AFCRL-65-724

INVESTIGATION OF METHODS TO MEASURE SIZE DISTRIBUTION OF FOG DROPLETS AND CONDENSATION NUCLEI

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505 King Avenue
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Contract No. AF 19(628)-344

Project No. 7605

Task No. 760501

FINAL REPORT

Period Covered: 1 April 1962 – 17 April 1965

15 May 1966

Prepared
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AIR FORCE CAMBRIDGE RESEARCH LABORATORIES
OFFICE OF AEROSPACE RESEARCH
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ABSTRACT

This report describes the development of a light-scattering cascade impactor to measure the size distribution of fog droplets in the range from 0.5 micron to 16 microns. The instrument comprises six impaction stages, each equipped with a forward-light-scattering optical system to measure fog droplets of size <16, <8, <4, <2, <1, and <0.5 micron, respectively. An impaction stage to remove particles larger than 16 microns is installed at the instrument intake. Two additional stages are furnished for calibration of the electronic and optical systems.

Feasibility of a condensation-nuclei counter based on the principle of electrostatic precipitation was investigated in the laboratory.

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INTRODUCTION

The research program covered in this report was concerned with the development of two instruments, one for measuring fog droplets and the other for counting condensation nuclei.

A field-tested version of the first instrument, including operating instructions and calibration curves, has been delivered to the Air Force Cambridge Research Laboratories. This instrument incorporates elements of two well-tested devices, the Battelle Cascade Impactor and the Naval Research Laboratories forward-light-scattering Penetrometer (see Appendix II). Some suggestions for the further improvement of this instrument are made.

The second instrument is based on the principle of electrostatic precipitation. The laboratory instrument that was assembled and tested demonstrated the feasibility of the general approach. However, two alternative systems for nuclei charging were considered, and it was not possible to incorporate both in the laboratory model. Therefore, laboratory investigation of the second alternative is recommended before a selection is made.

For convenience in discussing the two aspects of the research program, this report is organized in two major sections:

(1) Fog-Droplet Measurement

(2) Condensation Nuclei Counting.

RECOMMENDATIONS

Several possibilities for improving the operation of the light-scattering cascade impactor were revealed during the research program. It is recommended that work be continued as outlined in detail under "Further Improvement of the Light-Scattering Cascade Impactor", page 10.

With respect to the condensation-nuclei counting device, investigation is recommended relative to the feasibility of reducing the difficulties associated with background ion currents by bringing nuclei into equilibrium with a controlled ion atmosphere produced by a radioactive source.

FOG-DROPLET MEASUREMENT

The development of the light-scattering cascade impactor for fog-droplet measurement involved five principal steps, each of which is described in detail in this section of the report:

(1) Initial investigation with a three-stage laboratory device

(2) Construction and calibration of a seven-stage manual field instrument

(3) Field study

(4) Investigation of 45-degree light scattering

(5) Construction and calibration of fully automatic field instrument.

The fully automatic field version was delivered on June 27, 1964, for operation at Otis Air Force Base with instructions for operation as given in Appendix I. Figures 1 and 2 show this version; calibration curves are given in Figure 3. A number of additional improvements for this instrument were suggested during the final portion of the development program, and these are covered at the conclusion of this section under "Further Improvement of Light-Scattering Cascade Impactor". The instrument described here measures droplet sizes below 16 microns through 0.5 micron, with a printout of equivalent mass per cubic meter of air sampled. Light-scattering devices made to NRL specifications and purchased from National Instrument Laboratories of Washington, D. C., were incorporated in this instrument.

Initial Investigations

Initially, the AFCRL indicated that the Cascade Impactor and a readout for each impactor stage would be a practical means of continuous measurement of fog droplets. Since the Battelle Cascade Impactor(1) separates particles into size ranges, the major problem was how to measure the mass of droplets at each impactor stage. Three means of measurement were investigated: (1) light scattering, (2) electric-resistance changes of a wire grid, and (3) electric-resistance changes of lithium chloride.

It was decided that a combination of the Cascade Impactor and a light-scattering device would be the best approach to measurement of fog droplets.

A three-stage light-scattering cascade impactor was made with impaction slides in only the first two stages.

Figure 4 is a photograph of this device showing the arrangement. The aluminum-colored cascade impactor stages were designed and made at Battelle. Also, the black spot, wiring, and switch box shown below the "Solar" constant-voltage transformer were made in the Battelle laboratories. The light-source housing, optics, photomultiplier, and electronic indicator were the purchased items adapted for this device. At the bottom of the stand is a vacuum pump and vacuum gauge. The pump is used to draw air samples through the device for impaction and light-scattering analysis.

Figure 5 is a sketch showing a light-scattering stage.

Figure 6 is a sketch showing general arrangement of optics.

Initially, considerable difficulty was encountered while checking out the purchased instrument electronics. It was determined that the instrument was not wired correctly according to the Naval Research Laboratories (who designed the original instrument). A.c. current was carried to the d.c. meter through filament wires that had been soldered to the d.c. circuit. The wiring was revised to eliminate the problem.

The lens chambers were supplied without any means of calibrating the photomultiplier output. Accordingly, calibrating holes were made in dark spots on the condenser lenses as NRL instructions specified. Shields to cover the calibrating holes during operation of the instrument were designed and constructed.

Initial calibration runs were made using water aerosols tagged with a fluorescent dye, uranine. Finally, calibrations were made using a solution of 73 percent propylene glycol and 27 percent distilled water containing 0.126 g uranine/100 ml solution, which is in equilibrium at 60 percent relative humidity, to form the test aerosol. This solution was sprayed through three different nebulizers, a DeVilbiss No. 40, an Ohio, and a Deutreband, to vary aerosol mass flow rate and size distribution. The DeVilbiss was found to produce an aerosol with a mass median diameter of 3.5 micron. The mass median diameter of the Ohio Nebulizer aerosol was 3.2 micron whereas the Deutreband produced an aerosol with a mass median diameter of 2.0 micron.

Calibration of the light-scattering impactor with a standard Battelle cascade impactor established that the impaction characteristics of the light-scattering impactor were identical to those of a standard impactor

Figure 7 shows the calibration data obtained with the initial three-stage light-scattering impactor. In obtaining these data, the first light-scattering stage was placed at various locations within a standard six-stage cascade impactor. Stages 2 and 3 were always in the same location and indicated -1 and -0.5-micron sizes. Stage 2 shows a linear relationship of scattered light to mass flow. Stage 3 was the original horizontal stage purchased from NRL. The scatter of these points may be due to short test times, 3 minutes for each determination, or possibly to the fact that the large horizontal chamber did not reach equilibrium. However, the major interest of this calibration investigation was to establish the linearity of Stage 2 impactor design and to correlate this with standard impactors. This was satisfactorily demonstrated by the above work.

A Royco particle classifier was used to compare the particle counts given by the Royco, for uranine-tagged aerosols, with the outputs of the light-scattering impactor. A consistent correlation was obtained between the total count of the Royco and the reading from Stage 1 of the light-scattering impactor. However, the Royco did not count any particles with a diameter greater than approximately 2 microns, even when sampling an aerosol with 34 percent of its mass comprised of >2-micron particles.

Construction and Calibration of Manual Seven-Stage Field Instrument

The three-stage unit was revised mechanically, optically, and electronically for development of the field instrument. These changes produced stages of equal sensitivity, proportional response of all four ranges, and reproducible operation.

Mechanical looseness of the light source (lamps) was corrected by a change in the socket shape. Movement of the light source in the original unit contributed to erratic data.

A new optical filter screen was made to conform to the NRL specifications. This screen transmitted approximately 5 percent of the light and could be used only for the 100 percent range adjustment.

Changes made to the electronic system provide a continuous scale of four ranges from zero to 0.1, 1, 10, and 100 percent. The original unit had only three ranges, with no provision for a 10 percent range. A more precise balancing adjustment of the output from the photomultiplier tubes was accomplished by installing a potentiometer in the input to each photomultiplier tube. This provided better control of response than the previous potentiometers used only on the output of each tube.

After satisfactory operation of the preliminary three-stage light-scattering impactor, work was started on design of a field unit.

The Optics and Advanced Electronics groups at Battelle were consulted to establish refinements that would improve the field unit. Many changes were suggested that could produce a better and more sensitive field unit. However, at this time it was advisable to carry out only those refinements suggested for improving the laboratory model. In the future, after the field unit has been evaluated, some of the more advanced and sensitive devices may be incorporated in a later model.

Design Characteristics

Figure 8 is a photograph of the manually operated field instrument. This field unit of seven stages was designed incorporating a venturi in the sampling train at the light-scattering point in each stage. This venturi effect plus the addition of cone shapes in the light-scattering chamber reduced the amount of sample that passed into stagnant regions of the light-scattering chamber. It was decided that this stagnant region should be left open to the sample to eliminate the difficult problem of cleaning a proposed glass-enclosed sampling-tube arrangement that would be required to direct the sample through the focal point of the light-scattering chamber. Another disadvantage of a glass-enclosed sample tube would be a required radical change in the optics of the system. Because of time and funding limitations, no radical changes in optics or electronics were undertaken in the field model. The above-mentioned cones were designed to have antireflection threads cut in the surface to eliminate stray light from these areas.

An improved light socket was designed into the system to hold the lamp rigid. The optical filter and black spot plates were redesigned to reduce weight and facilitate their movement.

Camera shutters were incorporated in the light train to provide a positive cutoff of light to the photomultiplier tubes so that their operation could be continuous, thereby establishing a more stable operating condition. Only during recording of scattered-light intensity from a stage would the shutter be open. Also, it was envisioned that the iris adjustment on these shutters could be changed to alter mechanically the amount of light that each photomultiplier tube sees. This control could be used to balance or equalize all photomultiplier responses.

The electronic system was altered to provide for a seven-stage operation and to control individually both the input and output of the seven photomultiplier tubes. A constant-voltage transformer of greater capacity was used to handle seven lamps at one time. The lamps were on all the time, since off-on operation would reduce the life and uniformity of the bulb response. In future models a more elaborate electronics system may be warranted, as suggested by Battelle's Advanced Electronics Group.

The overall height of the field unit was reduced 1.3 inches per stage as compared with the preliminary unit. Practically all the parts were made of aluminum to reduce weight, except original purchased brass pieces. These aluminum parts were anodized and blackened to minimize reflection. A separate stand was made with seven supports to hold the seven stages when servicing slides and performing other operations.

A switch box containing a four-deck, eight-position switch and necessary electronics to zero and range seven-stage systems was designed and built. This was completely wired to provide an indicator white light for each stage and a red light to indicate when the optical filter should be in place. Outlets were also wired so that, in the future, they could be used for automatically operating solenoids to actuate shutters and optical filters in each stage of the light-scattering unit.

After the unit was assembled, it was run continuously during the working day for about 4 days while zeroing, and various ranging methods were tried. During this time, the lamp electric-contact springs in three stages became too hot and lost tension even though the stages were crisscross, as apparent in Figure 8. New lamp holders were made incorporating electrical connections directly soldered to the bulb. However, bulbs continued to burn out, probably because of overheating. A Sorensen MVR 500 constant-voltage regulator was utilized to light all seven lamps. With all seven lamps operating, the voltage was 5 Vac.

Calibration

Various impaction slides (e.g., glass and porous steel) were tried to establish their relative merits. It was decided that a porous slide was needed to hold droplets for extended periods of automatic recording of fog conditions.

Figure 9 shows the comparison of three different types of impaction-slide arrangements in the standard impactor and the light-scattering impactor.

Establishing these comparisons was of value because of the calibration technique. The calibration was accomplished by incorporating a tracer in the aerosol droplets. In order to use this technique, it was necessary to remove all the tracer from the slides. Glass slides were used because with glass slides it is comparatively easy to remove the tracer, but porous stainless steel slides retain the tracer, making analysis difficult.

Figure 10 is the calibration of the manual field model of the light-scattering impactor. Calibration was accomplished in the laboratory by sampling water fogs generated with a Hartman whistle and solid fluorescent aerosols generated by aerosol packages. These aerosols had mass mean particle diameters of 6 and 0.5 microns, respectively. Calibration with the Hartman whistle was accomplished by adding a known amount of fluorescent dye (uranine) to a measured amount of water before atomizing. The solution was atomized in a chamber that contained air saturated with water vapor. The chamber aerosol was sampled with a regular Battelle cascade impactor and with the light-scattering impactor. A fluorescence meter measured the amount of dye collected by each impactor stage. During the calibration, a simple method of zeroing and ranging the seven stages was developed. This method consisted of passing filtered air through all stages and zeroing by adjusting the voltage input to each photomultiplier tube. A clear lucite strip was placed in the sample and light stream to range the instrument. With the lucite strip in place, the maximum-range indication was set at 100 on the 100-scale of the meter when the optical-filter plate was not in place.

Field Study

Two field trips were made to Otis Air Force Base, Falmouth, Massachusetts, in the summer of 1963 to check the operation of the light-scattering impactor in fog. During the field trips it was soon determined that the maximum fog light-scattering reading would be about 1/100 of our maximum range setting with the optical-filter plate in place. With this information, it became apparent that the optical filter would not be required. Since the above ranging method required that all stages be separated from each other in order to insert the lucite strip, a second method of ranging was devised. With this method it was feasible to zero and range without disturbing the stages. The procedure consisted of using filtered air to zero all stages by adjustment of voltage input to each photomultiplier tube, and then ranging Stage 1, using the lucite strip, at 100 on the 100 percent scale by adjustment of photomultiplier output. Stage 1, at the top of the stack of stages, was the only stage accessible for insertion of the lucite strip (without disassembling). Therefore, the other stages were ranged by adjustment of the photomultiplier output to the same value as indicated by Stage 1 while cigarette smoke in the room was drawn into the system through a selective 5-micron filter. Stages 6 and 7 required a reduction in flow (25 inches on vacuum gauge) before the entire sample would pass through these stages without collecting on the slides. Then the ranges were set the same as indicated by Stage 1.

Field work indicated that, with fog, the meter output was approximately double that prior to fog formation. During one dense fog, Stage 1 output increased by a factor of four. During the field work there were only five periods of fog and three periods of mist and fog combined. Hence, data collection was limited.

The light-scattering impactor was compared with other instruments (capillary collector and J-W liquid-water meter) only near the end of the field trials when just mist or wet fog was present. The light-scattering impactor followed trends of the J-W meter more closely than it did the capillary collector, which suggests that large droplets either were not sensed or did not pass into the light-scattering chambers. However, at times a much higher reading was obtained than was indicated by the capillary collector. This reverse trend may be the result of air bubbles that collected in the capillary system most of the time.

During the first month of field work, the light-scattering impactor gave a steady reading on all stages. However, on Stages 6 and 7, meter indications were too low to be read satisfactorily. Therefore, the instrument was made more sensitive for the second field trip in September, 1963. Unfortunately, there was no fog to check the response. However, when sampling outside air, the response pulsated and was estimated at four times the original sensitivity. This estimate was made by comparison of the indications before and after sensitivity changes of outside air and also the increase associated with airplanes taxiing down the adjacent runway. Although the indicator circuitry was capable of much more sensitivity, all seven stages could not be zeroed and ranged at greater sensitivity, probably because of stray light. Also, it appears that photomultiplier differences are sufficient to make calibration tedious at higher sensitivity. It also became apparent that the slightly different photomultiplier tubes aged differently.

Two major difficulties with the light-scattering impactor in the field were failure of the 50-candlepower bulbs and the tedious work of zeroing and ranging all seven stages. A short laboratory study indicated that heat was a cause of early failure of the 100-hour-rated 50-cp bulbs. A solution was believed to have been found during the September field work with the installation of 32-candlepower bulbs, which have a rated life of 200 hours. During the final week of operation there were not bulb failures Another major problem was that the forward-light-scattering impactor appeared not to see droplets estimated to be larger than 100 microns. A minor problem of pulsations that are amplified by the more sensitive circuit should be resolved. It is believed that these pulsations can be reduced by a dampened circuit or by focusing the light scattered from a larger sample.

Investigation of 45-Degree Light Scattering

After the instrument was returned from the field, light scattering at an angle of 45 degrees, as suggested by Dr. J. R. Hodkinson, at that time of the School of Medicine, University of Rochester*, was investigated.

In this short study, three light-scattering conditions were compared. (Forward light scattering is defined as 0 degrees). The conditions investigated were light scattering from 45 degrees, 90 degrees, and nearly forward light scattering of about 20 degrees. It was advisable at this stage to determine the benefits associated with higher sensitivity and linear relation for large and small particles for these light-scattering angles. It was also advisable to determine whether the various arrangements could be readily incorporated in the present seven-stage light-scattering impactor. Figure 11 is a sketch showing the angles investigated.

For this work, three fiber optics of incoherent bundles, 1/8-inch in diameter and 18 inches long, were obtained from the Bausch and Lomb Company. The bundles are composed of 10-micron-diameter fibers. A 3-foot length of fiber transmitted slightly more than 50 percent of the light entering the fiber bundle. Light was emitted from the fiber bundle at about a 20-degree cone angle. This angle made it advisable to use a lens

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* Now at Virginia State College, Petersburg, Virginia.

to focus the light where needed. An optical bench was used to determine a satisfactory lens arrangement that would accommodate the 45- and 90-degree light-scattering angles of interest. Simple lenses were not satisfactory in the envisioned application. However, duplex and triplex lenses were satisfactory and eliminated troublesome diffraction rings.

A stage of the original three-stage light-scattering impactor was revised to accommodate fiber optics and both 45- and 90-degree light-scattering angles. The results of trials showed that transmitted light was only 0.07 of that emitted from the present forward-light-scattering impactor arrangement. This great difference results in part from the fact that with 45- and 90-degree scattering only a small conical section is viewed, while with forward scattering the entire conical toroid is observed.

Because an entirely new and better lens system would be required for all seven stages, the use of 45- and 90-degree light scattering was not considered economically feasible. Based on this limited investigation, no advantages were apparent during studies at these angles, as sensitivity was less and mass indication was no better than for forward scattering, even though less background light is associated with 90-degree angles. However, fiber optics was satisfactory when used with the present forward-light-scattering impactor.

Improved Automatic Field Instrument

Improvements included the use of single lamp and single photomultiplier tubes with the aid of fiber optics in conjunction with a mechanical motion to index the appropriate stage in sequence. Electronic improvements included removing the four ranges and incorporating a printout device that monitored the outputs of: (1) each stage; (2) a calibrated light source; and (3) the instrument's dark current. Figures 1 and 2 show the various parts of the fully automatic light-scattering cascade impactor.

Fiber Optics

Fiber optics were studied to reduce the number of lamps and photomultiplier tubes required. The seven lamps and seven photomultiplier tubes caused considerable practical trouble during field work at Otis Air Force Base because as a lamp would burn out realignment of a new lamp and optics was required to zero and range the stage. Other difficulties were apparent because individual photomultiplier tubes and individual lamps were slightly different and were aged differently.

Fiber optics of 1/8-inch diameter and 18-inch length of Bausch and Lomb's latest design were used. However, fabrication of the cycling device was finished before the fiber optics were received. These fiber optics had to be returned as they did not meet specifications and would not fit. When the requested fiber optics arrived their ends were not cut and polished. Because time was running short they were used as received.

Cycling Mechanism

An eight-station Geneva Motion and a 3-rpm motor were arranged to move the lamp in front of the seven fiber optics and, at the same time, the arrangement oriented the photomultiplier in front of corresponding light-pickup fiber optics. The eighth position was a blank where no light reached the photomultiplier; hence, only the instrument electronic dark current was recorded. Figure 2 shows this cycling mechanism.

The fiber optics were sealed and positioned by rubber sleeves that fastened onto the light-scattering units. At the lamp end, the fiber optics were retained by snap rings. At the photomultiplier end, both O rings and snap rings were used to seal and retain fiber optics.

Photomultiplier

Both 931A and 1P21 photomultiplier tubes were used. Because of lack of funds and time, it was decided to complete calibration of the instrument with the 931A photomultiplier tube. This tube was less sensitive to light saturation and could stand ten times the anode current. Hence, it was more readily incorporated in the instrument, but at a sacrifice of sensitivity.

Lamp

The lamp source initially used, 50- and 32-candlepower spotlight bulbs, was replaced by one 8-volt, 50-watt Phillip bulb, made in Holland. This projector bulb, which has a reflective coating on the outside, has a built-in lens. Since the bulb's rated life is short, to increase bulb life only 6 volts were applied to the filament. The bulb at this voltage was operated continuously for 6 weeks without showing signs of failure. However, during intermittent operation the reflective coating cracked. To correct this condition the bulb was wrapped with aluminum foil.

Electronics

The electronics of the old instrument was revised and made as simple as practical to eliminate the previous four ranges. A digital voltmeter (Hewlett Packard No. 405 CP dc meter) was connected in their place to provide a full range. Also, clips for resistors were placed in the stray-light circuits. These could be used with appropriate one percent resistors to take the place of optical filters by reducing sensitivity without overloading the photomultiplier. For the sake of expediency, the resistors were not used. Instead, optical filters that reduce sensitivity were placed in front of the lamp fiber optics of Stages 1 through 5. Both Stages 6 and 7 were left in the most sensitive arrangement without optical filters.

A Hewlett Packard digital recorder No. 561 BR was used to print the voltage equivalent to each stage of light scattering. One side of the digital recorder was vacant and the light-scattering indicator was rearranged to fit into this space. A flexible shaft connected the Geneva Motion mechanism with an eight-position rotary switch in the indicator section. This switch was connected to the stray-light adjustment of each stage. As installed, the original stray-light-adjustment potentiometers result in a coarse adjustment, and therefore were not used during calibration.

Calibration

Calibration was accomplished by utilizing a nebulizer to produce droplets that contained a fluorescent tracer. Three materials were combined to make the tracer solution: (1) dibutyl phthalate; (2) MDAC tracer purchased from Carlisle Chemical Works, Inc., Reading, Ohio; and (3) Fluorol 7GA tracer from General Aniline and Film Corporation, N.Y. The solution composition was 98.5 percent dibutyl phthalate, 1.4 percent MDAC tracer, and 0.1 percent Fluorol 7GA tracer. The mean diameter of droplets was 2.5 microns. Droplets of this solution were introduced into various amounts of filtered air that was drawn through the light-scattering impactor. By using various impactor stages ahead of the instrument, it was possible to vary the size and quantity of aerosol observed by the instrument. Analyses of the collected tracer on each stage and on the absolute filter gave a measurement of the aerosol mass in grams. Since the impactor samples at a known flow rate, the results were plotted in grams per cubic meter for the various voltages associated with the forward-light scattered for each stage. There was no radical difference in light scattering between stages because all stages see the smallest droplets.

In operation, Stage 0 is not a light-scattering stage and is used to remove the plus-16-micron sizes. Stages 1 through 7 are light-scattering stages. Stages 1 through 6 indicate particles of -16, -8, -4, -2, -1, and -0.5-micron sizes, respectively. Stage 7 is a reference used to indicate the condition of the light source and photomultiplier. Stage 8 permits no light to reach the photomultiplier; therefore, it is a reference stage that indicates both the electronic dark current of the instrument and the photomultiplier condition. The indications of all eight stages are recorded every cycle of 2 minutes and 4 seconds' duration.

Figure 3 shows the calibration curves for each stage. The voltage produced by light scattering is interpreted in terms of grams of droplets per cubic meter of air sampled. The impactor samples at the rate of 12.5 liters of air per minute.

Further Improvement of Light-Scattering Cascade Impactor

Efforts recommended to improve the light-scattering impactor, but not covered by this contract, are as follows:

(1) Polish fiber-optic ends.

(2) Investigate the optimum shape of fiber-optic ends.

(3) Record the digital-voltmeter calibration voltage at intervals.

(4) Record an indication of the voltage applied to the photomultiplier at intervals

(5) Develop a better flow pattern so that no aerosol circulates in the cone section of the light-scattering chambers.

  (a) Use, possibly, filtered-air bleed into the cone section to prevent circulation.

(b) Redesign lens system to eliminate cone sections.

(6) Modernize the electronics, including solid-state devices.

(7) Revise the system so that the photomultiplier output circuit is instantaneous instead of involving a 7-second lag.

(8) Use neutral-density or other appropriate filters if required.

(9) Investigate the use of smooth-acting stepping switches instead of Geneva Motion to move from stage to stage. It is believed that the jarring motion of some present-day stepping switches would not be compatible with the presently arranged lamp or photomultiplier tube. However, with the use of stepping switches, any sequence of stages could be recorded.

(10) Investigate fast scanning of stages with either direct printout or memory storage and then printout. It would be necessary to devise a fast-responding circuit for this application.

(11) Incorporate in the instrument one or more stages where the maximum light-scattering angle could be determined and recorded. Since the angle of maximum light given off varies for different materials, the maximum light angle would distinguish between droplets and dust or various nuclei material.

CONDENSATION-NUCLEI COUNTING

Small particles less than 1 micron in diameter in the atmosphere serve as nuclei for condensation of water in the formation of fog. However, their specific role has been illusive because instrumentation to monitor the concentrations of nuclei of different sizes has not been available.

The work described in this section was aimed at devising a laboratory instrument that would record the concentrations of particles in several size ranges smaller than 0.1 micron, based on an electrostatic-precipitation principle. Particles between 0.1 and 1 micron, which also serve as condensation nuclei, are not measurable by this technique.

During this program, an apparatus was assembled to charge and precipitate condensation nuclei in such a way that the amount of current accompanying the precipitation would depend on the concentration of nuclei. Although a relatively high background current was observed, presumably due to small ions not completely removed by an ion trap, the general approach was shown to have merit.

Two alternatives are possible. All nuclei can be charged to the same polarity, an approach that should give the instrument high sensitivity. The laboratory apparatus was constructed on this basis. However, interfering background-ion current was encountered. On the other hand, nuclei can be brought into equilibrium with a controlled ion atmosphere produced by a radioactive source. Though basically less sensitive, this system might not have the difficulty with background-ion currents, and it should be investigated before a choice is made between the two systems.

Electrostatic Precipitation

Electrostatic precipitation can form the basis for an automatic instrument for measuring the concentration of different-sized condensation nuclei in the atmosphere. The characteristic of the nuclei used to differentiate between sizes would be their mobility when charged and exposed to an electric field. Small nuclei, each carrying one electronic unit of charge, would move to a collection electrode more rapidly than similarly charged large nuclei. This effect can be used to cause different sizes of nuclei to collect on different electrodes in such a way that the electrical current from each electrode is a measure of the concentration of nuclei of a particular size.

In such a device it is important that all of the nuclei be charged, or at least that a known fraction be charged. This is not true of nuclei in the atmosphere; many are uncharged, and the relative numbers charged to positive and negative polarities vary widely. Accordingly, it is necessary to treat the nuclei in some manner to bring them to a known state of charge.

Two states of charge are desirable for the design of nucleus counters: (1) all of the nuclei are charged to one polarity; or (2) some are charged positively, some negatively, and some are uncharged, with the relative number in each group known and not varying. Monopolar charging of the condensation nuclei should make possible an instrument of higher sensitivity, but a special type of charging unit would have to be used to avoid deposition of the nuclei in the charging zone. Bringing the nuclei to a known distribution of charges would lead to an instrument having lower sensitivity, but a simpler charging unit could be used.

Both instruments would have two parts, a charging section and a precipitation section, but they would be quite different for the two approaches. The charging section to achieve monopolar charging could be a duct through which air passes as it moves toward the precipitation zone, the duct containing corona wires or other arrangements to provide a supply of ions of one polarity that would charge condensation nuclei by a diffusion-charging process. In the second approach, bipolar charging, the charging zone would be a chamber through which the air passes slowly and is exposed to both positive and negative ions produced by alpha- or beta-particle showers. The concentration of ions would be high enough that the nuclei would reach an equilibrium distribution during their time in the charging zone.

The precipitation section in both cases would consist of a laminar-flow duct of rectangular cross section with top and bottom plates being formed from pairs of electrodes for imposing a field and receiving charged nuclei of various mobilities. In the bipolar approach, the quantity measured to indicate concentration of nuclei of a given size would be the total current flowing to one of a pair of electrodes. The minimum concentration detectable would be determined by the background current flowing between the two electrodes due to ions produced in the duct by cosmic rays and radioactive contaminants and by leakage currents in the insulators supporting the electrodes. In the monopolar approach, the total amount of current flowing to each pair of electrodes would serve as the measure of nucleus concentration, and ions produced by cosmic rays or radioactive contaminants or leakage currents would not appear as background current. Thus, this approach should have much higher sensitivity. Because the higher sensitivity promised to ease several practical problems, it was decided to construct an instrument based on monopolar charging. Unfortunately, unexpected side effects encountered with the monopolar-charging device seriously interfere with the measurement, and in future work it would be well to reconsider the choice of approach.

The possibility of measuring concentrations of several sizes of condensation nuclei in this type of instrument depends on the ability to separate the different drift paths for nuclei having different mobilities. This separation could be made most effectively if the condensation nuclei are injected into the precipitation duct in a thin lamina of air, the rest of the air flowing into the duct being nuclei free. However, even with the duct filled with air containing charged nuclei, a reasonable separation into three sizes of nuclei can be achieved. Since the larger nuclei have the lowest mobility, they are the most difficult to precipitate. Most of the effort on this project was concerned with devising techniques for measuring concentrations of such larger nuclei. That done, design of an instrument to measure three ranges would be relatively straightforward.

Basic Instrument Concept

In its simplest form, the condensation-nucleus counter would consist of a duct through which the air to be sampled would be passed, the duct containing a charging section and a precipitation section. As indicated previously, the charging section would expose the condensation nuclei to ions of one polarity produced in a corona discharge, so that the nuclei would become charged by an ion-diffusion process. To differentiate between different-sized nuclei, all nuclei of a given size should be charged to the same level, preferably one electronic charge per nucleus.

The precipitation section of the duct would consist of a series of electrodes to provide the field to precipitate the charged nuclei. The electrodes also would be connected to sensitive current-measuring devices, so that the current produced by the flow of charged nuclei to the electrodes would be a measure of the concentration of nuclei in each size or mobility range. The number of size ranges that can be resolved would depend on (1) the noise level and drift rate of current-sensing devices (electrometers), (2) the noise level of the precipitation section of the duct, (3) the volume of air to be sampled, and (4) the minimum concentration of nuclei to be detected. It appears that a reasonably sized instrument could measure nuclei concentrations in three size ranges, less than 0.01 micron, 0.01 to 0.03 micron, and 0.03 to 0.1 micron. Particles or nuclei larger than 0.1 micron should be removed by an impaction stage, otherwise they would lead to ambiguous results because they have mobilities duplicating those of smaller nuclei.

Precipitation Section

One of the major difficulties in measuring small currents resulting from the precipitation of charged condensation nuclei is that in general the precipitation duct also acts as a conventional ionization chamber, and the current measured is a combination of nuclei current and background-ionization current. By going to monopolar charging of the nuclei, it is possible to use an arrangement that largely eliminates this ionization-chamber effect and also reduces the problems associated with adequately insulating the precipitation electrodes.

Figure 12 is a sketch of this arrangement that shows the negative electrodes forming the main duct of the precipitation section with a positive electrode inside and a grounded shield outside. Positively charged nuclei will be precipitated on the outer electrodes and their charges will produce a reading on the electrometer. However, an alpha-particle moving through the region between the electrodes (a frequent occurrence in any enclosure of a size practical for this work) will produce large numbers of positive and negative ions, but these will deposit on the inner and outer electrodes in equal numbers so as to produce no reading on the electrometer.

Similarly, the insulators supporting the inner electrode maintained at a high potential can be "leaky" without producing any effect on the electrometer. The chief source of background current will be ion flow and insulator leakage between the outer electrodes and the grounded duct; however, the potential difference between these two surfaces is never more than a small fraction of a volt, so the background current can be kept small.

Initial trials of precipitation sections of this general design showed that nuclei currents as small as 10^{-14} ampere could be measured using a vacuum-tube electrometer, such as the Keithley 600A. This current would be equivalent to about 63,000 nuclei per second, each carrying one electronic charge, depositing on the electrodes. Assuming that the measurement of concentrations in the neighborhood of 630 nuclei per cubic centimeter would be adequate in an instrument of this type, a volume flow rate of 100 cm^3/sec would be required.

To collect nuclei between 0.03 and 0.1-micron diameter (the most difficult to collect), the largest fields possible should be used. However, a self-contained (not line-powered) voltage supply is essential. A reasonable compromise was 900 volts (three 300-volt batteries) across a 1-centimeter spacing. At a field strength, E, of 900 volts/cm, the sideways velocity of a 0.1-micron-diameter nucleus having a mobility, M, of 8×10^{-5} cm/sec/volt/cm would be:

$$\begin{aligned} v_s &= ME \\ &= (8 \times 10^{-5})(900) \\ &= 7.2 \times 10^{-2} \text{ cm/sec.} \end{aligned}$$

To keep length reasonable, the average air velocity should be low; 5 cm/sec was chosen. The length, L, would then be:

$$L = \frac{v_a}{v_s} S$$

where:

v_a = air velocity, 5 cm/sec

v_s = sideways velocity, 7.2×10^{-2} cm/sec

S = distance between electrodes, 1 cm

$$L = \frac{5}{7.2 \times 10^{-2}} = 69 \text{ cm.}$$

At a velocity of 5 cm/sec and a flow rate of 100 cm^3/sec, the cross-section area would be 20 cm^2. Two ducts, back-to-back, each 10 centimeters wide and 1 centimeter between electrodes, would have the desired cross-sectional area.

Figure 13 is a cross-sectional sketch of the precipitation zone. The two inner ducts were fabricated from sheet aluminum and lucite insulators. These ducts and the battery box attached to them were held inside the grounded outer shield with polystyrene insulators. A shielded connection was made between the duct and a Keithley 600A electrometer operating on its most sensitive ammeter scale, where full scale is 10^{-13} amp. The electrometer output was fed to a Bausch and Lomb V.O.M. recorder. The background current, which was relatively constant and approximately 1.5×10^{-14} ampere, was suppressed with the electrometer zero setting. In operation, there was no indication of the unwanted current pulses due to alpha-particle ionization.

Charging Section

The precipitation section previously described requires that all nuclei be charged to one polarity, or that if some nuclei remain uncharged, the fraction of uncharged nuclei of any given size be known and remain constant. Since condensation nuclei do not naturally have such a charge distribution, some type of charging procedure is needed. The only practical way to charge the nuclei is by means of small ions, and the easiest way to get ample quantities of small ions of one polarity is by a corona discharge.

The charging of submicron particles such as condensation nuclei by ion diffusion or other processes that may be active when such particles are exposed to a corona discharge has not been studied well enough to predict the results that will be obtained for any given set of conditions. It seems certain that there must exist values for the parameters of time, ion concentration, and field strength, such that essentially all condensation nuclei will become charged to one polarity. Since knowledge of what these values may be was lacking, it was decided to explore possible charging arrangements that it was hoped would produce useful results.

A possible difficulty with charging by means of a corona discharge is that the high fields produced would tend to precipitate out any of the smaller charged nuclei before they could be conveyed to the precipitation section of the instrument. To avoid this difficulty, charging units were designed that would produce large quantities of ions without exposing the nuclei to large fields.

Figure 14 is a sketch of the type of charging unit used in several modifications in this application. The idea here is to provide in the duct a region of high ion concentration but relatively low field strength. A vigorous corona discharge is maintained in the two chambers shown and ions diffuse through the perforated duct walls. To increase the number of ions available to charge nuclei in the central duct, an alternating potential difference is applied between the two sides of the duct. To achieve a maximum efficiency in extracting ions, a square wave potential was applied with an amplitude and frequency such that the ions would be moved a significant fraction of the duct width, but the most mobile nuclei, which have a mobility less than one-tenth that of small ions, would not be greatly affected.

One undesired effect observed with this and other corona charging devices is that the ion current from the corona wires produces an ion wind that disturbs the laminar flow of air in the duct. Reducing the corona current to low values – less than 70 microamperes for the charging section shown – and inserting air guides between the charging section and the precipitation section appeared to control this effect.

Experimental Results

The charging and precipitation sections were put together with a section of duct containing two plates arranged to produce a field across the duct and remove small ions and the smaller condensation nuclei. The plates were 10 centimeters long and were spaced 2 centimeters apart; a potential difference of 90 volts was maintained between them. This ion trap was designed to remove all small ions plus nuclei having mobilities greater than about 10^{-2} cm/sec/volt/cm, equivalent to nuclei about 10^{-2} microns and smaller in diameter. In this experimental arrangement, all larger nuclei would be collected in the single-precipitation section. If favorable results were obtained with this arrangement, the next step would be construction of a three-range nucleus counter.

The input of the device was fed room air pumped with a diaphragm pump to roughly 2 atmospheres pressure, and passed through a critical flow impactor with a cutoff near 0.1 micron. Provision was made to recirculate air through the arrangement so that some indication would be gained of the fraction of the nuclei removed in a single pass. This was done because it was feared that limiting the corona current to avoid ion-wind turbulence would prevent charging of all the nuclei in one pass.

As is often the case when working with extremely small currents, numerous practical difficulties were encountered in putting this laboratory arrangement into operation. Most of these difficulties were overcome and will not be recounted here. However, one relatively serious problem casts its shadow over the general monopolar approach and it will be discussed.

Figure 15 is a reproduction of the recording for a run made when the device was working well. When the air flow was begun but the charging section not operating, the electrometer reading remained near zero. When the charging section was turned on, the current rose steadily and leveled off. A valve was then turned so that the air was recirculated. The electrometer reading promptly dropped noticeably and again leveled off. This result was unexpected. It was expected that when the air was recirculated the reading would drop slowly, exponentially, indicating that a fraction of the nuclei present were being charged with each pass until finally all of the nuclei were precipitated. Instead, the fact that the reading leveled off a second time indicates that charge was reaching the precipitation section in some other form in addition to charged nuclei. Although not definitely established, it is presumed that this additional charge is in the form of ions not removed by the ion trap.

Although the mobility of small ions is often given as a single value in the neighborhood of 2 cm/sec/volt/cm, the movement of ions through the air is a random process and some few ions will lag behind the majority. It is believed that these ions, far out on the tail of the distribution curve describing the rate of movement of a large number of ions, produced the continuing high electrometer reading.

The possibility of merely compensating for this high background level was considered. Tests showed that it varied somewhat from day to day. This might still be handled by frequent checks of a zero level, but two serious questions remained:

(1) Would this background ion current completely obscure readings for the normally lower nuclei content of outside air?

(2) Would not this background current be even larger for other stages added to capture smaller, more mobile nuclei?

Assuming the final current can be subtracted from the initial current, an estimate of nucleus concentration can be made. For this run, the electrometer was operated at one-tenth its highest sensitivity and a full-scale recorder reading was equivalent to 10^{-12} amperes. The difference between the readings before and after recirculating the air is about 1.4×10^{-13} ampere which, at the flow rate used (100 cm^3/sec), would be equivalent to 8900 nuclei/cm^3. Most runs that appeared at all reliable gave concentration levels in this general neighborhood or higher, which is reasonable for the type of atmosphere being sampled.

There is a possibility that variations in the design of the charging section and ion trap would reduce the undesirable background current. A long, low-field ion trap might remove more of the ions without significantly reducing the nucleus concentration. Depletion of funds prevented investigation of such variations.

Whether or not a bipolar approach would suffer a similar difficulty is problematical. It would appear that to bring positively and negatively charged nuclei to an equilibrium state might require a radioactively produced ion atmosphere only two or three times as concentrated as a normal ion atmosphere. If true, such a concentration would not be expected to produce a large background current.

Pertinent data are recorded in Battelle Laboratory Record Books Numbers 19224 and 20424 - Fog-Droplet Measurement section Number 20176 - Condensation-Nuclei Counting section.

APPENDIX I

OPERATING INSTRUCTIONS FOR LIGHT-SCATTERING CASCADE IMPACTOR

Automatic Recording

The instrument automatically records the mass of droplets in six size ranges, size being expressed as a minus quantity (e.g., -4.0-micron size). The eight light-scattering stages provide the following information:

Stage 8 - Reference stage; indicates reference voltage of 0.025. (dark current - no light reaches photomultiplier tube)

Stage 7 - Reference stage; indicates condition of lamps and photomultiplier tube

Stage 6 - Recording stage; -0.5-micron particles

Stage 5 - Recording stage; -1.0-micron particles

Stage 4 - Recording stage; -2.0-micron particles

Stage 3 - Recording stage; -4.0-micron particles

Stage 2 - Recording stage; -8.0-micron particles

Stage 1 - Recording stage; -16.0-micron particles

Stage 0 - No light scattering; removes particles above 16 microns. Since the size of particles above 16 microns would be unknown and light scattering is a function of surface area, light-scattering data for this stage, designed to remove particles larger than 16 microns, would be of little value.

Start Up

(1) Switch on voltmeter

(2) Switch voltmeter to automatic

(3) Switch on indicator

(4) Plug in lamp

(5) Put 5-micron filter on inlet

(6) Put absolute filter on 5-micron filter

(7) Plug in vacuum pump. Let run 45 minutes

(8) Press digital voltmeter-calibration button and adjust to 7.43 volts

(9) Insert banana plug in indicator Ground and "V" volt sockets

(10) Set indicator to 0.660 volt by turning coarse and fine gain knobs

(11) Place banana plug in PM and Ground sockets

(12) Plug in motor

(13) Switch on motor

(14) Switch on recorder

(15) Set recorder print at 1 (single space)

(16) Switch on recorder print.

Adjustment

(17) Recheck voltmeter calibration (7.43 volts).

(18) Check reference voltage (0.025 volt) Stage 8. Should be constant + 0.010. If not in this range after warm up:

(a) Check and correct electronics
or (b) Make compensation for difference.

(19) Recheck indicator; reading should be 0.660 volt. At this setting the photomultiplier tube, 931A, receives 1,000 volts.

(20) Adjust voltage output of Stage 7 with the instrument and vacuum pump operating to 0.250 volt by moving lamp in or out as required.

Note: For all reference readings, only absolute, filtered air should flow through the stages.

Daily Service

(21) Remove porous and glass impactor slides and wash in distilled water to remove material deposited on slides.

(22) Remove all-glass filter and note if moist; if moist, remove and clean more frequently. Drierite or other moisture-absorbing material can be used above and on the all-glass filter.

APPENDIX II

DESCRIPTION OF BASIC INSTRUMENTS - CASCADE IMPACTOR AND FORWARD-LIGHT-SCATTERING PENETROMETER

The Battelle Cascade Impactor*

The Battelle Cascade Impactor was developed between 1952 and 1956 for the U. S. Army's Fort Detrick. It has been used extensively in determining the size and amount of aerosols deposited in the respiratory tract and in calibration of therapeutic nebulizers. It is now being used in the determination of aerosol-particle size in air-pollution studies.

It was developed as a result of previous work at Battelle for the Wright Air Development Center on the effect of oil-droplet size on combustion in military jet engines. During that research certain deficiencies of the impactors then available were observed which led to the development work for Fort Detrick.

The impactor operates on the principle that particles in a moving air stream will impact upon a slide placed in their path provided the inertia of the particles is sufficient to overcome the drag exerted by the air stream that must pass around the slide.

Figure 16 is a drawing showing how the cascade impactor classifies particles of different sizes. As the moving particles approach the slide in the impactor, the larger particles approach the slide in the impactor, the larger particles impact on the slide, while the smaller particles are carried around the slide by the drag force of the air stream. Because each jet is smaller than the preceding one, the velocity of the air stream is increased as the aerosol passes through the various stages of the impactors, and a complete size classification of the particles is achieved. Submicron particles that are too small to impact on the last stage are collected on a high-efficiency filter.

The impactor is operated in an upright position and leakage between stages is prevented by O-ring seals. This impactor has a volume flow rate of 12.5 liters of air per minute and requires a vacuum source of about 17 inches of mercury vacuum.

The impactor covers the size range from submicron to about 30 microns. The cut-off size of the first stage is 16 microns, and all particles larger than 20 microns are collected on this stage. The collection efficiency of the complete cascade impactor is essentially 100 percent. The only material not removed from the air stream is the extremely small particles that manage to pass through the high-efficiency filter. The cut-off size for each stage is inversely proportional to the square root of the density of the particle. The Battelle cascade impactor was especially designed to minimize wall loss and to obtain a sharp classification of particles on each impaction stage. A calibration of the cascade impactor is not required so long as the orifice diameters and flow rate are held to a rigid tolerance.

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*Additional information on this Battelle Cascade Impactor is available in a paper by R. I. Mitchell and J. M. Pilcher entitled "Improved Cascade Impactor for Measuring Aerosol Particle Sizes", Ind. Eng. Chem., 51, 1039 (September, 1959).

The Naval Research Laboratory Forward-Light-Scattering Penetrometer[*]

The Naval Research Laboratory light-scattering device was developed in the 1940's and was described in NRL Report P-2642, Development of Smoke Penetration Meters, by H. K. Knudson and Locke White, Ens. U. S. N. R., dated September 14, 1945. Additional information is contained in a paper by Joseph K. Thompson, NRL, entitled "Determination of Aerosol Size Distributions by Jet Impactor - Light Scattering Technique". This device and subsequent models have been extensively used to evaluate the effectiveness of filters used by the armed forces.

The Naval Research Laboratory light-scattering device is shown in Figure 17. This is a forward-light-scattering instrument. The light is produced by a 6-volt, 50-candle-power automobile-spotlight bulb. Power is supplied by a 110-volt-input and 6-volt-output Solar constant-voltage transformer. The light passes through condenser lenses were a movable optical filter is used to reduce light for high range. A black spot is located on one condenser lens as shown. The black spot prevents light from reaching the photomultiplier tube unless particles are present near the light focal point at the location of optical light chopper. A lens at the photomultiplier end of the aerosol chamber aids in focusing aerosol at the optical light chopper on to the photomultiplier tube. Voltage to the photomultiplier is regulated at about 700 volts d. c. The output of the photomultiplier is indicated on a microammeter through range switches of 0.1, 1.0, and 100 percent readings. Aerosol is drawn into the chamber past the optical light chopper through a flow control and vacuum pump. The instrument has a sensitivity of about 10^4 particles per liter for dioctylphthalate aerosol of 0.3-micron diameter.

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[*] Information on the NRL light-scattering device was obtained from Mr. Neil L. Anderson of the NRL in Washington, D. C.

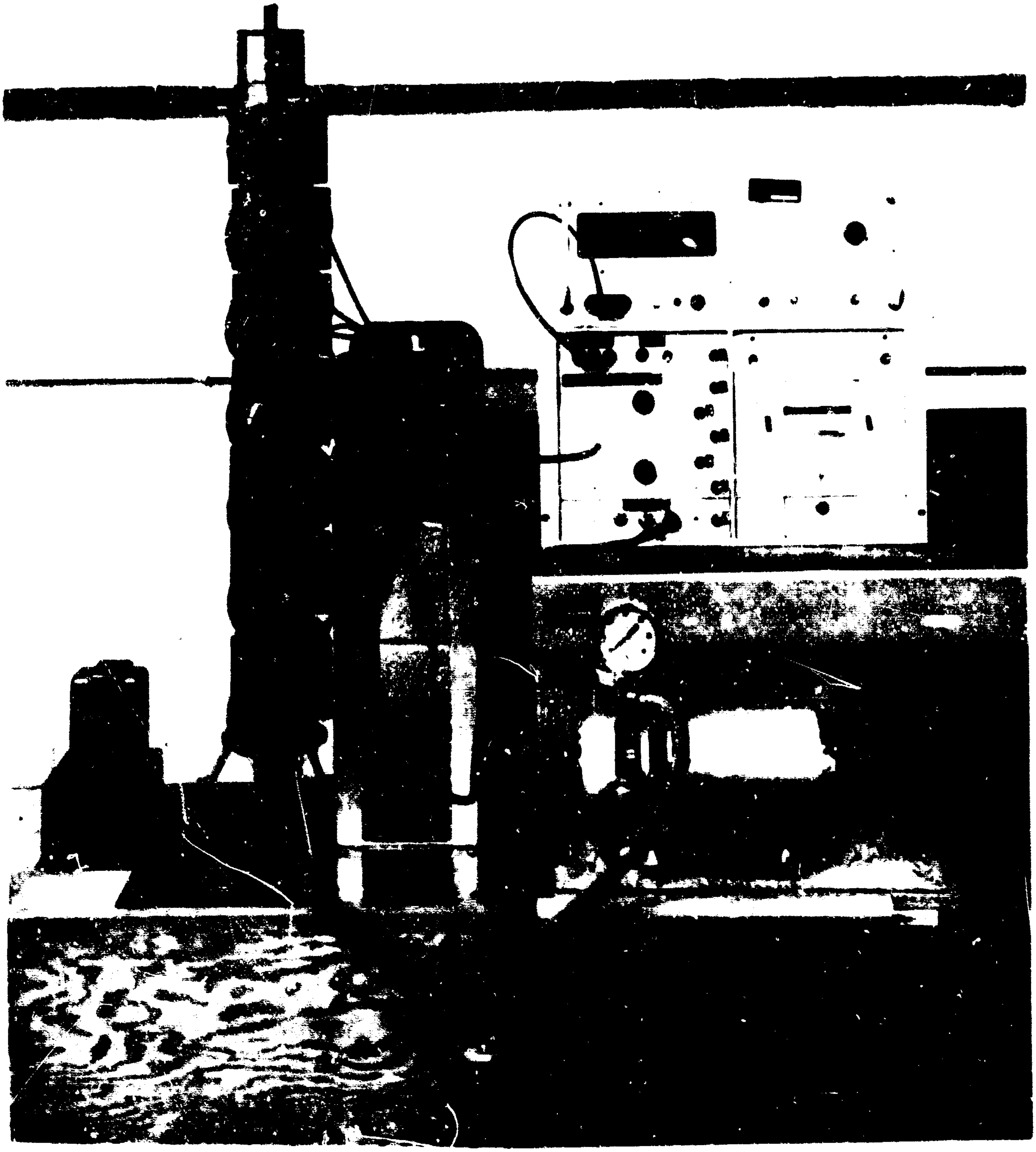

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FIGURE 1. AUTOMATIC FIELD INSTRUMENT

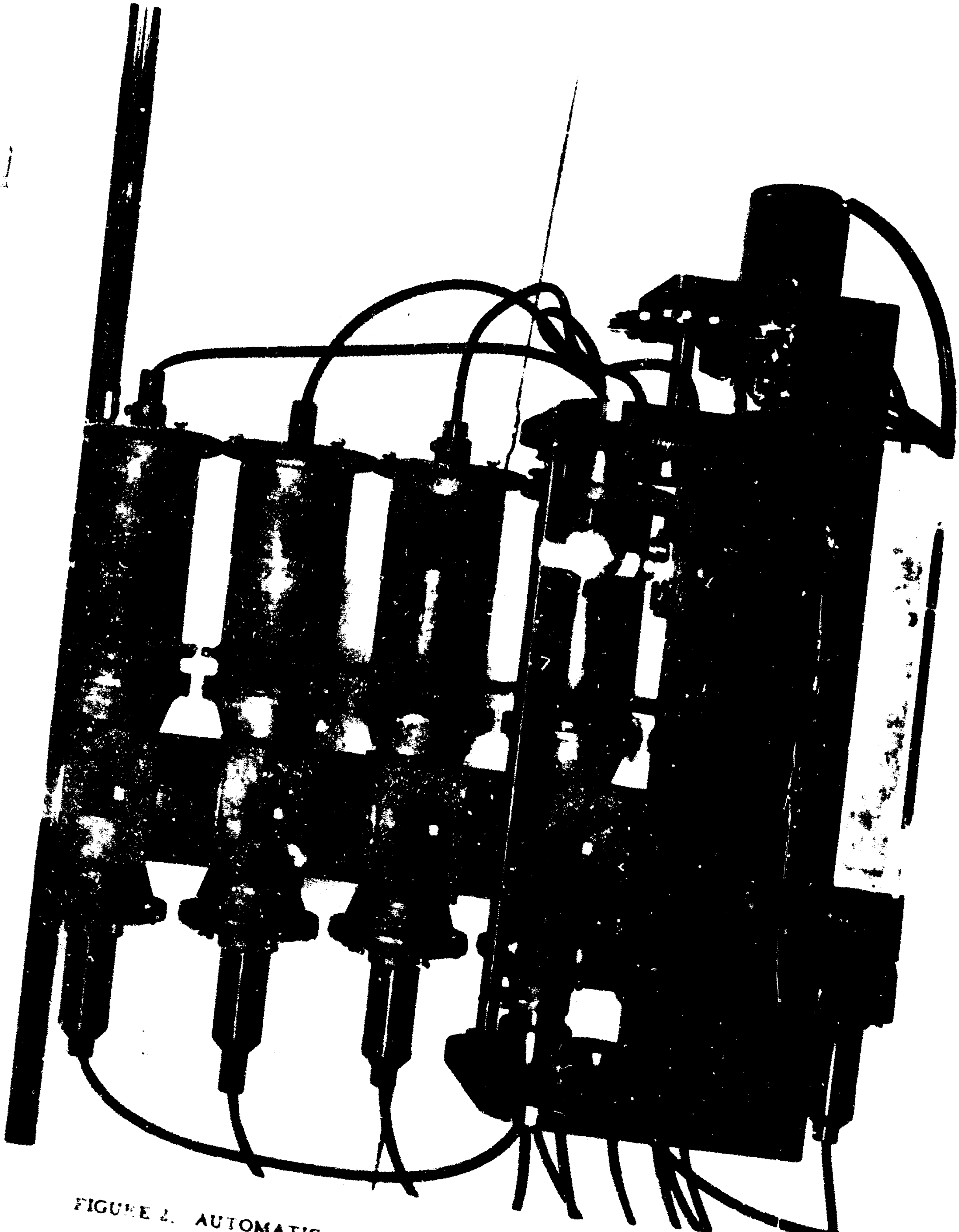

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FIGURE 8. AUTOMATIC FIELD INSTRUMENT INDEXING MECHANISM

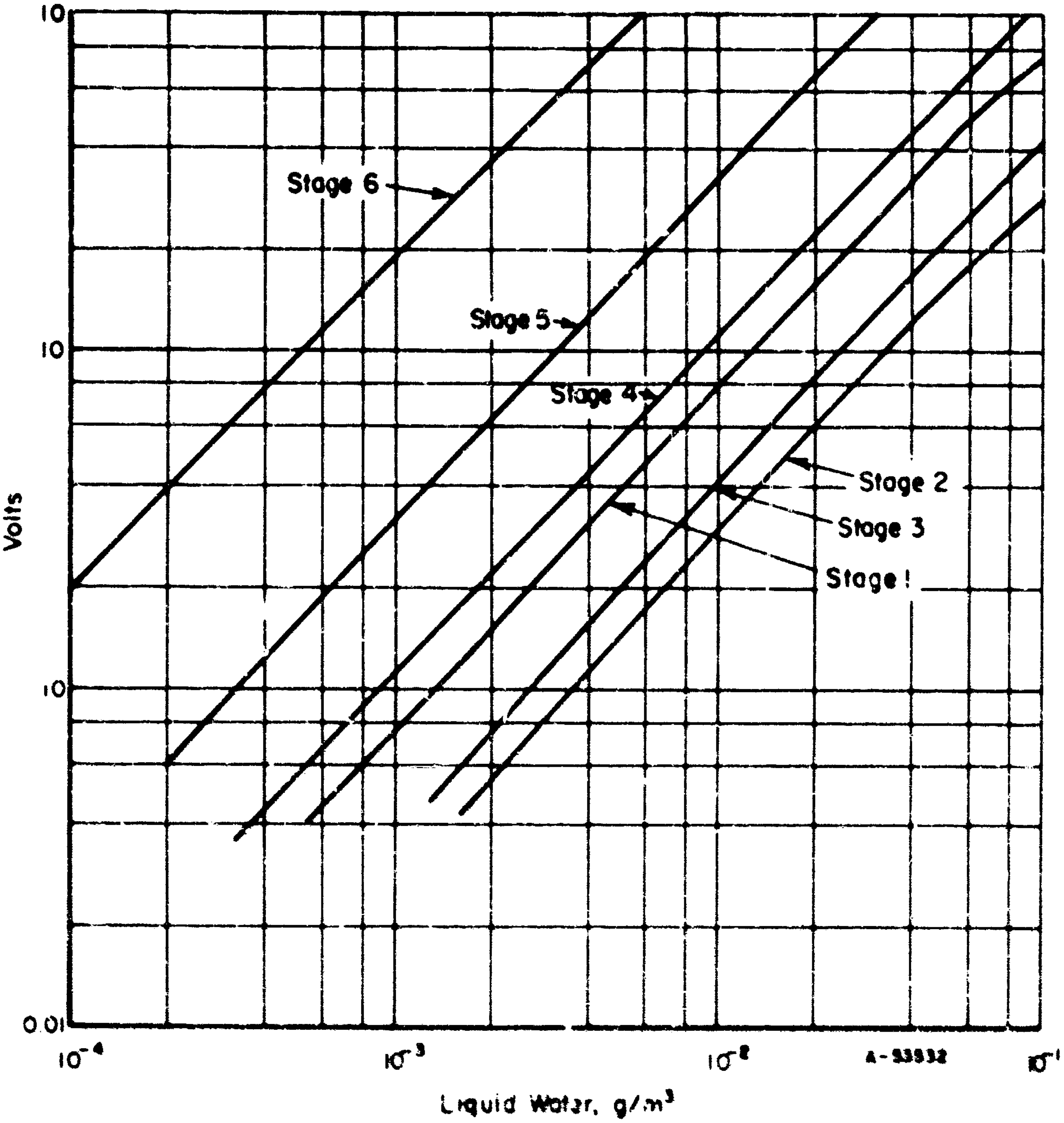

FIGURE 3. CALIBRATION CURVE FOR THE AUTOMATIC LIGHT-SCATTERING CASCADE IMPACTOR

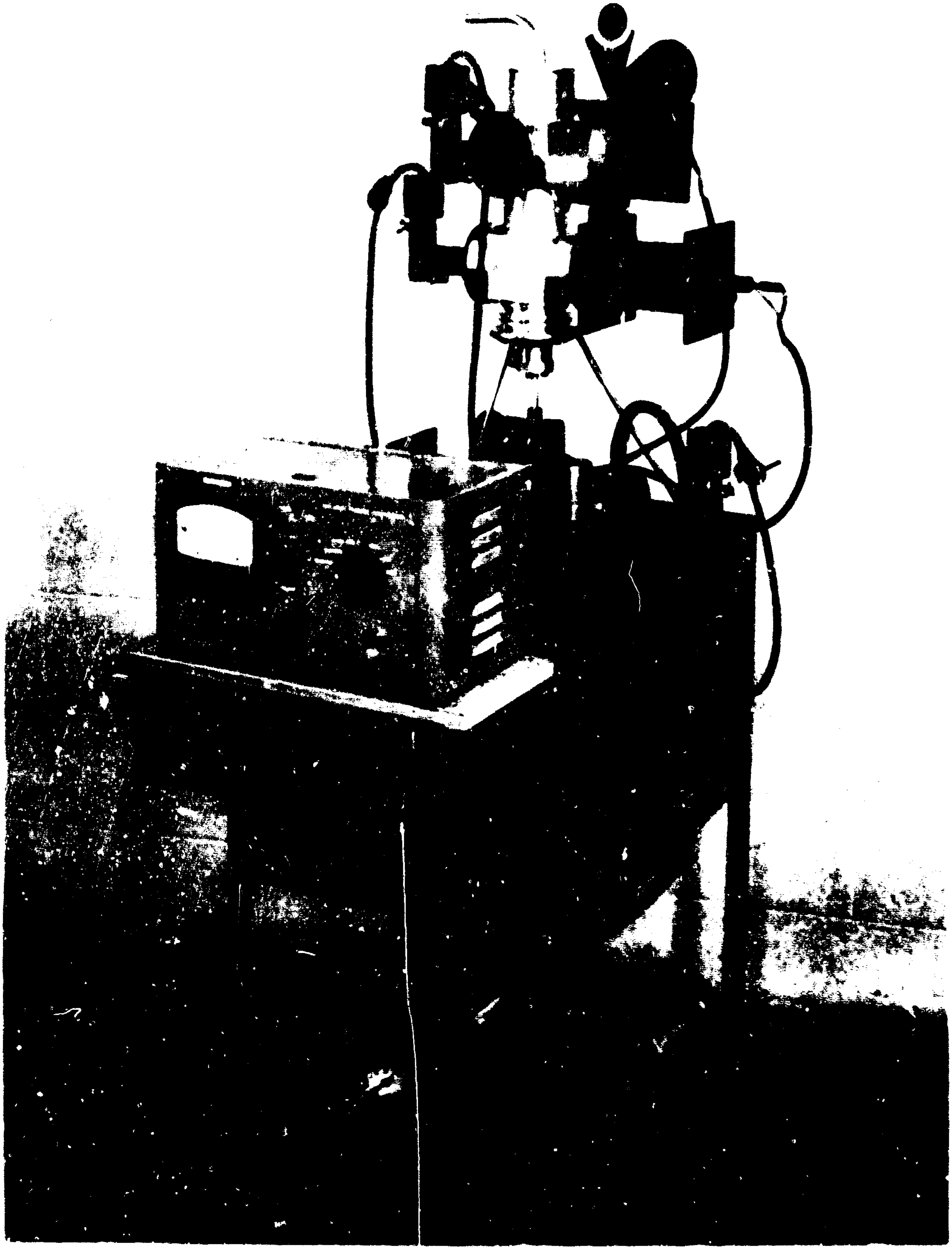

FIGURE 4. INITIAL THREE-STAGE LIGHT-SCATTERING IMPACTOR

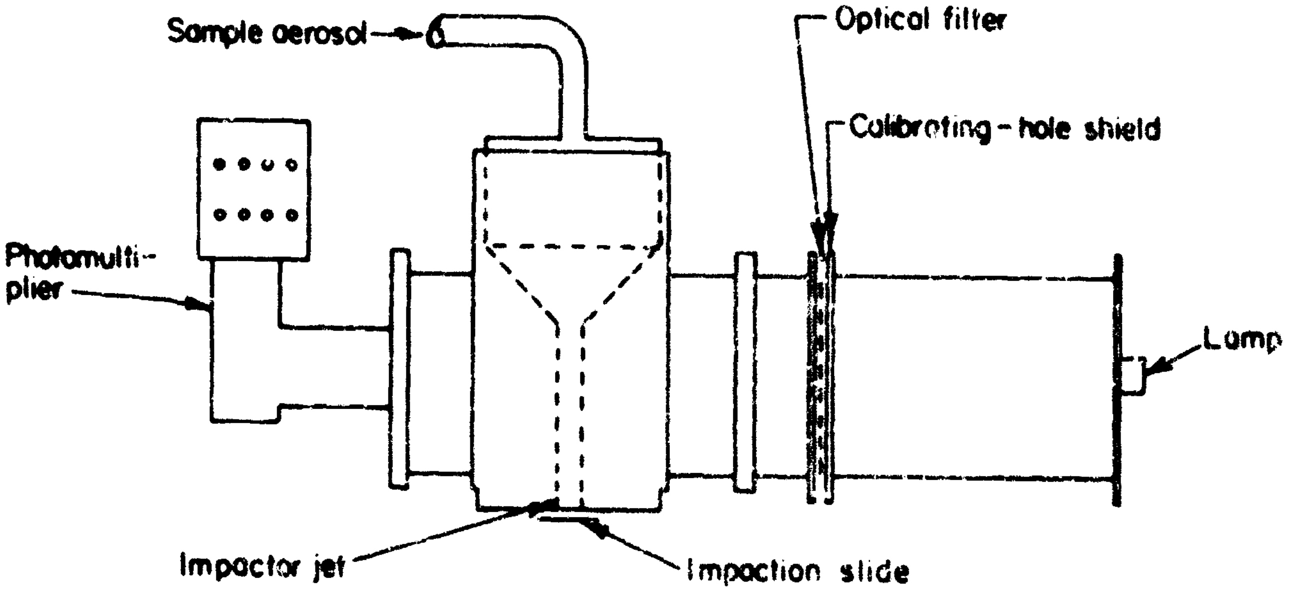

FIGURE 5. LIGHT-SCATTERING IMPACTOR STAGE

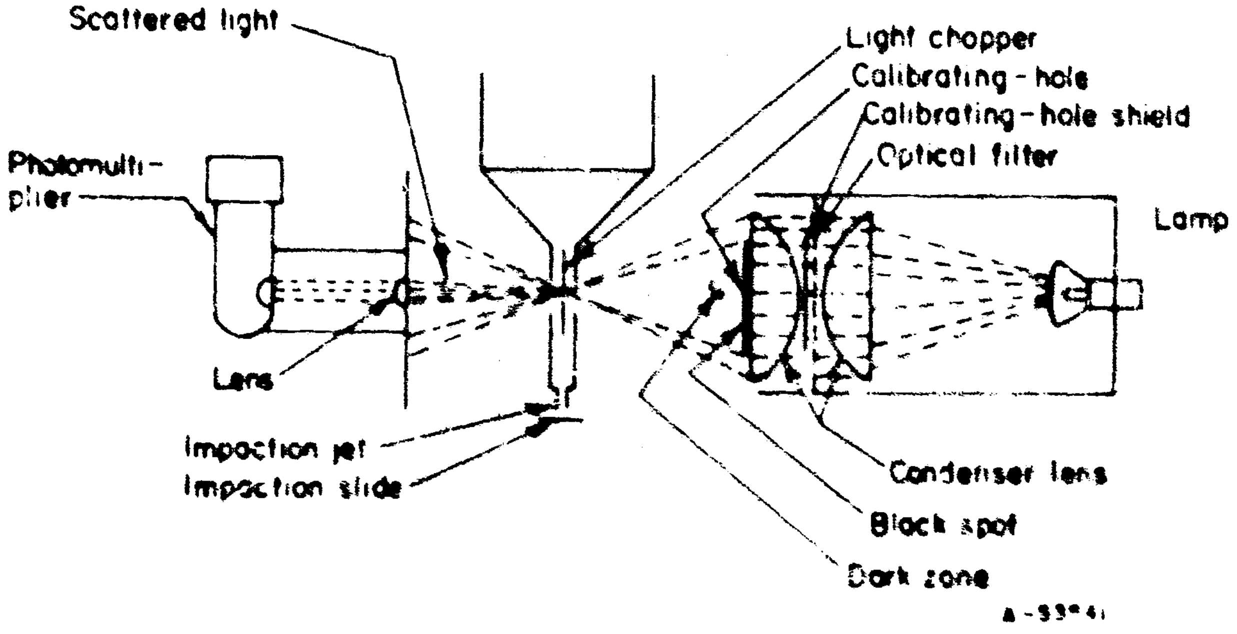

FIGURE 6. SKETCH SHOWING OPTIC ARRANGEMENT

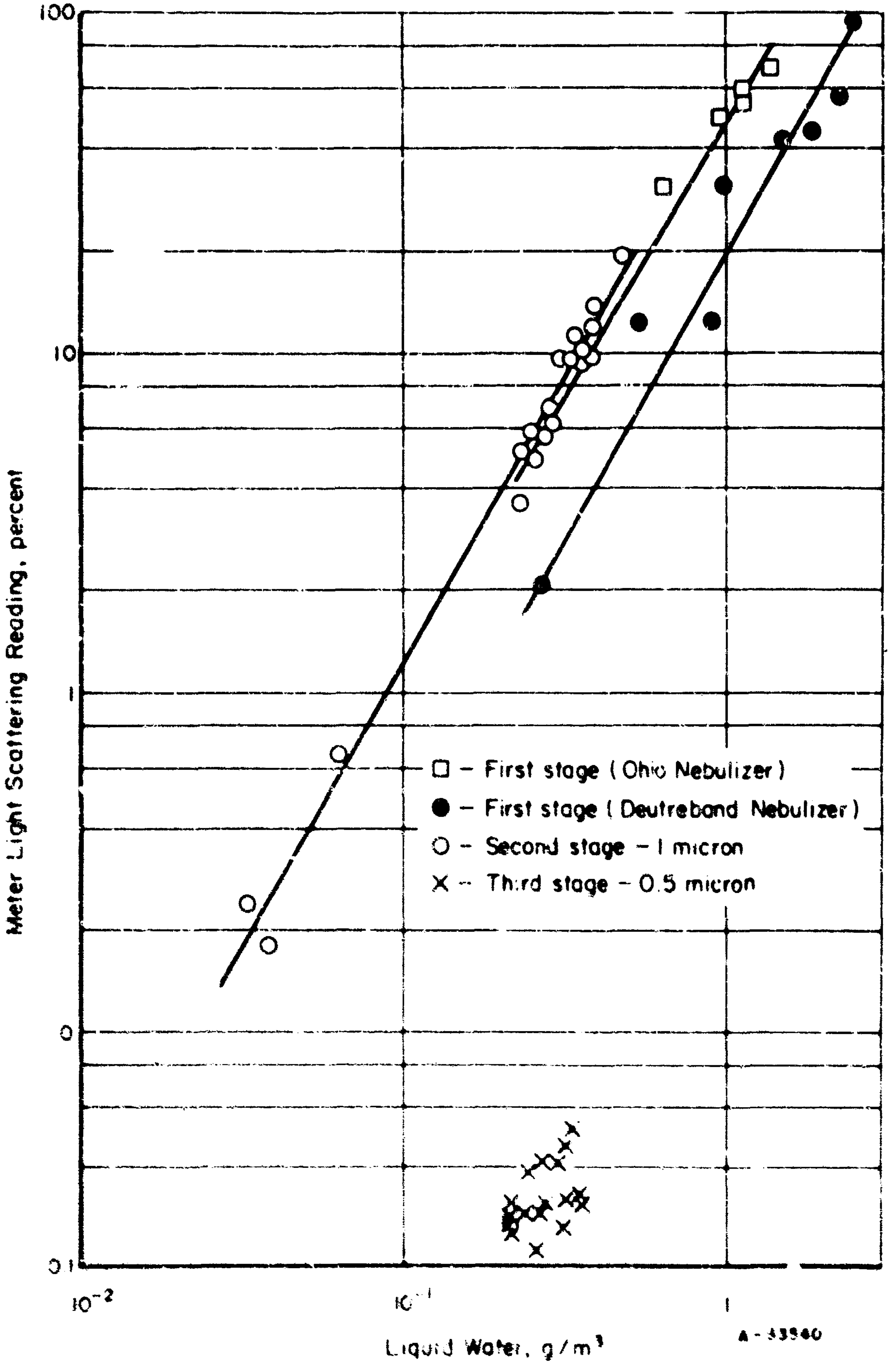

FIGURE 7. CALIBRATION INITIAL INSTRUMENT

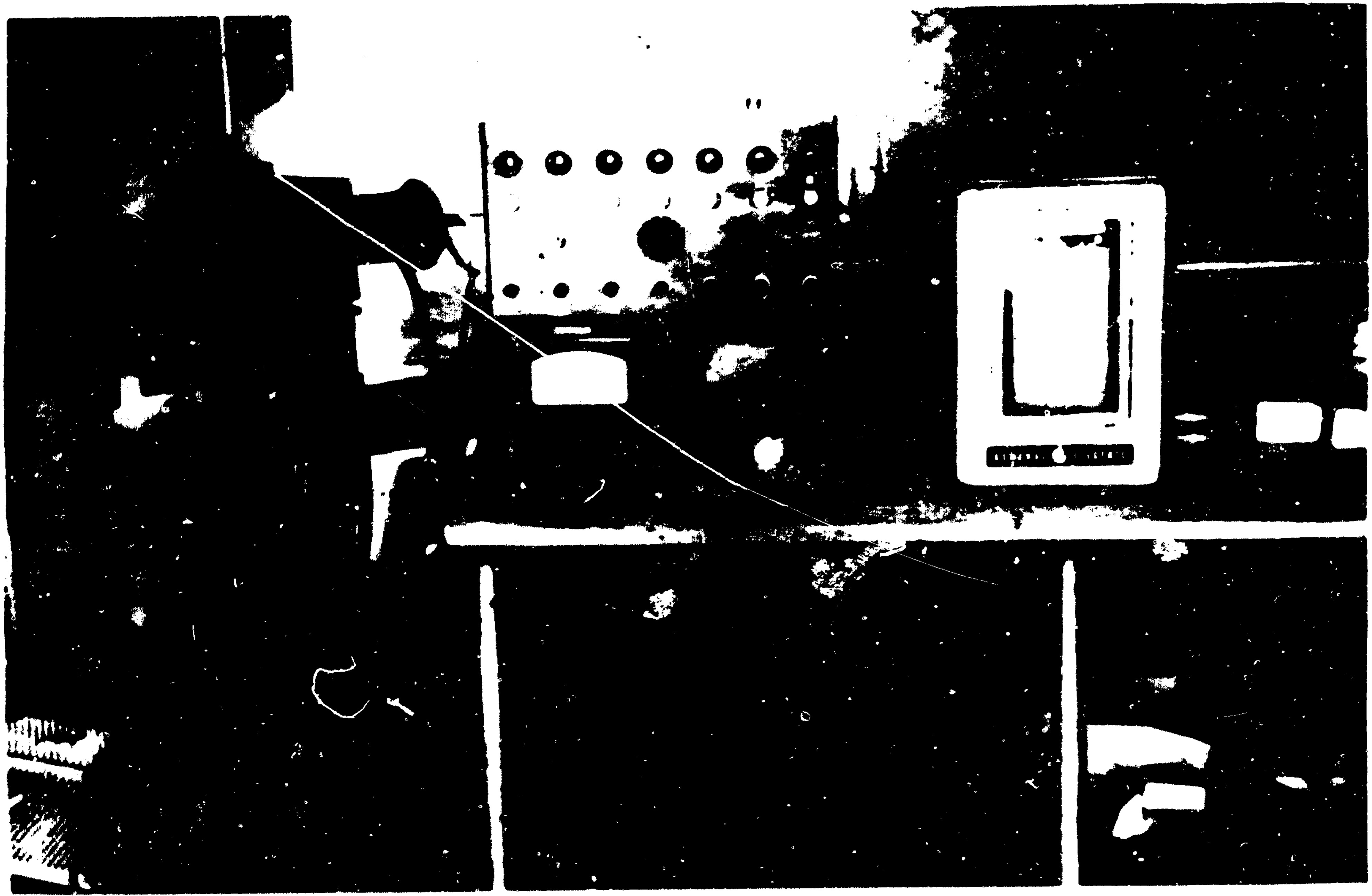

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FIGURE 8. MANUALLY OPERATED SEVEN-STAGE LIGHT-SCATTERING CASCADE IMPACTOR

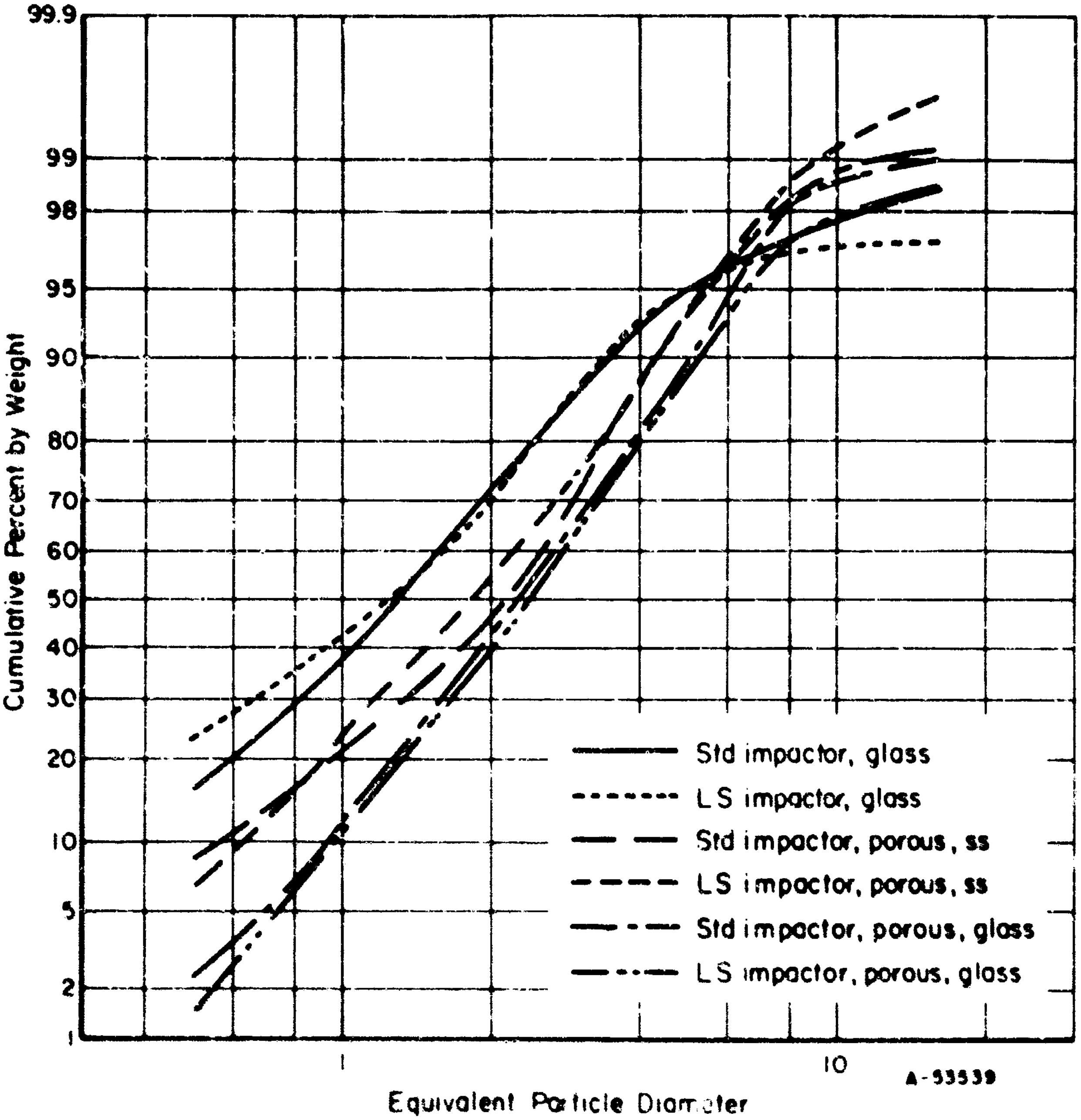


FIGURE 9. COMPARISON OF STANDARD CASCADE IMPACTOR AND LIGHT-SCATTERING CASCADE IMPACTOR WITH GLASS, POROUS STAINLESS STEEL, AND POROUS STAINLESS STEEL BACKED BY GLASS SLIDE

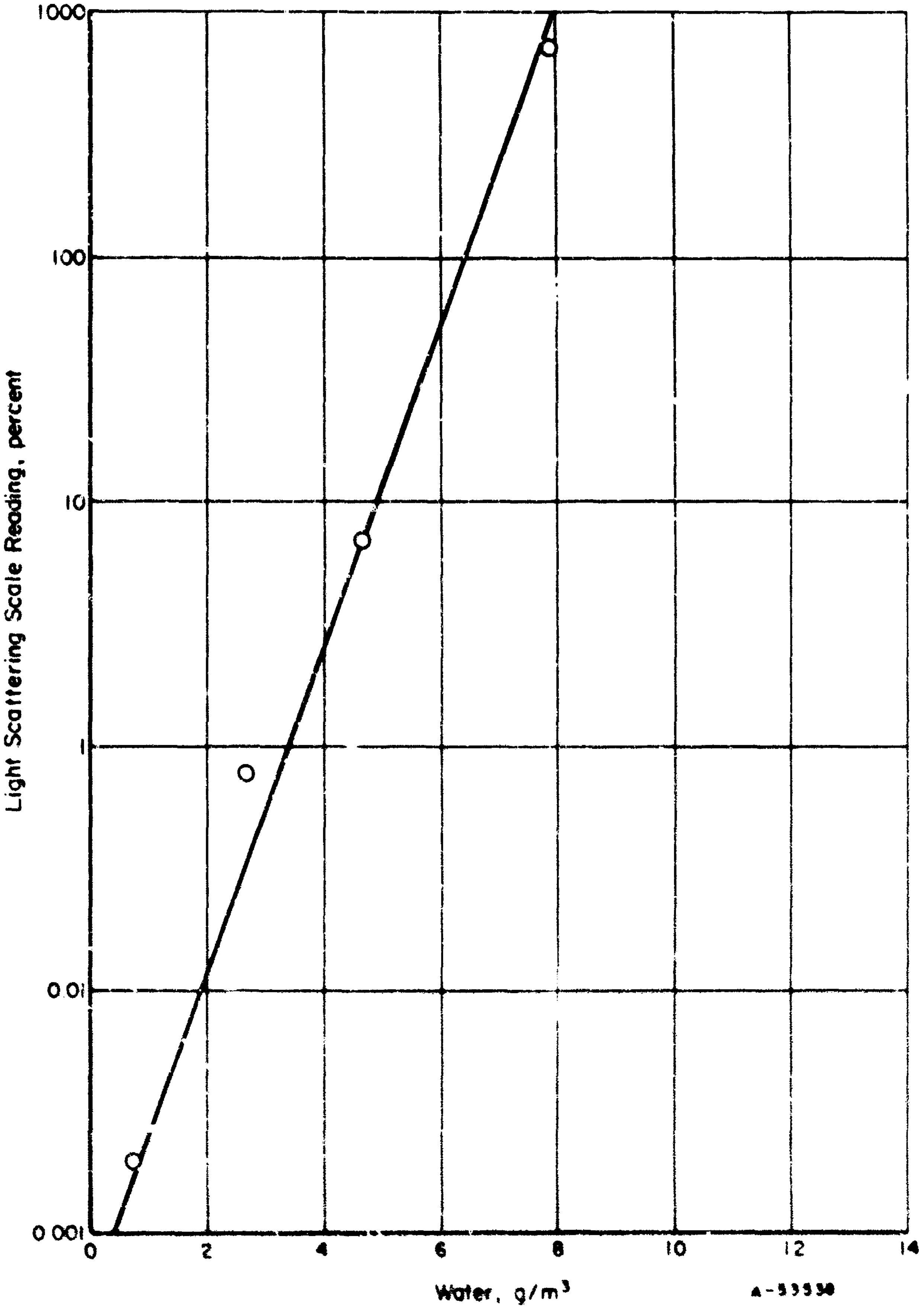
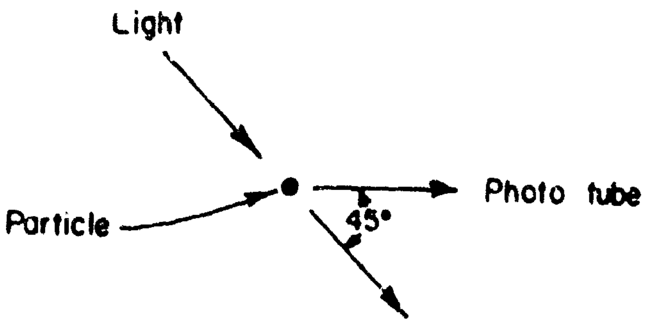
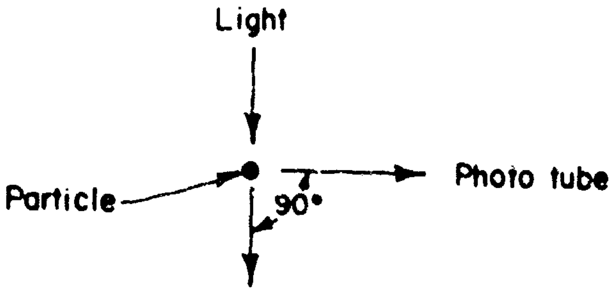


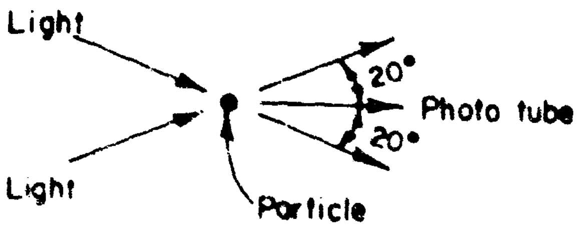
FIGURE 10. CALIBRATION CURVE OF MANUALLY OPERATED SEVEN-STAGE LIGHT-SCATTERING CASCADE IMPACTOR



45 – Degree Light Scattering



90 – Degree Light Scattering



20 – Degree Forward Light Scattering

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FIGURE 11. SKETCH SHOWING LIGHT-SCATTERING ANGLES INVESTIGATED

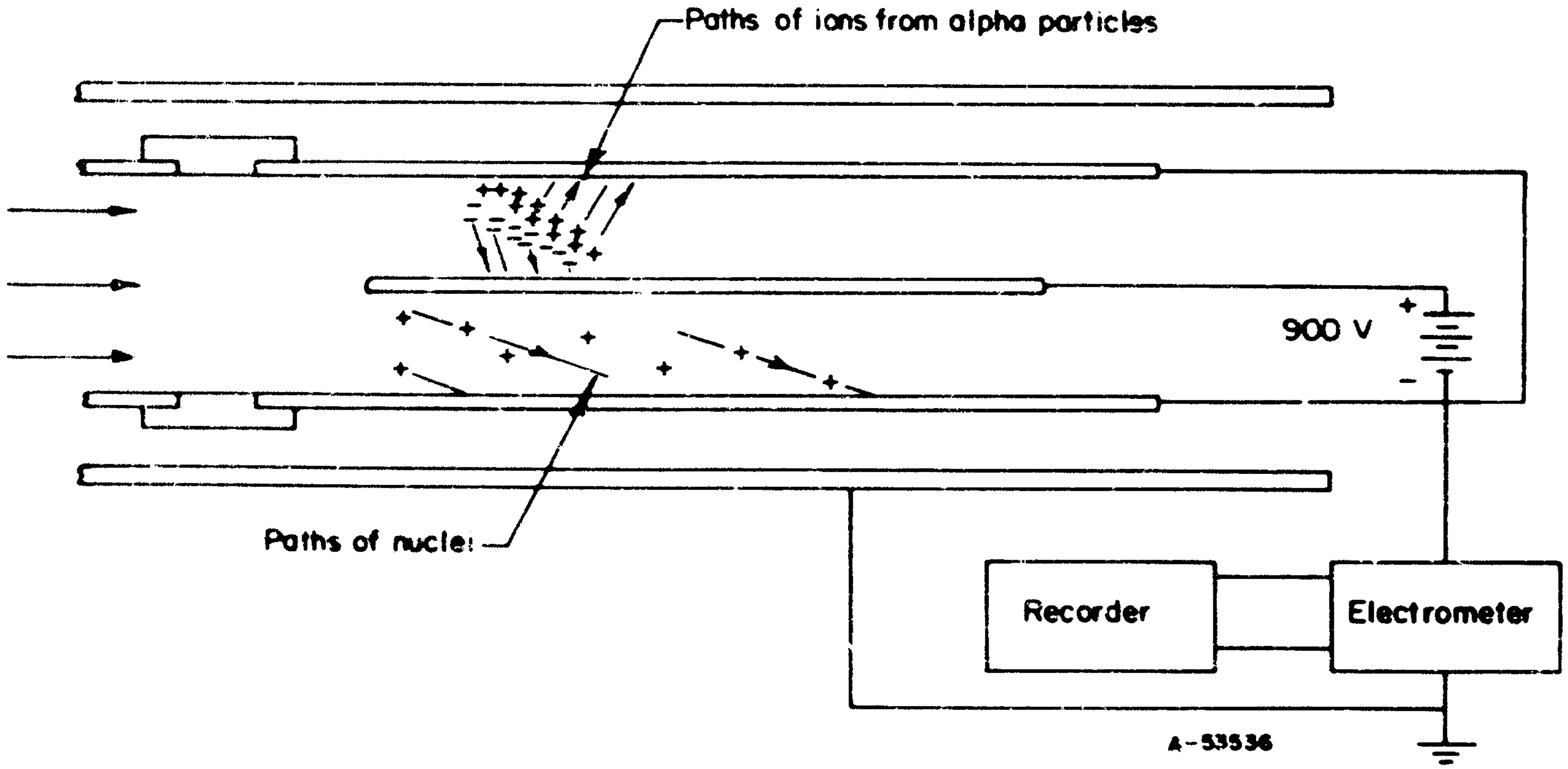

FIGURE 12. SEPARATION OF IONIZATION CURRENT FROM NUCLEI CURRENT

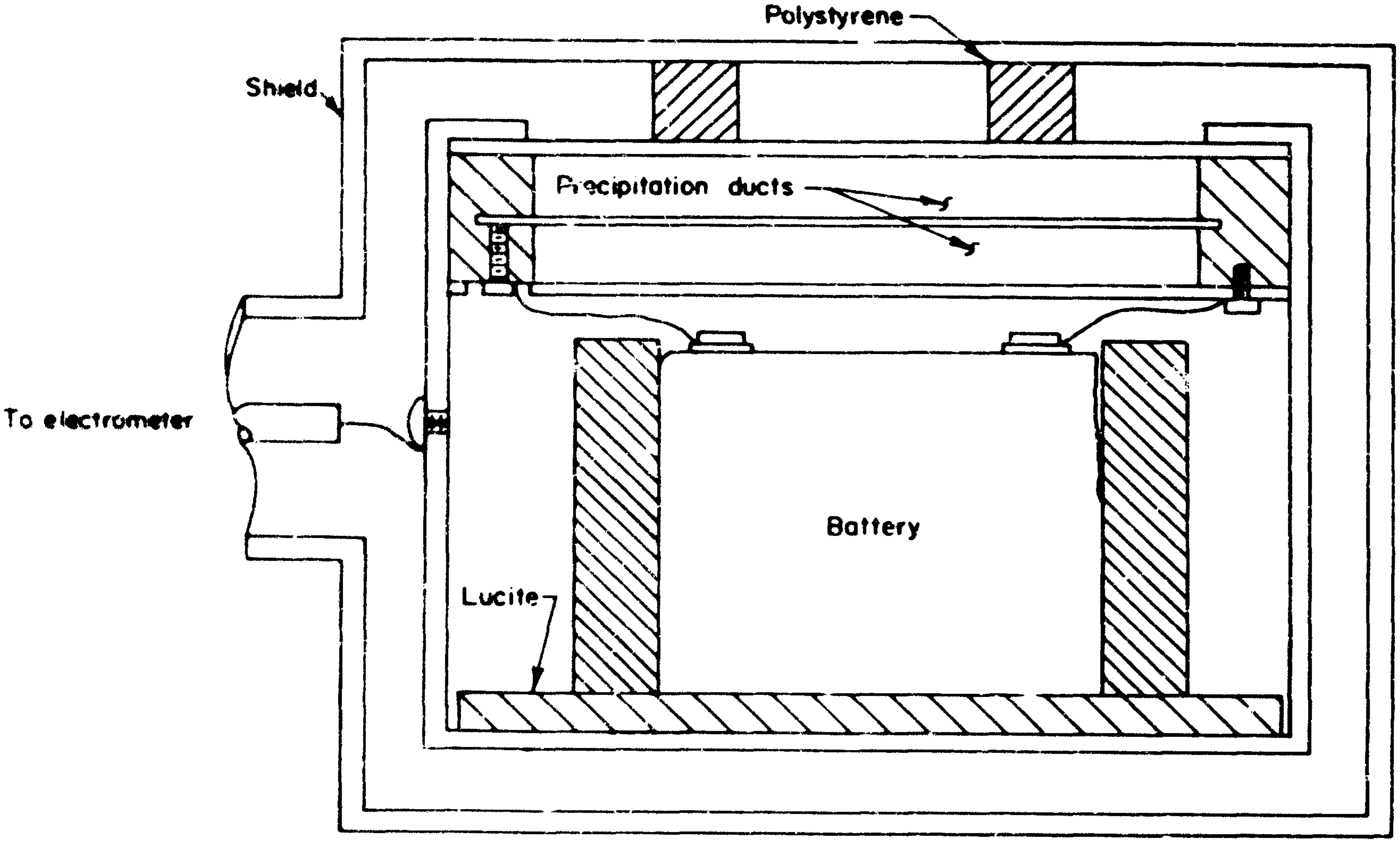

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FIGURE 13. PRECIPITATION ZONE

Transverse section.

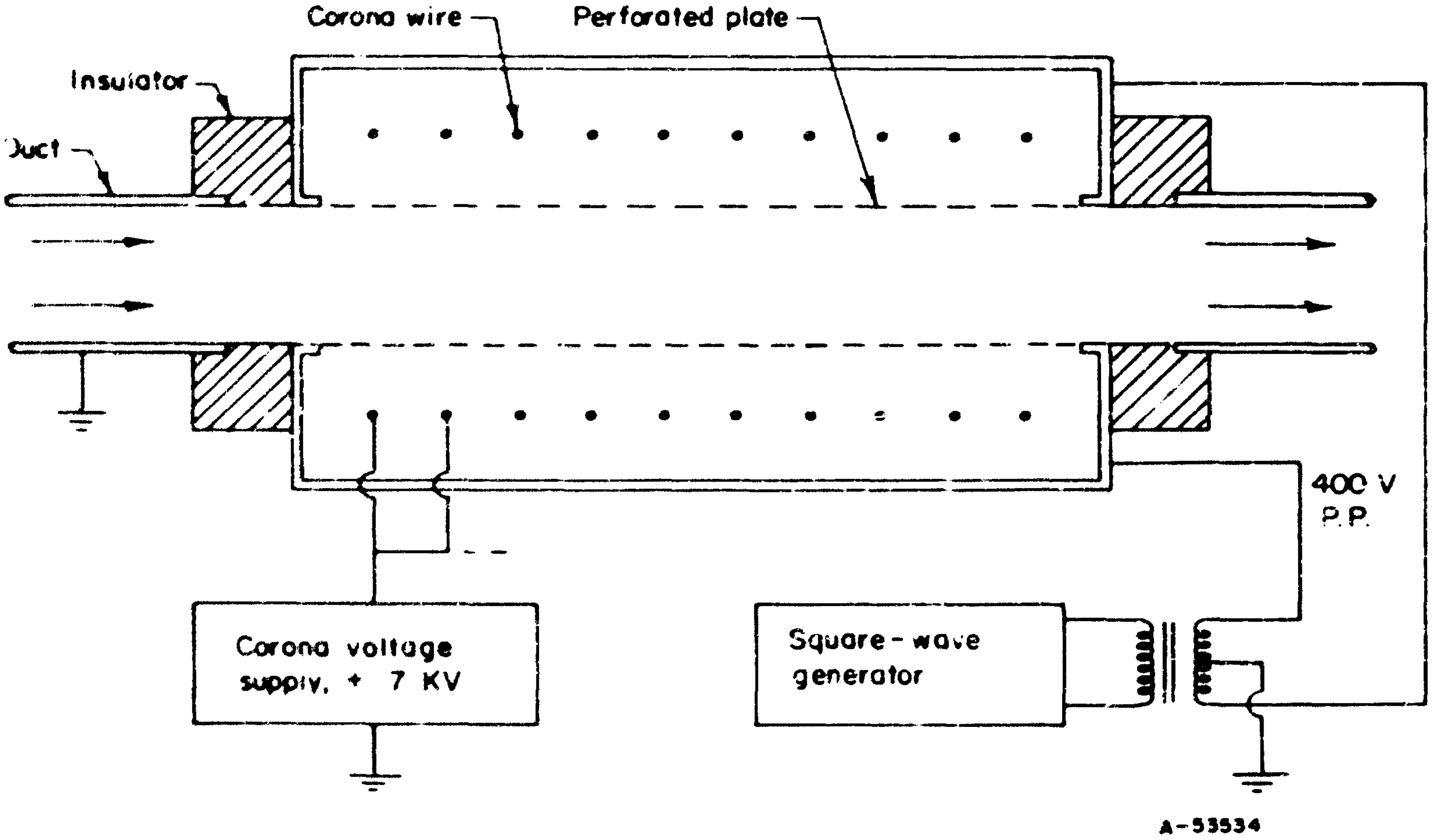

FIGURE 14. NUCLEUS CHARGING DEVICE

Longitudinal section.

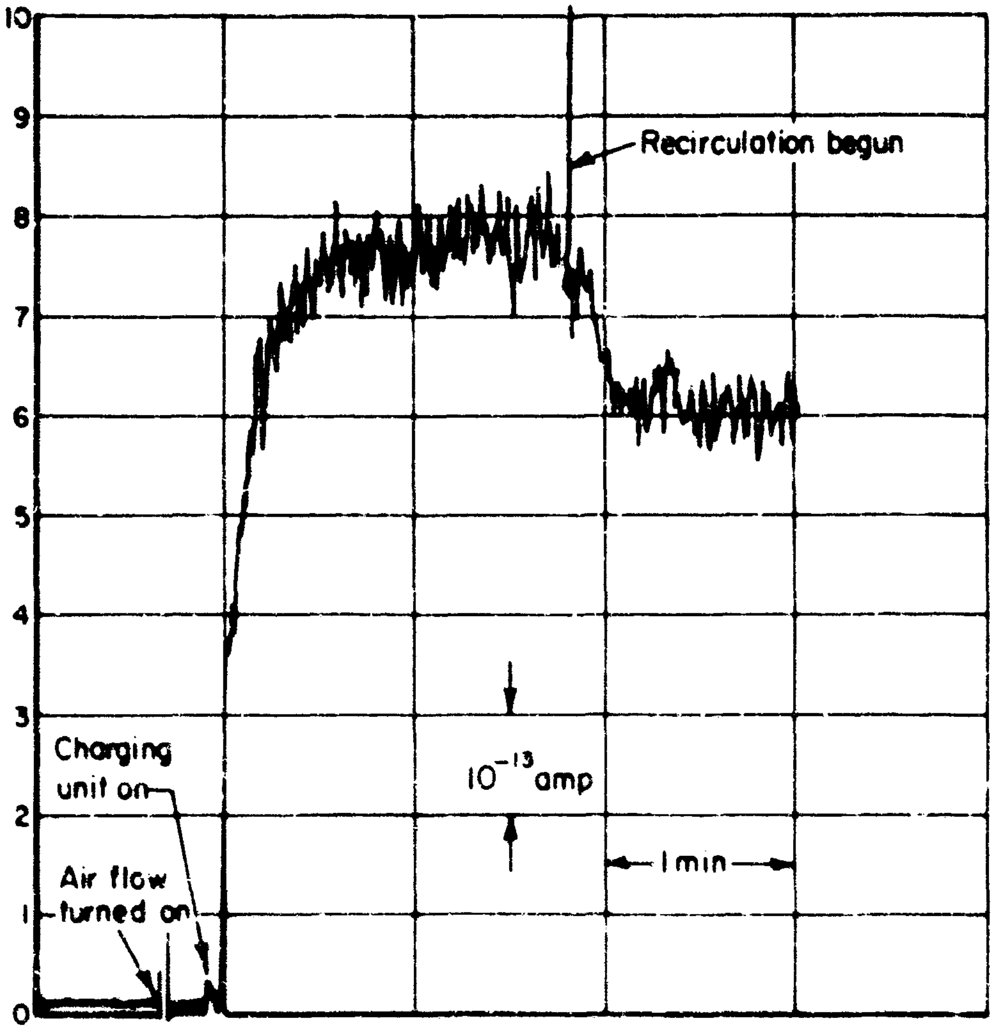

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FIGURE 15. TYPICAL RUN WITH CONDENSATION-NUCLEI COUNTER

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13 ABSTRACT

This report describes the development of a light-scattering cascade impactor to measure the size of fog droplets in the range from 0.5 micron to 16 microns. The instrument comprises six impaction stages, each equipped with a forward-light-scattering optical system to measure fog droplets of size <16, <8, <4, <2, <1, and <0.5 micron, respectively. An impaction stage to remove particles larger than 16 microns is installed at the instrument intake. Two additional stages are furnished for calibration of the electronic and optical systems.

Feasibility of a condensation-nuclei counter based on the principle of electrostatic precipitation was investigated in the laboratory.

Security Classification

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