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AEROSOL DISSEMINATION ASSESSMENT

Third Quarterly Progress Report

by

R. Dennis
D. Gordon

October 1966

PHYSICAL RESEARCH LABORATORIES
RESEARCH LABORATORIES
Edgewood Arsenal, Maryland 21010

Contract DA-18-035-AMC-376(A)

GCA CORPORATION
GCA TECHNOLOGY DIVISION
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FOREWORD

The work described in this report was authorized under Project 1B522301A081, Chemical Agent Dissemination (U). The work was started in December and completed in March 1966 for this reporting period.

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DIGEST

This report describes the work accomplished during the third three months of a program to develop, design and fabricate a complete chamber aerosol assessment system. The system is intended to be a major tool in the development of chemical devices, and applicable to a broad range of agents and dissemination methods. Aerosol description in terms of chemical composition and particle size properties should provide useful data on disseminator performance and be relatable to biological effects. Sensor output from instruments in the system should be amenable to either direct display or data storage and processing or both whenever the latter techniques prove to be the most effective methods for obtaining real-time measurement of aerosol-cloud properties.

During this period, the selection of instruments and experimental methods has been further reduced based upon results of the information survey and a laboratory test program. The reasons for selecting either specific instrumentation or techniques or both are cited. Similarly, the basis for rejection is indicated for certain methods, which do not appear applicable to the present system.

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I. INTRODUCTION

Basic program objectives which have been outlined in previous quarterly reports (Ref. 1,2) are restated here for immediate reference.

The objectives of this program are to develop, design and fabricate a complete chamber aerosol assessment system. The function of this system is to provide, by the selection and integration of appropriate instruments and analytical methods, a detailed description of chamber aerosols from as near as possible to the instant of dissemination to time periods up to one hour. By continuous or incremental sampling, the state of the aerosol should be definable over time intervals as small as one second.

Since the system is to be used as a major tool in the development of chemical devices, it should be adaptable to the assessment of a broad range of disseminators and agents and to most test chambers now used by United States Army Edgewood Arsenal groups and contractor organizations. Furthermore, extraneous effects attributable to chamber geometry, aerosol mixing, wall losses, electrostatic charge, agglomeration, etc., should not interfere with proper assessment of disseminator functions.

To provide a broad capability in aerosol assessment, component instrumentation should permit the following measurements: mass concentration of active agent, concentration of degraded agent or inert material, descriptive particle size parameters for active agents, and shape factor for solid particles. In addition to covering a broad range of particle size, < 1 to 100 microns (μ), particle dimensions should be relatable to those describing biological effectiveness of aerosol clouds.

Aside from the fact that system components should define aerosol cloud properties described above, sensor outputs from these instruments should be amenable to direct display and/or data processing and storage. Since the aerosol assessment system is to be furnished as a tested, prototype assembly at the end of a three-year period, the contract framework prohibits any extensive exploration of new or novel instrumental concepts. Therefore, we propose to select commercially available equipment or well-advanced experimental devices as the basic system components and to modify and calibrate these components in accordance with specific system needs. This approach implies that the extent to which ultimate program objectives may be realized is closely related to the present state-of-the-art of aerosol assessment. Thus, the thoroughness of the information survey, i.e., general literature search, review of DOD and contractual research programs and contact with instrument manufacturers, is of particular importance to this program.

II. PROGRAM REVIEW

During the third quarter of this program, several phases of the proposed experiment schedule outlined in our Second Quarterly Report (Ref. 2) were started. Our activities were directed first toward completing the 1-m^3 test chamber facility as well as the aerosol assessment instrumentation considered as tentative system components. Following the above operations, various test programs were begun to establish stirring conditions for optimum chamber mixing and to compare performance of different aerosol assessment techniques.

In the following sections of this report, details are furnished on (a) the structural and functional aspects of the test chamber, (b) the design and performance of the filter sampling system, (c) methods of test aerosol generation, (d) stirring fan configurations and operating characteristics, (e) calibration and performance of the GCA "in-line" cascade impactor, (f) preliminary cloud assessment by a light scattering particle counter with a special sampling system for chamber applications, and (g) reconstruction of particle size distributions of chamber aerosols by mathematical inversion of mass decay curves.

In addition to the above activities, review and updating of the technical literature were continued with particular emphasis on analytical methods for chemical agents and methods of sampling. According to appraisals of commercial apparatus, it appeared that one type of automatic analyzer would fulfill most of the analytical needs of the system provided that modifications suggested by our analytical chemist could be incorporated in the existing design. Tests were performed in the laboratory relating to modifications and improvements in existing methods for chemical agent detection.

Samples of a test liquid, "Bis," were sent to an instrument manufacturer to determine the feasibility and probable sensitivity of flame photometers as system components.

Tentative arrangements were made to schedule delivery of automatic analyzing equipment to GCA laboratories during August, 1966. Prior to delivery, plans were also made for our analytical chemist to attend an intensive training course in the use of this equipment.

Techniques for determining mass concentration by automatic gravimetric balances and by β-ray penetration of solid or liquid agents deposited on filter surfaces were investigated during this period. No existing commercial apparatus relying upon gravimetric response could meet the sensitivity requirements for the present system. However, a continuous tape sampler designed for sampling solid particles in industrial stacks (based on β penetration measurements) appeared promising (Ref. 3). Arrangements have been made to conduct joint tests with staff members of the Harvard School of Public Healts to study this technique.

III. EXPERIMENTAL TEST FACILITY

A. ONE CUBIC METER TEST CHAMBER AND RELATED APPARATUS

Assessment of chemical agent dissemination techniques at the GCA laboratories is usually performed in a special test chamber consisting of a concrete reinforced steel cylinder, 24 feet high and 20 feet in diameter, with adjoining glove box, control room, decontamination area, and analytical laboratory facilities. The design features of this chamber, however, did not provide the flexibility required to meet certain of the experimental needs of the current program because of its size, the time required to clean interior surfaces, and the extended decay periods for fine particles.

It was decided, therefore, to use a smaller chamber (1 m^3) for testing of instrumentation under conditions where experiments could be performed rapidly and with simpler manipulations.

The chamber, Figures 1 and 2, is constructed of 3/8-inch molded polyethylene which is spray coated with a colloidal dispersion of "DAG."* This treatment simulates a normal steel design which permits grounding of the chamber and thereby minimizes static charging of the walls. The chamber height and diameter are 115 cm and 106 cm respectively. It is equipped with a removable cover which slightly overlaps the tank walls and is sealed by use of a large, stainless-steel worm gear clamp.

Three, 2-inch sampling ports are placed in the front wall of the chamber at heights of 17.8 cm, 57.5 cm, and 97.2 cm from the bottom. These ports perform a dual function in that they are used for aerosol dissemination as well as sampling ports. Three additional ports, each separated by 90 degrees, are located in the chamber at the 57.5 cm level: a 3-inch port for venting purposes, a 2-inch port with a glass window for inside viewing, and a 2-inch illumination port.

The cover of the chamber is equipped with a pneumatic spray nozzle for washing the chamber when it is desirous to leave the chamber closed. Three mixing fans which may be adjusted externally to any desired height by means of a rod and packing gland are located symmetrically at radial distances of 40 cm from chamber centerline. They may also be rotated on axis to any desired angle by a swivel joint which connects to the supporting rod.

The light scattering particle counter is coupled directly to a boundary layer diluter which is inserted into the chamber bottom in a vertical position 38.5 cm from centerline. The diluter which connects to the cascade impactor is installed in the same manner. The tank bottom also contains a center drain to remove liquid washings.

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*"DAG" dispersion No. 41, Acheson Colloids Co., Fort Huron, Michigan.

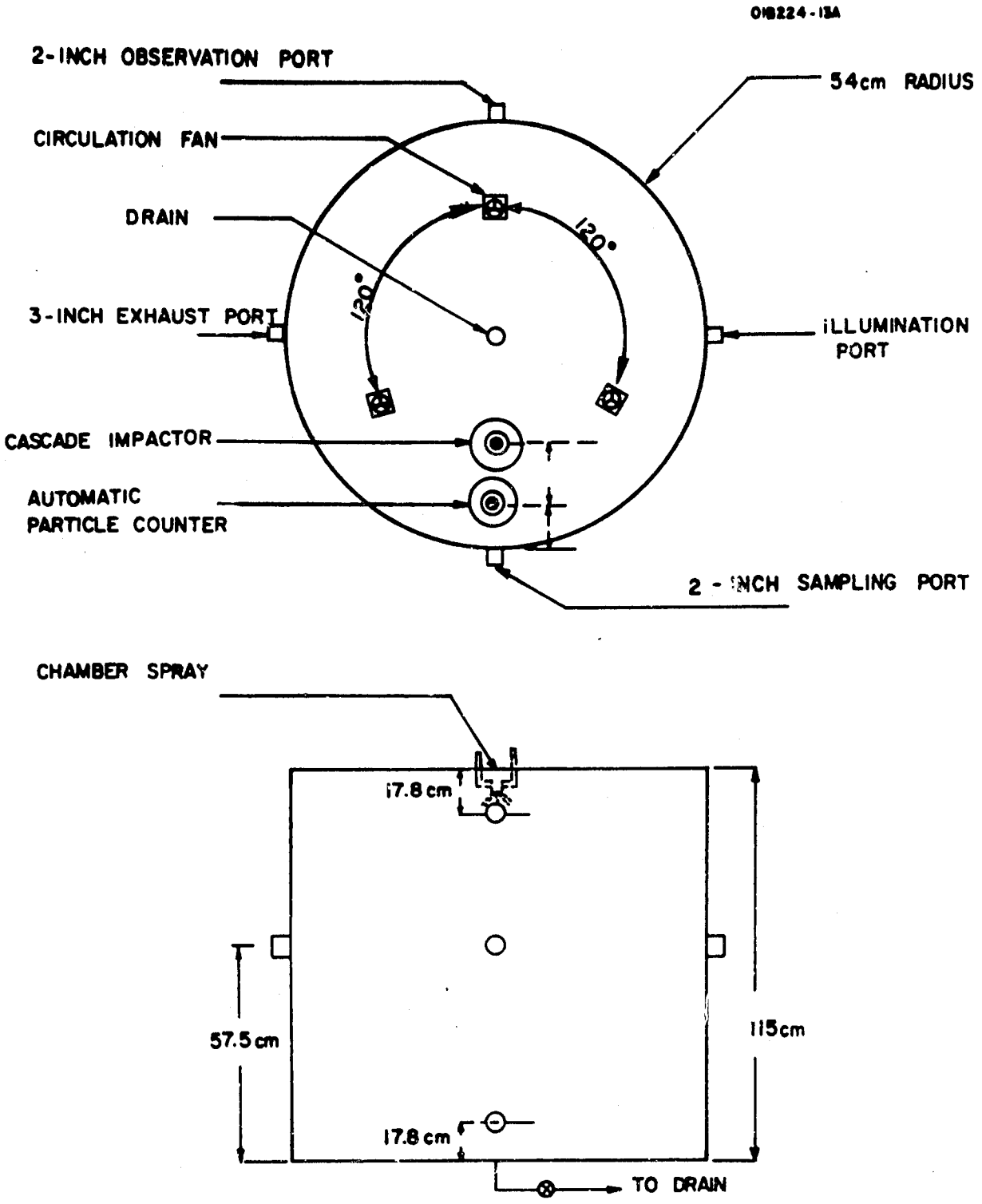

Figure 1. Schematic - cubic meter chamber.

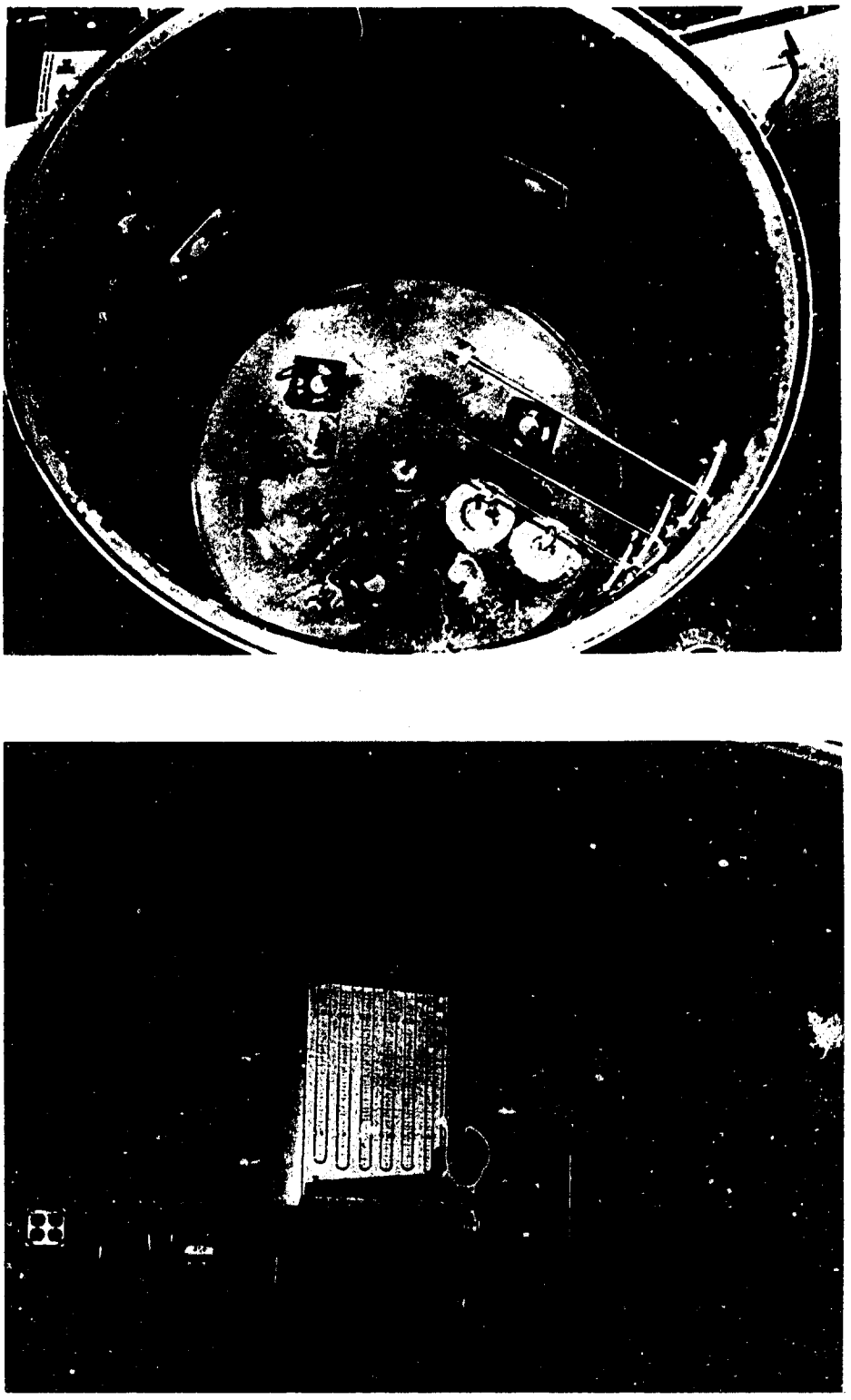

Figure 2. Exterior and interior views of one cubic meter chamber.

The chamber itself is supported on a wooden platform approximately 3 feet above floor level to facilitate sampling and maintenance operations.

B. FILTER SAMPLING SYSTEMS

1. Basic Design

Three probes (lances) were constructed for collection of filter samples of test aerosols within the 1 m^3 chamber, Figure 3. To the end of each 3-foot long, 3/8-inch diameter lance a standard taper section was attached which mated with a disposable Millipore filter holder designed for 3.7 cm diameter filter circles. The filter samplers were manifolded to a single suction pump. Individual or multiple sampler operation could be obtained by means of fast response solenoid valves, Figure 4. A limiting orifice in each sampling line maintained a constant sampling rate of 8.9 ℓ/min.

Individual filter assemblies were leak tested and calibrated prior to use. The above equipment was designed to permit rapid insertion and removal of filter probes so that dilution of chamber aerosols would be negligible during change periods.

Filter papers (glass or cellulose media) were sealed between machined plastic bearing surfaces by compressing in a vice since the plastic holders were used for one test only.

The filter assemblies were inserted through the 2-inch sampling ports described previously. Adjustable rubber sleeves permitted flexibility in sampler positioning and eliminated air leakage at the port locations.

A bleed line was placed in the manifold so that the pump could be turned on prior to sampling. This practice eliminates the initial flow surge associated with pump startup which can lead to serious flow errors during brief (0.1 minute) sampling periods.

It is emphasized that these samplers were built mainly to facilitate evaluation of system components and are not necessarily considered as prototype units for final designs.

2. Filter Shielding

A major problem in sampling chamber aerosols has been the shielding of filters from extraneous splatter during the dissemination period. To overcome this difficulty a filter holder was designed with an iris type shutter which can be opened and closed from outside the chamber. This enables the investigator to position the sampling filter before the test begins and thereby obtain a sample at an earlier time than would be otherwise possible. It also prevents leakage of the chamber

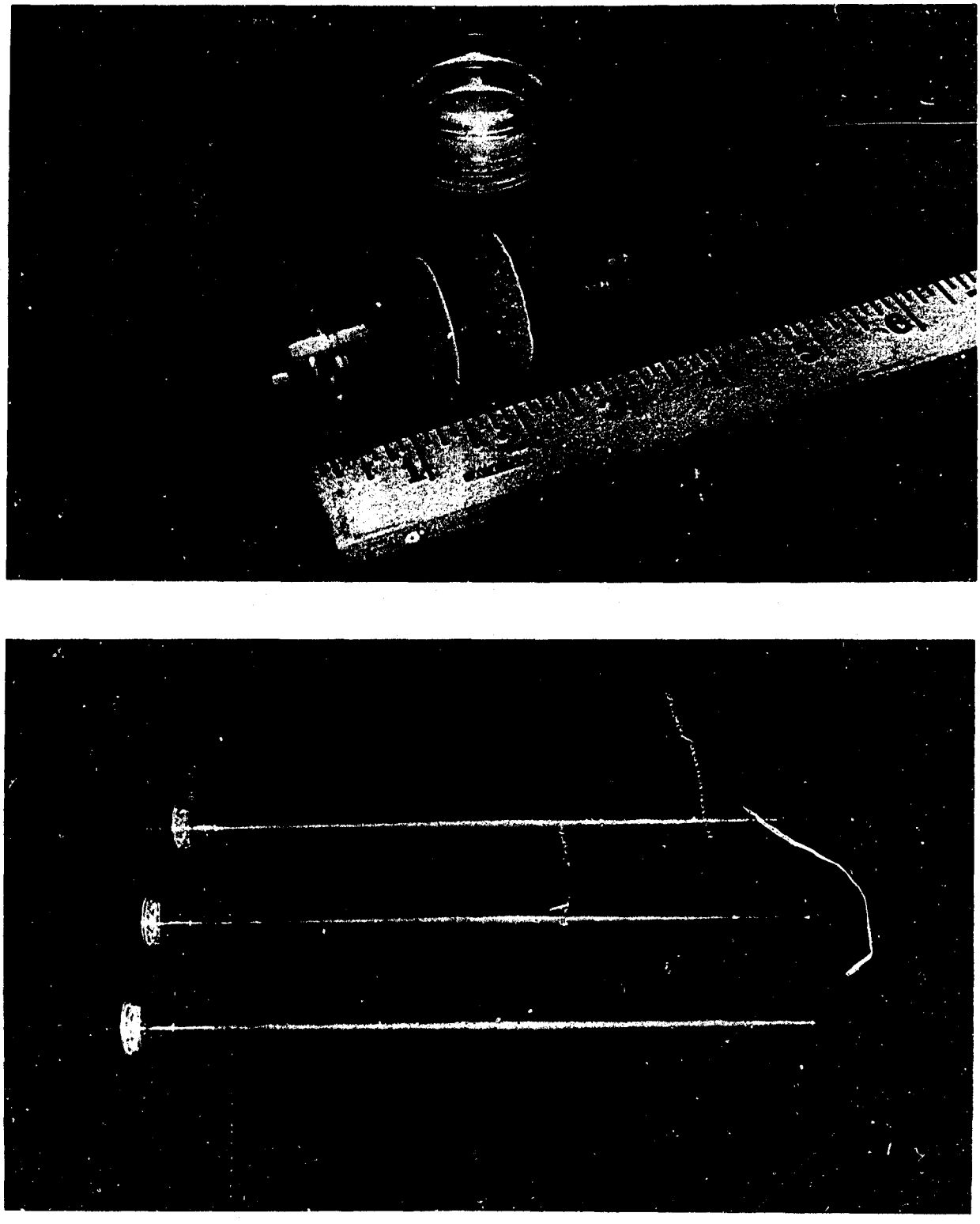

Figure 3. Sampling lance (probe) and filter holder (exploded view).

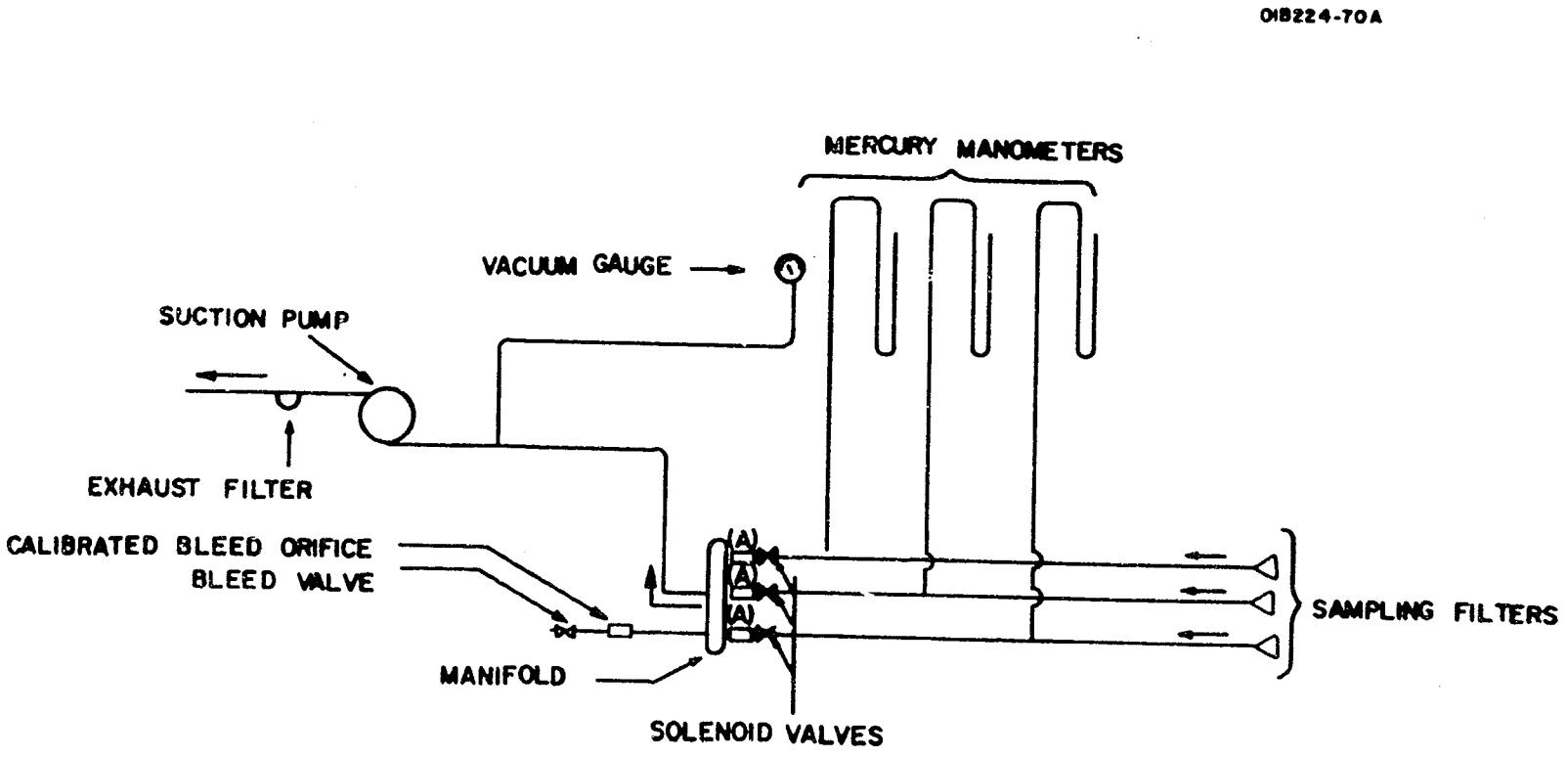

Figure 4. Schematic - aerosol sampling system.

aerosol when the sampling port is opened for probe insertion. Such loss may be significant when particle concentrations are high and the chamber mixing process is incomplete. Shielding of this type may be very important when, for example, the handling of toxic substances requires that no ports be opened following aerosol generation.

3. High Velocity Filter Samplers

A second special filter holder was constructed in which the diameter of the retained filter circle was reduced to 1.0 cm as compared to 3.7 cm for the regular samplers. The function of this design, which is intended to sample at 10 l/min, is to increase the sampling velocity to 1000 ft/min in contrast to 73 ft/min for the 3.7 cm diameter circles. Under these conditions, it is expected that the inertial loss of large particles prior to entering the sampler will be reduced (Ref. 4). It is planned to compare the results of low and high velocity sampling to assess the extent of such sampling errors. A critical factor in the design of this unit was the fabrication of a dependable filter gasketing and support system since the pressure loss is high for high velocity filtration processes (Ref. 5). Under these conditions, control of leakage may be difficult.

4. Sampling Media

Various types of filter media have been used for sampling test aerosols in the 1 m^3 chamber. These included all-glass paper (fired and unfired), membrane filters, and high efficiency cellulose filters.

All-glass filters are generally satisfactory when sampling high concentrations for gravimetric analyses provided that variations in moisture content are small in comparison to the weight of collected material. Gravimetric procedures, however, are applicable only when no significant cloud decay occurs over the period required to collect the sample and the cloud is composed of undegraded chemical agent. One minute sampling periods are suitable in a 24-foot-high chamber but not acceptable in a 4-foot chamber where the particle decay rate is six times as rapid.

When chemical or physical-chemical methods are used to provide both agent specificity and high sensitivity, it is necessary to ascertain what contamination, if any, is attributable to the filter media.

5. Filter Media Evaluation

Comparisons were made between two types of all-glass media commonly used for aerosol collection to determine the hygroscopic properties of fired and unfired filters. The firing process consisted of placing filters in a muffle furnace for a few minutes at 1500°F to burn

out the organic binder. Although this treatment is essential when filters are to be analyzed by chemical methods, it was decided to determine if firing could be avoided when filters were to be analyzed gravimetrically. Unfortunately, the firing process reduces the strength of the media, thus necessitating extreme care in handling. Repetitive weighings were made on two each of two commercially available filter types under fired and unfired conditions. During a two-week check period, relative humidities in the weighing atmosphere varied from 14 to 44 percent at ambient temperature levels of 70° to 75°F. Test results were inconclusive as far as showing any positive relationship between humidity and weight gain.

Tests summarized in Table 1 (performed with 5.0 cm diameter circles) indicated that filter weight variations in ambient atmospheres were greater with Type 1* media. Furthermore, the firing process appeared to reduce significantly the weight variations attributable in part to moisture absorption and desorption. On the other hand, firing of Type 2** media made little change in the observed weight variation although the absolute changes were less than noted for Type 1 media. Since a poor correlation was observed between relative humidity and filter weight, it is obvious that factors other than moisture absorption contribute to weight fluctuations, e.g., room temperature gradients, vibrations, electrostatic effects, and human error. From a practical viewpoint, however, the above data suggest the expected weighing precision that can be obtained with routine laboratory procedures.

A spectrophotometric technique was used to determine the amount of saccharin collected on various filter media. It was found that methanol could not be used to remove the treated saccharin*** from fired or unfired all-glass filters without first thoroughly pre-washing the media in methanol. Unfortunately, treated saccharin does not dissolve completely in water, which would eliminate the filter pre-wash step. (Water does not extract any noticable impurities from the filter.) Failure to pre-wash led to extraction of interfering substances from the filter itself which absorbed in the saccharin waveband. Cellulose media (acid washed, analytical grade) also required washing but to a lesser degree than that for the glass filters. Since membrane filters dissolve in methanol yielding interference products in the working waveband, they were not considered acceptable for this application.

Although the cellulose media represented the optimum choice, it was found that the methanol-washed glass paper could be used for most analyses. The cellulose paper presents the disadvantage of high resistance to air flow which requires that flow corrections be made in the sampling system for reduced gas density downstream of the media.

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*Type E glass filter, Gelman Instrument Co.

**1106B filter paper, Mine Safety Appliance Co.

***Treated with 3 percent Cab-O-Sil and 1 percent hexamethyldisilizane.

TABLE 1

WEIGHT VARIATIONS FOR FIRED AND UNFIRED ALL-GLASS FILTERS IN AMBIENT ATMOSPHERES

	Type 1		Type 2	
	Unfired	Fired	Unfired	Fired
Average* Weight (mg)	165.6	159.6	112.1	92.4
Percent Binder	3.6	-----	8.6	----
Average Deviation (mg)	1.1	0.90	0.73	0.70
Percent Deviation	0.66	0.56	0.65	0.76

*Filter Circle Diameter = 5.0 cm

Final liquid filtration of methanol sample extracts were made with prewashed Whatman paper to prevent particulate contamination from micron-sized glass fibers.

C. AEROSOL GENERATION

Several methods of aerosol generation were considered for evaluation of sampling systems tested in the 1 M^3 chamber. Regardless of the dispersion technique, it was necessary that equipment be provided to generate both liquid droplets and solid particles over the approximate size range 0.5 to 100 microns. Specific solids and liquids were also required which would simulate the physical properties of typical agent cloud components, with particular emphasis on refractive index, mass and bulk density and aerodynamic behavior. Where possible, the use of specifically prepared mono- or near monodisperse particles was preferred for calibration of instruments which are sensitive to particle geometry, e.g., light scattering particle counters or inertial fractionators such as the cascade impactor.

Two aerosol generator types were considered, the first based upon continuous operation with provision to bypass material into the chamber at predesignated intervals and the second designed as a "single shot" pulse type unit. The former system, although more complex in design, furnishes better reproducibility in determining total concentration and particle size properties at time zero. The pulse type generator, however, is much simpler to operate and is entirely suitable for estimating cloud decay properties for sampling periods starting one to two minutes after cloud formation.

1. Dry Power Ejector

The generator used for solids dissemination was constructed to provide a simple method for rapid dispersion of dry powders. Dispersion is attained by directing short pulses of cleaned and dried compressed air at 50 psig against 0.5 to 1 gram quantities of powder held in a small test tube. The tube is clamped in a metal holder attached to an extension tube, the latter also functioning as the air supply line. Construction details are shown in Figure 5, including the positioning of the 1/8-inch air injection line which extends 1/2-inch into the dust holding tube. Since the extension tube is 3-feet long, it is possible to disseminate the solids at several points within the test chamber. The generator does an effective job in removing the powder from the test tube. However, some of the powder does not become airborne and falls to the chamber floor; hence, 50 $\pm$3 percent aerosolization of particulate matter is about average for this device. Since experiments performed to date were intended to test the effectiveness of chamber mixing systems and the comparative efficiencies of various sampling devices, it was not practicable to use a more sophisticated generator.

2. Plastic Sphere Disseminator

A special generator was used to disseminate a liquid suspension of monodisperse 1.30 micron diameter polyvinyl latex spheres for calibration of the light scattering particle counter, Figure 6. The device consists of a Pen-i-sol nebulizer,* a glass bubble neck drying tube, and meters to regulate nebulizer and dilution air flow. Clean dry air from a single source is split to provide nebulizing air for the generator and dilution and drying air for the aerosol formed in the nebulizing chamber. The diluted aerosol passes through the bubble column which acts as a dryer, and the effluent stream is run directly into the counter inlet. The resultant aerosol concentration is reasonably constant and contains few agglomerates.

A similar device, which was designed and constructed at the Harvard School of Public Health, Department of Industrial Hygiene, was also tested with the light scattering counter. The Harvard unit is different from our generator in that special air flow controls were used to regulate particle concentration for a given liquid suspension.

3. Explosive Dissemination

Several preliminary tests were made to determine whether any type of explosive dissemination was feasible in the 1 m^3 test chamber. A 3-1/2-inch diameter by 3-inch steel cyclinder, Figure 7, in which a

*J.E. Emerson Co., Cambridge, Massachusetts.

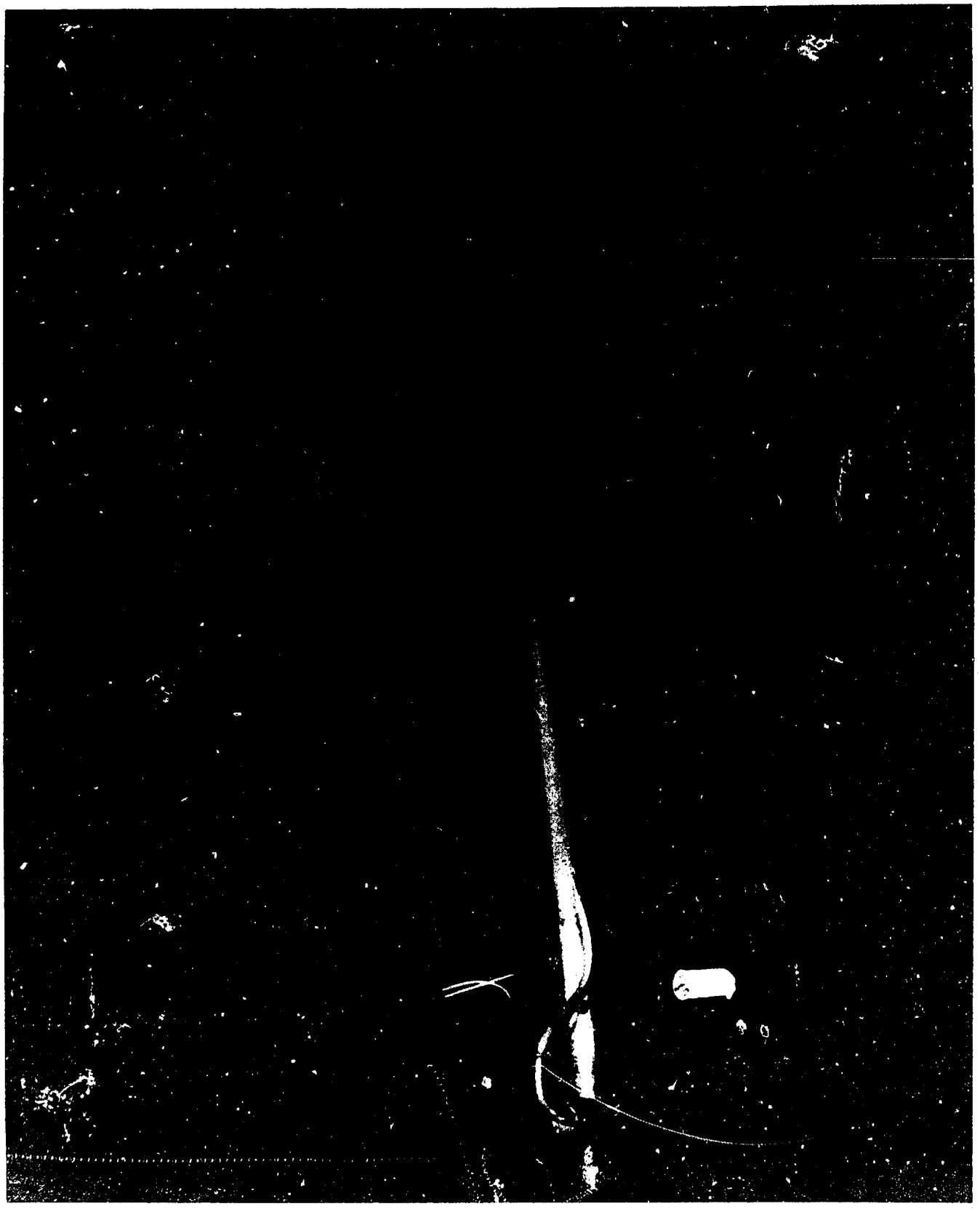


Figure 5. Dry powder ejector.

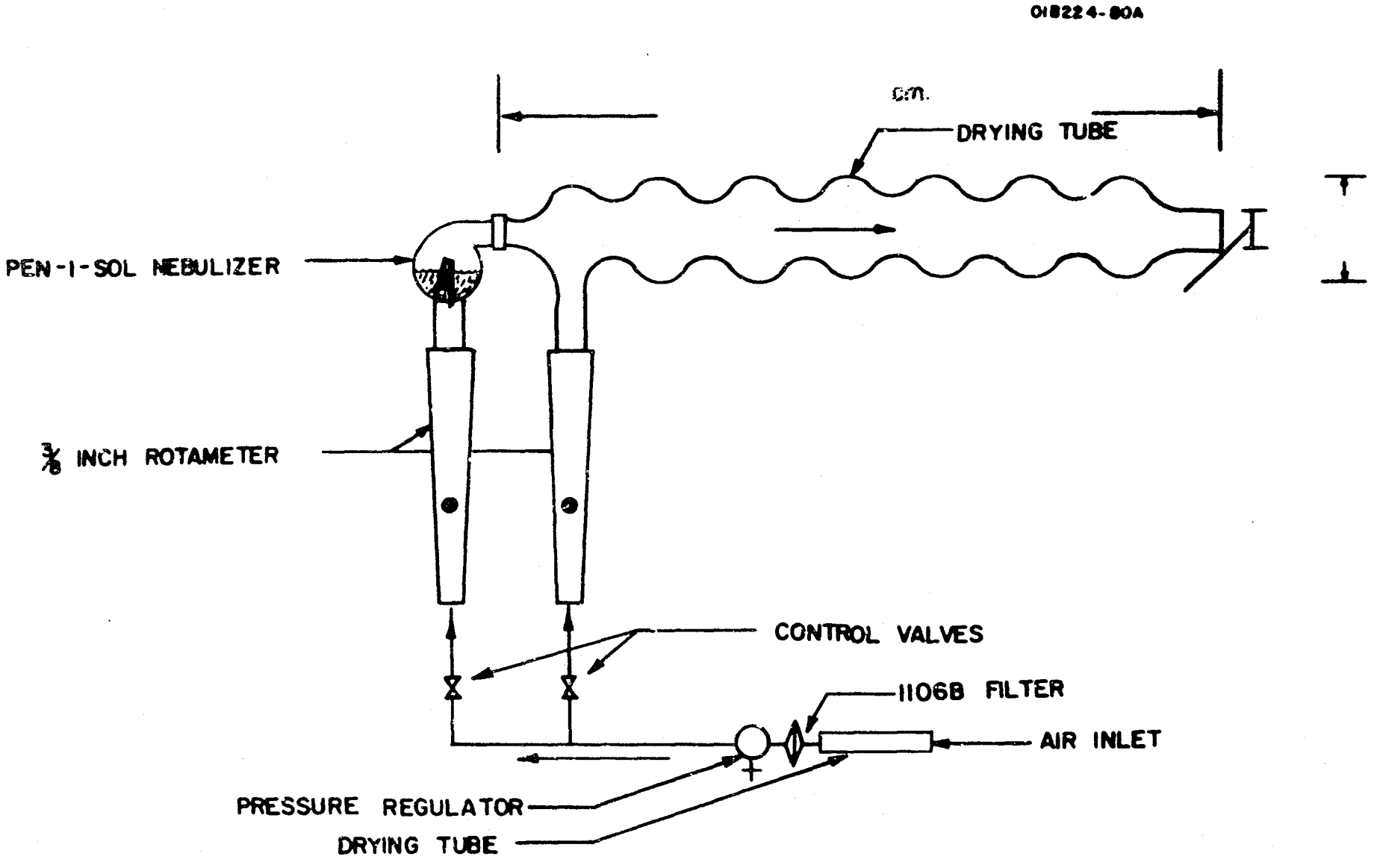
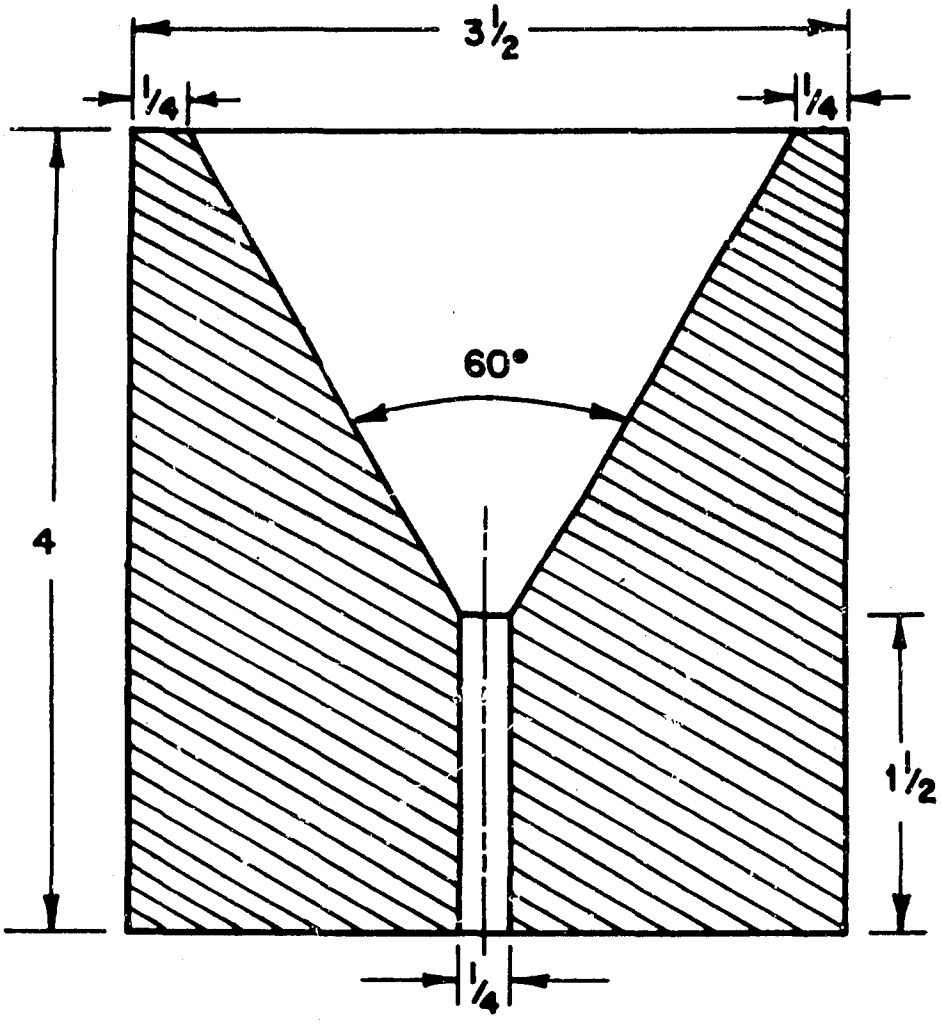


Figure 6. Schematic - plastic sphere generator.



Full Scale
Dimensions in Inches

Figure 7. Cylindrical explosive powder disseminator.

conical depression was made, was placed in a 5-gallon covered plastic container. An E-94 detonating cap was inserted at the apex of the conical section and covered with 1 gm of treated saccharin. The cap generated sufficient energy to disperse all of the saccharin without any sign of gross fallout and without any damage to the container. Although no quantitative measurements were made to assess the aerosol cloud, the initial tests showed that this method was suitable and safe for explosive generation of dry powders in our 1 m^3 chamber. The aerosol cloud produced under the above circumstances depicts the typical multi-component cloud generated by explosive devices. Size analyses of such clouds cannot be performed with light scattering detectors currently available.

4. Generation by Evaporation-Condensation

A versatile, all-glass aerosol generator, Figure 8, utilizing evaporation and recondensation to provide nearly homogeneous, micron and submicron aerosols was constructed on the basis of a design described by Rappaport and Weinstock (Ref. 6). This device uses a nebulizer for formation of the primary aerosol which is subsequently evaporated on passing through a heated tube. Recondensation of the vapor produces a homogeneous aerosol. This procedure eliminates the need for prolonged heating of the substance in bulk and provides high mass concentrations (limited volumes) of aerosolized material. It can function without the addition of condensation nuclei to produce aerosols greater than 0.5 micron diameter, and may be used to produce aerosols of alkyl phthalates, stearic acid, oleic acid, menthol, tri-cresyl phosphate, etc.

D. CHAMBER STIRRING SYSTEMS

1. Basic Considerations

In a previous report (Ref. 2), the importance of maintaining stirred settling conditions within aerosol test chambers has been discussed. It was pointed out that realistic interpretation of mass decay curves for particle size assessment depends upon concentration homogeneity within the chamber at any instant. Under these circumstances, one should expect to find identical mass concentrations and particle size distributions regardless of sampling location, provided that samples are collected over the same time intervals.

Basic concepts relating to mixing procedures which appear to be generally accepted are as follows:

(1) symmetrical location of fans
(2) low fan speeds to reduce particle collection on impellers
(3) fans located near bottom of chamber and discharging upwards.

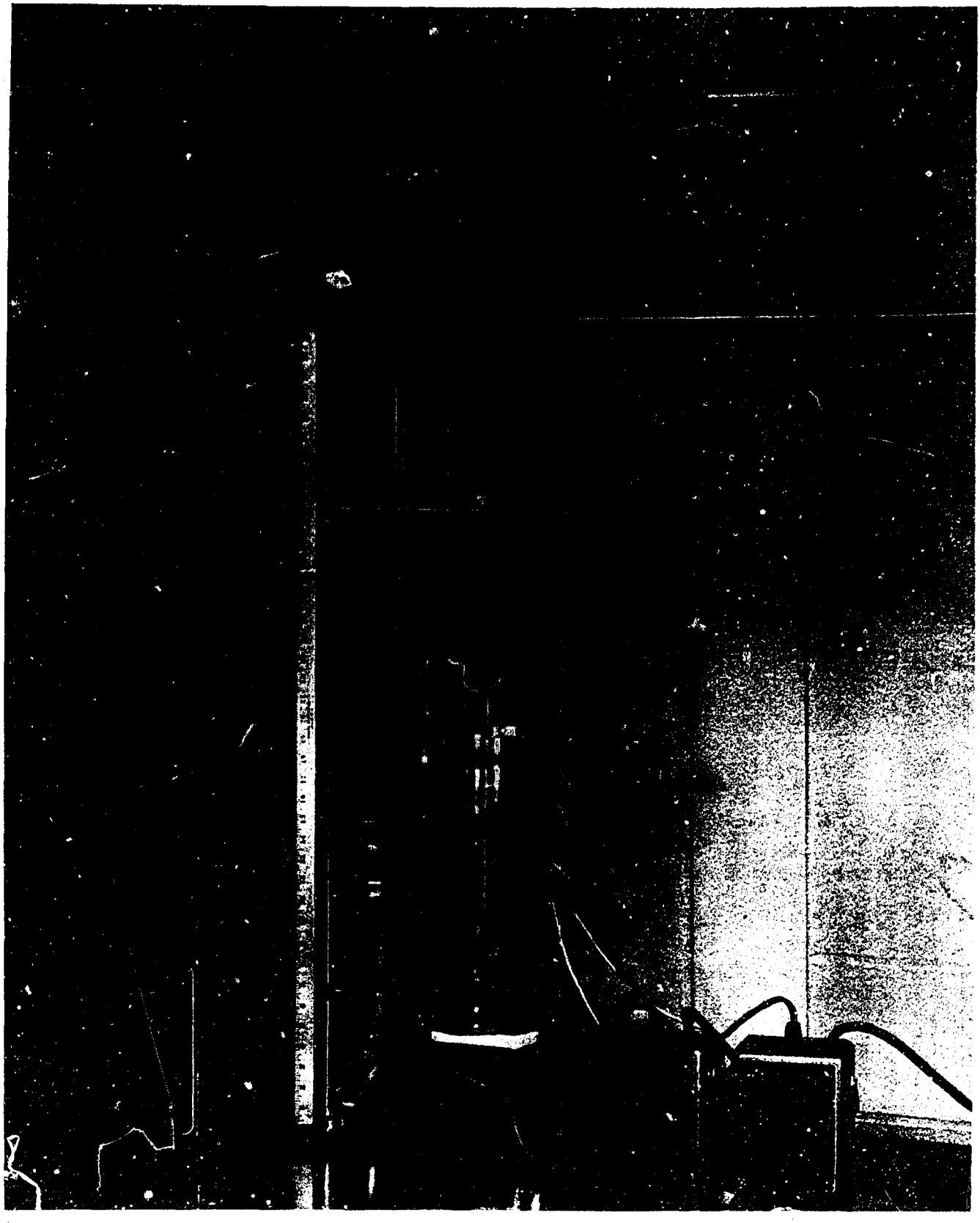


Figure 8. Monodisperse aerosol generator.

Reference to the literature indicated no proven methods for guaranteeing thorough mixing in chambers, particularly for a broad range of particle sizes. From a theoretical viewpoint, it is apparent that what constitutes an optimum mixing system must be a compromise between diametrically opposed effects induced by the stirring process. For example, the more intense stirring required to maintain a uniform suspension of coarse particles may increase the coagulation rate, cause excessive particle impingement upon the blades and scroll of the mixing fan, and lead to increased diffusion of the fine particle fractions to chamber walls. The above effects, if sufficiently large, may rule out any useful interpretation of mass decay curves.

The ideal mixing system would appear to be an array of several small fans, (turbulence generators) uniformly positioned throughout a test chamber. Such an arrangement would circumvent the current problem of excessive energy levels in the vicinity of a single fan accompanied by a gradual reduction in circulation rate and turbulence in the remote regions of the chamber, the latter effect leading to incomplete mixing.

For practical reasons, i.e., operational simplicity and minimal interference with test devices, sampling equipment,and decontamination apparatus, no more than two fans are used in most test chambers. These have been located in peripheral locations usually at the top or bottom of the chambers with some attempt at symmetrical placement.

2. Theoretical Considerations

It appears that the location of the fan(s) at the bottom of the chamber should be preferable to a top mounting. Both systems may interfere with normal testing or decontamination processes, e.g., collection of floor samples with settling trays or washing the chamber walls with a spray nozzle array located at the top of the chamber. The qualitative aspects of top and bottom mixing can be described by the following simplified models in which it is assumed that (a) the top half of the chamber is completely stirred while the lower half remains as a stagnant atmosphere and (b) the reverse of situation (a). In both cases, it is assumed that the aerosol is monodisperse and completely mixed at time zero.

A comparison is made first between the total mass (or number of particles) which reach the floor of the chamber during the time, t_1, required for a single particle to settle a distance one half the chamber height, h', under tranquil settling conditions at a terminal velocity, V_T. The fractional depletion from the upper half is represented by the relation

$$1 - C_u/C_o = 1 - \exp\left(-\frac{V_t\ t_1}{h'}\right) = 1 - \exp(-1) = 0.63$$

where $t_1 = h_1/V_T$

Particles which settle from the upper stirred section enter a tranquil settling region in the lower half of the chamber. Hence, no particles arriving from the upper section during the time interval, t_1, will have left the lower half of the chamber. Rather, they will be distributed according to an exponential concentration gradient with the maximum concentration immediately above the bottom of the chamber. Concurrently, all particles originally in the lower half of the chamber will have settled out (reached the bottom surface of the chamber) since the settling period t_1 is defined by h'/V_T. The net result, therefore, is that with top stirring 50 percent of the initial number of particles within the chamber is removed during the interval t_1.

If the mixing fan is placed in the lower half of the chamber, the particle concentration, C_ℓ, in this section may be treated in two fractions, the first representing settlement from the upper half and arriving at a constant rate over the time interval, t_1, and the second representing loss of particles from the lower half under stirred settling conditions, i.e.,

$$C_\ell = C_o\left[1 - \exp\left(-\frac{V_T\ t_1}{h'}\right) + \exp\left(-\frac{V_T\ t_1}{h'}\right)\right]$$

According to the above equation, no change in particle concentration takes place in the lower half of the chamber over the time interval, t_1. Since the upper half is void of particles at t_1 the net loss of material from the entire chamber is 50 percent, the same value predicted for top mixing.

Based upon the above analysis one can argue that bottom mixing offers no particular advantage and that the final selection of fan site would depend upon the location of other chamber accessories. It is apparent, however, that if sampling port(s) were located in the lower section of a chamber (a more convenient station with existing installations) the measured concentrations would be consistently higher than those observed under ideal stirred settling until the elapsed time corresponded to that required for the indicated particle size to settle through the complete height of the chamber, Figure 9 - Curves C and S. Subsequently, decay rates exceed the theoretical values for perfect mixing.

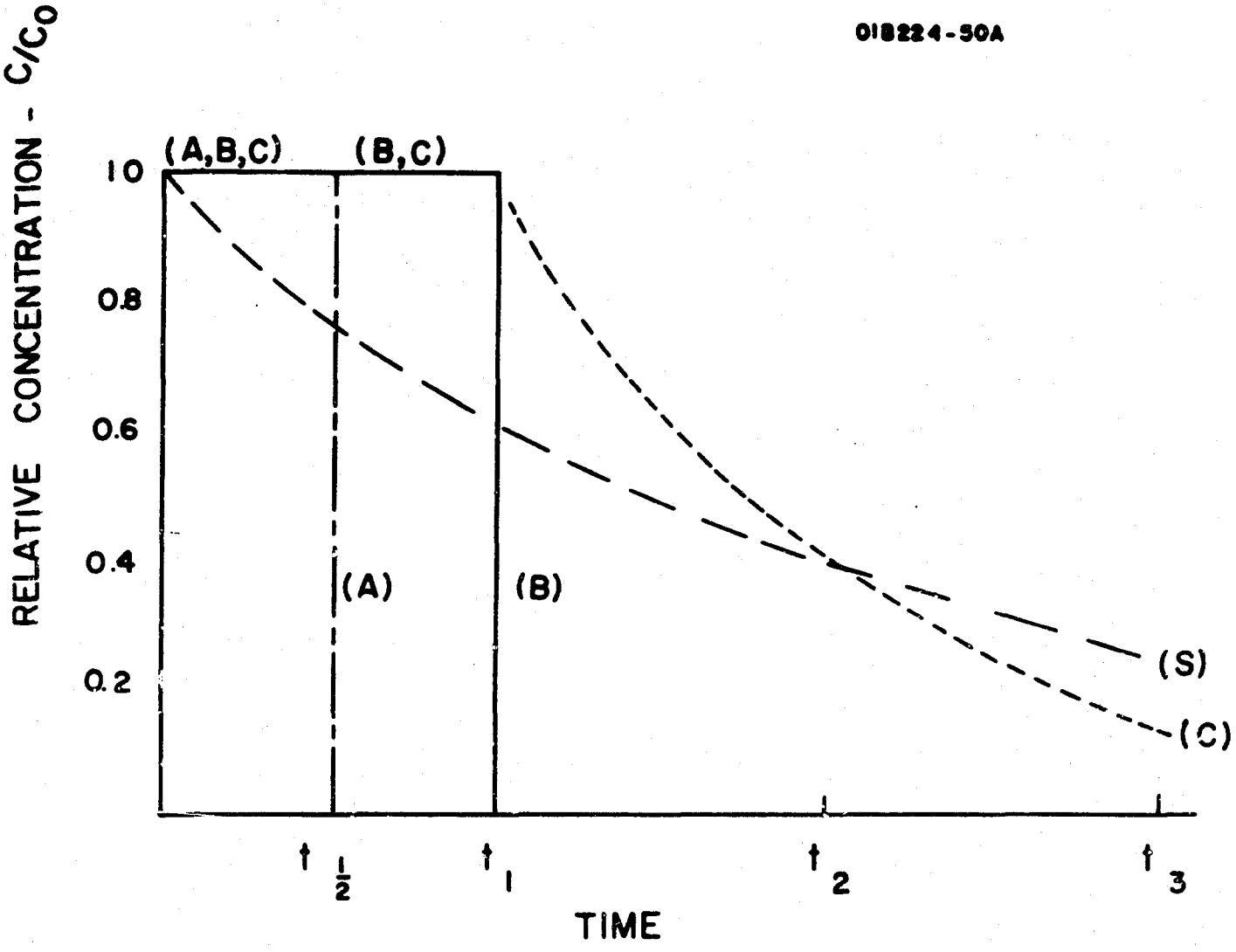


Figure 9. Theoretical fractional concentrations with bottom mixing versus decay time and sampling location.

On the other hand, use of a top mixing system, Figure 10 - curves A and S, would lead to consistently low results relative to ideal stirred settling for all measurements made above the chamber midpoint. Sampling below the chamber midpoint indicates variable results depending upon the location of ports, Figure 10 - curves A,B,C, e.g., the nearer the sampling port to the bottom of the chamber the longer the period of constant concentration before any mass decay is evidenced.

Although one would never expect the extremes in settling conditions predicted by these simplified models, the indicated trends serve as a guide for positioning sampling probes.

It should be noted that, in actual practice, single or multiple fan systems mounted either at the top or bottom of a chamber must provide sufficient circulation to mix the aerosol cloud at the opposite end of the chamber. This requirement leads automatically to relatively high velocities at the fan face which subsequently decay to near ambient levels (25 to 75 fpm) at points removed from the fan. The return circulation which flows through a much larger cross sectional area than that of the primary jet moves at a much lower velocity. Consequently, large particles entrained in the primary air flow of a downward directed fan are accelerated toward the bottom of the chamber at a greater rate than that produced by an upward directed fan. Qualitatively, the positive combination of gravity and flow fields should lead to erroneously high settling rates. Use of bottom positioned fans should reduce error from this source.

A series of tests is in progress in which the number of fans, their location and orientation, and volume deliveries are being investigated. During the current periods, three fans were positioned symmetrically 120 degrees apart, each 40 cm from the center of the chamber and located approximately 4 inches above the chamber floor. (See Figure 1.) The fans were simple axial types,* housed within a 4-inch wide, 2-inch deep casing and designed as cooling units for electronic equipment. An adjustable rod which extended through the bottom of the chamber allowed for height selection and a yoke attached to this rod permitted both rotation and inclination of the fan.

3. Test Program

Prior to installation, each fan was checked for blade rpm, volume flow, and velocity profile as a function of input voltage. Flow rate vs voltage data were used subsequently as the basis for calibration curves. Velocity profiles were determined for various volume deliveries to establish (1) the "throw" characteristics of the fans, i.e., the rate of velocity decay in the forward direction, and (2) the divergence of the jet as a function of the distance from the face. It is recognized that the above parameters as determined in free air do not represent fan operation in a confined chamber. These tests, however, characterize

*Rotron Axial Fan 115 CFM Free air, Cramer Electronics.

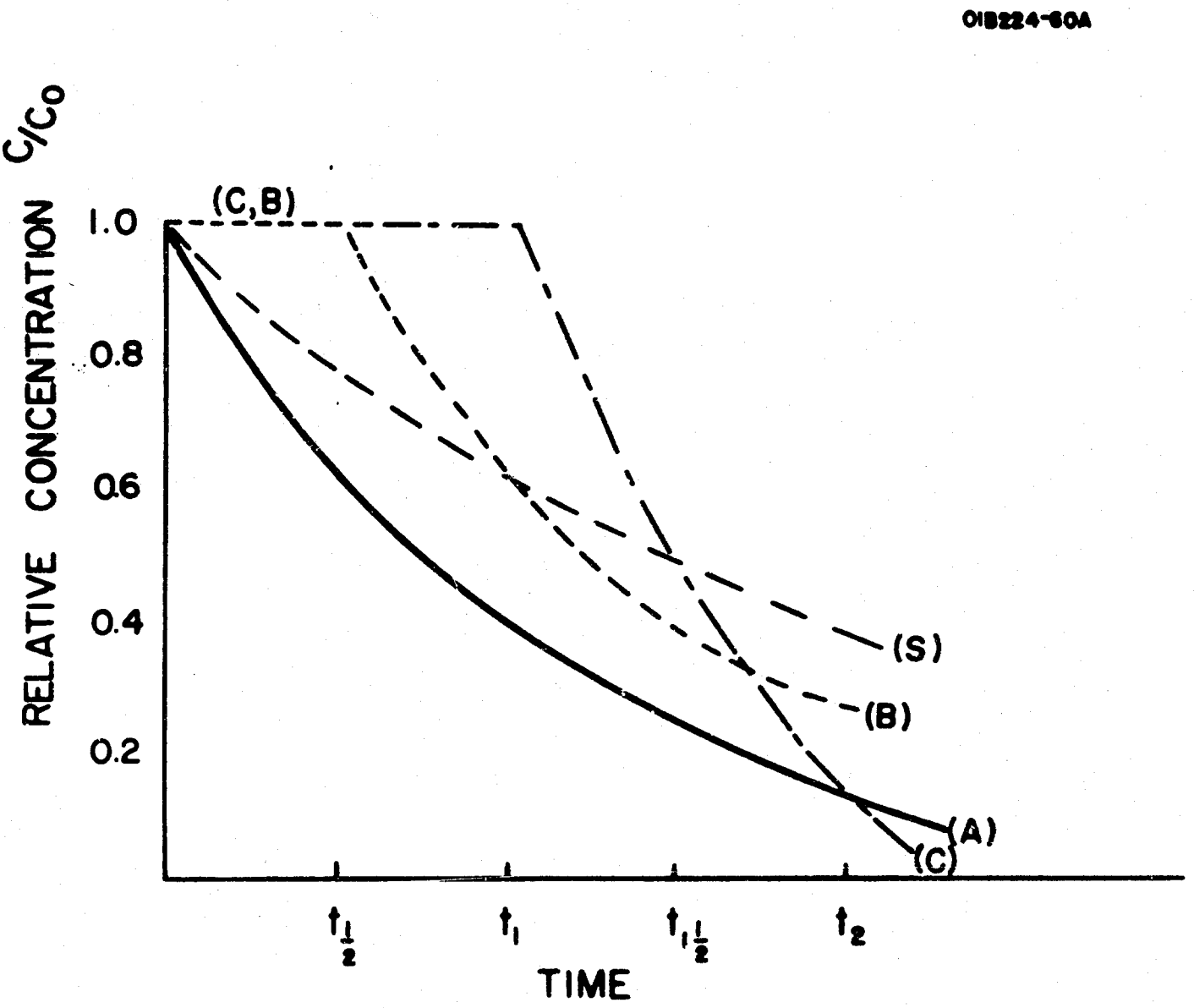


Figure 10. Theoretical fractional concentrations with top mixing versus decay time and sampling location.

fan performance and provide basic information from which stirring effectiveness may be related later to the energy input of the fan system. Figure 11 shows typical velocity profiles as measured 2 inches from the fan face at operating voltages (variac controlled) of 50, 80, and 130 volts. The characteristic dip at centerline is caused by the obstruction of the flow by the hub. In Figure 12, constant velocity contours vs radial position are shown for distances up to 2 feet from fan discharge. The principal application of these data was to determine the divergence and velocity decay of the jet. At a distance of 4 feet from the fan (not shown in Figure 12) air velocities decreased to the order of room air currents, 50 to 75 fpm.

4. Test Procedures

The first experimental fan arrangement had the three fans located at the bottom of the 1 M^3 chamber with their discharges focused at a point one third the distance from the top of the chamber and on the centerline. The rationale behind this system was that the distances from fans to walls were maximized and that a zone of strong turbulence could be generated in the central region of the chamber. Other fan arrangements are proposed for study, including vertically upward discharge (three fans) and single fan operation at the top and bottom of the chamber. In the latter tests, it is planned to compare the effectiveness of a single fan at the top of the chamber (downward flow) with a single fan at the bottom (upward flow).

One gram quantities of a saccharin powder were dispersed by the dry powder ejector described previously (Section III-C). Simultaneous filter samples were collected at three locations: top, center, and bottom of chamber. Filters were positioned in three arrays: (1) all three filters on centerline, (2) top and bottom filters on centerline and middle filter one inch from wall, (3) top and bottom filters one inch from wall and middle filter on centerline.

5. Test Results

Results of comparative concentration measurements under different mixing conditions are shown in Tables 2 and 3. Data reported in Table 2 refer to the simultaneous collection of saccharin particles over one minute periods with the filter probes described in Section III-B. Since fired all-glass circles were used, it was expected that the weighing of milligram quantities would show some variability (see Table 1). Therefore, two to three tests were performed for each of the four fan flows shown in Table 2 to improve the data quality.

Examination of centerline concentrations (expressed in terms of filter weight gain) showed no significant differences with respect to top, middle and bottom sampling stations for any fan flows. Averaging of concentrations for each station without regard to fan flow

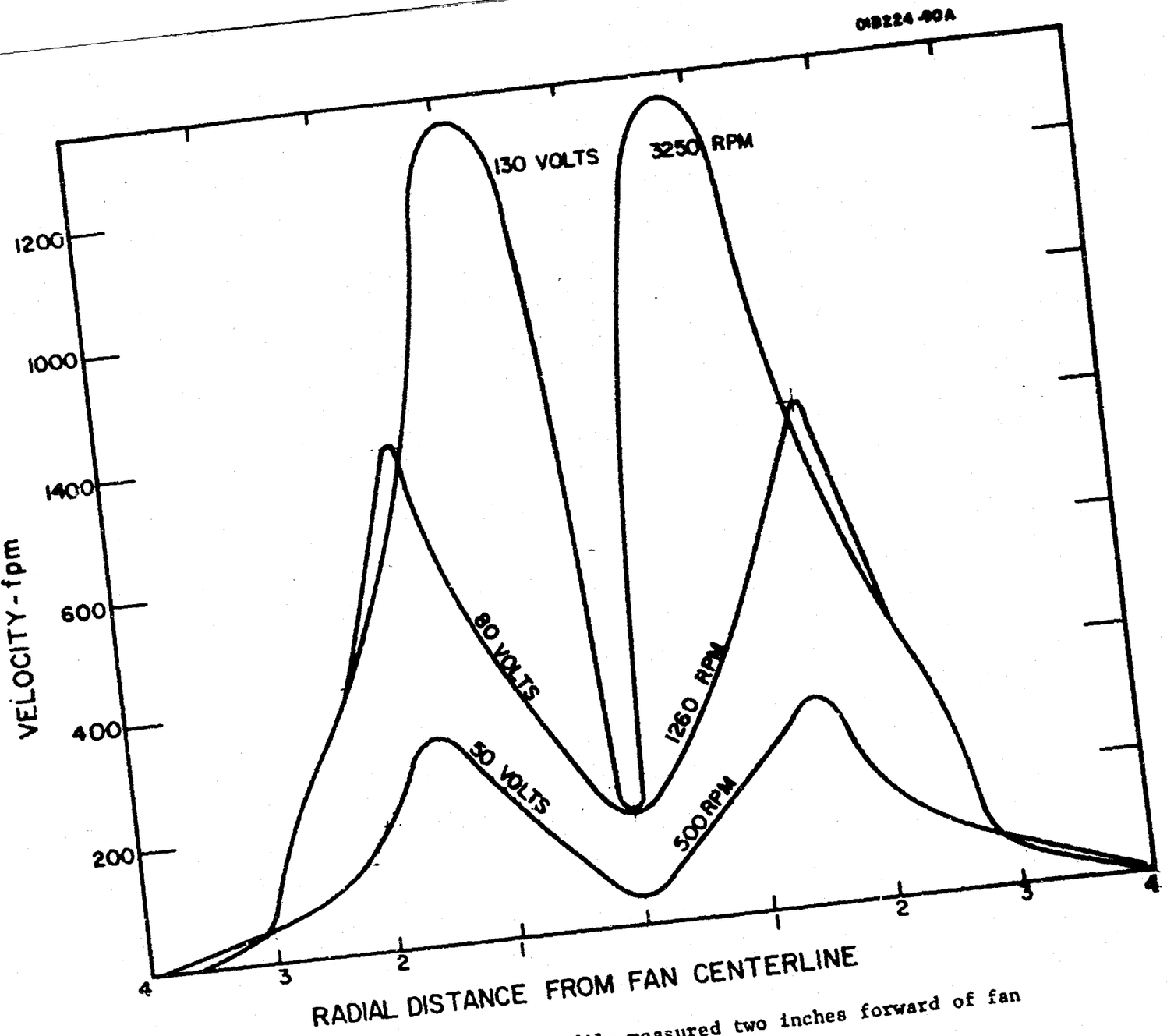


Figure 11. Fan velocity profile measured two inches forward of fan discharge.

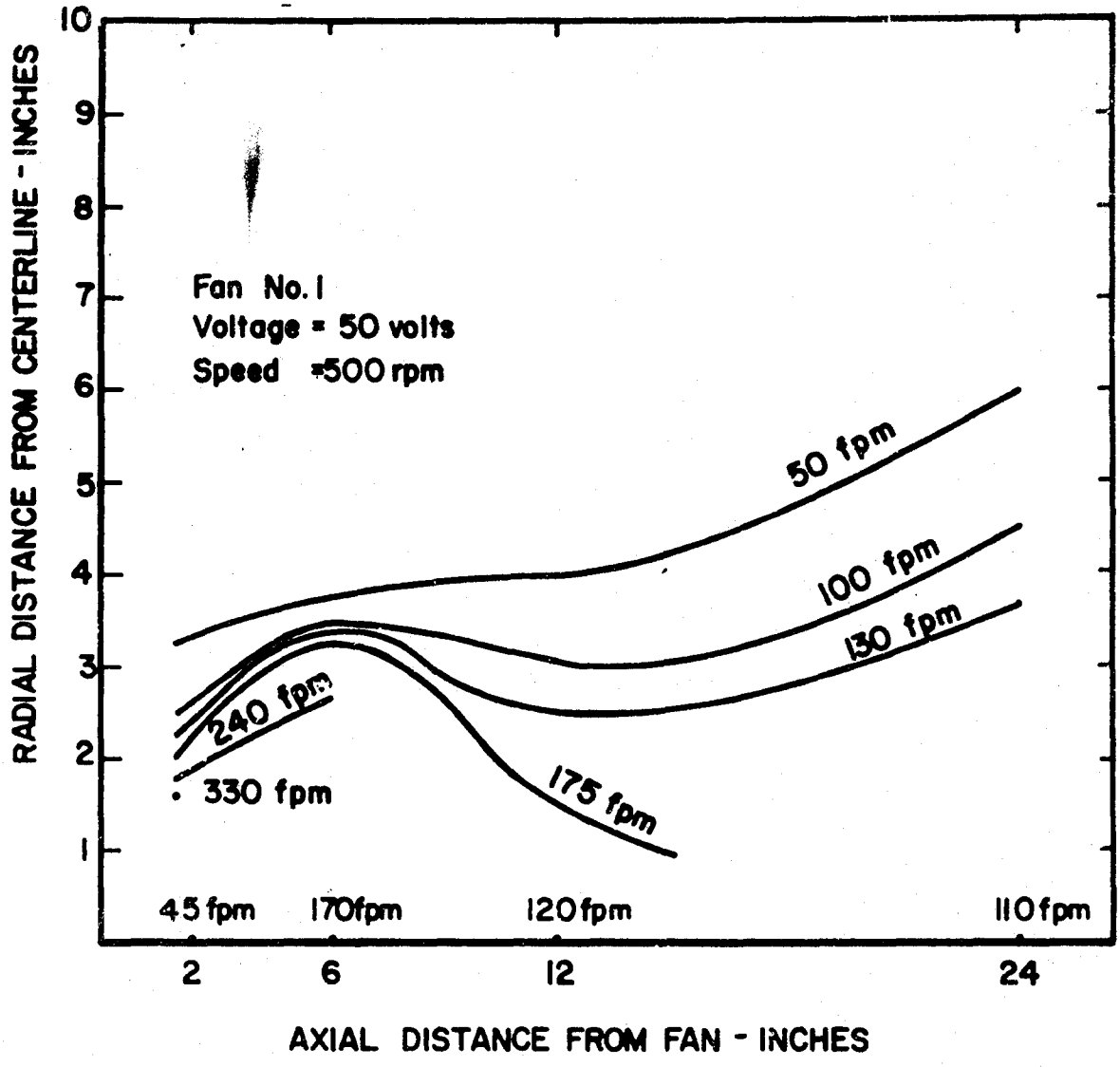


Figure 12. Fan velocity profile at constant voltage lines of constant velocity.

TABLE 2

CHAMBER CONCENTRATIONS AT THREE LOCATIONS WITH SIMULTANEOUS SAMPLING AND FOUR MIXING RATES

Fan Flow[(a)]-cu. ft/min	Filter Weight Gain[(b)] - Milligrams		
	A	B	C
115	2.8	2.9	2.6
95	3.7	3.9	3.7
75	2.7	3.0	3.0
50	2.4	2.3	2.5
Average	2.9	3.0	3.0

(a) Free volume delivery per fan
(b) Weight gain per filter for one minute samples at top (A), middle (B) and bottom (a) centerline of 1 m^3 chamber - gravimetric analysis

Note 1 Saccharin Aerosol, 1 gram dispersions
2 Three fans operating, focussed on centerline, 16 inches from top of chamber
3 Indicated test values are averages of 2-3 tests.

TABLE 3

CHAMBER CONCENTRATIONS AT THREE LOCATIONS WITH SIMULTANEOUS SAMPLING WITH SINGLE FAN OPERATION AT 50 CU FT/MIN

Operating Fan[(a)]	Filter Weight Gain - Milligrams[(b)]			
	A	B	C	Average
No. 1	0.45	0.45	0.43	0.44
No. 2	0.43	0.41	0.45	0.43
No. 3	0.46	0.48	0.44	0.46
Average	0.45	0.45	0.44	

(a) Each fan position the same as in three fan system (Table 2)
(b) Weight gain per filter for 0.1 minute sample at stations A, B and C (See Table 2) - Chemical analyses, spectrophotometric

Notes 1 Saccharin aerosol, 1 gram dispersions
2 Indicated test values are averages of 2 tests.

showed excellent agreement. Thus, it appears that the three fan stirring system provides acceptable uniformity in the test suspension. Furthermore, test data indicated that satisfactory mixing was attained at the lowest fan circulation rate, 50 cu.ft/min/fan.

Based upon the information appearing in Table 2, a second test series was undertaken in which one fan only was operated during each test. This approach provided an opportunity to study the effect of filter position relative to the air-flow pattern created by each of the three fans.

The only difference between these tests and those described in Table 3 was that sampling periods were reduced to 0.1 minute. This decrease was made possible by the use of a sensitive spectrophotometric method to determine the saccharin holding of each filter. It should be noted that the average concentrations for three different fan positions (and three separate saccharin disseminations) indicate good reproducibility with the dry powder ejector. For example, the average concentration during the period 1 to 1.1 minutes following dissemination was approximately 1.5 times greater than that noted for the 1-2 minute sampling period. Reference to Figure 13, which shows a typical mass decay curve for saccharin in the 1 m^3 chamber indicates that the above ratio is consistent for the decay periods covered by these tests.

The most important feature of the single fan tests was the fact that the stirring rate could be reduced threefold without detectable impairment of the mixing process. At the same time, it appears that insofar as a small chamber is concerned, location of the fan on some axis of symmetry is not necessary.

Preliminary tests were performed to determine whether the uniformity of mixing which prevailed along the vertical centerline axis was also attainable in the radial direction, Table 4. Fan operation during these tests corresponded to that indicated in Table 2. Although the data are limited, there is nothing to suggest that any distinct concentration gradients exist between wall and centerline stations. Additional tests are planned to verify the above results.

Since the saccharin aerosol at the time of sampling during these tests was characterized roughly by a mass median diameter, Mg', of 18.6μ and a geometric standard deviation, σ_g, of 1.6, it appears that the reported stirring conditions were adequate for particle diameters at least as large as 30μ. Particle size characteristics of the redispersed saccharin (M_g' = 9.4μ, M_g = 3.1μ) were larger than those for the parent material since many particles appeared as agglomerates of 2 to 4 particles. Agglomerates were described as single units whose effective diameters were based upon the estimated projected area. This reference basis was used to permit comparison of microscopic sizing data with that obtained from single particle light scattering measurements. In the latter case, instrument response for the size ranges of interest is mainly a function of particle surface area.

TABLE 4

COMPARATIVE CHAMBER CONCENTRATIONS AT WALL AND CENTERLINE, SACCHARIN AEROSOL

Test[(a)]	Radial Station	Filter Weight Gain-Milligrams A	B	C	Sampling Time Minutes	Sampling Analysis
1	Wall	0.37	----	0.34	0.1	Spectro-
	℄	----	0.36	----		photometric
2	Wall	----	0.39	----	0.1	Spectro-
	℄	0.25	----	0.39		photometric
3	Wall	3.0	----	3.3	1.0	Gravi-
	℄	----	4.0	----		metric

(a) Three fans operating, each at 50 cu.ft/min - same arrangement as for Table 2 tests.

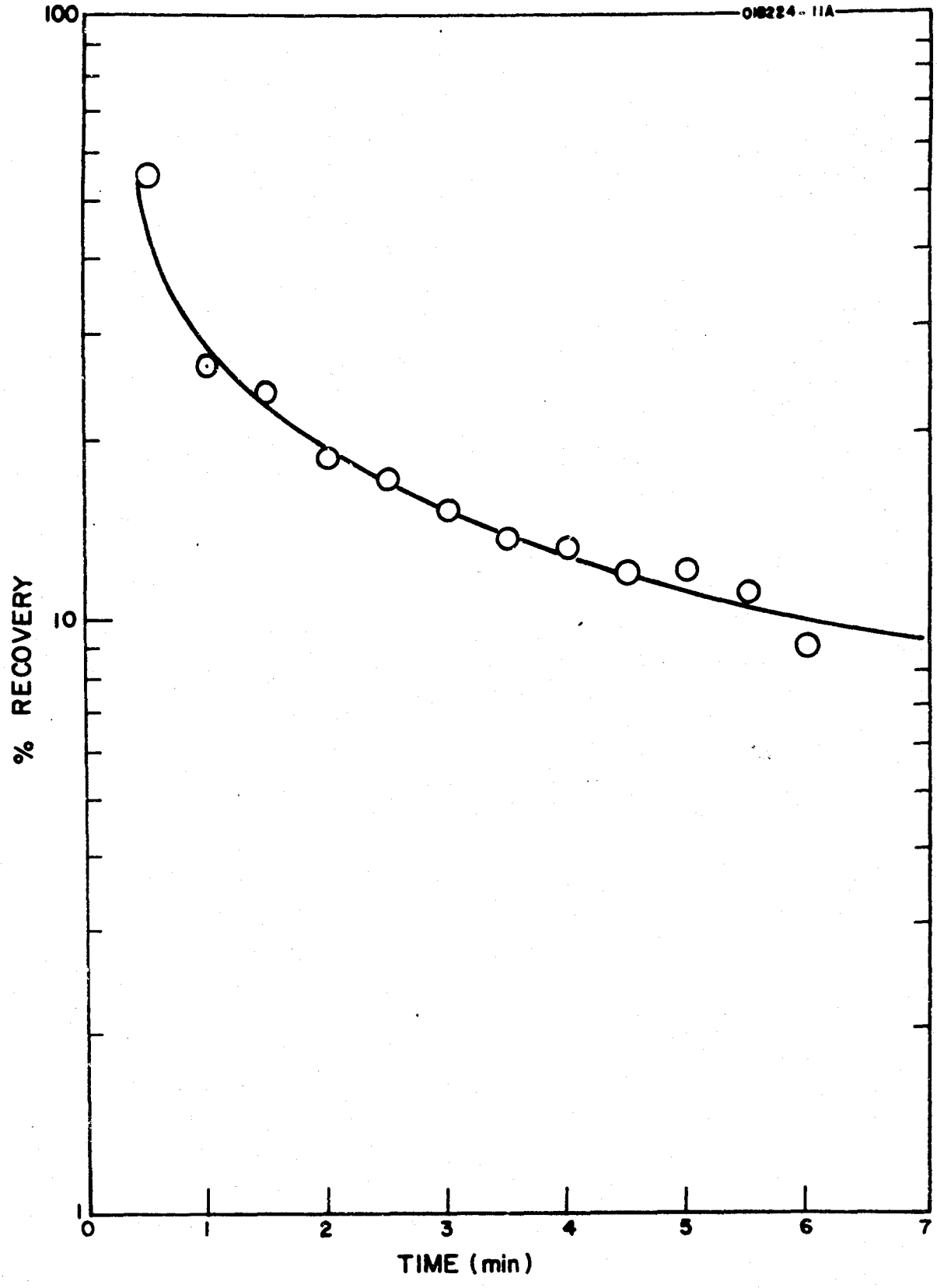


Figure 13. Mass decay curve for treated saccharin aerosol in one cubic meter chamber.

IV. CASCADE IMPACTOR STUDIES

A. DESIGN CONSIDERATIONS

We have indicated in our Second Quarterly Progress Report (Ref. 2) that a properly designed cascade impactor should be a useful device for the size measurement of particles ranging from 0.5 to 25 microns in diameter. Aside from selecting this instrument as a calibration device for system components, it now appears feasible to use it as a supporting unit for assessing certain chamber aerosols. Although we do not believe it practicable to automate the impactor system insofar as transfering sampled material from individual stages is concerned, it is recommended that samples be collected at predetermined times during the decay cycle. Use of an automatic chemical analyzing system will decrease greatly the time ordinarily required for mass analyses of the various stages.

It is expected that sizing information supplied by the impactor following 15 to 20 minutes of aerosol decay will improve the quality of data obtained by decay curve analysis by proposed inversion techniques. In fact, our current thinking is that the use of two independent sizing methods is the only effective way of defining the aerosol state immediatly after dissemination.

The cascade impactor undergoing evaluation in our 1 m^3 chamber was constructed specifically for sampling in aerosol test chambers for microscopic analyses, Figures 14 and 15. The design is sufficiently flexible, however, to permit sampling for mass analyses of stages provided that sensitive physical or chemical analytical procedures are available.

The preferred impactor design seemed to be that of Mitchell and Pilcher (Ref. 7) (Battelle design) in which round jets and an S/D ratio of 0.375 was used. (S/D = distance from jet to plate/diameter of jet.) This design has a calculated impaction parameter of $\varphi^{1/2} = 0.29$ which has been demonstrated as reasonably constant over a volumetric flow range of 2 orders of magnitude.

Mitchell and Pilcher also studied the intrastage loss factor relating collector plate diameter to impactor diameter. According to their results, wall losses were minimized when the plate and impactor diameters were 38 and 82 mm, respectively. Here, the distance between the edge of the collector plate and the impactor wall, D_{WS}, was 22 mm. In their final design, however, the value of D_{WS} was reduced to 12.7 mm for practical reasons. According to Battelle experiments performed with a bacterial aerosol, intrastage deposition losses were of the order of 4 percent. The GCA impactor was scaled approximately to the Battelle design and included Mercer's recommendation that the efficiencies of successive stages be displaced by a factor of two (Ref. 8). Since the GCA

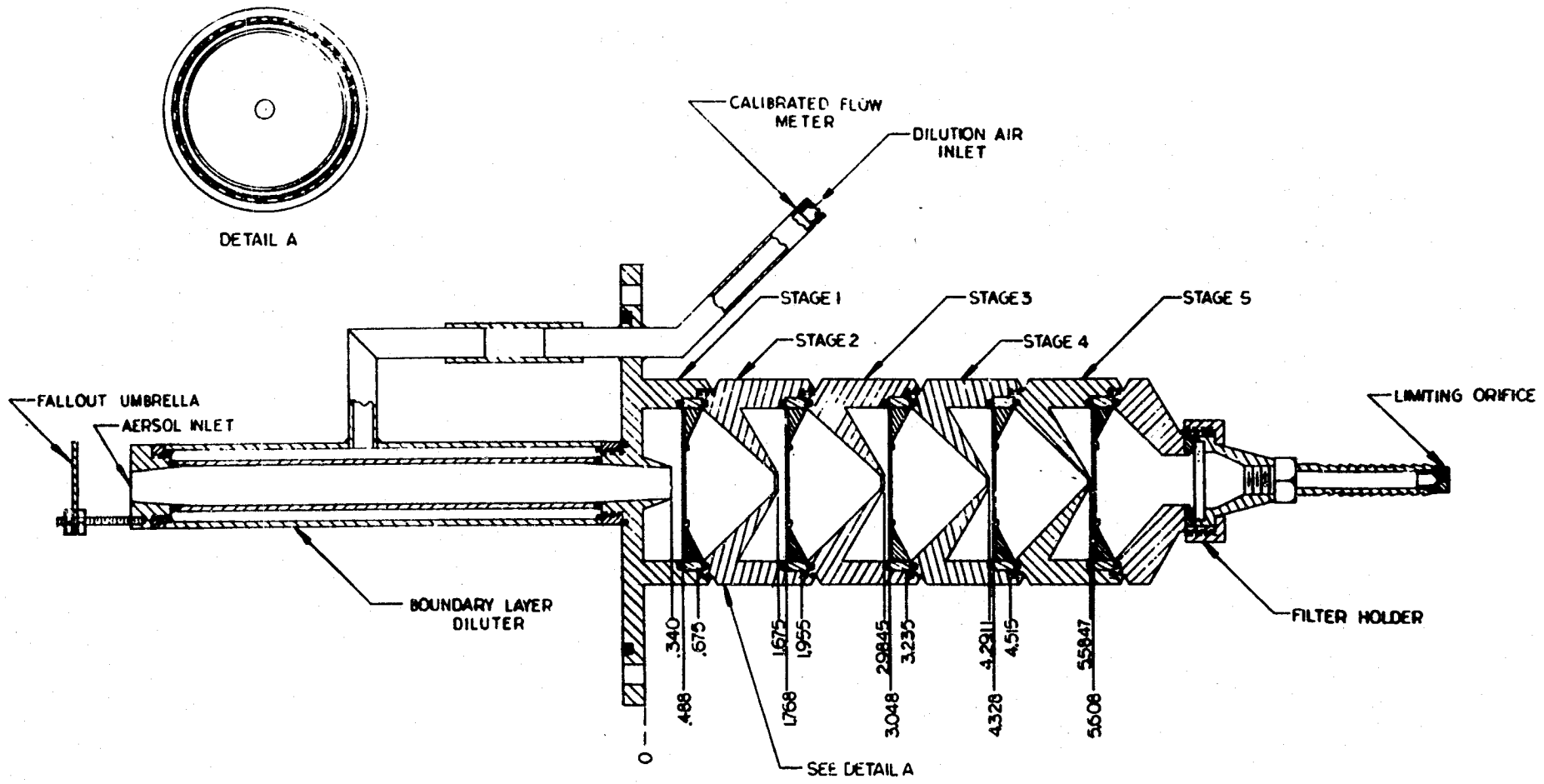


Figure 14. Cascade impactor with boundary layer diluter.

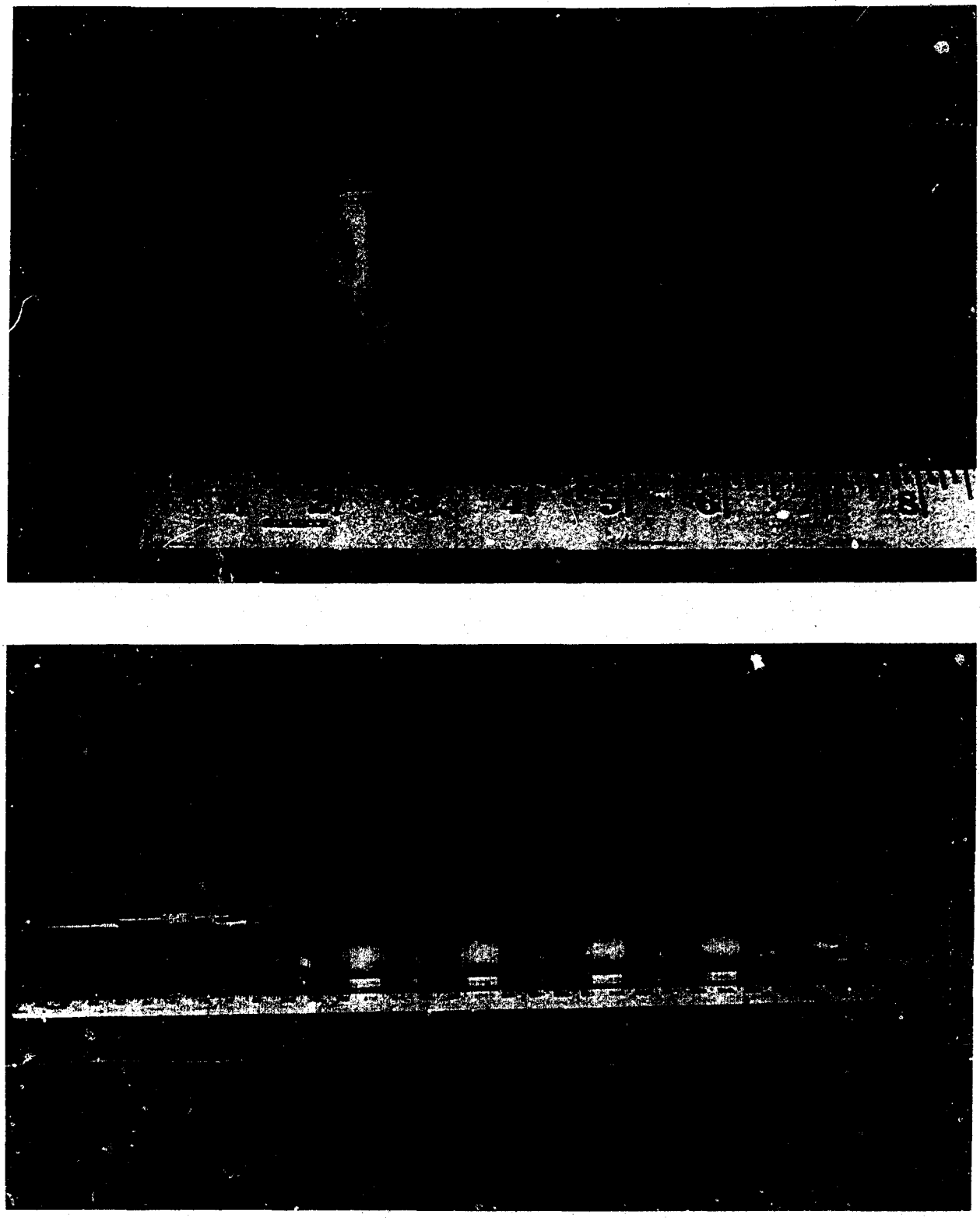


Figure 15. Cascade impactor - assembled and exploded views.

plate diameter was 25 mm, we selected an impactor diameter of 50 mm. Thus, the distance between plate edge and wall, D_{ws}, was 12.5 mm. As a result, the ratio of plate diameter to impactor diameter in the GCA unit, 0.5, was a compromise between the optimum and actual values given by Mitchell and Pilcher, 0.46 and 0.60, respectively.

The flow rate for the GCA impactor was scaled from the Battelle design on the basis of the ratio of the squares of the plate diameters.

$$Q = 12.5 \text{ lpm } (25/38)^2 = 5.4 \text{ } \ell/\text{min}$$

In our instrument, the sampling rate was assumed to be 5.0 ℓ/min for determination of key dimensions. Since the design was based upon the median value of the impaction parameter which is a function of the effective cut-off diameter (ECD) (Ref. 8), the following expressions were used to relate ECD values to particle density, ρ_p, and sampling rate, Q.

$$\varphi^{1/2} = (\rho_p \, V/18\mu D)^{1/2} \, d_p = 0.29 \text{ at 50 percent stage efficiency} \quad (1)$$

$$V = 4Q/\pi D^2 = \text{average jet velocity} \quad (2)$$

Combining and simplifying Equations (1) and (2) gives an expression for estimating the cut-off diameter d_p for each stage.

$$d_p = (1.28 \, D^3/\rho_p \, Q)^{1/2} \quad (3)$$

Definition of terms in the above equations are as follows:

D = jet diameter, mm

d_p = particle diameter, μ

Q = sampling rate, ℓ/min

ρ_p = particle density g/cc

TABLE 5

SUMMARY OF DESIGN PARAMETERS FOR GCA CASCADE IMPACTOR

Stage	Jet Diameter mm	Cut-off Size - Microns (50 percent Count)	(50 percent Mass, σ_g = 1.5)
1	10.0	16	26.7
2	6.30	8	13.2
3	3.97	4	6.6
4	2.50	2	3.3
5	1.57	1	1.6

B. CONSTRUCTION FEATURES

The main body of the cascade impactor was composed of separate, threaded aluminum sections and the stages (which are interchangeable) were made from 316 stainless steel. Each stage consisted of an inner ring with a machined shoulder to support the collecting plate at the proper distance from the jet. Four faired rods connected this holder to an outer ring or stage section which was placed between the body sections of the impactor.

The outer stage sections were sealed on both sides against O-rings in captive grooves milled in the jet section of adjacent segments of the impactor. The system was designed so that the proper jet-to-plate distance was obtained with metal-to-metal contact between the stage and jet sections. The removable stages were provided with spring clips to hold the collecting plates firmly in position. An alternate design was fabricated in which the collecting plates were permanently cemented to the stage. This method is useful for microscope counting since the entire stage can be carried to the microscope with minimal handling of the collection plate.

A special diluter section precedes the first stage of the impactor so that inlet particle concentration may be reduced to levels suitable for microscopic examination of the collecting plates, Figure 16. This provides a capability to reduce particle counts on a given stage by as much as a factor of ten if necessary. When particulate concentrations are very high, one can extend sampling periods to sufficiently long intervals, 0.25 to 0.5 minutes, to minimize sampling volume errors attendant with starting and stopping the unit.

The diluter section was made from 316 stainless steel and similar in design to that described for the light scattering particle counter. A separate pump delivers a prefiltered air supply at preset flow rates ranging from 0 to 5 ℓ/min to a porous ceramic diluter tube. Concurrently,

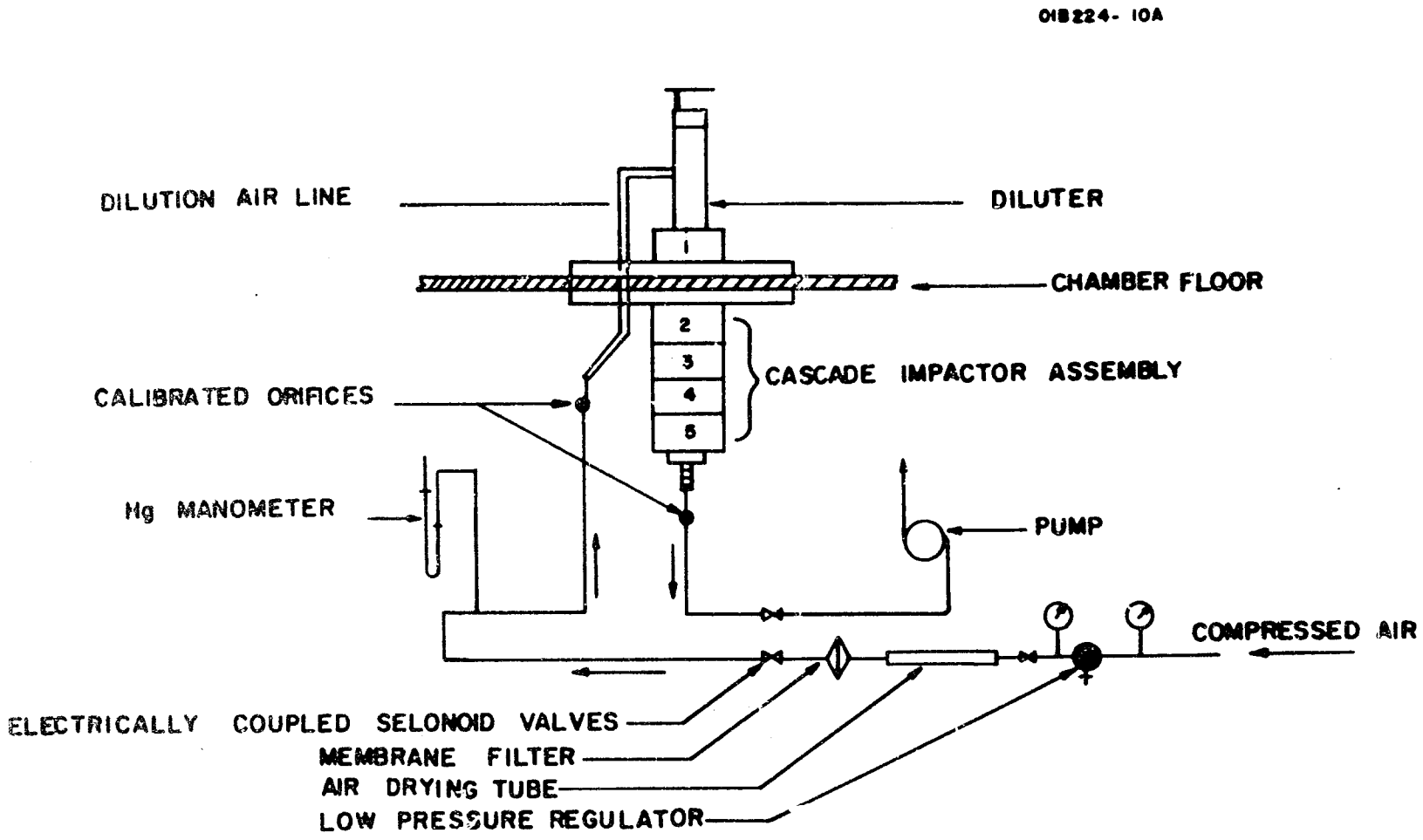


Figure 16. Schematic - cascade impactor - diluter system.

the main air flow through the cascade impactor is controlled by a critical flow orifice which guarantees a constant flow rate through the impactor at all times of approximately 5 ℓ/min. In order to dilute the inlet particle concentration by a factor of two, for example, the delivered diluter flow would be adjusted to 2.5 ℓ/min. Since the main flow through the impactor is always constant at 5 ℓ/min, the true sampling rate from the test atmosphere is restricted to 2.5 /min.

C. LABORATORY TESTING

The first step in the rating of the GCA impactor was to check the leak characteristics of the unit. This test was performed by assembling the unit in its normal sampling configuration which included an all-glass filter in a connecting holder as the sixth stage. With the inlet stoppered, there was no appreciable loss of vacuum at 15 inches of Hg. The observed sampling rate as governed by the critical orifice was 5.44 l/min. This was considered to be sufficiently close to the design objective of 5 l/min since final values for stage constants were to be determined by microscope calibration.

A preliminary determination of stage constants was made by sampling a saccharin aerosol cloud established in the 1 m^3 chamber. Parallel samples were collected on AA Millipore filters for comparison of particle size properties. Those samples which were intended for microscope examination only were collected on glass slides coated with Dow Corning 50 centistoke lubricant to minimize bounce-off and reentrainment. Glycerine coatings were applied to slides which were to be analyzed by chemical or physical methods for the weight of chemical agent present. In order to obtain a thin, uniform glycerin film, glass slides were etched by dipping in 20 percent hydrofluoric acid for 30 seconds, followed by dipping in a 10 percent glycerine in methanol solution. The residual coating after evaporation of the methanol provided a good adhesive surface from which deposited particles could be completely dissolved by selection of a suitable solvent (methanol).

Particle sizing data for saccharin particles collected on impactor stages 2 through 5 are shown in Table 6.

TABLE 6

SIZE PARAMETERS FOR SACCHARIN PARTICLES COLLECTED BY CASCADE IMPACTOR (DIAMETER MICRON)

Stage	Count Median[a]	Mass Median[a]	50 percent Cut-off[b]	Effective Drop (Particle) Size (EDS)[c]
2	10.7	36.9	8	26
3	6.9	13.0	4	15
4	3.4	6.6	2	10
5	2.4	3.6	1	6.0

a. Based on light field microscopy
b. Based on design and operating parameters
c. Based on absolute calibration technique

Count and mass median diameters based upon light field microscopic measurements of individual impactor stages appear in columns 2 and 3. It should be noted that the count median values are consistently greater than the 50 percent cut-off sizes (Column 4) determined by theoretical considerations. These findings substantiate Mercer's data (Ref. 8) and illustrate the fact that 50 percent cut-off sizes are useful only for comparing different cascade impactors. They do not, however, depict characterizing stage diameters which, in conjunction with chemical or gravimetric analyses, can be used to construct a mass distribution curve for the sampled aerosol.

Better estimations of stage constants are the mass median diameters for individual stages (Column 3) although these values are sometimes poor approximations depending upon the size properties of the parent aerosol. The most reliable estimates of stage constants for a specified aerosol cloud are the effective droplet sizes (EDS), (Column 5). In our case, these values were obtained by combining the results of a separate particle size analysis (based upon microscopic observation of a membrane filter sample) with the chemical analyses of the impactor stages. The count and derived mass distribution curves based upon membrane filter samples were considered to represent the true size distributions. The average of three such curves is shown in Figure 17. A second curve depicts the best estimate of the aerosol size properties as determined by simultaneous cascade impactor measurements. In the latter case, the mass median diameter values were used as stage constants (Column 3, Table 6). Although the mass median diameters are in good agreement for both distributions, significant variations appear in the tail regions.

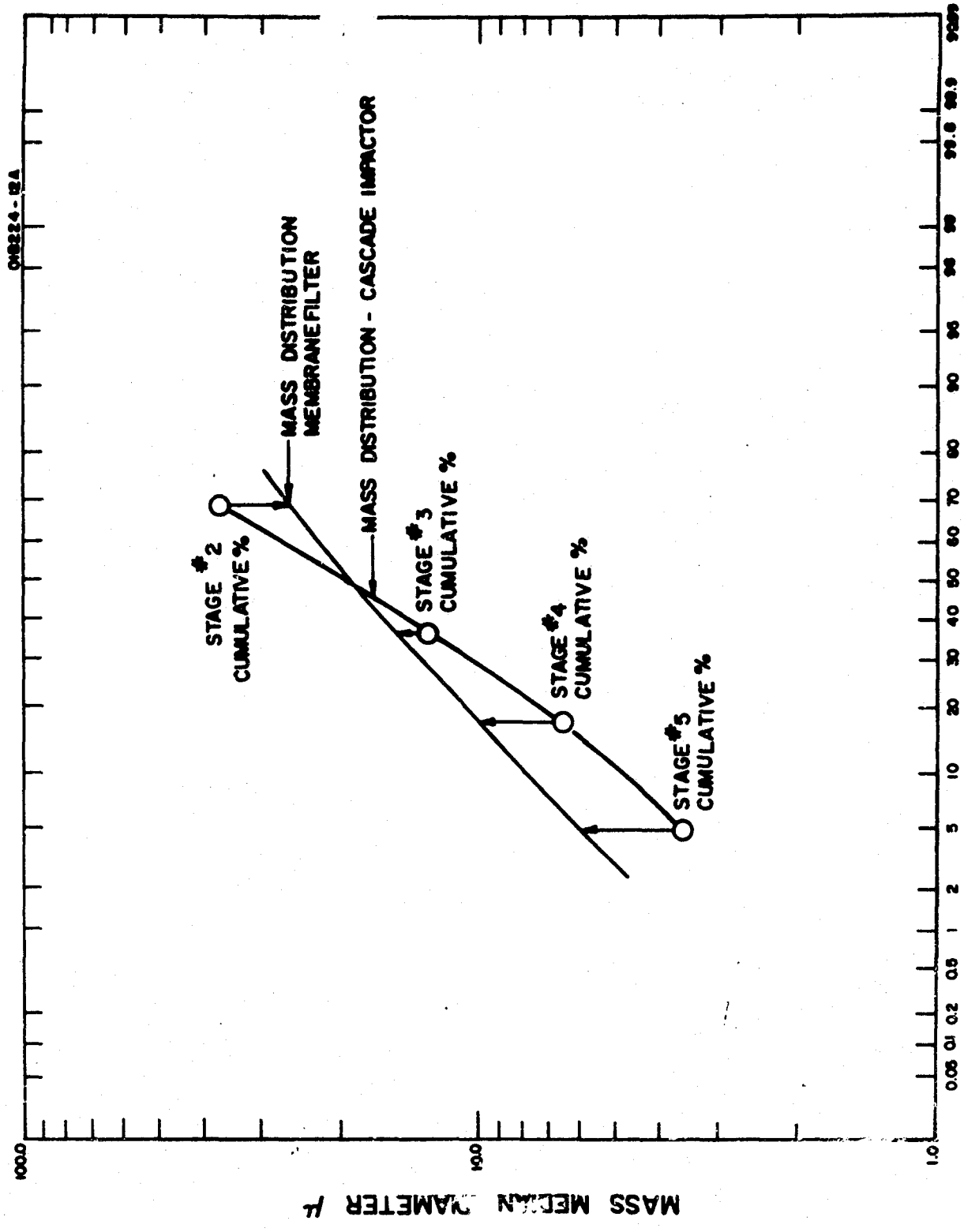


Figure 17. Comparative size measurements, membrane filter and cascade impactor on saccharin aerosol.

In order to obtain the correct representation of stage constants for the specific saccharin aerosol under study, one draws a vertical line from the cumulative percentage value indicated for a given stage of the cascade impactor to the point where it intersects the curve determined by membrane filter sizing. The particle diameters corresponding to these points are the best estimates of the stage constants for the aerosol in question. It should be noted, however, that the term "constant" is not entirely correct since significant changes in the parent distribution will require a new calibration.

Although the data presented above are preliminary, the results are considered very encouraging insofar as impactor performance is concerned. First, the saccharin concentrations in the chamber were nearly identical for simultaneous filter and cascade impactor samples. This implies that very little material was lost due to intrastage deposition and that flow errors attributable to leakage were negligible. Second, the estimates of mass median diameters by impactor and membrane filter samples were in excellent agreement. Third, it appears that errors associated with inertial losses when sampling anisokinetically were not serious for particles diameter less than 50 microns.

V. LIGHT SCATTERING PARTICLE COUNTER

A. TECHNICAL DISCUSSION

Light scattering particle counters (Ref. 1,2), which furnish information on both number concentration and particle size distribution, provide a unique analytical capability for certain chamber aerosols. Insofar as working range is concerned, these instruments can, with suitable modifications and proper calibration, be applied to a broad range of particle diameters, 0.5 to > 50μ. Their major limitation as applied to chamber aerosol studies lies in the fact that the relation of light scattering intensity to particle size may be obscured by particle geometry, surface roughness, refractive index, and dielectric and absorbing properties. Hence, only in the case of aerosol clouds which are homogeneous with respect to chemical and/or physical properties can particle size properties be established with certainty. For example, mechanical dissemination, such as atomizing or condensation, techniques produce essentially single component clouds which can be assessed accurately by light scattering counters. On the other hand, analyses of aerosol clouds generated by explosive means are usually complicated by the presence of foreign particles which cannot be isolated from the particles of interest. Although it is possible in some instances to make corrections for non-agent particles, light scattering data for such mixed aerosols is usually interpreted on a relative basis.

Despite recognized limitations in commercially available light scattering counters, we have undertaken a program to determine how this type of instrument might be integrated with a chamber assessment system. Since a commercial unit which performed size operations on the basis of 90° light scatter was available for testing, these studies were begun during the first part of our laboratory evaluation program.

The first step in the adaptation of this device was to build a dependable sampling-dilution system which would permit selection and control of particle number concentrations entering the sensing elements of the light scattering counter.

The special dilution system constructed for use in this program was a second generation system based upon an earlier GCA design, Contract DA-108-AMC-249 (A), the major changes lying in the miniaturization of the device. The principal construction details of the diluter section are shown in Figures 18 and 19. Sampled air enters the unit at flow rates ranging from 0 to 300 cc/min, through a 0.444 mm diameter orifice and passes downward through the interior of a 15.2 cm

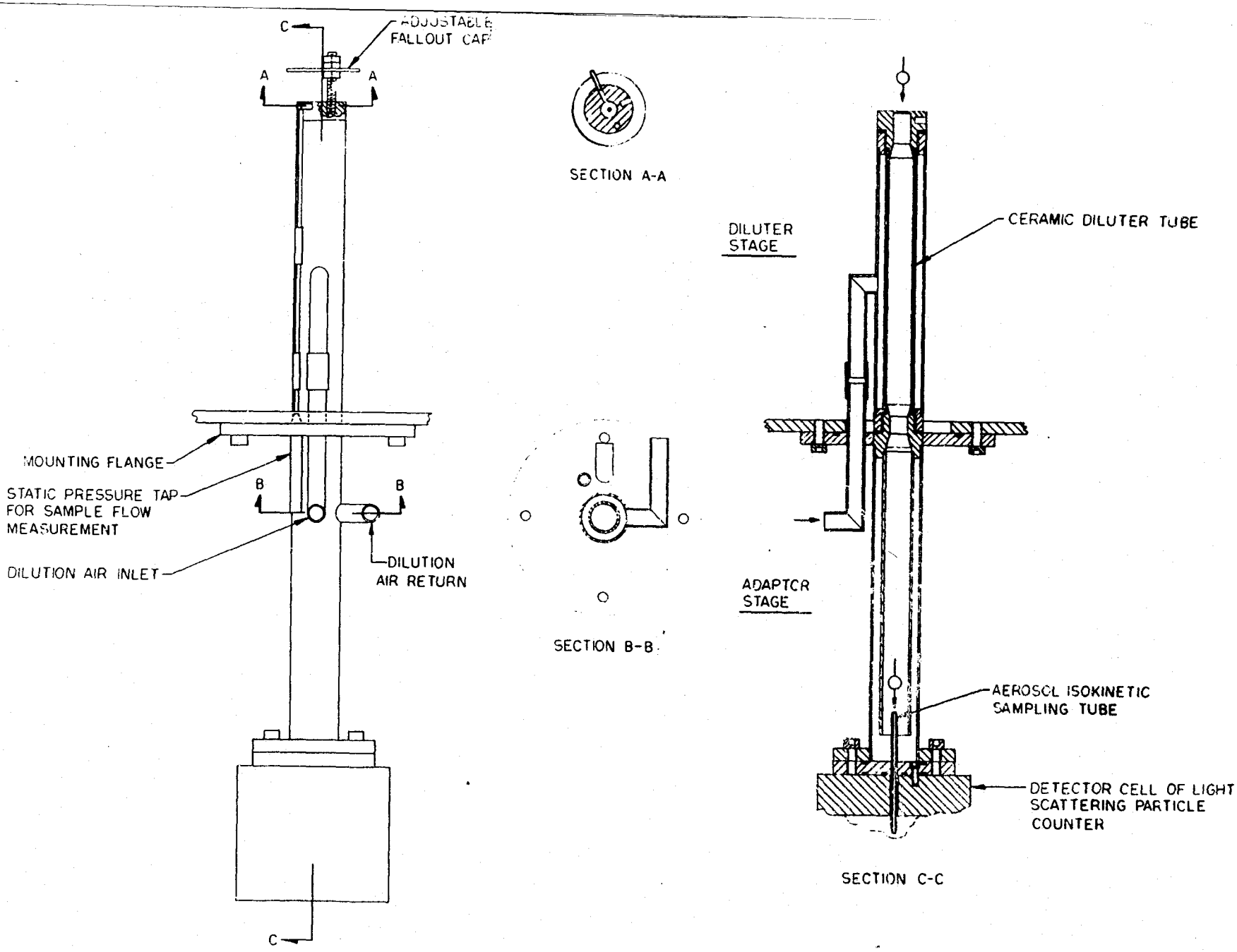


Figure 18. Aerosol sampling diluter.

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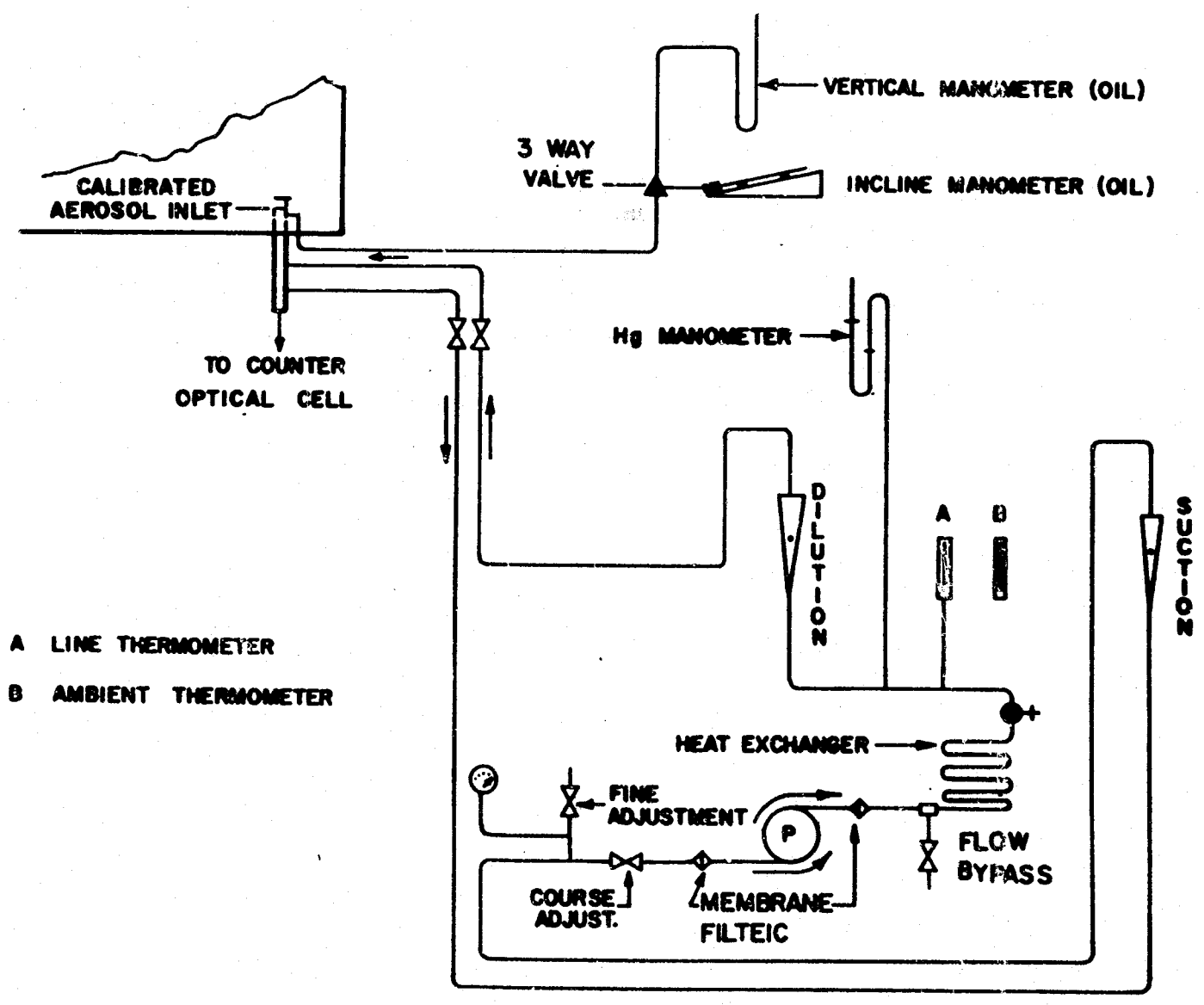

Figure 19. Closed loop diluter system.

long, 1.22 cm diameter porous ceramic tube. Filtered dilution air, cooled to ambient temperature, is delivered at a flow rate of ≈ 30 ℓ/min, depending on the desired dilution, to the annular chamber surrounding the ceramic tube. Passage of dilution air through the porous walls provides a boundary or sheath air layer which minimizes deposition of particles on the wall during the mixing process. After leaving the diluter section, the flow continues in a vertical path for another 6 in. at which point an aliquot is sampled isokinetically at 291 cc/min for admission to the optical sensing chamber of the particle counter. The remaining (and the major portion) of the flow is withdrawn by a separate pump and recycled to the diluter section.

The inlet orifice to the system in conjunction with a static pressure measurement made immediately downstream of the orifice provides a pressure reading directly relatable to sampling flow rate. The ratio of sampling flow to diluter flow provides the multiplying factor to be applied to indicated particle concentrations. The dilution capability for this device ranges from 60 to 1000.

Since the present light scattering counter is equipped with a single channel display (0.32 minutes counting per channel) the total time required to observe 10 or 20 different size channels is 4.0 or 8.0 min, including 0.08 minute/channel for readout and channel changing time respectively. Should the aerosol cloud contain large particles which settle rapidly, sequential readout of various size channels introduces errors which must be corrected for the time lag factor. It is preferred that a multi-channel readout capability be used for chamber aerosol studies to circumvent this problem. Commercial instrumentation is available to perform this operation. A new concept has been suggested whereby all sizing information from the light scattering sensor unit can be put on magnetic tape and introduced at a later time to the gating and size discriminating circuit of the counter for selected real-time readout of particle size data. GCA has examined various means by which multi-channel display might be achieved without prohibitive cost. One method is to provide a simultaneous display for 10 to 15 channels which should be adequate to define the major range of particle diameters in a given size distribution. Since this range would not be sufficient to cover the complete range of sizes possible for all dissemination techniques, it is proposed that a scaling adjustment be added to the discriminating circuitry so that arbitrary diameters can be assigned to individual channel readout. It is presumed here that either a priori knowledge of the aerosol properties be available or that pilot tests will be performed to establish the approximate characteristic of the chamber aerosol.

Optimum performance of light scattering counters (or any other assessment techniques depending upon sampling and transport of a fixed aerosol volume to a collection or measuring unit) depends upon a minimum straight length of sampling line. A vertical passage is necessary when the possibility of particle loss by gravity settling is added to the problem of diffusional deposition in sampling lines. For this reason the optical sensing head was removed from the main instrument casing and placed directly beneath the 1 m^3 chamber. Although it would be preferable to keep this equipment outside the test chamber, particularly where explosive dissemination techniques are employed, we believe that the optical head can be encased in a shrapnel-proof housing and shock mounted for use within any test chamber. Experience at Edgewood Arsenal with shockmounted holographic camera equipment located within a few feet of explosive events would appear to support this contention. (Ref. 9).

E. EXPERIMENTAL PROGRAM

The diluter system was evaluated to determine its accuracy by making comparative measurements over a broad range of cloud dilutions, 60/1 to 1000/1. These tests were performed by injecting 1 gram quantities of a saccharin powder, $M_g' = 9.4\mu$ $M_g = 3.1\mu$ into the 1 m^3 test chamber. Sizing data reported here are based on microscopic measurements of an oil dispersion of the powdered material in which only discrete particles were sized. Aerosol sampling started one minute after dissemination at which time mass concentrations ranged from 300 to 500 mg/m^3. In order to minimize the correction required for aerosol decay, only one size channel, 4-5μ, was monitored. The observed decrease in number concentration for the above size range over the 3 to 4 minutes required to make 10 separate count measurements was of the order of 10 percent. Five successive counts were made at one of the preset dilution conditions shown in Table 7, followed immediately by five similar measurements on the undiluted cloud.

Test results indicated good agreement with theoretical dilution ratios in view of the number of variables which might, singly, or in combination lead to experimental errors. A statistical problem is introduced in that the electronic counting system becomes inoperative or "chokes" when the counting rate exceeds 15,000 particles/min/channel. Dilution of this limiting concentration by a factor of 100 reduces the count rate to 150 particles/min with a standard error of the mean of 8.2 percent. However, at a 1000:1 dilution, the expected counting rate is 15 particles/min. Here the standard error increases to about 26 percent.

TABLE 7

FLOW RATES FOR SAMPLING DILUTER SYSTEM, LIGHT SCATTERING PARTICLE COUNTER

Theoretical Dilution Ratio	Dilution Flow l/min	Sample Flow cm^3/min	Relative Particle Number Concentration[a] Diluted Sample	Undiluted Sample	Observed Dilution Ratio
60:1	28.6	485	52	2911	56
100:1	28.8	291	31	3174	102
500:1	29.0	58	7.6	4185	560
1000:1	29.1	29.1	5.2	5785	1110

(a) Number of particles counted during 0.32 minute period.

Since the aerosol cloud is undergoing decay while successive counts are made, the average concentration value for diluted samples should be about 4 percent greater than the value at the end of the 2 minute counting period. Therefore, undiluted concentration measurements made immediately afterwards (within 0.4 minute) should have been compared to the corrected value for the diluted sample. Since these tests represented preliminary evaluation of the overall effectiveness of the dilution system the above refinement was omitted.

Examination of the theoretical and observed dilution ratios in Table 7 shows that the observed values are smaller than expected at low dilutions and, conversely, greater at high dilutions. It appears at this time that the differences in sampling rates from the test chamber may explain this factor. Noting that actual sampling rates decrease from 485 cc/min at 60:1 dilution to 2911 cc/min at 1000:1 dilution, one would expect to encounter the smallest sampling error at the highest sampling velocity according to Levin (Ref. 4). This problem has been discussed in our Second Quarterly Progress Report (Ref. 2). Although the magnitude of the errors is not large based upon our limited data, it is planned to pursue the effect of sampling rate and velocity on sampling accuracy. Qualitatively, it is expected that low velocity sampling from stagnant or slow-moving gas streams may prevent representative collection of the larger size fractions of suspended particulates.

VI. MASS-DECAY CURVE ANALYSIS

The generation of mass decay curves for given particle size distributions as a result of gravitational precipitation under conditions of continuous stirring is a comparatively simple process. For particles undergoing stirred settling in a cylindrical chamber of height, h, the mass decay curve is given by

$$\frac{M(\tau)}{M(0)} = \int_a^b \left[\exp\left(-\beta\, d^2\tau\right)\right] f(d)\delta d \qquad (1)$$

where $M(\tau)$ = mass concentration at time τ

$M(0)$ = mass concentration at time $\tau=0$

d = particle diameter

$f(d)$ = distribution function

$\exp(-\beta\, d^2\tau)$ = decay function based on Stokes' settling

β = constant

and where a and b define a range of expected diameters. These decay curves may sometimes be fitted to the experimental data by adjusting parameters associated with the given distribution. Such an approach requires *a priori* information as to the type of distribution represented by the test cloud.

Direct inversion of decay curves to obtain the distribution functions would exclude the above requirement. By expressing the decay function, $\exp(-\beta\, d^2\tau)$, as $F(d,\tau)$

$$\frac{M(\tau)}{M(0)} = \int_a^b F(d,\tau)\, f(d)\, \delta d \qquad (2)$$

Equation (2) appears as a Fredholm equation of the first kind. The problem of obtaining $f(d)$ when $M(\tau)$ and $F(d,\tau)$ are known has not been examined rigorously. Two basic approaches have been used to obtain the distribution function from decay curves resulting from gravitational settling of particles under stirred settling conditions, a graphical method and an analytical method.

A simple graphical technique for direct inversion of decay curves has been advanced by Dimmick, et al (Ref. 10). This method yields a rough approximation with the better definition of size distribution on log-probability paper. Only three or four points can be obtained by this method. Therefore, it would be desirable either to improve Dimmick's method or to formulate a new graphical or semi-graphical approach.

Equation (2) may be replaced by the following system of equations.

$$\frac{M(\tau_j)}{M(0)} = \sum_{i=o}^{i=n} F(d_i,\tau_j)\ f(d_i) \qquad j = 0,1,\ldots n \tag{3}$$

where a reasonable range of diameters can be estimated from knowledge of the range of expected diameters. If a mass decay curve is plotted on semi-log paper, it can be treated as if it were composed of a number of monodisperse aerosols whose decay would appear as linear functions. A log-normal distribution having a mass median diameter, Mg′, of 50 microns and a geometric standard deviation, σ_g of 3.0 was decayed in this manner according to the corresponding monodisperse aerosols represented by arbitrary size fractions. Here, the parent distribution was broken down into size intervals in which the ratio of maximum to minimum diameter was $\sqrt{2}$. Settling characteristics of all particles within each size range were assumed to be approximated by the arithmetic mean of the range. The mass decay curve so obtained was in good agreement with that obtained by a Simpson's Rule approximation to Equation 2. In the latter case, the accuracy should be better since average diameters for the range were estimated by a more refined curve fitting technique. A direct inversion technique appeared attainable from a study of these monodisperse aerosols and their relations to the composite mass decay curve.

The first approach at inversion utilized the first step in Dimmick's method, i.e., construction of a "final" exponential slope line. It appears that this final exponential slope is a satisfactory representation of the smaller particles present in the aerosol cloud. If the point of tangency occurs at a time $\tau = \tau_o$

$$\frac{M(\tau_o)}{M(0)} = f(d_o)\ \exp(-\beta\, d_o^{\,2}\, \tau_o) \tag{4}$$

from which d_o is determined.

It should be noted that the symbol τ_o refers to the time coordinate at which the first tangent line is drawn. It should not be confused with time zero ($\tau = 0$) relating to mass concentration, M(0) at time zero.

At an earlier time, τ_1, in the decay cycle, and for a selected diameter d_1

$$\frac{M(\tau_1)}{M(0)} = f(d_1)\ \exp\ (-\beta d_1^{\,2}\, \tau_1) + f(d_o)\ \exp\ (-\beta d_o^{\,2}\, \tau_o) \tag{5}$$

which yields $f(d_1)$. The process was continued for successive (d,t) intervals to evaluate the term $f(d_i)$. The latter value describes the upper limit of particle size which can be defined by this method. Reconstruction of the mass decay curve in this manner led to negative values of $f(d_i)$. No simple process for determining τ_i and d_i values was developed except for rather time consuming empirical approaches which were not suitable for the present program.

It appeared that the tangent to the mass decay curve at some time, τ_i, represented the modal diameter, d_i, of the distribution at time τ_i. At a later time, t_{i-1}, the modal diameter, d_{i-1}, will have decreased for a unimodal distribution. The straight lines representing these two diameters must intersect at sometime τ such that $t_i < \tau < \tau_{i-1}$. This concept was utilized in a second approach similar in some respects to the one described above. Here the effect on the mass decay curve of the various monodisperse aerosols was taken into account over the entire curve. Again, a final exponential curve was drawn. The resulting distribution from the second reconstruction process was constrained to a unimodal system, and furthermore, gave no information on the distribution of particles greater than Mg'. Since the quality of the final data was of the order of that obtained by Dimmick's method, the above approaches were discontinued.

The most obvious analytical approach is that of the straightforward matrix approximation to Equation 2. Introducing the following matrix notation

$$f = f(d_j) \quad F = F(d_j,\tau_i) \quad M = M(\tau_i)$$

Equation 2 can be written in the following form:

$$f = F^{-1}M \tag{6}$$

It was recognized that the solutions for f depend greatly on the accuracy of M. This method did not yield useful results in the present case unless $M(\tau)/M(0)$ (constant vector) values were known to the order of 6-8 decimal places. Since it is highly unlikely that $M(\tau)$ can be defined to better than 99 percent accuracy, Equation 2 should be expressed in the following form.

$$M(\tau) + \epsilon(\tau) = \int_a^b F(d,\tau)\, f(d)\delta d \tag{7}$$

A technique for the solution of an equation of this form has been presented by Phillips (Ref. 11). Although the kernels used by Phillips are not those that describe aerosol decay, substitution of the appropriate kernels should not invalidate his approach.

LITERATURE CITED

1. Dennis, R., Euling, R., Gussman, R.A., and Hommel, C.O., "Aerosol Dissemination Assessment," First Quarterly Progress Report, Contract DA 18-035-AMC-376(A), (December 1965).

2. Dennis, R., and Hommel, C.O., "Aerosol Dissemination Assessment," Second Quarterly Progress Report, Contract DA-18-035-AMC-376(A), (May 1966).

3. Bulba, E., and Silverman, L., "A Mass Recording Stack Monitoring System for Particulates," presented at the 58th APCA Meeting, Toronto, Canada. (June 20-24, 1965).

4. Levin, L.M., "The Intake of Aerosol Samples," Translated by K. Syers from Izv. Akad. Nauk. SSSR Sera Geofiz. No. 7, 914-925, (1957). (Reprint Order Number GEO 112.)

5. Gussman, R.A., Dennis, R., and Silverman, L., "Notes on the Design and Leak Testing of Sampling Filter Holders," AIHA J. 23, 480-81, (December 1962).

6. Rapaport, E., and Weinstock, S.E. (Devir), "A Generator for Homogeneous Aerosols," Experimentation (Switzerland) 11, 363, (1955).

7. Mitchell, R.I., and Pilcher, J.M., "Improved Cascade Impactor for Measuring Aerosol Particle Sizes in Air Pollutants, Commercial Smokes and Cigarette Smoke," Ind. Eng. Chem. 5, 1039-1042, (1959).

8. Mercer, T.T., "On the Calibration of Cascade Impactors," Ann. Occup. Hyg. (Pergamon Press, Ltd.,) 6, 1-14, (1964).

9. Zinky, W.R., "Hologram Camera and Reconstruction System for Assessment of Explosively Generated Aerosols," Final Report, Contract Number DA-18-035-AMC-256(A). (7 October 1965).

10. Dimmick, R.L., Hatch, M.T., and James, N.G., "A Particle-Sizing Method for Aerosols and Fine Powders," A.M.A. Arch. of Ind. Health, 18, 23, (July 1958).

11. Phillips, D.L., "A Technique for Numerical Solution of Certain Integral Equations of the First Kind," J. Assoc. Comp. Mach., 9, 84-97 (1962).

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13. ABSTRACT
This report describes the work accomplished during the third three months of the program to develop, design and fabricate a complete chamber in the development of chemical devices, and applicable to a broad range of agents and dissemination methods. Aerosol description in terms of chemical composition and particle-size properties should provide useful data on disseminator performance and be relatable to biological effects. Sensor output from instruments in the system should be amenable to either direct display or data storage and processing or both whenever the latter techniques prove to be the most effective methods for obtaining real-time measurement of aerosol cloud properties. During this period, the selection of instruments and experimental methods has been further reduced based upon results of the information survey and a laboratory test program. The reasons for selecting either specific instrumentation or techniques or both are cited. Similarly, the basis for rejection is indicated for certain methos which do not appear applicable to the present system.

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