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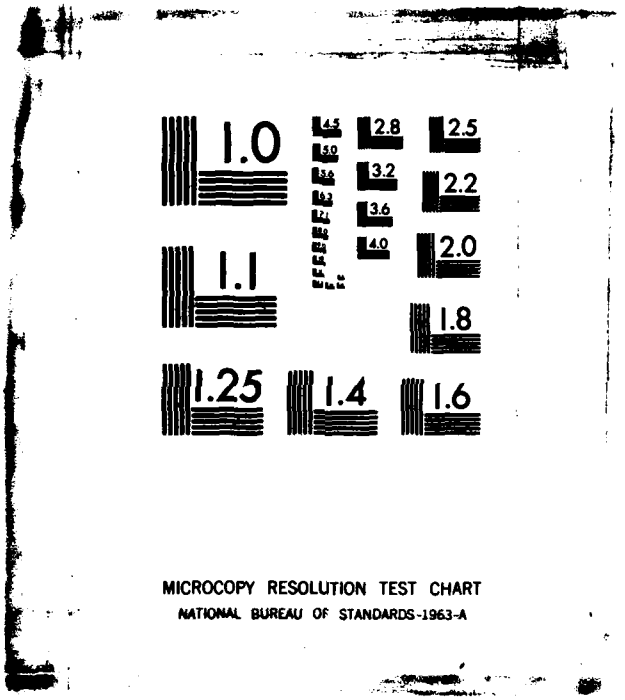

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A MOLECULAR IDENTIFICATION DEVICE FOR INDIVIDUAL SUB-MICRON AEROSOLS: ENGINEERING BREADBOARD DEVICE

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

Progress on the second phase development effort of an instrument for determining both the size and the chemical composition of individual sub-micron particulates, is reported. A description of improvements incorporated in the engineering breadboard following recommendation issuing from the first phase effort, together with a very preliminary set of specifications for a flight prototype instrument, are included.

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Also included is a brief discussion on the corrolary question of the size distribution prevalent in the stratosphere, and its relation to design aspects of the instrument.

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I. INTRODUCTION

This report describes work performed during the second phase of a long range development effort leading ultimately to the simultaneous capability of identifying and sizing individual submicron particulates in the stratosphere. The first-phase effort, whose end result indicated that the concept was feasible in general, has been described in a previous report (Ref. 1). Briefly, the concept involves: 1) the very gentle sampling of air through a sampling flow tube in which laminar flow is assiduously maintained, 2) the detection of individual particulates contained in the sample by observing the occurance of scattered light as each traverses a carefully controlled illumination region, 3) an independent determination of the size of each particle by measuring the quantity of forward scattered light collected within two carefully controlled annular apertures, 4) an appropriate decision logic (employing a criterion based upon the particle size) to actuate the identification procedure, 5) evaporation of the particle by its absorption of energy from a very high fluence excitation flashlamp, and 6) the subsequent examination of the rapidly spreading gaseous particulate material by absorption spectroscopy using a separate flashlamp illuminator.

The aforementioned feasibility study produced experimental results that were sufficiently encouraging to warrant continuation of the effort: the gaseous materials evaporated from micron-sized individual particulates of several different molecular species were identified spectroscopically. However, it was difficult to detect the presence of these spectral identification features imbedded within the "noise" (film fog, graininess, etc.) without the benefit of a practiced eye. These results pointed to the necessity for improving the S/N ratio of the gaseous identification procedure as the first step toward the ultimate achievement of automated recognition of spectral data.

Thus, the present contractual effort centered upon incorporating several improvements in the breadboard device. In addition, two other thrusts were included, so that this second-phase effort was directed along three lines simultaneously: 1) publication of previous stratospheric particulate size distribution results as well as pertinent instrumentation factors in the open

Ref. 1: Miranda, Henry A., Jr., "A Molecular Identification Device for Individual Sub-Micron Aerosols: Feasibility Study", Final Report, AFGL-TR-80-0155, May 1980, ADAD90018.

literature, 2) producing proof-of-principle results that would be obvious even under the most casual perusal, and 3) generating requisite engineering data for use in designing a flight prototype instrument. These three aspects are discussed separately below.

II. SIZE DISTRIBUTION PUBLICATIONS

The publication effort constituted a logically coherent ingredient of the overall development program because these prior results pointed to the distinct possibility that a much larger percentage of the total stratospheric mass resided in particles smaller than 0.3 - 0.4 μm diameter than is currently believed to be the case by the scientific community. This in turn would indicate that the exchange rate between solid/liquid and gaseous state of volatile particulates in the stratosphere would be significantly greater than generally thought to be the case, thereby rendering much more exigent an understanding of the chemical constituency of these smaller particles.

It was believed that a thorough objective airing of the rather complex (and subtle) set of instrumental considerations among members of the scientific community would permit a meaningful resolution of this important issue. It was hoped that this route would lead eventually to a set of final instrumentation specifications more closely matched to the realities under investigation, than would be possible in the absence of such a consensus. This particular nexus will be elucidated further in Section IV below.

Unfortunately this did not take place. In the meantime, a considerably greater amount of contract effort was expended in this activity than had been anticipated at the outset, thereby leaving a lesser amount for hardware-related activity.

III. PROOF-OF-PRINCIPLE BREADBOARD DEVICE

Several specific improvements were incorporated in the original breadboard device described in Ref. 1, following more or less along the lines recommended therein. These are described below.

1. Spectrograph improvements

The natural line widths comprising the spectral identification data of interest (in this instance, the vibrational lines of electronic transistions for the various molecular species under investigation), are much narrower than the spectral resolving power of the spectrograph. It followed therefore that detection capability would improve in a linear fashion with increasing spectrographic resolution. This in turn was achieved by simply increasing the focal length of the collimator/imaging optics, since the inherent resolving power of the transmission grating was sufficiently large as to be far from the limiting factor thereof.

Because of the very small object field of view ($\sim$ 1 mm), it was possible to incorporate a long focal length (one meter for both the collimator lens and the imaging lens) into a very compact multiple-folded optical module. The modified spectral configuration resulted in a plate factor of 16.34 Å/mm. This represents an improvement of 11.6 as compared with the 189 Å/nn plate factor of the earlier model.

This improvement was achieved at the expense of two separate disadvantages: 1) the absence of zero-order imaging, and 2) the necessarily more restricted spectral coverage range across the Polaroid film ($\sim$1500 Å), as compared to the full range (i.e., 1800 Å - 6500 Å) in the previous case. The former rendered somewhat more complicated the task of system alignment, and the latter required the fabrication of an articulated adjustment apparatus to permit different spectral regions to be covered. Since most chemical species of interest will yield useful spectral information over a range of 1500 Å, the latter restriction is not regarded as serious for the present breadboard application.

A series of tests was conducted to ensure that the spectrograph optics were properly aligned, and also that the spectral image was in proper focus. In addition a very careful spectrographic calibration was performed, using a Mercury-Argon lamp as a spectral source. This was checked against the characteristic emission lines of the Xenon flashlamp with excellent results.

A spectrometer entrance slit was also introduced. The object field region of about one cubic millimeter(at the intersection of the sampling flow cylinder and the laser illumination ribbon) was imaged onto this slit by an equal-conjugate biconvex relay lens. A spectrograph entrance slit width of 0.1 ($\pm$0.005)mm produced a spectral resolution of 1.63 Å.

2. Optical Configuration

The improved breadboard device can be conceived as a set of separate orthogonal modules having a single common region of interest ($\sim$ one cubic millimeter), as shown schematically in Figure 1. Two of these are at right angles to each other, and lie in the same plane (the horizontal). The first one is the HeNe Laser ribbon illumination/scattered light receiver optics; and the second is the spectrograph/xenon flash illuminator optics. A third (tilted) system is the vertical Xenon flashlamp excitation optics. The last is the sampling flow tube system, which is set in a skew plane and is interrupted by a gap. The axis of this system passes through the center of the common region.

The action is as follows: As each particle passes across the laser ribbon (which is situated athwart the flow) a pulse of scattered light is received by the PM tube optical system. The latter is a two-channel forward-scatter detector; the simultaneous occurance of a signal in both channels signifies the passage of a particle, which action sets in motion the remainder of the chemical identification apparatus. (Meanwhile the two-channel scattered light detector develops the requisite information for measuring the size of the particle.)

Upon receiving a logic trigger signal, the excitation flashlamp is actuated within several microseconds, (the time delay being adjustable). After the passage of a selectable number of microseconds (depending upon the molecular relaxation and other phenomena), the illumination flashlamp is actuated. The pulse duration of both flashlamps, as well as their respective operating voltages, are also adjustable (within certain obvious limits) to favor one or more optical parameters of import.

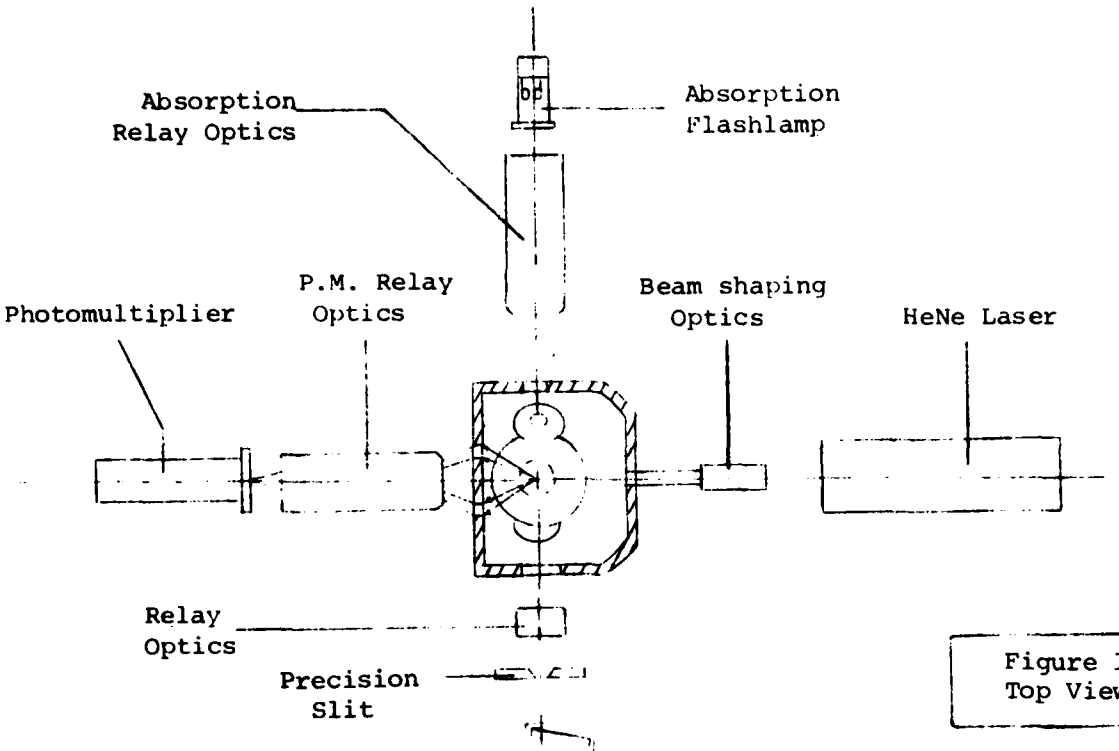
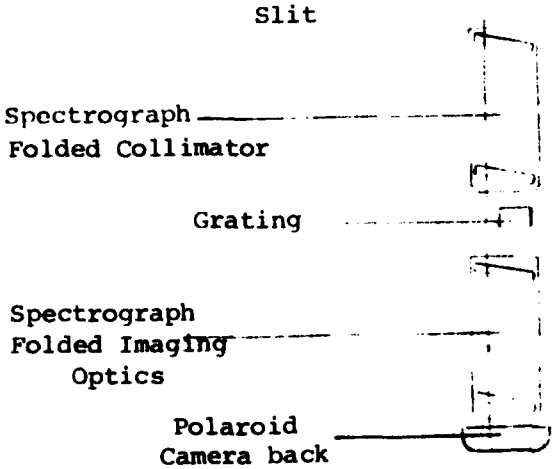


Figure 1a
Top View



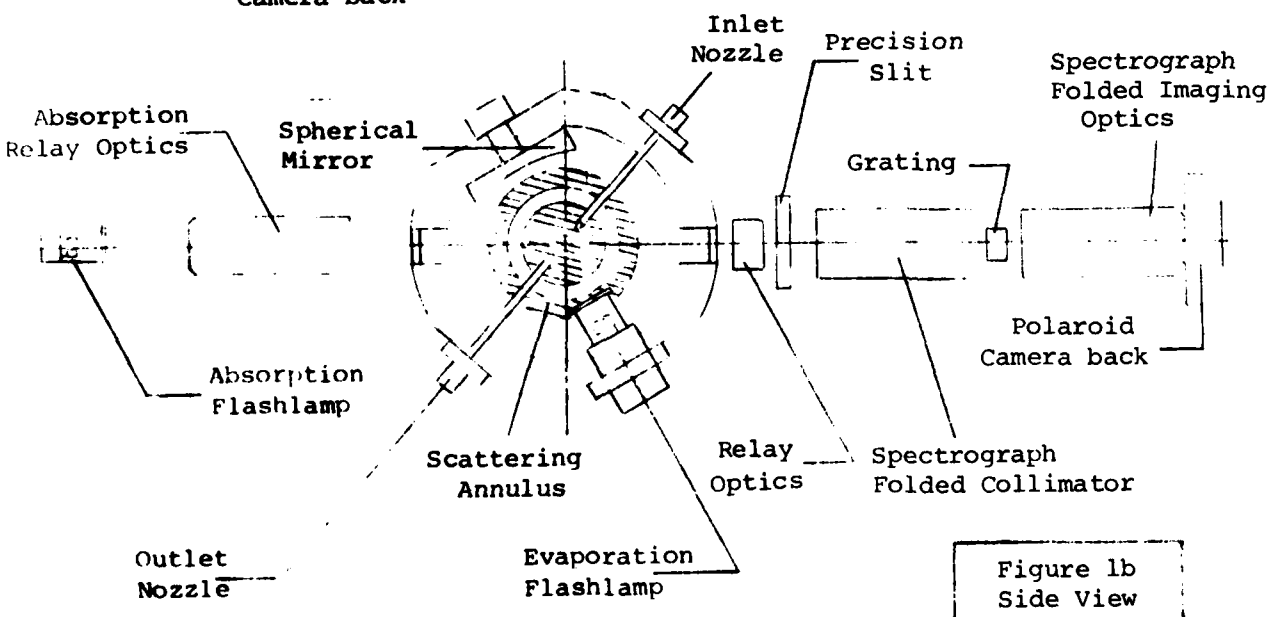


Figure 1b
Side View

With regard to the latter question, one may wish for example to optimize the UV excitation (in which case a higher voltage, short-duration flash would be used). In the case of the illumination flashlamp, one overriding criterion must be satisfied at all times: the need to produce a continuum. This places its own set of constraints upon the electrical/temporal parameters.

3. Flashlamp Modifications

The linear excitation flashlamp used in the earlier device, was replaced by the EG&G FX 265 with Quartz window. This lamp is much smaller in size (producing a cylinder of light roughly 1/16" diameter and 1/8" long), but is much more efficient in terms of output per unit area of illuminated region. Thus it is better matched to the requirements of this program, where only a very small region must be illuminated.

The flashlamp efficiency was measured to be about 6% between 2000 Å and 3000 Å, (an additional 6% of electrical input energy was found between 4000 Å and 8000 Å) for an input of 2.25 joules (1500 V; and 2 μf capacitor). With a mirror acceptance angle of $\Pi/4$ ster, an image/object reduction factor of 2.7, and a throughput of 80%, the image fluence is 5×10^{-2} joules/cm^2. This is at least an order of magnitude greater than the previous system (on a per-Å basis).

A second identical flashlamp (with different voltage and capacitor values, as discussed in subsection 2 above) is used for the illuminator. In this instance high fluence is not required per se. On the other hand, the parameters must be such as to produce a pulse whose spectral output includes a sizeable continuum component. This requirement is generally translated in terms of slightly different electrical parameters, whose values need to be determined by resort to experiment.

4. Sampling Inlet Tube Structure

This was modified to elminate the generation of rotary motion of the inlet airflow. Indication of this behavior had been suggested by the existence of pulse-shape pecularities for particles transiting the HeNe laser ribbon sensing gap, the onset of which peculiarities seemed to be correlated with flow rate. The corrective measure consisted of replacing the original U-shaped sampling tube with a straight-through configuration. (This latter configuration was facilitated by elmination of the CO_2 laser as well as other modifications relating to optical parameters in the interaction chamber).

An improved nozzle design was also implemented. Observations of the flow conditions using cigarette smoke illuminated as it transited the sensing zone were made which permitted detailed observation of the flow dynamics in the plane of the HeNe ribbon illumination beam. Visual inspection of the flow region revealed greatly reduced turbulence within the sensing volume: eddy currents observed in previous apparatus were entirely eliminated. These observations were then corroborated by the shape of the scattered light pulses (generated by the passage of each particle as it crosses the gaussian illumination profile), which were found to be considerably smoother.

The interposition of funding and other constraints noted above forced a curtailment of effort before arriving at the point in time where requisite checkout tests of the modified breadboard design could be completed. For the same reason it was also not possible to perform that portion of the effort relating to the extraction of engineering data permitting more refined specifications of the flight prototype instrument to be generated.

IV. ENGINEERING DATA

1. Flight Prototype Specifications

As noted previously, circumstances beyond our control precluded the execution of those engineering tests required for developing a detailed set of specifications. Despite this, however, it is possible to establish the general features of the device. Also certain isolated items of a more specific nature have emerged from the experimental work performed to date; this information has been incorporated for the sake of completeness. Thus the material presented below is of a somewhat inhomogeneous nature, certain items containing much more detail than others.

The instrument will:

1) be balloonborne, and designed to function in the troposphere and the stratosphere,
2) sample the ambient air at a known rate and measure the concentration of particulate matter utilizing forward scattered light from a suitable laser source,
3) detect their presence individually over a nominal concentration range of 0.1 - 1000 per cm^3,
4) break up the particulate sizes into a set of adjacent sizing bins over the nominal size range between 0.25 μm diameter and 1.0 μm diameter. (The number of such bins shall be sufficient to determine size distributions which, when fitted to a power-law, exhibit slopes steeper than $(D/D_o)^{-15}$),
5) include an on-board tape recorder having a capacity compatible with flight duration, and having the capability of recording all the scientific sizing data in digital form, as well as sampled housekeeping data, (incorporate on-board pre-processing of data to the extent deemed feasible),
6) provide a separate output compatible with the AFGL digital telemetry system for use as a data recording back-up, and as a real-time quick-look adjunct for flight parameter adjustments on ascent/descent,
7) collect spectroscopic data on selected particles in real time, and record in a manner that permits subsequent analysis to determine the chemical constituency of each said particle,
    a. employ a suitable criterion (e.g., random, sequential, or size-related) for particle selection (the collection of sizing infor-

mation is to be computed and digested prior to initiation of spectroscopic data processing),

b. illuminate particles selected under criterion (7a) with a xenon flashlamp delivering a fluence of about 1 x 10^{-5} joules/cm^2 Å,

c. illuminate, (after a suitable delay), with a second xenon flashlamp yielding a suitable spectral continuum component to permit identification of photodissociation products by means of absorption.

8) record sizing information of each particle for which spectroscopic data are collected, in a manner that permits adequate correlation of chemical constituency with particle size distribution,

9) emphasize (initially) the following materials:

H_2SO_4

HNO_3

H_2O

chlorofluoromethanes (F-11; F-12)

10) determine the relative content of the various constituents to the extent possible.

2. Discussion

Certain of the specifications are of a provisional nature, stemming as they do from our own best judgement on the matter. Owing to the basic research aspects of this work, there exist a number of items whose definitions are necessarily unclear. A specific case in point is Item 4, which we believe to be of singular importance, namely: the stipulation of a capability for observing such a steeply-sloping size distribution as $(D/D_o)^{-15}$. This represents a very significant departure from what would seem to be prudent if one were to restrict one's perspective to be in conformity with the perception common to most workers in this field, namely, that the observed size distributions in the stratosphere are much less steep.

In contrast to this view, our past work on this specific point (wherein extremely careful pains were taken over a protracted period to examine very critically those judgemental aspects from which controversy could have arisen) gives strong support to the existence of very steep size distributions.

Additional support for our own view has surfaced in the form of a critical in-house review and examination of our prior data (Ref. 2). This work, which

Ref. 2: Dearborn, Frank K., "Aerosol Measurements With a High Resolution Spectrometer - A Summary Report", AFGL-TR-82-0235, Final Report, August 1982, ADA123691.

was largely motivated by the desire to correct residual size and concentration errors arising from flow meter behavior and related effects in the balloon flights of 1973/75; produced unexpected fruit in two areas which would have remained hidden in the absence of high sizing resolution: 1) a remarkable degree of uniformity, persisting over a long time span, for particles in the neighborhood of 0.3 μm diameter, and 2) evidence of particle growth by multiple successive binary coagulation events involving a single small kernel particle size.

On the other hand, the acceptance of steep size-distribution slopes (with the attendant stringent resolution requirement as manifested in Item 4), introduces added complexity and expense to a molecular identification device. Thus in spite of such added support for the validity of our views, prudence would seem to dictate a search for consensus as an integral part of the overall design effort. The stimulation of a substantive discussion among members of the scientific community would have served to resolve this exceedingly important point of apparent fluidity prior to freezing the particular numeric value of Item 4. However, this discussion did not take place. Nevertheless, in spite of apparent consensus to the contrary, we find no reason to dispute our assertions: An analysis has been published showing the detailed origins of several specific instrumentation deficiencies that render instrumentation prone to yielding drastically erroneous size distributions (Ref. 3), and that are known to be present to a greater or lesser extent in devices currently being used by other prominent workers in the field.

In summary, we believe that any relaxation of Item 4 in instrument design specifications be approached with caution.

Ref. 3: H. A. Miranda, Jr., "Instrumentation Design Factors Affecting the Accuracy of Particulate Size Distribution Measurements", Pg. 361, Atmospheric Aerosols: Their Formation, Optical Properties, and Effects, A. Deepak, Ed., Spectrum Press, (1982).