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STANFORD RESEARCH INSTITUTE,
MENLO PARK, CALIFORNIA

NOVEMBER 1976

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EDGEWOOD ARSENAL CONTRACTOR REPORT
ED-CR-76103

OPTICAL BASES FOR REMOTE BIOLOGICAL AEROSOL DETECTION

Quarterly Progress Report 2

By:

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November 1976

STANFORD RESEARCH INSTITUTE
Menlo Park, California 94025

Contract DAAA15-76-C-0042

DEPARTMENT OF THE ARMY
Headquarters, Edgewood Arsenal
Aberdeen Proving Ground, Maryland 21010

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1. REPORT NUMBER: ED-CR-76103	2. GOVT ACCESSION NO.
3. RECIPIENT'S CATALOG NUMBER	
4. TITLE (and Subtitle): OPTICAL BASES FOR REMOTE BIOLOGICAL AEROSOL DETECTION	5. TYPE OF REPORT & PERIOD COVERED: Quarterly Progress Report 2 Covering the Period 17 March through 2 July 1976
	6. PERFORMING ORG. REPORT NUMBER: SRI Project 4805
7. AUTHOR(s): William B. Grant	8. CONTRACT OR GRANT NUMBER(s): Contract DAAA15-76-C-0042
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Stanford Research Institute, Menlo Park, California 94025	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS: IW762711 AD34-02
11. CONTROLLING OFFICE NAME AND ADDRESS: Commander, Edgewood Arsenal, Attn: SAREA-TS-R, Aberdeen Proving Ground, Maryland 21010	12. REPORT DATE: November 1976; 13. NO. OF PAGES: 18
	15. SECURITY CLASS. (of this report): UNCLASSIFIED
14. MONITORING AGENCY NAME & ADDRESS (if diff. from Controlling Office): Commander, Edgewood Arsenal, Attn: SAREA-DE-DB, Aberdeen Proving Ground, Maryland 21010, Stanford Mumford, Project Officer (671-3358)	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE: N/A

16. DISTRIBUTION STATEMENT (of this report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Lidar techniques	MAPP (program)	Baird-Atomic Fluorispec SF-1
Fluorescence	Tryptophan	
Biological aerosols	Bacillus subtilis	
Spectrofluorimeter	Growth medium	

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Measurements and calculations are being made to determine whether lidar techniques based on fluorescence and scatter can remotely detect and identify biological aerosols. The spectrofluorimeter has been calibrated. A forward-scatter instrument has been assembled to monitor aerosol flow rate. The facilities for generating aerosols have been assembled, tested, and calibrated. Aerosol concentrations up to 80 mg/m^3 have been generated from 1% solutions of ammonium fluorescein. The mass median diameters of the dried particles are

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19. KEY WORDS (Continued)

20 ABSTRACT (Continued)

in the range of 2 to 5 μm. Preliminary measurements of the excitation and fluorescence spectra of aerosols of ammonium fluorescein and tryptophan have been made. A 20-m cell for the measurement of extinction by aerosols has been purchased.

PREFACE

The research covered by this report was authorized under Task 1W762711AD34-02, Detection and Warning Investigations. The work was conducted during the period 17 March through 2 July 1976. The original notes are recorded in Notebooks 510 and 660.

The use of trade names in this report does not constitute an official endorsement or approval of the use of such commercial hardware or software. This report may not be cited for purposes of advertisement.

Acknowledgments

Charles E. Lapple, Clyde Witham, and Donald Hutson helped with the manufacture of the forward-scatter aerosol monitor. James Centis helped with the assembly of the aerosol-generation equipment. Clyde Witham calibrated the aerosol-generation equipment and assisted in the measurements of aerosol properties.

OPTICAL BASES FOR REMOTE BIOLOGICAL AEROSOL DETECTION

I INTRODUCTION

The object of work on this project is to make all the necessary measurements and calculations to determine whether lidar techniques can be used to detect and identify biological aerosols remotely. Aerosols of simulant biological organisms will be generated in the laboratory and passed through a spectrofluorimeter. Fluorescence and scatter will be carefully measured, as will excitation and extinction. Absolute cross sections will be determined. The SRI Modular Atmospheric Propagation Program (MAPP) will then be used to calculate lidar sensitivity for various targets and conditions.

II ACTIVITIES DURING PERIOD REPORTED

A. Calibration of the Spectrofluorimeter

The Baird-Atomic spectrofluorimeter, Model SF-1, has been well characterized for the purposes of this project. Both the source intensity and the detector sensitivity have been measured.

The source intensity was measured using a Schottky barrier photodiode (Model PIN-10UV, from United Detector Technology). It was calibrated using a radiometer (EG&G Model 550) that in turn was calibrated using standards traceable to the National Bureau of Standards. The calibration curve for narrow slits (2 nm spectral width) is shown in Figure 1. The light source was a xenon lamp. The lamp has several intense lines (470, 495, 535, and 630 nm) as well as a black-body continuum. The monochrometer gratings are blazed for 300 nm. The relative measurement uncertainty is estimated to be ±5%. During operation, the intensity of the signal reaching the sample chamber changes because of xenon arc wander and thermal expansion of spectrofluorimeter components. This does not present a serious experimental problem as long as the system sensitivity is checked periodically and adjustments in lamp position are made.

The effect of slit width on source intensity was also investigated. Using 8-mm slits instead of 2-mm slits increased the source intensity by a factor of 15.5 ± 0.6 for wavelengths spaced 50 nm apart from 300 to 700 nm. (There are two slits for the two monochrometers on the source side.) Using one 32-nm and one 8-nm slit increased the intensity by another factor of 2.85 ± 0.5 for a total increase of 44.2 ± 1.1.

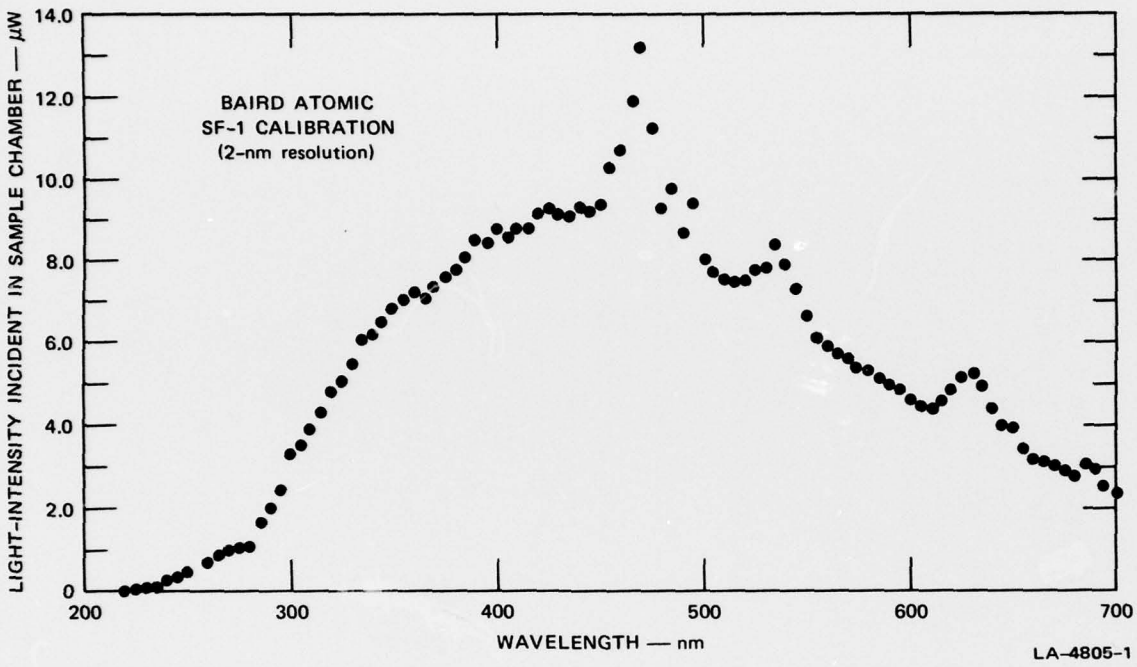

FIGURE 1 CALIBRATION CURVE FOR THE EXCITATION HALF OF THE BAIRD ATOMIC SPECTROFLUORIMETER

Extrapolation to encompass the use of two 32-nm slits indicates that 126 times as much light is available for 32-nm slits as for the 2-nm slits. The use of 32-nm slits would give 1.17 mW for the continuum near 400 nm.

The detector half of the spectrofluorimeter was calibrated using two types of targets in the sample compartment--an MgO powder (Eastman Kodak #6080), and transparent liquids that have known Raman scattering efficiency. The MgO powder was applied to a glass slide. It acts as a Lambertian reflector with greater than 90% reflectance for the 220-to-700 nm range. A graph of the reflectance as a function of wavelength is provided by Eastman Kodak. The liquids (cyclohexane and benzene) have been measured and reported in the literature.* The values given for cyclohexane and MgO agreed very well at 450 nm, while benzene gave a value approximately 40% lower than expected for the same conditions. Three series of measurements using the MgO target gave variations on the order of ±15%. This, along with the Raman scattering discrepancies, implies that these measurements have an uncertainty at this time of ±25%. The calibration will be improved if warranted by later experimental results. The peak sensitivity of the detector is 0.15 $\mu W/\mu A$ at 450 nm, falling nearly symmetrically to 75 $\mu W/\mu A$ at 220 nm and 120 $\mu W/\mu A$ at 700 nm for a perfect Lambertian target in the sample compartment.

B. Aerosol Flow Monitors

Meters that measure aerosol flow using forward scatter of light have been fabricated and tested. The minimum sensitivity of these meters corresponds to an aerosol density of about 0.5 mg/m^3. A reference scatterer made of teflon produces a signal of approximately 1 nA. This corresponds to 1 g/m^3 in previous calibrations on coal dust, reported in "Portable Mine Dust Concentration Instrument," Final Report, SRI Project PYU-1267, Contract HO111688, for the U.S. Bureau of Mines. Water-based biological aerosols will give somewhat different scattering-to-mass ratios than coal dust. While these devices will give some information on the amount of aerosol flow, their primary purpose is to monitor the relative aerosol flow rates.

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* M. J. Colles and J. E. Griffiths, J. Chem. Phys., Vol. 56, No. 7, p. 3384 (April 1972).

C. Fluorescence and Excitation

Ammonium fluorescein was used for preliminary measurements of system sensitivity. It was chosen rather than sodium fluorescein because it produces particles that are more spherical and less hygroscopic.* The aerosol was kept at pH 11 for maximum fluorescence efficiency.

The fluorescence peaked at 514 nm. One absorption band peaked near 490 nm, and another near 325 nm. The instrument response was 27 times greater for excitation at 490 nm than for excitation at 325 nm. The light intensities for these two wavelengths were in the ratio of 1.8 to 1, which means that for the same incident power levels the fluorescence was 15 times greater at 490 nm than at 325 nm. In terms of photons emitted relative to incident photons, the ratio was only 10 to 1. The absorption in the two bands has not been measured. With 8-nm slits, 10^{-11} g/cm^3 of ammonium fluorescein can be detected with a signal-to-noise ratio of one at a signal level of 0.1 nA.

Aerosols were then generated using 1% by weight solutions of ammonium fluorescein in water with ammonium hydroxide added to maintain a pH of 11. Aerosols that were dried by the addition of compressed air with relative humidity of 12% gave no measurable fluorescence. Aerosols with no dilution air added gave a sizable fluorescence signal, with a peak of about 1.5 nA and a noise level of 0.16 nA. The fluorescein aerosol gave a signal equivalent to 7% by weight of fluorescein in water. For the entire band, the signal-to-noise ratio was about fifty to one. When dilution air was added to reduce the aerosol concentration as measured by the forward-scatter device to about 60% of its nondiluted value, the fluorescence signal decreased to 10% of the nondiluted value. The most likely explanation is that when the aerosol is wet the fluorescein molecules are distributed diffusely in each droplet, allowing good optical penetration. It was noted that such aerosols were wet when collected on filters. When the aerosols are dry, which seems to happen with even a small amount of dilution air, it is surmised that the particles are too thick to fluoresce well. To check this, a 1-cm-thick sample of 2×10^{-4} g/cm^3 solution of ammonium fluorescein in water at a pH of 11 (equivalent to a 2-μm-thick plate) was measured for absorption. It was opaque for wavelengths shorter than 520 nm, and reached 50% transmission at 532 nm. For this condition, only a small surface depth that is in the field of view of both the source and the detector can be effective in providing a fluorescence signal. (For such particles, the backscattered fluorescence signal would be roughly twice as large as the 90° signal.)

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* W. Stöber and H. Flachsbart, Atmospheric Environment, Vol. 7, p. 737 (1973).

Preliminary measurements of the excitation and fluorescence spectra of tryptophan aerosols were made. The signals were easily measurable down to the lowest aerosol concentrations used (~ 6 mg/m^3). The excitation maximum occurs in the spectrofluorimeter at 287.5 nm for wet aerosols, and at 297.5 nm for dry aerosols. The 90^o scatter and fluorescence signals increased linearly with concentration for relative humidities from 20% to about 67%. For wet aerosols, the fluorescence signal was approximately half as strong as for the same mass of dry aerosol, and the 90^o scatter was almost twice as large as for the same mass of dry aerosol.

Since the aerosol concentrations used in the experiments would produce only a very small extinction for a two-foot path length (on the order of one to two percent) it was decided to order a multipass cell to increase the optical path length. A Wilks Scientific Corporation 20-m cell has been ordered and delivered. It has a box length of 3/4 m. It will be mounted vertically above the Baird Atomic spectrofluorimeter, which will be used as the light source. A PIN-10UV photodiode will be mounted on the device as the detector.

D. Scatter

Ninety-degree light scatter shows up readily when aerosols are in the sample cell in the Baird Atomic spectrofluorimeter. It is planned to measure 180^o light scatter by displacing a sample cell out along the entrance beam of the spectrofluorimeter and using a mirror to send 180^o-scattered light into the detector of the spectrofluorimeter.

E. General Test Facilities

The test facilities have all been designed, fabricated, installed, and put into operating condition.

A photograph of the facilities is shown in Figure 2. Figure 3 presents a schematic diagram of the facilities. In Figure 3 an attempt has been made to maintain the scale and arrangement of the various components insofar as possible, consistent with clarity of presentation. This should permit identification of all the major items in the photograph of Figure 2. The generator with its auxiliaries, the dilution chamber, and the humidifier are separately housed in an enclosure with a plastic window. This enclosure is vented through an absolute filter to the atmosphere by means of a separate fan. This enclosure was provided to protect personnel from any potential hazards of biological or chemical

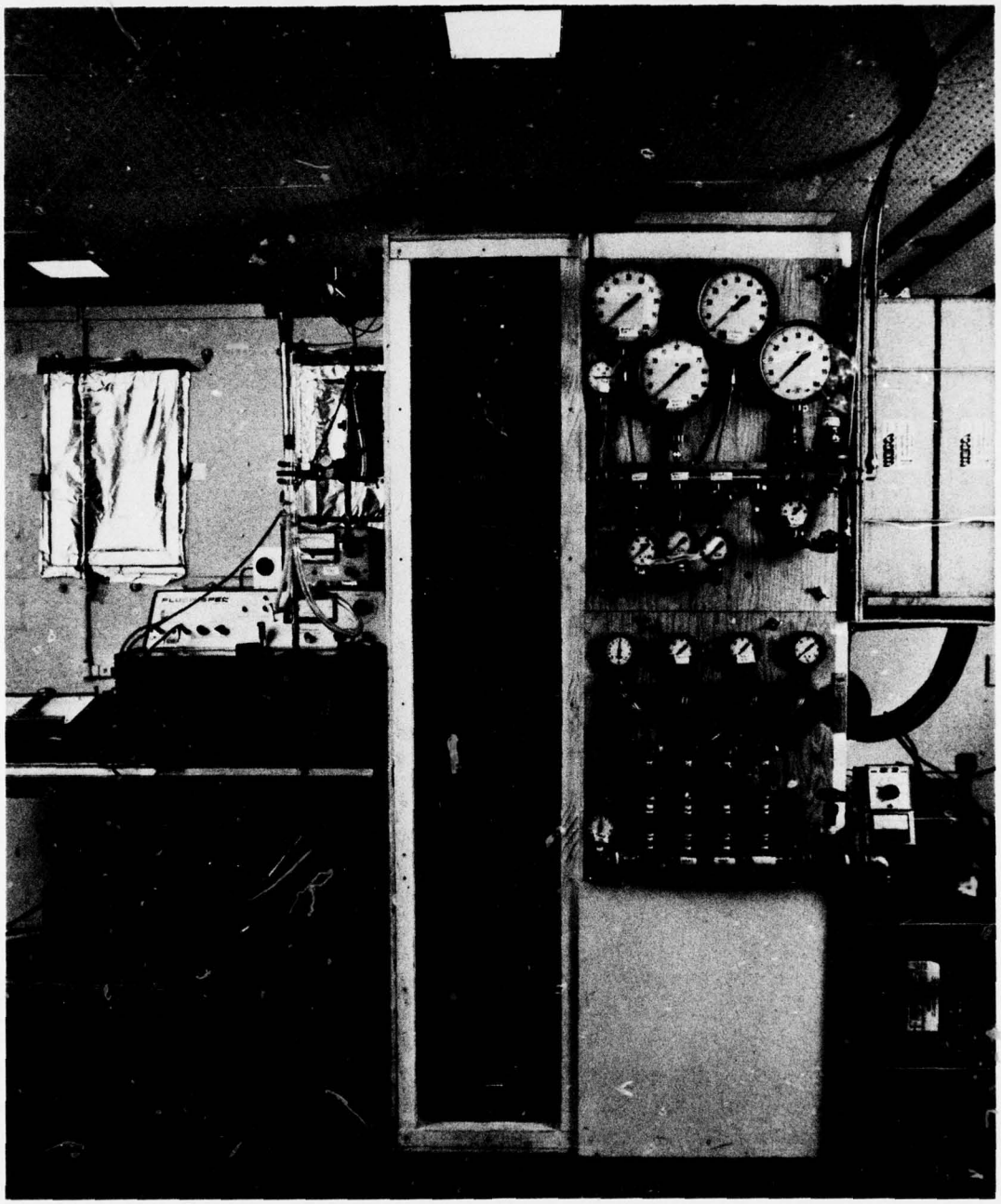

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FIGURE 2 PHOTOGRAPH OF THE EXPERIMENTAL APPARATUS

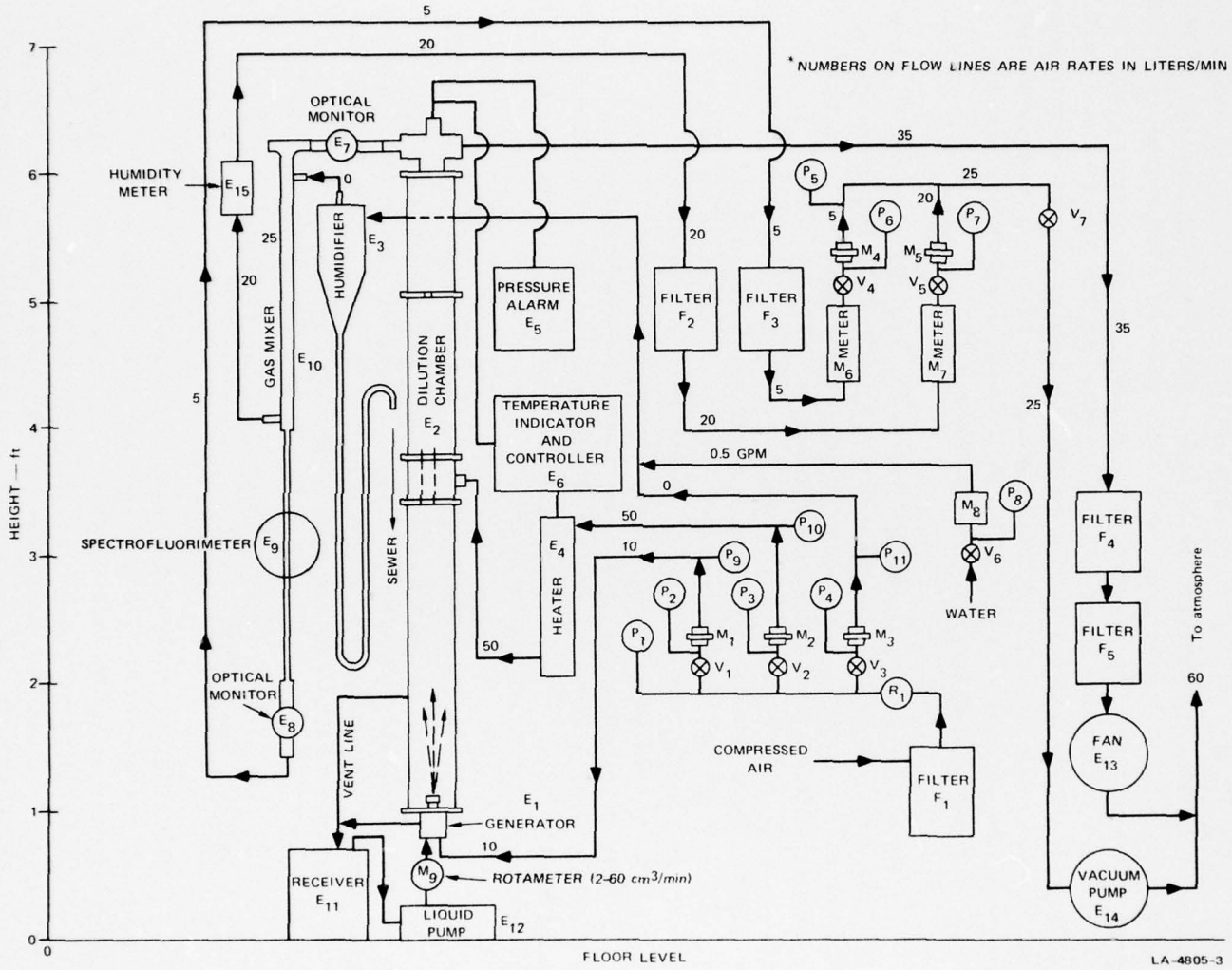

FIGURE 3 BLOCK DIAGRAM OF THE EXPERIMENTAL APPARATUS

materials that might be used. The experimental system is also kept under negative pressure by means of a separate fan (E-13). A warning alarm that would sound if the vacuum dropped below 0.2 inch of water is shown in Figure 3, but was not and will not be installed.

A pneumatic atomizer discharges a spray into the bottom of the dilution chamber. A bottle located in this section of the chamber serves to separate out super-coarse droplets. Halfway up the chamber, additional heated air is introduced to dry the droplets. At the top of the dilution chamber part of the aerosol is led to the test equipment at predetermined rates by means of critical orifices discharging into a vacuum pump; the balance of the aerosol is discharged to the atmosphere after passing through two absolute filters in series (F4 and F5).

As a simple and convenient expedient, it was originally intended to control humidity by diluting a given aerosol with various amounts of saturated air. However, this would result in lower aerosol concentrations. In view of the fact that achieving maximum aerosol concentrations have become important in permitting measurement of aerosol fluorescence, this approach has been abandoned and the humidifier shown in Figure 3 is not being used. Instead, a humidity meter has been installed to monitor a stream of aerosol at the outlet of the gas mixer. The dilution air rate will then be varied to give the desired humidity for this aerosol stream. The humidity meter consists of a glass unit in which the aerosol flows successively over a dry- and a wet-bulb thermometer. A plastic-strand immersion type of unit was first considered but proved to be too slow in response.

A forward-scattering light meter was placed both ahead of the gas mixer and behind the spectrofluorimeter. These meters are used to monitor the uniformity of aerosol concentrations with time and position. Actual aerosol concentrations are determined by separate "Nucleopore" filter samples taken before the second optical monitor.

F. Aerosol Generation

The original aerosol generator contained a 0.020-inch-diameter air orifice. In an attempt to increase aerosol production rates, this was replaced with a 0.028-inch orifice. The original system also provided for internal recycling of liquid by the pneumatic nozzle, and the recirculation rate was not measured. It was later found that through reduction of this rate a denser aerosol was generated. The original system was therefore replaced with one in which the liquid feed rate to the atomizer nozzle was both controlled and metered.

For reasons of analytical convenience, ammonium fluorescein has been used as the aerosol material for purposes of assessing and developing the generator. In the early tests, aerosol concentrations of 2 to 80 mg/m^3 were obtained with mass median diameters of the dried particles in the range of 2 to 5 μm. However, because of the many changes that were made simultaneously, these data were too spotty to provide any systematic evaluation of the generator performance. A series of tests is currently underway to establish particle size systematically (by visual microscopic estimate), mass concentration (by filter measurement), and optical response (forward light scattering), as a function of atomizing gas pressure and liquid circulation rate. Once this is completed with a 1% ammonium fluorescein solution, a 1% tryptophan solution will be used for further experimental work.

The first measurements with an ammonium fluorescein solution and a 0.028-inch atomizing nozzle were quantitatively ambiguous because, as the result of evaporation, the 1% starting solution had concentrated to 1.6% by the time the tests were completed. These tests, however, did indicate that an optimum aerosolization rate was obtained for liquid feed rates of 10 to 40 cm^3/min. The optimum aerosolization rate was in the range of 0.02 to 0.2 cm^3/min for an atomizing air-pressure range of 10 to 80 psig, the lowest rate being obtained at the lowest pressure. From simultaneous measurements of relative humidity with a 50-ℓ/min dilution-air flow rate, it was concluded that saturation of the atomization air occurred primarily from liquid recycled to the atomizer rather than from the ultimate aerosolized drops. With the dilution-air flow rate of 50 ℓ/min, aerosol concentrations of as high as 30 mg/m^3 were obtained with particle mass median diameters (as estimated from visual microscopic observation) of the order of 4 μm.

For the purpose of achieving speed and conserving funds, all size characterization in the preliminary generator development work has been by visual estimates of mass median diameter from simple microscopic examination. For any final test aerosols, a more quantitative characterization of size distribution will be made. Continuous gravity settling and impaction are being considered. Each has its own limitations. The anticipated size distribution of the dried aerosol will be predominantly in the range of 2 to 10 μm, with some particles as large as 15 μm. A continuous gravity chamber is useful for this range, but suffers from potential problems of eddy circulation arising from small temperature differences. We have therefore designed and built a new type of unit that utilizes a radial outward flow from the center of a 2-ft-diameter sink, 1/2 inch high. Clean air is admitted radially outward through a porous plug located at the center and at a fixed rate in the range 2 to 20 ℓ/min. Aerosol is admitted at the top center of the channel and also

flows radially outward at a rate of the order of 0.1 ℓ/min. Convective currents will be minimized by providing water cooling of the floor of the chamber, thereby setting up the equivalent of an atmospheric temperature inversion in the chamber. The contemplated arrangement has the following advantages: (1) settling is on a differential rather than an integrated basis (i.e., only particles of a given size should deposit on the floor of the chamber at any given radius); (2) a single channel or operating condition will permit analyzing a wider range of particle sizes, since settled-particle diameter should vary inversely as the radius rather than inversely as the square root of length as in the conventional rectangular channel; (3) thermal convective currents should be minimized; and (4) the absence of side walls should prevent lateral velocity gradients (assuming that an initial uniform radial velocity is provided by the porous plug)--in this sense such a radial channel corresponds to a rectangular channel of infinite aspect ratio.

An impactor poses problems with the coarsest particles, caused by particle acceleration and possible particle bounce. SRI had originally planned to use an Andersen impactor that is available. However, Edgewood Arsenal is lending a Casella unit, which will be used for these measurements instead of the Andersen unit.

It is not intended to devote more than one week of development time to any particle-size-measuring technique. Should major problems arise, either the technique would be abandoned or the results would be taken with recognized limitations. Counting of particles in the photomicrographs will then be resorted to, although in the case of an aerosol having a wide size distribution, measurement of the mass median size will be less precise.

G. Maximum Aerosol Concentrations

Pneumatic atomization to the desired size range of 2 to 10 μm will require that the initial suspension or solution of biological materials be less than 10%, possibly less than 1% in order to avoid excessively viscous mixtures. Thus, the bulk of the liquid must be evaporated by the combination of atomizing and subsequent dilution with air. This need for evaporative capacity on the part of air leads to a maximum possible aerosol concentration that can be achieved for any desired relative humidity as given by

$$c_a = \rho \, c_\ell \, M \, (R_f - R_o)$$

where

c_a = Maximum aerosol concentration, kg/m^3

c_ℓ = Fractional concentration of aerosol material in initial liquid being atomized, weight fraction

ρ = Air density at ambient conditions, 1.18 kg/m^3

M = Concentration of water in saturated air at ambient conditions, approximately 0.020 kg water/kg air

R = Relative humidity of air in final aerosol, fraction of saturation

R_o = Initial relative humidity of air, approximately 0.12 for 110-psi compressed air.

From this relationship the following are the maximum dried aerosol concentrations that can be achieved for any final relative humidity of the aerosol system for a 1% solution (or suspension) of the dry aerosol material:

Final Relative Humidity (%)	Maximum Dry Aerosol Concentration (mg/m^3)
12	0
20	19
35	54
50	90
85	172
100	208

Only a small increase in aerosol concentration could be achieved by using completely dried air rather than that obtainable from ordinary compressed air. By going to higher initial solution strength, however, correspondingly higher aerosol concentrations could be achieved, provided the higher resultant viscosity does not prevent achieving the desired degree of atomization (or particle size). Any evaporation of liquid from recycled drops will result in lower aerosol concentrations than the values calculated above.

III CONTRACT EXPENDITURES TO DATE

As of 2 July 1976 a total of 1,417 man-hours, representing 56.4% of the man-hour allocation, had been expended; funds in the amount of $48,243, representing 60.6% of the total contract amount of $79,610, had been spent.

IV PLANS FOR THE NEXT QUARTER

The remaining pieces of experimental apparatus will be mounted, tested, and calibrated. This includes the Wilks 20-m cell, the back-scatter cell, and the gravity-settling particle size analyzer.

Optical properties will then be measured for aerosols of ammonium fluorescein, tryptophan in water, growth medium (casein partial hydrolysate), egg tissue medium, Bacillus subtilis, Escherichia coli, and Pasturella tularensis. Liquid suspensions of the above materials will also be investigated to aid in understanding the properties of the aerosols.

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