AN ABSTRACT OF THE THESIS OF

Chris Shinya Tomine for the degree of Doctor of Philosophy

in Mechanical Engineering presented on October 19, 1973

Title: Integrated Forward Light Scattering As A Measure

of Aerosol Mass Concentration.

Abstract approved: **Redacted for privacy**

Dr. Richard W. Boubel

A mathematical model was developed that relates the
ratio, R, of two light flux measurements to the mass con-
centration, m, of the atmospheric aerosol particles between
a source of parallel light (e.g., the sun) and the measur-
ing system. R is defined as the ratio of the light fluxes
passing through the pinhole and open positions of an iris
diaphragm located in the focal plane of a lens system.
The following relationship between the two variables was
derived:

$$R = 1/(1 + Km)$$

where K is a constant that depends on the area of the lens
surface, the mean wavelength of the incident light, the
thickness of the turbid layer of air through which the
light travels, and the parameters of the assumed Junge
particle size distribution, including the specific gravity
of the aerosol.

An experimental investigation was conducted under limited laboratory conditions. The results were in agreement with the functional form of the mathematically derived relationship. Quantitative agreement was not achieved between the experimental and mathematical values of K due to the dissimilarities in the experimental and atmospheric aerosol size distributions.

Integrated Forward Light Scattering as a Measure of
Aerosol Mass Concentration

by

Chris Shinya Tomine

A THESIS

submitted to

Oregon State University

in partial fulfillment
of the requirements for the
degree of

Doctor of Philosophy

June 1974

APPROVED:

Professor of Mechanical Engineering in charge of major

Head of Department of Mechanical Engineering

Dean of Graduate School

Date thesis is presented _Oct. 19, 1973_

Typed by Ellie Peck for Chris Shinya Tomine

ACKNOWLEDGMENT

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

INTEGRATED FORWARD LIGHT SCATTERING AS A MEASURE OF AEROSOL MASS CONCENTRATION

I.   INTRODUCTION

Statement of Problem

From common experience it is known that an aerosol (a suspension of small particles in the air) introduced into the path of a light wave results in a scattering of a certain fraction of the light from its original path. For example, on clear days shadows are sharp and distinct due to the nearly parallel rays of the sun. However, on hazy, foggy, or smoggy days, i.e., days when there are many particles in the air, shadows become more and more diffuse with increasing aerosol concentration as light is scattered away from its original path and into the shadow area. This sunlight that is scattered from its original path appears to come from all directions and is called "air-light" (Middleton, 1952). Since the amount of air-light is apparently related to the aerosol concentration, the question arises: is there a monotonic, well-defined relationship between the air-light flux and the aerosol concentration?

Objective and Scope of Investigation

The objective of this dissertation is: (1) to develop
a mathematical model relating quantitatively, the air-light
flux and the aerosol concentration for atmospheric aero-
sols, and (2) to investigate the relationship experimentally
in the laboratory.

The scope of the experimental investigation is limited
to results obtained in a confined laboratory test chamber
with aerosols produced with a nebulizer-type generator,
and aerosol concentrations measured with a Sinclair-Phoenix
aerosol photometer in units of milligrams of aerosol per
liter of air. The mathematical model is based on an em-
pirical aerosol size distribution deduced from measurements
made in the real atmosphere. It is beyond the scope of
this investigation to produce such a distribution, however,
the distribution used will allow conclusions to be reached
concerning the effect of narrowing the range of particle
diameters in the aerosol. The mathematical model developed
in this manner however, has potentially a much greater
applicability, albeit beyond the limits of this experiment.

II. BACKGROUND

Light Scattering-Qualitative Description

The mechanism of light scattering is relatively simple
to describe qualitatively. When light, an electromagnetic
wave, is incident on a particle, and has a frequency that
does not approach any of the resonant frequencies of lat-
tice vibration or electronic transition, then the electrons
in the particle are forced to oscillate at the same fre-
quency as the impressed electromagnetic oscillation. This
follows directly from the fact that the electron, bearing
an electrical charge, experiences a force in the electric
field parallel to the field, and in the magnetic field,
perpendicular to the field. Since light consists of an
electric field and a magnetic field perpendicular to each
other that oscillate at the same frequency, the electron,
or any other charged particle under its influence, will be
forced to oscillate at the same frequency. These acceler-
ating electrons in turn radiate light in all directions
of the same frequency, as any accelerating charge will do.
The intensity of this re-radiated or scattered light, how-
ever, is not the same in all directions. Light from each
of the electrons in a given particle originate at different
locations within the particle and thus, pathlengths to any

observation point and the phase relationships among the waves will vary. If the angle of scattering is such that the waves from individual electrons are in phase, then the superposition of the waves results in an intensity maximum. If the angle is such that the waves are 180° out of phase, then an intensity minimum results. Light scattered by small particles then, is omni-directional, but varies in intensity with the scattering angle.

Light Scattering - Quantitative

A rigorous mathematical description of the phenomenon of light scattering by small particles requires the solution of Maxwell's equations of electromagnetic theory. The solution must then be subjected to the requisite boundary conditions on the electric and magnetic field vectors at the particle-air interface. This problem can be reduced to the solution of a three-dimensional vector wave equation, which is solved most easily in spherical coordinates due to the nature of the solution, i.e., an out-going spherical wave (Stratton, 1941; Van De Hulst, 1957; Kerker, 1969). The application of the boundary conditions, in actual practice, becomes extremely complex except in the simplest case of a spherical particle. The details of this problem of light scattering by small (dimensions on the same order of magnitude as the wavelength of the light) spherical particles

were worked out by Gustav Mie in 1908, following the treatment of Debye's 1908 thesis. These solutions, however, are most commonly referred to as the Mie solutions. These Mie solutions give the intensity of scattered light as a function of the scattering angle, θ, the index of refraction, n, and the parameter $\alpha = \frac{2\pi r}{\lambda}$, where r is the particle radius and λ is the wavelength of light. The form of these solutions, however, is a very slowly converging infinite series of associated Le Gendre polynomials, spherical Bessel functions and exponential functions, and consequently, numerical values in closed form were not obtainable until the advent of high speed computers in the mid-1940s. Since then, numerous tables of the numerical solutions for various values of the parameters and n, have been prepared. Typical scattering patterns calculated from the Mie solutions are shown as plots of intensity vs scattering angle in Figure 1.

Several conclusions can be drawn from these figures: (1) angular maxima and minima in the intensity are present as predicted by the qualitative arguments, (2) the number and angular location of the maxima are dependent on r and λ through α, and (3) there exists an omnipresent and predominant forward scattering lobe (in the 0° direction).

As indicated, the preceding discussion applies only to single spherical particles. The problem of scattering from single particles of arbitrary, or even regular but

α=1.0

10¹

10⁻²

60°   120°   180°

scattering
angle

scattered intensity (arbitrary units)

i₁
i₂

total
scattered
intensity = i₁ + i₂

α=3.0

10²

10¹

10⁰

10⁻¹

10⁻²

60°   120°   180°

scattering
angle

10⁴

10³

10²

10¹

10⁰

10⁻¹

α=10.0

60°   120°   180°

scattering
angle
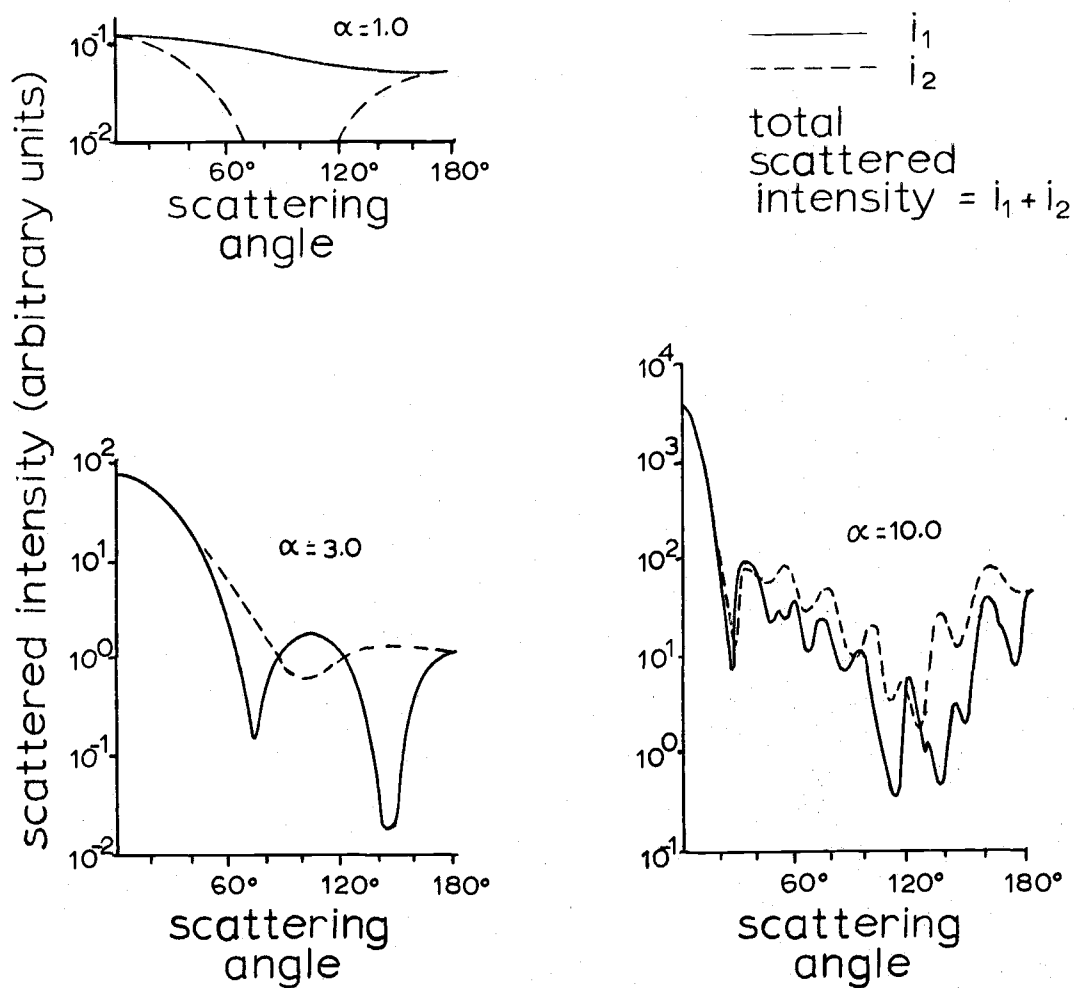
Figure 1. Angular distribution of monochromatic light
scattered by single spheres of refractive index
1.5.

non-spherical shape becomes almost impossible to solve.
Hodkinson (1966) however, has determined that a large num-
ber of randomly oriented non-spherical particles together
as a group has a resultant scattering pattern that closely
approximates that of a polydisperse (i.e., of many dif-
ferent diameters) assembly of spheres of equal mean volume.
The explanation for this lies in the fact that the scatter-
ing pattern of an assembly of particles is a simple super-
position of the scattering patterns of the individual
particles, and the smoothing effect that the variation in
orientation produces on the scattering pattern of the
assembly of non-spherical particles is comparable to the
smoothing effect that the variation in sizes produces in
the assembly of spherical particles. These smoothing
effects are a direct result of the superposition of the
scattering patterns from the individual particles. In the
case of the spherical particles, different particles have
scattering patterns that differ in the number and location
of its maxima and minima by virtue of the differences in
the diameters. As a result, a large assembly of such par-
ticles has a combined scattering pattern that loses its
rapidly oscillating maximum and minimum intensities
(Figure 2). Hodkinson's conclusion implies then that an
assembly of randomly oriented non-spherical particles
scatters light in a manner that is similar to a polydis-
perse assembly of spherical particles and therefore
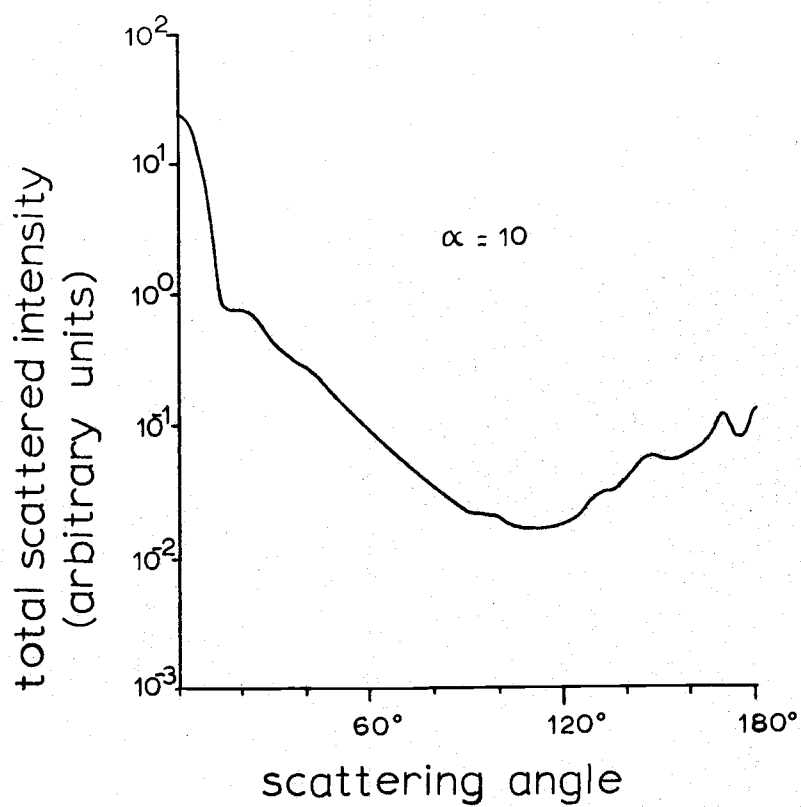
Figure 2. Angular distribtuion of monochromatic light by poly-disperse spheres of refractive index 1.33.

following the Mie theory. Hodkinson adds further that if
the smoothing effect of polychromatic light is added to
that of the orientation and size polydispersity, the
resemblance of the two scattering patterns becomes even
closer (recall that the pattern of maxima and minima de-
pends on λ as well as diameter). Cadle (1965) reaches the
same conclusion for light scattering in the forward lobe.

Thus, using the Mie solutions as guides, certain
conclusions can be drawn concerning the passage of light
through an aerosol "cloud." First, the intensity of the
scattered light is the greatest about the forward direc-
tion. This is due both to the constant occurrence of the
forward lobe in the scattering patterns of all particles
and also to the large relative magnitude of the forward
lobe. Hodkinson (1966) estimates that the light flux in
the forward lobe is about 84% of the flux geometrically
incident on the particles. The second conclusion is that
the occurrence of the forward lobe is independent of λ,
n or d (the magnitude, of course, is not). The third
conclusions follows from the first, and it is that in the
forward direction, multiple scattering becomes relatively
negligible, i.e., the addition of light that has been
scattered successively by two or more particles into the
forward direction, to the light that has been scattered
only once into the forward direction, is negligible. It

is evident that the intensity of the once-scattered light
is much greater than that of the twice-scattered light.
The difference becomes proportionally greater for higher
orders of multiple scattering. The subtraction of light,
however, from that which has been scattered only once into
the forward direction, due to subsequent scattering, is
not negligible, but can be accounted for quite simply by
the well known exponential attenuation law. This law
(Cadle, 1965; Hodkinson, 1966; Bullrich, 1964; Van De
Hulst, 1957), which is also known as Bouguer's or Beer's
or the Beer-Lambert law, states that light is removed
from a beam in passing through a turbid medium in such a
manner that:

$$f = f_0 exp-bl \qquad (1)$$

where: f = light flux at distance l
f_0 = light flux at distance 0
b = attenuation coefficient
l = pathlength of light

The fourth conclusion is that the use of white light would
make the Mie solution (spherical particles) more applicable
to arbitrary particles.

The conclusions suggest that the measurement of air-
light would be most feasible in directions making only
small angles with the incident light and with polychromatic
or white light.

It should be noted that the term "forward scattered light" as used in the preceding discussion should not be confused with or be interpreted as referring to the "unscattered light." These two terms must be strictly differentiated in any application of light scattering or extinction principles. When a beam of light passes through a volume of air containing suspended particles, some of the light is scattered by the particles but some of the light passes through without encountering any particles in its path. The proportion of the light that is scattered of course depends on the concentration of the particles in the volume. Of the light that is scattered, some will be scattered in the forward direction, i.e., in the same direction as the original beam and therefore in the same direction as the unscattered light. Since the Mie solutions and the scattering patterns are applicable to and predict only the intensities of the <u>scattered</u> light, and since the exponential light attenuation law is applicable only to the <u>unscattered</u> light, it becomes absolutely essential to distinguish between the forward scattered light and the unscattered light when applying either or both of these mathematical descriptions to light measurements in the forward directions.

Applications to Aerosol Measurements

The principles of light scattering have been applied
quite extensively to the measurement of various aerosol
parameters. True scattering measurements, i.e., measure-
ments of the scattered light, however, have been largely
limited to single particle measurements or to measurements
on a very small volume of aerosol-laden air, both within
the confines of an instrument test chamber. Measurements
in the atmosphere over extended distances, on the other
hand, have generally been extinction measurements, i.e.,
measurements of the light that remains after scattering
removes a certain fraction of the original light.

Sinclair (1953, 1967) describes a method for measur-
ing aerosol mass concentrations in which aerosol-laden
air is passed through the focal point of a converging
light beam so that only one particle at a time can enter
the zone where it can scatter light. A photomultiplier
tube is placed opposite the light source on the axis of
the light beam, but normally registers no light because it
is just covered by a shadow created by an opaque screen
placed on a part of the condensing lens. When the par-
ticle passes through the light beam, between the condensing
lens and the photomultiplier tube, a pulse of light is
scattered into the shadow region and thus registers on the
photomultiplier tube. The size and frequency of these

pulses indicate the aerosol mass concentration. Similar

methods are discussed by Gucker and Rose (1954) and

Clarenburg and Princen (1963).

A particle analyzer described by Gucker (1956) util-

izes a similar principle, except that the photomultiplier

tube is at right angles to the axis of the converging

light beam, and the necessity of the opaque screen is

eliminated. O'Konski, Briton and Higuchi (1958), Fisher,

Katz, Lieberman and Alexander (1955), and Zinky (1962) have

also described 90° scattering instruments.

30° aerosol counters have been designed by Berek,

Maennchen and Schaefer (1936), Stuke (1955), and Stuke and

Rzeznik (1964).

45° aerosol counters have been designed by Koshi

(1958), Thomas and Oglesby (1959), Thomas, Bira, Collins

and Rice (1961) and Randall and Keller (1965).

The integrating nephelometer of Beutell and Brewer

(1949) and Charlson et al. (1967) measures the scattering

coefficient of aerosol-laden air by illuminating a four

liter volume of the air with a zenon flash lamp and sensing

the light scattered into a photomultiplier tube placed at

right angles to the flash. The measured value of light

flux in this instrument is that which is integrated over

all of the particles in the volume sensed by the photo-

multiplier tube, as well as all of the angles of acceptance

of the photomultiplier tube.

As stated earlier, all of these light scattering studies referenced in the preceding paragraphs concern point measurements, i.e., measurements made on the air from only one point or location in the atmosphere. One exception is the work of Collis et al. (Collis and Ligda, 1964; Collis, Fernald and Ligda, 1964) who have utilized back-scattering from a Q-switched ruby laser beam as an optical radar for detecting discontinuities in the atmospheric aerosol loading. Quantitative measurements of the aerosol concentration, however, are not obtained.

The most common light measurements over extended distances in the atmosphere are the extinction measurements. Volz (1959) describes a photometer that in essence measures the extinction of sunlight due to its passage through the atmosphere. By measuring the flux of unscattered light at ground level and utilizing estimates of the fixed value of extra-terrestial solar flux and its path length based on the elevation angle of the sun at the time of measurement, the attenuation coefficient of the atmosphere is obtained from the exponential attenuation law (Equation 1).

The transmissometer (Rockney, 1959) operates on the same principle as the Volz sun photometer except that a ground level collimated light source is used instead of sunlight.

One study, that of Chinn et al. (1955) merits further discussion due to its similarities to this dissertation. Chinn proposes a method for obtaining particle size distributions for particles that are large compared to the wavelength of the incident light, by illuminating the test atmosphere with a collimated light source and measuring the scattered flux contained in the forward cone as a function of the cone angle. Using a mathematical model based on a modified Bouguer's law and an involved graphical differentiation and integration of plotted functions of the measured quantities, the size distribution is obtained. The similarity of Chinn's study to this investigation lies in the similar but independently conceived optical systems (described in section IV) used to measure the scattered light, and consequently the similar measurements that are made. However, where Chinn makes measurements for many values of the cone angle, in this investigation a measurement is made at only one angle. The flux measured at this cone angle is then divided by the value of the unscattered flux and correlated directly with the desired parameter, which in this investigation is the mass concentration. The mathematical model here is not integral to the data reduction as is the case in Chinn's work. Lastly, it should be noted that the results of this investigation, although providing different information about the aerosol

than the size distribution, may have greater practical use in the field of air pollution, since: (1) air quality standards, both Federal and California, are expressed in terms of mass concentrations, and (2) Chinn's model is subject to the condition that the measured particles be large compared to the wavelength of the light. It has been shown that particles that are approximately equal to the wavelength of light have the greatest effect on health (Hatch, 1961; Hilding, 1965; Nadel, 1965) and visibility (Steffens, 1956; Steffens and Rubin, 1949).

III. MATHEMATICAL MODEL

Assumptions

The following mathematical model is based on three
assumptions. The first assumption is that the Mie solution
will be applicable to large numbers of non-spherical par-
ticles if they are randomly oriented and the light is poly-
chromatic. The second assumption is that aerosol particles
will be distributed in size according to the Junge distri-
bution. The third assumption is that the index of refrac-
tion of the aerosol will have very little effect on the
experimental results, and therefore an average value of 1.5
(Bullrich, 1964) may be used in the calculations. This
follows from the conclusion of Pueschel and Noll (1967)
that the scattering coefficient of tropospheric aerosols
is nearly independent of the index of refraction if the
aerosol has a Junge's size distribution. Bullrich (1964)
concludes that the effect of the index of refraction
diminishes as the aerosol becomes more polydisperse.
Hodkinson (1966) explains that the scattering in the for-
ward lobe is independent of the optical properties of the
particle because it is due to Fraunhofer diffraction which
arises from light passing near the particle rather from

rays undergoing reflection and refraction.

The Junge size distribution (Junge, 1955, 1958, 1963; Friedlander and Wang, 1966; Whitby and Clark, 1966; Peterson and Paulus, 1967) is an empirically determined particle size distribution based on experiments on atmospheric aerosols and is commonly used in calculations where an analytical expression is required for aerosol size distributions (McCormick and Baulch, 1962; Bullrich, 1964; Charlson, 1968). The expression is written as follows:

$$dn(r) = cr^{-\nu} d(\log r) \tag{2}$$

where: $dn(r)$ = the number of particles per unit volume
with radii between r and $r+dr$

c = a constant of proportionality depending on the total number of particles per unit volume of air

ν = a number between 2.5 and 4.0

Junge (1955, 1958) found that the value of 3.0 for the exponent ν gave the best fit to his experimental data. Bullrich (1964) states that "the majority of direct aerosol measurements and indirect optical measurements indicate that ν = 3 gives the best fit to the natural size distribution."

Due to the empirical nature of equation (2), the results developed in the following sections are not

entirely theoretical, although the word "theoretical" is used for convenience to differentiate between the analytically and experimentally obtained results.

Choice of Parameters

The first and most important parameter that must be chosen is that which provides some quantitative measure of "air-light." Some insight into this choice is provided by the discussion at the end of section II, which suggests that: (1) air-light of all wavelengths be measured (white light), (2) air-light making only small angles with the incident light be measured, and (3) scattered and unscattered light be distinguished. It will be seen that the satisfaction of requirement (3) will suggest a means for the satisfaction of requirement (2) which will subsequently lead to the definition of the parameter under search.

The requirement (3) is quite simple to achieve utilizing a lens-pinhole system as shown in Figure 3. The pinhole is located in the focal plane of the lens and is centered on the lens. The result is that only light incident on the lens parallel to the lens-pinhole axis can pass through the pinhole. Any light not parallel to the axis, focuses at a point in the focal plan whose distance from the axis, and thus the pinhole, is given by:

$$x = y \tan \theta \qquad (3)$$
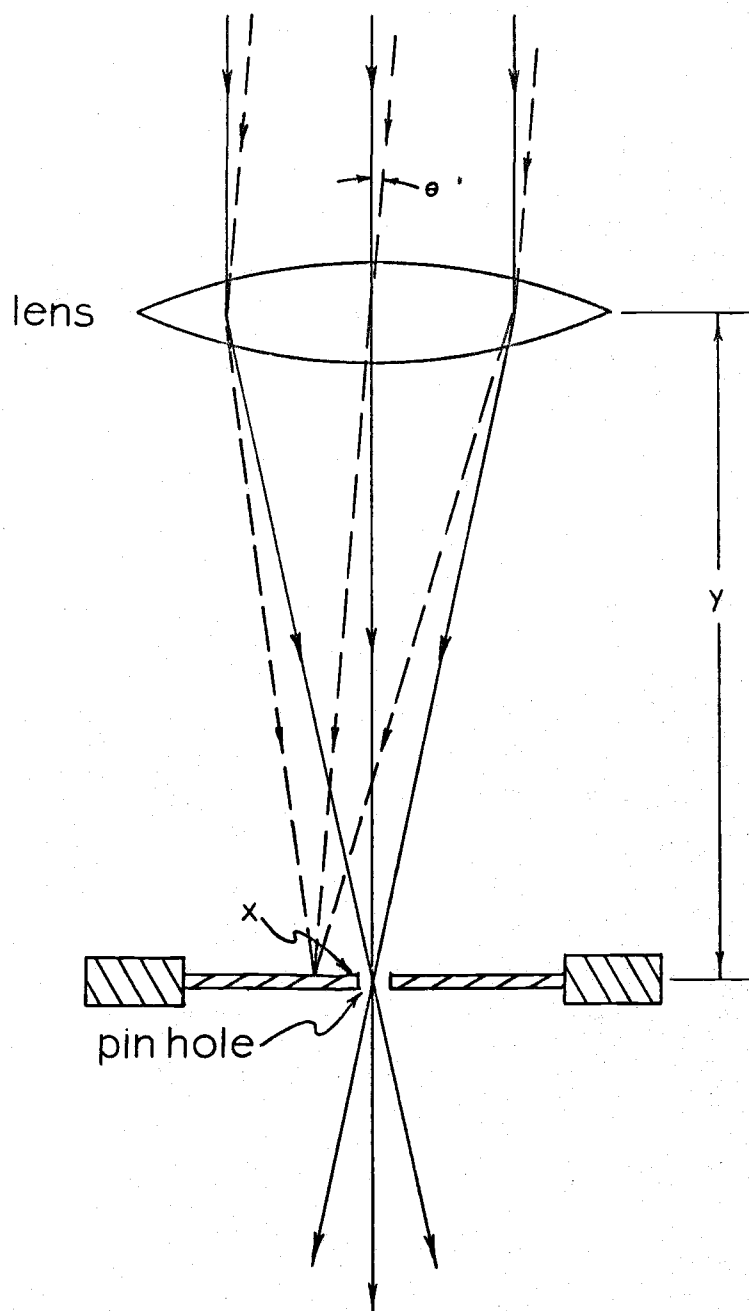
Figure 3. Lens-pinhole system.

where: y = focal length of the lens

    θ = angle of incidence of the light

If the axis of the lens-pinhole system is aligned with a
parallel beam of light, then essentially, only light that
has not been scattered passes through the pinhole. The
word "essentially" is used because a very minute fraction
of the light scattered through small angles does pass
through the pinhole partly because some of it is exactly
parallel to the incident beam but mostly because the finite
size of the pinhole does allow light that is slightly off
of parallel to pass through as well. A numerical calcula-
tion will give an indication of how small this fraction is
in this investigation. A 1 mm diameter pinhole and a
100 mm focal length lens were used which allowed light up
to an angle of 0.17° to pass through the pinhole. Figure
2 shows that the forward lobe representing about 84% of
scattered light lies within approximately 30° of the for-
ward of 0° direction. The ratio of the solid angles cor-
responding to cones of half-angles 0.17° and 30°, is only
0.00007. Thus, the scattered light that was allowed to
pass through the pinhole, as a result of its finite size,
was that which was contained in a solid angle that was only
0.007% of the forward lobe.

    If the pinhole in Figure 3 is replaced by an iris
diaphragm, then it can be opened up to admit light that

has been scattered through angles greater than 0.17°. (Of
course, this scattered light will still be in addition to
the unscattered light.) In this investigation, the dia-
phragm was opened up to admit light with an angle of
incidence up to 5°. The light that passed through the iris
diaphragm in the open position then was the unscattered
light incident on the lens plus the "air-light" incident
on the lens with angles of incidence less than 5°. A
measurement of this light flux passing through the open
diaphragm could be chosen as the parameter being sought
but the mathematical model developed in the next section
suggests that the value of the original flux, f_0, in the
term for the unscattered light (refer back to Equation 1),
can be eliminated by dividing the flux passing through the
diaphragm in the pinhole position by that passing through
in the open position. Consequently, it becomes beneficial
to choose as the parameter measuring airlight, the ratio

$$R = f_{pinhole}/f_{open} \tag{4}$$

where: $f_{pinhole}$ = light flux passing through pinhole

f_{open} = light flux passing through diaphragm in
the open position

The parameter used to measure the aerosol concentra-
tion in this investigation is the mass loading or concen-
tration, i.e., the mass of aerosol per unit volume of air,

and will be designated by the letter m. This choice is
made primarily because in air pollution work, this is the
standard parameter for specifying suspended particulate
concentrations.

Physical Description

The physical description of the model follows directly
from the definition of the ratio R. Recall that the ratio
R is defined from an operational standpoint as the ratio
of the light flux measured with the iris diaphragm in the
pinhole position to the light flux measured with the dia-
phragm in the open or 5° acceptance angle position. The
same ratio can be expressed in terms of the incident light
as the ratio of the unscattered light flux incident on the
objective lens to the sum of the scattered light flux
(scattered through less than 5°) and the unscattered light
flux incident on the objective lens, i.e.,

$$R = f_u/(f_s + f_u) \tag{5}$$

where: f_u = unscattered light flux incident on the objec-
tive lens

f_s = scattered light flux (through less than 5°)
incident on the objective lens

The relationship between the flux passing through the dia-
phragm and that incident on the objective lens has already

been discussed in the preceding section.

The flux f_u represents the portion of the light originally traveling towards the objective lens parallel to the optical system axis that actually reaches the objective lens without encountering any particles. This flux decreases with the distance between the light source and the lens since more of the light will be scattered away from its original path by the aerosol. This attenuation follows Bouguer's law (Equation 1) as is well known.

The value of the flux f_s, however, is not known, and constitutes the heart of the mathematical model. The method by which this value will be obtained is to find the contribution that a volume element of air, dV, makes to f_s, and then integrate these contributions df_s over the total volume of air from which contributions can be made.

The contribution from dV can be derived from the knowledge of how individual particles within dV scatter light. This information is provided by the Mie solutions which give the intensity of scattered light in any given direction as a function of the particle radius. Junge's size distribution gives the number of particles of a given radius in dV, and since light flux combines by simple superposition or addition, the flux scattered toward the optical system by all of the particles in dV can be obtained by multiplying the flux scattered in that direction

by a particle of radius r by the number of particles having that radius r, and integrating over all r. This light is attenuated in traveling over the pathlength between the volume element dV and the optical system according to Bouguer's law.

The total volume whose volume elements contribute to f_s is determined by the angle of acceptance of the optical system. Figure 4 shows that this total volume is a right, circular cone with its vertex located at the optical system and whose axis coincides with the optical system axis. The vertex half-angle of the cone is equal to the acceptance angle of the optical system, which in this experiment will be 5°, and the height of the cone is equal to the height of the aerosol chamber. All volume elements dV_i within this cone scatter light toward the vertex (and hence the optical system) through an angle of 5° or less which consequently is measured by a pyranometer. All volume elements dV_o outside of this cone scatter light toward the optical system through angles greater than 5° which therefore is not measured as part of f_s.

The light scattered into the optical system is slightly underestimated in choosing this cone as the volume of integration due to the finite area of the objective lens. This underestimation will be discussed in the last part of this section.
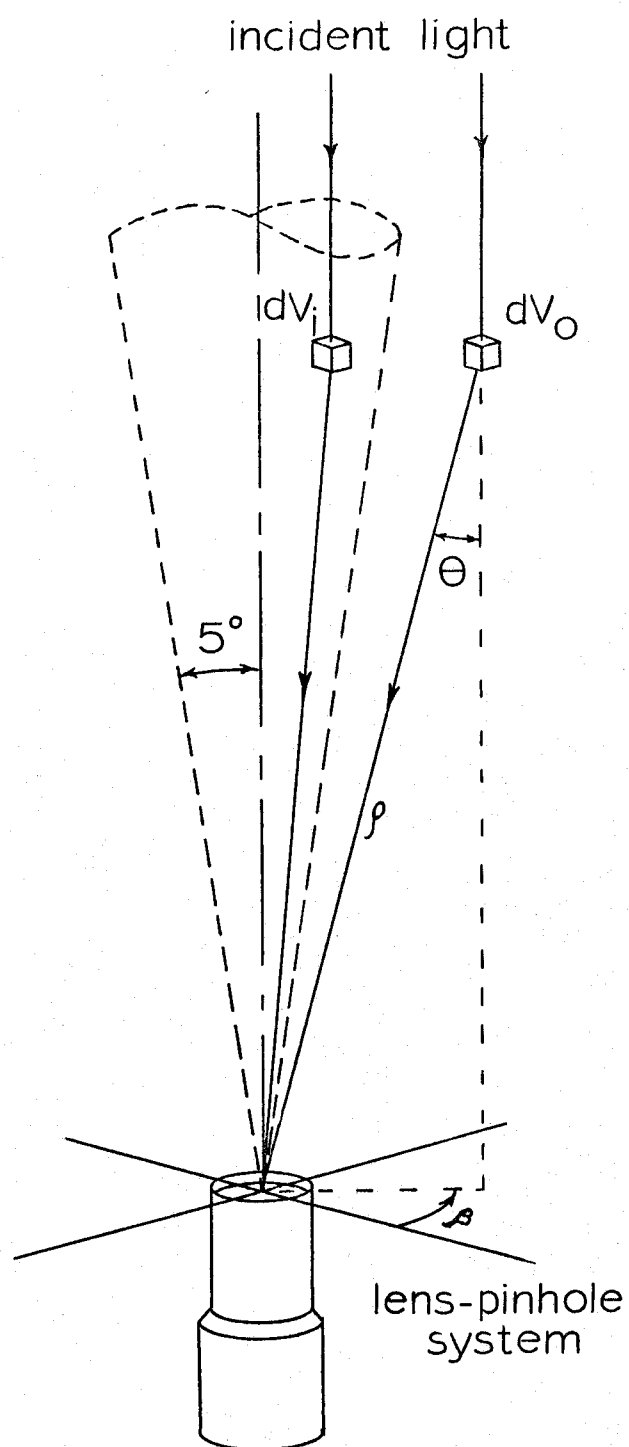
Figure 4. Volume of integration for determining f_s.

Analytical Derivation

The unscattered light flux, f_u, is given by Bouguer's law:

$$f_u = f_o \, exp\text{-}bh \tag{6}$$

where: f_u = flux at the optical system objective lens

f_o = original flux at $z = h$

h = height of the cone

b = scattering coefficient

Here, the attenuation coefficient, b, is taken as a scattering coefficient and absorption is neglected as is commonly done (Charlson, Horvath and Pueschel, 1967; Robinson, 1968) due to the colorless, transparent nature of the experimental aerosols. The height of the cone, h, in this experiment is the distance between the optical system and the top of the aerosol chamber.

In order to derive the analytical expression for df_s (the contribution to f_s made by dV), the angular scattering distribution as a function of particle radius must be multiplied by the particle size distribution and integrated over all radii. The Mie solution itself, however, is commonly expressed as $i(\phi,r)$ the scattered intensity per unit of incident flux per particle divided by $\lambda^2/8\pi^2$, where λ is the wavelength of the incident light. Intensity is

defined as the flux per unit solid angle emanating from a point, therefore, $i(\phi,r)$ must first be multiplied by f_i (the flux incident on dV), $\lambda^2/8\pi^2$, and the solid angle, Ω, subtended by the objective lens of the optical system in order to give the flux per particle scattered in the direction ϕ. The size distribution given as $dn(r)$ by the Junge equation (Equation 2) expresses the number of particles <u>per unit volume</u> with radii between r and r+dr and thus has to be multiplied by the volume dV in order to give the total number of particles in this size range in dV. Making these multiplications, the flux scattered in the direction of the optical system by the volume element dV is given by:

$$(df_s)_o = \int_r i(\phi,r) \, f_i(\lambda^2/8\pi^2)\Omega \, dV \, dn(r) \qquad (7)$$

Equation (7) gives the expression for the scattered flux leaving dV as a function of ϕ, the scattering angle. Now if the variable ϕ is replaced by the particular angle θ that locates the declination of the volume element dV from the optical system axis, the equation (7) gives the expression for the flux scattered by dV in the direction of the optical system.

$$(df_s)_o = \int_r i(\theta,r) \, f_i(\lambda^2/8\pi^2)\Omega \, dV \, dn(r) \qquad (8)$$

The flux actually reaching the lens, however, has been attenuated in traveling over the pathlength ρ according to Bouguer's law. The contribution to f_s made by the volume

element dV is then:

$$df_S = (df_S)_o \exp [-b\rho] \qquad (9)$$

or

$$df_S = \exp[-b\rho] \int_r i(\theta,r) \, f_i(\lambda^2/8\pi^2)\Omega \, dV \, dn(r) \qquad (10)$$

The flux, f_i, incident on the volume element dV is simply the original flux, f_o, attenuated according to Bouguer's law also, but in traveling from source at $z = h$ to dV. The pathlength is a function of the location of dV, and is given by:

$$1 = h - \rho\cos\theta \qquad (11)$$

where: h = the height of the cone

ρ and θ are the spherical coordinates shown in Figure 4.

Thus, the incident flux is given by:

$$f_i = f_o \exp{-b(h-\rho\cos\theta)} \qquad (12)$$

The solid angle, Ω, subtended by the objective lens of the optical system can be expressed in terms of the coordinates of dV as:

$$\Omega = A\cos\theta/\rho^2 \qquad (13)$$

where: A = the area of the objective lens

ρ and θ are the spherical coordinates that locate dV

The use of the whole area of the objective lens results in a slight overestimation of the light scattered into the optical system and tends to be cancelled by the underestimation mentioned previously. This overestimation follows from the fact that all of the light from dV incident on A will not be measured since some of it may strike A at an angle greater than 5°. A detailed discussion will be pursued at the end of this section.

The expression for the volume element dV in spherical coordinates is the usual one given by:

$$dV = \rho^2 \sin\theta \, d\theta \, d\beta \, d\rho \qquad (14)$$

where β is used as the azimuthal angle in the spherical coordinate system in place of the customary designation ϕ.

Since equation (10) involves an integration over the particle radii, it is helpful to recast the Junge equation (Equation 2) in terms of the differential dr, instead of dlogr. This is accomplished easily by first converting from base 10 logarithms to natural logarithms and using the identity, $d\ln r = dr/r$. Equation (2) then becomes:

$$dn(r) = 0.434cr^{-(\nu+1)}dr \qquad (15)$$

Grouping the terms containing r, and substituting equation (15) in equation (10) yields:

$$df_s = \exp(-b\rho)0.434cf_i\Omega \, dV(\lambda^2/8\pi^2)\int_r i(\theta,r)r^{-(\nu+1)}dr \qquad (16)$$

The integral in equation (16) can be evaluated using the results of Bullrich (1964) if a change of variables is made from r to α, where $\alpha = 2\pi r/\lambda$. Then:

$$df_s = \exp(-b\rho)0.43cf_1\Omega dV\, 1/2(2\pi/\lambda)^{\nu-2}\eta(\lambda,\theta) \tag{17}$$

where: $\eta(\lambda,\theta) = \int i'(\theta,\alpha)/\alpha^{\nu+1}d\alpha$ \hfill (18)

and, $i'(\theta,\alpha)$ is the Mie solution expressed in terms of α.

Equation (18) has been integrated numerically by Bullrich (1964) with limits of integration corresponding to radii of 0.04 microns and 10 microns. The results for an index of refraction of 1.5 are shown in Table I and graphically in Figure 5.

Substituting equations (12), (13) and (14) into equation (17) gives:

$$df_s = \exp(-b\rho)0.434cf_0\exp(-bh+b\rho\cos\theta)(A\cos\theta/\rho^2)x$$
$$\rho^2\sin\theta d\theta d\beta d\rho\, 1/2(2\pi/\lambda)^{\nu-2}\eta(\lambda,\theta) \tag{19}$$

Grouping the constant terms, equation (19) becomes:

$$df_s = K_1\exp(-b\rho-b\rho\cos\theta)\cos\theta\sin\theta\eta(\lambda,\theta)\,d\theta d\beta d\rho \tag{20}$$

where: $K_1 = f_0\exp(-bh)\, 0.434cA\, 1/2(2\pi/\lambda)^{\nu-2}$ \hfill (21)

Integrating equation (20) over the conical volume shown in Figure 4, the flux f_s is obtained:

$$f_s = K_1\int_{\theta=0}^{5°}\int_{\rho=0}^{h/\cos}\exp-b\rho(1-\cos\theta)\cos\theta\sin\theta\eta(\lambda,\theta)d\theta d\rho\int_0^{2\pi}d\beta \tag{22}$$

TABLE I

Numerical Values of the Angular Function η(θ,λ)

| θ | η | | | | | |
| | ν=2.5 | | ν=3.0 | | ν=3.5 | |
	λ=.55	λ=.65	λ=.55	λ=.65	λ=.55	λ=.65
1	324.05	286.09	54.91	51.21	12.56	12.20
4	64.16	63.92	19.77	19.75	7.406	7.407
7	33.71	33.62	12.96	12.95	5.742	5.745
10	22.91	22.83	9.841	9.835	4.758	4.760
30	6.072	6.038	3.106	3.104	1.802	1.805
60	1.405	1.398	0.7568	0.7575	0.4821	0.4842
80	0.6263	0.6247	0.3479	0.3489	0.2306	0.2323
90	0.4514	0.4503	0.2547	0.2556	0.1719	0.1736
110	0.2843	0.2845	0.1688	0.1699	0.1190	0.1208
120	0.2600	0.2604	0.1565	0.1577	0.1119	0.1139
130	0.2662	0.2667	0.1592	0.1607	0.1135	0.1157
150	0.4323	0.4329	0.2219	0.2236	0.1414	0.1442
180	1.1613	1.1472	0.4423	0.4428	0.2213	0.2243
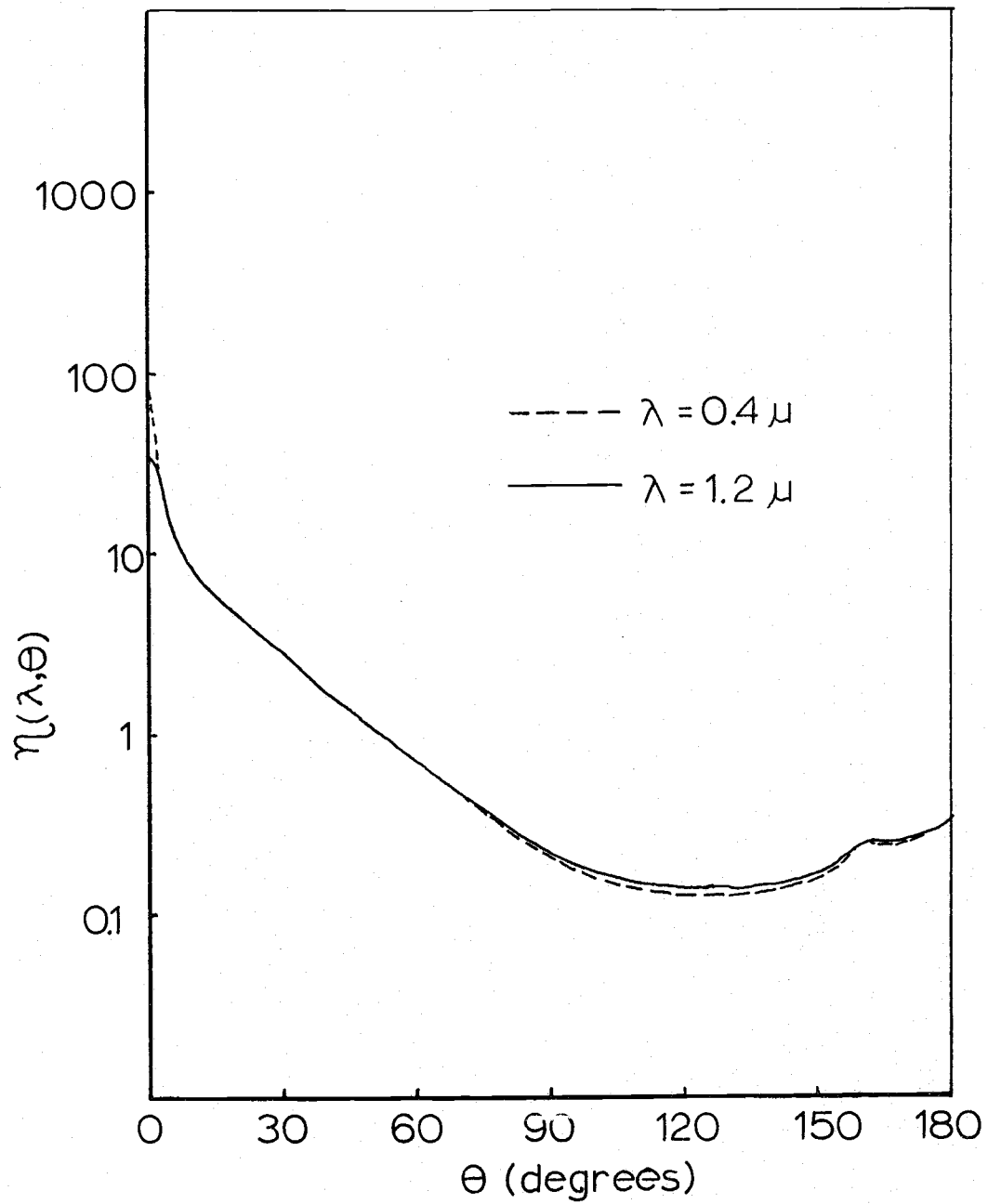
Figure 5. Angular variation of the parameter $\eta(\lambda,\theta)$ for
$\nu = 3.0$.

The integration over β can be carried out immediately to yield:

$$f_s = K_1 2\pi \int_{\theta=0}^{5°} \int_{\rho=0}^{h/\cos} \exp{-b\rho(1-\cos\theta)}\ \cos\theta\sin\theta\eta(\lambda,\theta)d\theta d\rho \qquad (23)$$

The integration over ρ yields:

$$f_s = K_1 2\pi \int_{\theta=0}^{5°} \cos\theta\sin\theta\eta(\lambda,\theta)\ \left[\frac{\exp{-b\rho(1-\cos\theta)}}{-b(1-\cos\theta)}\right]_0^{h/\cos\theta} d\theta \qquad (24)$$

or:

$$f_s = K_1 2\pi \int_{\theta=0}^{5°} \cos\theta\sin\theta\eta(\lambda,\theta)\left[\frac{(\exp{+bh(\cos\theta-1)/\cos\theta})-1}{b(\cos\theta-1)}\right]d\theta \qquad (25)$$

Substituting the series expansion for the exponential term and allowing the term $(\cos\theta-1)/\cos\theta$ to be called X for the moment yields:

$$f_s = K_1 2\pi \int_{\theta=0}^{5°} \frac{\sin\theta\eta(\lambda,\theta)}{bX}\ (1+hbX+\frac{h^2 b^2 X^2}{2!}+\dots\ -1)d\theta \qquad (26)$$

or:

$$f_s = K_1 2\pi \int_{\theta=0}^{5°} \frac{\sin\theta\eta(\lambda,\theta)}{bX}\ (hbX+\frac{h^2 b^2 X^2}{2!}+\dots)d\theta \qquad (27)$$

Cancelling bX in the numerator and denominator gives:

$$f_s = K_1 2\pi \int_{\theta=0}^{5°} \sin\theta\eta(\lambda,\theta)\ (1+\frac{hbX}{2!}+\frac{h^2 b^2 X^2}{3!}+\dots)d\theta \qquad (28)$$

or:

$$f_s = K_1 2\pi h \sum_{n=0}^{\infty} \frac{(hb)^n}{(n+1)!}I_n = K_1 2\pi h \sum_{n=0}^{\infty} S_n \qquad (29)$$

where: $\qquad I_n = \int_{\theta=0}^{5°} \sin\theta\eta(\lambda,\theta)X^n d\theta \qquad (30)$

or: $\qquad I_n = \int_{\theta=0}^{5^\circ} \sin\theta\, \eta(\lambda,\theta)\, (\frac{\cos\theta-1}{\cos\theta})^n d\theta$ (31)

and $\qquad S_n = \frac{(hb)^n}{(n+1)!}\, I_n$ (32)

The behavior of the series in equation (29) can be examined more easily if the first few integrals, I_n, are evaluated. Since only tabulated values of the function $\eta(\lambda,\theta)$ are available, and the functional form is not known, the integrals, I_n, must be solved numerically. The first four integrals in the series were evaluated using Simpson's rule with ten intervals of width 0.5° for three values of the exponent ν, 2.5, 3.0, and 3.5, and two values of the wavelength λ, 0.55 microns and 0.65 microns. The results are shown in Table II. The values for $\eta(\lambda,\theta)$ at 0.5° intervals were interpolated from an enlarged section of figure (5) between 0° and 5°.

The coefficients of the integrals in the summation can be evaluated for the average value of the scattering coefficient, b, encountered in this experiment. This value of b can be approximated using the linear relation between the scattering coefficient and the mass loading used by Charlson (1967) and McCormick and Baulch (1962). This relationship, which is presented as a conversion factor between b and m by Meteorological Research, Inc. with their commercial model of the integrating nephelometer, is as follows:

TABLE II

Values of I_n, for $\lambda = 0.55$, 0.65 Microns,
$\nu = 2.5$, 3.0, 3.5

	LAMBDA = .55 MICRONS		LAMBDA = .65 MICRONS	
	$I_0 =$	$2.3793086E\ 01$	$I_0 =$	$2.2445194E\ 01$
	$I_1 =$	$-2.9880817E\text{-}02$	$I_1 =$	$-2.9279500E\text{-}02$
NU = 2.5	$I_2 =$	$6.5791815E\text{-}05$	$I_2 =$	$6.5182517E\text{-}05$
	$I_3 =$	$-1.7653394E\text{-}07$	$I_3 =$	$-1.7547430E\text{-}07$
	$I_0 =$	$6.0235972E\ 00$	$I_0 =$	$5.8718988E\ 00$
	$I_1 =$	$-8.9117630E\text{-}03$	$I_1 =$	$-8.8250098E\text{-}03$
NU = 3.0	$I_2 =$	$2.0480619E\text{-}05$	$I_2 =$	$2.0392037E\text{-}05$
	$I_3 =$	$-5.5932891E\text{-}08$	$I_3 =$	$-5.5815547E\text{-}08$
	$I_0 =$	$1.8524265E\ 00$	$I_0 =$	$1.8322071E\ 00$
	$I_1 =$	$-3.1521791E\text{-}03$	$I_1 =$	$-3.1425358E\text{-}03$
NU = 3.5	$I_2 =$	$7.7000265E\text{-}06$	$I_2 =$	$7.6922913E\text{-}06$
	$I_3 =$	$-2.1634077E\text{-}08$	$I_3 =$	$-2.1625912E\text{-}08$

$$m = 0.38 \times 10^4 b \qquad\qquad (33)$$

where: m = mass loading in micrograms per cubic meter

b = scattering coefficient in meters^{-1}

The average value of the mass loading for the one-hundred and fourteen measurements made in this experiment is 28 micrograms per liter. Using equation (33), this gives a value of 7.36 meters^{-1} or 0.0736 cm^{-1} for the scattering coefficient.

The value of h, the height of the conical test volume, is fixed in this experiment at 178 cm.

Using these values, calculations show (see Table IIIa) that the ratio of the absolute values of the second term in the series to the first term is equal to 0.00969, and the third to the first, 9.76 x 10^{-5}, and the fourth to the first, 8.74 x 10^{-7}. This is using the integrals for ν= 3.0 and λ= 0.55 microns. It can be seen that the second and higher order terms are neglibigle when compared to the first term, especially when the alternating nature of the signs on successive terms is considered. Even if the worst possible case is considered, i.e., the highest single mass concentration measured in this experiment (0.116 mg/l, with the corresponding value of b = 0.305 cm^{-1}) and the most slowly decreasing series of integrals (ν = 3.5 and λ= 0.65 microns), it can be seen (see

TABLE IIIa

S_n and $|S_n/S_o|$, for $\lambda = 0.55$ Microns,
$\nu = 3.0$, $b = 0.0736$ cm^{-1}

n	$(hb)^n/(n+1)!$	I_n	$I_n(hb)^n/(n+1)!$	$|S_n/S_o|$
0	1	6.024	6.024	1
1	6.55	-8.912×10^{-3}	-5.84×10^{-2}	9.69×10^{-3}
2	28.7	2.048×10^{-5}	5.88×10^{-4}	9.76×10^{-5}
3	93.9	-5.593×10^{-8}	-5.26×10^{-6}	8.74×10^{-7}

TABLE IIIb

S_n and $|S_n/S_o|$, for $\lambda = 0.65$ Microns,
$\nu = 3.5$, $b = 0.305$ cm^{-1}

n	$(hb)^n/(n+1)!$	I_n	$I_n(hb)^n/(n-1)!$	$|S_n/S_o|$
0	1	1.832	1.832	1
1	27.15	-3.143×10^{-3}	-8.53×10^{-2}	4.66×10^{-2}
2	492	7.692×10^{-6}	3.79×10^{-3}	2.07×10^{-3}
3	6670	-2.163×10^{-8}	-1.45×10^{-4}	7.91×10^{-4}

Table IIIb) that the ratio of the second term to the first is still only 0.0466.

It should be noted that even the average value of b used in these calculations is extremely large when compared to values found in actual atmospheric measurements. Since light scattering is dependent on the total number of particles in the light path, the short pathlength used in this experiment, due to the physical limitations of the laboratory, is compensated for by a high mass loading. In the real atmosphere, a visibility of a half-mile would correspond to a value for b of 4.87×10^{-5} cm^{-1}. This would be considered extremely high. Using this value of b and the integrals for $\nu = 3.0$ and $\lambda = 0.55$ microns calculations show (Table IV) that the ratio of the second term to the first becomes 6.41×10^{-6}.

Neglecting then the second and higher order terms in the summation in equation (29), the expression for the scattered flux, f_s, becomes:

$$f_s = K_1 2\pi h I_o \qquad (34)$$

or substituting for K_1:

$$f_s = f_o \exp(-bh) \; 0.434cA \; 1/2(2\pi/\lambda)^{\nu-2} \; 2\pi h I_o \qquad (35)$$

On first glance it appears that the scattered flux, f_s, is dependent on the mass concentration only through the scattering coefficient in the exponential term pre-

TABLE IV

S_n and $|S_n/S_o|$, for $\lambda = 0.55$ Microns,
$\nu = 3.0$, $b = 4 \times 10^{-5} \text{cm}^{-1}$

n	$(hb)^n/(n+1)!$	I_n	$I_n(hb)^n/(n-1)!$	$|S_n/S_o|$
0	1	6.02	6.02	1
1	4.335×10^{-3}	-8.912×10^{-3}	-3.86×10^{-5}	6.41×10^{-6}

cisely as in Bouguer's law, however, a dependence does exist through the "constant" c. Recall that c was considered a constant in equation (2), the Junge size distribution equation, in that it is independent of the variable r, the particle radius, and in equation (16), it was independent of the spatial variables of integration. The dependence of c on the mass concentration, m, can be obtained by returning to equation (2). Note that in equation (2), the shape of the distribution curve is indicated by the terms containing r, but that c serves as a constant of proportionality or normalization constant that raises or lowers the curve to make the area under the curve correspond to the total number of particles in a unit volume of air. This implies that if the Junge distribution (which gives the number of particles of radius r per unit volume of air) is multiplied by the mass of one particle of radius r, then c would have that value which would make the area under this new curve equal to the total mass per unit volume of air. Using this approach, the functional dependence of c on m is obtained by first writing the expression for the mass of the particles of radius r between r and r+dr per unit volume of air:

$$dm = 4/3\pi r^3 \gamma dn(r) \tag{36}$$

where: $4/3\pi r^3$ = volume of one particle

γ = mass density of the bulk material of which the particle is composed

$dn(r)$ = number of particles of radius r between r and r+dr

Substituting equation (15) in equation (36) gives:

$$dm = 4/3\pi\gamma 0.43c \; r^{(2-\nu)}dr \qquad (37)$$

Integrating equation (37) over r yields:

$$m = 4/3\pi\gamma 0.434c\int_{r_1}^{r_2} r^{(2-\nu)}dr \qquad (38)$$

where the limits of integration are chosen to correspond with the integration of equation (18).

Transposing equation (35) gives the desired dependence of c on m:

$$c = m(4/3\pi\gamma 0.434 \int_{r_1}^{r_2} r^{(2-\nu)}dr)^{-1} \qquad (39)$$

which can be evaluated explicitly for any given value of ν.

Note that the units of the quantity c depend on the value of ν as $cm^{(\nu-3)}$, and c is dimensionless for a size distribution with $\nu = 3$.

Equation (39) can now be substituted back in equation (35) to give:

$$f_s = \frac{f_o\exp(-bh) \; 0.434A1/2(2\pi/\lambda)^{\nu-2}2\pi hI_o \; m}{4/3\pi\gamma 0.434\int_{r_1}^{r_2} r^{(2-\nu)}dr} \qquad (40)$$

or:

$$f_s = f_o \exp(-bh) K_2 \, m \tag{41}$$

where:

$$K_2 = \frac{3 \ A(2\pi/\lambda)^{\nu-2} h I_o}{4 \ \gamma \int_{r_1}^{r_2} r^{(2-\nu)} dr} \tag{42}$$

Now that the expression for f_s as a function of m has been derived, it can be substituted along with equation (6), the expression for f_u, into equation (5) to give:

$$R = \frac{f_u}{f_s + f_u} = \frac{f_o \exp(-bh)}{f_o \exp(-bh) + f_o \exp(-bh) K_2 \, m} \tag{43}$$

or on cancelling:

$$R = 1/(1 + K_2 m) \tag{44}$$

Equation (44) represents the desired model equation for the ratio R; however, the relationship between R and m is hyperbolic. In order to facilitate comparisons between the model and the experimental measurements, equation (44) can be recast into a linear form with K_2 as the slope. This is accomplished by defining a new variable:

$$R' \equiv 2 - (1/R) = 1 - K_2 m \tag{45}$$

which decreases from unity with increasing m, as did the original variable R, except linearly and with a slope of negative K_2. In terms of experimental measurements, R'

can be expressed as:

$$R' = 2-(f_{open}/f_{pinhole})$$ (46)

where equation (4) has been substituted for R, or:

$$R' = 2-\frac{f_s+f_u}{f_u} = 1 - \frac{f_s}{f_u}$$ (47)

where equation (5) has been substituted for R.

Equation (45) represents the relationship between the airlight variable R' and the mass concentration of the air based on the mathematical model developed in this section. It predicts a negative linear dependence of R' on m, with the restriction on the particles that they have a Jungian size distribution and assuming that the index of refraction of the aerosol will have a negligible effect on the relationship and consequently that the value of 1.5 will be representative.

Discussion of Inaccuracies in the Mathematical Model Due to Finite Size of Objective Lens

As mentioned in previous sections, the finite size of the objective lens in the optical system results in two inaccuracies in the mathematical model. These inaccuracies, however, have the effect of cancelling each other in part, resulting in a combined error that is small.

An underestimation of the flux f_s results from assuming that only those volume elements contained in the

cone shown in Figure 4 contribute to f_s. This assumption would be valid if the objective lens area were a point; however, the finite extent of the objective lens does allow volume elements from the volume, V' surrounding the cone, to make contributions to f_s also (see Figure 6). Recalling that the optical system was designed so that only light striking the objective lens at an angle of 5° or less is detected, it can be seen that all volume elements inside the cone V, and no volume elements outside the cone can scatter light toward a point located at the vertex of the cone through an angle less than or equal to 5°. With a finite objective lens, however, light does not have to be scattered toward the vertex of the cone in order to be detected; light scattered into any part of the objective lens through an angle of 5° or less is detected. Thus the volume whose elements contribute to f_s must be extended to include V'. All volume elements outside V', however, continue to make no contribution to f_s.

An overestimation of the flux f_s results from assuming that for each volume element, the scattered flux contained within the solid angle subtended at that volume element by the whole objective lens will contribute to f_s. This is an attempt to account for the finite extent of the objective lens or equivalently, for the solid angle containing the scattered flux, since the Mie solutions are
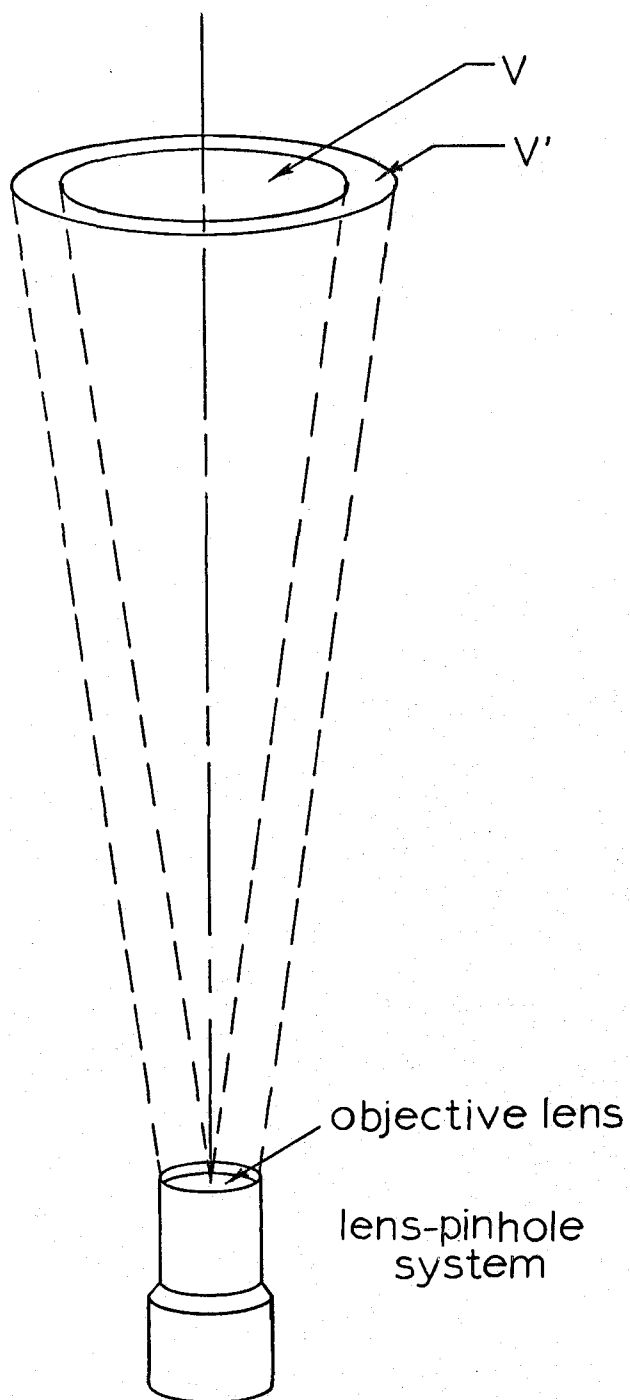
Figure 6. Extension of the volume of integration due to
the finite extent of the objective lens.

expressed in terms of scattered flux per unit solid angle. If this assumption were valid, all light rays incident on any part of the objective lens after being scattered by volume elements within the conical volume V would contribute to f_s. Some of this light, however, may strike the objective lens at an angle greater than 5° depending on the location of the volume element, and actually will not be detected by the optical system.

The degree to which these two inaccuracies cancel one another can be estimated by examining the true contributions to the measured value of f_s made by volume elements at arbitrary altitudes, i.e., by comparing the assumed with the actual contributions made by volume elements at several fixed values of z, where $z = r \cos\theta$. This is illustrated in Figure 7 for an arbitrary altitude z' greater than $z = r/\tan 5°$, where r is the radius of the objective lens. According to the assumptions made in the mathematical model, the contribution made to f_s by light scattered from the volume element dV shown in Figure 7 is that flux contained in the solid angle Ω_1 subtended by the entire objective lens. Due to the limitations of the 5° acceptance angle, however, only the flux contained in the solid angle Ω_2, or equivalently, the flux striking the intersection of the objective lens and a right circular cone of half-angle 5° emanating vertically downward from
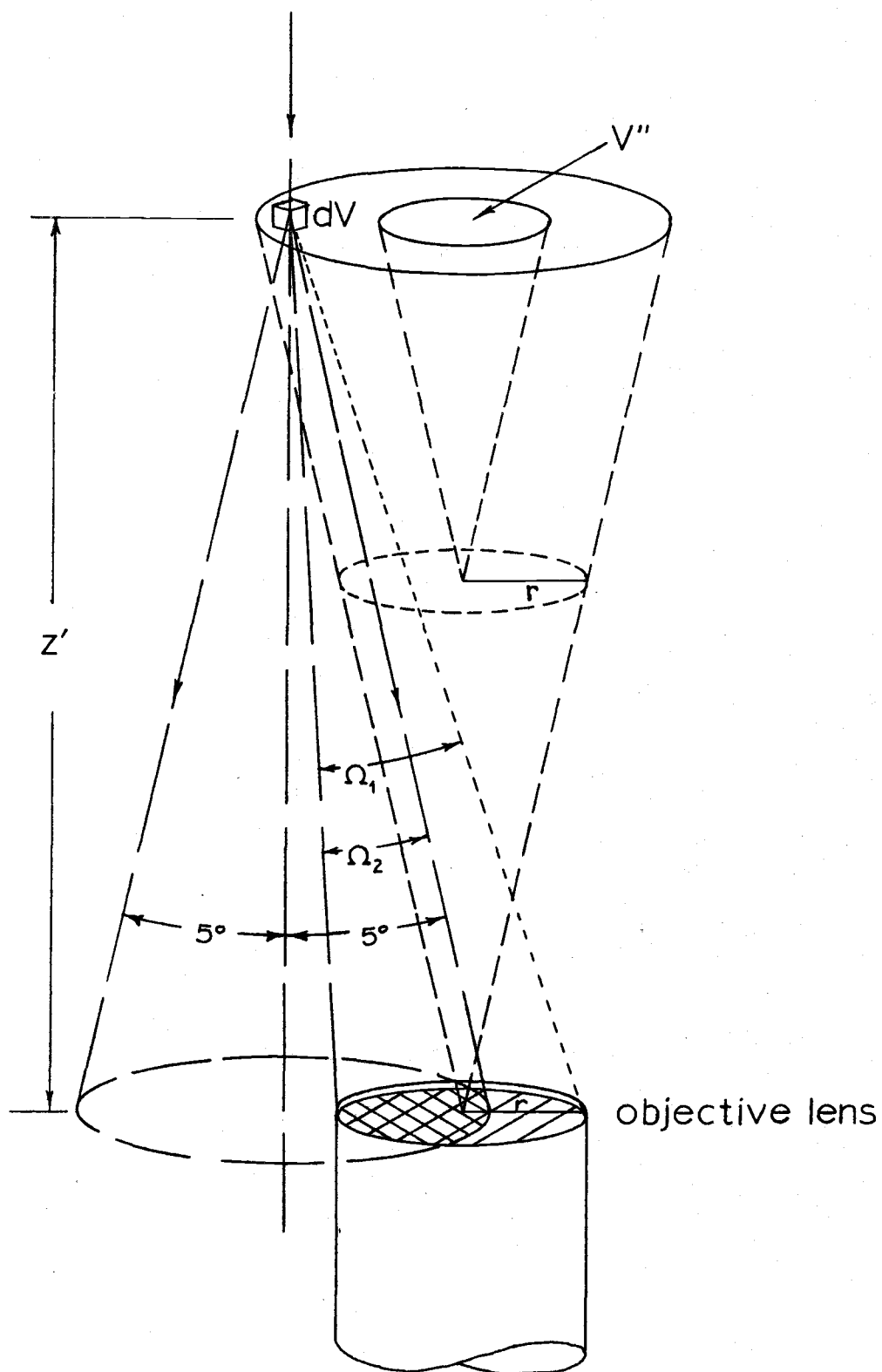
Figure 7. True contribution to f_s made by a volume element.

dV, is actually detected. This follows from the fact that
all rays from dV making an angle of 5° or less with the
vertical are confined to this downward 5° cone, and thus
only those rays within this cone that strike the objective
lens are measured. Note that for all volume elements with-
in the conical volume V" with its vertex at the altitude
$z = r/\tan 5°$ the objective lens does lie completely within
the downward 5° cone and thus the assumption of the mathe-
matical model is entirely valid. It can be seen that it
is the volume elements in the region between the cones V
and V" that produce the solid angle errors. For every
volume element dV in this region, however, there exists
a volume element dV' (in the volume V' surrounding the
cone V) in the same horizontal plane as dV, diametrically
opposite dV and located outside the perimeter of cone V
a distance equal to the distance that dV is inside the
perimeter of cone V, such that the intersection, A_1, of
its downward 5° cone and the objective lens is tangent to
the intersection, A_2, of the objective lens and the down-
ward 5° cone from the volume element dV (see Figure 8).
The result is that the surface of the objective lens
still is not completely covered by detectable contribu-
tions to f_s from dV as assumed in the mathematical model,
but the actual area covered, A_1, is supplemented by de-
tectable light from dV' striking A_2, and the sum of A_1 and
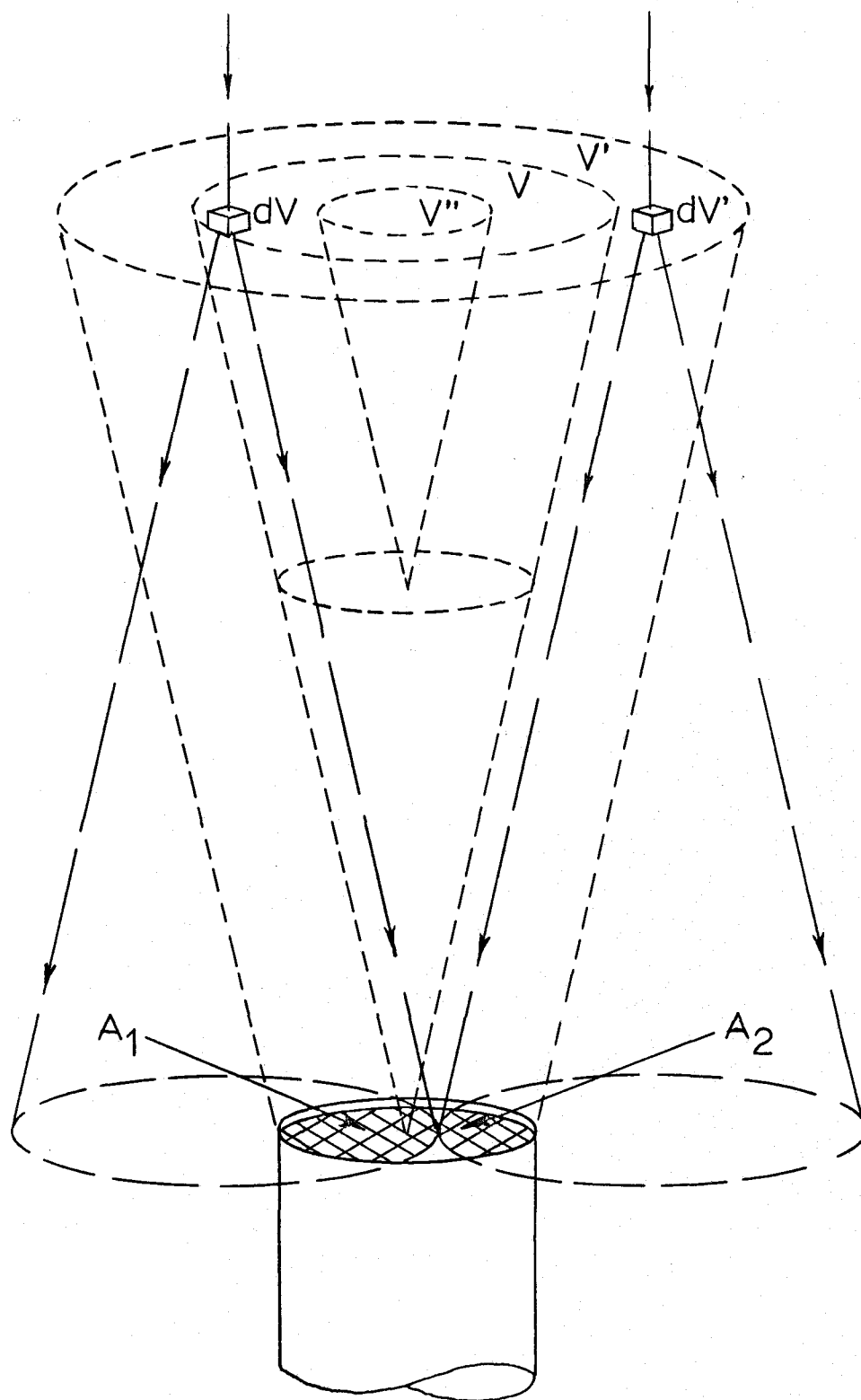
Figure 8. Combination of over- and under-estimated light fluxes.

A_2 approaches complete coverage as z increases. Thus, two

conclusions are reached: (1) the solid angle error does

not result from all of the volume elements and, in fact,

the region from which the error causing elements come,

becomes a smaller fraction of the total volume as z in-

creases, and (2) for every volume element for which such

an error results, there exists another volume element, not

considered by the mathematical model, that partially

compensates for the error, and this compensation becomes

more complete also as z increases. As an indication of

the degree of compensation, Figure 9 shows a plot of the

fraction of the objective lens receiving contributions to

f_s from a volume element just inside the perimeter of

volume V at an altitude z and from its companion element

in volume V' versus the altitude z, i.e., $(A_1 + A_2)/A_{obj}$

vs z. It can be seen that the mathematical model is in

error by approximately 50% in estimating the contributions

to f_s by volume elements inside V but outside V" at the

altitude z = 11.5 cm, but only about 10% for volume ele-

ments at z = 40 cm. At first glance, this does not

appear to be negligible; however, noting that the volume

of a cone of fixed vertex angle increases as the cube of

its altitude, it can be seen that the volume elements for

which the mathematical model is most in error (those near

the vertex of the cone V) make up only a very small per-
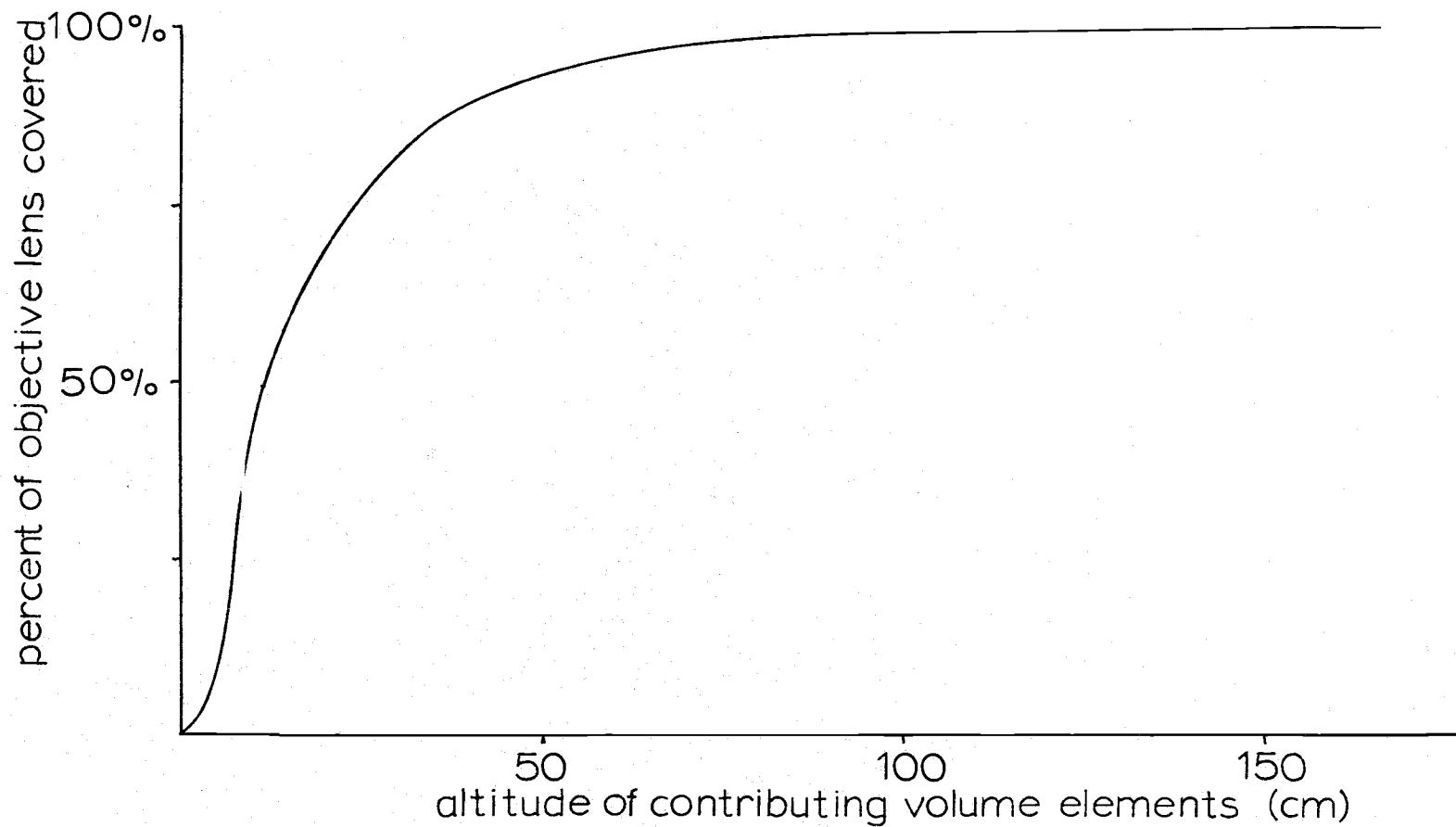
Figure 9. Degree of cancellation of over- and under-estimation of light flux.

centage of the total volume. Thus, since the altitude of the cone V in this experiment is 178 cm, for 99.97% of the total volume, the mathematical model is in error less than 50%; for 98.84% of the volume, the error is less than 10%; and for 91.08% of the volume, the error is less than 0.2%. Since the mathematical model is not in error at all for volume elements in V", 73.75% of the volume has zero error. These numbers are illustrated graphically in Figure 10.
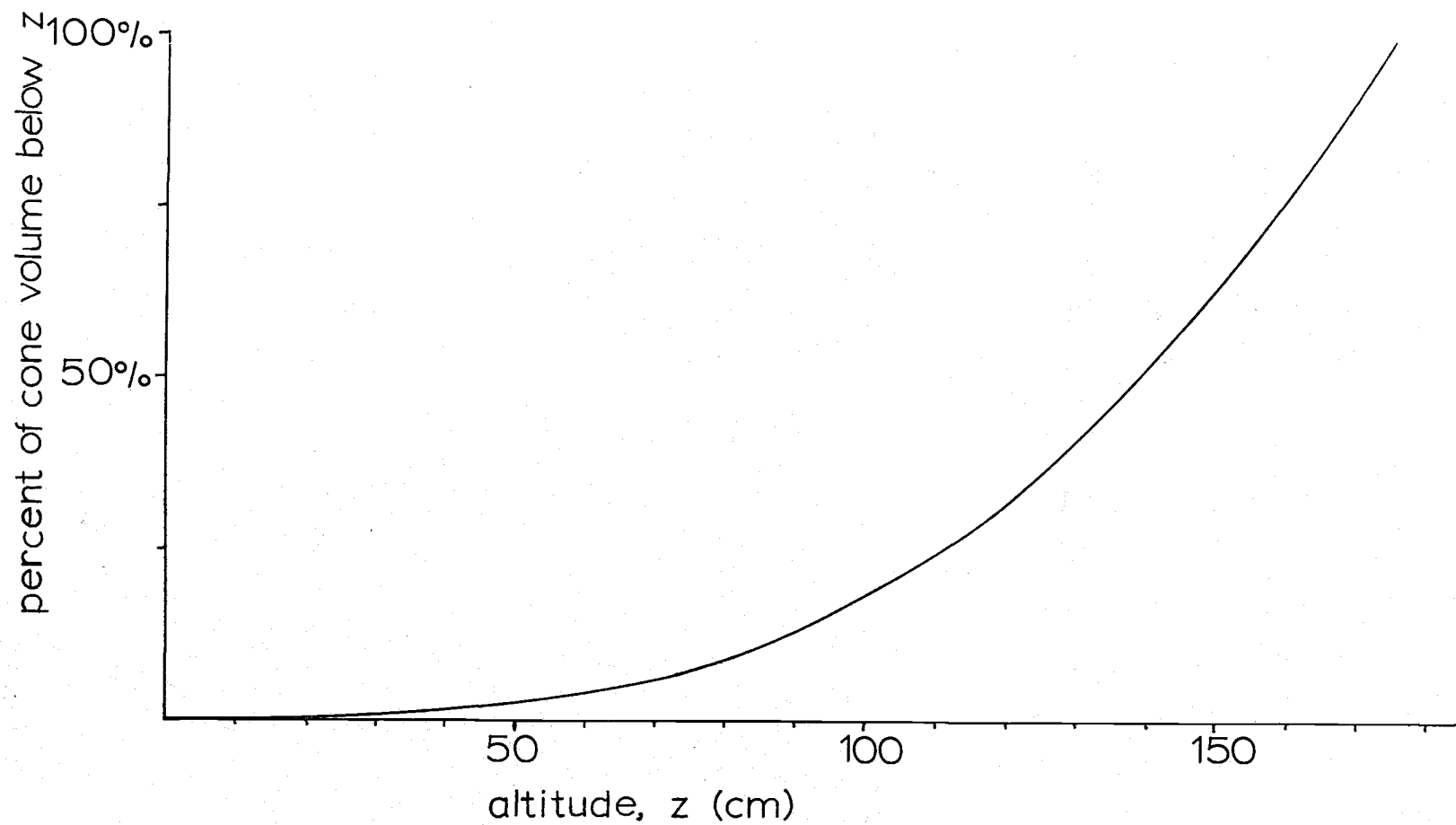
Figure 10. Percentage of volume of integration below a given altitude as a function of altitude.

IV. METHODOLOGY

Experimental Design

The design of the experiment was very simple and con-
sisted of simultaneous R and m measurements for varying
values of m for a given aerosol, i.e., a given index of re-
fraction and size distribution. This process was carried
out for nine different aerosols--three different size
distributions for each of three indices of refraction. Each
size distribution is characterized by the mean diameter.
Regression analyses were then performed to determine the
least-squares curves that provided the best fit for each of
the nine sets of data points and also the correlation co-
efficients and indices of determination.

The three aerosol materials used in this experiment
were potassium chloride, sodium chloride and ammonium
chloride. These three compounds were selected because they
are very similar in their light scattering properties ex-
cept that their indices of refraction are different. They
all have identical crystal structures (cubic), they are all
colorless, and they are all transparent. The indices of
refraction for KCl, NaCl, and NH$_4$Cl, are 1.49, 1.54, and
1.64, respectively. Thus the variations in the experimental
measurements due to particle shape and light absorption

(color) were eliminated leaving only the variations due to
index of refraction and particle size. A final considera-
tion that was made in the selection of these compounds was
their solubility in water. The method of generating the
aerosols in this experiment (see Experimental Apparatus in
this section) requires that the aerosol material be
soluble enough to make at least a 20% by weight solution
in water at room temperature. The different aerosol size
distributions are determined by the concentration of this
solution.

Experimental Procedure

In order to maintain control over the experimental
variables, the experiment was performed in an airtight
aerosol chamber (see Experimental Apparatus). The measure-
ments were made by first purging the aerosol chamber with
air that had been filtered and desiccated, and then charging
the chamber with an aerosol of a given index of refraction
and size distribution. A fan located inside the chamber
was switched on for fifteen seconds to mix the aerosol with
the chamber air, and insure a uniform concentration. Five
minutes was allowed for spurious and unusually large par-
ticles to settle out and for equilibrium to be established.
The collimated light source was then switched on at the
upper end of the aerosol chamber. The light fluxes in the

open and closed positions of diaphragm were then measured
at the lower end, and the light switched off. The mass
loading, m, of the test atmosphere was then measured with
the Sinclair-Phoenix aerosol photometer.

The chamber was again charged with the same aerosol,
i.e., more of the same aerosol was added to "old" aerosol,
to increase the concentration. This was again mixed with
the fan and allowed to stand for five minutes. The light
fluxes and m were then measured again using the same
procedure as in the first measurement.

The process of recharging and measuring the light
fluxes and m was repeated until the concentration in the
chamber approached the upper limit of the Sinclair-
Phoenix photometer.

Similar experimental runs were performed for each of
the remaining eight aerosols.

The purge air is filtered for the obvious reason of
providing a clean background for the test aerosol. It is
desicated to allow the test aerosol to exist in a dry,
solid state, with fixed diameters and indices of refrac-
tion. Junge (1958), Bullrich (1964), and Charlson (1968)
have concluded that soluble aerosols can exhibit deli-
quescent behavior when the relative humidity is approxi-
mately 70% or greater, resulting in a growth of the par-
ticles by the absorption of water from the atmosphere and
a reduction of the index of refraction toward the value

for water (1.33).

Experimental Apparatus

Aerosol Chamber. The aerosol chamber (Figure 11)
consisted of a 3/8" plywood enclosure with inside dimen-
sions of 23.5"x23.5"x70.25". The chamber was sealed at the
seams with GE-RTV silicon seal, and at the door, with 1/8"
foam insulation. All interior surfaces were painted with
flat black latex to minimize reflections. A 16"x18"x20"
expansion chamber of collapsable polyethylene was provided
to allow the removal of air for sampling without the neces-
sity of introducing replacement air which would alter the
aerosol concentration. A nine-inch diameter four-blade
fan was located in an upper corner of the main chamber to
mix the test atmosphere to insure a uniform aerosol con-
centration within the chamber. The temperature and rela-
tive humidity inside the chamber were measured by means of
a Weston model 4300 dial thermometer and a Frieze hair
hygrometer respectively. The hygrometer was first cali-
brated against a sling psychrometer and psychrometric
chart before installation. The relative humidity readings
were made through a two inch square glass window in the
chamber wall which was also sealed with GE RTV silicon
seal. Stray light was prevented from entering the chamber
through the window by a hinged opaque cover on the outside,
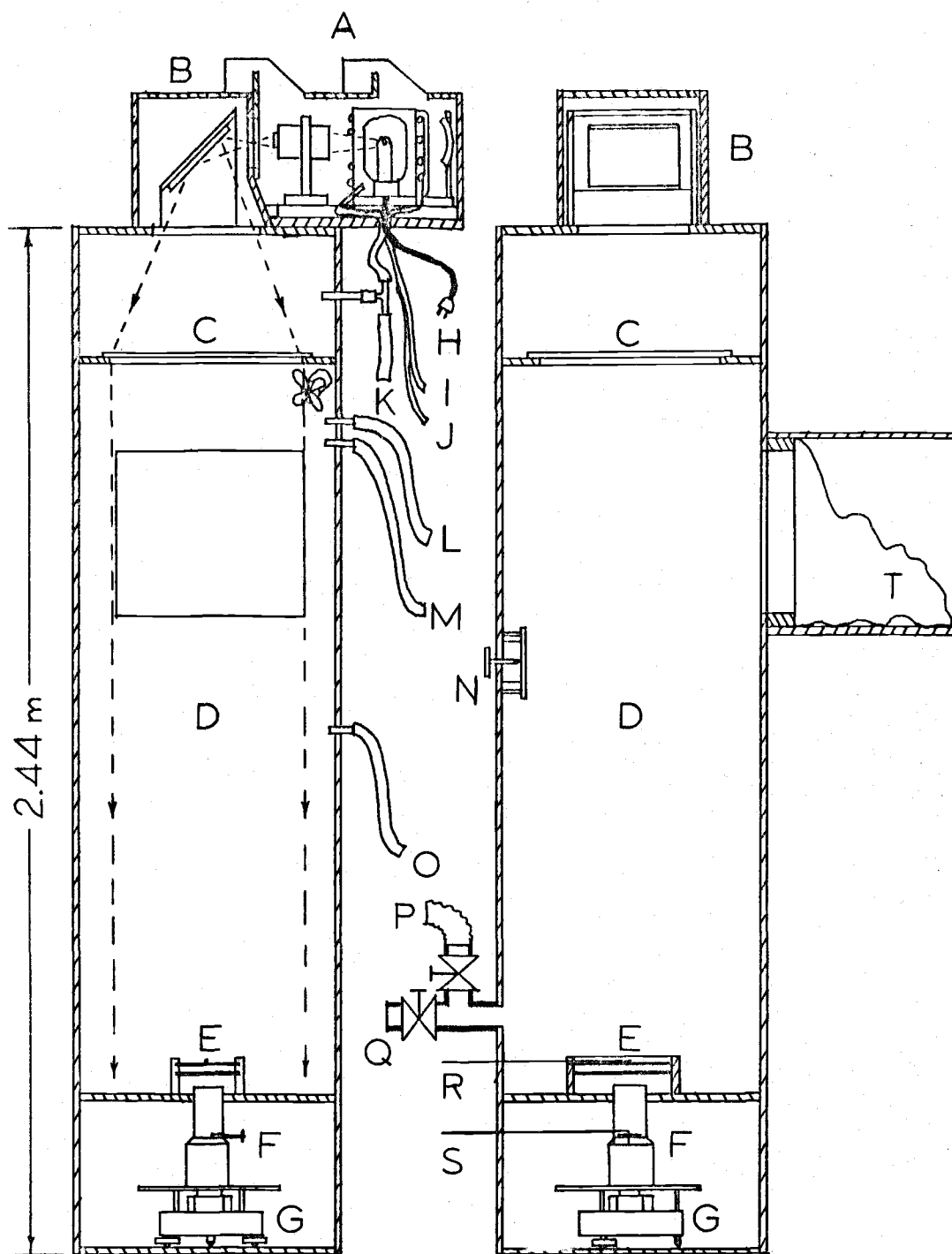
Figure 11. Aerosol chamber (Legend on following page).

Legend for Figure 11

A   Light source
B   Front surface plane mirror
C   Fresnel lens
D   Test chamber
E   I-R filter and dust cover
F   Optical system
G   Pyranometer
H   Light source power
I   Cooling water, in
J   Cooling water, out
K   Cooling air, in
L   Purge air, in
M   Aerosol, in
N   Thermometer and hygrometer
O   Sinclair-Phoenix sample, out
P   Main exhaust
Q   Purge exhaust
R   Dust cover control rod
S   Iris diaphragm control rod
T   Collapsable expansion chamber

and a 3/8" plywood screen behind the hygrometer on the inside.

Purge air and aerosol charging inlets, and sampling outlets were 3/8" tygon tubing connected to short lengths of copper tubing that passed through the chamber wall through Swage-Lok fittings.

Air was exhausted from the chamber during the purging process through standard 1/2" pipe fittings and valve located at the bottom of the chamber. During the clearing-out procedure between runs of different aerosols, a Pullman industrial vacuum cleaner was used to exhaust the chamber through standard 1" pipe fittings and valve also located at the bottom of the chamber.

Light Source. The light source was a 1000 watt incandescent projection bulb with a square array of tungsten coil filaments one centimeter on a side. In order to minimize intensity fluctuations due to line voltage fluctuations, current to the bulb was supplied by a 1000 volt-amp Sola harmonic neutralized constant voltage transformer. In order to dissipate the heat generated by the bulb, a continuous flow of air was maintained around the bulb and the bulb was enclosed laterally by a sheet brass, water-cooled jacket. The system is shown in Figure 12. A two-inch diameter front surface spherical mirror was positioned behind the bulb so that the filament fell in the focal
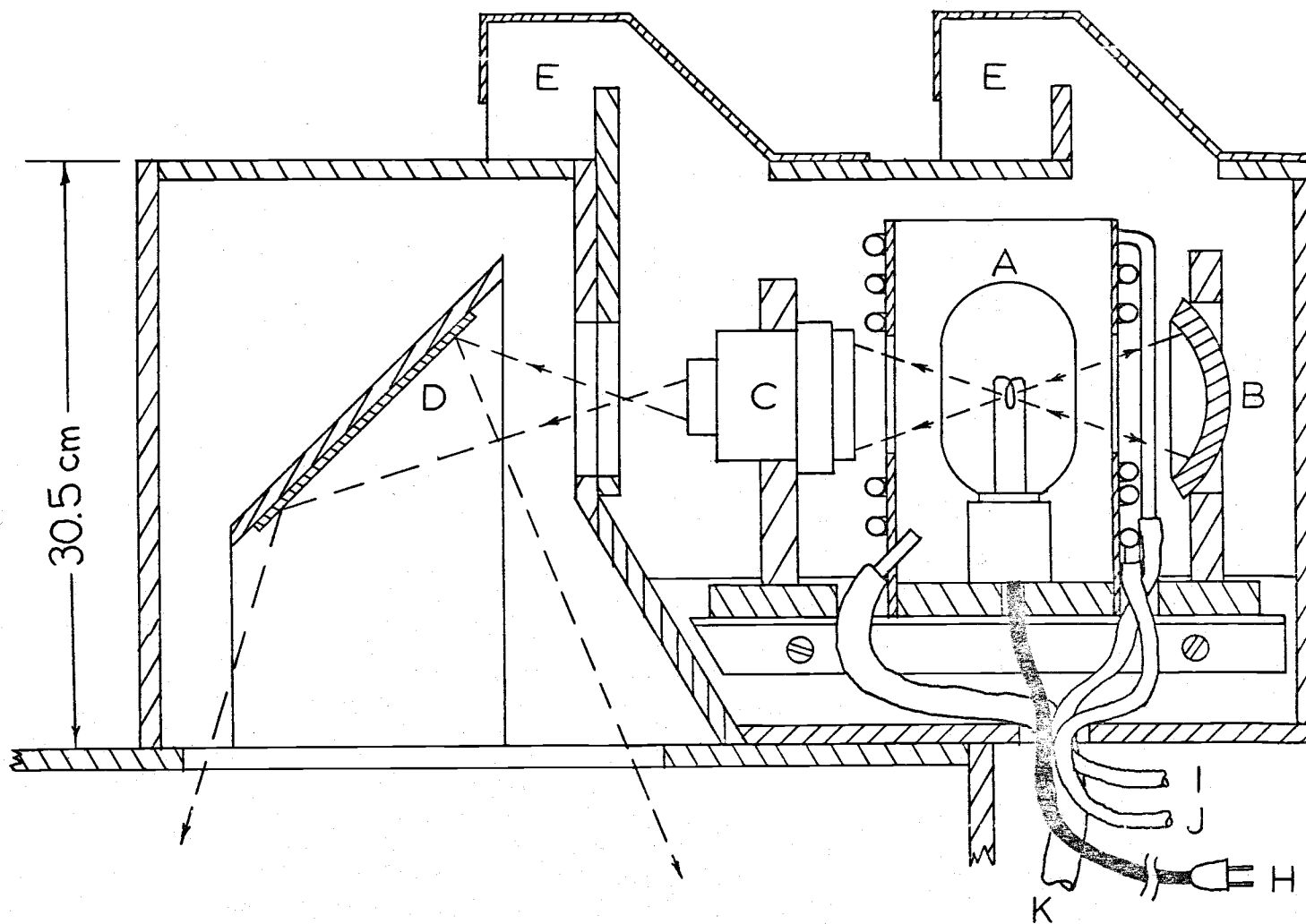
Figure 12.   Light source (Legend on following page).

Legend for Figure 12

A   Light bulb
B   Spherical mirror
C   Condensing lens
D   Plane mirror
E   Exhaust louvers
H   Power
I   Cooling water, in
J   Cooling water, out
K   Cooling air, in

plane of the mirror. This effectively doubled the intensity of the light in the forward direction, which passed through a hole in the cooling jacket, and then through a condensing lens system that produced an areal reduction of approximately 45% in the filament image. The horizontal beam diverging from the image was reflected downward in a vertical direction by a 5"x7" plane front surface mirror oriented at a 45° angle. An 18-1/4" diameter collimating Fresnel lens located so that the filament image was at its focal point produced a parallel beam of light directed along the vertical axis of the aerosol chamber. The alignment of the beam with the axis of the aerosol chamber was accomplished by adjusting the position and angle of the reflecting mirror so that the spot of light created by the beam on the aerosol chamber floor was centered on the R measuring system located at the center of the floor. The collimation of the beam was adjusted by moving the condensing lens system and the light source along the horizontal track until a twelve-inch steel scale held in the vertical beam perpendicular to its axis at the upper part of the chamber cast the sharpest and most accurately sized shadow on the chamber floor. This was determined by placing a similar scale on the floor and making adjustments until the shadow of the upper scale coincided with the scale on the floor.

The entire light source system was enclosed in a 3/8"
plywood cover to prevent any extraneous light from enter-
ing the aerosol chamber. All of the interior surfaces of
the light source enclosure were also painted flat black to
prevent reflections.

R' Measurement System. The R' measurement system
(Figure 13) consisted of three components: (1) the infra-
red filter and dust cover assembly, (2) the optical system
and (3) the sensing device.

The filter-dust cover assembly was essentially a
holder for an Oriel 2"x2" infra-red cutoff filter with a
sliding dust cover that could be operated from outside of
the aerosol chamber by means of a 1/32" diameter steel rod.
The cover was kept closed between measurements to prevent
undue deposition of the aerosol on the filter. The rod
passed through the chamber wall through a rubber gasket
and a sliding-fit copper tube held in place by a Swage-Lok
fitting. The rod was lubricated with silicon vacuum grease.
The infra-red cutoff filter was used to bring the incan-
descent spectrum of the projection bulb into closer conform-
ity with the solar spectrum.

The optical system was essentially the lens-iris
diaphragm system described in the preceding section, with
the addition of an aspheric condensing lens system follow-
ing the iris diaphragm to condense the diverging rays into
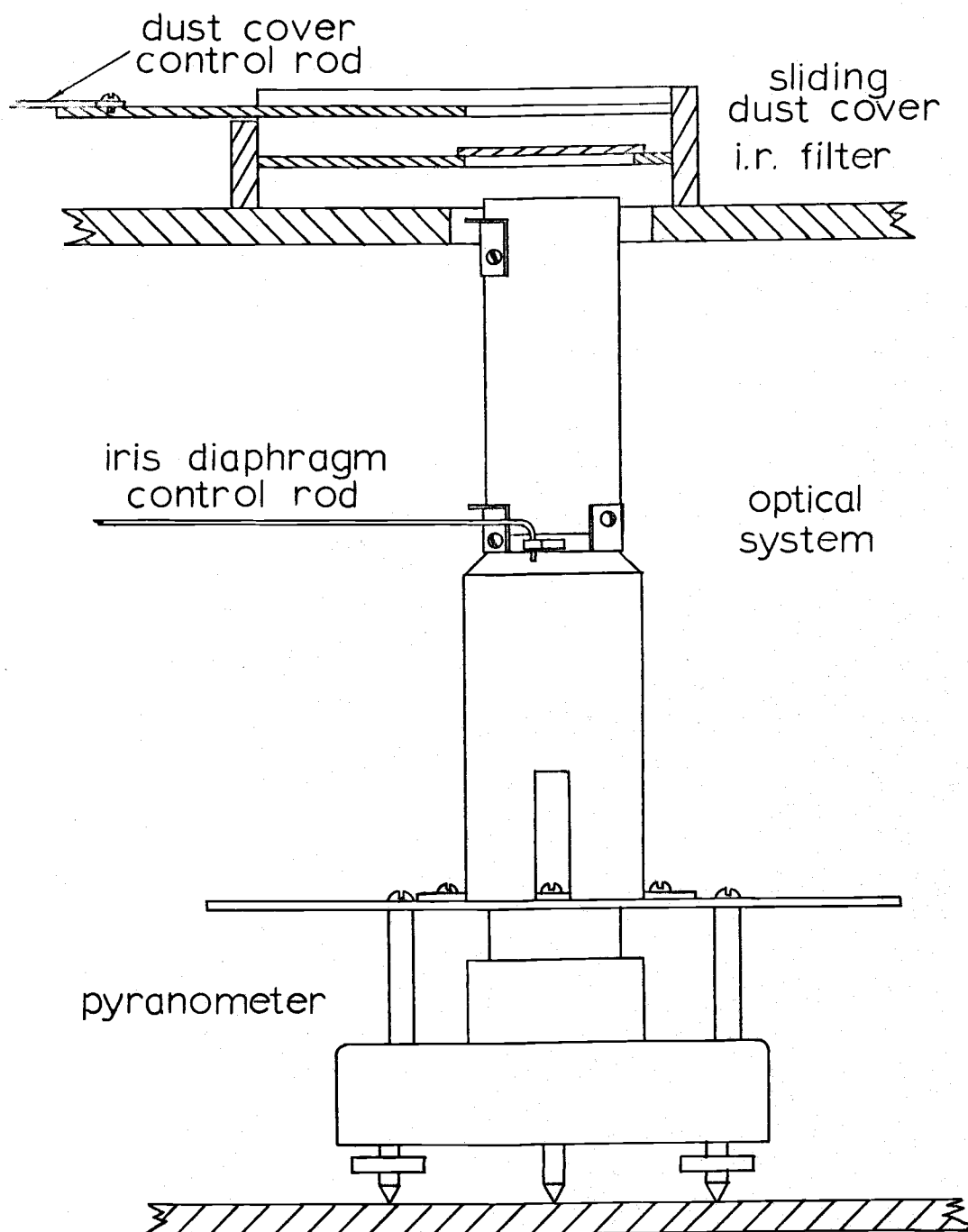
Figure 13. R' measurement system.

a beam with a small enough cross section to be completely detected by the sensing element. The objective lens was actually a compound lens made up of two 50 mm diameter binocular objective lenses with a combined focal length of 100 mm. The optical system was mounted in a two-piece, machined aluminum barrel as shown in Figure 14, with all of the inside surfaces painted flat black to minimize internal reflections. Allen head set screws were used to hold the retainer rings in place and to hold the two sections of the barrel together. The brass alignment guides were similar to those found on the Volz sun photometer, and provided a quick means of aligning the optical axis of the lens-diaphragm system with the axis of the incoming light beam, i.e., so that the parallel light rays did indeed focus within the pinhole. The guides themselves were two sheet brass plates mounted at right angles to the axis on the exterior of the front barrel section. The front plate had a 1/16" hole drilled through it while the back plate had a slightly smaller spot inked onto it. The hole and the spot were positioned so that when the optical system was aligned with the light beam, the light passing through the hole created a spot of light centered on the ink spot. The alignment of the hole and spot was initially set on an optical bench with a collimated light source. This was accomplished by placing the forward barrel section on the optical bench, aimed at the light source. A circular piece of translucent
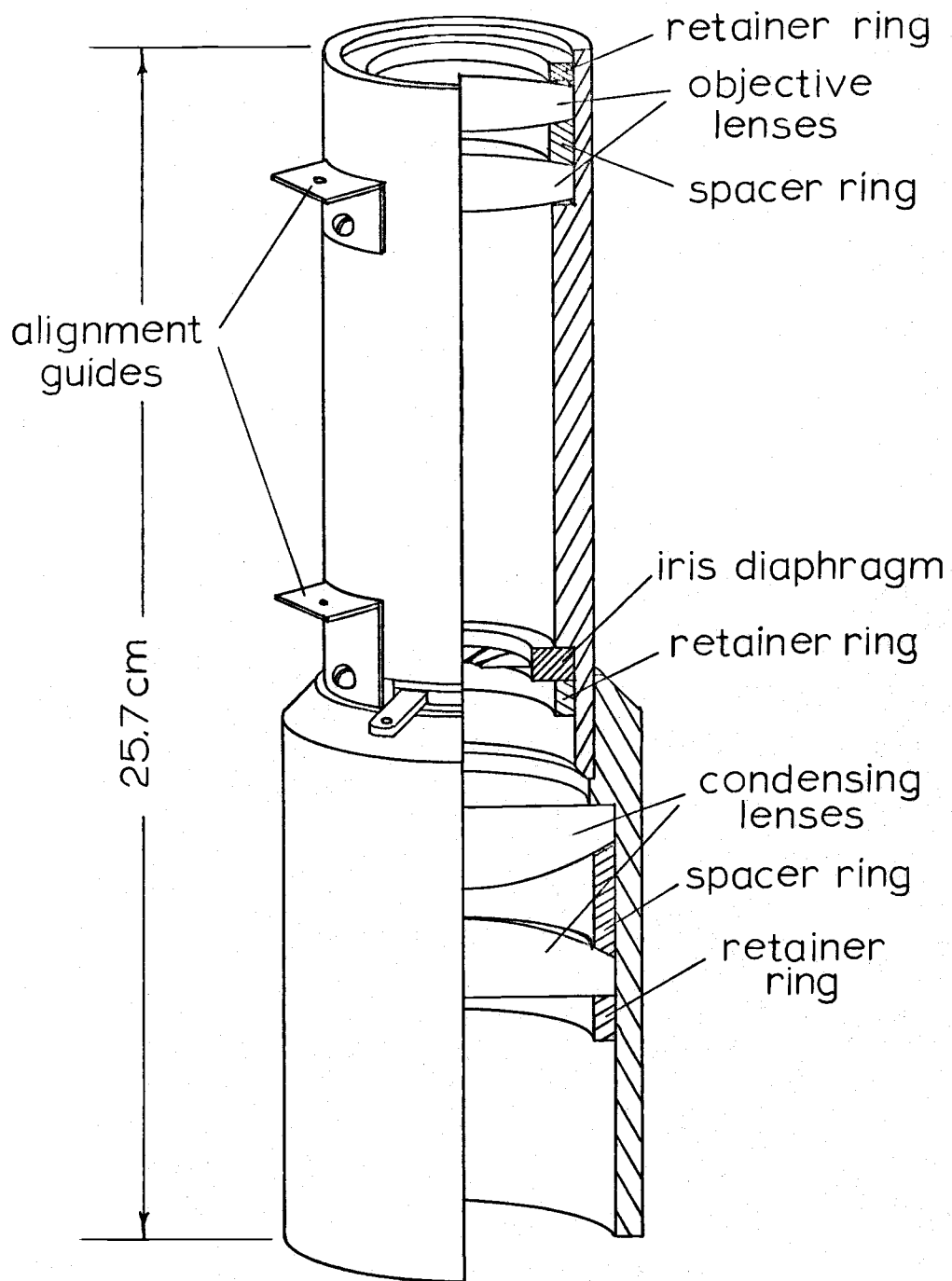
Figure 14. Optical system.

paper was placed against the diaphragm leaves to act as a screen on which the point of focused light could be seen from the back. When this point of light fell exactly in the center of the pinhole, the ink spot was marked onto the back brass plate in the center of the spot of light.

The back plate also acted as a positive stop for the diaphragm lever corresponding to a 5° acceptance angle for incoming light. This setting was also made on the optical bench with a collimated light source. The barrel was mounted this time, however, on the rotating table of a surveying transit. With the paper screen in place on the diaphragm leaves, the optical system was aligned with the light beam again by centering the point of focused light in the pinhole. The diaphragm aperture was then opened up and the transit table rotated through a horizontal angle of 5° as indicated on the vernier scale of the transit. The diaphragm was then closed down until the leaves just touched the point of light which was now off center by a distance x = 100 mm tan 5° according to equation 3. As a check on symmetry, the table was rotated also 5° in the opposite direction from the original position, and when the point of light just touched the diaphragm leaves on the opposite side of the aperture, then the brass stop was secured against the diaphragm lever in this position.

The diaphragm lever was operated during the experiment by means of a 1/32" steel rod, with one end hooked through a hole drilled at the end of the lever, passing through the aerosol chamber wall through a rubber gasket and a sliding fit copper tube held in place by a Swage-Lok fitting.

The light sensing device in this experiment was a Kipp-Zonen pyranometer, a standard solar radiation measuring instrument used in meteorology (Hewson, 1968). This instrument consists of a fourteen junction manganin-constantan thermopile that has the "hot" junctions imbedded in flat plate which is painted black and exposed to the solar radiation. The "cold" junctions are located on the underside of the instrument and are encased in a massive brass enclosure to prevent exposure to sudden variations in temperature due to winds and variations due to the effects of the solar radiation. The "hot" junctions are sealed from the ambient air by two concentric hemispherical glass domes. The incident solar radiation is absorbed by the black surface and creates a temperature difference between the "hot" and "cold" junctions, resulting in a potential difference at the two leads proportional to the light flux.

The optical system was seated on the pyranometer on a specially constructed platform of 1/8" steel that was attached to the pyranometer base. It was centered and held in place by three adjustable brackets. The platform was

constructed of 1/8" steel to provide a rigid base for the optical system that would not allow it to rock while the diaphragm lever was being operated.

The alignment of the optical system with the light beam inside of the aerosol chamber was accomplished by means of the leveling screws that make up two points of the three point base on the pyranometer. With the colli- mated light beam turned on, the system was roughly aligned by using the external brass alignment guides, and then more precisely, by obtaining the maximum reading from the pyranometer while the diaphragm was in the pinhole position.

Aerosol Generator and Air Supply. The aerosol gener- ation system consisted of a DeVilbiss nebulizer, and an air filtration system. The nebulizer (Figure 15) is es- sentially a liquid atomizer that transforms a liquid into a fine mist. The nebulizer can be used as a generator of a solid aerosol by atomizing a solution of the desired solid dissolved in water. When the mist enters a dry atmosphere, the water evaporates leaving an aerosol of solid particles. The index of refraction of a test aerosol gener- ated by this method is determined by the choice of the solute used in the nebulizer solution.

The mean particle diameter for the solid aerosol is determined by the concentration of the solute in the nebu- lizer solution. Studies have shown (Lauterbach, et al.,

mist
out

nebulizer
(glass)

rubber
stopper

12.7 cm

aerosol
solution

air in →
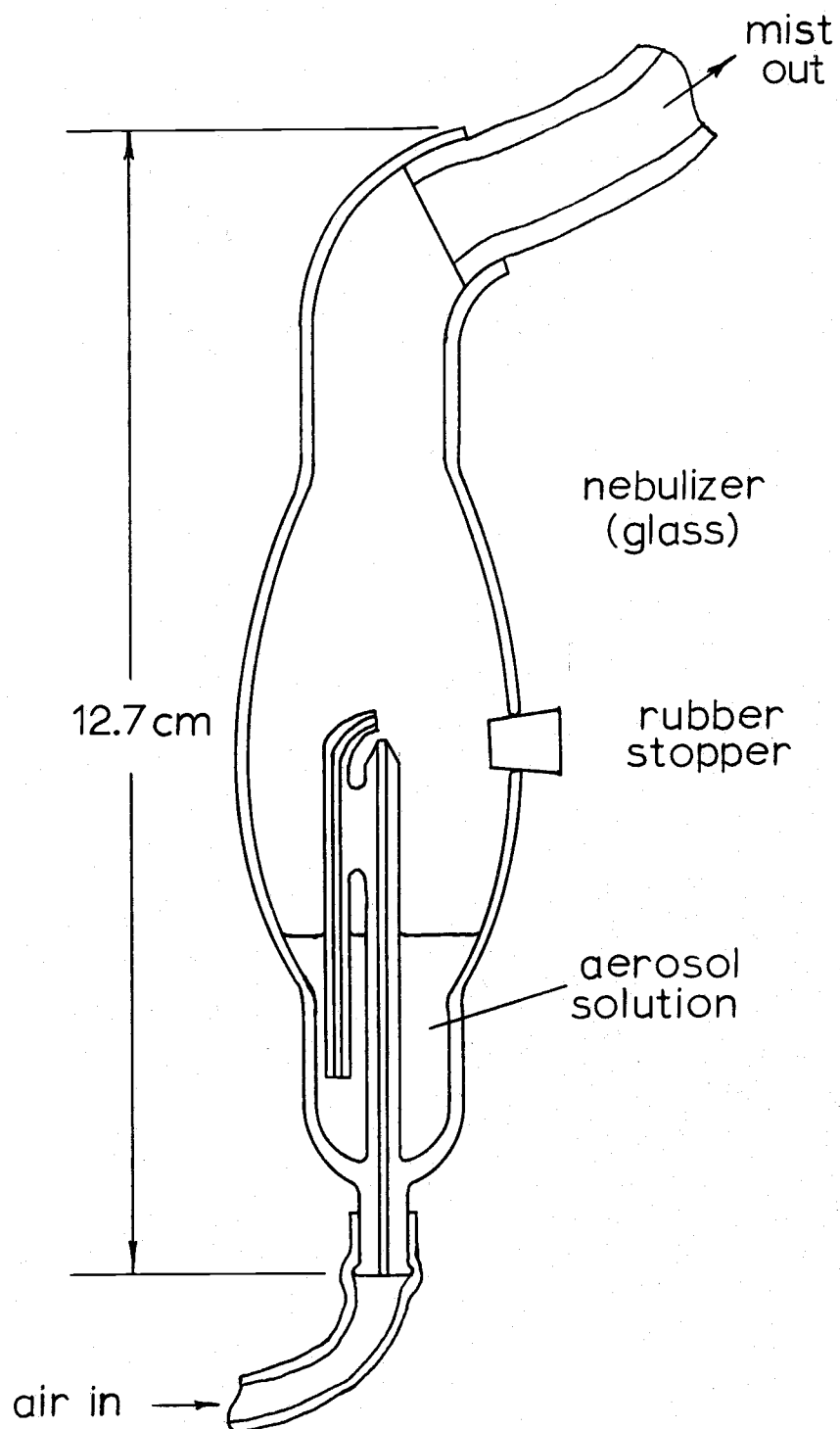
Figure 15.   Aerosol generator.

1956 and Mercer, et al., 1963) that the mean particle dia-
meter is a direct function of the solution concentration,
and moreover, it has been shown by Mercer, et al., that the
other two parameters, the nozzle size and the air pressure
or flowrate, while having a significant effect on the total
output of the atomizer, have only a minor effect on the
aerosol size distribution. In this investigation, these
minor effects were eliminated maintaining a constant air
pressure of 18 psi through a fixed nozzle, and the mean
aerosol diameter was varied by varying the concentration
of the nebulizer solution.

The air supply for the nebulizer was first filtered to
prevent any aerosols present in the air from contaminating
the test aerosol. The filtration system is shown in Figure
16. Compressed air from the laboratory wall outlet first
passed through a liquid trap and then a pressure regulator.
The air then passed through a two-inch i.d.packed-tube
filter containing fourteen inches of glass fiber wool fol-
lowed by fourteen inches of calcium sulfate granules. The
calcium sulfate is a desiccant used to absorb water vapor
from the air stream. The desiccant was followed by three
more inches of glass fiber wool and a wire screen to pre-
vent particles of the desiccant from being entrained in
the air stream. This packed-tube filter was followed by a
similar filter but smaller in size, made of glass tubing
and filled with an indicating calcium sulfate desiccant

clean, dry air

glass fiber filter

indicating desiccant

glass wool

glass wool   desiccant   glass wool   air from pressure regulator and liquid trap

79 cm
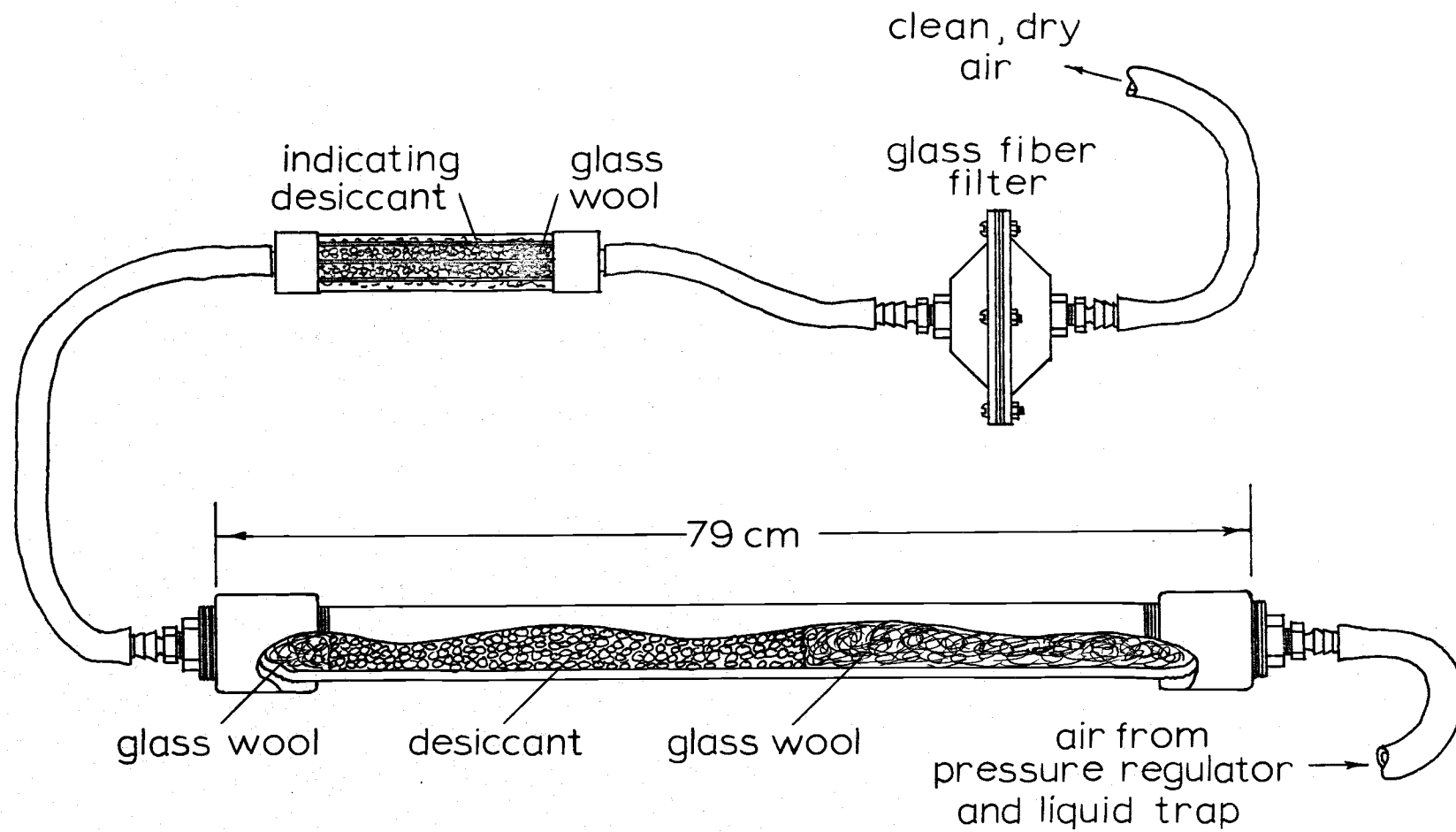
Figure 16.   Air filtration system.

that gradually changed color as it becomes saturated. This
served as an indicator showing the effectiveness of the
main body of desiccant which loses efficiency as it absorbs
water. The final filter was a four and one-half inch
diameter glass fiber filter (Gelman type A) that has a
collection efficiency of over 99% for particles as small as
0.3 microns in diameter (Rossano, 1969).

The purpose of the desiccant was to provide dry air for
the aerosol chamber. The filtration system described above
conditioned air not only for the aerosol generator but also
for the purging of the aerosol chamber prior to the experi-
mental runs and thus the filtration and the desiccation
were both included in the same system. Dry air was neces-
sary in the aerosol chamber for two reasons. First, it was
necessary to eliminate deliquescence in the test aerosols
whereby the aerosol particles increase in mass and size, and
undergo a change in their index of refraction due to their
absorption of water. The commonly used value of relative
humidity below which deliquescence may be neglected is 70%
(Bullrich, 1964; Charlson, 1968; California Ambient Air
Quality Standards, 1973), this assumption is supported by
the investigation of Lundgren and Cooper (1969). Relative
humidities between 35% and 40% were achieved with the
system described above. The second reason for the des-
iccation of the chamber air was to provide the dry atmos-
phere necessary to evaporate the water from the mist

the Sinclair-Phoenix photometer. The entire sample was passed through a membrane filter (Gelman Metricel GA-4) which was placed in the sample flow line in a filter holder provided for this purpose. The filter was weighed before and after sampling to obtain the mass of the particulate contained in the sample. From this mass and the sampling flowrate and time, the mass concentration in the sample was calculated. The photometer scale reading corresponding to this mass concentration was converted to a value proportional to the mass concentration by means of the calibration curve supplied with the instrument. Since the proportional scale is linear with respect to the mass concentration, and now the mass concentration corresponding to two points on the scale was known (the one just found and zero mass concentration at zero scale reading), the calibration of the proportional scale was complete and any subsequent photometer readings made on the same aerosol could be converted to mass concentration. The same procedure was followed for each of the remaining eight aerosols.

In order to determine the average photometer reading from the instantaneous variations, the output was fed into a strip chart recorder.

The light sensing instrumentation consisted of a Tektronix Type 1A7A High Gain Differential Amplifier with

droplets from the nebulizer in the formation of the solid
aerosol particles. This evaporation had the effect of in-
creasing the relative humidity of the air in the aerosol
chamber to a maximum value of approximately 45% over one
experimental run.

Instrumentation. The instrumentation for this experi-
ment fell into two groups: (1) that used to measure the
aerosol mass concentration and (2) that used to amplify and
measure the signal from the light sensing system.

A Sinclair-Phoenix aerosol photometer was used to
measure the aerosol mass concentration inside the aerosol
chamber. The Sinclair-Phoenix aerosol photometer, a light
scattering instrument, is described in principle in
section II, but since light scattering is dependent on the
physical characteristics of the aerosol, such as size,
shape, and index of refraction, it was necessary to cali-
brate the instrument for each specific aerosol. The in-
strumental scale readings, however, are not proportional
to the mass loading and a non-linear calibration curve is
supplied with the instrument that relates the instrument
scale to one that is proportional to the mass loading.
This second scale is calibrated by the user for specific
aerosols. The calibration was accomplished in this experi-
ment by charging the aerosol chamber with one of the nine
aerosols and sampling this atmosphere for ten minutes with

a Tektronix Type 133 plug-in power supply, and a strip
chart recorder. The optical system and the light sensor is
described in the preceding section. The signal of about
0.15 millivots from the pyranometer was amplified to regis-
ter approximately three-quarter scale on the 1 volt full
scale strip chart recorder, i.e., about a 5K amplification.

V. DATA ANALYSIS AND RESULTS

R and R' Measurements

The measured light flux signals from the pyranometer
were amplified and recorded on strip charts. Due to ampli-
fier drift and line voltage variations, three measurements
of the ratio R were made for each value of m. A typical
trace is shown in Figure 17. The first plateau represents
light flux measured with the optical system diaphragm in
the pinhole position. The second plateau represents the
flux measured with the diaphragm in the open or 5° accep-
tance angle position. The third and fourth plateaus
represent the same measurements as in the first and second
plateaus except they were made with the amplifier in the
DC offset mode. In this mode, the amplifier output is off-
set in such a manner that the baseline on the strip chart
corresponds to an input voltage higher than zero, i.e., the
strip chart pen is activated only when the input voltage
reaches a predetermined value. Thus, since only the upper
part of a trace is recorded, the amplification can be in-
creased and still have the trace fit on the chart. The
third and fourth plateaus therefore are repeat measure-
ments of the first and second plateaus but showing only
the upper portions and amplified by a factor of five.
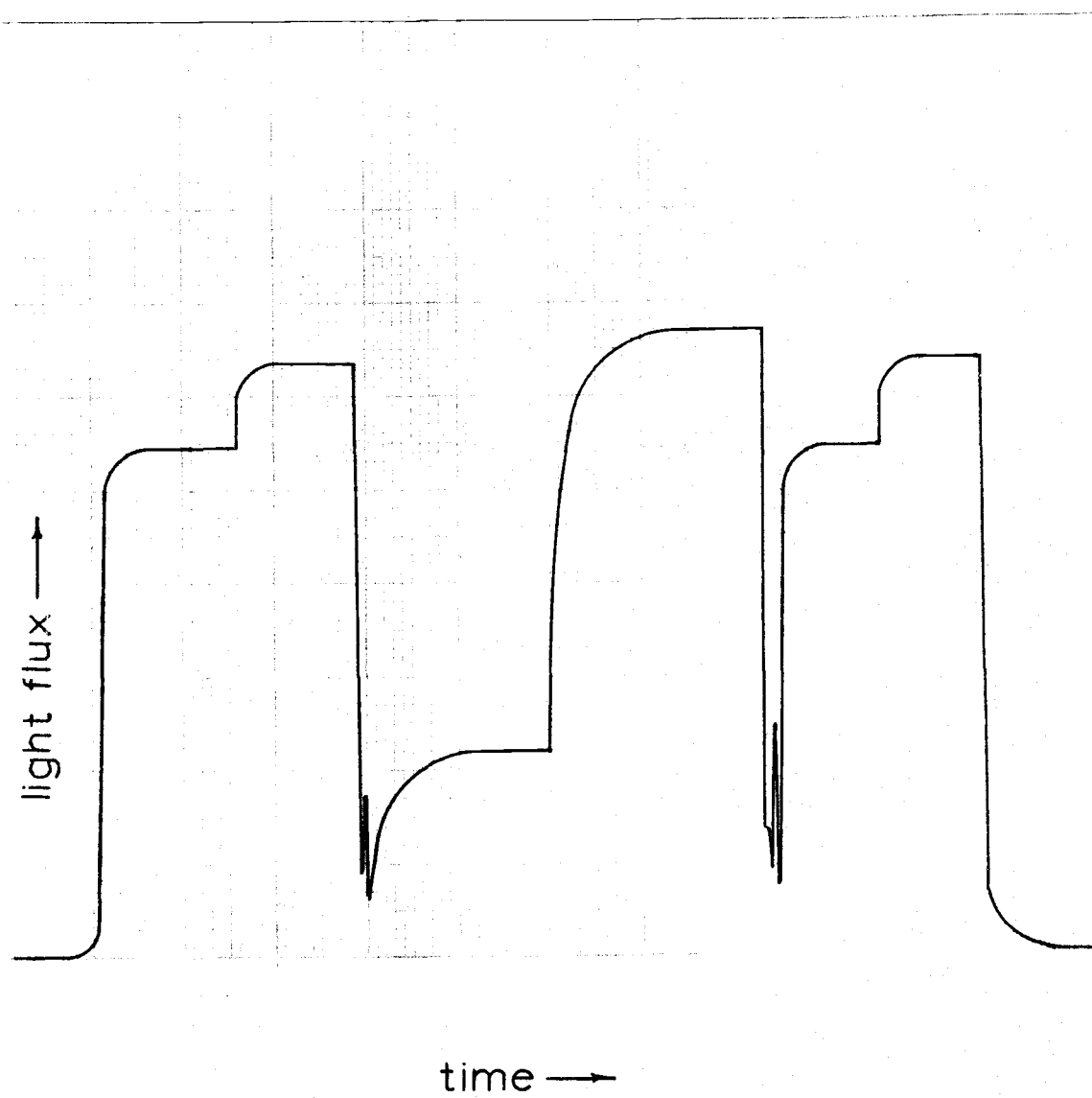
Figure 17. Strip chart trace of pyranometer output.

The fifth and sixth plateaus are a repeat of the first and second full trace measurements. When agreement was reached between the higher resolution offset trace and at least one of the two full traces on the difference between the plateaus (agreement within 0.03 units when full scale represented 10.0 units) then the observation was considered acceptable. A total of 30 observations out of 195 were rejected using this criterion.

Since the output voltage from the pyranometer is proportional to the light flux striking the sensor, the results, expressed as the ratio R, were calculated by dividing the height or voltage of the lower plateau by that of the higher plateau. R' was calculated by substituting these values of R into equation (45). Graphs of the ratio R' as a function of m for the nine experimental runs are presented in Figures 18 through 26. The curves that are shown represent the linear least squares regression lines for the data points with m greater than 1.0 micrograms/liter. The reason for the omission of the data points below 0.001 milligrams/liter are discussed in section VI. Table V lists the constants for the linear regression equations as obtained by least squares analysis and also the correlation coefficients for each regression line and its data points. Appendix A lists all of the experimental R vs m data points along with the temperature and relative humidity. Appendix B lists all of the experimental R' vs
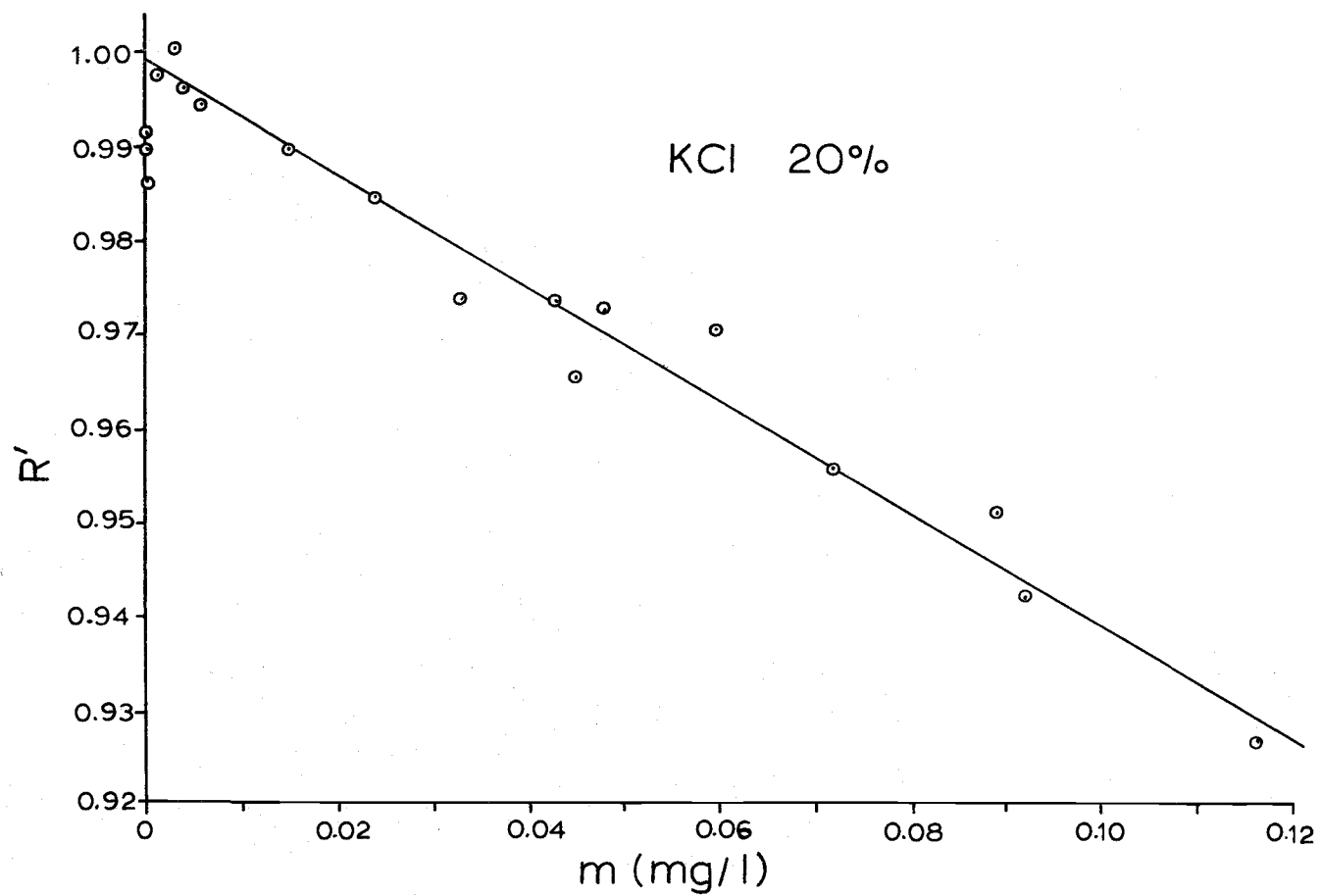
Figure 18.   Plot of experimental data for KCl 20%.
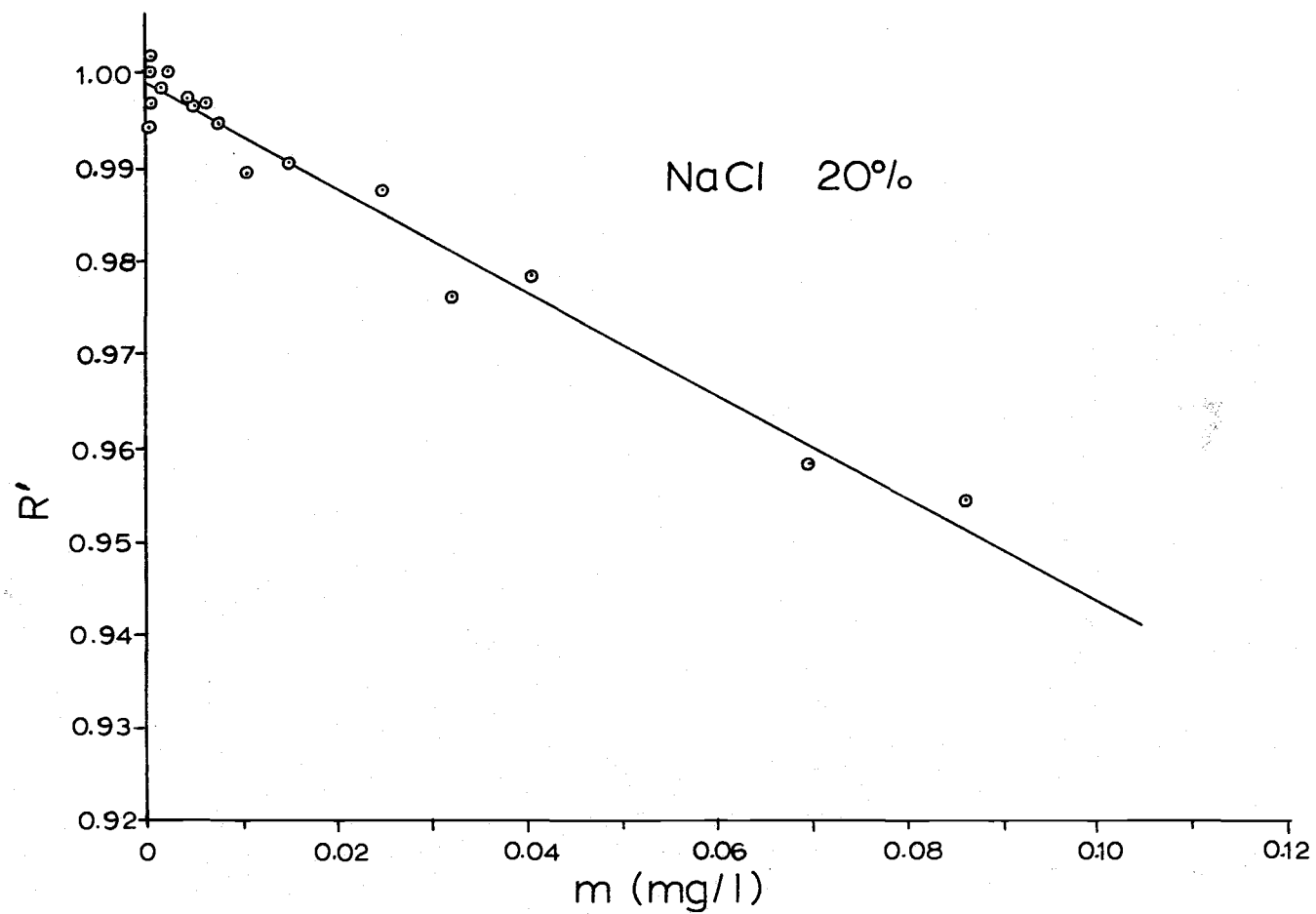
Figure 19. Plot of experimental data for NaCl 20%.
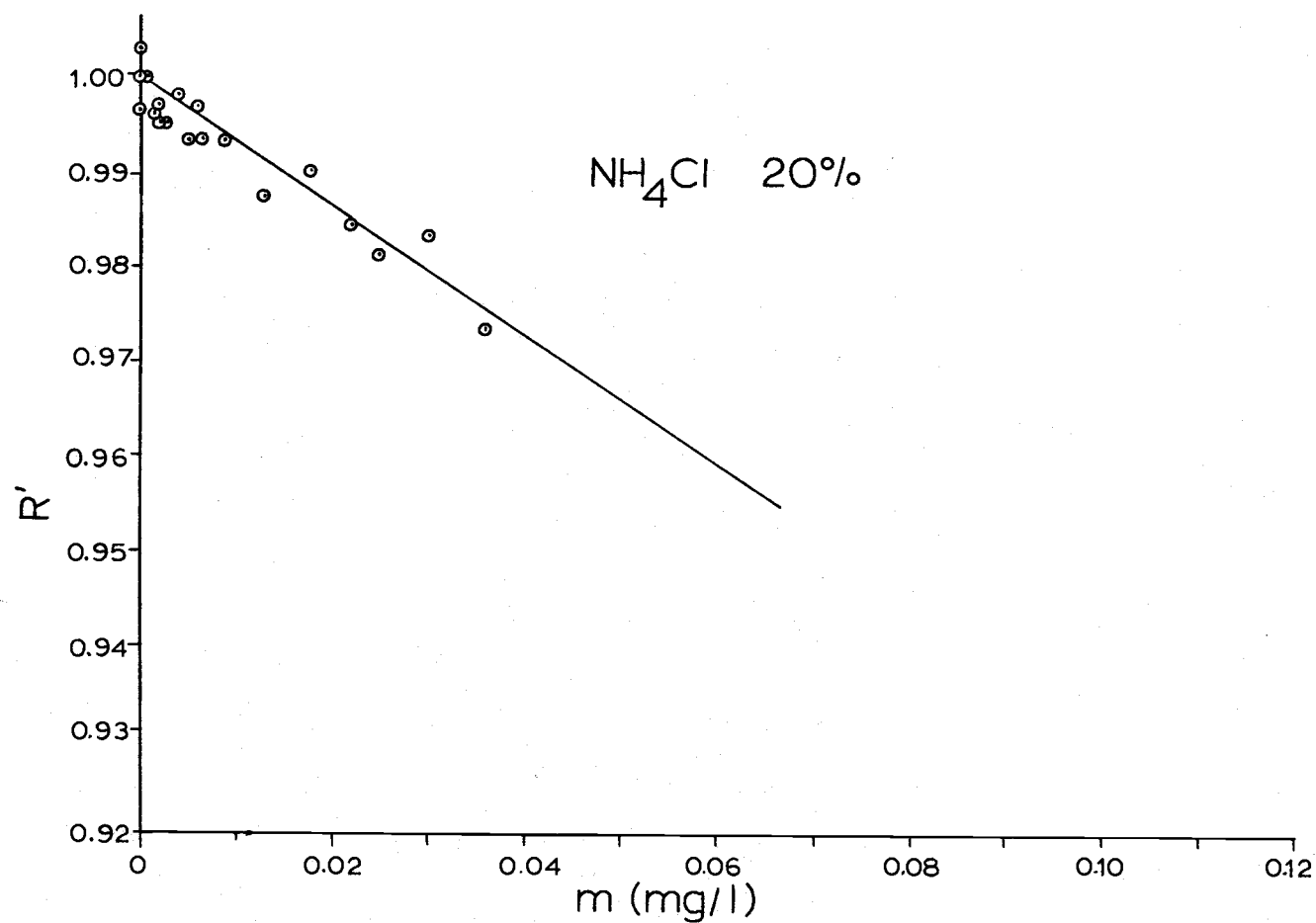
Figure 20.   Plot of experimental data for NH_4Cl 20%.
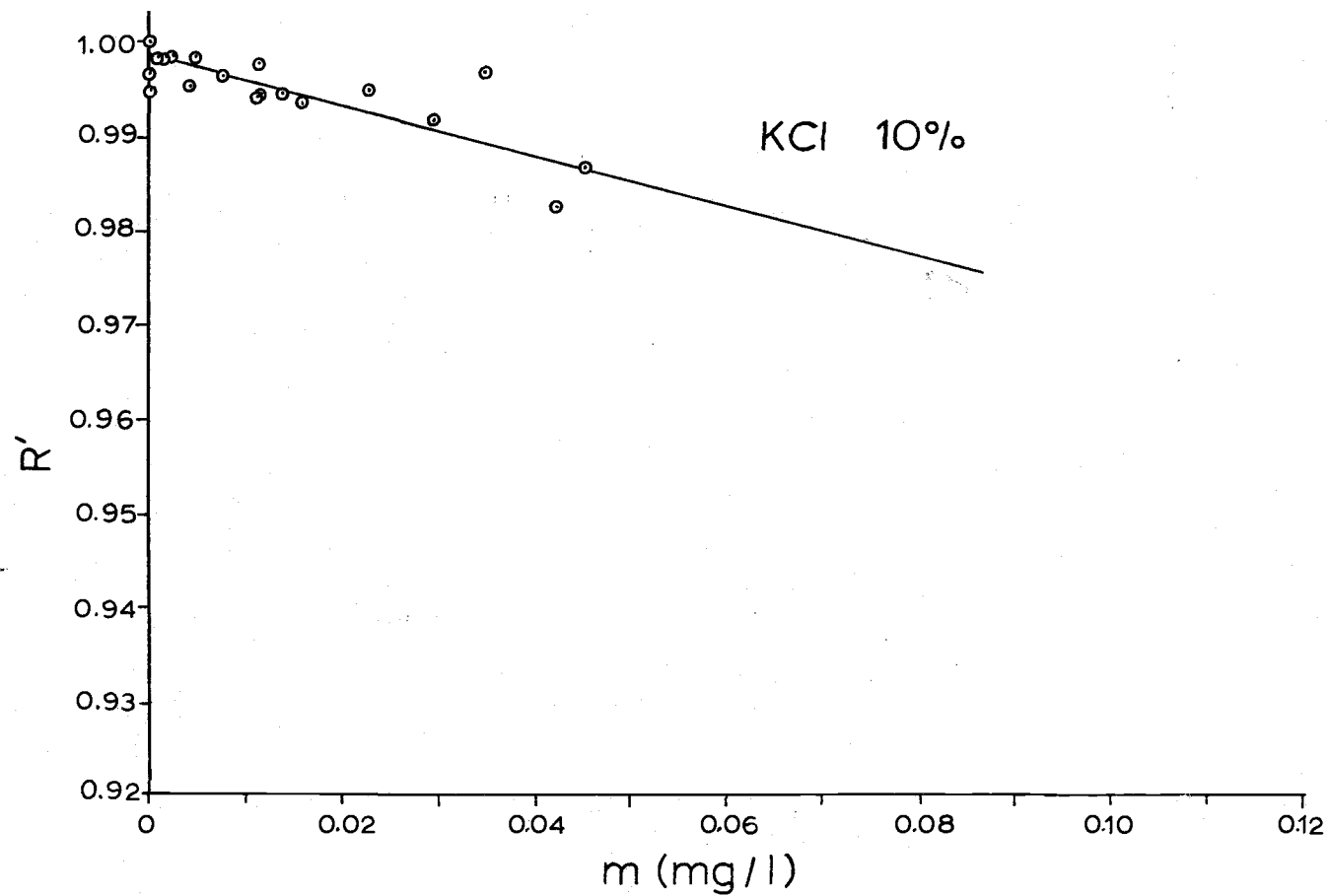
Figure 21.   Plot of experimental data for KCl 10%.
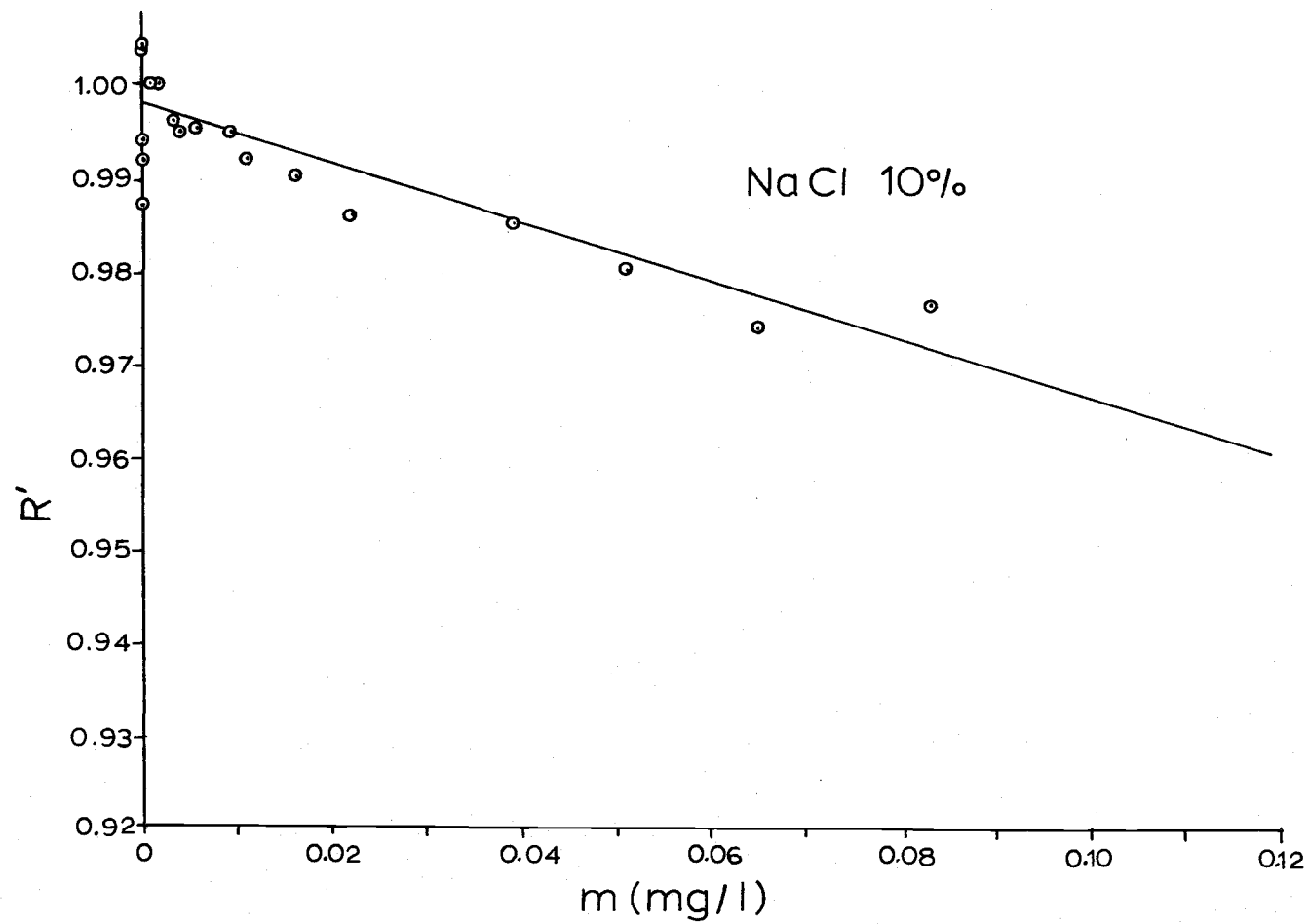
Figure 22. Plot of experimental data for NaCl 10%.
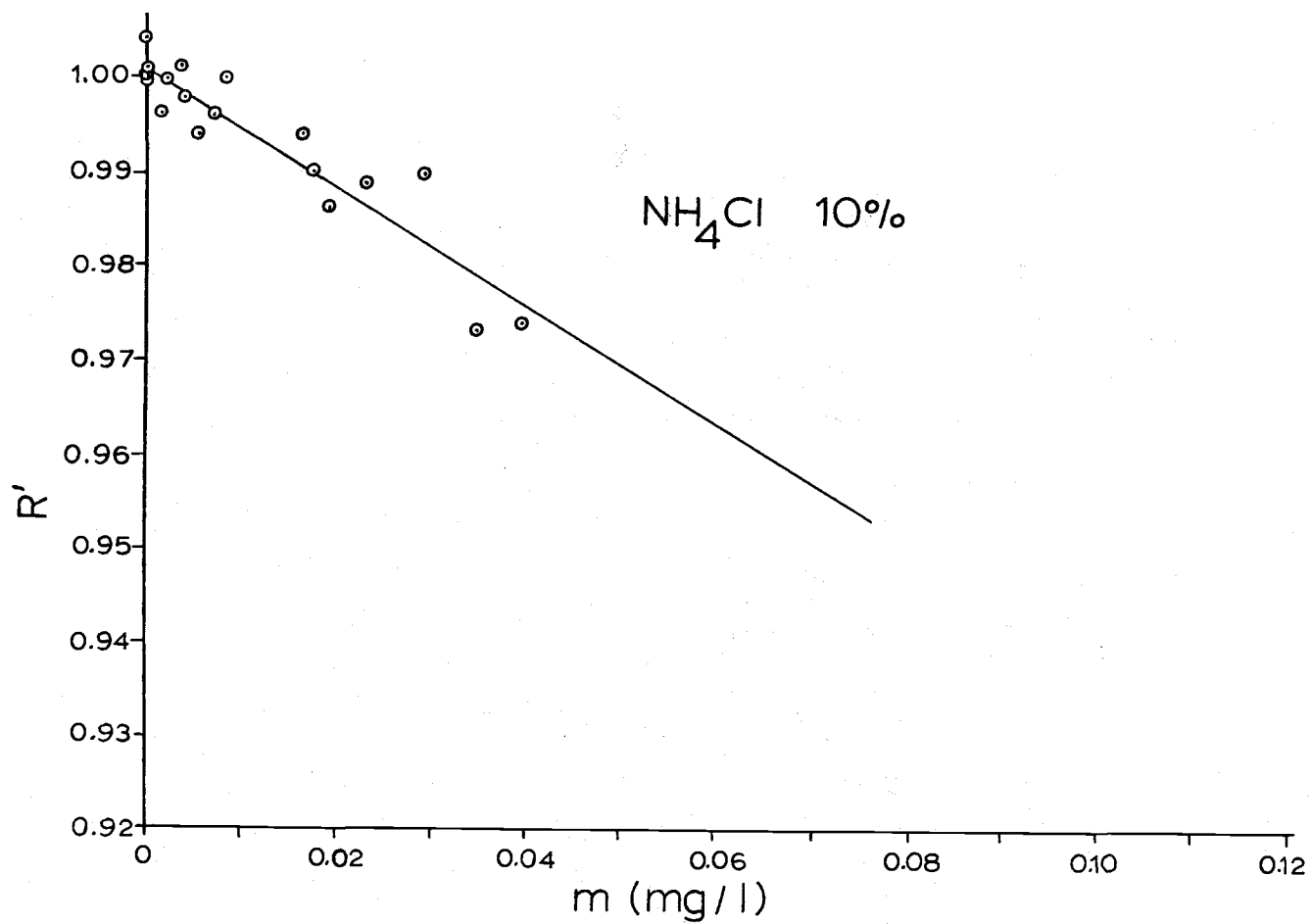
Figure 23.   Plot of experimental data for NH_4Cl 10%.
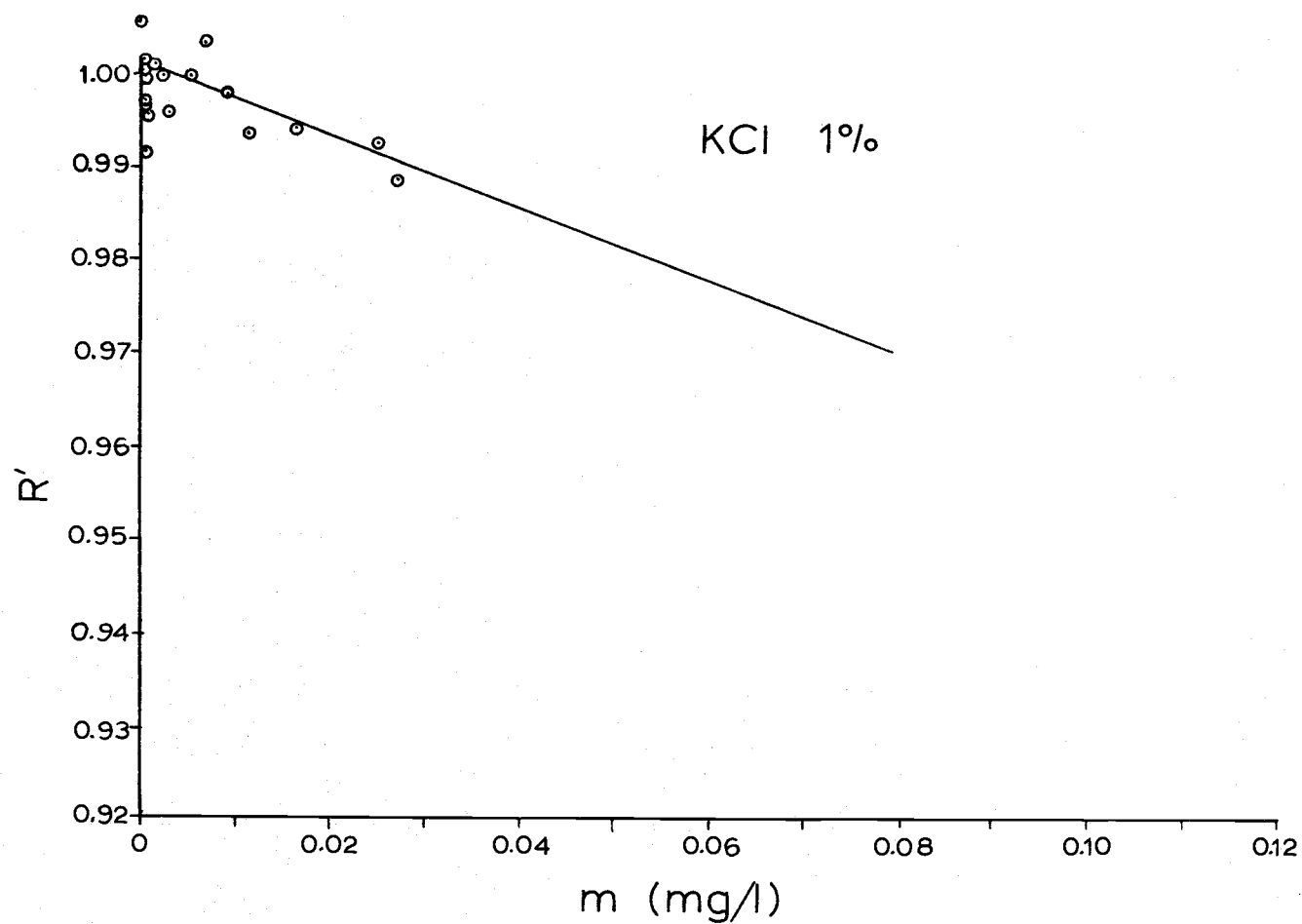
Figure 24.   Plot of experimental data for KCl 1%.
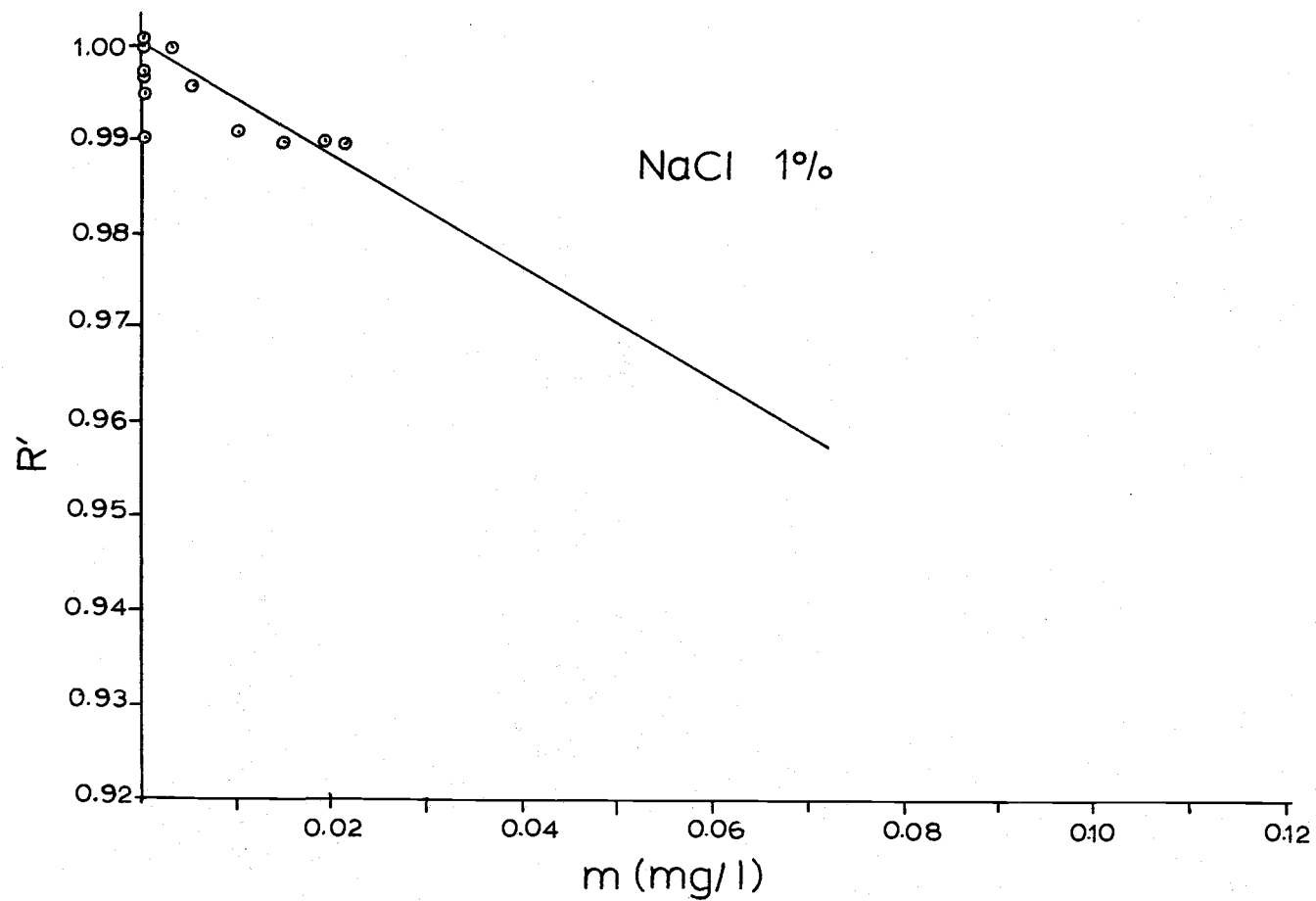
Figure 25.   Plot of experimental data for NaCl 1%.
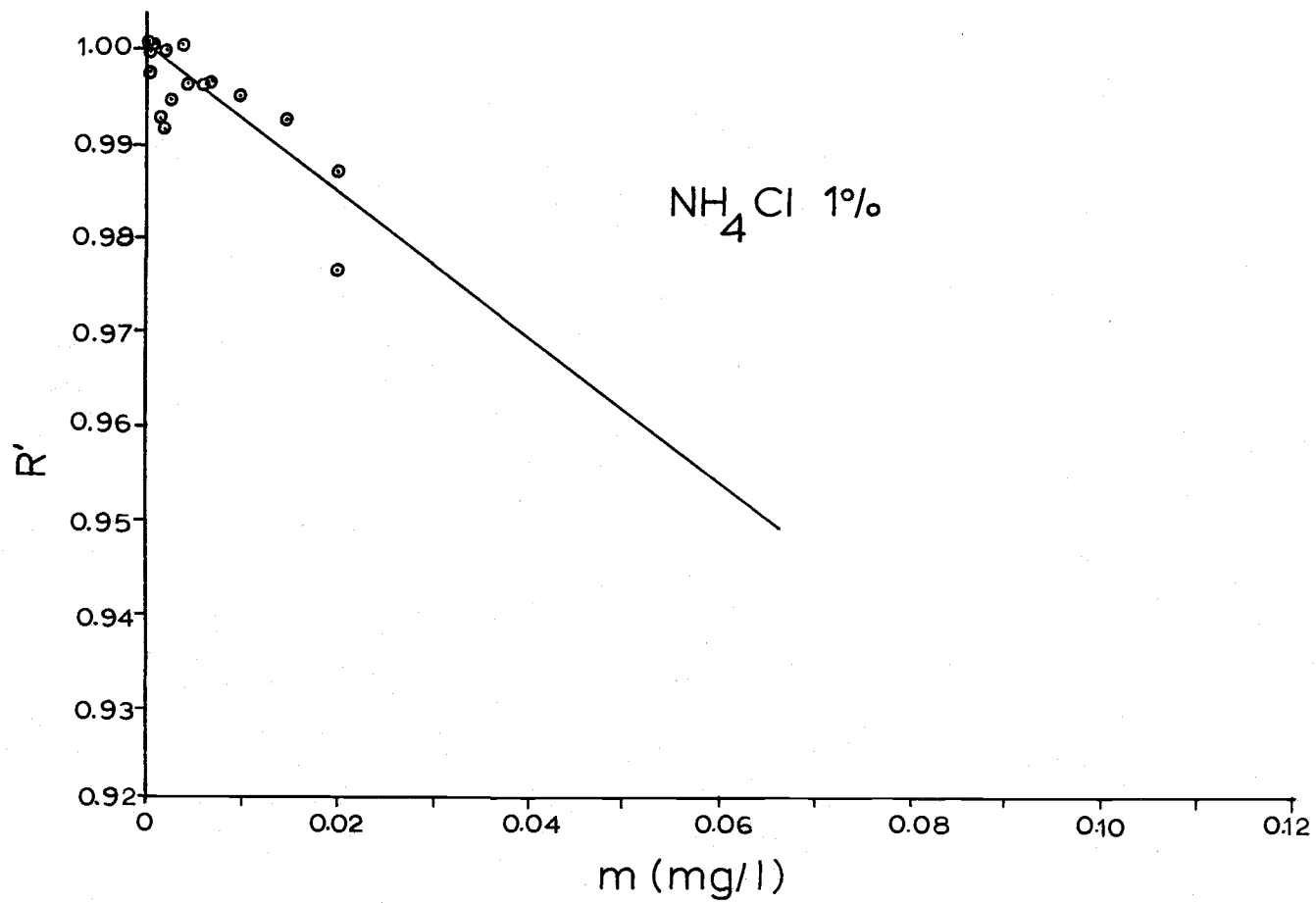
Figure 26. Plot of experimental data for NH_4Cl 1%.

TABLE V

Linear Regression Equations and Correlation Coefficients

Aerosol	Percent	(R' = A + B m) A	B (1/mg)	Correlation Coefficient
NaCl	20	0.9990	-0.5476	0.986
NH₄Cl	20	1.0002	-0.6790	0.909
KCl	20	0.9990	-0.6008	0.987
NaCl	10	0.9977	-0.3139	0.957
NH₄Cl	10	1.0016	-0.6316	0.902
KCl	10	0.9987	-0.2617	0.861
NaCl	1	1.0010	-0.5898	0.893
NH₄Cl	1	0.9998	-0.7697	0.811
KCl	1	1.0005	-0.3842	0.813

m data points and the corresponding R' values from the
least squares line and the per cent difference between the
two values of R' for each data point.

In each of these curves, the data has been corrected
so that they all have the common intercept on the ordinate
of 1.0. The fact that R(0) is not equal to 1.0 experimen-
tally is due to the fact that the incident light is not
perfectly parallel. The reasons for this deviation from
perfect collimation are: (1) the spherical and chromatic
aberrations associated with the condensing and particularly
the Fresnel lenses, and (2) the finite size of the light
source, i.e., the lamp filament, even after condensing, is
not a true point source. These effects were observed and
are illustrated in Figure 27. If the light were parallel,
the flux recorded by the pyranometer at m = 0 would be the
same for both the pinhole and the open position of the
diaphragm since all of the light incident on the objective
lens would be focused at a point within the pinhole. Thus
the ratio of the two fluxes would equal 1.0. When the in-
cident light has a non-parallel component, however, this
non-parallel component is not detected with the diaphragm
in the pinhole position. With the diaphragm in the open
position, both the parallel and the non-parallel components
are detected resulting in a ratio R (or R') that is less
than 1.0. The correction that has been applied in Figures

spherical
aberration

chromatic
aberration

red
yellow
blue

white
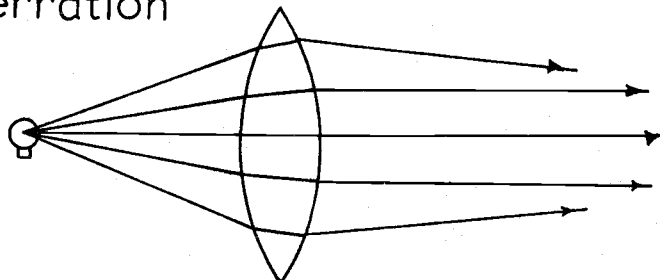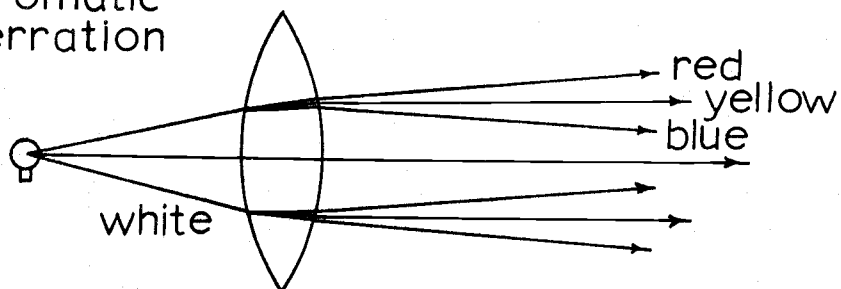
non-point
source
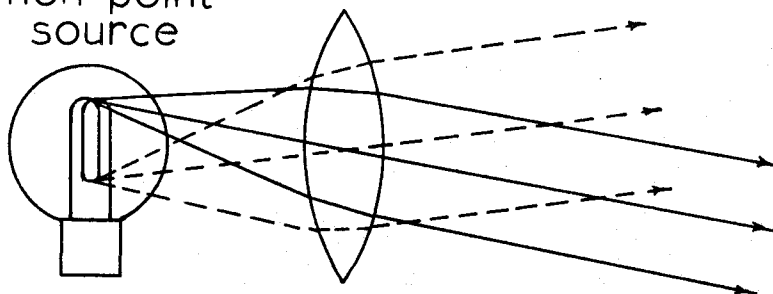
Figure 27.   Causes for imperfect collimation.

18 through 26 is a simple addition of constant additive

correction factor F, to all R' values, whose magnitude

brings R'(0) up to 1.0. Thus the relationship of all the

data points remains exactly as measured. The magnitude of

this correction, however, is not truly constant over all m

and the slope of the R' vs m curve must be multiplied by

1/R'(0). This can be seen by examining the regression

equation of the measured values of R' on m:

$$R'(m) = R'(0) - C\ m$$
$$= R'(0)\ (1 - \frac{C}{R'(0)}\ m) \tag{48}$$

where: C is the uncorrected slope.

This can be rearranged to give:

$$R'(m)/R'(0) = 1 - \frac{C}{R'(0)}\ m \tag{49}$$

which is of the same form as equation (45) if $K_2 = \frac{C}{R'(0)}$.

Both the additive correction factor [1-R'(0)] and

the slope correction factor [1/R'(0)] were obtained by

first plotting the uncorrected R' vs m curves and deter-

mining R'(0) from the regression equations. These values

of R'(0) were then used in plotting Figures 18 through 26.

The experimentally measured value of the slope for each R'

vs m curve along with its slope correction factor, its

additive correction factor, and the corrected value of the

slope are listed in Table VI.

In order to facilitate the comparison of the results of the nine experimental runs, all of the data from the nine runs are plotted together in Figure 28. The least squares straight line for the aggregate data is also indicated. Appendix C lists each experimental data point used to find this least squares line, its corresponding point (with the same value of m) on the regression line, and the percent difference between the two.

Experimental Particle Size Distributions

A sample size distribution for each of the nine aerosols used in this experiment was obtained by microscopic examination using a Porton graticule to obtain projected area diameters. Due to time limitations, a size distribution was not obtained for each of the 114 measurements of R and m. The samples were collected on Metricel GA-4 triacetate membrane filters (Gelman Instrument Company) which were made transparent on the microscope slide with dioxane. These size distributions are shown in Figures 29, 30, and 31. The tables of the data points are in Appendix E.

TABLE VI

Experimental Values of dR'/dm and Correction Factors

Aerosol	Percent	Experimental Slope (1/mg)	Slope Correction, 1/R'(0)(1/mg)	Corrected Slope (1/mg)	Additive Correction, F (1/mg)
NaCl	20	-0.5476	1.1899	-0.6516	0.1596
NH_4Cl	20	-0.6790	1.2073	-0.8198	0.1717
KCl	20	-0.6008	1.1838	-0.7113	0.1553
NaCl	10	-0.3139	1.1932	-0.3745	0.1619
NH_4Cl	10	-0.6316	1.2133	-0.7663	0.1758
KCl	10	-0.2617	1.1798	-0.3088	0.1524
NaCl	1	-0.5898	1.1891	-0.7013	0.1590
NH_4Cl	1	-0.7697	1.2034	-0.9262	0.1690
KCl	1	-0.3842	1.1905	-0.4574	0.1600
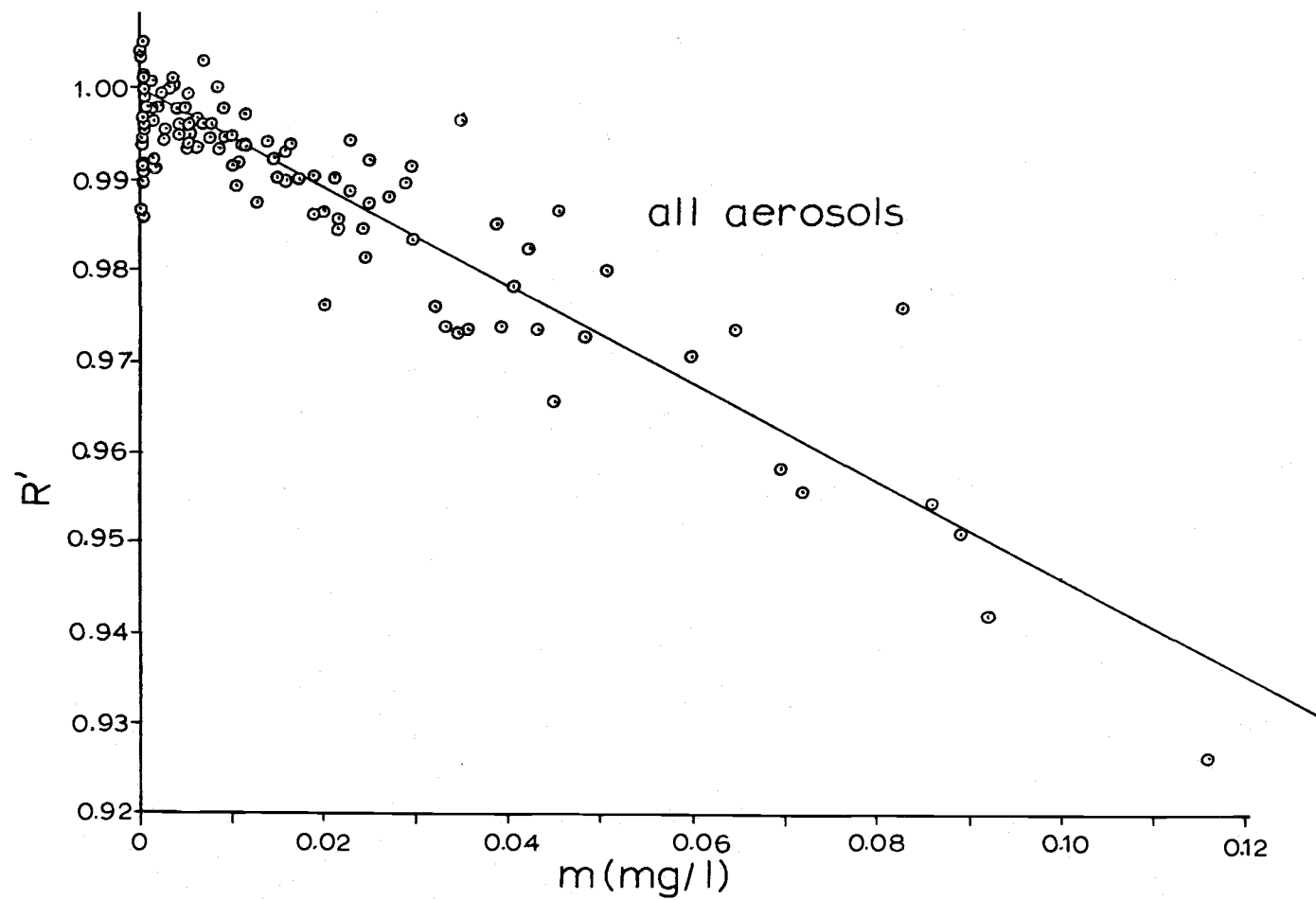
Figure 28. Plot of experimental data for all aerosols.
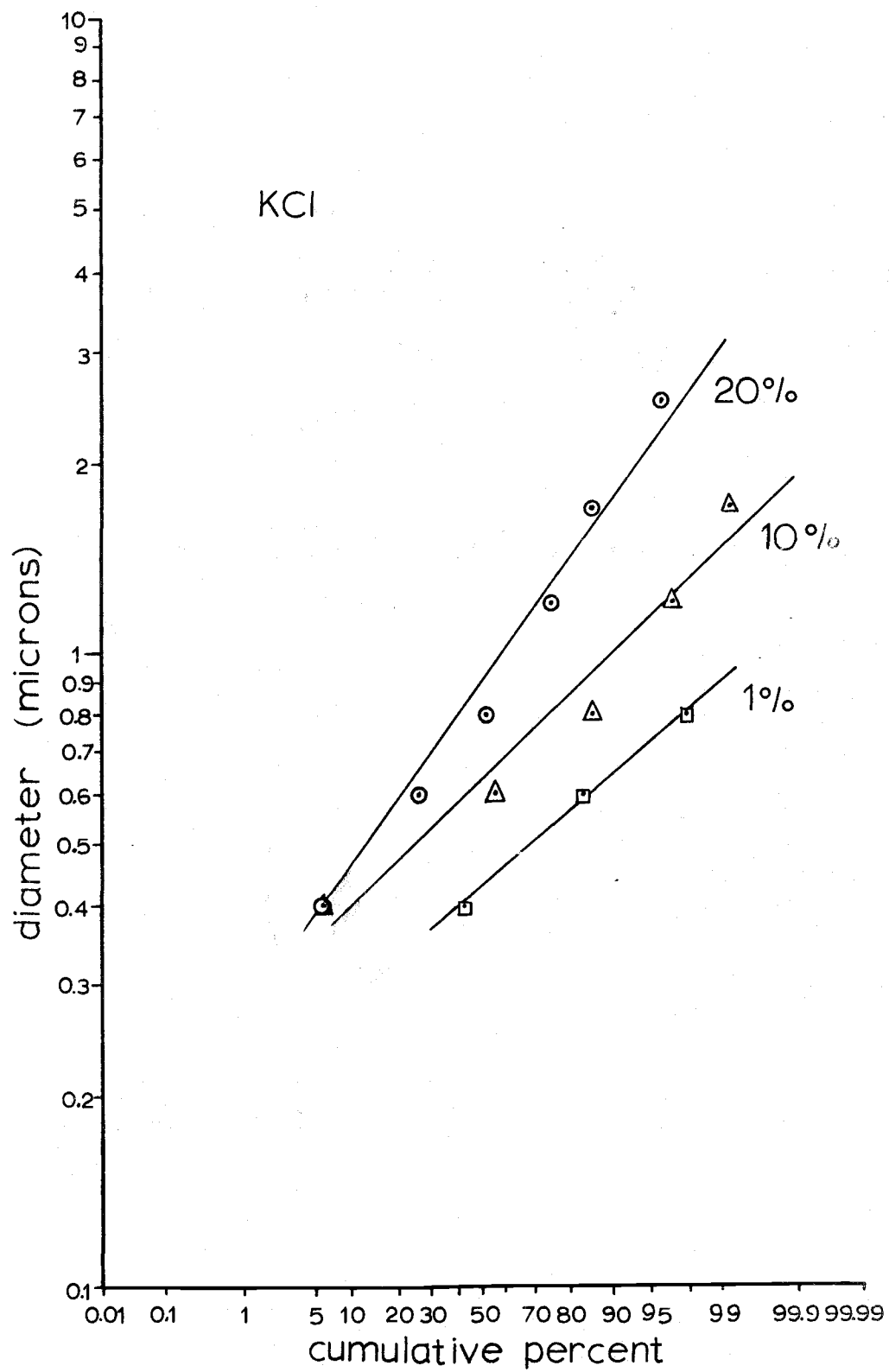
Figure 29. Sample particle size distributions for KCl.
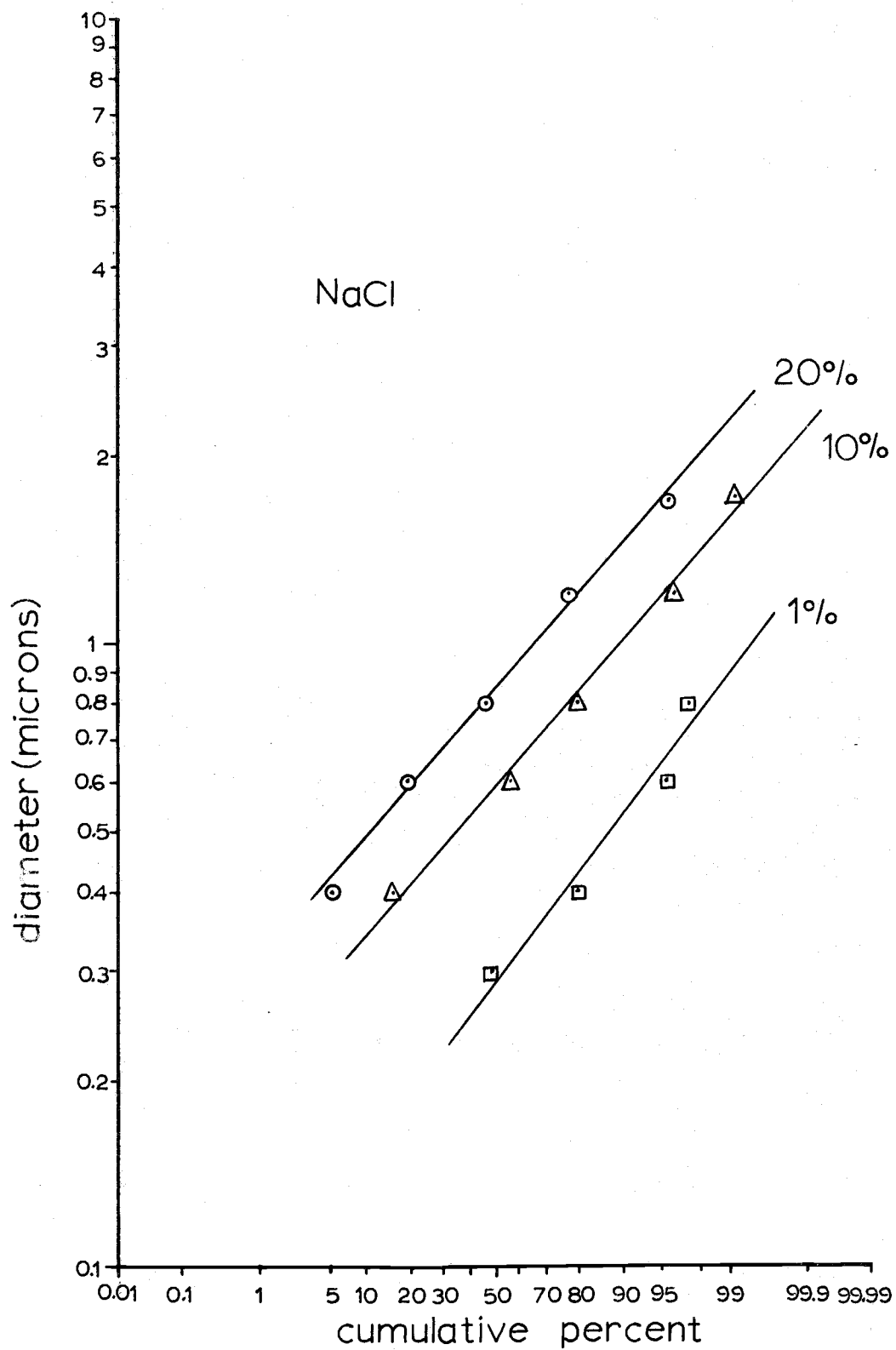
Figure 30. Sample particle size distributions for NaCl.
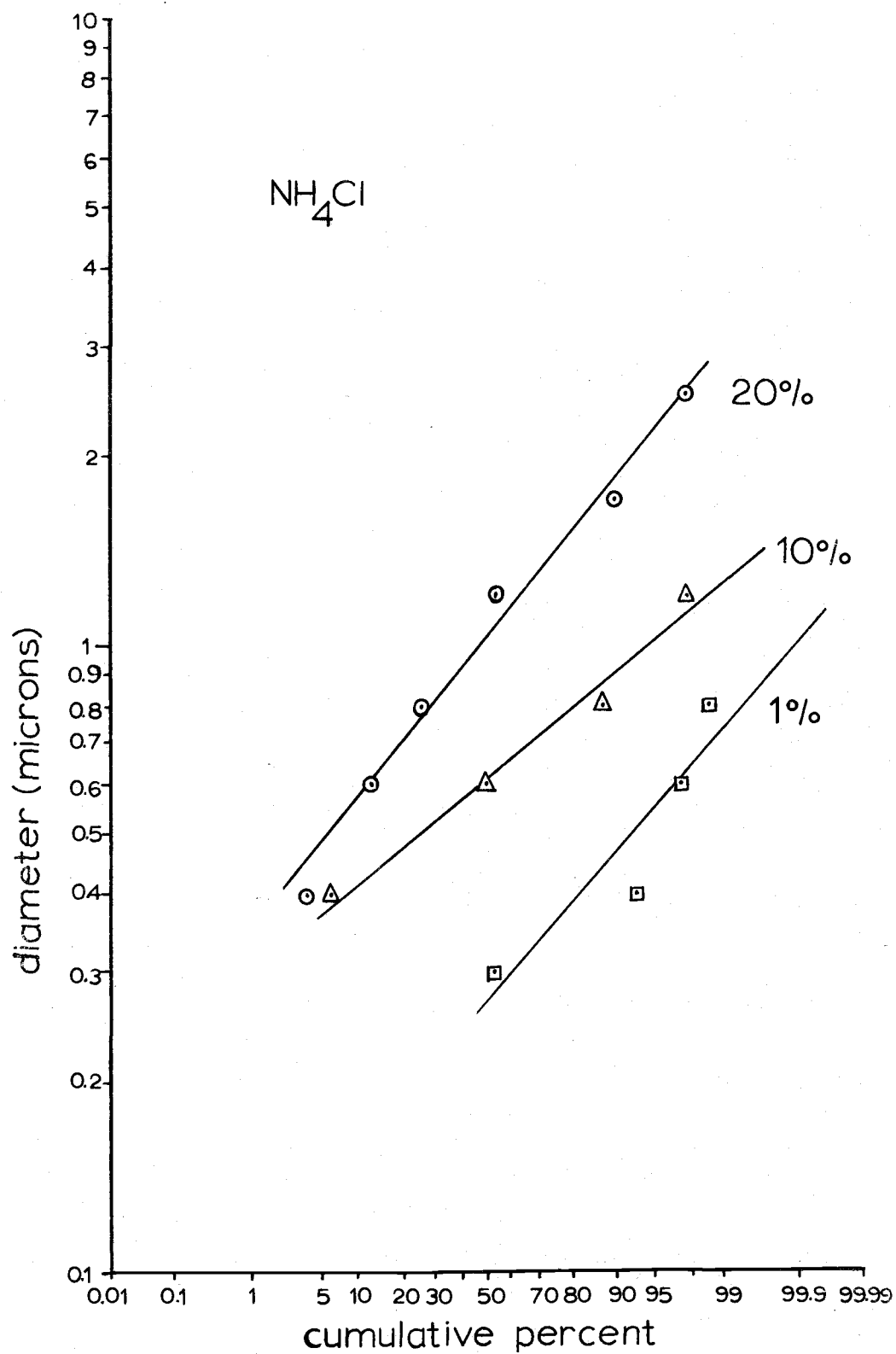
Figure 31.   Sample particle size distributions for NH$_4$Cl.

VI. DISCUSSION

Discussion of Experimental Results

The experimental data from this investigation yields two salient results. The first and most apparent is the linearity of the dependence of R' on m. The second is the dependence that the results have on the aerosol index of refraction and the mean aerosol diameter.

The linearity of the individual R' vs m curves is apparent both from the scatter diagrams and from Table VI which lists the correlation coefficients for each set of data and its least squares line. Five out of the nine correlation coefficients are greater than 0.90, and none are below 0.811. For the number of data points involved, these coefficients indicate correlation at a 99% confidence level in all but one case. For the one exception, correlation is indicated at a 95% confidence level. Appendix B, which lists each data point and its corresponding point on the least squares line, demonstrates the correlation further by indicating that only two experimental R' value out of the 114 total were more than 0.7% above or below their respective least squares line. This evidence supports the contention that a correlation between the ratio R and the aerosol mass concentration does exist, and,

moreover, indicates that the functional form of the dependence of R and R' on m fits the prediction of the mathematical model.

The dependence of the experimental results on the aerosol index of refraction and on the mean aerosol diameter shows up as small changes in the slope of the R' vs m curve. This dependence is illustrated in Figure 32, which is a plot of $|dR'/dm|$ as a function of the aerosol mean diameter for the three indices of refraction used in this experiment. It can be seen that the variations in the slope due to variations in either the index of refraction or the mean diameter are large relative to one another, but quite small on an absolute scale. For a given index of refraction, the greatest change in slope due to a change in mean diameter is 0.3825 1/mg for KCl. This means that a change of 0.01 mg/l in the aerosol concentration results in changes in R' for the two KCl aerosols that differ by 0.003825. This is approximately 10% of the total variation in R' encountered in this experiment, or about 0.3% of the actual R' values. It should be noted that a change in aerosol concentration of 0.01 mg/l is very large in terms of atmospheric concentrations. The maximum allowable concentration of lead aerosols under the California Ambient Air Quality Standards is 1.5 x 10^{-6} mg/l averaged over 30 days. The smallest change in slope
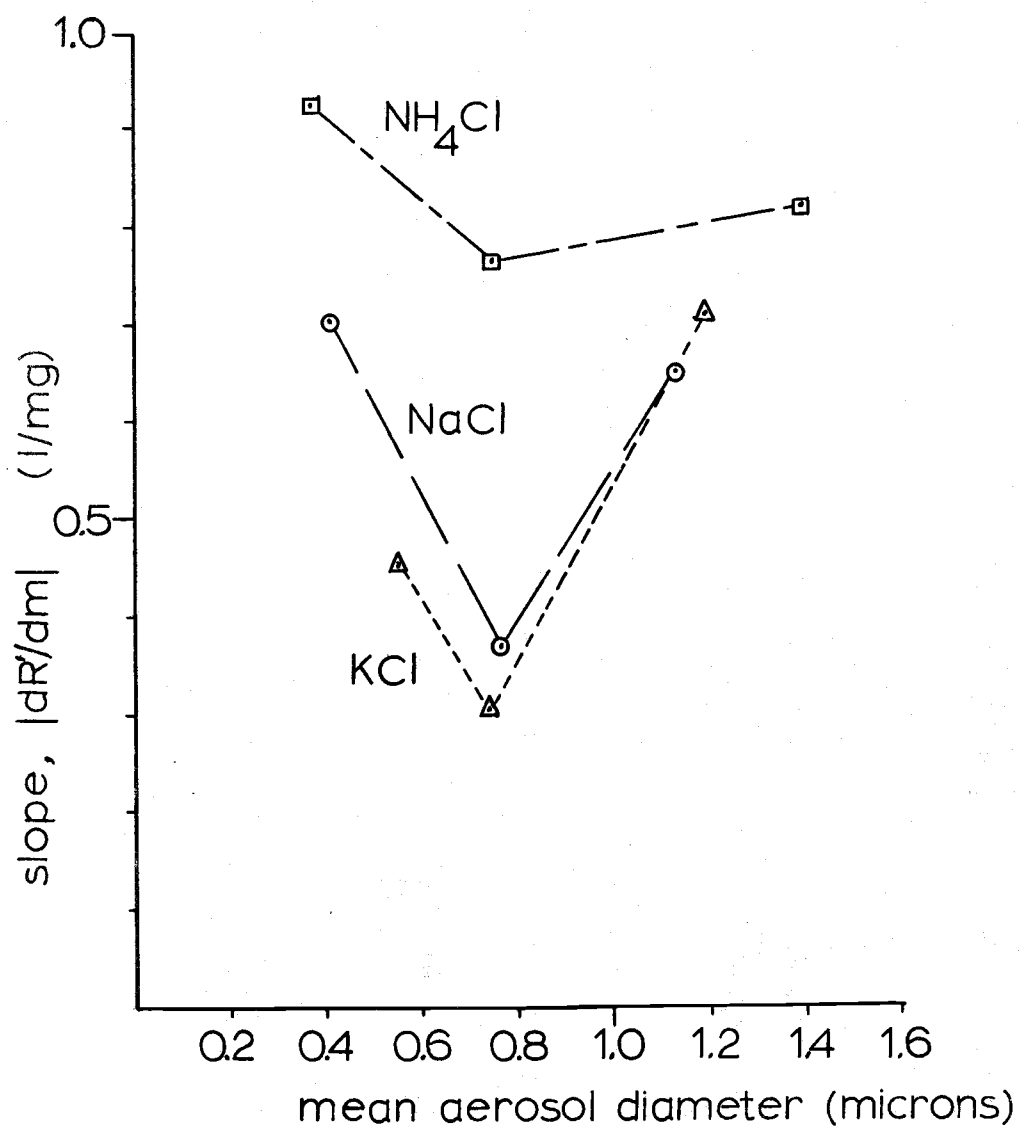
Figure 32. Experimental values of the slope of R' vs m
as a function of mean aerosol diameter.

is 0.0535 1/mg and is for NH_4Cl.

The greatest difference in slope due to a difference in index of refraction (for a given mean diameter) is approximately 0.46 1/mg at d_m = 0.75 microns.

The second result indicated by Figure 32 is that the R' vs m slope is not a monotonic function of the mean aerosol diameter. For each of the indices of refraction, the R' vs m slope decreases with increasing d_m from the 1% nebulizer concentration to the 10% concentration then increases with a further increase in d_m from its 10% nebulizer concentration to the 20% concentration.

Figure 32 also shows that the dependence of the R' vs m slope on the index of refraction is not uniform over all d_m, but appears to be roughtly proportional at least for the lower values of d_m. (The indices of refraction for KCl, NaCl, and NH_4Cl are 1.49, 1.54, and 1.64, respectively.) In the range of d_m values obtained with the 20% nebulizer concentrations, however, the order of the slopes for the three indices of refraction appear to begin changing.

The weak dependence of the results on the aerosol index of refraction and mean aerosol diameter is also illustrated in Figure 28 which is a scatter diagram of all the data points from the nine experimental runs. The linear least-squares regression line for the aggregate

data is also shown. A visual inspection shows the close grouping of the data points about the least-squares line independent of the index of refraction and the mean diameter of the aerosol. More significant is the high correlation coefficient (0.936), since it is independent of the units used. A graphical plot of the data, on the other hand, could be constructed to appear closely grouped by choosing a scale that compresses the ordinate.

Comparison of Experimental and Theoretical Results

In comparing the experimental results with the mathematical model, it can be seen that the general functional relationship between R' and m is in agreement; however, the experimental points below m = 0.001 mg/l have been omitted due to the obvious non-linearity, the theoretical and experimental slopes of the R' vs m curves are in disagreement, and a dependence of the slope on the aerosol index of refraction and mean diameter not predicted by the mathematical model appears experimentally.

The non-linearity of the R' vs m data for low values of m can be explained by considering the assumption of the Junge size distribution which was made in the mathematical model. The Junge particle size distribution is based on a steady state in the shape of the distribution which is arrived at through the mechanisms of particle diffusion,

coagulation and sedimentation, and applies to so-called "aged" aerosols, i.e., aerosols that have had sufficient time to allow the mechanisms to operate. According to Smoluchowsky's theory (Smoluchowsky, 1918), the rate of coagulation is proportional to the square of the particle concentration and thus a low concentration would require a longer time to reach the steady state distribution. Since all measurements were made after relatively short aging times (five minutes) due to total time limitations, it is expected that at the lowest concentrations, the aerosol may not have had sufficient time to reach the Junge-shaped distribution. The result is a relative excess of the smaller particles (Junge, 1958), which in this experiment are in the size range (0.3 to 1.0 microns) that scatter light most efficiently (Volz, 1954; Junge, 1955; Pueschel and Noll, 1966; Horvath, 1966). Since R' is defined in equation (44) as $1-(f_s/f_u)$, the resultant increase in f_s and decrease in f_u lead to a decrease in R'. This result is seen in the R' vs m scatter diagrams.

The disagreement between the theoretical and experimental values for the slopes of the R' vs m curves is shown in Table VII which lists both the theoretical and experimental slopes. In calculating the theoretical slopes, λ was taken as 0.55 microns and ν was taken as 3.0 following Junge (1955, 1958). The difference in the three theoretical values is a result of the difference in the

TABLE VII

Experimental and Theoretical Values of dR'/dm

Aerosol	Experience Slope (1/mg)	Theoretical Slope (1/mg)
	-0.6516 (20%)	
NaCl	-0.3745 (10%)	-89.2
	-0.7013 (1%)	
	-0.8198 (20%)	
NH_4Cl	-0.7663 (10%)	-125.6
	-0.9262 (1%)	
	-0.7113 (20%)	
KCl	-0.3088 (10%)	-97.3
	-0.4574 (1%)	

The percentages in parentheses are the nebulizer concentrations.

specific gravities of the three aerosol materials. The explanation for this disagreement between experiment and theory is not as simple as that for the non-linear behavior at low values of m. Several possible explanations exist.

The most probable cause of the disagreement is the difference in the width of the aerosol size distributions assumed by the model and generated in the laboratory. Following Bullrich (1964), the Junge distribution in the mathematical model was taken to have the boundary radii, 0.04 and 10.0 microns. A sample of a typical experimental distribution, NH4Cl-10%, was found to have the boundary radii, 0.2 and 0.85 microns. Figure 33 shows this distribution along with the Junge distribution having the same slope and representing the same total mass. The location of the Junge distribution on this graph with respect to the experimental distribution was determined by equating the total mass of the particles in each distribution as given by equation (38) to yield:

$$4/3\pi\gamma 0.434 \; c_{th}\int_{0.04}^{10.0} r^{(2-\nu)}dr = 4/3\pi\gamma 0.434 \; c_{ex}\int_{0.2}^{0.85} r^{(2-\nu)}dr \quad (50)$$

or:

$$c_{th}\int_{0.04}^{10.0} r^{(2-\nu)}dr = c_{ex}\int_{0.2}^{0.85} r^{(2-\nu)}dr \quad (51)$$

where: c_{th} = the theoretical constant of proportionality

c_{ex} = the experimental constant of proportionality

ν = the Junge exponent and was found to be 3.5 from the experimental distribution
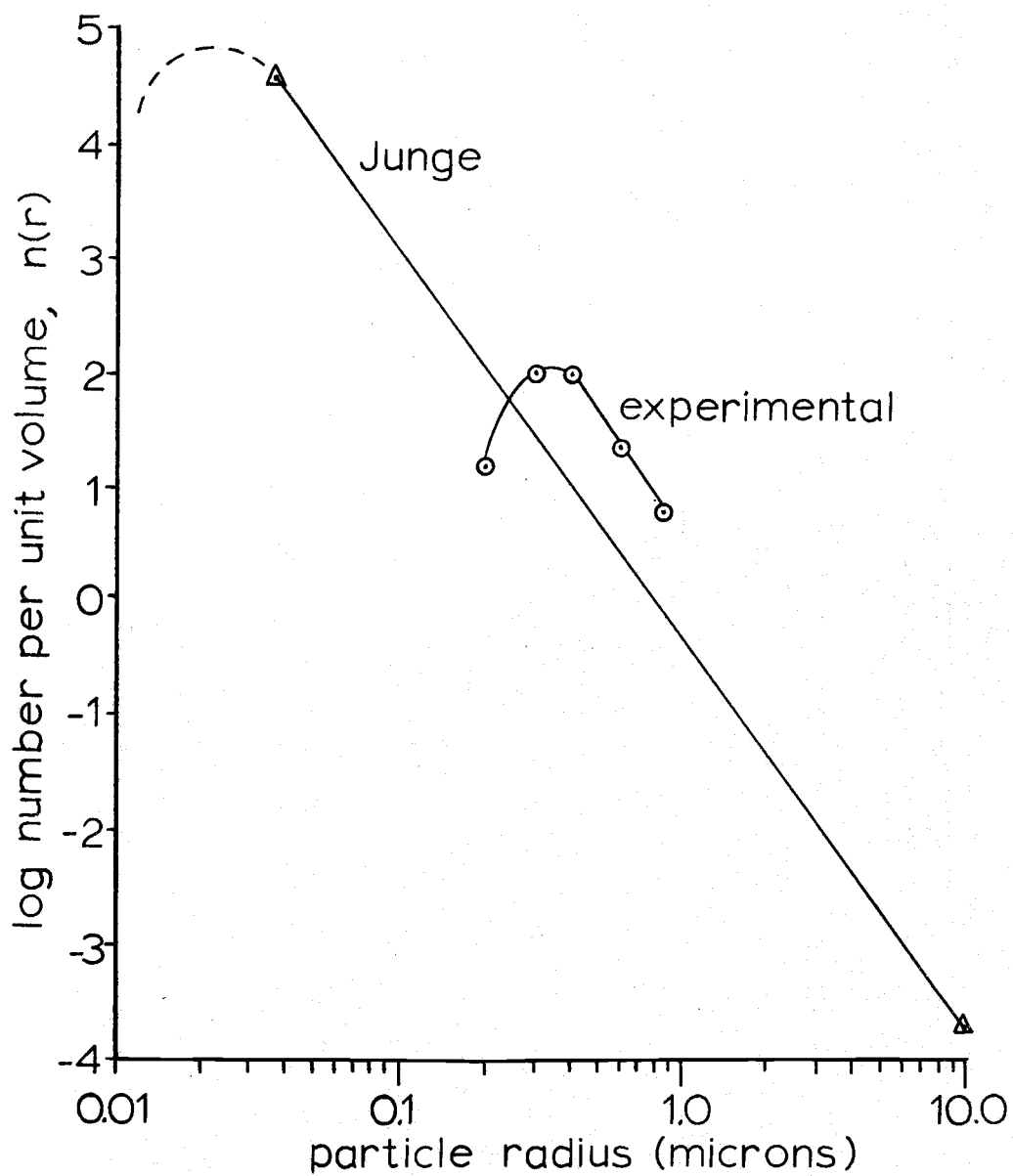
Figure 33. Comparison of experimental and "theoretical" particle size distributions on logarithmic axes.

Carrying out the integration and substituting the limits
results in the following ratio:

$$c_{ex}/c_{th} = 6.38 \qquad (52)$$

Since $n(r)$ is proportional to c, the ratio of the $n(r)$
values of the two distributions for any given value r
is equal to the ratio of their constants c.

Figure 34 shows the two curves on linear graph paper
to give an undistorted visual comparison. A rough calcu-
lation of the area under the two curves shows that the
Junge distribution has on the order of 10^2 times as many
light scattering particles as the experimental distribution.
All particles, however, do not scatter light with the same
efficiency. The light scattering efficiency Q, defined
as the ratio of the light scattered by a particle to the
light gemoetrically incident of the particle, is an os-
cillating function of r (see Figure 35) whose general shape
can be calculated (see Appendix E) following Penndorf's
method (Penndorf, 1958). Depending on the location of the
two size distribution curves with respect to maxima and
minima in the efficiency curves, it can be seen that the
fewer particles in the experimental distribution could
result in less light being scattered, and thus a smaller
value of f_s/f_u, and thus a smaller R' vs m slope than for
the theoretical distribution by an amount more than suf-
ficient to account for the disagreement in the theoret-
ical and experimental R' vs m slopes. Two obstacles,
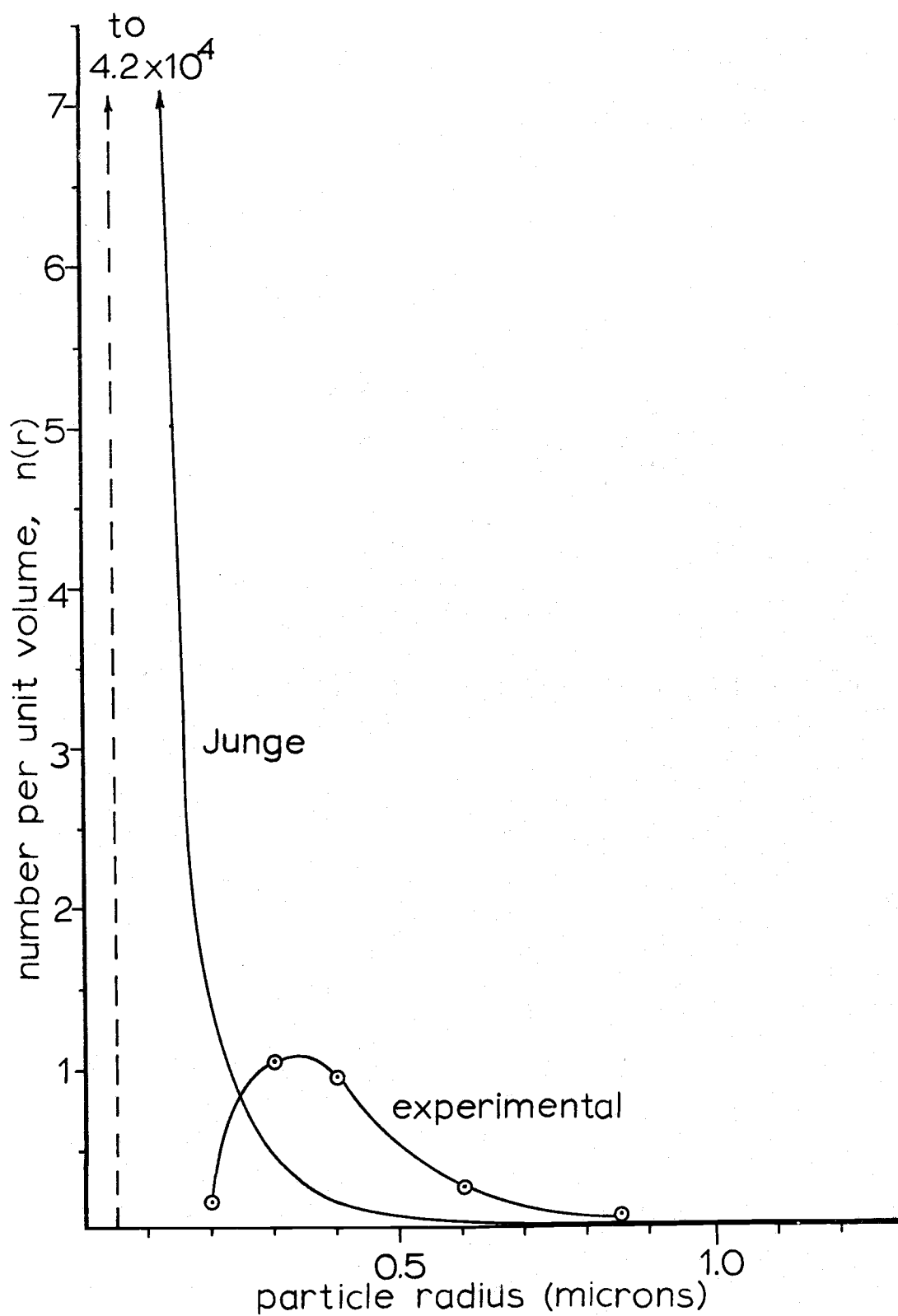
Figure 34. Comparison of experimental and "theoretical"
           particle size distributions on linear axes.
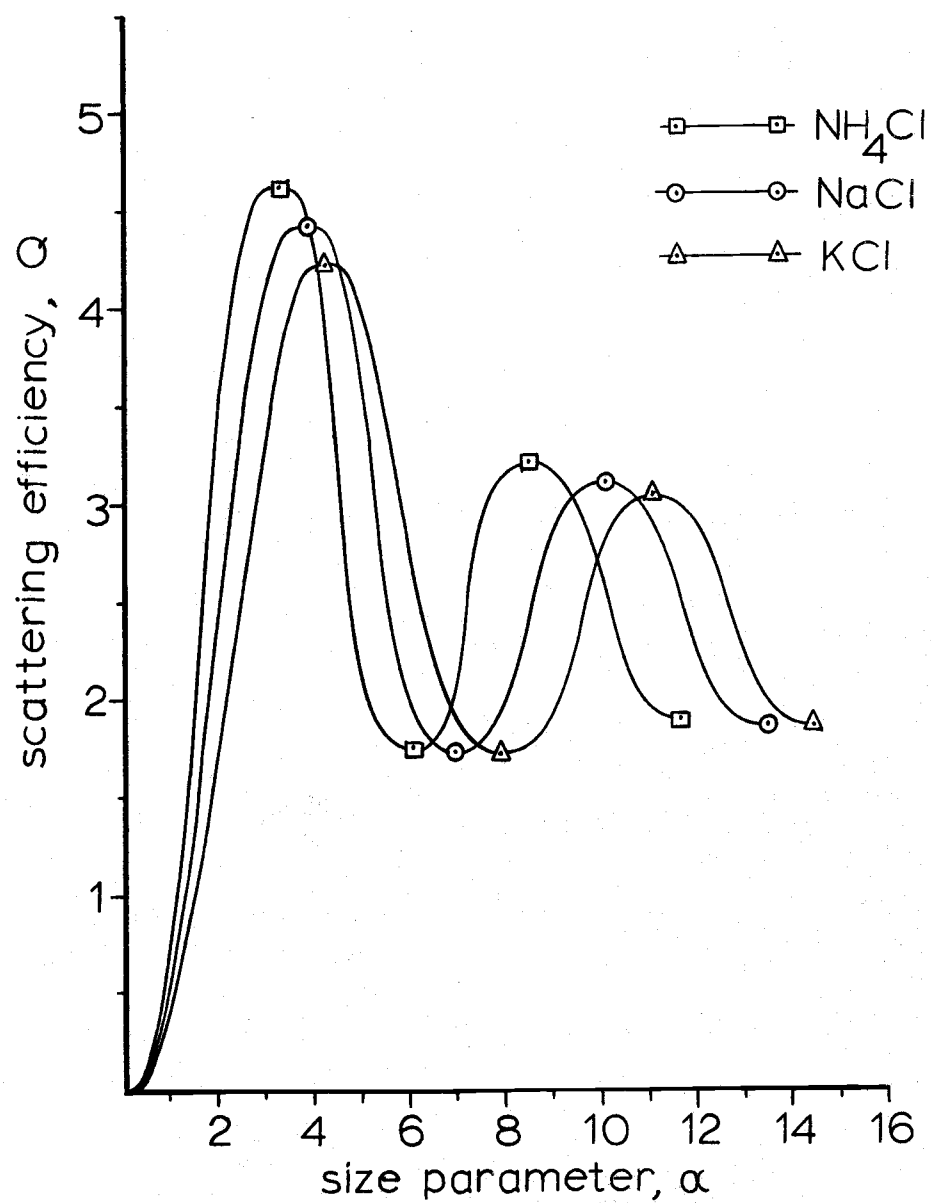
Figure 35. Scattering efficiencies of the experimental
aerosols as a function of α.

however, stand in the way of performing an exact, quantitative analysis of this source of disagreement. First, the location of the experimental distribution on the r axis is not known precisely since it was determined at only one concentration for each run and consequently may not be representative of the average over the whole run. The distribution, indeed, may not have been constant over any given run. The second obstacle is the scattering efficiency curve. Penndorf's equation for the scattering efficiency curve provides only the general trends for the curve and neglects the so-called ripple structure. Figure 36 shows an exact curve and a smoothed curve obtained by Penndorf's method for spherical particles of index of refraction 2.0. The ripple structure presents a problem in that it fluctuates so violently and also that it is not possible to obtain exact curves with the ripple structure for particles of arbitrary indices of refraction without solving numerically a horrendous infinite series from the Mie solution.

A second difference between the theoretical and experimental aerosols that might lead to a difference in the theoretical and experimental R' vs m slopes lies in the magnitude of the aerosol concentrations. The mathematical model assumes that the aerosol is dilute enough so that multiple scattering may be neglected. Due to the short pathlength for the light in the experiment, the experi-
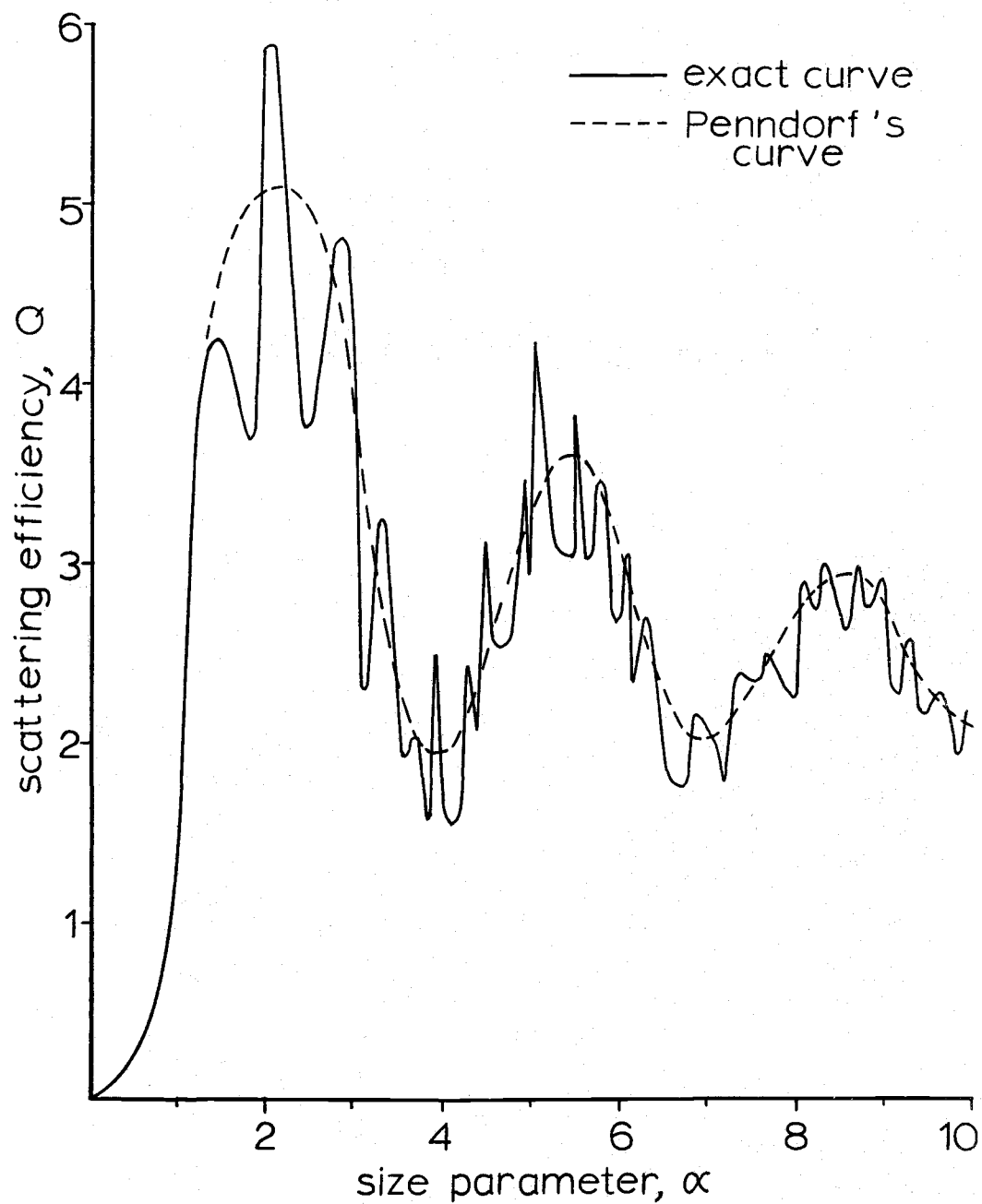
Figure 36. Comparison of scattering efficiencies calculated
by Penndorf's method and by the Mie theory.

mental aerosol was used at a high concentration to provide measureable scattered light. The measurement of scattered light in the forward direction may make an individual multiple scattering event relatively insignificant, but with large particle concentrations the probability of such an event may increase to significant proportions. The range of concentrations measured in this experiment extended from 0.001 mg/l to 0.116 mg/l. Observed concentrations in the atmosphere range from 10^{-5} mg/l to 0.005 mg/l (Hendrickson, 1968).

A third difference between the theoretical and experimental aerosols lies in the shape of the individual particles. The mathematical model was derived for spherical particles, but from a miscroscopic examination, the experimental particles were found to be either irregular in shape or cubic and rectangular. Without further experimental investigation it is not possible to estimate the error introduced in assuming that randomly oriented particles of arbitrary shape will scatter light as spheres.

A fourth possible source of disagreement between the theoretical and experimental R' vs m slopes lies in the measurement of the Junge exponent ν. The value of ν for a given aerosol is the value of the slope of the size distribution curve plotted on log-log axes. Since, as indicated earlier, the measured size distributions may not

have been representative of the average distributions over their respective runs, and it is not known whether the distribution was actually constant over any given run, a small uncertainty is introduced in using the measured values of ν to calculate the theoretical R' vs m slopes for comparison with the experimental values of the slopes.

A fifth possible source of the disagreement lies in the construction of the mathematical model. In the model, it was assumed that only those volume elements inside the conical volume V (see Figure 4) contribute to f_s, and that the contribution made by these elements is the flux scattered through the solid angle subtended by the entire objective lens. The error in these assumptions, while tending to cancel one another, over estimates f_s, which has the effect of increasing the theoretical slope. This error is discussed in detail in section III.

A sixth possible source of error is in the assumption that the light scattering for polychromatic light could be approximated by the light scattering at the mean wavelength. This assumption was made in the mathematical model based on the "smoothed" scattering curves for polydisperse aerosols (Figure 5). However, since the wavelengths of light used in this experiment (0.4 to 0.7 microns) cover a small range, and the corresponding scattering curves vary gradually and continuously, the error

introduced is expected to be small.

Thus, only a qualitative discussion of the possible sources of the discrepancy between the theoretical and experimental values of dR'/dm is offered. A more extensive (and prohibitively time consuming) investigation of the instantaneous size distributions, a comparative study of the light scattering characteristics of spheres and cubes or irregular particles, and a detailed examination of multiple scattering would be necessary in order to present a quantitative analysis of the relative magnitudes of the effects mentioned in the preceeding paragraphs. A more profitable investigation, however, might be conducted on actual atmospheric aerosols.

The dependence of the R' vs m slopes on the aerosol index of refraction and the mean aerosol diameter follows directly from the narrowness of the experimental aerosol size distributions. The mathematical model does not predict such a dependence because of the very broad Junge size distribution assumed (see section III). The Junge distribution, covering a range of aerosol diameters from 0.04 to 10.0 microns, scatters light with an efficiency that is averaged over a large part of the scattering efficiency curve for individual particles (Figure 34), and becomes independent of the mean aerosol diameter and aerosol index of refraction. A very narrow distribution, on

the other hand, will have an efficiency that is averaged over only a very small part of the efficiency curve and its light scattering characteristics will follow these curves directly. The experimental aerosols used in this investigation fall in between these two extremes. From Figure 35, it can be seen that the scattering efficiencies are not monotonic functions of the particle diameters, and that the relationship between the efficiency and the index of refraction changes for different values of the particle diameter. The scattering efficiency is related to the R' vs m slope in that a high efficiency implies that more light is scattered and more light is removed from the incident beam. This increases f_s and decreases f_u resulting in lower values of R', which increases the slope since R' is fixed at a value of 1.0 at m = 0 and the slope is negative. Thus the dependence of the slope on the aerosol index of refraction and mean diameter can be explained in part by the similar behavior of the scattering efficiency curves. The fact that the dependence did not follow the efficiency curves exactly can be explained by the imprecise knowledge of the experimental size distributions as discussed previously, the poly-chromatic incident light, the existing width of the experimental aerosol size distributions, the fact that the width of the distributions increased proportionally with the mean diameter due to the method of generation, and the increase

in _forward_ light scattering with particle diameter.

The effect of the polychromatic light is to necessitate the averaging of the scattering efficiency curves over the _wavelengths_ of light. Since the scattering efficiency curve is a function strictly of α (where $\alpha=\pi d/\lambda$), the abscissa can be expressed as a diameter scale only for a fixed value of λ, and all curves for other wavelengths of light drawn on this abscissa will be compressed or expanded in the direction of the abscissa depending on whether the wavelength is less than or greater than that used to determine the scale originally. Thus, when polychromatic light is used, a curve averaged over the curves of all the component wavelengths and weighted by the relative intensity of that component must be used. The result of this averaging is a curve with damped oscillations with maxima and minima located where they would occur for the single curve for the weighted mean wavelength.

In order to compare the total light scattering by two polydisperse aerosols, the area under curves formed by product of each size distribution curve and the efficiency curve must be compared. This is especially true in the case where the width or spread of the two size distributions is not equal. Due to the method of varying the size distribution in this experiment, the width indeed was not constant.

The last consideration, the increase in forward scattering with increasing particle size, is necessary since

the scattered flux f_s, measured in this experiment is in

fact light which has been scattered through an angle of 5°

or less and is well within the domain of forward scatter-

ing.   Thus, a group of small particles may scatter an

equal amount of light in total as a group of large par-

ticles, but the value of f_s measured for the large par-

ticles will be greater.   A possible result of this effect

is seen in Figure 32 in that the slopes for the R' vs m

curves for the 20% nebulizer concentrations in all three

cases are approximately equal to or greater than the

slopes for the 1% nebulizer concentrations, even though

the peak scattering efficiencies decrease with increasing

d.

The preceding considerations can also be cited in

explaining the weakness of the dependency of the R' vs m

slopes on the mean aerosol diameter and the index of re-

fraction.   The damping effect of polychromatic light on

the scattering efficiency curves, and the increasing for-

ward scattering with increasing particle diameter, both

tend to level out the effects of the scattering efficiency

curves, which in turn reduces any variation in R' due to

variations in d_m for any given mass loading.   The result

of this is that the R' vs m slopes for aerosols with dif-

ferent values of d_m will not differ greatly.   These same

effects also tend to bring the scattering efficiency

curves for the three different indices of refraction, which

already lie close together, into closer proximity. This decreases the difference in the scattering characteristics among the different indices of refraction, resulting in the weak dependence of the R' vs m slopes on the index of refraction.

Discussion of Errors

There are several sources of random errors that contribute to the scattering of the data points about the regression lines. The precision with which the values of f_{open} and $f_{pinhole}$ could be read from the strip chart was $\pm$ 0.001 volts, where the flux values ranged from about 0.35 volts to 6.0 volts. The aerosol photometer scale could be read to $\pm$ 0.01 scale units, which translates to $\pm$ 0.2% on the filter transmission scale at a photometer scale reading of 4.5, and $\pm$ 0.0001% at 1.5. These values become $\pm$ 0.00189 mg/l and $\pm$ 9.0 x 10^{-5} mg/l respectively, at their largest, in the case of the 20% NH_4Cl aerosol. Other sources of random errors are: amplifier drift, line voltage fluctuations (affecting the intensity of the light source), and non-homogeneous distribution of the aerosol in the test chamber.

An indication of the magnitude of the combined effect of these errors is given by the index of determination. The index of determination, which is the square of the correlation coefficient, can be interpreted as the fractional amount of the variation in the R' values that can be attributed to the linear regression of R' on m (Wine, 1964).

Thus, subtracting the index of determination from unity,
gives the fractional amount of the R' variation that is due
to non-linear effects such as the superposition of higher
order ploynomial or transcendental functions, or to <u>random</u>
<u>errors</u>. If it is accepted that the mathematical model is
an accurate representation of the physical experiment, then
the linear term in the power series expansion of R' is
approximately three orders of magnitude greater than the
quadratic term and even more so greater than the cubic
and all other higher order terms. Since the model also
predicts an absence of transcendental terms, the quantity
(1 - index of determination) can be taken as the frac-
tional amount of the R' variation that is due to random
errors. The mathematical model, however, is not a com-
pletely accurate representation of the physical experiment,
as seen by the lack of quantitative agreement between the
theoretical and experimental results. Therefore it can be
assumed that the fractional amount of the R' variation
that is due to random errors is actually less than the
quantity (1 - index of determination), and that this quan-
tity may be considered to be only an upper bound. Table
VIII lists the indices of determination and the quantity
(1 - index of determination) for each of the nine experi-
mental runs, and for the aggregate data. Using the ag-
gregate data as an example, the numbers in the table can
be interpreted as follows: 87.6% of the variation in the

TABLE VIII

Maximum Estimates of Random Errors

Aerosol	Percent	Index of Determination	Maximum Fraction of Regression Due to Random Errors
NaCl	20	0.9723	0.0277
NH$_4$Cl	20	0.8268	0.1732
KCl	20	0.9735	0.0265
NaCl	10	0.9162	0.0838
NH$_4$Cl	10	0.8133	0.1867
KCl	10	0.7420	0.2580
NaCl	1	0.7979	0.2021
NH$_4$Cl	1	0.6578	0.3422
KCl	1	0.6616	0.3384
Aggregate Data		0.8760	0.1240

ratio R', when all of the data from the nine experimental runs are plotted together, is due to the linear regression of R' on m, and <u>at most</u>, 12.4% of the variation is due to random errors. This error is comparable to that found in commercial aerosol measuring instruments such as the integrating nephelometer.

VII. CONCLUSION

Results

A mathematical model, based on the Mie theory of light scattering and the Junge aerosol size distribution, was developed that predicted the following functional dependence of R and R' on the mass concentration m:

$$R = 1/(1+Km) \tag{52}$$

and,

$$R' = 2-(1/R) = 1-Km \tag{53}$$

where: K is a constant depending on the specific gravity of the aerosol, the aerosol size distribution, and the average wavelength of incident light and the pathlength of light and the lens area of the measuring system.

Experimental measurements of R' as a function of m were made. A linear correlation between R' and m was indicated by the correlation coefficients for the nine experimental runs that ranged from 0.811 to 0.987. All but one of these coefficients indicated correlation at a 99% confidence level; the exception was at a 95% confidence level.

The values of K found experimentally ranged from 0.3088 to 0.9262 1/mg and were found to have a weak

dependence on the mean aerosol diameter and the aerosol index of refraction. The relationship between K and the mean aerosol diameter was not monotonic and followed qualitatively, the scattering efficiency curves. The relationship between K and the aerosol index of refraction appeared to be roughly proportional for the 1% and 10% nebulizer concentrations but the relationship began to deteriorate at the 20% concentration. This again followed qualitatively the behavior of the scattering efficiency curves.

Since the mathematical model was developed for atmospheric aerosols, the values of K were in quantitative disagreement with the experimental values; however, the functional relationship between R' and m indicated experimentally was identical to that in the model.

Recommendations

Based on the conclusion that R or R' and m appear to follow well defined relationships and that the relationships become independent of the aerosol index of refraction and mean aerosol diameter for broad aerosol size distributions as found in the atmosphere, it appears that a possibility exists for using measurements of the ratio R or R' as an indicator of the aerosol mass concentration. The feasibility of these measurements in the atmosphere using sunlight is even more strongly implied by the results

of the mathematical model. The model predicts that the variations in R or R' with variations in m will be much greater (by a factor of approximately 10^2) in the atmosphere than in the laboratory, resulting in a greater sensitivity in determining m from R or R', and reducing the effect of random errors in the measurement of R or R'. It is thus recommended that atmospheric study of the relationships examined in this investigation be undertaken.

Another application that might warrant further investigation is the substitution of these or similar light flux _ratio_ measurements for transmissometer measurements due to the inherent error in the lens-pinhole arrangement used in the transmissometer to eliminate scattered light. It has been found that these errors may become significant, especially in high aerosol concentrations such as those found in stack discharges (Uthe, 1973).

BIBLIOGRAPHY

BIBLIOGRAPHY

Berek, M., K. Maennchen and W. Schaefer. 1936. Tyndallo-
    metric measurement of the dust content of the air,
    and a new dust measuring apparatus. Zeitschrift fur
    Instrumentenkunde 56:49-56.

Beutell, R. G., and A. W. Brewer. 1949. Instruments for
    the measurement of visual range. Journal of Scien-
    tific Instrumentation 26:357-359.

Bullrich, K. 1964. Scattered radiation in the atmosphere
    and the natural aerosol. In: Advances in geophysics,
    ed. by H. E. Lundsberg and J. Van Miegham. Vol. 10.
    New York, Academic Press, p. 99-260.

Cadle, R. D. 1965. Particle size: theory and industrial
    applications. New York, R. Reinhold. 390 p.

California Ambient Air Quality Standards. 1973. In: Air
    pollution in California: Annual report 1972.
    Sacramento, California Air Resources Board. 37 p.

Charlson, R. J., H. Horvath and R. F. Pueschel. 1967. The
    direct measurement of atmospheric light scattering
    coefficient for studies of visibility and pollution.
    Atmospheric Environment 1:469-478.

Charlson, R. J. 1968. Visibility. Unpublished manuscript.
    Seattle, Washington.

Chinn, J. H., C. M. Sliepcevich and M. Tribus. 1955.
    Particle size distributions from angular variation of
    intensity of forward scattered light at very small
    angles. Journal of Physical Chemistry 59:841-844.

Clarenburg, L. A., and L. H. Princen. 1963. An improved
    instrument for the measurement of the mass concentra-
    tion of aerosols. Staub, Reinhaltung und der Luft
    23:234-236.

Collis, R. T. H., and M. G. H. Ligda. 1964. Laser radar
    echoes from the clear atmosphere. Nature 203:508.

Collis, R. T. H., F. G. Fernald and M. G. H. Ligda. 1964. Laser radar echoes from a stratified clear atmosphere. Nature 203:1274-1275.

Debye, P. 1909. Der Lichtendruck auf Kugeln von beliebigem Material. Annalen der Physik 30:57-61. (Microfilm)

Fisher, M. A., S. Katz, A. Lieberman and N. E. Alexander. 1955. The aerosoloscope: an instrument for the automatic counting and sizing of aerosol particles. In: Proceedings of the Third National Air Pollution Symposium, Pasadena. p. 112-119.

Friedlander, S. K., and C. S. Wang. 1966. The self-preserving size distribution for coagulation and Brownian motion. Journal of Colloid and Interface Science 22:126-132.

Gucker, F. T., and D. G. Rose. 1954. A photoelectric instrument for counting and sizing aerosol particles. British Journal of Applied Physics, Supplement No. 3, 138:S138-S143.

Gucker, F. T. 1956. Aerosol particle counters. In: Proceedings of the Eleventh Industrial Waste Utilization Conference, Purdue University, Lafayette, Indiana. p. 284-312.

Hatch, T. F. 1961. Dust and disease. In: The air we breathe, ed. by S. M. Farber and R. H. C. Wilson, Springfield, Illinois, Thomas. p. 115-125.

Hendrickson, E. R. 1968. Air sampling and quantity measurement. In: Air pollution, Vol. II, ed. by A. C. Stern, New York, Academic Press. p. 3-51.

Hewson, E. W. 1968. Meteorological measurements. In: Air pollution, Vol. II, ed. by A. C. Stern, New York, Academic Press. p. 329-387.

Hilding, A. C. 1965. Mucociliary insufficiency and its possible relation to chronic bronchitis and emphysema. Medicina Thoracalis 22:329-346.

Hodkinson, J. K. 1966. The optical measurement of aerosols. In: Aerosol science, ed. by C. N. Davies, New York, Academic Press. p. 287-357.

Hodkinson, J. K. 1966. Particle sizing by means of the forward scattering lobe. Applied Optics 5:839-843.

Horvath, H. 1966. Untersuchengung an Naturlichen und Anthropogenen Aerosolen. Doctoral dissertation. Vienna, University of Vienna. (Cited in: Charlson, R. J., H. Horvath and R. F. Pueschel. 1967. The direct measurement of atmospheric light scattering coefficient for studies of visibility and pollution. Atmospheric Environment 1:469-478.)

Junge, C. E. 1955. The size distribution and aging of natural aerosols as determined from electrical and optical data on the atmosphere. Journal of Meterology 12:13-25.

Junge, C. E. 1958. Atmospheric chemistry. In: Advances in Geophysics, ed. by H. E. Lundsberg and J. Van Miegham, Vol. 4. New York, Academic Press. p. 1-108.

Junge, C. E. 1963. Air chemistry and radioactivity. New York, Academic Press. 328 p.

Kerker, M. 1969. The scattering of light and other electro-magnetic radiation. New York, Academic Press. 666 p.

Koshi, S. 1958. A new portable aerosol photometer. Bulletin of the National Institute of Industrial Health (Japan) 1:26-32. (Cited in: Davies, C. N. 1966. Aerosol science. New York, Academic Press. 468 p.)

Lundgren, D. A., and D. W. Cooper. 1969. Effect of humidity on light scattering methods of measuring particle concentrations. Journal of the Air Pollution Control Association 19:243-247.

Lauterbach, K. E., A. D. Hayes and M. A. Coelho. 1956. An improved aerosol generator. Archives of Industrial Health 13:156-160. (Microfilm)

McCormick, R. A., and D. M. Baulch. 1962. The variation with height of the dust loading over a city as determined from the atmospheric turbidity. Journal of the Air Pollution Control Association 12:492-496.

Mercer, T. T., M. I. Tillery and M. A. Fores. 1963. An electrostatic precipitator for the collection of aerosol samples for particle size analysis. Atomic Energy Commission Research and Development Report LF-6. Alburquerque, Lovelace Foundation. 13 p.

Middleton, W. E. K. 1952. Vision through the atmosphere. Toronto, University of Toronto Press. 250 p.

Mie, G. 1908. A contribution to the optics of turbid media, especially colloidal metallic suspensions. Annalen der Physik 25:377-445. (Microfilm)

Nadel, J. A. 1965. Structure-function relationships in the airways: bronchoconstriction mediated via vagus nerves or bronchial arteries; peripheral lung constriction mediated via pulmonary arteries. Medicina Thoracalis 22:329-346.

O'Konski, C. T., M. D. Briton and W. I. Higuchi. 1958. Light scattering instrumentation for particle size distribution measurements. In: American society for Testing and Materials special technical publication No. 250, Particle size measurements, Philadelphia. p. 2-28.

Penndorf, R. 1958. An approximation method to the Mie theory for colloidal spheres. Journal of Physical Chemistry 62:1537-1542.

Peterson, C. M., and H. J. Paulus. 1967. Micrometeorological variables applied to the analysis of variations in aerosol concentration and size. Presented at the sixtieth Annual Meeting of the Air Pollution Control Association, Cleveland. Paper No. 67-133, 19 numbered pages.

Pueschel, R. F., and K. E. Noll. 1966. Comparison of calculated and observed visibilities based on measured size distributions. Presented at the annual meeting of the Pacific Northwest International Section of the Air Pollution Control Association, Seattle, Washington, November 1966. (Cited in: Charlson, R. J. 1968. Visibility, unpublished manuscript, Seattle, Washington.)

Pueschel, R. F., and K. E. Noll. 1967. Visibility and aerosol size frequency distributions. Journal of Applied Meteorology 6:1045-1052.

Randall, J. M., and J. D. Keller. 1965. Unpublished manuscript. (Cited in: Hodkinson, J. K. 1966. The optical measurement of aerosols. In: Aerosol science, ed. by C. N. Davies, New York, Academic Press. p. 287-357.)

Robinson, E. 1968. Effect on the physical properties of the atmosphere. In: Air pollution, ed. by A. C. Stern, Vol. I, New York, Academic Press. p. 349-399.

Rockney, V. D. 1959. New developments in observations and instrumentation in the Weather Bureau. Bulletin of the American Meteorological Society 40:554-560.

Rossano, A. T., Jr. 1969. Air pollution control: guidebook for management. Stamford, Conneticut, Environmental Science Services. 214 p.

Sinclair, D. 1953. Light scattering instruments as an aid in air pollution measurements. Air Repair 3(1):51-56.

Sinclair, D. 1967. A new photometer for aerosol particle size analysis. Journal of the Air Pollution Control Association 17:105-108.

Smoluchowsky, M. Von. 1918. Versuch einer mathematischen Theorie der Koagulationskinetik Kolloider Losungen. Zeitschrift fur Physikalische Chemie 92:129-168. (Microfilm)

Steffens, C., and S. Rubin. 1949. Visibility and air pollution. In: Proceedings of the First National Air Pollution Symposium, Pasadena. Stanford Research Institute. p. 103-108.

Steffens, C. 1956. Visibility and air pollution. In: Air pollution handbook, ed. by Magill, P. L., F. R. Holden and C. Ackley. New York, McGraw-Hill. p. 6.1-6.43.

Stratton, J. A. 1941. Electromagnetic theory. New York, McGraw-Hill. 615 p.

Stuke, J. 1955. The Tyndalloscope II, a new improved construction of the dust measuring instrument. Gluckauf 91:1405-1406.

Stuke, J., and J. Rzeznik. 1964. The Tyndallograph, an optical dust measuring apparatus with electrical indicator. Staub, Reinhaltung und der Luft 24:366-368.

Thomas, A. L., and S. Oglesby. 1959. Permissible-type dust counter for coal mines. Mining Engineering 11:328-331.

Thomas, A. L., N. Bira, R. H. Collins and P. Rice. 1961. A portable photometer and particle analyzer. Journal of the Instrument Society of America 8(7):52-56.

Uthe, E. 1973. Meteorologist, Stanford Research Institute. Personal communication.

Van De Hulst, H. C. 1957. Light scattering by small particles. New York, Wiley and Sons. 470 p.

Volz, F. 1959. Photometer mit Selen-Photoelement zur spektralen Messung der Sonnenstrahlung und zur Bestimmung der Wellenlanenabhangigkeit der Dunsttrubung. Archiv fur Meteorologie, Geophysik und Bioklimatologie, series B, 10:100-131. (Cited in: McCormick, R. A., and D. M. Baulch. 1962. The variation with height of the dust loading over a city as determined from the atmospheric turbidity. Journal of the Air Pollution Control Association 12:492-496.

Whitby, K. T., and W. E. Clark. 1966. Electric aerosol particle counting and size distribution measurement system for 0.015 to 1 micron range. Tellus 18: 573-586.

Wine, R. L. 1964. Statistics for scientists and engineers. New Jersey, Prentice-Hall. 671 p.

Zinky, W. R. 1962. A new tool for air pollution control: the aerosol particle counter. Journal of the Air Pollution Control Association 12:578-583.

APPENDICES

APPENDIX A: R vs m data

m $(10^{-3}mg/l)$	R	Relative Humidity(%)	Temperature($^{\circ}$F)
NaCl 20%			
0.112	0.8636	36.5	72
0.400	0.8627	37.0	72
0.768	0.8577	38.5	72
0.920	0.8597	39.0	72
1.240	0.8611	39.0	72
2.240	0.8636	39.0	72
4.16	0.8601		72
4.880	0.8595	40.0	72
6.480	0.8595	40.0	72
7.680	0.8581	40.0	72
10.80	0.8546	41.0	72.5
15.20	0.8550	41.0	72.5
25.60	0.8532	41.0	72.5
32.0	0.8455	41.0	72.5
40.80	0.8464	41.5	72.5
70.40	0.8323	42.0	72.5
86.40	0.8547	42.0	72.5
NH_4Cl 20%			
0.161	0.8511	43.0	73.0
0.514	0.8525	43.0	73.0
0.857	0.8557	43.0	73.0

APPENDIX A (continued)

m $(10^{-3}$mg/l)	R	Relative Humidity(%)	Temperature(°F)
0.911	0.8541	44.0	73.0
1.098	0.8547	44.0	73.0
1.553	0.8527	44.0	73.5
1.875	0.8507	44.0	74.0
2.143	0.8512	44.0	74.0
2.624	0.8498	44.0	74.0
3.480	0.8503	44.0	74.0
4.390	0.8525	44.0	74.0
5.140	0.8489	44.0	74.0
6.000	0.8514	44.0	74.0
7.230	0.8490	44.0	74.0
9.370	0.8486	44.0	74.0
13.92	0.8447	44.0	74.0
18.75	0.8464	44.0	74.0
22.50	0.8423	44.0	74.0
25.45	0.8400	44.0	74.0
30.52	0.8415	44.5	74.0
35.88	0.8353	44.5	74.0
KCl 20%			
0.240	0.8574	43.0	72.0
0.356	0.8556	44.0	72.0

APPENDIX A (continued)

m	(10⁻³mg/l) R	Relative Humidity(%)	Temperature(°F)
0.788	0.8594	44.0	72.0
1.200	0.8573	44.5	72.0
1.817	0.8636	45.0	72.0
3.150	0.8666	46.0	73.0
4.175	0.8632	46.0	73.0
6.040	0.8615	46.0	73.0
15.77	0.8576	46.0	73.0
23.65	0.8543	46.0	73.0
33.24	0.8460	46.0	73.0
45.25	0.8406	46.0	73.0
48.00	0.8485	46.0	73.0
43.80	0.8458	46.5	75.0
58.90	0.8441	47.75	75.0
72.00	0.8331	47.0	76.0
89.10	0.8302	47.0	76.5
92.50	0.8235	47.0	77.0
116.60	0.8141	47.5	77.0

NaCl 10%

m	(10⁻³mg/l) R	Relative Humidity(%)	Temperature(°F)
0.148	0.8638	38.0	71.5
0.296	0.8558	39.0	71.5
0.363	0.8546	39.0	71.5
0.472	0.8509	39.5	72.0

APPENDIX A (continued)

m	$(10^{-3}mg/l)$	R	Relative Humidity(%)	Temperature($^{\circ}$F)
0.787		0.8633	40.0	72.0
1.390		0.8610	40.5	72.0
1.936		0.8610	41.0	72.0
3.330		0.8578	41.0	72.0
3.995		0.8575	42.0	72.0
5.810		0.8590	42.0	72.0
8.890		0.8574	42.0	72.0
11.50		0.8551	42.0	72.0
16.94		0.8534	42.0	72.0
22.68		0.8509	42.5	72.0
39.3		0.8503	43.0	72.0
51.40		0.8462	44.0	72.0
65.30		0.8419	44.0	72.0
83.50		0.8432	44.0	72.0
NH$_4$Cl 10%				
0.175		0.8511	43.0	72.0
0.389		0.8515	44.0	73.0
0.486		0.8538	44.0	73.0
0.974		0.8478	44.0	73.0
2.630		0.8506	44.0	73.0
3.460		0.8523	44.0	73.0
3.990		0.8490	44.0	73.0

APPENDIX A (continued)

m	$(10^{-3}mg/l)$	R	Relative Humidity(%)	Temperature($^{\circ}$F)
5.500		0.8463	44.0	73.0
6.470		0.8482	44.0	73.0
7.680		0.8521	44.0	73.0
16.06		0.8468	44.0	73.0
17.50		0.8434	44.5	73.0
19.95		0.8408	44.5	73.0
23.83		0.8431	45.0	73.0
34.50		0.8318	45.0	73.0
39.90		0.8326	45.0	73.0
KCl 10%				
0.328		0.8674	39.0	73.5
0.618		0.8635	40.0	73.5
0.965		0.8684	40.0	73.5
1.512		0.8668	41.0	73.0
2.207		0.8666	41.0	73.0
2.521		0.8663	41.0	73.0
4.410		0.8644	41.0	73.0
4.915		0.8664	41.0	73.0
8.510		0.8649	41.0	73.0
11.66		0.8656	42.0	73.5
14.69		0.8639	42.0	73.5
11.66		0.8633	44.0	74.0

APPENDIX A (continued)

m	$(10^{-3}mg/l)$	R	Relative Humidity(%)	Temperature(°F)
11.72		0.8633	44.0	75.0
16.40		0.8627	44.0	75.5
22.70		0.8634	44.0	76.0
35.30		0.8643	44.0	76.0
42.80		0.8548	44.5	76.5
45.40		0.8577	44.5	76.5
NaCl 1%				
0.0854		0.8561	43.0	73.5
0.121		0.8598	43.0	73.5
0.208		0.8630	43.0	73.5
0.219		0.8610	43.0	73.5
0.314		0.8609	43.0	73.5
0.383		0.8625	44.0	74.0
0.458		0.8643	44.0	74.0
0.574		0.8610	44.0	74.0
0.786		0.8641	44.5	74.0
3.076		0.8641	44.5	74.0
5.740		0.8605	44.5	74.0
10.940		0.8568	46.0	73.5
15.370		0.8555	46.0	73.5
19.47		0.8557	46.0	73.0
21.20		0.8557	46.0	73.0

APPENDIX A (continued)

m $(10^{-3}mg/1)$	R	Relative Humidity(%)	Temperature($^{\circ}$F)
NH$_4$Cl 1%			
0.065	0.8564	38.0	73.5
0.128	0.8576	40.0	73.5
0.196	0.8556	41.0	74.0
0.266	0.8558	41.0	74.0
0.427	0.8540	41.0	74.0
0.498	0.8551	41.0	74.0
0.723	0.8559	41.0	74.0
0.978	0.8592	41.0	74.0
2.015	0.8493	42.0	73.5
2.37	0.8551	42.0	73.5
2.962	0.8512	42.0	73.5
3.67	0.8556	42.5	73.5
4.80	0.8529	43.0	73.5
6.04	0.8528	43.0	73.5
7.11	0.8528	43.0	73.5
15.40	0.8525	44.0	73.0
20.73	0.8386	44.0	73.0
9.59	0.8512	45.0	78.5
20.70	0.8457	46.0	79.5

APPENDIX A (continued)

m	$(10^{-3}mg/l)$ R	Relative Humidity(%)	Temperature($^\circ$F)
KCl 1%			
0.056	0.8621	45.0	71.0
0.114	0.8620	45.0	72.0
0.158	0.8606	45.5	72.0
0.192	0.8661	46.0	72.0
0.239	0.8621	46.0	72.0
0.269	0.8606	46.0	72.0
0.224	0.8556	46.0	72.0
0.341	0.8622	46.0	72.0
0.436	0.8609	46.0	72.0
0.614	0.8587	46.0	72.0
0.896	0.8595	46.0	72.0
1.45	0.8621	46.0	72.0
2.30	0.8614	46.0	72.0
3.16	0.8590	46.0	72.0
4.52	0.8615	46.0	72.0
11.52	0.8572	47.0	73.0
9.82	0.8601	47.0	76.0
16.23	0.8579	48.0	77.5
25.59	0.8564	48.5	79.0
27.30	0.8534	48.5	79.0
7.51	0.8646	47.0	73.0

APPENDIX B: R' vs m Data and Comparison with
              Least-Squares Line

m (10^{-3}mg/1) (measured)	R' (measured)	R' (least-squares)	Difference	
NaCl 20%				
1.24000E+00	9.98300E-01	9.98320E-01	0.00%	****EVEN****
2.24000E+00	1.00160E+00	9.97773E-01	0.40%	****ABOVE***
4.16000E+00	9.97000E-01	9.96721E-01	0.00%	****EVEN****
6.48000E+00	9.96100E-01	9.95451E-01	0.00%	****EVEN****
7.68000E+00	9.94300E-01	9.94794E-01	0.00%	****EVEN****
1.08000E+01	9.89400E-01	9.93085E-01	0.30%	****BELOW***
1.52000E+01	9.90000E-01	9.90676E-01	0.00%	****EVEN****
2.56000E+01	9.87600E-01	9.84981E-01	0.20%	****ABOVE***
3.20000E+01	9.76900E-01	9.81477E-01	0.40%	****BELOW***
4.08000E+01	9.78100E-01	9.76658E-01	0.10%	****ABOVE***
7.04000E+01	9.58100E-01	9.60449E-01	0.20%	****BELOW***
8.64000E+01	9.54300E-01	9.51688E-01	0.20%	****ABOVE***
4.88000E+00	9.96100E-01	9.96327E-01	0.00%	****EVEN****
NH$_4$Cl 20%				
1.09800E+00	1.00170E+00	9.99473E-01	0.20%	****ABOVE***
1.55300E+00	9.98900E-01	9.99164E-01	0.00%	****EVEN****
1.87500E+00	9.96200E-01	9.98945E-01	0.20%	****BELOW***
2.14300E+00	9.96900E-01	9.98763E-01	0.10%	****BELOW***
2.62400E+00	9.95000E-01	9.98437E-01	0.30%	****BELOW***
3.48000E+00	9.95600E-01	9.97855E-01	0.20%	****BELOW***
4.39000E+00	9.98700E-01	9.97238E-01	0.10%	****ABOVE***
5.14000E+00	9.93700E-01	9.96728E-01	0.30%	****BELOW***
6.00000E+00	9.97100E-01	9.96144E-01	0.00%	****EVEN****
7.23000E+00	9.93800E-01	9.95309E-01	0.10%	****BELOW***
9.37000E+00	9.93300E-01	9.93856E-01	0.00%	****EVEN****
1.39200E+01	9.87900E-01	9.90767E-01	0.20%	****BELOW***
1.87500E+01	9.90200E-01	9.87488E-01	0.20%	****ABOVE***
2.25000E+01	9.84500E-01	9.84941E-01	0.00%	****EVEN****
2.54500E+01	9.81300E-01	9.82938E-01	0.10%	****BELOW***
3.05200E+01	9.83300E-01	9.79496E-01	0.30%	****ABOVE***
3.58800E+01	9.74500E-01	9.75857E-01	0.10%	****BELOW***

APPENDIX B (continued)

m (10^{-3}mg/l) (measured)	R' (measured)	R' (least-squares)	Difference	
KCl 20%				
1.81700E+00	9.97300E-01	9.97917E-01	0.00%	****EVEN****
3.15000E+00	1.00130E+00	9.97116E-01	0.40%	****ABOVE***
4.17500E+00	9.96800E-01	9.96501E-01	0.00%	****EVEN****
6.04000E+00	9.94600E-01	9.95380E-01	0.00%	****EVEN****
1.57700E+01	9.89300E-01	9.89534E-01	0.00%	****EVEN****
2.36500E+01	9.84700E-01	9.84799E-01	0.00%	****EVEN****
3.32400E+01	9.73200E-01	9.79038E-01	0.50%	****BELOW***
4.52500E+01	9.65700E-01	9.71822E-01	0.60%	****BELOW***
4.80000E+01	9.72000E-01	9.70169E-01	0.10%	****ABOVE***
4.38000E+01	9.73000E-01	9.72693E-01	0.00%	****EVEN****
5.89000E+01	9.70600E-01	9.63620E-01	0.70%	****ABOVE***
7.20000E+01	9.55000E-01	9.55750E-01	0.00%	****EVEN****
8.91000E+01	9.50800E-01	9.45475E-01	0.50%	****ABOVE***
9.25000E+01	9.41000E-01	9.43433E-01	0.20%	****BELOW***
1.16600E+02	9.26900E-01	9.28953E-01	0.20%	****BELOW***
NaCl 10%				
1.39000E+00	1.00050E+00	9.97239E-01	0.30%	****ABOVE***
1.93600E+00	1.00040E+00	9.97068E-01	0.30%	****ABOVE***
3.33000E+00	9.96100E-01	9.96630E-01	0.00%	****EVEN****
3.99500E+00	9.95700E-01	9.96421E-01	0.00%	****EVEN****
5.81000E+00	9.97000E-01	9.95852E-01	0.10%	****ABOVE***
8.89000E+00	9.95600E-01	9.94885E-01	0.00%	****EVEN****
1.15000E+01	9.92400E-01	9.94066E-01	0.10%	****BELOW***
1.69400E+01	9.90100E-01	9.92358E-01	0.20%	****BELOW***
2.26800E+01	9.86700E-01	9.90556E-01	0.30%	****BELOW***
3.93000E+01	9.85900E-01	9.85340E-01	0.00%	****EVEN****
5.14000E+01	9.80100E-01	9.81542E-01	0.10%	****BELOW***
6.53000E+01	9.74100E-01	9.77179E-01	0.30%	****BELOW***
8.35000E+01	9.76000E-01	9.71466E-01	0.40%	****ABOVE***

APPENDIX B (continued)

m $(10^{-3}mg/l)$ (measured)	R' (measured)	R' (least-squares)	Difference	
NH$_4$Cl 10%				
2.63000E+00	1.00010E+00	9.99983E-01	0.00%	****EVEN****
3.46000E+00	1.00150E+00	9.99459E-01	0.20%	****ABOVE****
3.99000E+00	9.97900E-01	9.99124E-01	0.10%	****BELOW****
5.50000E+00	9.94200E-01	9.98170E-01	0.30%	****BELOW****
6.47000E+00	9.96900E-01	9.97558E-01	0.00%	****EVEN****
7.68000E+00	1.00220E+00	9.96793E-01	0.50%	****ABOVE****
1.60600E+01	9.94900E-01	9.91500E-01	0.30%	****ABOVE****
1.75000E+01	9.90100E-01	9.90591E-01	0.00%	****EVEN****
1.99500E+01	9.86500E-01	9.89043E-01	0.20%	****BELOW****
2.38300E+01	9.89700E-01	9.86593E-01	0.30%	****ABOVE****
2.92000E+01	9.90500E-01	9.83201E-01	0.70%	****ABOVE****
3.45000E+01	9.73600E-01	9.79853E-01	0.60%	****BELOW****
3.99000E+01	9.74500E-01	9.76442E-01	0.10%	****BELOW****
KCl 10%				
1.51200E+00	9.98700E-01	9.98259E-01	0.00%	****EVEN****
2.20700E+00	9.98500E-01	9.98077E-01	0.00%	****EVEN****
2.52100E+00	9.98100E-01	9.97995E-01	0.00%	****EVEN****
4.41000E+00	9.95600E-01	9.97501E-01	0.10%	****BELOW****
4.91500E+00	9.98200E-01	9.97368E-01	0.00%	****EVEN****
8.51000E+00	9.96200E-01	9.96428E-01	0.00%	****EVEN****
1.16600E+01	9.97200E-01	9.95603E-01	0.10%	****ABOVE****
1.46900E+01	9.94900E-01	9.94811E-01	0.00%	****EVEN****
1.16600E+01	9.94100E-01	9.95603E-01	0.10%	****BELOW****
1.17200E+01	9.94100E-01	9.95588E-01	0.10%	****BELOW****
1.64000E+01	9.93200E-01	9.94363E-01	0.10%	****BELOW****
2.27000E+01	9.94200E-01	9.92715E-01	0.10%	****ABOVE****
3.02400E+01	9.91200E-01	9.90742E-01	0.00%	****EVEN****
3.53000E+01	9.95400E-01	9.89418E-01	0.60%	****ABOVE****
4.28000E+01	9.82600E-01	9.87455E-01	0.40%	****BELOW****
4.54000E+01	9.86500E-01	9.86775E-01	0.00%	****EVEN****

APPENDIX B (continued)

m (10^{-3}mg/l) (measured)	R' (measured)	R' (least-squares)	Difference	
NaCl 1%				
3.07600E+00	1.00200E+00	9.99236E-01	0.20%	****ABOVE***
5.74000E+00	9.96900E-01	9.97665E-01	0.00%	****EVEN****
1.09400E+01	9.91800E-01	9.94598E-01	0.20%	****BELOW***
1.53700E+01	9.90100E-01	9.91986E-01	0.10%	****BELOW***
1.94700E+01	9.90400E-01	9.89568E-01	0.00%	****EVEN****
2.12000E+01	9.90400E-01	9.88547E-01	0.10%	****ABOVE***
NH$_4$Cl 1%				
2.01500E+00	9.91600E-01	9.98221E-01	0.60%	****BELOW***
2.37000E+00	9.99500E-01	9.97948E-01	0.10%	****ABOVE***
2.96200E+00	9.94200E-01	9.97492E-01	0.30%	****BELOW***
3.67000E+00	1.00030E+00	9.96948E-01	0.30%	****ABOVE***
4.80000E+00	9.96500E-01	9.96078E-01	0.00%	****EVEN****
6.04000E+00	9.96400E-01	9.95123E-01	0.10%	****ABOVE***
7.11000E+00	9.96400E-01	9.94300E-01	0.20%	****ABOVE***
1.54000E+01	9.92000E-01	9.87919E-01	0.40%	****ABOVE***
2.07300E+01	9.76500E-01	9.83817E-01	0.70%	****BELOW***
9.59000E+00	9.94200E-01	9.92314E-01	0.10%	****ABOVE***
2.07000E+01	9.86400E-01	9.83840E-01	0.20%	****ABOVE***
KCl 1%				
1.45000E+00	1.00000E+00	9.99946E-01	0.00%	****EVEN****
2.30500E+00	9.99100E-01	9.99618E-01	0.00%	****EVEN****
3.16000E+00	9.95800E-01	9.99289E-01	0.30%	****BELOW***
4.52000E+00	9.99200E-01	9.98767E-01	0.00%	****EVEN****
7.51000E+00	1.00340E+00	9.97618E-01	0.50%	****ABOVE***
9.82000E+00	9.97400E-01	9.96730E-01	0.00%	****EVEN****
1.15200E+01	9.93400E-01	9.96077E-01	0.20%	****BELOW***
1.62300E+01	9.94200E-01	9.94268E-01	0.00%	****EVEN****
2.55900E+01	9.92300E-01	9.90672E-01	0.10%	****ABOVE***
2.73000E+01	9.88200E-01	9.90015E-01	0.10%	****BELOW***

APPENDIX C: Aggregate R' vs m Data and Comparison
              with Least-Squares Line

m (10^{-3}mg/1) (measured)	R' (measured)	R' (least-squares)	Difference	
1.67000E+00	9.96300E-01	9.99365E-01	0.30%	****BELOW***
2.63000E+00	1.00010E+00	9.98846E-01	0.10%	****ABOVE***
3.46000E+00	1.00150E+00	9.98398E-01	0.30%	****ABOVE***
3.99000E+00	9.97900E-01	9.98111E-01	0.00%	****EVEN****
5.50000E+00	9.94200E-01	9.97294E-01	0.30%	****BELOW***
6.47000E+00	9.96900E-01	9.96770E-01	0.00%	****EVEN****
7.68000E+00	1.00220E+00	9.96116E-01	0.60%	****ABOVE***
1.60600E+01	9.94900E-01	9.91584E-01	0.30%	****ABOVE***
1.75000E+01	9.90100E-01	9.90806E-01	0.00%	****EVEN****
1.99500E+01	9.86500E-01	9.89481E-01	0.30%	****BELOW***
2.38300E+01	9.89700E-01	9.87383E-01	0.20%	****ABOVE***
2.92000E+01	9.90500E-01	9.84479E-01	0.60%	****ABOVE***
3.45000E+01	9.73600E-01	9.81613E-01	0.80%	****BELOW***
3.99000E+01	9.74500E-01	9.78694E-01	0.40%	****BELOW***
1.39000E+00	1.00050E+00	9.99517E-01	0.00%	****EVEN****
1.93600E+00	1.00040E+00	9.99222E-01	0.10%	****ABOVE***
3.33000E+00	9.96100E-01	9.98468E-01	0.20%	****BELOW***
3.99500E+00	9.95700E-01	9.98108E-01	0.20%	****BELOW***
5.81000E+00	9.97000E-01	9.97127E-01	0.00%	****EVEN****
8.89000E+00	9.95600E-01	9.95461E-01	0.00%	****EVEN****
1.15000E+01	9.92400E-01	9.94050E-01	0.10%	****BELOW***
1.69400E+01	9.90100E-01	9.91109E-01	0.10%	****BELOW***
2.26800E+01	9.86700E-01	9.88005E-01	0.10%	****BELOW***
3.93000E+01	9.85900E-01	9.79018E-01	0.70%	****ABOVE***
5.14000E+01	9.80100E-01	9.72475E-01	0.70%	****ABOVE***
6.53000E+01	9.74100E-01	9.64959E-01	0.90%	****ABOVE***
8.35000E+01	9.76000E-01	9.55118E-01	2.10%	****ABOVE***
1.81700E+00	9.97300E-01	9.99286E-01	0.10%	****BELOW***
3.15000E+00	1.00130E+00	9.98565E-01	0.20%	****ABOVE***
4.17500E+00	9.96800E-01	9.98011E-01	0.10%	****BELOW***
6.04000E+00	9.94600E-01	9.97002E-01	0.20%	****BELOW***
1.57700E+01	9.89300E-01	9.91741E-01	0.20%	****BELOW***
2.36500E+01	9.84700E-01	9.87480E-01	0.20%	****BELOW***
3.32400E+01	9.73200E-01	9.82295E-01	0.90%	****BELOW***
4.52500E+01	9.65700E-01	9.75801E-01	1.00%	****BELOW***
4.80000E+01	9.72000E-01	9.74314E-01	0.20%	****BELOW***
4.38000E+01	9.73000E-01	9.76585E-01	0.30%	****BELOW***
5.89000E+01	9.70600E-01	9.68420E-01	0.20%	****ABOVE***
7.20000E+01	9.55000E-01	9.61336E-01	0.60%	****BELOW***
8.91000E+01	9.50800E-01	9.52090E-01	0.10%	****BELOW***
9.25000E+01	9.41000E-01	9.50252E-01	0.90%	****BELOW***
1.16600E+02	9.26900E-01	9.37220E-01	1.10%	****BELOW***

APPENDIX C (continued)

m (10^{-3}mg/1) (measured)	R' (measured)	R' (least-squares)	Difference	
1.09800E+00	1.00170E+00	9.99675E-01	0.20%	****ABOVE***
1.55300E+00	1.00970E+00	9.99429E-01	1.00%	****ABOVE***
1.87500E+00	9.96200E-01	9.99255E-01	0.30%	****BELOW***
2.14300E+00	9.96900E-01	9.99110E-01	0.20%	****BELOW***
2.62400E+00	9.95000E-01	9.98850E-01	0.30%	****BELOW***
3.48000E+00	9.95600E-01	9.98387E-01	0.20%	****BELOW***
4.39000E+00	9.98700E-01	9.97895E-01	0.00%	****EVEN****
5.14000E+00	9.93700E-01	9.97489E-01	0.30%	****BELOW***
6.00000E+00	9.97100E-01	9.97024E-01	0.00%	****EVEN****
7.23000E+00	9.93800E-01	9.96359E-01	0.20%	****BELOW***
9.37000E+00	9.93300E-01	9.95202E-01	0.10%	****BELOW***
1.39200E+01	9.87900E-01	9.92742E-01	0.40%	****BELOW***
1.87500E+01	9.90200E-01	9.90130E-01	0.00%	****EVEN****
2.25000E+01	9.84500E-01	9.88102E-01	0.30%	****BELOW***
2.54500E+01	9.81300E-01	9.86507E-01	0.50%	****BELOW***
3.05200E+01	9.83300E-01	9.83766E-01	0.00%	****EVEN****
3.58800E+01	9.74500E-01	9.80867E-01	0.60%	****BELOW***
1.24000E+00	9.98300E-01	9.99598E-01	0.10%	****BELOW***
2.24000E+00	1.00160E+00	9.99057E-01	0.30%	****ABOVE***
4.16000E+00	9.97000E-01	9.98019E-01	0.10%	****BELOW***
4.88000E+00	9.96100E-01	9.97630E-01	0.10%	****BELOW***
6.48000E+00	9.96100E-01	9.96765E-01	0.00%	****EVEN****
7.68000E+00	9.94300E-01	9.96116E-01	0.10%	****BELOW***
1.08000E+01	9.89400E-01	9.94429E-01	0.50%	****BELOW***
1.52000E+01	9.90000E-01	9.92049E-01	0.20%	****BELOW***
2.56000E+01	9.87600E-01	9.86426E-01	0.10%	****ABOVE***
3.20000E+01	9.76900E-01	9.82965E-01	0.60%	****BELOW***
4.08000E+01	9.78100E-01	9.78207E-01	0.00%	****EVEN****
7.04000E+01	9.58100E-01	9.62202E-01	0.40%	****BELOW***
8.64000E+01	9.54300E-01	9.53550E-01	0.00%	****EVEN****
1.51200E+00	9.98700E-01	9.99451E-01	0.00%	****EVEN****
2.20700E+00	9.98500E-01	9.99075E-01	0.00%	****EVEN****
2.52100E+00	9.98100E-01	9.98905E-01	0.00%	****EVEN****
3.41000E+00	9.95600E-01	9.97884E-01	0.20%	****BELOW***
4.91500E+00	9.98200E-01	9.97611E-01	0.00%	****EVEN****
8.51000E+00	9.96200E-01	9.95667E-01	0.00%	****EVEN****
1.16600E+01	9.97200E-01	9.93964E-01	0.30%	****ABOVE***
1.46900E+01	9.94900E-01	9.92325E-01	0.20%	****ABOVE***
1.16600E+01	9.94100E-01	9.93964E-01	0.00%	****EVEN****
1.17200E+01	9.94100E-01	9.93931E-01	0.00%	****EVEN****
1.64000E+01	9.93200E-01	9.91401E-01	0.10%	****ABOVE***
2.27000E+01	9.94200E-01	9.87994E-01	0.60%	****ABOVE***
3.02400E+01	9.91200E-01	9.83917E-01	0.70%	****ABOVE***
3.53000E+01	9.95400E-01	9.81181E-01	1.40%	****ABOVE***
4.28000E+01	9.82600E-01	9.77125E-01	0.50%	****ABOVE***
4.54000E+01	9.86500E-01	9.75720E-01	1.10%	****ABOVE***

APPENDIX C (continued)

m (10^{-3}mg/l) (measured)	R' (measured)	R' (least-squares)	Difference	
1.45000E+00	9.99500E-01	9.99484E-01	0.00%	****EVEN****
2.30500E+00	9.98600E-01	9.99022E-01	0.00%	****EVEN****
3.16000E+00	9.95300E-01	9.98560E-01	0.30%	****BELOW***
4.52000E+00	9.98700E-01	9.97824E-01	0.00%	****EVEN****
7.51000E+00	1.00290E+00	9.96208E-01	0.60%	****ABOVE***
1.15200E+01	9.92900E-01	9.94039E-01	0.10%	****BELOW***
9.82000E+00	9.96900E-01	9.94959E-01	0.10%	****ABOVE***
1.62300E+01	9.93700E-01	9.91492E-01	0.20%	****ABOVE***
2.55900E+01	9.91800E-01	9.86431E-01	0.50%	****ABOVE***
2.73000E+01	9.87700E-01	9.85507E-01	0.20%	****ABOVE***
2.01500E+00	9.93500E-01	9.99179E-01	0.50%	****BELOW***
2.37000E+00	1.00140E+00	9.98987E-01	0.20%	****ABOVE***
2.96200E+00	9.96100E-01	9.98667E-01	0.20%	****BELOW***
3.67000E+00	1.00220E+00	9.98284E-01	0.30%	****ABOVE***
4.80000E+00	9.98400E-01	9.97673E-01	0.00%	****EVEN****
6.04000E+00	9.98300E-01	9.97002E-01	0.10%	****ABOVE***
7.11000E+00	9.98300E-01	9.96424E-01	0.10%	****ABOVE***
1.54000E+01	9.97900E-01	9.91941E-01	0.60%	****ABOVE***
9.69000E+00	9.96100E-01	9.95029E-01	0.10%	****ABOVE***
2.07000E+01	9.88300E-01	9.89075E-01	0.00%	****EVEN****
3.07600E+00	1.00100E+00	9.98605E-01	0.20%	****ABOVE***
5.74000E+00	9.95900E-01	9.97165E-01	0.10%	****BELOW***
1.09400E+01	9.90800E-01	9.94353E-01	0.30%	****BELOW***
1.53700E+01	9.89100E-01	9.91958E-01	0.20%	****BELOW***
1.94700E+01	9.89400E-01	9.89741E-01	0.00%	****EVEN****
2.12000E+01	9.89400E-01	9.88805E-01	0.00%	****EVEN****

APPENDIX D: Particle Size Distribution Data

| Diameter | Number Count |||||||||
| | NaCl ||| NH4Cl ||| KCl |||
(microns)	1%	10%	20%	1%	10%	20%	1%	10%	20%
0.3	115			120					
0.4	74	36	11	92	15	7	98	16	13
0.6	37	88	28	10	103	16	94	130	45
0.8	4	55	58	4	93	24	32	73	50
1.2	7	37	67	3	24	52	5	26	47
1.7		7	42		6	62		6	22
2.5		2	10			13		2	19
3.5						5			9
Mean (microns)	0.41	0.76	1.13	0.37	0.75	1.39	0.55	0.74	1.19

APPENDIX E: Penndorf's Method for Calculating the
               General Shape of the Scattering
               Efficiency Curve.

Penndorf (1958) has developed empirical expressions

for the values of $\rho = 2\alpha(n-1)$, where n is the particle

index of refraction, at which the extrema of the scat-

tering efficiency curve occur and the value of the scat-

tering efficiency at these extrema.

The equation for the location of the extrema is given

by:

$$\rho_y(n) = \rho_y(1) + 0.3(n-1) \qquad\qquad (E-1)$$

where: $\rho_y(n)$ is the value of ρ corresponding to the yth

          max. or min.

          n is the particle index of refraction

          $\rho_y(1)$ is given by the following table:

TABLE E-1

maxima y	y(1)	minima y	y(1)
1	4.08	1	7.62
2	10.79	2	14.0
3	17.16	3	20.32
4	23.47	4	26.63
5	29.78	5	32.93

α is obtained from the definition of ρ.

The equation for the values of $Q_y(n)$, the scattering efficiency at the yth maxima or minima, is given by:

$$Q_y(n) = a + bM \qquad\qquad (E-2)$$

where: $M = (n^2-1)/(n^2+2)$ $\qquad\qquad$ (E-3)

a and b are given by the following table:

TABLE E-2

y	at the maxima		at the minima	
	a	b	a	b
1	3.173	4.02	1.542	0.579
2	2.404	2.25	1.734	0.433
3	2.247	1.52	1.813	0.328
4	2.178	1.14	1.855	0.262
5	2.139	0.92	1.822	0.218

Kerker (1969) states that an accuracy of 3% or better can be obtained by drawing a smooth curve between the values of the extrema.

Using equation (E-1) and table (E-1), and equations (E-2) and (E-3) and table (E-2), the following table can be constructed for the three values of n corresponding to the three experimental aerosol materials:

TABLE E-3

Aerosol	n	y	Maximum		Minimum	
			α	Q	α	Q
KCl	1.49	1	4.31	4.23	7.93	1.71
		2	11.18	3.05	14.43	1.85
NH_4Cl	1.64	1	3.34	4.62	6.10	1.75
		2	8.58	3.21	11.67	1.89
NaCl	1.54	1	3.92	4.43	7.20	1.72
		2	10.13	3.10	13.53	1.86

Drawing smooth curves through these extrema results in Figure 35.