probe electrification, light scattering, light extinction, and light scintillation. The report covers seventeen instruments in detail. It compares all the instruments by quoting the detection limits in terms of mass loading, mg/m^3. Table I lists those instruments covered with their minimum and maximum mass loading capabilities. The information derived from the study shows that while all of the instruments reviewed could possibly be used upstream of a HEPA filter only two are suitable for use downstream. Only one of these, the Sick, Inc. RM210, appears to be suitable if the upstream loading is less than the Nuclear Air Cleaning Handbook (NACH) HEPA maximum loading design limit of 23 mg/m^3.[2]

The emission rates downstream of a HEPA filter are much lower than the minimum detection limits listed in Table I. However, the uncertainty associated with their measurement remains a source of significant public concern. In order to provide scientific credibility to the process of issuing operating permits for facilities subject to the HWC MACT, DOE and EPA commissioned a study to evaluate and monitor particulate matter emission rates downstream of HEPA filters under varying conditions utilizing the most sensitive instrumentation available. This paper presents comparative data collected during this study. Instrumentation and measurement techniques employed include: (1) RM5i, (2) ELPI, (3) SMPS, (4) diffusion battery, and (5) CPC.

The Diagnostic Instrumentation and Analysis Laboratory (DIAL) at Mississippi State University has undertaken the task of evaluating the most commonly used standard method for particulate measurement, USEPA RM5i, and comparing this method with several, possibly more accurate, instrumental techniques. These include the particle counting and sizing techniques of the differential mobility analyzer/condensation particle counter (DMA/CPC), the electrical low pressure impactor (ELPI), and the diffusion battery (DB). Also included for comparative measurements is the total particle counting technique or stand-alone CPC.

REFERENCE METHODS

USEPA Reference Method 5

EPA has traditionally quantified the total particulate matter contained in gaseous emissions from stationary sources by the use of extractive sampling techniques. The standard method, Reference Method 5, consists of a sampling train composed of: (1) a right angle sample nozzle attached to a heated sample probe that is inserted into the emissions stack/flue perpendicular to the flow, (2) a filter substrate that is contained in an oven maintained above the dew point of the gas, 248 +/-25 ^{0}F, (3) a set of impingers that condense the water vapor from the gas stream prior to reaching the metering console and facilitate determination of the water content of the flue gas, (4) a metering console used to control and measure the sampling flow rate and volume ensuring that the sample is collected isokinetically and is sufficiently large, and (5) a rotary vacuum pump used to draw the gas through the apparatus.

Table I Minimum and Maximum PM loadings for PM CEMS

	Particulate Loading (mg/m^3)		**Particulate Loading (#/cm^3)**			
			Monodisperse Aerosol, d = 1.0µm		**Polydisperse Aerosol, μ_g = 1.0µm, σ_g = 2µm**	
PM CEM	**Min**	**Max**	**Min**	**Max**	**Min**	**Max**
Mechanical Systems, Inc. BetaGuard PM Beta Attenuation	1	500	9.65E+02	4.82E+05	1.11E+02	5.55E+04
Sigrist KTNR and CTNR Extractive Light Scatter	*	0.1		9.65E+01		1.11E+01
Durag DR-300-40 In-situ Light Scatter	*	1		9.65E+02		1.11E+02
Environmental Systems Corporation P5 In-situ Light Scatter	0.5	2000	4.82E+02	1.93E+06	5.55E+01	2.22E+05
Sick Inc. RM210 In-situ Light Scatter	0.0001	200	9.65E-02	1.93E+05	1.11E-02	2.22E+04
Sick Inc. FW100 and FWE200 Light Scatter	0.1	5	9.65E+01	4.82E+03	1.11E+01	5.55E+02
Grimm Technologies Inc. Model 6300 In-situ Light Scatter	*	1	*	9.65E+02		1.11E+02
Monitor Labs Model 300L In-situ Light Scatter	*	20	*	1.93E+04		2.22E+03
BHA Group CPM 5000 Scintillation	2.5		2.41E+03		2.78E+02	
PCME Scintilla SC600 Scintillation	2.5		2.41E+03		2.78E+02	
Insitec TESS In-situ or Extractive Laser Light Extinction-Scatter	1.3		1.25E+03		1.44E+02	
PCME DustAlert-90 Electrostatic Induction	0.02		1.93E+01		2.22E+00	
Auburn International Triboguard III or II In-situ Triboelectric	0.005		4.82E+00		5.55E-01	
Codel StakGuard Triboelectric Dust Monitor	0.1		9.65E+01		1.11E+01	
Opacity monitors	1.2		1.16E+03		1.33E+02	

* Note that for several monitors only the nominal loading range is given with a zero lower limit, but no indication of the resolution is given (nor can be determined from readily available literature).

The gaseous components pass through the filter membrane and the particulate is retained on the filter substrate. In addition to the PM deposited on the filter, some PM impacts walls of the sampling apparatus and is retained in the probe nozzle, probe liner, sampling line, and the upstream half of the filter housing. PM retained in the probe nozzle, probe liner, and front half of the filter housing is collected by washing these parts with a solvent. This sample is reduced to dryness and desiccated to constant mass along with the sample filter. These two fractions are analyzed gravimetrically and the total particulate mass per volume of dry gas is reported. The gaseous sample is extracted from the stack isokinetically to minimize the inertial separation of the particles based on particle size (momentum) from the flow and prevent introduction of systematic bias to the extracted sample.

The methodology developed by USEPA to ensure adequate characterization of flue gas flow patterns for collection of a representative sample has been canonized in Reference Methods 1 through 5. Procedures outlined in this suite of methods take into account duct size and gas velocities and establish minimum requirements for collection of valid samples. Nearly all of the other EPA extractive, stack sampling methods are derivatives of RM5; RM23 and 23A for Dioxins and Furans, RM29 for multi-metals, VOST for volatile organics and the organic methods 0010, 0020, 0030, etc. found in SW 846. It is important to note that one of the central considerations for validating the sample is whether or not the sample was collected isokinetically. If $90\% \leq I \leq 110\%$, the results are acceptable.

USEPA Reference Method 5i

RM5 does not provide reliable results in systems with low PM concentrations because of problems associated with gravimetric analysis of the solvent and filter substrate and/or the time required to collect a sample. Method 5i was developed to address these low PM applications. This method is performed using a paired train configuration, and the RSD of paired data is used to quantify data precision. The enhanced sensitivity of RM5i is achieved by (1) improved sample handling procedures, (2) the use of a light-weight sample filter assembly, and (3) the use of low residue grade acetone.

The Environmental Protection Agency has determined that Method 5i has a calculated practical quantitation limit (PQL) of 3 mg of PM and a minimum detection limit (MDL) of 1 mg. Therefore, EPA has concluded that for PM testing the target catch must be no less than 3 mg. In certain cases for determining source compliance, a sample catch of 1mg to 3 mg may be used, but results will have a high degree of uncertainty. For the purposes of this HEPA filter performance project, a minimum target catch of 3 mg will be used. The sample rate for an EPA type stack sampler is 0.75 cfm and a typical sample extraction time is 30 – 60 minutes. This results in a PQL limit of 2.4 mg/m^3 for a sampling time of one hour. The NACH lists the maximum PM concentration that should be delivered to a HEPA filter as 23 mg/m^3 and the HWC MACT sets 34 mg/m^3 as an emission limit.[3] From this it can be seen that RM5i PQL compares favorably with expected upstream concentrations, but is orders of magnitude greater than the corresponding downstream filter concentration of 0.0069 to 0.0102 mg/m^3 (0.03% of the NACH recommended maximum and the HWC MACT, respectively). Table II illustrates the sampling times required to meet the above conditions.

RM5 is most effective for total PM catches of 50 mg or less. This procedure incorporates all components of Reference Methods 1 through 4 and the majority of the elements of Reference Method 5. The filtering system for Method 5i differs from the standard Method 5 holder in both size and assembly. The Method 5i filter holder is fitted for a 47 mm filter compared to 110 mm for the RM5 unit. The entire filter holder assembly is weighed for RM5i versus only the filter for RM5. The uniquely numbered 5i filter holder assembly is oven-dried and desiccated before and after sample collection. An impinger system follows the filter to collect moisture in the sample gas stream. The moisture content is determined by measuring the amount of water collected in the impinger train using volumetric or gravimetric procedures.

For conditions in which the aerosol concentration is above the HWC MACT, RM5i may not be applicable. In this case it may be necessary to employ RM5.

Table II Sampling Times Required for USEPA RM5i to Achieve PQL of 3 mg and MDL of 1 mg/m^3 at a Sampling Rate of 0.75 cfm

Stack concentration (mg/m^3)	Sampling time		
	(minutes)	(hours)	(days)
100	1.412		
50	2.824		
10	14.12		
1	141.2	2.353	
0.1	1412	23.53	
0.01 [HWC MACT]	14120	235.3	9.81
0.0069 [NACH]	20464	341	14.2
0.001	141200	2353	98.1

INSTRUMENTAL TECHNIQUES

Differential Mobility Analyzer/Condensation Particle Counter (DMA/CPC)

Principle of Operation

Within a DMA/CPC system particles are classified, or sized, using an electrical mobility technique. A bipolar charger within the electrostatic classifier (EC) charges particles to a known charge distribution. The particles are the sized within the DMA according to their ability to traverse an electrical field and the concentration, or number density, is measured with a CPC. The system is automated using a personal computer and software to control the instrumentation, collect and store data, and perform data reduction. Further data reduction is most easily performed by exporting data to another application such as Microsoft Excel.

The DMA/CPC system is commercially available from TSI, Inc. of Minneapolis, MN as a Scanning Mobility Particle Sizer or SMPS. Table III lists the specifications of the three commercially available SMPS systems from TSI. These systems offer a particle sizing range from 0.003 to 1.0 μm within a concentration range of one particle/cm^3 to 10^8 particles/cm^3 depending on the combination of DMA/CPC. The measurement cycle time for the systems are selectable from 60 to 600 sec with a resolution of up to 162 channels and scan times from 10 to 300 sec.

Applications

The DMA/CPC particle sizing and measuring system predominantly finds application as a laboratory instrument with limited field applications. Most applications of DMA/CPC particle sizing and counting are in the areas of combustion studies, atmospheric aerosol measurements, indoor-air research, filter testing, and inhalation toxicology.

Advantages and Disadvantages

As compared to the commercially available PM CEMS discussed earlier, the DMA/CPC system offers the ability to far more accurately size and count particles orders of magnitude smaller and at concentrations far below most PM CEMS. The applicable size counting range (see Table III) is, however, much narrower. Particle size distribution, number density, and mass loading rates of particles with an aerodynamic diameter larger than one μm can only be projected from measured DMA/CPC data .

The following are possible sources of error encountered in the use of a DMA/CPC system: multiple charges, coincidence in CPC, efficiency of CPC, voltage in DMA, timing, flows, and air viscosity/density values. Those related specifically to the CPC are discussed in a later section on the CPC as a stand-alone instrument.

As noted earlier, the DMA sizes particles according to their ability to traverse an electrical field. Before particles enter the DMA, a bipolar equilibrium charge level on the particles must be produced. This is accomplished by passing the aerosol through a radioactive (Kr-85) bipolar charger or neutralizer. Nearly all particles from 2 to 300 nanometers in diameter receive either a single positive, single negative, or zero charge. These particles then enter the DMA and are separated according to their electrical mobility. The electrical mobility of particles in a constant electric field depends on both the size and charge, causing larger multiply-charged aerosols to be output with smaller particles holding single charges. Correction can be applied to account for this effect assuming the particles have neutralized to a Fuchs charge distribution.

With the DMA/CPC system, particles between 15 and 700 nm are detected and classified into up to 64 size bins per decade. The discrimination between particles is based on the electrical mobility diameter. Over a period of two to five minutes, the DMA voltage is ramped exponentially to produce a scan of the particle size distribution. The DMA/CPC system, consequently, cannot provide accurate data on transient aerosols in this scanning configuration.

Unlike PM-CEMS, the portability of the system is limited. In addition, factors such as operating temperature and humidity must be closely controlled to insure accuracy of measurements. The DMA/CPC system is designed for indoor use. The required temperatures and humidity for operation are 10^0C to 37^0C and 0 – 90%, noncondensing.

Table III TSI Model 3936 SMPS Specifications [4]

Concentration range	Model 3010 1 particle/cm^3 to 10^7 particles/cm^3 Model 3022A 2 particles/cm^3 to 10^8 particles/cm^3 Model 3025A 20 particles/cm^3 to 10^7 particles/cm^3
Particle diameter range	Model 3010 10 nm to 1000 nm Model 3022A 7 nm to 1000 nm Model 3025A 3 nm to 1000 nm
Displayed resolution	4, 8, 16, 32, or 64 geometrically equal channels per decade
Inlet flowrate of the CPC	Model 3010 1.0 lpm Model 3022A 0.3 lpm or 1.5 lpm Model 3025A 0.3 lpm or 1.5 lpm
Flowrate of the Electrostatic Classifier Aerosol Sheath air	Model 3010 0.20 to 2.0 lpm Model 3022A 0.2 to 2.0 lpm Model 3025A 0.2 to 2.0 lpm 10 times aerosol flow (nominal) 2–15 lpm
Measurement cycle time	Total: 60 to 600 seconds, user selectable. Up scan: 10 to 300 seconds
Sample averaging	One sample can average 1 to 999 scans

Calibration and Maintenance

Calibration of the DMA/CPC system requires calibration of each of its three major components; classifier, DMA, and CPC. The sheath, bypass, and impactor flows of the electrostatic classifier are

calibrated using a bubble flowmeter such as a Gilman Gilibrator Electronic Bubble Flowmeter. Calibration of the DMA voltage requires a high-voltage multimeter and specialized wire lead. Calibration of the CPC will be discussed in later section of this paper.

Periodic maintenance of the DMA/CPC is necessary to insure proper performance of the system. The frequency of service depends on the concentration of aerosol entering the classifier and length of sampling time. Frequent cleaning of the impactor and nozzle is recommended and is essential under high aerosol concentration conditions. High aerosol concentrations and/or long sampling times may also result in a build up of particles in the DMA requiring its disassembly and cleaning. The HEPA filters used to filter the sheath air and bypass air should be regularly inspected and/or replaced. TSI, for example, recommends changing the filters at least after every 8000 hours of operation or earlier depending on conditions of use.

Electrical Low Pressure Impactor

Principle of Operation

The electrical low pressure impactor is based on charging particles within an aerosol stream before passing the particles through a cascade impactor where collection of particles on the stages of the impactor is detected by a series of electrometers. The incoming aerosol stream initially passes through a corona charger wherein the particles are electrically charged by a Wolfram needle. Passing from the charger to the 13-stage cascade impactor, the particles are classified or sized according to their aerodynamic diameters. The stages of the impactor are electrically insolated allowing electrometers to measure the accumulated current on each stage of the impactor. This current value (fA) for each stage corresponds to the number of particles collected for each stage. These values allow for computation of a size distribution for the particles in the aerosol stream. Table IV lists the cutpoint diameters of the impactor for a 30 lpm flowrate.

Table IV ELPI 30 lpm Impactor Cutpoint Diameters

Stage	d_{50}	Stage	d_{50}
13	10.04	6	0.386
12	6.65	5	0.265
11	4.03	4	0.158
10	2.41	3	0.0939
9	1.61	2	0.0556
8	0.957	1	0.0285
7	0.619		

The electrical low pressure impactor is commercially available from Dekati Ltd of Finland as the ELPI Model 3935. This instrument offers a broad measurement range of particle sizes, from 0.03 to 10 µm with 12 channels corresponding to the 12 stages of its inertial impactor. This size range can be extended down to 0.008 µm with the use of an electrical filter stage available from Dekati. The ELPI operates in a concentration range of 80 to $1x10^7$ particles/cm^3. The instrument is made up of essentially three components; a corona charger, low pressure cascade impactor, and a multi-channel electrometer. Table V lists the general specifications of the Dekati ELPI.

Applications

The most common application of the ELPI has been in the area of particle size distribution measurements of combustion and automotive exhaust. It has also found application in the areas of particle size distribution measurements in pharmaceutical inhaler studies, filter grade efficiency, particle charge measurements, and indoor/outdoor air quality measurements.

Advantages and Disadvantages

The ELPI is capable of measuring particle size distribution and concentration in real time from 70 nm up to 10 µm. It is much more compact and portable than the TSI SMPS. Table VI gives a comparison summary of the two TSI SMPS systems and the Dekati ELPI.

The ELPI sizes particles based on aerodynamic diameter but measures particle concentration using Stokes diameter. This requires a known density for the particle being sampled. If the density varies with diameter additional uncertainty is introduced into the measurement.

For the ELPI, diffusional losses in the impactor due to the charge of the aerosol cause small particles to collect in the upper stages. The ELPI corrects the measurement for these losses. Particle bounce, a phenomenon where particles fail to adhere to the stage collection substrate and fall to a lower stage, causes higher readings in the lower stages. Aluminum foil substrates greased with a toluene-Apiezon grease mixture are used to minimize the effect of bounce.

Table V Dekati ELPI General Specifications [5]

Nominal air flow	10 or 30 lpm
Particle size range	0.03-10 µm with filter stage 0.008-10 µm
Number of channels Time resolution	12 < 5 s
Operation conditions, instrument: Ambient temperature Ambient humidity	 0-40 °C 0-60%, non condensing
Aerosol conditions: Gas temperature	 < 60 °C with heated impactor < 200 °C

Table VI Comparison Summary of SMPS and ELPI Specifications

	SMPS		
Model	**3936L22**	**3936L10**	**ELPI 3935**
CPC included	3022A-S	3010-S	n/a
DMA included	3081	3081	n/a
Particle size range (µm)	0.007 – 1.0	0.01 – 1.0	0.03 – 10
Particle concentration (#/cm^3)	2 to 10^8	1 to 10^7	80 - 10^7
Measurement cycle time	120 – 600 sec	60 – 600 sec	>5 sec (programmable)
Resolution (total channels measured)	up to 162		12
Channels displayed per decade	4, 8, 16, 32, or 64 (selectable)		n/a

Calibration and Maintenance

Aside from routine maintenance, the Dekati ELPI requires no calibration. The manufacturer recommends the instrument be serviced by either Dekati or an authorized distributor annually, biennially, or every three years depending on frequency of use.

The manufacturer provides detailed instructions on cleaning the impactor and corona charger. For routine cleaning, the charger should be rinsed with isopropanol and allowed to dry. Water must never be used to clean the charger. If needed, the charger may be dismantled and cleaned more thoroughly. Again this should be done with isopropanol and detailed instructions on dismantling are found in the ELPI instruction manual.

In order to insure proper function and accuracy of results, the ELPI impactor must be properly cleaned and assembled. The stainless steel parts of the impactor may be cleaned with water but it is recommended that these parts be rinsed with isopropanol and allowed to dry. Under some sampling conditions, it may become necessary to clean the impactor stages with a more aggressive solvent and/or in an ultrasonic cleaner. Compressed air may be used to remove particles from the holes of the impactor stages. Usually a combination of these techniques works best. The Teflon insulators between the impactor stages must never be cleaned with water. If cleaning of the insulators is necessary, they should be rinsed with isopropanol and allowed to dry.

The impactor should be checked before use to insure that it is properly assembled and does not leak. Also before each measurement, the ELPI electrometer must be zeroed. There are three methods for zeroing the electrometer: (1) Main Reset, (2) Zero, and (3) All Zero. Main Reset zeroes all four measuring ranges and is accessed from the front panel display of the ELPI. The results of this zeroing method are saved to an ELPI onboard computer and the values are used if no other zeroing method is employed. The manufacturer recommends using either the Zero or All Zero method which is accessed from the software's (ELPI VI) control panel. All zero conducts zeroing to all four measuring ranges while Zero zeroes only the measuring range in use. Procedures for each of these tasks are described in the Dekati ELPI instruction manual.

Condensation Particle Counter (CPC)

Principle of Operation

The condensation particle counter (CPC), also known as a condensation nucleus counter (CNC), is an instrument that saturates an aerosol stream with alcohol or water vapor, causing particles in the sample flow to serve as condensation nuclei. The enlarged particles (~ 10 – 20 μm in aerodynamic diameter) are then counted optically.

The largest manufacturer of CPCs, TSI, offers three models (3010, 3022A, and 3025A) for standard particle counting that can be used with their SMPS systems and two models (3760A and 3762) intended for clean room monitoring and extremely low level applications. The parameters for these instruments are summarized in Table VII.

Table VII Summary of operating parameters of TSI CPC Models

TSI Model	**3010**	**3022A**	**3025A**	**3760A**	**3762**
Minimum Particle size, nm (50% efficiency)	10	7	3	11	11
Aerosol Flow Rate, cm^3/min	1000	300	30	1500	3000
Upper Concentration Limit, #/cm^3	10^4	10^7	10^5	10^4	$5x10^3$
False Background Count Level, #/cm^3	<0.00001	<0.01	<0.01	<0.00005	<0.00005
Response time (95% response), s	< 5	< 1.3	1	< 3	< 1.5

Applications

CPCs are capable of measuring across a broad range of concentrations (see Table VIII), consequently, finding applications in clean-room monitoring, atmospheric aerosol measurements, and filter efficiency testing. Real-time response allows for continuous monitoring of aerosol in systems.

Table VIII Concentration Range of CPC

Mass, mg/m^3	Number, #/cm^3
0.1	1390
0.5	6940
1.0	13900
2.0	27800

Advantages and Disadvantages

Error in CPC measurements derives from statistical and calibration considerations. For low particle concentrations, Poisson statistics dictates the error in the measurement:

sigma = sqrt(N) (Eq. 1)

where sigma is the standard error of the measurement and N is the true value. The effect of coincidence—two particles entering the control volume of the optical sensor close enough in time to be read as a single particle—need also be corrected. Calibration is applied to two elements of a CPC: the aerosol flow through the instrument and the measurement of the optical sensor. In the TSI 3022A CPC, for example, calibration of concentration is only necessary above 10000 #/cm^3, where a photodetector compares the light scattered from the aerosol to a calibration value. Below 10000 #/cm^3, the CPC eliminates the need for calibration by counting each particle.

Calibration and Maintenance

Aside from factory-performed optics alignment, the TSI 3010 CPC requires no calibration other than periodic flow verification. This is accomplished by connecting a low pressure-drop flowmeter to the CPC inlet. A bubble or thermal flowmeter works best. The flowrate should be 1.0 lpm +/- 10%.

In applications of prolonged use and/or very high aerosol concentrations, it may become necessary to clean the optics of the CPC. To avoid delays and cost of returning to manufacturer, cleaning may be performed by the user. The user's first attempt at performing this task should only be done under the direct guidance of a TSI technical representative.

Unlike the TSI 3010 CPC, the TSI 3022A CPC is equipped with a configuration mode which allows the user to select various microprocessor outputs and controls. It also allows the user to calibrate the D/A and A/D converters and the flow meter. These configuration changes and calibrations are explained in detail in Chapter 4 of the TSI 3022A CPC instruction manual.[6] Chapter 7 of this manual provides a short description of the procedures used by TSI to calibrate the sensors of the 3022A CPC. For the typical user, it is recommended that these calibrations be performed only by the manufacturer.

As with the 3010 CPA, in applications of prolonged use and/or use in very high aerosol concentrations, it may become necessary to clean the optics of the CPC. This may be accomplished by the user. Consultation with TSI is recommended before proceeding.

Diffusion Battery

Principle of Operation

Diffusion batteries rely on diffusion to remove small particles from a laminar-flow aerosol stream. They were designed to measure the diffusion coefficient of particles less than 0.1 μm and can be used to determine particle size distribution by converting the diffusion coefficient to particle size.

Applications
Diffusion batteries are applicable to particle sizes from 0.002 to 0.2 μm. There are several types of diffusion batteries including rectangular channel, parallel disks, cylindrical tubes, and screen type. In typical applications a CPC is used to measure the number concentration from each stage of a diffusion battery. This is accomplished by employing a system of diffusion battery, automatic switching valve, and CPC.

Advantages and Disadvantages
As noted earlier, diffusion batteries rely on diffusion to remove small particles from a laminar-flow aerosol stream. Diffusion is the dominant mechanism controlling the motion of particles less than 0.1 μm making the diffusion battery a very effective measurement technique. The disadvantages to diffusion batteries include lengthy sampling times and other instrumentation required to automate diffusion battery measurements. Under conditions of very low aerosol concentration, sampling times for measuring particle concentration behind each stage of a diffusion battery can range from 30 minutes to several hours. For this reason, automation of the diffusion battery measurements is necessary.

Up to the mid-1990s, TSI manufactured a Diffusional Particle Sizer (DPS) system which included a TSI Model 3040 or 3041 screen-type diffusion battery, a TSI Model 3042A switching valve, and either a TSI Model 3022A or 3025 CPC. Currently, no one manufactures such a system. Attempts to locate any company that manufactures a switching valve to be used in combination with a CPC and diffusion battery have been unsuccessful.

InTox Products in New Mexico is the only company known to still manufacturing screen-type diffusion batteries. The Model 02-190C InTox Products diffusion battery is a 10-stage diffusion battery in which stainless steel wire mesh screens are used as collecting surfaces. The four available screen sizes are 145, 200, 325, and 400 mesh. The battery has an aerosol flow diameter of 2.5 inches and is designed for an aerosol flow rate of 11 lpm.

EXPERIMENTAL DATA

Each of the measurement techniques discussed above are applicable to measurement of particulate matter downstream of HEPA filters. In an effort to demonstrate the capabilities and limitations of these techniques, a series of experiments have been conducted, each of which are discussed below.

ELPI, SMPS, and EPA RM5i

A small-scale test stand was designed to allow for simultaneous measurements with each of these instruments. A potassium chloride aerosol stream was generated using a TSI 9306A six-jet atomizer then passed through a diffusion dryer and a TSI 3054 neutralizer. A series of KCL solutions ranging in concentration from 3% to 0.073% were used to generate aerosol streams of varying number density. For all measurements, one jet of the atomizer was used along with makeup air to produce a constant output of 80 lpm. The aerosol was introduced to each of the sampling systems by use of a four-way flow splitter.

A series of measurements was first made comparing EPA RM5i with a DMA/CPC. The DMA/CPC used was a TSI 3936L22 Scanning Mobility Particle Sizer (SMPS) equipped with a TSI 3022A CPC and 0.071 cm impactor inlet. The sheath and sample flow rates of the SMPS were maintained at 3.0 and 0.3 lpm respectively. For each measurement, a dual train EPA RM5i setup was used. Sampling times for the RM5i ranged from 18 minutes with the 3.0% solution to 135 minutes with the 0.073% solution. Three tests runs were made for each solution concentration. These measurements were repeated comparing the ELPI and RM5i. The ELPI used was a Dekati 3935 with a 30 lpm sample flow rate.

Table IX gives, in tabular format, the mass loading rates as measured by each of the three techniques.

Table IX Results from ELPI, SMPS, RM5i Comparison

		RM5i		SMPS			ELPI (First two minutes)			ELPI (Entire range)		
Run ID	Solution (% KCl)	Loading rate (mg/m^3)	Sampling time	Loading rate (mg/m^3)	CMD (nm)	GSD	Loading rate (mg/m^3)	CMD (nm)	GSD	Loading rate (mg/m^3)	CMD (nm)	GSD
126(1)	3	51.48	12	36.12	85	2.05	174	45.58	1.83	183.33	54.55	1.97
126(2)	3	55.94	12	41.97	85	2.09	142	44.67	1.82	169.63	50.66	1.99
126(3)	3	53.33	12	35.54	82	2.06	131	42.15	1.79	160.29	49.00	1.96
127(1)	1.16	22.64	18	22.90	69	2.08	54.31	39.66	1.71	77.46	49.43	1.97
127(2)	1.16	23.31	18	22.76	70	2.07	51.09	39.51	1.71	69.33	47.51	1.94
127(3)	1.16	22.52	18	22.52	68	2.13	44.96	40.15	1.71	67.08	48.28	1.93
129(1)	0.27	6.03	60	7.41	54	1.99	7.62	38.68	1.54	21.69	52.13	1.97
129(2)	0.27	5.67	60	7.51	54	2.00	8.13	38.66	1.58	20.97	52.27	2.00
129(3)	0.27	6.30	60	8.46	55	2.01	8.78	37.92	1.56	21.47	51.01	1.99
128(1)	0.07	1.93	135	2.39	45	1.89	2.05	36.15	1.43	5.82	48.94	1.79
128(2)	0.07	2.22	135	2.74	45	1.90	2.85	35.88	1.45	8.47	48.56	1.81
128(3)	0.07	2.03	135	3.01	46	1.91	2.32	35.98	1.44	8.69	50.71	1.80

Figure 1A gives a log plot of the results plotting each data point as well as a line through the average. As can be seen from the data in Table IX, agreement between mass concentrations measured by the SMPS (using TSI Model 3022A CPC) and RM5i becomes much better as solution concentration, thus number density, decreases. As noted earlier in this paper, the accuracy of particle counting for the CPC (TSI Model 3022A) increases as number density decreases. It should be noted that sampling times for the RM5i measurements greatly increased as number density decreased. For example, the sampling time for RM5i with a 3% KCl solution was only 12 minutes while with the 0.073% solution, the RM5i sampling time required was 2 hours and 15 minutes.

As illustrated by Figure 1B, the number densities measured by the ELPI were either near or above the upper detection for the ELPI particularly in the lower stages; i.e., smallest particles size bins. In addition, the loading rate of the ELPI impactor was extremely high for the higher number densities. The ELPI data given in Table IX are those measured during the first two minutes of sampling as well as during the entire run time of the RM5i. For the lower number densities, if one looks at just the first two minutes of ELPI loading, a much closer agreement between ELPI and RM5i mass loading rates is observed for the smaller number density. For example, with the 0.073% KCl solution, the first two minutes of sampling with the ELPI yielded an average mass loading rate of 2.41 mg/m^3 as compared to an average of 2.71 mg/m^3 for the SMPS and 2.06 mg/m^3 for RM5i. Figure 1C illustrates the comparison of the SMPS and ELPI data to that of the RM5i. As can also be seen from the data in Table IX, the particle size distribution (PSD) shifts downward as the number density decreases resulting in a decrease in count median diameter (CMD) and geometric standard deviation (GSD). This occurs for the SMPS measurements as well as the ELPI during the first two minutes of sampling.

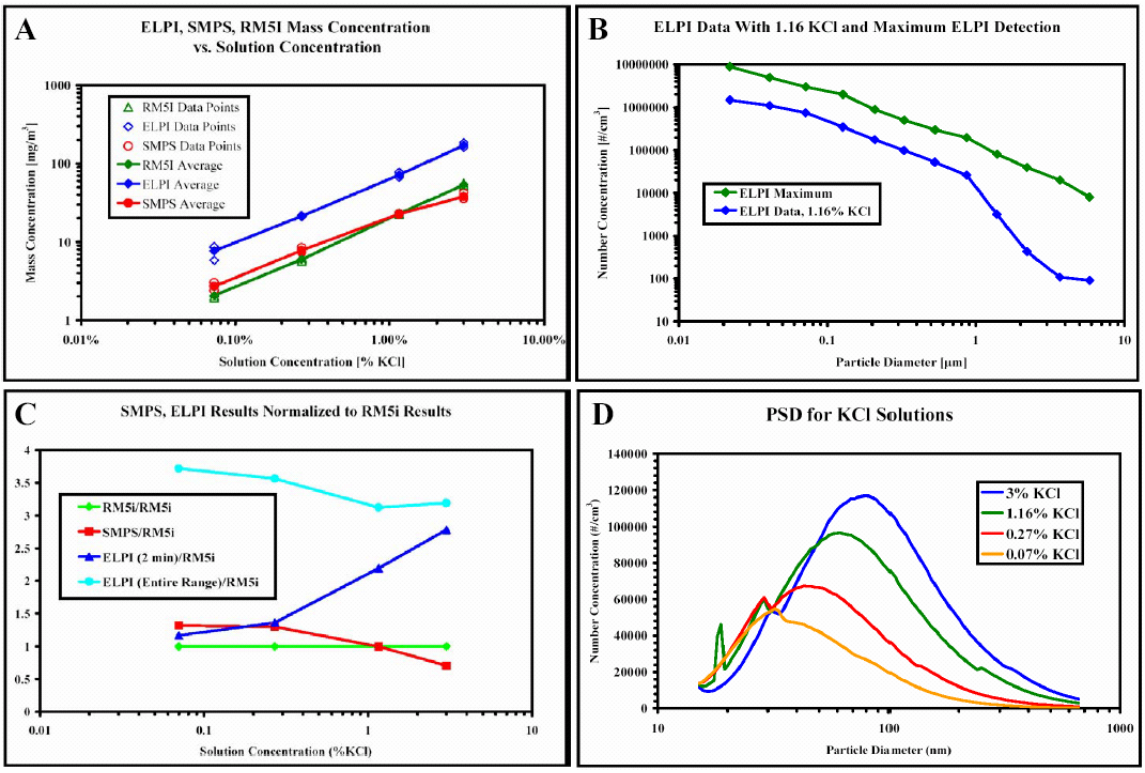
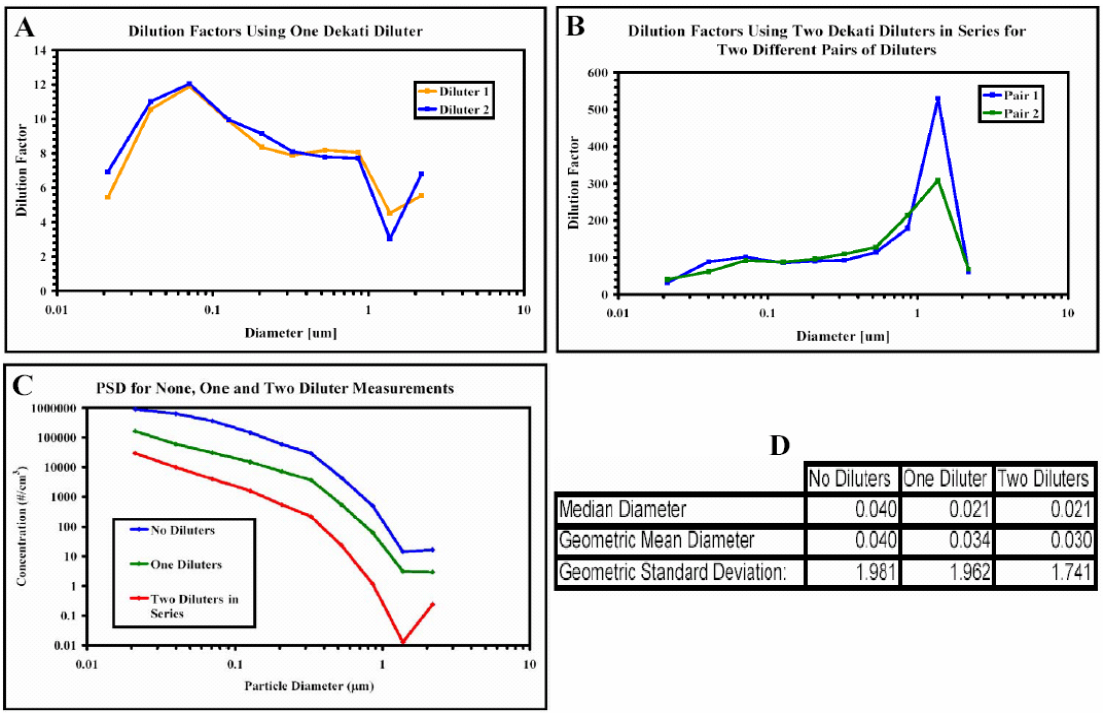


Fig. 1A Log-log plot of ELPI, SMPS, RM5i mass concentration vs. solution concentration. B: ELPI data from 1.16% KCl solution compared to maximum ELPI detection limit. C: Comparison of ELPI, SMPS, and RM5i results. D: Particle size distributions comparison of SMPS results for each KCl solution.

The decay in quality of the ELPI measurements with time under high particle loadings can be partially corrected using diluters on the ELPI. Dekati manufactures cylindrical, ejector-type aerosol diluters intended for use with the ELPI that have a nominal dilution factor of approximately 8.5. Substituting a TSI 3076 atomizer for the TSI 9306A, the same generation setup as before was used to take ELPI measurements without diluters, with one diluter, and with two diluters in series. Figure 2A shows the dilution factor vs. diameter for two single diluters while Figure 2B shows the dilution factor vs diameter for two sets of diluters used in series; i.e., paired. These figures illustrate a problem encountered when using diluters. The diluters do not have constant influence on all impactor stages. Additionally, the upper stages (i.e., larger particles) have higher uncertainty associated with the dilution factor due to the low concentrations measured with diluters. Figure 2C illustrates the downward shift (decrease in CMD) in PSD as diluters are used. Figure 2D gives the CMD and GSD for the ELPI distributions shown in 2C.



D

	No Diluters	One Diluter	Two Diluters
Median Diameter	0.040	0.021	0.021
Geometric Mean Diameter	0.040	0.034	0.030
Geometric Standard Deviation:	1.981	1.962	1.741

Fig. 2A Plot comparing dilution factors measured for two different Dekati DI-1000 diluters. B: Plot comparing dilution factors for two Dekati DI-1000 diluters in series for two different pairs. C: Comparison of particle size distributions measured by ELPI with no, one and two diluters. D: Statistical comparison of ELPI particle size distributions in 2C.

Measurement Of Low Number Density Particulate Downstream Of A HEPA Filter Using a Diffusion Battery As Compared To Measurement With A DMA/CPC

Each of these measurements was conducted on the DIAL-MSU HEPA test facility. The main parameters that went into the design of the test stand include:

1) Flow rate range = 50-375 cfm (250 cfm nominal)
2) Inlet temperature = ambient to 300 F
3) Relative humidity = 15%-90%
4) Filter size = 12"x12"x11 ½ " thick
5) Port availability for making multiple, simultaneous, measurements upstream and downstream of the filter.
6) Particle injection.

The test stand uses a Flanders AG1 series filter housing to accommodate standard 12"x12"x11.5" nuclear grade HEPA filters for testing. Pressure across the test HEPA filter is measured with a dual set of differential pressure transducers along with a magnehelic pressure transmitter.

Inlet air to the test stand is entrained from room air and conditioned by passage through an 85% ASHRAE filter, a nuclear grade HEPA filter and finally an ULPA filter. The air is drawn into the test stand by a 10Hp, vortex blower located on the outlet of the test stand. Flow through the test stand is controlled via a by-pass valve just upstream of the blower and monitored by two venturi flow meters, one upstream and one downstream of the test HEPA filter.

The upstream and downstream sections of the test stand are equipped with ports fitted to allow sampling by a variety of instrumentation. These sections are also equipped with sensors for monitoring temperature, humidity and, as previously mentioned, volumetric flow. Measurement, control and data logging of these parameters are performed on a Lonworks, network based computer system.

For this series of tests a KCl aerosol stream of approximately 30 mg/m^3 was generated by the DIAL particle generator. This generator was customized in-house from a 30"x38" stainless steel tank. The aerosol is produced from a 30% KCl solution fed to an atomizing nozzle at a rate of 10 ml/min with 30 lpm of dry air. Along with heating the walls of the tank, hot dry air is introduced into the tank to facilitate drying of aerosol particles and reduce agglomeration of particles on the tank wall. Aerosol exits the generator via a 1" stainless steel tube. Before entering the test stand, the aerosol passes through a cyclone, also designed in-house, which removes particles from the aerosol larger than 3 to 5 micrometers.

All measurements were made at ambient temperature, a relative humidity of approximately 15%, and a flow rate of 250 cfm.

Instrumentation used included a TSI 3936L22 SMPS and Model 02-190C Intox Products diffusion battery. Each of these instruments is described in a previous section of this paper. The sheath and sample flows of the SMPS were 15 and 1.5 lpm, respectively. A 0.071 cm impactor inlet was used. Measurement of particulate concentration from each stage of the diffusion battery was made using a TSI 3022A CPC. The total flow rate through the diffusion battery was maintained at 11 lpm using a Welch Model 1399 vacuum pump and an Aalborg Model GFC47 mass flow controller. Stainless steel screens of 325 mesh were used in the diffusion battery. The arrangement of screens employed is noted in Tables X and XI.

Table X lists number of particles measured during a 70 sec sampling interval from each stage of the diffusion battery during loading of a new HEPA filter with KCl. A total of five different measurements were made. As can be seen from these measurements, the downstream number density was decreasing as the measurements were being made; i.e., the efficiency of the filter was increasing. Table XI lists the number of particles measured during a 90 sec sampling interval from each stage during the second day of loading of the same filter. As can be seen from the results, the efficiency of filter had increased to the point that even with an increased sampling time, few particles were detected. As discussed earlier, at such a low number density, sampling time for each stage of the diffusion battery would have to be increased to several minutes, possibly hours, necessitating automation of the diffusion battery.

Table X Downstream Diffusion Battery data taken during Initial Loading (Day 1) of a HEPA Filter

Stage	Screens Before Stage	Number of Particles Detected in 70 sec Sample				
		Run 1	Run 2	Run 3	Run 4	Run 5
1	0	386	177	80	46	17
2	1	294	256	102	72	21
3	3	330	150	79	56	20
4	6	316	196	121	32	30
5	10	280	122	92	58	34
6	15	444	120	60	44	25
7	21	227	172	72	26	17
8	28	392	197	84	49	18
9	36	395	109	58	34	11
10	45	333	92	49	32	20

Table XI Downstream Diffusion Battery data taken during second day of Particulate Loading

Stage	Screens Before Stage	Number of Particles Detected in 90 sec Sample		
		Run 1	Run 2	Run 3
1	0	0	0	1
2	1	1	0	3
3	3	0	0	0
4	6	2	3	1
5	10	3	0	1
6	15	3	3	0
7	21	2	0	0
8	28	3	2	0
9	36	2	1	0
10	45	1	0	0

Lower Detection Limits for ELPI, DMA/CPC and RM5i

Each of these measurements was made using the same setup and conditions as those for the diffusion battery and DMA/CPC. A series of four metal shims measuring 0.03" x 0.75" x 12.125" each were placed between the left front seal of the HEPA filter and the filter housing to simulate small seal leaks. Data was obtained for 'leaks' produced first by one shim, then two, three and four respectively. The top of Figure 3 illustrates the comparison of number densities measured downstream of the HEPA filter by ELPI and TSI 3010 CPC for each series of measurements. As shown, the ELPI and CPC measured number densities of <100 particles/cm^3 nearing the minimum detection limit of the ELPI but still well within the range of the CPC. The bottom portion of Figure 3 illustrates a comparison of subsequent ELPI and TSI 3936L22 SMPS measurements for each series of 'leaks'. Here we see less agreement between measurements as we are well below the minimum detection limit of the ELPI and near that of the SMPS.

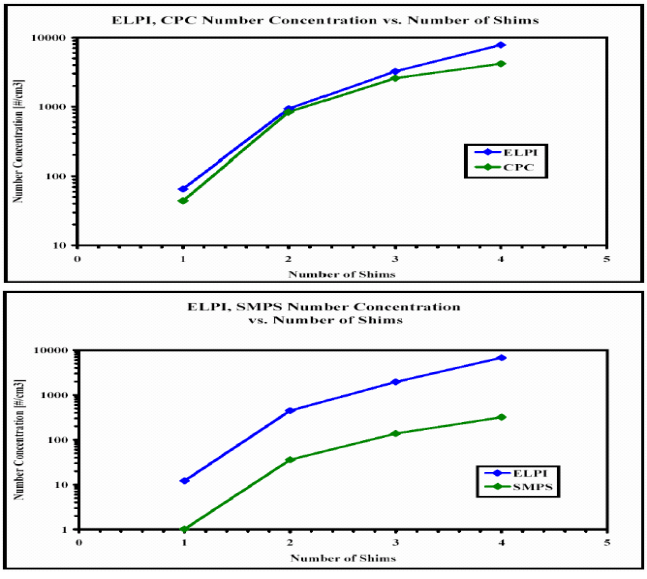


Fig. 3 Top: ELPI and CPC number concentration vs. number of shims Bottom: ELPI and SMPS number concentration vs. number of shims

HEPA Filter Particulate Loading

Each of these measurements was made using the same setup and conditions as those for the diffusion battery and DMA/CPC. For these series of tests, measurements were made for number density and particle size distributions both upstream and downstream of the HEPA filter at a sequence of particulate loading stages measured as pressure drop (ΔP) across the filter. Measurements were made at ΔPs of 1", 2", 3", 4", 5", and 6".

As illustrated by the top portion of Figure 4, as a HEPA filter loads, downstream number density decreases with time. Consequently, as illustrated in the bottom portion of Figure 3, the filtering efficiency increases with time. This was observed during measurements made at ΔPs of 1", 2",…, 6" respectively. At each increasing ΔP, the initial number density measured downstream was less than the initial number density measured at the previous ΔP. This indicates that the HEPA filter actually becomes more efficient as it loads. These measurements were repeated for four different new HEPA filters with similar results for each. As can also be seen from the top portion of Figure 4, the accuracy of the CPC measurement decreases as the HEPA filter loads and its efficient increases; i.e., number density approaches zero. A longer averaging time is needed to counter the statistical inaccuracy of measuring lower concentrations; however, this would decrease the time-sensitivity of the measurement technique.

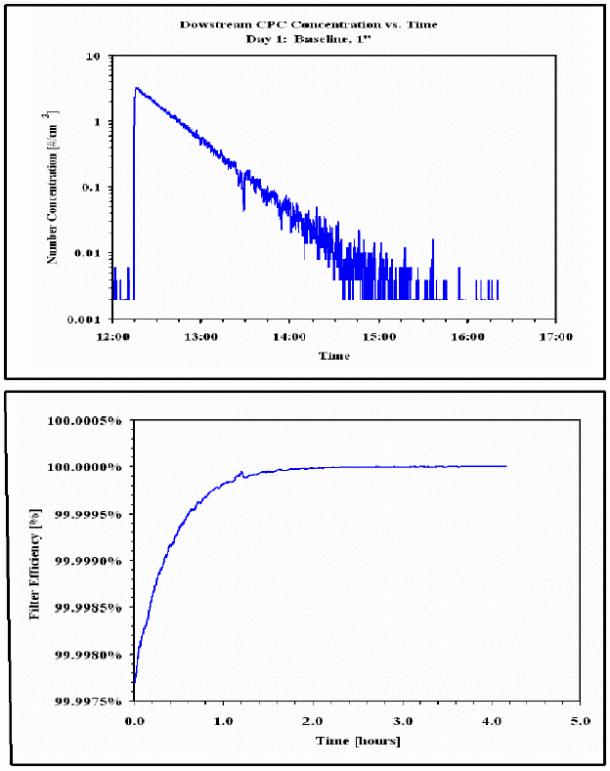


Fig. 4 Top: Log of number density vs. time of particulate loading onto HEPA filter
Bottom: Efficiency of same filter

SUMMARY

As discussed, public concern regarding possible emissions from facilities employing HEPA filtration systems and increasing regulatory scrutiny (HWC MACT) of such facilities has caused many to question

the viability of monitoring particle concentration downstream of such HEPA filters. We have employed this suite of instruments (ELPI, SMPS, diffusions battery, CPC, RM5i) to evaluate measurement uncertainty of PM downstream of HEPA filters. We have compared the results from each measurement technique and have identified a variety of advantages and disadvantages for each. PM mass emission rates downstream of HEPA filters will be on the order of 0.001 mg/m^3. This emission rate is well below the detection limit for EPA RM5i. This emission rate would require approximately 100 days of sampling time to collect the RM5i PQL of a 3 mg catch. RM5i is US EPA's required method for determining PM emission rates from low emission sources and each of the instrumental techniques has been compared to it in regard to overall performance.

While instrumental techniques such as ELPI and DMA/CPC certainly demonstrate the capability to quantify low PM concentrations downstream of HEPA filters, it must be remembered that these units are predominantly designed for research applications. Frequent routine maintenance and calibration are essentially to insuring their proper operation. Diffusion batteries are particularly sensitive to very low PM concentrations, however their operational requirements make automation both expensive and logistically difficulty. Further, diffusion batteries are best suited to characterizing aerosols with small (<300 nm aerodynamic diameters). This dramatically limits their applicability to particle size distributions greater than one micrometer.

The work described in this paper has demonstrated the tendency of the techniques that have been evaluate to become overwhelmed at relatively low PM loading rates. Both the ELPI and SMPS perform best when incoming total particle concentration is $<10^3$ particles/cc, each tends to develop baseline drift when aerosol concentrations exceed that value. Dilution can be used to control aerosol concentration reaching the instrumentation, however, use of these devices may alter the PSD and projected mass loading rates. While not addressed in this paper, it is reasonable to anticipate the tendency of diluters to alter the measured PSD will be a function of both the type diluter employed and how the aerosols are formed. Clearly the SMPS and ELPI are superior measurement techniques for extremely low PM concentrations (as compared to RM5i). But even they fail to provide PSD data downstream of functioning HEPA filters because of the low particle count rates. In general the only unit evaluated in this study that is capable of providing filter efficiency data is the CPC.

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