INTERIM REPORT September 24, 1980

Accession No. _____
Contractors Report No. _____

Contract Program or Project Title: Accident Aerosol Characterization

Subject of this Document: Progress reported for August 1980

Type of Document: Informal monthly progress report

Author(s): Jofu Mishima

Date of Document: September 12, 1980

Responsible NRC Individual and NRC Office or Division:

G. S. Lewis, Systems Performance Research Branch, SAFER:RES

This document was prepared primarily for preliminary or internal use. It has not received full review and approval. Since there may be sutstantive changes, this document should not be considered final.

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NRC FIN No. B2287

INTERIM REPORT

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September 12, 1980

G. S. Lewis
Systems Performance Branch
Safeguards, Fuel Cycle and
Environmental Research
U.S. Nuclear Regulatory Commission
MS 1130-SS
Washington, DC 20555

Dear Lew:

ACCIDENT AEROSOL CHARACTERIZATION-AUGUST MONTHLY REPORT

Approximately 88% ($381K) of the authorized operating funds for FY-80 have been spent.

TASK A. PROJECT MANAGEMENT

As of 8-28-80, approximately $381K of the $435K authorized operating funds have been spent. Approximately $54K remains of which $15K are committed for services outside the section.

Authorization for the additional $10K for combustion product experimental capital equipment has been received.

S.L. Sutter and J.A. Glissmeyer attended the Aerosol Technology meeting held in Boston, Massachusetts on August 18 through 20. A trip report is attached to this report (Attachment 1).

Notification of meetings at Lawrence Livermore Laboratory on October 24 to discuss their fire experiments and at NMSU at Las Cruces on October 27 and 28 (regular RRG meeting) was received.

Summary and Justification of NRC Fuel Cycle Facility Accident Research Program Plans and Proposed Experiments has been issued.

TASK B. DEFINE MAJOR, CREDIBLE ACCIDENTS

Comments on ORNL mid-August "data dump" were written and transmitted 8-28-80.

TASK C. LITERATURE REVIEW

Attached is the NTIS cover page for the EPA report Sue Sutter mentioned as a potential handbook-type format during the last RRG meeting (Attachment 2).

Literature reviews on four topics have been assigned -- Fire Generated Combustion Products, J. Mishima; Fire Models, P.C. Owzarski; Particulate Generation Under Accident Stresses, S.L. Sutter; and Particulate Behavior (with emphasis on constrained volumes), J.A. Glissmeyer.

Over 120 articles, documents, and books on fires/combustion and their products have been accumulated.

TASK D. EXPERIMENTS TO CHARACTERIZE ACCIDENT GENERATED AEROSOLS

Half of the 14 powder experiments outlined in "Experiments Proposed to Measure Aerosols Generated by Free Fall Spills of Uranium Powder and Natural Uranium Solutions" have been completed. Preliminary gravimetric analysis indicates the airborne fraction during the free fall of depleted uranium dioxide, DUO (density 10.9 g/cm^3) is approximately 20% of the amount airborne using TiO_2 (density 4.3 g/cm^3) under comparable conditions. It is anticipated that the full matrix of 28 powder and liquid free fall experiments proposed with data reduction will be completed by December 1980.

A laser analytical technique currently under testing in the Chemical Technology Department shows promise as a useful uranium content measurement technique. Recoveries of 98% and 102% were obtained for DUO samples submitted containing 0.02 and 0.05 g. Testings will be completed by mid-September and, if results warrant, the technique will be used to measure the uranium from the experimental program.

TASK E. TIME DEPENDENT BEHAVIOR OF PARTICULATE MATERIAL

Primary emphasis during this reporting period has been on identifying and selecting appropriate instrumentation for the simultaneous, real-time measurement of airborne particles from several locations during aerosol behavior experiments. Discussions held with attendees at the 10th Aerosol Technology Meeting (see Attachment 1) and a supplemental literature search narrowed the field to 4 techniques. The relative strengths and weaknesses of the techniques are discussed in Attachment 3. Based upon the programmatic requirements and funds available, the light scattering particle sizer manufactured by Climet (range 0.3 to 20 μm) appear to be the best candidate. It appears to have the best capacity for use in a multi-sensor system with the broadest application in our experimental program. A demonstration of the instrument is scheduled

in the 1st half of September. When used in conjunction with an Electric Aerosol Analyzer (Thermal Systems Inc. Model 3030), a size range of 0.003 to 20 μm can be spanned.

If you have any questions, please contact me.

Sincerely,

Jofu

Jofu Mishima
Applied Meteorology & Emissions Assessment
Atmospheric Sciences Department

JM:dh

Enclosures

cc: W.S. Gregory/R.A. Martin - LASL
H.W. Godbee/E.J. Fredrick - ORNL

ATTACHMENT 1

Battelle
Pacific Northwest Laboratories

Project Number

Internal Distribution

GA Glissmeyer
RK Hadlock
J Misnima
PC Owzarski
SL Sutter

Date August 27, 1980

To Distribution

From SL Sutter/JA Glissmeyer

Subject 13th Annual Aerosol Technology Conference, August 18-20

During the subject meeting we made several contacts of interest to us in our accident analysis contract work. There was some interest in Sue's free fall work that we want you to be aware of.

Melvin First, Harvard School of Public Health, was very interested in Sue's experiments. He suggested investigating the same powder volume rather than the same weight as a basis of relationships, because of drag effects, etc. He said the results of these tests might be far more useful than we might have comprehended.

Regarding John's sampling in Paducah, Mel hasn't heard a word from Ed Vallario nor received a copy of the drafted report. John told him Vallario instructed Lysle not to send Mel a copy. (Vallario said no one should be sent copies prior to the official DOE printing and distribution.) Mel gets the feeling he's been had. John indicated similar feeling.

Janet M. Macher was instructed by Dr. First to keep in contact with us and follow the study. She is beginning graduate study with him. They are investigating leakage from safety chambers. Sue has Janet's address and gave her her business card. She gave Sue some names to follow in her literature search.

Clyde Roe, staff consultant on consumer product safety at United States Gypsum Co. was interested in these experiments, and asked for the final report. Sue did not have an opportunity to talk further with him.

Fred Gelbard, MIT, would like Sue's report when it is finished. He is interested in modeling aerosol behavior in chambers.

Bob Gussman, BGI Corp., is familar with the carbon brush disintegration Sue noticed in her experiments. He suggested a remedy that has been used if this becomes a problem. Moving the hi-vol motor outside the RART and connecting a flexible hose to the sampler inside would overcome this problem. The carbon could become a problem if she used particle size instrumentation. The hi-vol motors have a 900 hour life. He knows of people who polish the armature parts and extend the motor life to 1800 hours.

Marv Tillery, LASL, has been contacted by Dick Martin and Bill Gregory. However, he is not really involved in the accident aerosol program. Sue discussed sampling with him. He felt that it is important to keep the same sampling method over a range of comparative experiments, since each one has a built-in bias.

Chuck Fairchild, LASL, operates one of the wind tunnel facilities Dick Martin and Bill Gregory are planning to use for the accident aerosol program. His present

work uses spherical aluminum metal in two size ranges. He will send us information on these tracers and on the wind tunnel, etc. He weighs the collection on 47 mm filters and uses eosin tracing. He has recently compiled an article for journal publication and he will send Sue a copy.

Chuck has used PMS's CSASP and ASAS-X light scattering probes as tools in his experiments. He did not feel competent to make any recommendation, however. He has visited Jim Wedding to discuss the use of the light scattering particle sizer but was disappointed at not being shown calibration data. We had not seen such data in the past either, although Bob Smith (HEDL) had paid Jim some $10K to formulate specs for the device.

Jim Wedding, Colorado State Univ., tried to sell Sue a wind tunnel on credit. Apparently Jim feels his light scattering particle sizer is developed to the point of commercialization.

Virgil Marple, University of Minnesota, will send Sue final reports on coal dust instrumentation. Sue gave him a literature reference where he reports on this work and he said to call him if she does not receive it to remind him.

Virgil gave a talk on the performance of Berkeley Industries QCM impactor. He found the stage cut-points were sharp and agreed well with theory up to 1 µm. While above 1 µm the cuts were slightly lower than predicted, they were sharp and reliable. Interstage losses were highest for the larger particles and peaked at 45% for around 10 µm and decreased as size increased. For sampling at 250 cc/min the cuts for the ten stages ranged from 0.12 µm to 15 µm. He referred John to Don Wallace to get a copy of the results of the study.

Don Wallace, Berkeley Industries, pointed out that the interstage losses for particles >3.5 µm in the QCM impactor result in poor agreement with concentration measurements obtained gravimetrically. He thinks we need a real time sizer like his QCM, of course, and will send a copy of Virgil's results. (Berkeley seems more interested in this kind of instrument testing than their competitor, California Measurements.)

Ray Chuan, Brunswick Corp., (one of the developers of the QCM impactor) discussed the virtues of the QCM impactor with John while they strolled through Bloomingdale's and waited for a table at Legal Seafoods. He has extensive data on the use of the QCM for measuring fire combustion aerosol sizes as well as a number of aerosols in the troposphere and stratosphere. He will send John some of his reports on combustion aerosols. He feels the California Measurements QCM impactor would perform the same as Berkeley Industries model because they use the same impactor design. However, the electronics and crystal mounts in the California Measurements unit are redesigned and improved over Berkeley's allowing for easier operation. California Measurements are very good at custom applications, he feels. Ray gave John some tips on using the QCM at high concentrations (gram/m^3) by pulse sampling and on quickly renewing the collection crystal.

Sue talked to John McCarthy, MIT, who works with coal combustion aerosols. Sue did not get much information from him because she was unfamiliar with Jo's combustion problem. John has had some problems with early versions of PMS's

CSASP and ASASP probes because of internal leakage. He favors using the Climet 208 but with added signal capabilities, vis a vis multichannel analyzer.

In McCarthy's paper he described his apparatus for studying coal combustion aerosols. His combustor was a downdraft laminar flow tube furnace. Aerosol analysis was by EAA, gravimetric filters, Andersen impactor, and SEM microscopy with image analysis. The image analysis was used for sizing chain aggregate aerosols and used the EAA for monitoring relative aerosol stability. Agreement between the two was not assured. Samples were extracted via a combustion aerosol "quencher" which used a porous stainless steel tube for dilution.

John discussed optical particle counters with Ed Avol, Rancho Los Amigos Hospital. He has done a market search similar to ours; however, he's had a chance to try some of the instruments. He's settled on a Climet 208 but for signal processing uses a computer as a multi-channel analyzer. He has done extensive comparison between Climet and Royco.

John also discussed optical particle counter applications with J. Gebhart, Gesselshaft fur Strahlen u Umweltforschung, Frankfurt. He used to work for Battelle-Frankfurt and was of the opinion that any young scientist interested in research should start with a few years at Battelle for an education. (How about that?) Although John lost the battle to stay awake during his paper, he showed some modifications he made to the Karlsruhe OPC (marketed by PolyTec) that permitted under certain circumstances to operate at concentrations up to 10^6 particles/cc.

Jugal Agarwal, TSI, indicated they may be able to take orders for the laser doppler velocimeter aerodynamic particle sizer early next year. Delivery may take a few months.

Klaus Willeke, Univ. of Cincinnati, requested copies of our final powder leak reports. From his experiences with PMS's optical particle counters, their major problem in early models was internal leakage.

David Bright, NBS, briefly described to John that he sized individual large particles by their rate of fall along a vertical lazer beam located axially in a vertical laminar flow tube a few mm in diameter.

G. Kasper, SUNY at Buffalo, gave a paper describing his use of a cylindrical aerosol centrifuge to measure the relationship between AED and long chain aggregate particles. He showed there was poor agreement between the centrifuge and the electrical aerosol analyzer (EAA). The volume mean diameters measured by electrical mobility (EAA) range from 0.11-0.14 μm and when measured by the centrifuge, the range was 0.4-0.9 μm.

John discussed the use of EAA with Gilmore Sem, TSI. Gil described the functions of the computerized data reduction system for the EAA. Gil and John got to practice their Norwegian on each other while touring the GCA plant.

John discussed the performance of the GCA real time monitor (RAM-1) with George Quackenbos and Pedro Lilienfield, GCA. They offered no data on RAM-1 performance versus particle size and refractive index. They suggested the best

way to calibrate the RAM-1 was against the actual aerosol using a filter sample as a standard.

On August 21 John visited with Bruce Weiner, Malvern Scientific Corp., at Ronkonkoma on Long Island. They discussed the application of the Malvern ST 1800 particle and droplet sizer to our experiments. The claimed sizing range of interest to us was 1.2-118 μm. John had taken samples of TiO_2 and ZnS and tried unsuccessfully to get a response from the instrument by blowing some of the powder through the sensing volume. This was a disappointing but perhaps poorly conducted test. When the powders were slurried, the results were about as expected from John's earlier data using the Quantimet Image Analyzer and a hi-vol impactor. The volume mean diameter was 5.5 μm for ZnS and below range for TiO_2. The instrument does not measure concentrations at this time but a data reduction program in the works will add this capability in a half year. If John had a Wright dust feeder with him, perhaps a better demonstration would have resulted.

Sue took the Battelle Staff Association Mount St. Helens slides to the Aerosol Technology meeting and showed them at a special session she and Ray Chuan put on. He is doing volcano plume sampling via aircraft and had movies of his work. There was very much interst in this session.

ATTACHMENT 3

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.	2.	3. PB 275 525
AP-42		

4. TITLE AND SUBTITLE	5. REPORT DATE
Compilation of Air Pollutant Emission Factors Third Edition (Including Supplements 1-7)	August 1977
	6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S)	8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS U.S. Environmental Protection Agency Office of Air Quality Planning and Standards Monitoring and Data Analysis Division Research Triangle Park, N. C. 27711	10. PROGRAM ELEMENT NO.
	11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS U.S. Environmental Protection Agency Office of Air and Waste Management Office of Air Quality Planning and Standards Research Triangle Park, N. C. 27711	13. TYPE OF REPORT AND PERIOD COVERED
	14. SPONSORING AGENCY CODE 200/04

15. SUPPLEMENTARY NOTES

16. ABSTRACT

Emission data obtained from source tests, material balance studies, engineering estimates, etc., have been compiled for use by individuals and groups responsible for conducting air pollution emission inventories. Emission factors given in this document, the result of the expansion and continuation of earlier work, cover most of the common emission categories: fuel combustion by stationary and mobile sources; combustion of solid wastes; evaporation of fuels, solvents, and other volatile substances; various industrial processes; and miscellaneous sources. When no specific source-test data are available, these factors can be used to estimate the quantities of primary pollutants (particulates, CO, SO_2, NO_x, and hydrocarbons) being released from a source or source group.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Fuel combustion Emissions Emission factors Mobile sources Stationary sources		

18. DISTRIBUTION STATEMENT	19. SECURITY CLASS (This Report)	21
Release Unlimited	Unclassified	
	20. SECURITY CLASS (This page)	22. PRICE
	Unclassified	

EPA Form 2220-1 (9-73)

ATTACHMENT 3

QUARTZ CRYSTAL MICROBALANE (QCM) SYSTEM

The ten stage QCM impactor has the following theoretical aerodynamic euqivalent cutpoints: 0.07, 0.14, 0.28, 0.57, 1.1, 2.3, 4.5, 9.1, 17.7 and 35.4 microns. Rubow and Marple (1980) found the cuts to range from 0.12 to 15 μm at a flow of 0.25 ℓ/min. Impactors with fewer stages up to 4.5 μm are also available. Cutpoints are sharp but interstage losses are significant above a few microns and peak at 45% at 10 microns. These losses tend to make size data for the larger particles unreliable. Perhaps then the six stage versions present the useful limit, but estimates given here will be in terms of the ten stage units. At least two of the submicron stages are operating at low pressure (~0.5 atm) which may result in problems when sampling combustion aerosols. Some submicron combustion aerosols will evaporate at reduced pressure and this may cause erroneous readings in the low pressure stages if it is desired to obtain the size data at atmospheric pressure.

The impactors measure aerodynamic diameters rather than equivalent area or volume spheres measured optically. This is a plus for our data needs. Data output for the California unit is in the form of concentration (g/m^3) per stage and a small histogram is plotted normalized to total concentration. Also printed are total concentration and sampling time. Printed format is the roughly 2-inch wide paper tape. (I'm not sure the same format applies to the Berkeley unit.)

Each impactor comes with its own electronics and data reduction packages. Thus, a system would compose of n impactors and n electronic and data packages. It would be a custom package to have several impactors feeding a single data unit.

The standard intake is through a ball value. Both Berkeley Industries and California Measurements have options where the valve can be operated remotely. The Berkeley unit has an optional timer/programmer to govern valve operation.

The California unit has an optional inlet valve which permits pulse sampling allowing the monitoring of concentrations up to 10 g/m^3 (the standard is 60 $\mu g/m^3$ per stage). The special valve is just a sliding sampling valve used in gas chromatography with a sample loop volume of 30 microliters.

An inherent problem with QCM's is that they have a limited operating time. For a given aerosol, the curve of frequency change versus collected mass is linear up to a given Δf characteristic of the aerosol. For nearly all aerosols the lowest limit is a Δf of 1200 Hz. Dry carbon black has no linear response (Sem, Tsurabayashi, Homma 1977). A limit in the linear frequency change limits the mass that should be collected per stage to about 5 μm. Thus, if a 1 mg/m^3 aerosol were collected evenly over ten stages, the maximum length of continuous sampling time is 2,000 min. When the limit is reached, the collection surface should be renewed. This can be done rapidly by replacing the stage, turning the crystal over, or interchanging the sensing and reference crystals. (I'm not sure this is possible in the Berkeley unit. Each stage has two crystals, one used for temperature reference and the other for mass collection. Each crystal has two faces giving a total of four collection surfaces available per stage according to Ray Chuan.) Each crystal is usually coated with a special grease. The California unit has a built-in coating facility. The Berkeley unit comes with a coating kit.

The output tape of either QCM can be monitored to see when surface renewal is required. California Measurements would build a controller that regulates sampling time that would include the ability to automatically stop sampling once a preset sampling time or Δf on any stage is encountered.

According to Ray Chuan, the impactor design of both QCM's is the same, but the California Measurements has a second generation electronics/data package and routinely builds custom units for him. Virgil Marple indicated that the impactor design of the Berkeley unit is "cleaner" than the California unit in terms of flow protrusions in the impactor. The Berkeley Industries people seem to be the most active in obtaining performance data on the QCM.

The pricing and specs given here are based on non-custom units without any automatic control. The level of data reduction included in the prices is limited to the description in an earlier paragraph. Both units have a continuous readout of frequency change of one selectable stage.

Specifications

- 10 stages with aerodynamic cutpoints of 0.07, 0.14, 0.28, 0.57, 1.1, 2.3, 4.5, 9.1, 17.7, and 35.4 microns
- 0.24 to 0.25 ℓ/min sample flow
- maximum sample temperature 80°C
- warm-up time 10-20 min.

Pricing

	1 Unit	3 Units
Berkeley Industries 10 stage*	$13,840	$41,520
California Measurements 10 stage	$13,750	$41,250

* May not reduce data beyond Δf per stage. California Measurements offers the high conc. option (sliding sampler valve) for $950 for one impactor or $2850 for three units. Diaphram pumps cost about $185 and matched pairs of crystals cost $60/pair.

Advantages

- measures aerodynamic size directly
- measures concentration directly
- shouldn't require as much recalibration as OPC's.

Disadvantages

- possibility for significant inlet losses of large particles because sample must be extracted and piped into sensor.
- inaccurately measures sizes larger than a few microns because of large interstage losses.
- sampling may be interrupted because of crystal overloading when accurate dilution or pulse sampling are not provided.
- possible erroneous readings for submicron volatile aerosols.

CLIMET SYSTEM

This system could be built around the Climet 208 particle counter. The standard size ranges are: 0.3, 0.5, 1, 3, 5, >10 microns. (For $100 extra, an additional range of 0.65 to 20 microns.) Both ranges cannot be run simultaneously.

Each size range is monitored in turn with a digital display showing the number of particles greater than the selected size. In the standard model each range is selected manually but an option is available for automatic sequencing through the ranges. The concentration upper limit before coincidence errors must be taken into account is 10^4 particles/cc. A dilutor should be added upon placing the order to enable concentrations up to 10^6 particles/cc. A high temperature version of the 208 is available for up to 350°C. This would allow measuring combustion aerosols without prior quenching. This model is not considered here because quenching is necessary for the EAA anyway. Also, the dilutor cannot be used simultaneously with the high temperature option. It should be remembered that quenching does allow the condensation of vapors and hence the introduction of new aerosols.

If the 208 is to be used as a single package, a Model 237 printer should be considered. This printer has an internal clock and capability for setting alarm thresholds. A Model 236 printer is available without the interal clock or alarm contacts. A clock is worthwhile since the printed record has the sampling time recorded.

The analog output of the 208 can be fed into a Model 210 Multichannel Analyzer. The Model 210 has an internal printer. It also has a capacity for 48 analog input channels of particle size data. The number of particle size channels per 208 sensor is available in multiples of 8. The standard input board allows for 8 size channels with ranges of 0.3 to 0.5, 0.5 to 0.7, 0.7 to 1, 1 to 2, 2 to 3, 3 to 5, 5 to 10 and >10 microns. (The channels are, of course, different for the 0.67 to 20 μm range.) Further finer subdivisions of the range is obtained in multiples of eight. With a three-sensor system we could collect either 8 or 16 channels of size data per sensor. The output data is in terms of particle count per channel. A microprocessor selects the sequencing of data printout. Sampling time is printed as well as particle count per channel. All size channels are monitored simultaneously.

A built-in digital display on the 210 lets the user see the particle counts in one channel. Of course, the display on each of the Model 208 sensors displays particle count > selected size so the operator can watch those displays too. If this is found to be unnecessary, we might as well buy the Model 225 particle counter instead of the 208. It supplies the same sizing data to the 210 but doesn't have a built-in size range selector and digital display. The 225 costs $1950 less than the 208. (Climet recommends using the 225 for a system such as we are building.) Inasmuch as the 225 has no built-in display, it must be used with the 210--it cannot just be used with the printers. The 225 is available in the high temperature configuration. It is also available with the dilutor but that would lengthen delivery time. Again, the dilutor and high temperature cannot be used simultaneously.

Specifications

flow rate	0.25	SCFM	standard
	0.0025 - 0.25	SCFM	with dilutor
size ranges	0.3 to 10 µm and 0.65 to 20 µm		subdivided into 5 "greater than" ranges or multiples of 8 channels
optical config.	quartz halogen lamp, elliptical mirror, photomultiplier tube, collecting angle of 20 to 105°		

Pricing

Prices are as of August 1980 based on 0.3 to 10 and 0.65 to 20 micron dual range capability.

Item	3 Unit System	One Unit System Allowing Expansion	One Stand-Alone Unit
Model 208	$18,150	$6,050	$6,050
Dilutor	2,700	900	900
210 MCA 24 channel	7,500	8-Channel 5,450	
Printer			2,250
	$28,350	$12,400	$9,200

Item	3 Unit System	One Unit System Allowing Expansion	One Stand-Alone Unit
Model 225	$12,300	$4,100	
Dilutor	2,700	900	
210 MCA 24-channel	7,500	8-channel 5,450	
	$22,500	$10,350	

Advantages

- when used with 210 can monitor particle size in several channels simultaneously.
- can come with manufacturer installed dilutor
- compared to other OPC's is least sensitive to variations in real refractive indix. This might lessen the need for calibration with every aerosol. See the figures below.

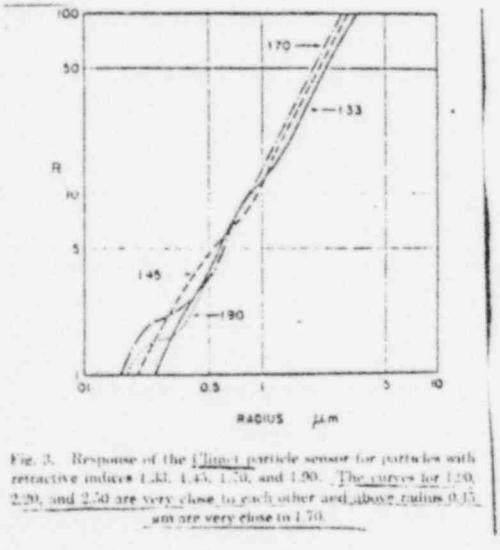
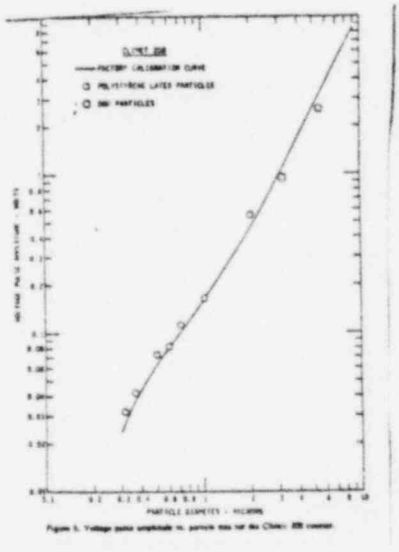


Fig. 3. Response of the [illegible] particle sensor for particles with refractive indices 1.33, 1.45, 1.50, and 1.90. The curves for 1.50, 2.20, and 2.50 are very close to each other and above radius 0.35 μm are very close to 1.70.



From Cooke and Kerker, 1975

From Clark and Avol, 1979

- the particle size per response voltage is single valued as shown above. This is because the wide diffraction angle observed.
- inexpensive to build a system.
- calibrating using Marple's method could yield aerodynamic size data.

Disadvantages

- upper limit of size range is only 20 μm.
- separate MCA required if calibration by Marple's method is attempted.
- Model 210 MCA channel voltages are factory set. It would be very difficult to change in the field if required due to using special calibration.
- possibility for significant inlet losses of large particles because sample must be extracted and piped into sensor.
- if exposed to dense combustion aerosol of chain agglomerates, the optical chamber will probably require cleaning.
- size resolution decreases as particle shape becomes less spherical.
- theoretically sensitive to variation in imaginary index of refraction which may be a problem for absorbing particles (carbon) as shown below.

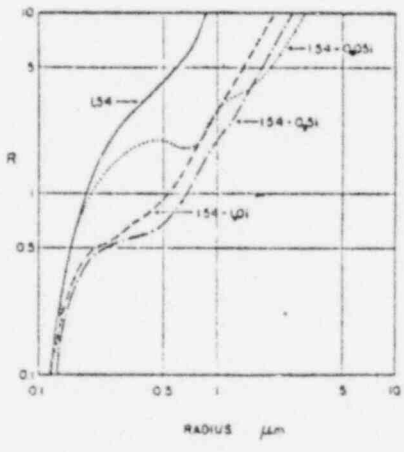


Fig. 8. Response of the Climet particle sensor for particles with refractive indices 1.54, 1.54–0.05i, 1.54–0.5i, and 1.54–1.0i

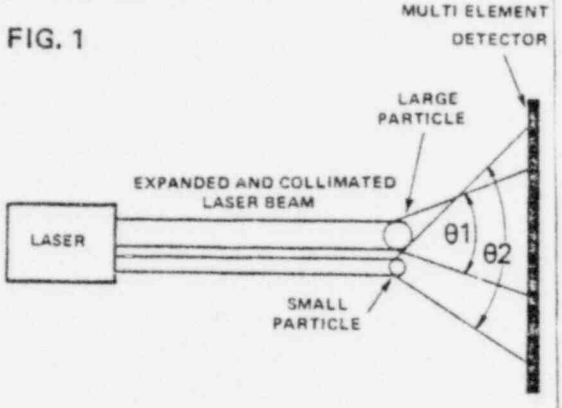
From Cooke and Kerker, 1975

MALVERN SYSTEM

This system would be built around the Malvern ST 1800 Particle and Droplet Sizer. The ST 1800 uses an optical technique for particle sizing but it is not a single particle counter. Instead, the sensitive volume contains many particles at one time. Sizing is based on the Fraunhofer diffraction intensities of a large number of randomly oriented particles. Since this technique is new to all of us, the following quote from the manufacturer's literature describes the method.

PRINCIPLE OF OPERATION

FIG. 1



Diffraction angle Θ' small for large particles.
Diffraction angle Θ' large for small particles.

SCHEMATIC OF MALVERN PARTICLE SIZER

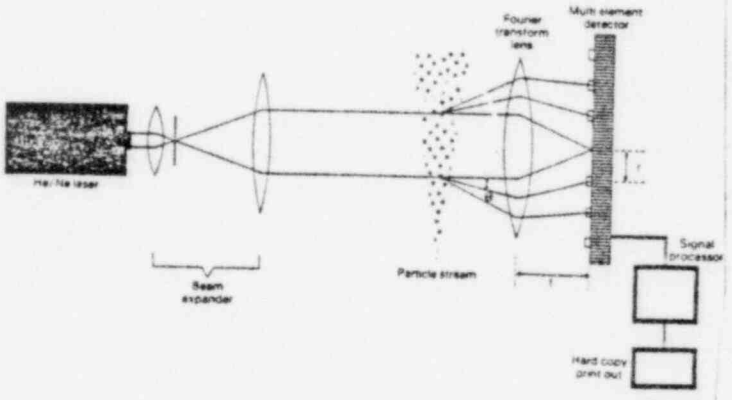


FIG. 2

If a laser beam falls on a spherical or non circular particle a diffraction pattern is formed, whereby some of the light is deflected by an amount dependent upon the size of the particle. Fig. 1. If a suitable lens is placed in the light path behind the particle and a screen or detector placed at the focal plane, then light not deflected by the particle is brought to a point focus on axis and the conically diffracted light by the particle is "focussed" around the centre spot at a distance which is a function of that particular particle size. If particles of different diameters are present in the laser beam, then a series of "focussed" rings will be generated at various radii, each focussed ring being a function of different particle sizes. Since the pattern produced on the detector is obtained by focusing parallel rays, it is a far field pattern and is known as Fraunhofer Diffraction. The light energy contribution at any of the focussed rings in the focal plane is simply the sum of the contributions from each individual particle, of all sizes, and although any size of particle diffracts a certain amount of light to all radii, the energy distribution curve peaks at one particular radius. There is therefore an optimum radius for detection of each particular particle size.

In the Malvern Particle Sizer this light energy information is extracted from the diffraction pattern by using a specially developed multi element detector consisting of a series of detectors at increasing radius. The detectors are rapidly scanned electronically and the output taken to the signal processor, the output of this processor provides hard copy read out.

When particles are above a couple of microns in size, the light scattering in the forward direction as predicted by Mie theory is closely approximated by the Fraunhofer diffraction theory for discs. Hodkinson (1966, pg. 324) stated:

> "Their value lies in providing a simplified, general description of the scattering patterns of polydisperse systems of fine particles larger than 2 or 3 microns in diameter. Such particles show a forward mixiumum corresponding to Fraunhofer diffraction, increasing in intensity and decreasing in angular spread as the particle size increases and depending very little on the material of the particle."

On Fraunhofer diffraction, van der Hulst (1957, p. 104) has pointed out that:

> "The intensity distribution in this diffraction pattern depends on the form and size of particles but is independent of its composition or the nature of its surface. For example, a black (fully absorbing) body, a white (diffusely reflecting) body, a totally reflecting body, or a glass body of the same shape, all have identical diffraction patterns."

This technique would then seem to be appealing for measuring particles larger than a micron. This method has been used principally for measuring slurries or dry powders in the Leeds and Northrup Microtrac instruments and in the Malvern instrument.

For airborne particles the advertised sizing range is 1.2 to 1800 microns divided into three subranges by the use of different lenses. The subrange of potential interest to us is 1.2 to 118 microns divided into 15 size brackets. (Thresholds of 1.2, 1.5, 1.9, 2.4, 3.1, 3.9, 5.0, 6.4, 8.2, 11, 14, 18, 24, 34, 55, and 118). The top useful size is 55 microns.

Generally, the amount of light scattered into the receiver is proportional to the equivalent projected area of a particle. Because many particles are in the sensing volume simultaneously, and their orientation is random, light is scattered at many angles and many intensities. The deconvoluted particle size distribution is then proportional to equivalent volume spheres.

The sampling interval can vary from 12 milliseconds to 50 seconds. Data reduction is performed by a dedicated PDP-8 minicomputer and output is via a

teletype. Reduced data can also be outputed to other peripherals. Data processing is handled by two different methods: 1) no special distribution assumed, and 2) the data are force-fitted into a Rosin-Rammler distribution. In either case, the results are presented in terms of a volume percent per bracket and cumulative volume percent per bracket. Where the Rosin-Rammler distribution is used, a histogram is also presented. I'm told a log-normal distribution can be fitted with some custom programming. Also in the near future relative concentration data will also be presented. (It seems this latter option would only be accurate if flow through the viewing volume is constant.) With some custom programming the data reduction system can be made to handle several sensors operating sequentially. Data from 64 sizings can be stored before processing, permitting rapid sequencing.

The laser and receiver are mounted on an optical bench and the "throw" between the two is about three feet. The "viewing volume" extends for 1 to 2 cm along the laser beam at a distance of 6.3 cm from the laser expanding lens. The actual laser and receiver can be placed in several geometries if proper mirrors are used and alignment can be maintained. The instrument can be used by placing the "viewing volume" in a dust chamber, liquid cell, wind tunnel, a glass slide, or a variety of configurations.

This all sounds very good except that during a demonstration in New York we couldn't get the instrument to respond when I blew zinc sulfide across the beam. The demonstration may not have been very fair because I may not have kept the dust in the viewing volume for very long. A better demonstration would result if Malvern could try their instrument during one of Sue's experiments or if I could take a Wright Dust Feeder with me to the instrument. (The closest one is in Los Angeles--but then we might have to do that for a demonstration on the QCM's. Climet would bring theirs to us.)

Specifications

Power:	220 VAC, 50 Hz, 1000 Watts (110 operation optional)	
Weight:	transmitter/receiver	31 kg
	processor	25 kg
Accuracy:	computed mean diameter (Rosin-Rammler) 4%	
Temperature:	15 to 30°C	

Pricing - July 1980

	Triple Unit	Single Unit
1st unit + processor	$34,390	$34,390
additional sensors (2)	24,000	
	$58,390	$34,390

Advantages

- no extraction losses should allow accurate sizing of large particles.
- variable sample geometries possible.
- equivalent volume sphere size data.
- size range will accommodate largest particles.
- no calibration necessary.
- sizing independent of refractive index.
- some flexibility in programming and sequencing.

Disadvantages

- had an unconvincing demonstration.
- upper concentration limit is vague because there is no set velocity through the system. Might be about 10^4 particles/cc, might not.
- not much published literature concerning instrument, either supportive or critical. (This can be seen as an opportunity for us to start filling the vacuum.)
- no submicron sensitivity negates any use for well dispersed TiO_2 or other submicron aerosols. This can be compensated for by employing an EAA for this range. There are probably several experiments we'll want to do where it would be far more convenient to cover the tenths to tens of microns (0.3 to 20) with a single instrument.

PMS SYSTEM

The system described here is based on the Particle Measurement Systems (PMS) Classical Scattering Aerosol Spectrometer Probe - 100 (CSASP-100) which comes in two configurations and can be mated to a variety of data acquisition systems. I'll describe the standard CSASP-100 configuration first.

The CSASP-100 uses illumination from a 5 mVHe-Ne laser operating in high order multimode*. Forward scattered light is collected in the range of 4 to 22° off axis. The aerosol is drawn through the sensing volume two ways: 1) by a miniature trumpet-shaped wind tunnel (with 3 cm min. diam.) for ambient sampling (276 ℓ/min) and 2) an insert permitting use as a plumbed-in unit (5 ℓ/min). The unit has four overlapping size ranges (typically 0.3 to 0.75, 0.5 to 2.75, 1 to 12.25, 2 to 20 microns) with fifteen equally spaced channels each. All channels are monitored simultaneously and the sequencing through the ranges is automatic. The normal temperature range is -25 to 40°C. A high temperature version is available for temperatures up to 400°C. As explained for the Climet system, the high temperature version is not included in this price estimate.

A second configuration for the CSASP is one where the laser and receiver can be mounted separately on a bench or across a duct. The farther apart the laser and receiver are, the sensitivity for small particles decreases. For example, the smallest size of 0.3 microns increased to 0.5 microns when the instrument was mounted across an 11 inch duct. In addition, velocity must be maintained constant and at least 1 m/sec across the sensitive volume. This would require us to aspirate at least a 1.8 ℓ/min sample anyway (given a 3 mm x 10 mm cross section) to get a sample to that velocity and maintain it. This configuration is more applicable to a small wind tunnel. The advantage of this configuration to the standard one is that it may allow us to have a shorter distance between intake and sensor. This may be important for large particles.

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* The intensity across a laser beam is usually normally distributed. High order multimode means there are several intensity maxima which tend to flatten the distribution.

There are a variety of data reduction systems available depending on the approach we take. If we have a single sensor and just want particle count per channel, $9,800 PSD-200 would be sufficient. If we want several probes and particle count per channel, a DAS-64 would cost $14,000. If particle concentration per channel, size distribution plots, calculations, four probes, a PDS-300 with a printer would run $17,300. PMS is now doing a custom system where data from a CSAS and a TSI-EAA are integrated. I was given no estimate of cost for this, but the development work is already paid for by another customer.

Pinnick and Auvermann (1979) have done the most thorough evaluation of the CSASP-100 so far. Pinnick has told me they were rather pleased with their instruments. He warns that the resolution of the CSAS doesn't merit 60 size channels over the whole range (because of multivaluedness of size versus response) but he groups some channels together for a total of 32. Also, because of sensitivity to refractive index, the groupings may have to be different for different aerosols. This is illustrated in the following two figures from Pinnick.

It is clear that a given instrument response correlates to different sizes depending on the aerosol index of refraction. Figure 4 shows experimental data for three aerosols plotted next to theoretically derived curves. Figure 12 shows theoretical calculations for a variety of indices. It may turn out that experimental response curves may show less response than the theoretical ones. (Pinnick has offered to do these calculations for us if we furnish the indices of refraction to him.) In any case, it would be prudent to do some calibrating of our own using the Marple method.

Specifications

Size Ranges:	0.3 to 0.75 0.5 to 2.75 1.0 to 12.25 2.0 to 20 microns
Power:	115 VAC, 60 Hz, 1 amp

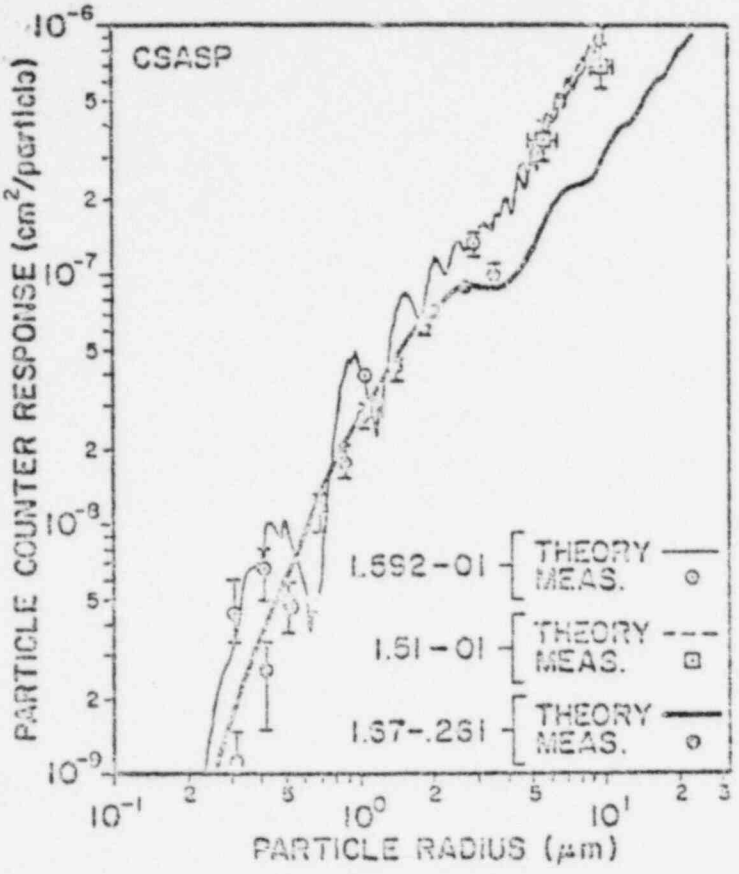

Figure 4. Knollenberg CSASP response: measured (circles and squares) and calculated using Mie scattering theory (curves) for single spherical particles versus particle size. The measurements have been normalized for best fit to the calculated response for polystyrene latex particles with refractive index n = 1.592-0i. The theoretical curve for glass beads with refractive index 1.51-0i extends down only to about 5μm radius.

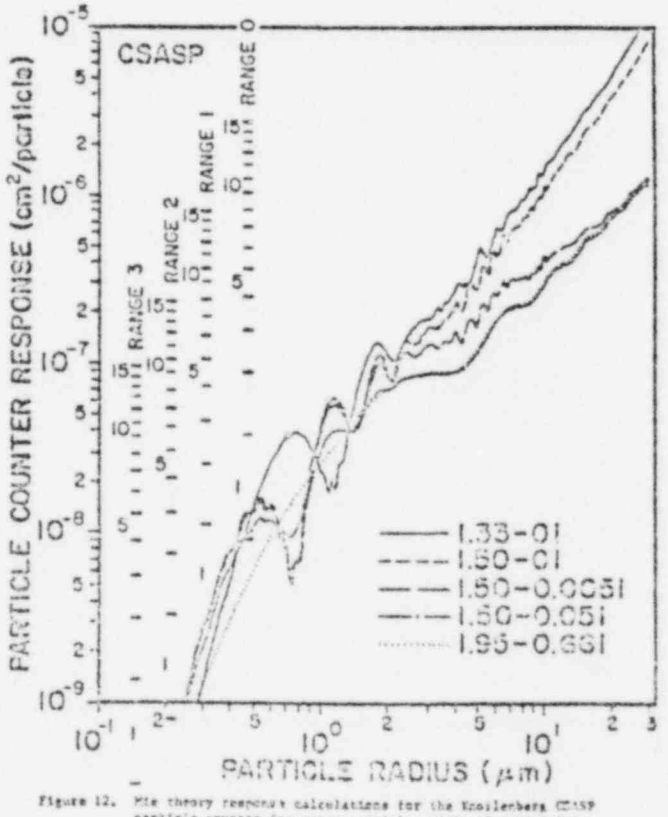

Figure 12. Mie theory response calculations for the Knollenberg CSASP particle counter for water particles with refractive index 1.33-0i, ammonium sulfate with appropriate index 1.5-0i, atmospheric dust with indexes 1.50-0.005i and 1.5-0.05i, and carbon with index 1.95-0.66i. The tick marks indicate the pulse height discriminator levels as set by the manufacturer for the counter. Channels 1, 5, 10, and 15 are labeled between the appropriate tick marks for the different ranges of the instrument. The heavy tick marks indicate the pulse height discriminator levels used to avoid regions of multivalued response under the assumption that particles are water.

- don't have an off-the-shelf dilution system.
- some possibility for losses of large particles during extraction.
- maximum concentration 10^6 pt/cc<3 μm, 10^4 pt/cc >3 μm.

Price

	Triple Unit	Single With Growth Capability	Single Stand-Alone Unit
CSASP-100	$33,000	$11,000	$11,000
Data System	17,300 PDS-300	17,300 PDS-300	9,800 PDS-200
	$50,300	$28,300	$20,800

Advantages

- high resolution of particle size (32 useful channels instead of 8 or 16 for Climet for essentially the same range).
- low extractive flow rate (5 ℓpm)
- sophisticated signal processing.
- simultaneous monitoring of 15 channels from each sensor--automatic sequencing through each sensor range.
- calibrating using Marple's method could yield aerodynamic size data.
- less of a problem for large particle loss than either the Climet or QCM's because of large flow channel through optics. If we used the open configuration of the probe with an abbreviated accelerating and sampling nozzle, that would improve our chances of sampling large particles even more.
- for small particles a higher concentration without dilution is acceptable (10^6 pt/cc <3 μm).

Disadvantages

- upper limit of size range is only 20 μm
- separate MCA required if calibration by Marple's method is attempted.
- would require competent instrument man to change MCA channels
- some sensitivity to index of refraction as shown in the figures.

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