United States Environmental Protection Agency
Industrial Environmental Research Laboratory
Research Triangle Park NC 27711
EPA-600/7-78-130
July 1978

EPA

Evaluation of the PILLS IV

Interagency Energy/Environment R&D Program Report

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EPA-600/7-78-130
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Evaluation of the PILLS IV

by

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Contract No. 68-02-2131
T.D. 11007
Program Element No. EHE624

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Research Triangle Park, NC 27711

Prepared for

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Research and Development
Washington, DC 20460

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FIGURES (Continued)

ACKNOWLEDGMENTS

This work was carried out with the assistance in the laboratory of Douglas R. Sittason. Gratitude is also expressed for the helpful criticisms of Joseph D. McCain, Larry G. Felix, and Kenneth M. Cushing at Southern Research Institute. The theoretical evaluation of the PILLS IV could not have been carried out in its present form without the information supplied by Dr. G. Kreikebaum, formerly at Environmental Systems Corporation (ESC) in Knoxville, Tennessee, and Dr. H. W. Schmitt at ESC.

We also appreciate the guidance of our Project Officer, Mr. D. Bruce Harris.

SUMMARY

The operating characteristics of the PILLS IV in situ particle sizing instrument have been investigated theoretically and experimentally. The results of both types of work show large errors in this instrument's ability to size particles. Attempts to correlate the experimental findings with qualitative theoretical explanations have been successful. This investigation established a sensitivity to particle refractive index and detector response that seems to account for the observed characteristics of the instrument. Further measurements would be required to test this explanation quantitatively.

The prototype device, an extension of the PILLS (Particulate Instrumentation by Laser Light Scattering) technology to fine particles, was designed to measure particle size using the ratio of intensities of light scattered from a particle at two small angles (14° and 7°) with respect to an incident laser beam. The intensity ratio was chosen as the sizing parameter because of its relative independence of particle refractive index. However, the magnitude of the scattered intensity at 14° is also used for several important decisions in the electronic processing logic, which, for this particular optical system, render it especially sensitive to refractive index and detector variations for determinations of particle size distribution. Possible solutions to these problems with only minor hardware changes are offered.

SECTION I

INTRODUCTION

The PILLS* IV is a prototype instrument designed to provide on-line, in situ determinations of particulate size distributions in process gas streams. This study of the PILLS IV was undertaken to investigate discrepancies in a set of previous measurements[1] in which this device was simultaneously tested with a cascade impactor. In that work the size distribution of resuspended fly ash in a wind tunnel was obtained with both devices. The results of those tests are illustrated in Figure 1 where the number frequency per cm^3 is plotted versus particle diameter (D) on semilog scales. The number frequency, $\Delta N(D)/\Delta D$, is the number per cm^3, per diameter increment. The study reported here was performed to determine if modifications to the PILLS IV to eliminate such discrepancies were feasible.

An evaluation program was carried out in which the PILLS IV response was investigated experimentally and theoretically. In the experimental work, monodisperse aerosols of PSL spheres were sampled and checks of the instrument hardware were performed, thus eliminating several equipment related malfunctions as the source of the observed discrepancies. In the theoretical work mathematical expressions were developed for the counting rate of each channel as a function of the aerosol light scattering properties. Upon analysis of these expressions, a basic design problem involving a counting criterion was found to be capable of producing the large errors indicated in Figure 1. In fact, the PILLS IV data in that figure can be explained quantitatively. Recommendations that could provide acceptable measurements of size distributions of polydisperse aerosols are given at the end of this report.

In Section II, background information related to our theoretical study is presented. Then in Section III the relevant components and operation of the instrument are described. Section IV summarizes the experimental work performed in this study. Section V gives a theoretical description of the instrument's response; Section VI gives a comparison of this theoretical response to the observed response; and Section VII gives our recommendations to anyone performing measurements with the PILLS IV.

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*Particulate Instrumentation by Laser Light Scattering developed by Environmental Systems Corporation, 1212 Pierce Parkway, Knoxville, TN 37921.

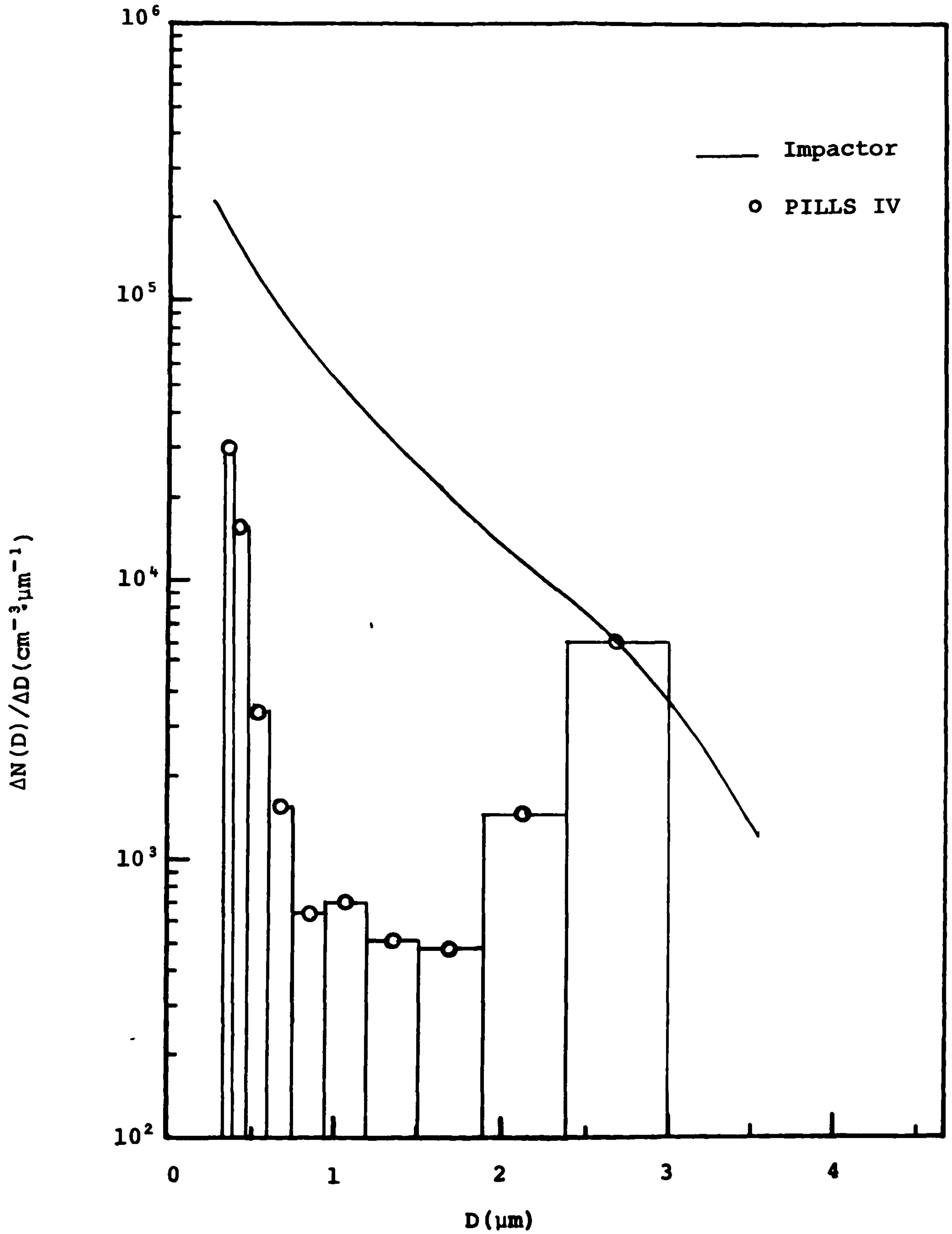

Figure 1. The Behavior of the PILLS IV Compared to an Inertial Cascade Impactor When Sampling Fly Ash. (After Gooding.[1])

SECTION II

BACKGROUND

Optical techniques provide the potential for particle sizing systems where representative data can be obtained in real time with no perturbation of the aerosol. The PILLS IV is one such instrument. However, interpretation of optical data to obtain size is not only difficult but sometimes restricted by ambiguities, as illustrated in this section.

When a beam of light is incident upon a particle, part of the light is scattered and part is absorbed. The nature of the scattered light is dependent upon the particle size and other parameters, including chemical composition, which is characterized in optical work through the refractive index m (a complex number expressed by $m = n - in'$). Figure 2 illustrates this relationship. The average, scattered intensity at angles of 20-40° and 75-100° from the beam direction is plotted as a function of particle size for various refractive indices. The imaginary part of the refractive index, n', determines the amount of absorption by the particle, which reduces the overall level of scattering. Of course such sensitivity to particle composition is undesirable for particle size measurements, so methods have been sought to minimize that effect.

At angles close to the incident beam the scattered wave is determined mainly by the diffracted wave, which passes outside the particle, and the wave emanating from the particle itself. For a large, homogeneous particle this latter wave is influenced by diffraction also, to some extent, but unless $|m-1| \ll 1$, refraction at the particle-medium boundary is the dominant factor. If absorption occurs, the refracted wave passing through the particle is attenuated. The combined wave observed in the far field, the result of adding the wave passing around the particle to that passing through it, is thus determined mainly by diffraction at small angles, and refraction at larger angles. The above discussion leads to the well known characteristic that scattering at small angles (within the forward lobe) is less sensitive to refractive index that at larger angles, as illustrated in Figure 3, where scattered intensity for spheres is plotted versus the scattering angle θ and two very different refractive indices.

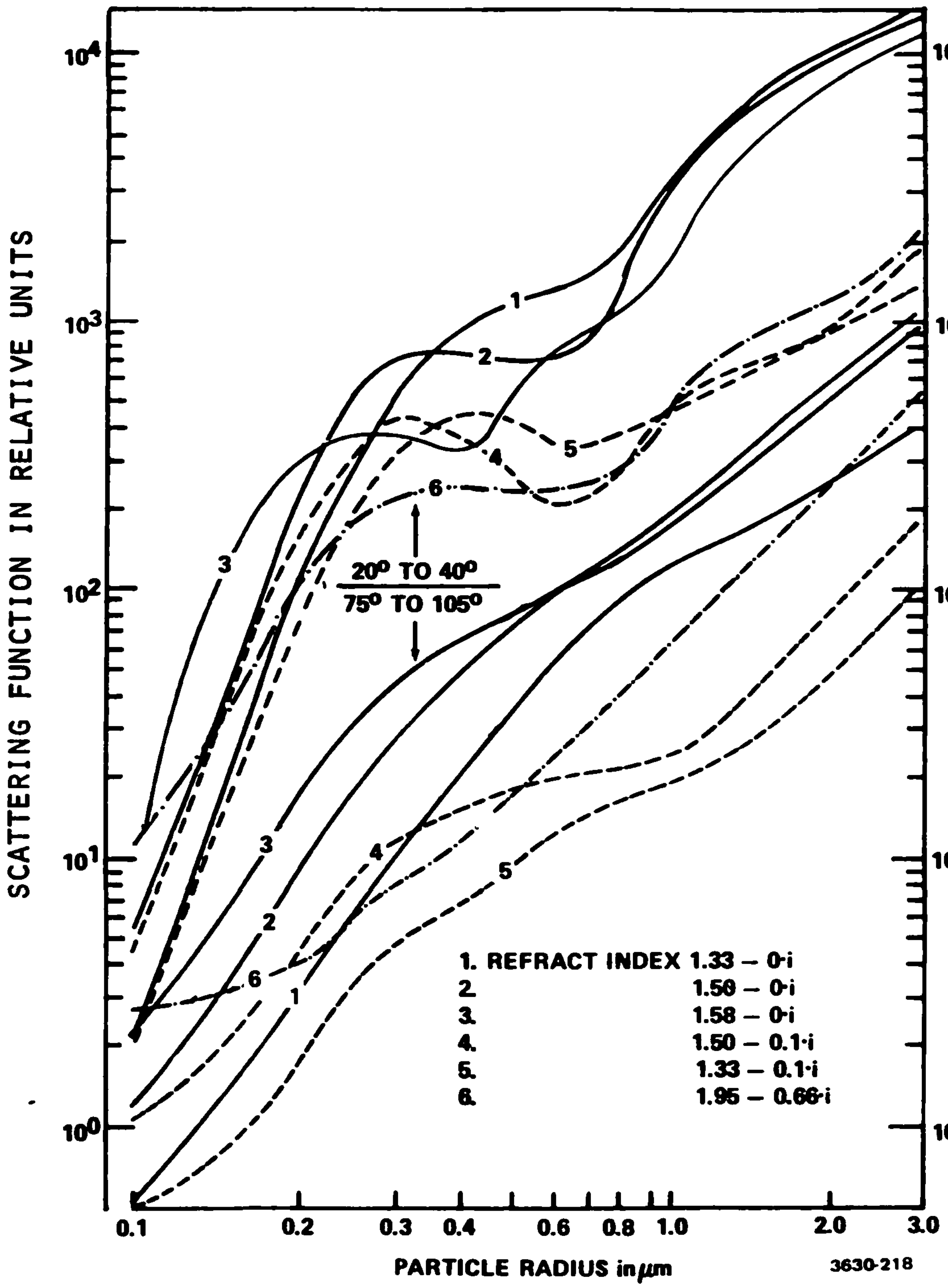

Figure 2. Mean Values of the Scattering Functions of Single Particles vs. Particle Size. The Scattering Angles are 20° to 40° and 75° to 105° (after Quenzel[2]).

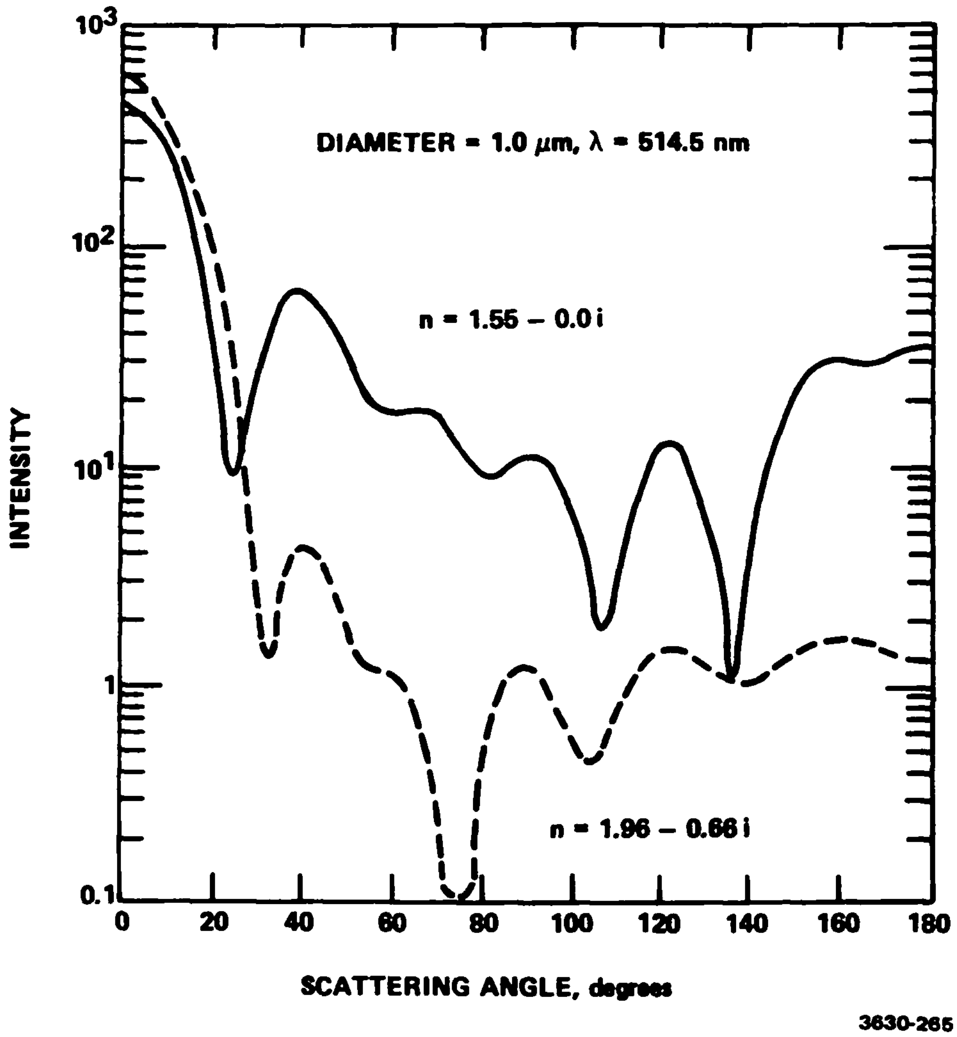

Figure 3. Scattered Intensity as a Function of Scattering Angle in the Plane of Polarization. m = index of refraction. λ = wavelength of incident radiation. (After Gravatt.[3])

Hodkinson[4] was the first to point out that although refractive index does have some effect on the magnitude of scattering in the forward lobe, the shape of the lobe does not change. Thus the ratio of scattered intensities at two angles within the forward lobe, while sensitive to particle size, is practically insensitive to refractive index. Figure 4 illustrates this point in a plot of the ratio versus the dimensionless size parameter $\alpha(=\pi D/\lambda)$ for two refractive indices.

Gravatt,[3] Shofner et al,[5] and Chan[6] have developed prototype systems for particle sizing that are based on the intensity ratio concept of Hodkinson. Figure 5 is a schematic of Shofner's system, the PILLS IV. The intensities of the scattered light pulses at the angles θ_1 and θ_2 are normalized to the reference pulse at $\theta = 0°$ for synchronization and to account for fluctuations in intensity of the laser source. The optics and sensors are kept clean and cool by the use of a purge air system.

The laser used in the PILLS IV is a semiconductor junction diode ($\lambda = 0.9$ μm). The useful size range for particle sizing is from 0.2 to 3.0 μm. Shofner states that the view volume of his system is approximately 2×10^{-7} cm^3. The upper concentration limit for single particle counters is determined by the requirement that the probability of more than one particle appearing in the view volume at a given time be much less than unity. For Shofner's system this requirement and the dimensions of the view volume would set the concentration limit at approximately 10^6 particles/cm^3, a value much higher than for conventional single particle counters.

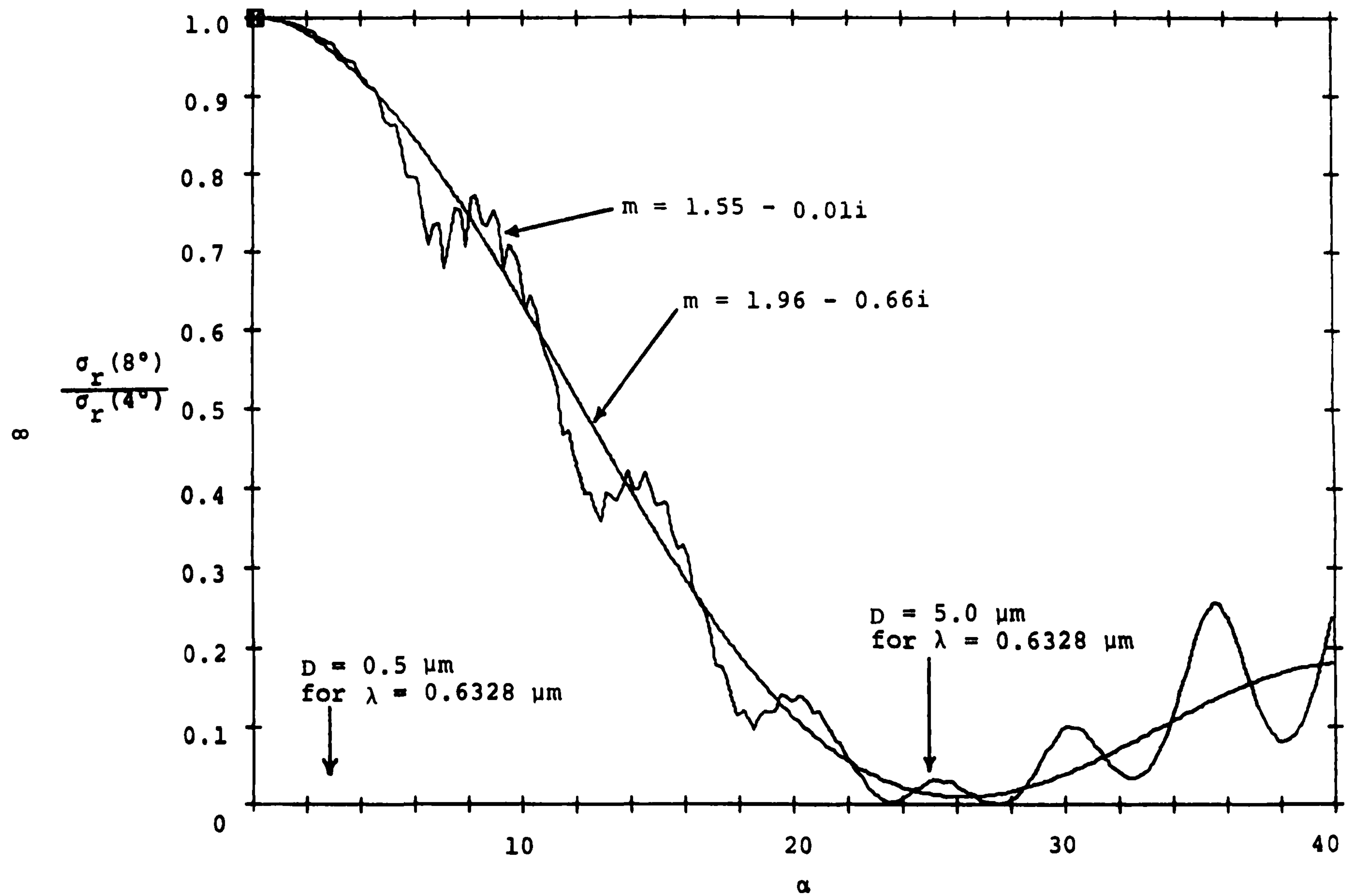

Figure 4. Ratio of Scattering Cross Sections for the Refractive Indices of Figure 3.

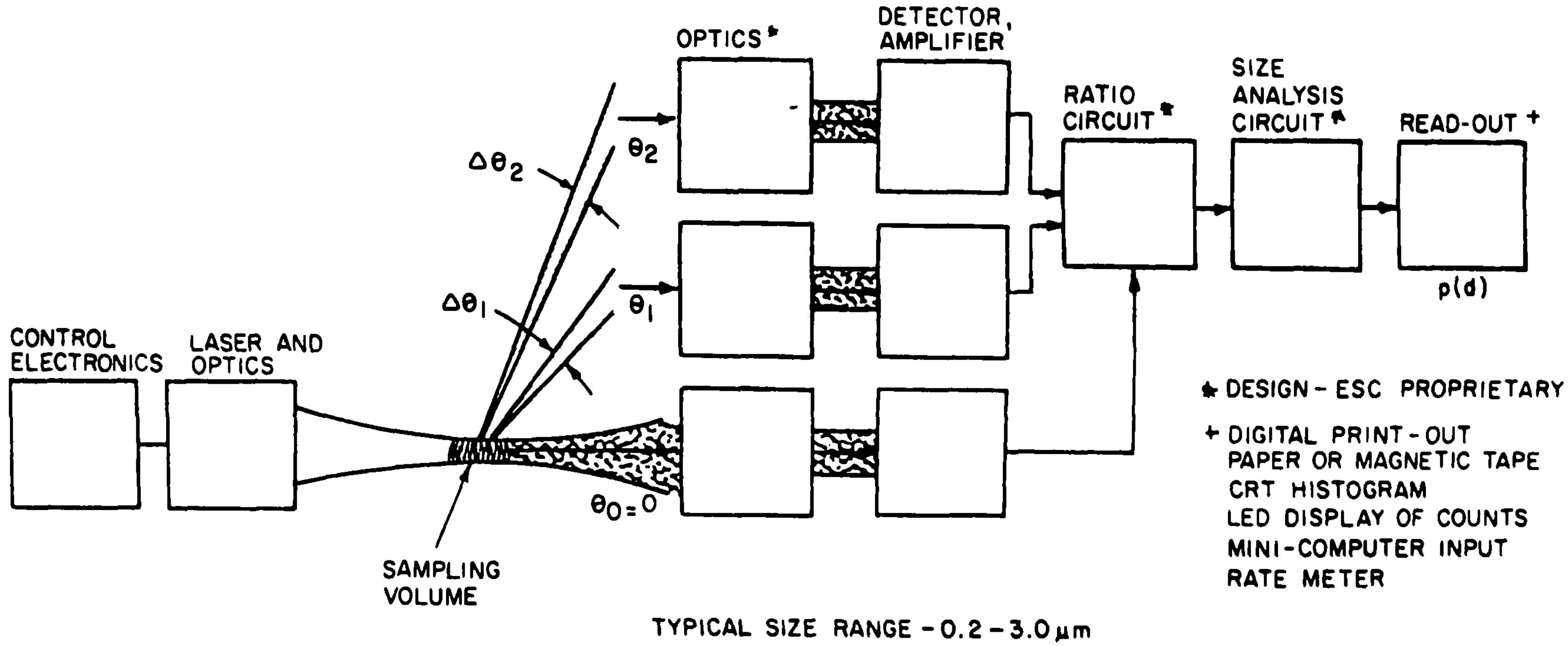

Figure 5. The PILLS IV Optical Particle Counter. (After Shofner et al.[5])

SECTION III

DESCRIPTION OF THE PILLS IV

A. Basic Optical Components

The light source of the PILLS IV is a GaAs laser diode with a lens system that focuses the beam to a very small cross-section on the order of tens of microns in diameter. Such a beam is normally described as a Gaussian beam because the intensity profile perpendicular to the beam direction is given by

$$I = I_0 e^{-2(r/B_0)^2} \quad , \qquad (1)$$

where r is the distance of the point of interest from the beam axis and B_0 is a measure of the beam width at its focus.

There are three photo detectors at angles of 14°, 7°, and 0° from the incident beam. The detector at 0° is used to monitor the undeviated beam to establish a reference for the 14° and 7° detector signals. The collection optics of the 14° and 7° detectors are of the annular design so that the detector fields of view illustrated in Figure 6 form hollow cones of thickness 2b with the beam passing through the common apices of the cones and along their common axis. It is important to note that although the beam is drawn with a finite width of $2B_0$, significant levels of radiation (determined by Equation 1) propagate outside of this column. The width of the beam as determined from measurements by the manufacturer is 46 μm for $2B_0$ ($I/I_0 = 0.5$ for $r =$ 13.5 μm) and the width of the detector fields of view are approximately 200 μm (for 2b).

B. Counting Scheme

As the laser is pulsed (10^3 times/sec), the 14° and 7° detectors respond with signals i_{14} and i_7 to the scattered light. A baseline for i_{14} (or i_7) is established by background subtraction in which the average of i_{14} (or i_7) for the two pulses, before and after the pulse of interest, is subtracted from i_{14} (or i_7). The background subtraction technique is based on the assumption that if a particle of interest was in the sizing volume daring a pulse of the beam, then no particle(s) of significant

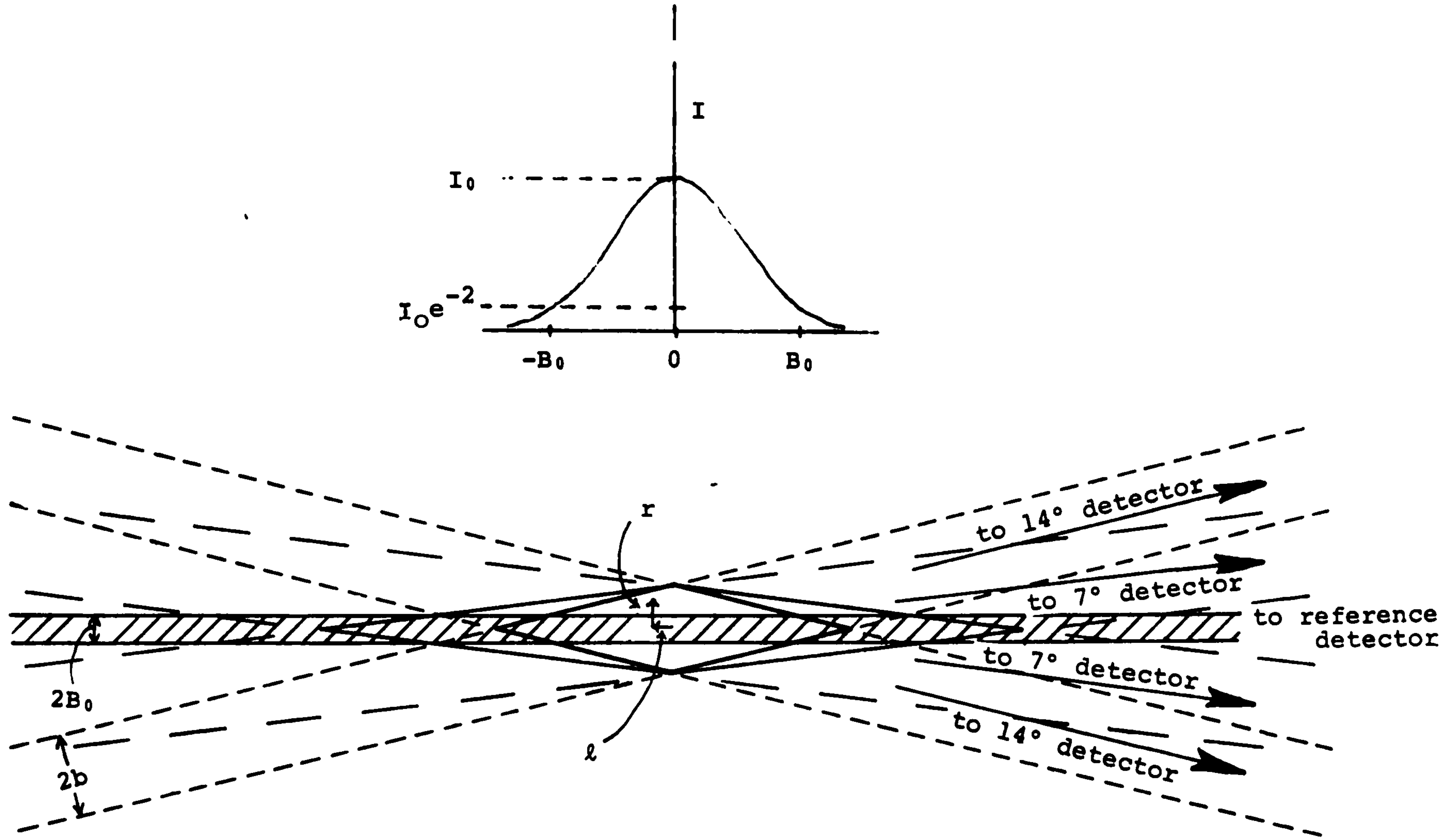

Figure 6. A Cross Section of the Idealized PILLS IV View Volume. The beam has a Gaussian intensity profile (i.e., $I = I_0 e^{-2(r/B_0)^2}$).

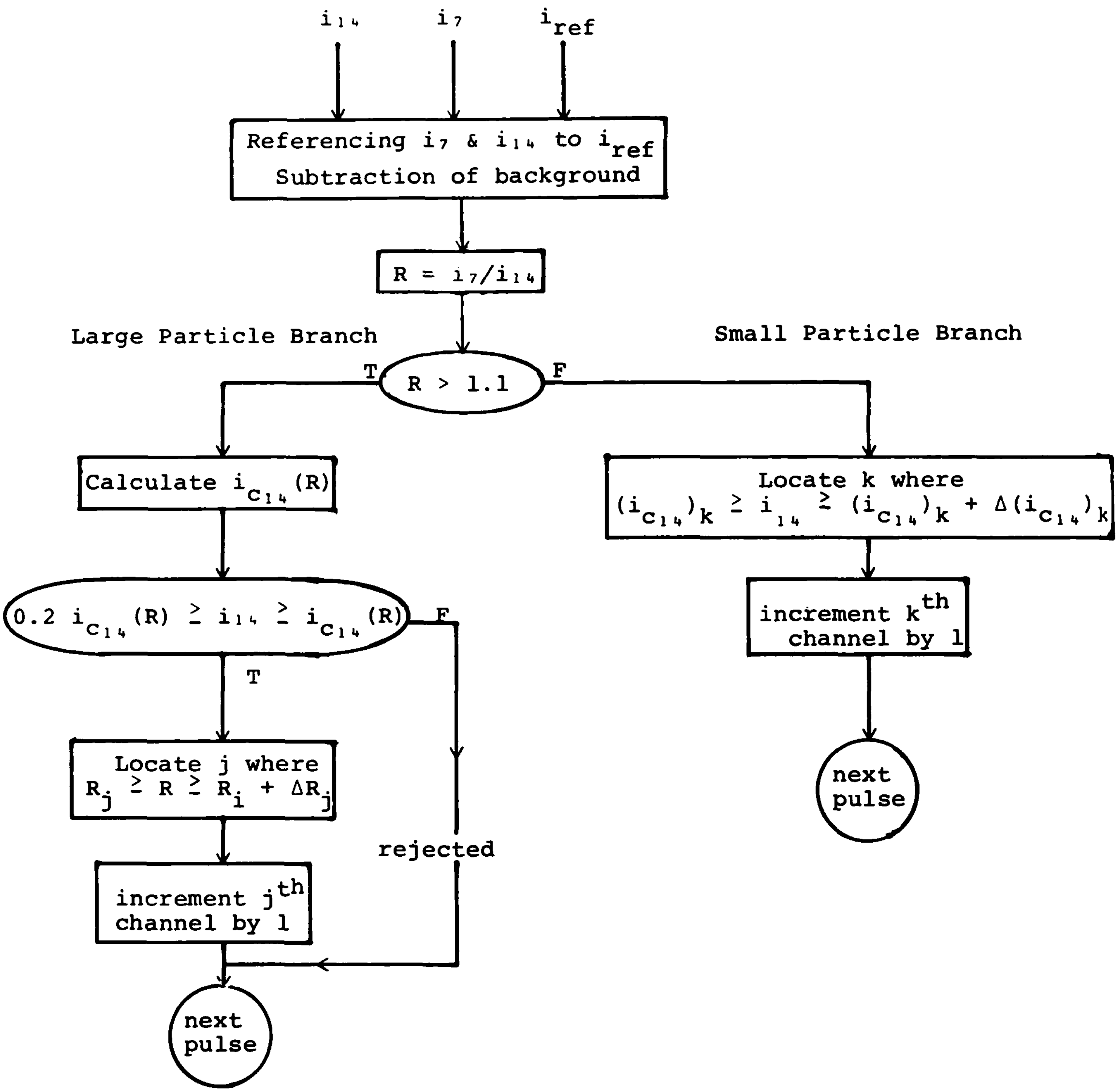

Figure 7. Counting Logic of PILLS IV.

size were in the sizing volume on either the preceding or following pulses. That assumption is valid when the aerosol concentration is below the limit discussed at the end of Section II. The counting logic after background subtraction and referencing of i_{14} and i_7 to the beam monitoring detector is depicted in Figure 7. If the ratio i_7/i_{14}, denoted by R, is greater than 1.1, then a particle size is inferred from its value. If the ratio is less than 1.1, the parameter i_{14} is used to determine particle size. Note that the PILLS IV uses the inverse of the parameter plotted in Figure 4.

A further criterion is invoked for the instrument to count a particle whose detector scattering ratio is above 1.1. The measured signal i_{14} must be in the range

$$0.2 i_{C14}(R) \leq i_{14} \leq i_{C14}(R) \qquad (2)$$

where $i_{C14}(R)$ is a predetermined function of R. The purpose of this criterion is two-fold. It attempts to provide a definition of the instrument sizing volume by eliminating counts when particles are near the edge of or outside of the 14° detector view volume but well within the 7° detector view. It also prevents the sizing of small particles as larger ones due to large errors in R produced by system noise. If i_{14} produced by a particle with ratio i_7/i_{14} satisfies the criterion then the instrument adds one increment in the size channel corresponding to the observed intensity ratio. This criterion is not applied to signals for which R is less than 1.1.

The output of the PILLS IV is the number of particles per channel in two series of channels, one series corresponding to values of R and one series to values of i_{14}. The upper and lower limits of each channel can be varied by the operator. When R > 1.1 the channel number to be incremented is determined by the value of R; that is,

$$R_j \leq R \leq R_j + \Delta R_j \qquad (3)$$

where j denotes the channel to be incremented. When R < 1.1 the channel number to be incremented is determined by the value of i_{14}; that is,

$$(ic_{14})_k \leq i_{14} \leq (ic_{14})_k + \Delta(ic_{14})_k \qquad (4)$$

where k denotes the channel to be incremented. The operator then relates the channels, j or k, to calibration tables for particle sizes.

SECTION IV

EXPERIMENTAL INVESTIGATION OF THE PILLS IV RESPONSE FUNCTION

An aerosol generation system was developed to evaluate the PILLS IV empirically. The system, depicted in Figure 8, consists of a nebulizer* containing a suspension of uniform latex spheres,[†] a drying chamber, rectangular ducts to and from the PILLS IV probe, and an optical particle counter** with diluter. The rectangular ducts were constructed with the same shape and size as the opening in the probe which contained the sensing region. The entire aerosol stream flowed downward to eliminate settling losses. The aerosol stream exiting the PILLS IV probe was sampled isokinetically by a particle counter or filter for independent determination of particle concentration.

Sampling experiments were performed with the PILLS IV purge air lines stopped and an aerosol stream velocity of 25 cm/sec. Velocity measurements made in the duct with a thermal-anemometer[††] demonstrated a flat profile across the duct within ±10%. The nebulizer flowrate of 10 LPM introduced 0.61 cm of the latex sphere water suspension per minute into a total air flow of 50 l/min. This gave a residence time of 8 seconds before particles passed through the PILLS IV. For each size of PSL spheres, aerosols of two concentrations were produced. One was low in particle concentration for calibrating the dilutor, and one high in concentration to evaluate the PILLS IV.

Figures 9-14 give aerosol size distributions in terms of ΔN/ΔD as derived from the PILLS IV and the Climet Instrument in

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* Retec X70/N; Civitron Burton Division, Retec Development Laboratory, Van Nuys, CA 91406.

† Polystyrene (0.357 μm, 760 μm, or 1.099 μm dia.), Polyvinyltoluene (2.02 μm dia.), or Styrene/Vinyltoluene (2.77 μm dia.); Dow Diagnostics, P.O. Box 68511, Indianapolis, IN 46268.

** Climet Instruments 0208A Particle Analyzer; Climet Instruments Co., Redlands, CA 92373

†† Sierra Instruments Model 440/441; Sierra Instruments, Inc., P. O. Box 909, Carmel, CA 93924.

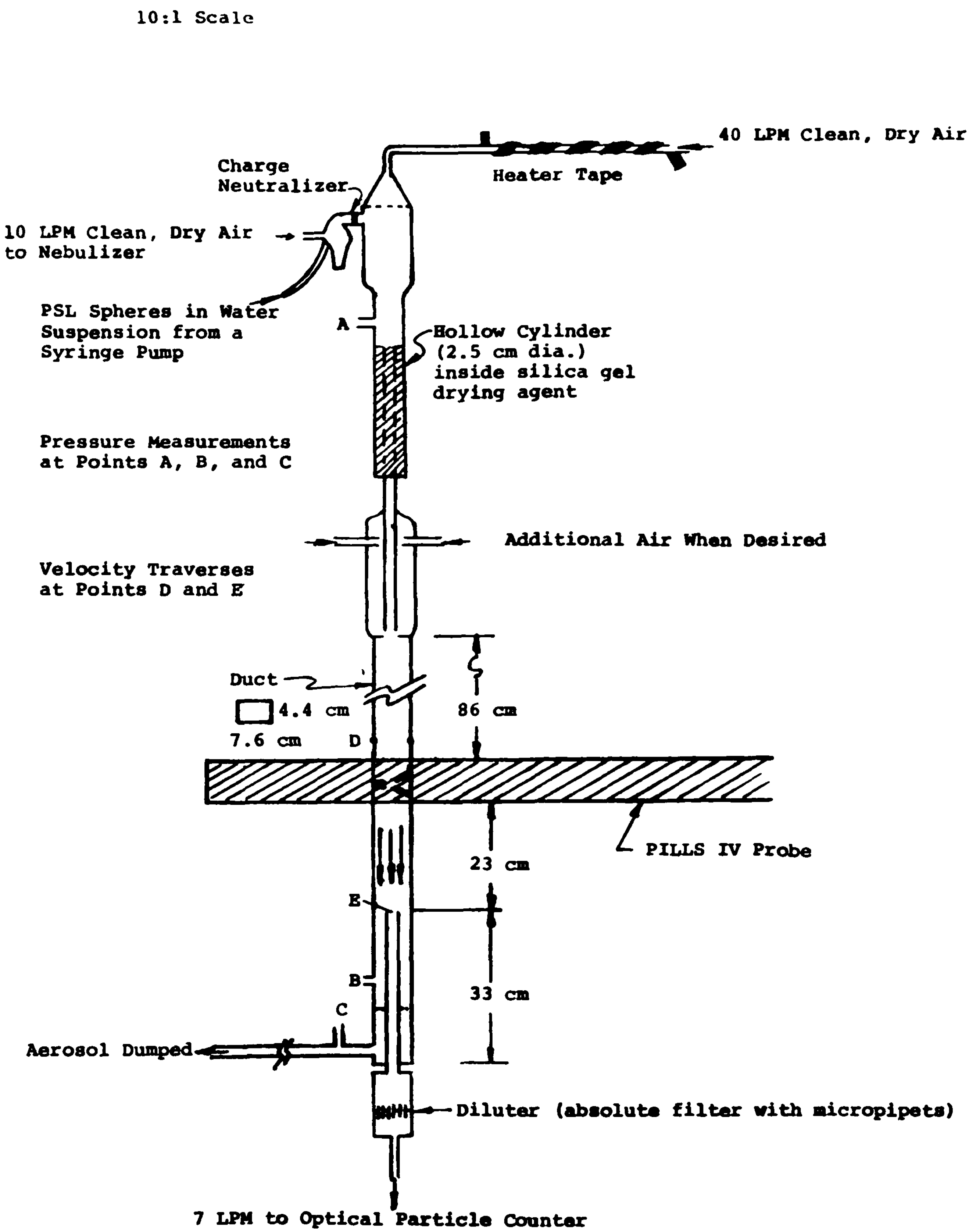

Figure 8. Aerosol Generation and Sampling System for PILLS IV Evaluation. Note: The Diameter of the Sampling Tube at Point E is Varied According to Desired Velocity to Preserve Isokinetic Sampling.

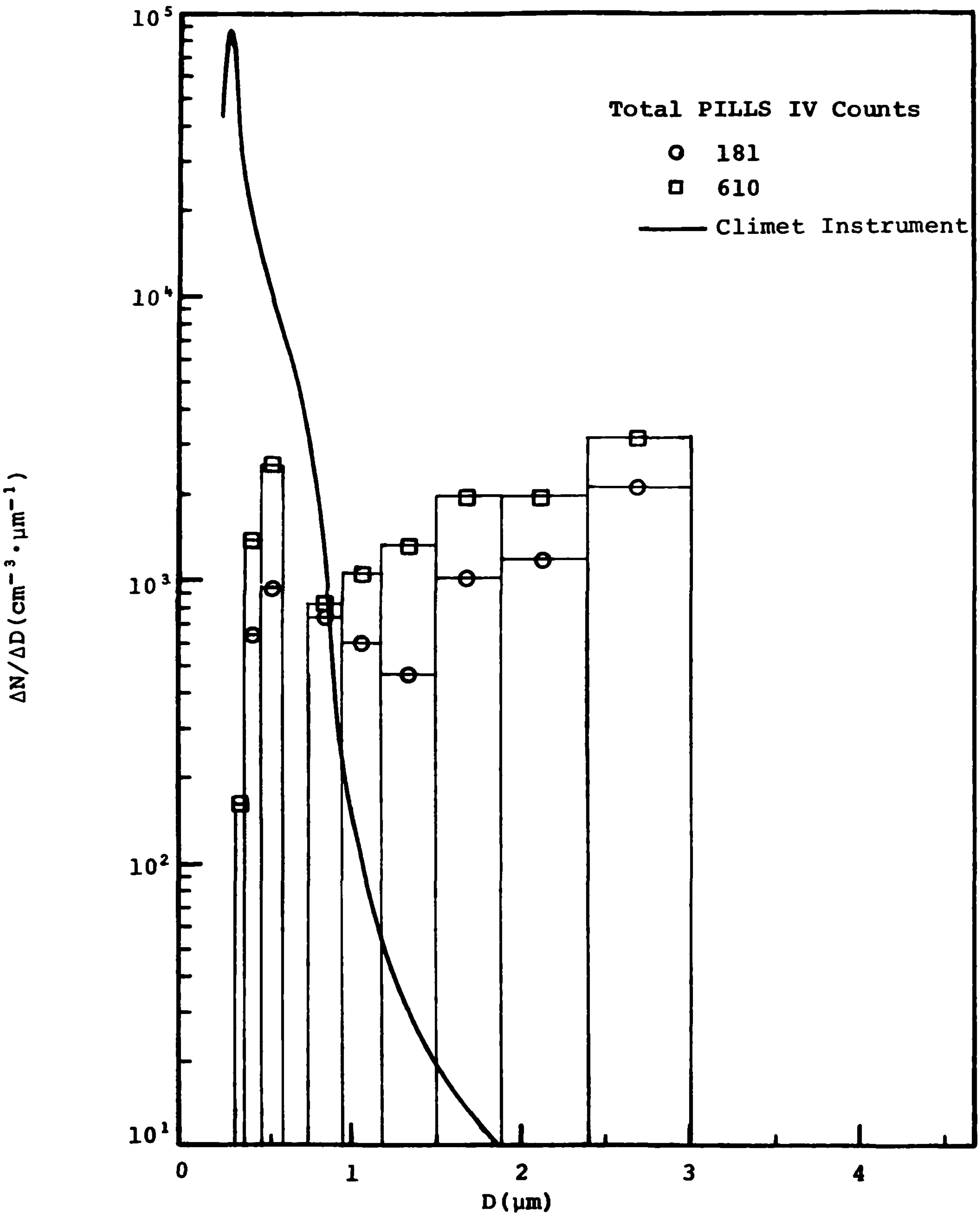

Figure 9. Comparison of PILLS IV and Climet Responses to 0.357 μm Latex Particles (back to back runs with 2.0 x 10^9 particles/cm^3 in nebulizer solution).

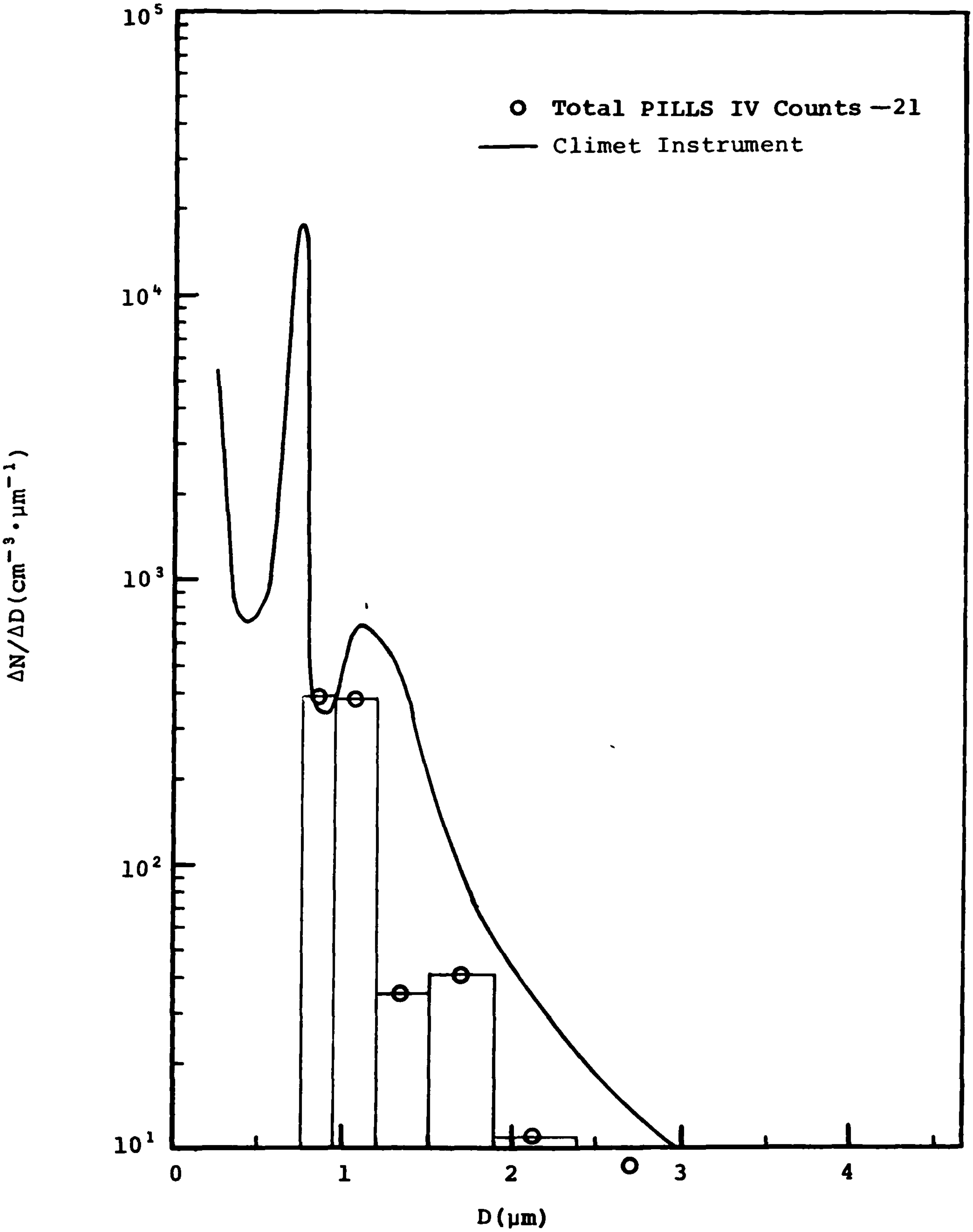

Figure 10. Comparison of PILLS IV and Climet Responses to 0.760 μm Latex Particles. (2.7×10^9 particles/cm^3 in nebulizer solution).

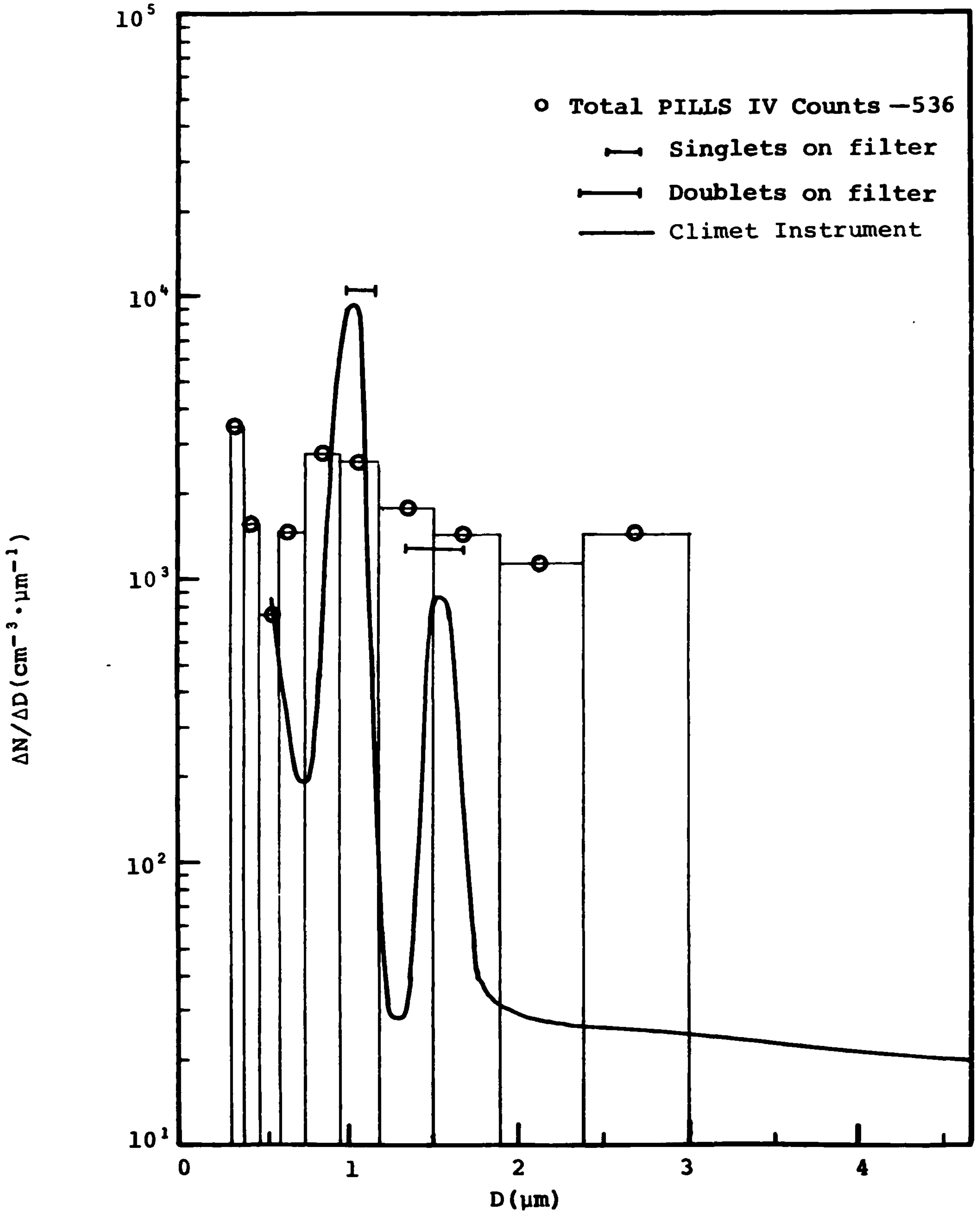

Figure 11. Comparison of PILLS IV and Climet Responses to 1.099 μm 1.099 μm Latex Particles. (1.12 x 10^9 particles/cm^3 in nebulizer solution.)

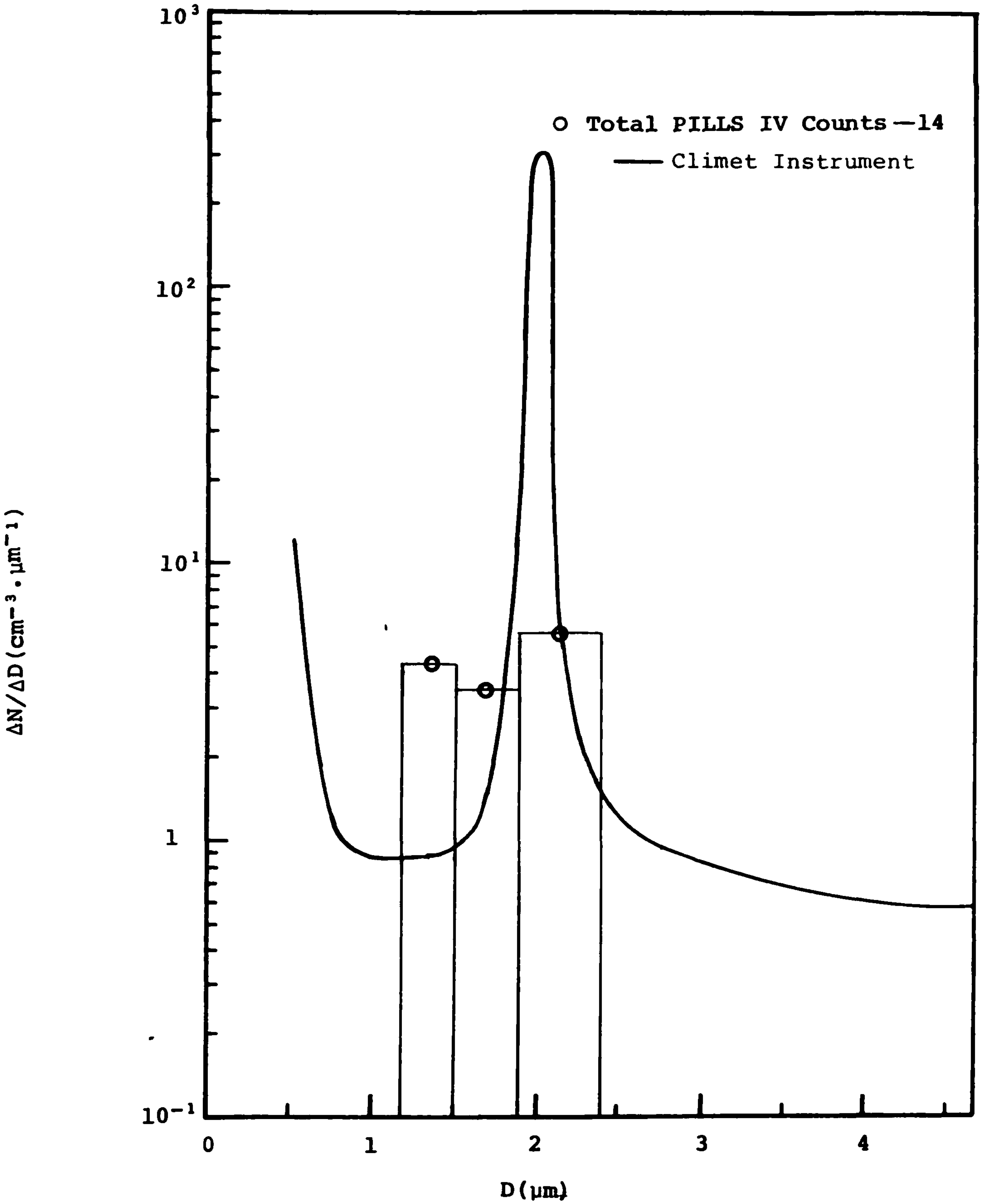

Figure 12. Comparison of PILLS IV and Climet Responses to 2.02 μm Latex Particles at Low Concentration. (1.43×10^7 particles/cm^3 in nebulizer solution.)

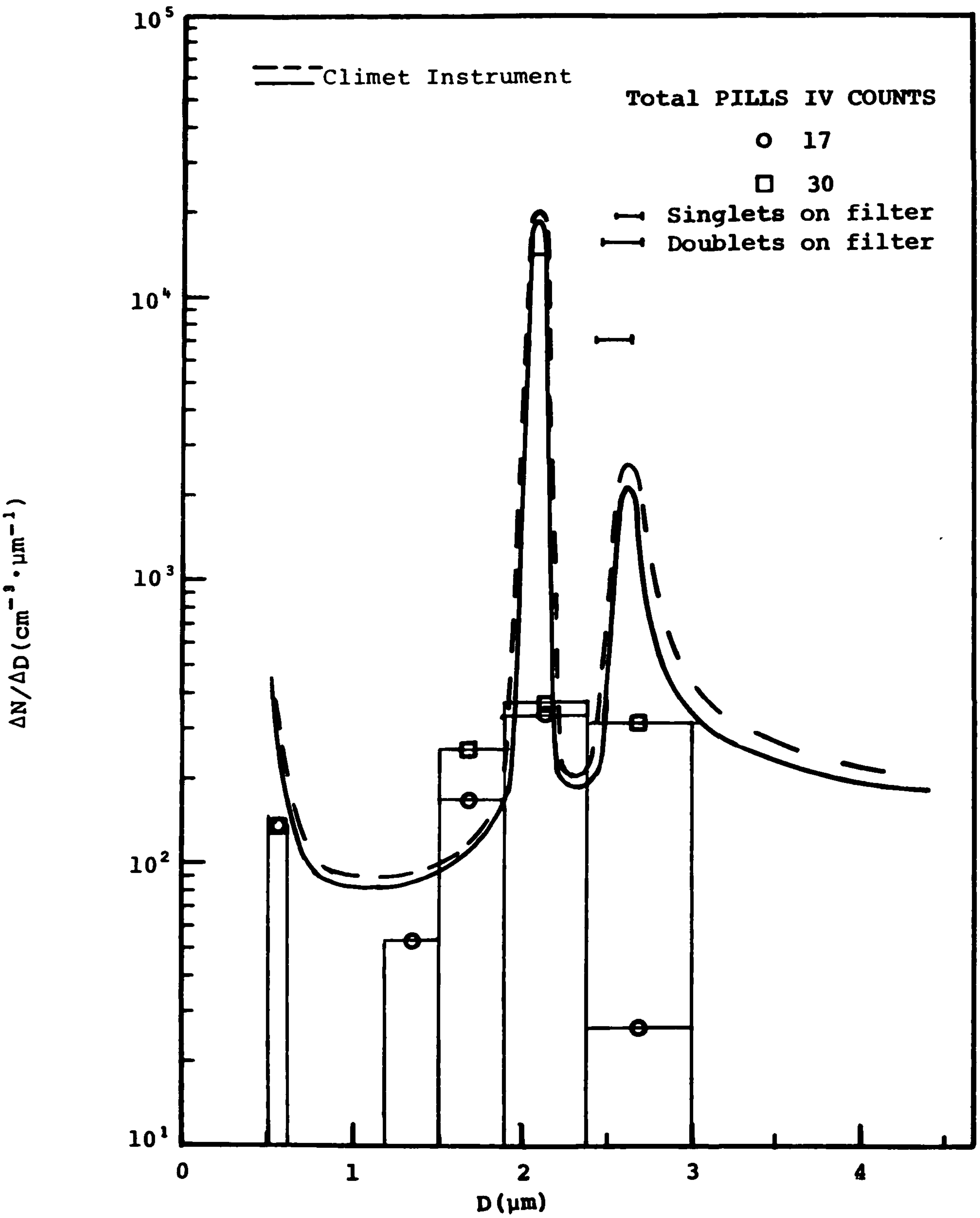

Figure 13. Comparison of PILLS IV and Climet Responses to 2.02 µm Latex Particles. (back to back runs with 1.43×10^9 particles/cm^3 in nebulizer solution.

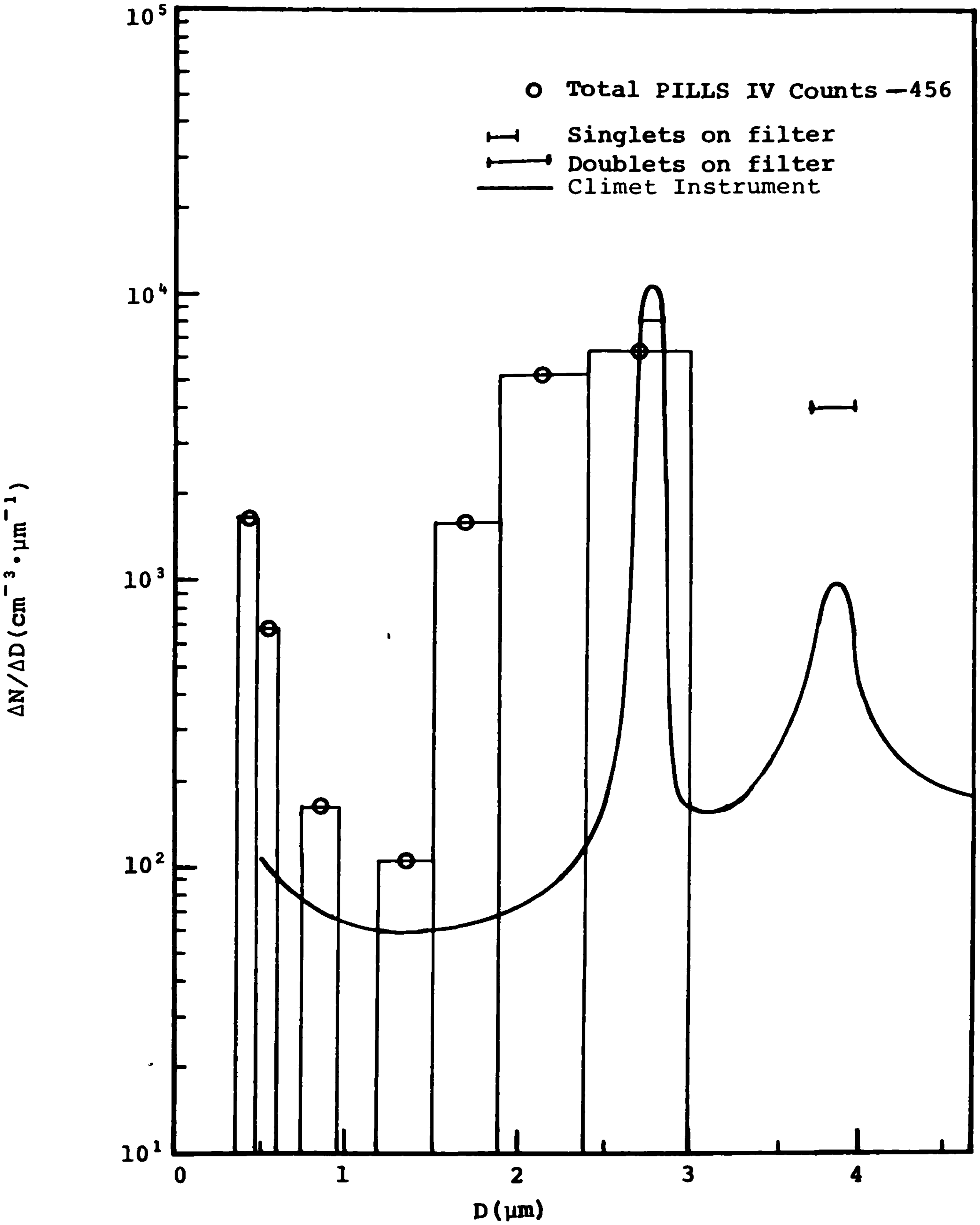

Figure 14. Comparison of PILLS IV and Climet Responses to 2.77 μm Latex Particles.
(1.11×10^9 particles/cm^3 in nebulizer solution.)

conjunction with a multi-channel analyzer. In some cases, the concentration of PSL spheres was also determined from filter catches. The PILLS IV sizing volume was assumed to be that given by the manufacturer, 2×10^{-7} cm^3.

The general features of the particle size distributions obtained with the Climet Instrument are as one might expect from the aerosol generator. There is a high concentration of particles counted around the diameter of the nebulized latex particles. The presence of doublets is usually observable as distinct peaks of considerably lower concentration. There are also substantial concentrations of particles apparently produced by dissolved wetting agents in the stock solution in which the latex particles must be stored.[7] When a droplet from the nebulizer is dried the wetting agent forms a particle of unknown structure with light scattering properties which makes most of them appear to be below 0.3 μm as sized by the Climet instrument. This explanation also agrees with measurements in our laboratory where a stock solution from which the latex particles had been filtered was nebulized and dried. The number of "small" particles seen by the Climet was unaffected. Further, it was found that dilution of the stock solution with water reduced the number of small particles detected by the Climet instrument.

The data in Figures 9-14 show two major characteristics of the PILLS IV relative to the Climet Instrument. Its responses to PSL spheres with diameters of 2.77 μm (Figure 13) and 1.099 μm (Figure 11) are comparable to that of the Climet Instrument, while its responses to the other PSL spheres, 2.02 μm (Figures 12 and 13), 0.760 μm (Figure 10), and 0.357 μm (Figure 9) are one to two orders of magnitude low. Such responses to latex particles are qualitatively explained by the theoretical response function given in the next section. Sizing of the three largest size particles is performed by the ratio i_7/i_{14} with the counting criterion invoked on i_{14}. The PILLS IV response to the two smaller sizes of latex particles depends on i_{14} only. The other major characteristic of the data given in Figures 9-14 is that relative to the Climet Instrument, the PILLS IV frequently indicates a large concentration of particles of sizes other than that of the latex particles under study. The origin of these counts is not known, although a possible source is discussed in Section VI.

SECTION V

THEORETICAL INVESTIGATION

The results of the experimental investigations revealed large errors in sizing particles with the PILLS IV. However, it was not determined whether the discrepancy is due to a basic design problem, an error in calibration, or a subtle malfunctioning of a component. In order to determine if the design or logic of the PILLS IV prototype was faulty, a theoretical analysis of the system has been performed.

Equations (5) represent the total signal from the three instrument detectors on the n^{th} pulse of the beam:

$$i_{14}(n) = i_{e_{14}}(n) + i_{m_{14}}(n) + C_{14}\sigma_{14}f_{14}(r,\ell)e^{-2(r/B_0)^2}I_0e^{-T}$$

$$i_7(n) = i_{e_7}(n) + i'_{m_7}(n) + C_7\sigma_7f_7(r,\ell)e^{-2(r/B_0)^2}I_0e^{-T} \qquad (5)$$

$$i_0(n) = i_{e_0} + C_0I_0e^{-T}$$

Of all the parameters given, only σ_{14} and σ_7, the differential scattering cross sections at 14° and 7°, are directly related to the particle of interest. The other parameters are defined in Table 1. Through background subtraction and referencing of i_{14} and i_7 to i_0, the processed signals, $i_{p_{14}}$ and i_{p_7}, can be obtained with errors denoted by $\Delta i_{p_{14}}$ and Δi_{p_7}:

$$\text{where } i_{p_{14}} = \frac{C_{14}}{C_0}\sigma_{14}f_{14}(r,\ell)e^{-2(r/B_0)^2} \pm \Delta i_{p_{14}} \qquad (6)$$

$$\text{and } i_{p_7} = \frac{C_7}{C_0}\sigma_7f_7(r,\ell)e^{-2(r/B_0)^2} \pm \Delta i_{p_7} \quad .$$

Optimum performance is obtained from the instrument when $\Delta i_{p_{14}} = \Delta i_{p_7} = 0$, and accurate values of $f_7(r,\ell)/f_{14}(r,\ell)$, C_0, C_7, and C_{14} have been determined by calibration measurements. As described in Section III, the ratio of signals $i_{p_7}/i_{p_{14}}$ and $i_{p_{14}}$ are used to deduce a particle size and to decide if the particle should be counted. In this analysis, the counting

TABLE 1.

DEFINITIONS

$i_{e_{14}}$, i_{e_7}, and i_{e_0}	- The portions of the signals due to electronic background noise.
$i_{m_{14}}$ and i_{m_7}	- The portions due to aerosol particles other than the one of interest on the n^{th} pulse.
i_{p_7} and $i_{p_{14}}$	- The signals from the 7° and 14° detectors after background subtraction and referencing to the 0° detector signal.
C_{14}, C_7, and C_0	- System conversion factors that relate light intensity at the detectors to the signals which are processed for size evaluation.
$I_0 e^{-T}$	- Intensity of the beam after transmission through the aerosol.
$e^{-2(r/B_0)^2}$	- Variation of beam intensity with position in the aerosol. (r is depicted in Figure 6).
$f_7(r,\ell)$ and $f_{14}(r,\ell)$	- Optical transfer functions which relate actual scattered intensity to that reaching the detector (r and ℓ are depicted in Figure 6). The functions are expected to decrease monotonically as r and ℓ increase.
σ_7 and σ_{14}	- The differential scattering cross section of a particle at 7° and 14° with respect to the incident beam. The units are those of radiant sterance, radiant flux per unit area per unit solid angle.
$\sigma_{c_{14}}$	- A reference function programmed into the PILLS IV, aside from an adjustment for the detector response, to establish a counting criterion which the 14° detector signal must satisfy.

scheme is first analyzed assuming the optimum conditions, and then the changes in behavior, which would result if those assumptions are violated are discussed. Also, it is assumed that the programmed function $i_{c_{14}}(R)$ was established for particles at the center of the view volume where $i_{p_{14}} = (C_{14}/C_0)\sigma_{14}$ and $i_{p_7} = (C_7/C_0)\sigma_7$.

Ideally, the ratio R $(= i_{p_7}/i_{p_{14}})$ is determined by σ_7/σ_{14}. When this ratio is above 1.1 then the particle is counted if $i_{p_{14}}$ is in the range $0.2 i_{c_{14}}(R) \leq i_{p_{14}} \leq i_{c_{14}}(R)$. However, since $i_{p_{14}}$ is a function of r and ℓ, the number of particles having a given value of R which are counted after many pulses depends upon the position of each in the beam as well as the value of σ_{14}. In this manner a sizing volume of the instrument is established. On the other hand, if $\sigma_{14}(R)$ varies due to different composition or shape from that used in calibration to establish $i_{c_{14}}(R)$, then the sizing volume varies, decreasing with $\sigma_{14}(R)$ for a given R. More specifically, the volume in which particles are counted is determined by values of r and ℓ for which

$$0.2 \leq \frac{\sigma_{14}}{\sigma_{c_{14}}(R)} f_{14}(r,\ell) e^{-2(r/B_0)^2} \leq 1 \quad . \tag{7}$$

Particles for which R < 1.1 produce results whose analysis is somewhat more complicated. The count in channel k is incremented when the criterian:

$$1 \leq \frac{\sigma_{14}}{(\sigma_{c_{14}})_k} f_{14}(r,\ell) e^{-2(r/B_0)^2} \leq 1 + \frac{\Delta(\sigma_{c_{14}})_k}{(\sigma_{c_{14}})_k} \tag{8}$$

is satisfied where $(\sigma_{c_{14}})_k$ and $(\sigma_{c_{14}})_k + \Delta(\sigma_{c_{14}})_k$ correspond to the lower and upper limits of the k^{th} channel. Thus we see that in principal the channel in which a particle is counted depends upon r and ℓ as well as the value of σ_{14}, which is determined by the particle's properties. For example, a particle with a high σ_{14}, positioned near the edge of the instrument view volume where $f_{14}(r,\ell)e^{-2(r/B_0)^2}$ is small, could not be discriminated from a particle with a low σ_{14}, positioned at the center of the view volume where $f_{14}(r,\ell)e^{-2)r/B_0)^2}$ is maximum. Of course, detailed descriptions of $f_{14}(r,\ell)$ and $\sigma_{14}(D)$ are necessary to evaluate the degree to which this property of the counting scheme effects the results.

We see from Equations 7 and 8 that the two functions $f_{14}(r,\ell)$ and σ_{14}/σ_{C14} must be known or otherwise accounted for to reduce PILLS IV data in terms of the aerosol size distribution and concentration. The most important question is whether for a given particle size σ_{14}/σ_{C14}, which is determined by the composition of the particles themselves, varies enough to cause significant changes in the response of the PILLS IV.

A. Variation of σ_{14}/σ_{C14} with Refractive Index

The exact function σ_{C14} programmed into the PILLS IV was not available. However, Dr. G. Kreikebaum, one of the developers, kindly provided a description of the procedure by which it was obtained. That procedure was reproduced in this study as follows:

σ_{C14} for Particles with $\sigma_7/\sigma_{14} > 1.1$

The differential scattering cross sections σ_{14} and σ_7 proportional to the respective scattered intensities, were calculated using Mie theory for a broad range of refractive indices* which one might expect the PILLS IV to encounter and for the diameters 0.3 µm - 4.0 µm.† From these, the function σ_{14} versus R was obtained for each refractive index. Then for each R-value, σ_{C14} (R) was set equal to the maximum value of σ_{14} versus refractive index for that value of R. Values of R > 10 were excluded from this analysis since a unique size cannot be associated with those R values.[4]

σ_{C14} for Particles with $\sigma_7/\sigma_{14} < 1.1$

The calibration curve $\sigma_{C14}(R)$ for R < 1.1 was set equal to σ_{14} for a particle refractive index of 1.59 - 0.0i.

The resulting function $\sigma_{C14}(R)$ is given in Figure 15. The top scale shows σ_{C14} in relation to particle diameter, D, assuming the relationship between D and R predicted by diffraction theory or by Mie theory with high absorption. Knowledge of

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*The refractive index m is a complex number, n-in'. The values of n' employed were 0.0, 0.01, 0.05, 0.1, 0.5, and 1.0. For n' = 0.0, n was varied from 1.33 to 1.96 in increments of 0.01. For the other values of n', n values of 1.33, 1.40, 1.45, 1.50 to 1.65 in increments of 0.01 were employed. In addition, calculations for the refractive index of carbon, 1.96-0.66i, were included. These may not be the exact refractive indices used for programming the PILLS IV; however, the differences are believed to be insignificant.

†The calculations were carried out by varying the Mie size parameter $\alpha(\pi D/\lambda)$ in increments of 0.2 from 0.9 to 13.9 where D is the particle diameter and λ is the wavelength, 0.904 µm.

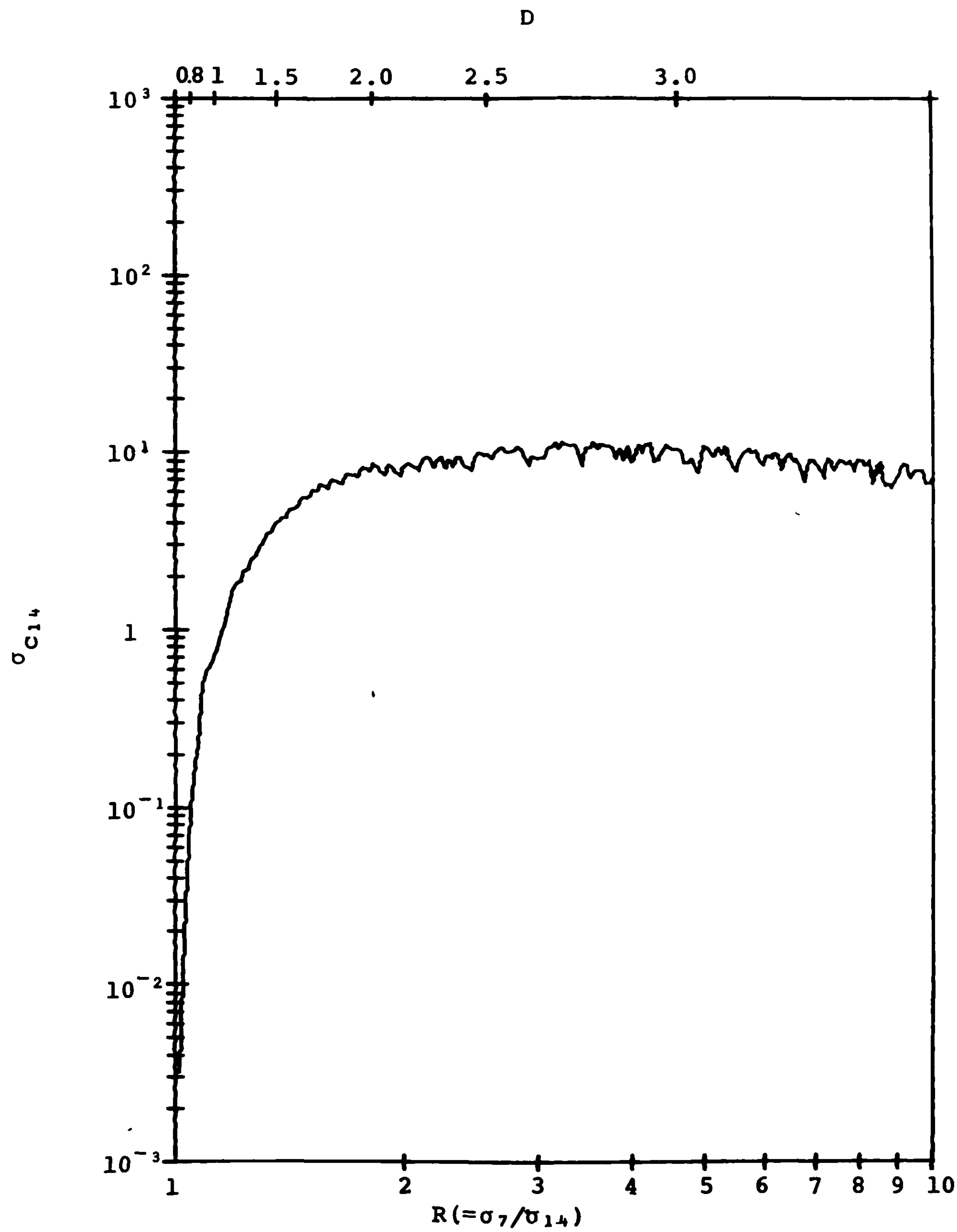

Figure 15. The Reference Function of the PILLS IV Versus the Ratio R. It determines the counting volume for particles of any diameter D. The top scale gives the equivalent diameter for any R-value programmed into the instrument.

$\sigma_{C14}(R)$ enables one to determine the relative response of the PILLS IV to particles of differing refractive index.

To predict σ_{14}/σ_{C14} the results of the Mie theory were used again. In addition, some calculations using diffraction theory were performed to check the appropriateness of Mie theory. The Mie theory of scattering assumes that the incident radiation is an infinite plane wave, and some differences in the actual particle scattering could be expected because the intensity of the PILLS IV beam is Gaussian in form and the intensity of light is not uniform across the scattering region associated with the particle. The results of the diffraction calculations show no significant change in σ_7/σ_{14} at different positions in the Gaussian beam (B_0 = 23 μm) until the particle diameter reaches 6 μm or higher. Thus Mie theory using the intensity of the wave at the center of the particle is suitable for the particles of interest here.

Figures 16-21 give theoretical response functions versus particle diameter for particles of five representative refractive indices, as expressed by the ratio $\sigma_{14}/\sigma_{C14}(R)$. The value $\sigma_{C14}(R)$ is given in Figure 15 using the appropriate value of R for each D. The ratio R versus D is also given for these refractive indices. When σ_{14}/σ_{C14} has a value of one and $R > 1.1$, particles of that size and refractive index are counted with maximum sensitivity; that is, the sizing volume for these is maximum. The maximum sizing volume V_{max} is determined by the optical-field transfer function of the collection optics and the beam profile as expressed in Equations (7)-(8). As $\sigma_{14}/\sigma_{C14}(R)$ decreases, the effective sizing volume decreases, since for a smaller value of σ_{14} the particle must be closer to the more intense central portion of the beam. When σ_{14}/σ_{C14} is below 0.2, no particles with $R > 1.1$ would be counted.

The significance of σ_{14}/σ_{C14} for particles in which $R < 1.1$ is most directly related to the deduced particle size. The higher values of σ_{14}/σ_{C14} for the same position in the view volume are counted in channels of the instrument corresponding to larger particles and two particles with the same value of σ_{14}/σ_{C14} could be counted in different channels if one were positioned closer to the center of the view volume than the other.

B. The Optical Transfer Function $f_{14}(r,\ell)$

The fluctuations in σ_{14}/σ_{C14} are important only if they cause a significant change in the counting volume associated with each channel of the PILLS IV output. To determine these volumes precisely one must know the optical transfer function $f_{14}(r,\ell)$.

The manufacturer performed measurements in which a 2 μm latex particle on a glass slide was traversed through the view

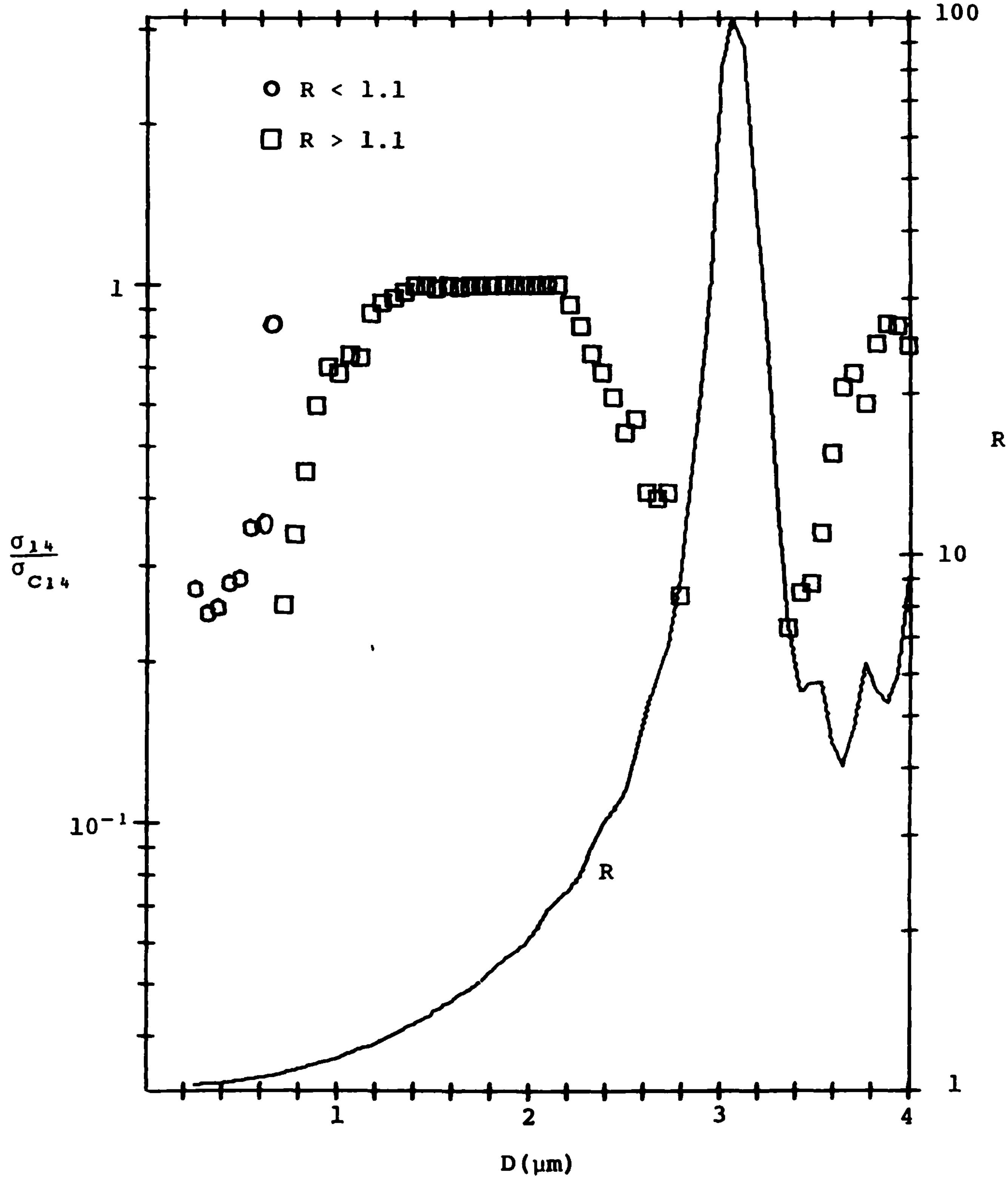

Figure 16. σ_{14}/σ_{C14} and R Versus D for a Refractive Index of 1.33-0.0i. The point symbols give σ_{14}/σ_{C14}.

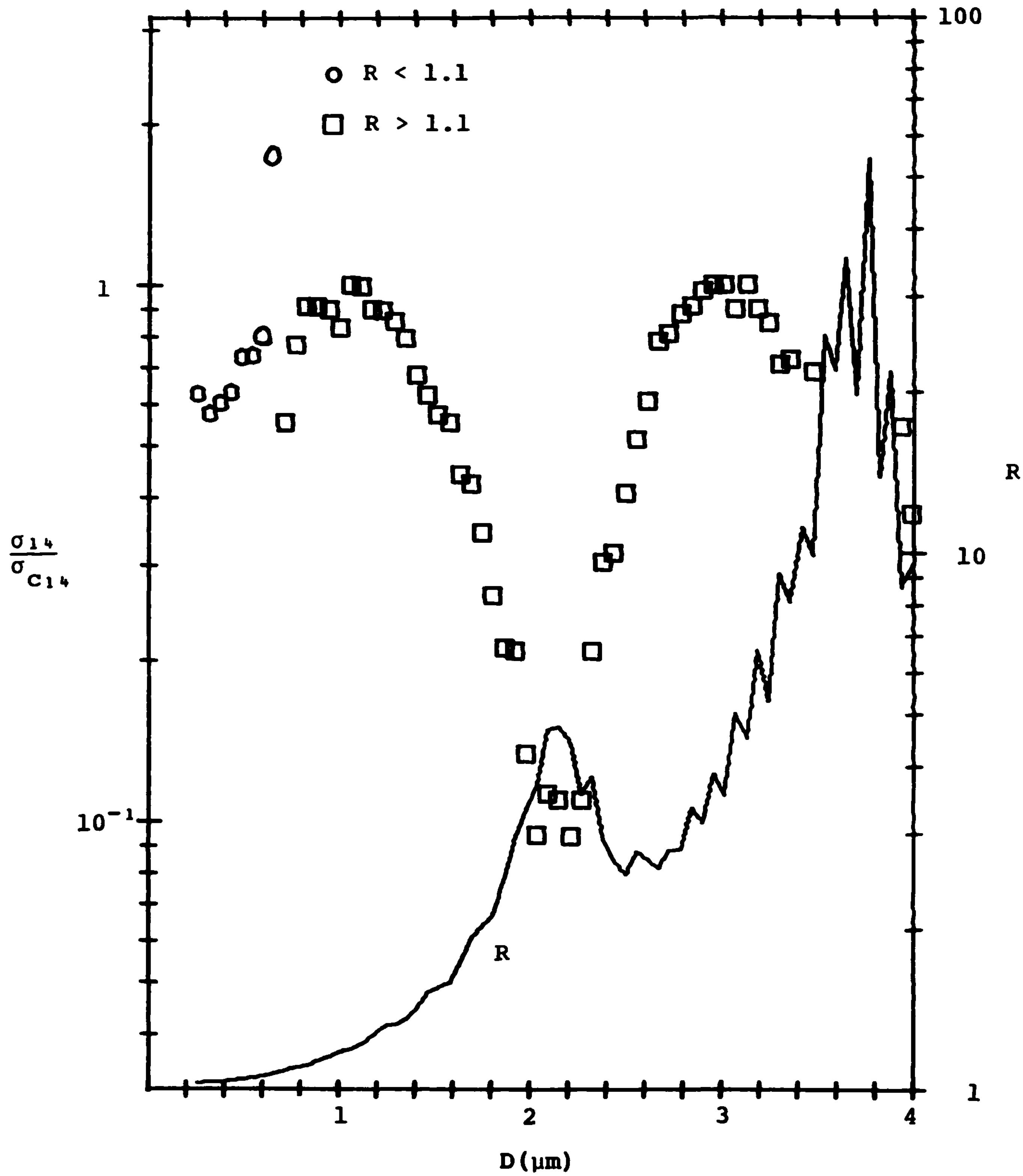

Figure 17. σ_{14}/σ_{C14} and R Versus D for a Refractive Index of 1.50-0.0i. The point symbols give σ_{14}/σ_{C14}.

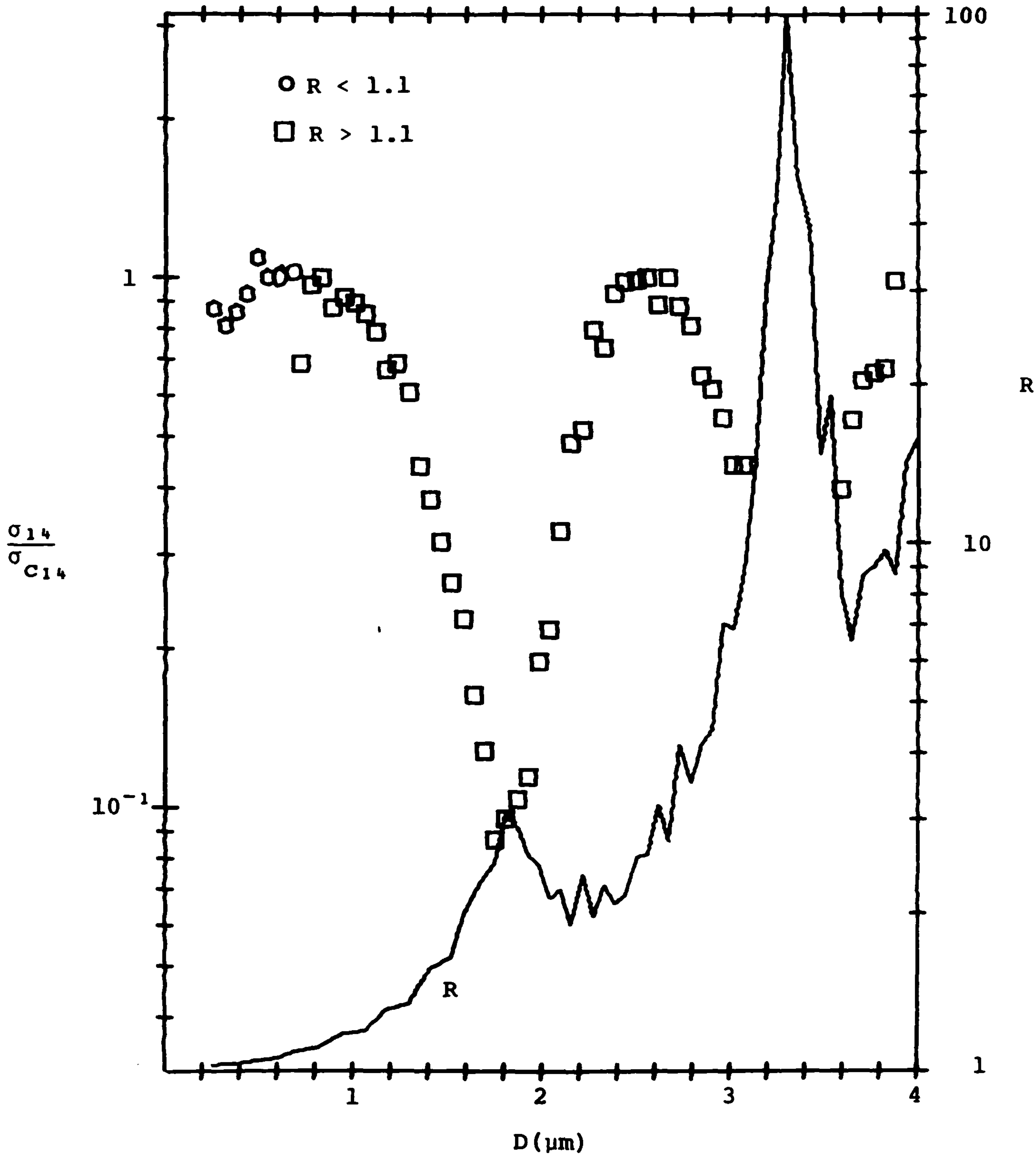

Figure 18. $\sigma_{14}/\sigma_{C_{14}}$ and R Versus D for a Refractive Index of 1.59-0.0i. The point symbols give $\sigma_{14}/\sigma_{C_{14}}$.

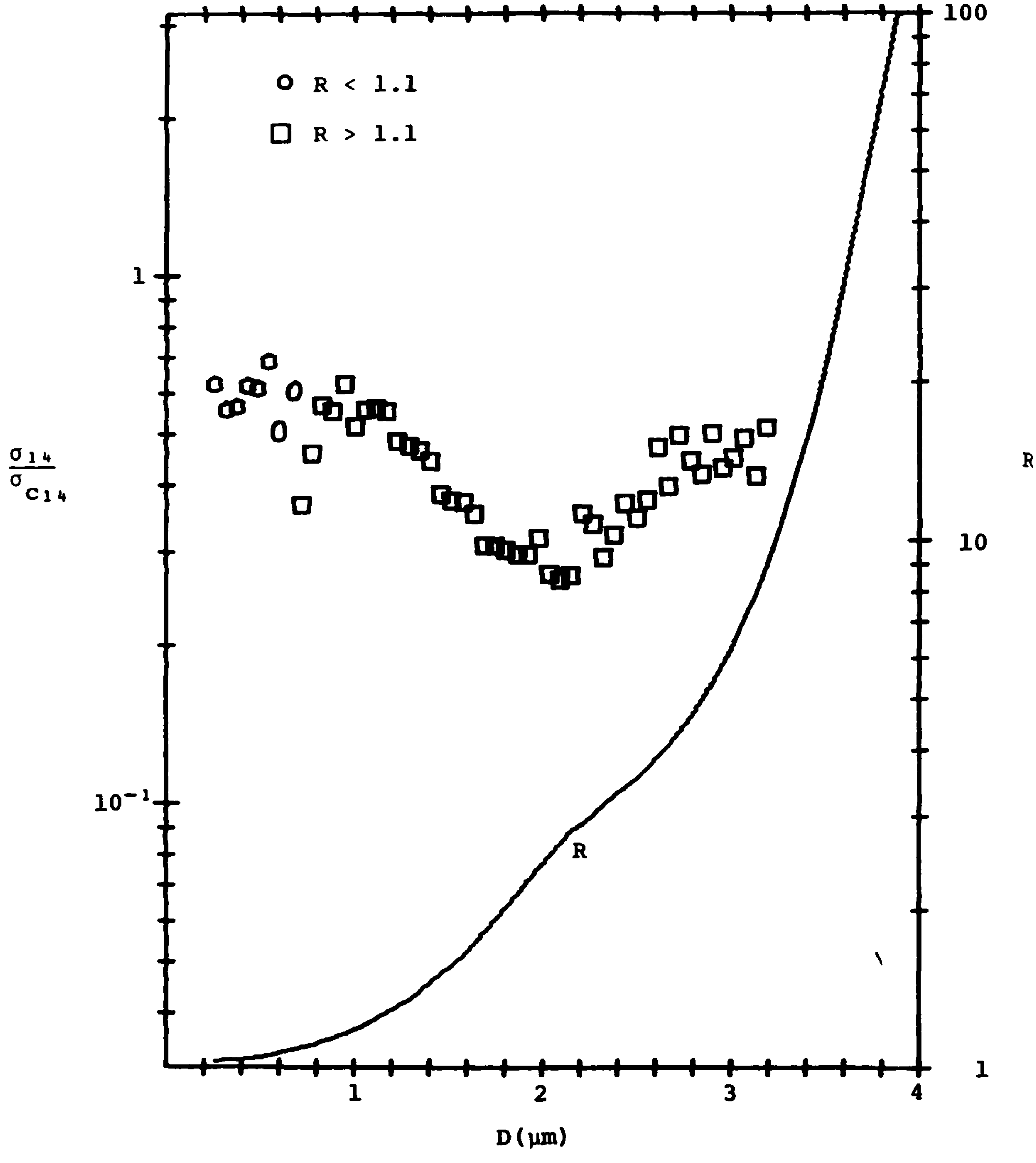

Figure 19. σ_{14}/σ_{C14} and R Versus D for a Refractive Index of 1.50-0.1i. The point symbols give σ_{14}/σ_{C14}.

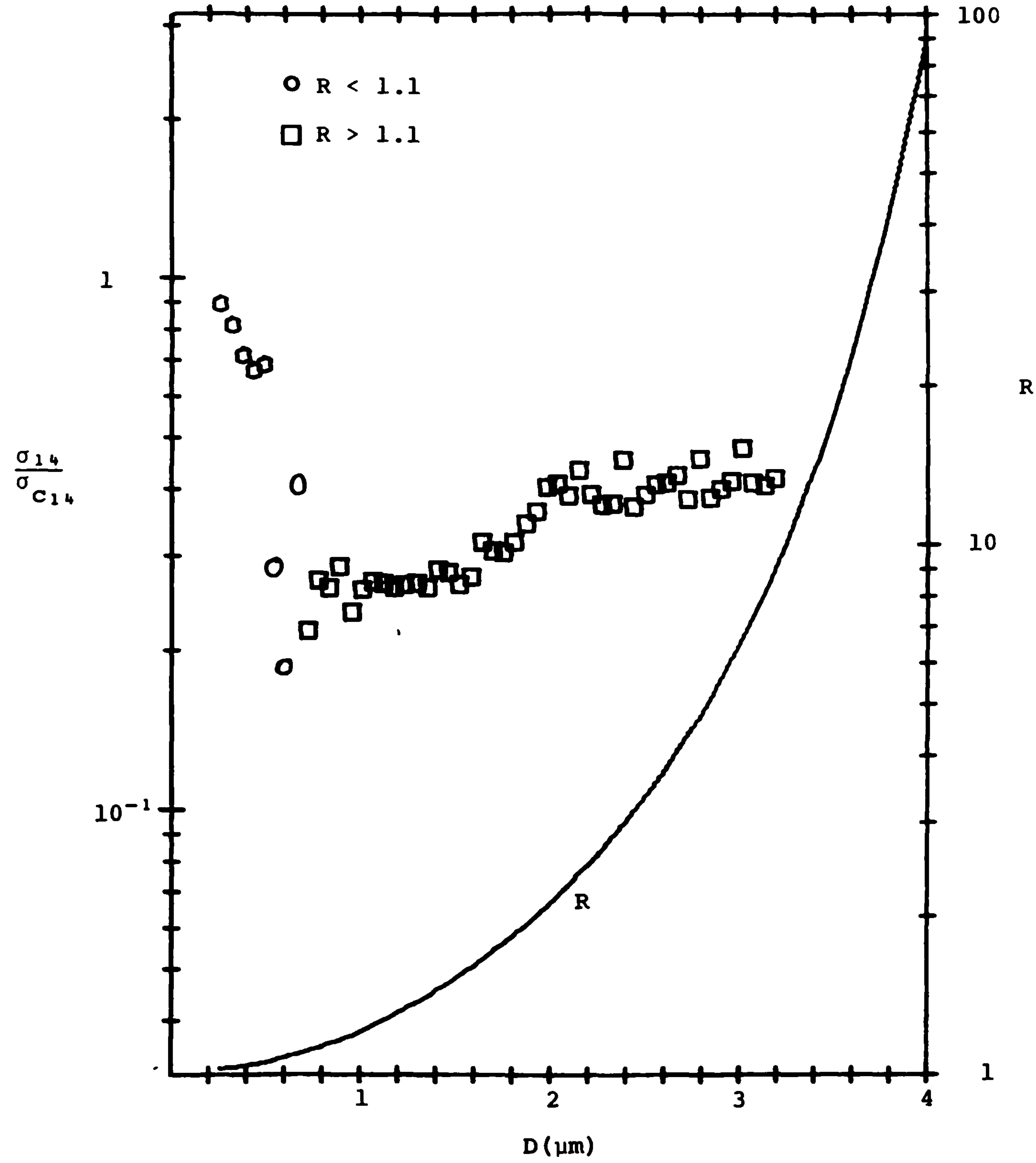

Figure 20. $\sigma_{14}/\sigma_{C_{14}}$ and R Versus D for a Refractive Index of 1.50-0.5i. The point symbols give $\sigma_{14}/\sigma_{C_{14}}$.

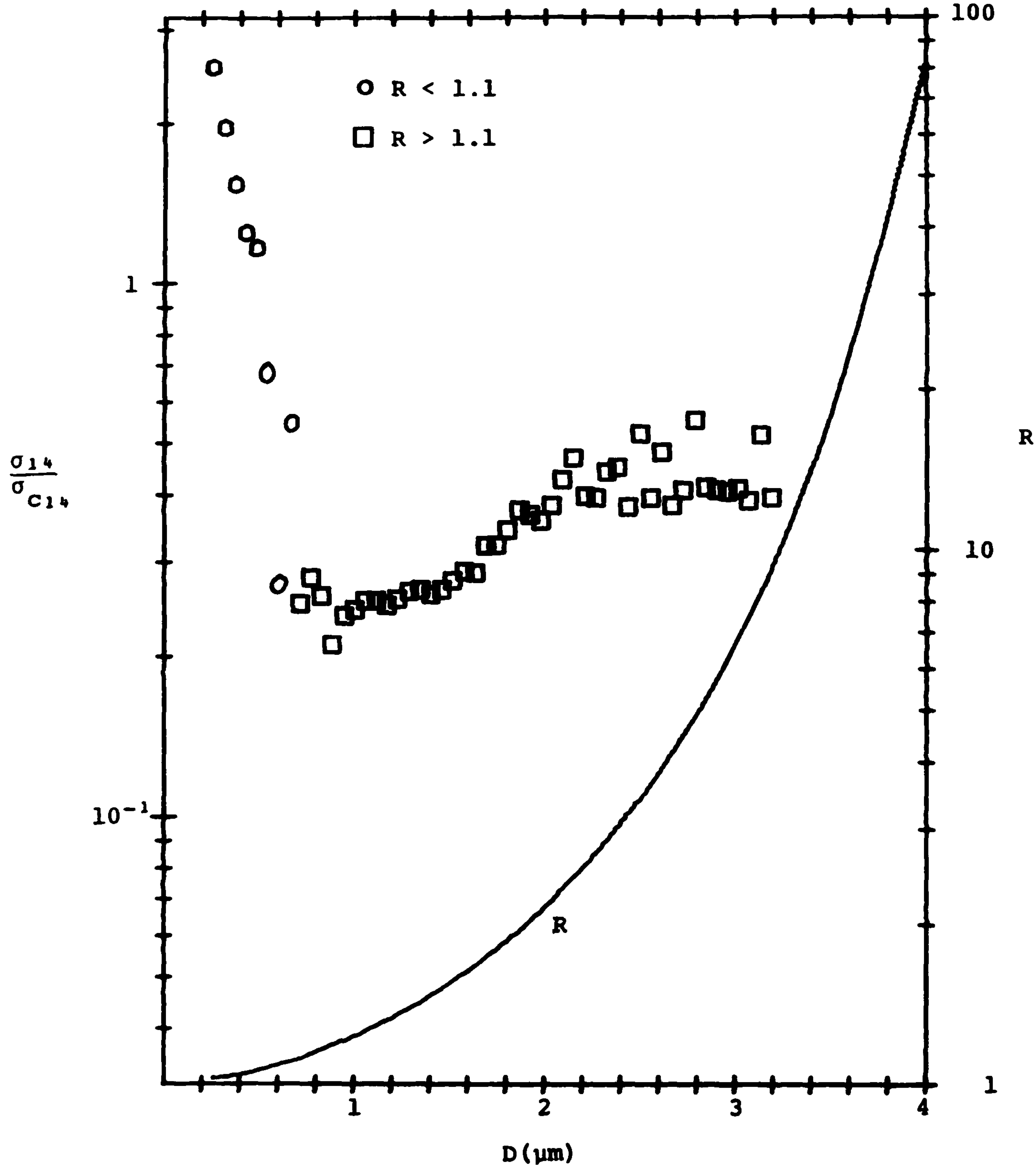

Figure 21. $\sigma_{14}/\sigma_{C_{14}}$ and R Versus D for a Refractive Index of 1.96-0.66i. The point symbols give $\sigma_{14}/\sigma_{C_{14}}$.

volume while monitoring the detected intensity of scattered light. It was found that values of r and ℓ for which

$$0.2 \le f_{14}(r,\ell)(\sigma_{14}/\sigma_{C14})e^{-2(r,\ B_0)^2} \le 1 \qquad (9)$$

formed a cylindrical surface 20 μm in diameter and 600 μm in length. This suggests that $f_{14}(r,\ell)$ is uniform in the central diamond-shaped portion of the view volume and decreases very rapidly near the edges of this region. Attempts to measure the view volume in this study were not conclusive but were consistent with the manufacturer's results, i.e., $f_{14}(r,\ell)$ is given by

$$\begin{aligned} f_{14}(r,\ell) &= 1 \text{ for } r < (b/\sin 14° - \ell)\tan 14° \\ &= 0 \text{ for } 0 > r > (b/\sin 14° - \ell)\tan 14°, \end{aligned} \qquad (10)$$

where b = 82 μm.

C. The Response Function of the PILLS IV

With $f_{14}(r,\ell)$, R(D), and $\sigma_{14}(D)/\sigma_{C14}(D)$ known, theoretical behavior of the PILLS IV for a given aerosol can be predicted from Equations (7) and (8) and the relationship of R as a function of diameter D. Equations (7) and (8) determine the counting volume for each particle size with given light scattering properties described by $\sigma_7'(D)$ and $\sigma_{14}(D)$. For channels in which R > 1.1 the count rate is the product of the particle concentration in the channel-size interval and the average counting volume over the size interval. For channels in which R < 1.1 the count rate is more complicated, because the channel in which a particle is counted depends upon its position in the beam. Thus particles having the same $\sigma_7(D)$ and $\sigma_{14}(D)$ can be counted in different channels. The counting volume for each particle size depends upon the lower and upper limits of the channel of interest. The total count rate of a channel is then the summation over all size intervals (for which R < 1.1) of the products of particle concentration in each size interval times the average counting volume over the size interval. This may be considered unusual behavior, but the count rate of each channel is actually similar to a measurement of the cumulative concentration of particles with sizes between that for which R = 1.1 and the size associated with each channel. A method for obtaining the size distribution from the count rate is discussed further in Section VII. The remainder of this section is concerned with characterizing the counting volumes as a function of particle light scattering properties and the channel limits. The most desirable behavior is obtained when V is independent of these parameters.

The counting volume is cylindrically symmetric about the optical axis. The intersection of its bounding surface with any plane parallel to the optical axis forms a trapezoid (see Figure 6) whose base and top are determined by minimum and maximum values of r which satisfy Equations (7) or (8). The size of this volume is given by

$$V = \frac{2\pi}{\tan 14°} \left[b(r^2_{max}-r^2_{min}) - \frac{1}{3}(r^3_{max}-r^3_{min})\right]. \qquad (11)$$

where $r_{min} \geq 0$. By inserting $f_{14}(r,\ell)$ of Equation (10) into Equations (7) and (8) the values of r_{max} and r_{min} can be obtained from

$$-\frac{B_0^2}{2}\ln\left[\sigma_{C14}(R)/\sigma_{14}(D)\right] \leq r^2 \leq -\frac{B_0^2}{2}\ln\left[0.2\sigma_{C14}(R)/\sigma_{14}(D)\right] \qquad (12)$$

for channels in which R > 1.1 and from

$$-\frac{B_0^2}{2}\ln\left[\frac{(\sigma_{C14})_k}{\sigma_{14}(D)} + \frac{\Delta(\sigma_{C14})_k}{\sigma_{14}(D)}\right] \leq r^2 \leq -\frac{B_0^2}{2}\ln\left[(\sigma_{C14})_k/\sigma_{14}(D)\right] \qquad (13)$$

for channels in which R < 1.1. Characterization of the PILLS IV behavior is straightforward for the upper channels governed by Equation (12), because the channel limits, set by the operator, are not involved. However, for the lower channels the behavior depends upon the channel limits. Characterization of the instrument's full potential then requires including the channel limits as a parameter.

Equation (13) can be put into the same form as (12) by noting in Figure 15 that σ_{C14} for R < 1.1 is proportional to D^5 and by identifying particle diameters $D_C \rightarrow D_C + \Delta D_C$ from calibration tables associated with $(\sigma_{C14})_k \rightarrow (\sigma_{C14})_k + \Delta(\sigma_{C14})_k$ for channel k. Then we find that

$$\frac{(\sigma_{C14})_k}{\sigma_{14}(D)} = \frac{\sigma_{C14}(D_C)}{\sigma_{14}(D)} = \frac{\sigma_{C14}(D)}{\sigma_{14}(D)}\left(\frac{D_C}{D}\right)^5$$

and

$$\frac{\Delta(\sigma_{C14})_k}{\sigma_{14}(D)} = \frac{\sigma_{C14}(D)}{\sigma_{14}(D)}\left(\frac{D_C}{D}\right)^5\left[\left(1 + \frac{\Delta D_C}{D_C}\right)^5 - 1\right]$$

Thus, Equations (12) and (13) actually have the same form expressed by

$$\frac{-B_0^2}{2}\ln\left[L(R)\sigma_{C14}/\sigma_{14}\right] \leq r^2 \leq \frac{-B_0^2}{2}\ln\left[L(R)H(R)\sigma_{C14}/\sigma_{14}\right] \qquad (14)$$

where

$$L(R) = (D_C/D)^5 (1 + \Delta D_C/D_C)^5$$
$$H(R) = (1 + \Delta D_C/D_C)^{-5}$$

for R < 1.1

and $L(R) = 1$

$$\left.\begin{array}{l} L(R) = 1 \\ H(R) = 0.2 \end{array}\right\} \quad \text{for } R > 1.1$$

Equation (14) inserted into (11) gives V as a function of $\sigma_{14}(D)$ for the upper channels and as a function of $\sigma_{14}(D)$, D, and the channel limits for the lower channels. V is given in Figure 22 in the form of V/V_{max} where V_{max} is the maximum counting volume occurring independent of particle light scattering properties. V_{max}, given by

$$V_{max} = \frac{2\pi}{\tan 14°} \left[-\frac{bB_0{}^2}{2} \ln H(R) - 1/3 \left(-\frac{B_0{}^2}{2} \ln H(R) \right)^{3/2} \right], \quad (15)$$

depends upon H(R), constant for the upper channels and variable for the lower channels. Note that V approaches zero when the abscissa approaches H(R). That is when r_{max} approaches zero.

To describe the response to particles with $R > 1.1$ from Figure 22, the curve for $H(R) = 0.2$ is employed. Sizing of particles is performed from R, but the number counted, proportional to V, depends upon σ_{14}/σ_{C14}. If σ_{14}/σ_{C14} is below one (see Figures 16-21) then V changes rapidly for small changes in σ_{14}/σ_{C14}. Indeed, for the calibration function σ_{C14} presently programmed into the PILLS IV σ_{14}/σ_{C14} is always less than or equal to one. It is desirable, however, that if $\sigma_{C14}(R > 1.1)$ were reduced so that σ_{14}/σ_{C14} is always greater than one, V/V_{max} is essentially constant for all refractive indices.

All of the curves in Figure 22 depict the variation of the counting volume of lower channels. Each channel has a curve determined by the channel limits through H(R). Note that ΔD_C, the width of each channel, must be decreased to increase H(R). The counting volume depends upon the independent variable $(\sigma_{14}/\sigma_{C14})(D/D_C)^5 H(R)$. To facilitate understanding of the dependance of V on σ_{14}/σ_{C14} and D the upper scales in Figure 22 show how V varies with $(\sigma_{14}/\sigma_{C14})(D/D_C)^5$ for specific values of H(R). Each scale refers to a different curve.

As an example, consider a monodispersed aerosol of latex particles where σ_{14}/σ_{C14} is 1 for all diameters and $D_{1.1}$, the diameter at which $R = 1.1$, is 0.75 μm. Then the number counted in the channel with associated diameters $D_C \rightarrow D_C + \Delta D_C$, assuming $\Delta D_C/D_C = 0.27$, is given by the curve labeled $H(R) = 0.3$ in Figure 22. A channel for which $D_C = D$ would receive no counts since V approaches 0 when the independent variable approaches H(R) or less. The number of counts would increase for decreasing D_C until $D_C = D[H(R)]^{1/5} = 0.87D$ where it would become essentially constant for lower channels. For a polydisperse aerosol of latex spheres the counts in any channel would be due to particles for which $D_C \leq D \leq D_{1.1}$. The number of counts in a given channel increases as D_C decreases for two reasons:

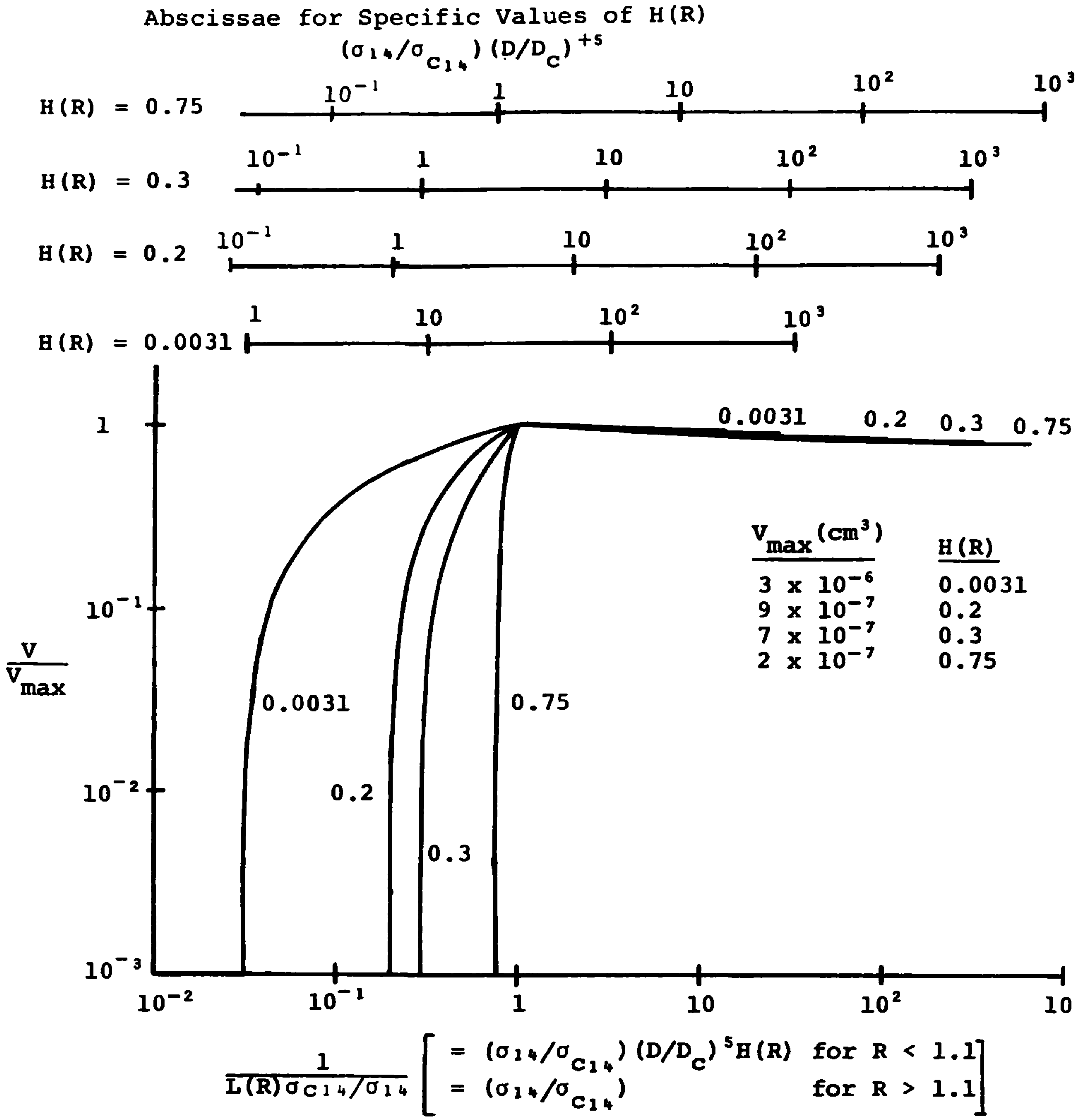

Figure 22. The Counting Volume of the PILLS IV Expressed in Terms of $\sigma_{14}/\sigma_{C_{14}}$, D, D_C, and ΔD_C for R < 1.1 and in Terms of $\sigma_{14}/\sigma_{C_{14}}$ for R > 1.1. V/V_{max} is given for 4 values of H. The top scales show V/V_{max} versus $(\sigma_{14}/\sigma_{C_{14}})(D/D_C)^5$ for specific H-values. If $\sigma_{14}/\sigma_{C_{14}}$ were one, then V goes from $0 \rightarrow V_{max}$ as D goes from $D_C \rightarrow D_C + \Delta D_C$.

because V/V_{max} is larger for lower D_C - values, and also the range of D-values which contribute to the channel is larger for lower D_C - values. Inspection of the curves shows that the one for which H(R) = 0.75 or higher is the most desirable for each channel since V is nearly a step function. Then for a polydisperse aerosol of latex spheres the count rate of each channel would give the cumulative concentration of particles with diameters in the range $D_C \leq D \leq D_{1.1}$. The differences in count rate between successive channels would give the concentration in the size range D_C to $D_C + \Delta D_C$. This approach is discussed further in Section VII.

For particles other than latex spheres $\sigma_{14}(D)/\sigma_{C14}(b)$ is not one and it varies with D as seen in Figures 16-21. For such particles the lower channels count particles for which $(\sigma_{14}/\sigma_{C14})^{-1/5} D_C \leq D \leq D_{1.1}$. This means variation in particle type produces some variation in the lower sizes of particles which a channel counts.

SECTION VI

THEORETICAL VERSUS OBSERVED RESPONSE

The theoretical description of the preceeding section explains the instrument's observed response to fly ash in Figure 1 if the function for $\sigma_{14}(D)/\sigma_{C14}(D)$ given in Figure 23 is chosen. Since very little is known about the optical properties of flyash the accuracy of this deduced function cannot be evaluated with certainty. In comparison to homogeneous spheres it agrees within 20% with the scattering of spheres having refractive index m = 1.5 - 0.5i. With the exception of a very small region around 0.8 μm, it could also be constructed of a combination of refractive indices with low imaginary parts and real parts varying from 1.5 to 1.96. Other factors effecting σ_{14}/σ_{C14} are particle shape and internal structure. It must also be recalled that the function given in Figure 23 is deduced with the assumption that the detector constants C_{14} and C_0 of Equation (6) remain constant after calibration as well as the ratio $f_7(r,\ell)/f_{14}(r,\ell)$ related to optical alignment of the lenses. These factors that can effect R are especially critical for sizes around 0.75 μm. For example, in this region a 15% lower value of $\sigma_{14}(D)$ causes R to be 18% high. If R > 1.1 this particular error would cause $\sigma_{C_{14}}(R)$ (see Figure 15) to be more than a factor of 5 higher, and σ_{14}/σ_{C14} would be less than 0.2 of its correct value. Thus, such an error in the deduced $\sigma_{14}(D)$ would eliminate essentially all counts. It appears then that such an error would also cause σ_{14}/σ_{C14} for m = 1.5 - 0.1i (Figure 19) to appear similar to that in Figure 23.

The response function [Equation (11)] also qualitatively explains the response to latex spheres, determined in this study. This is seen by comparing the values of σ_{14}/σ_{C14} at the appropriate diameter (Figure 18) to the response of the PILLS IV at the five diameters studied, given in Figures 9-14. In the case of 0.760 μm particles, it appears again as described above for fly ash that R was deduced to be slightly too high due to a shift in the optical constants, causing the chosen σ_{C14} to be so high that σ_{14}/σ_{C14} was too low for counting to occur.

The high count rate, which occurred in some instances, at sizes other than that of the nebulized latex, can only be explained by the presence of particles other than latex. The light scattering by these other particles must be such that the deduced size with the Climet is much smaller than with the ratio σ_7/σ_{14} used

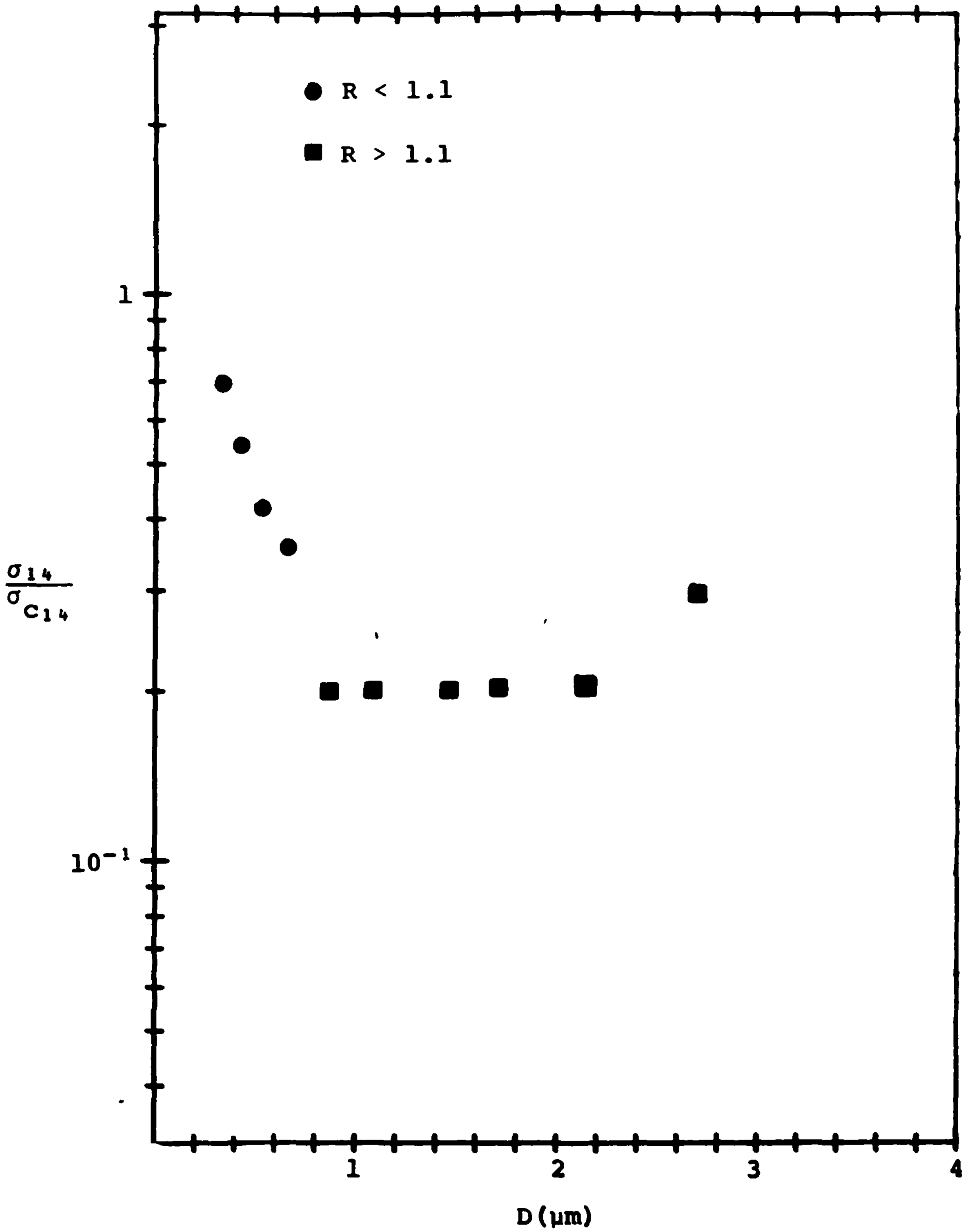

Figure 23. A $\sigma_{14}/\sigma_{C_{14}}$ Versus D Relationship that would Produce the PILLS IV Data from the Impactor Data in Figure 1. For D > 0.8, $\sigma_{14}/\sigma_{C_{14}}$ is unique while many possibilities for D < 0.8 could explain Figure 1.

by the PILLS IV. This suggests that the high PILLS IV counts compared to the Climet counts were due to particles formed from wetting agents in the stock solutions in which the latex particles are stored. Fuch's[7] discussion of these particles (called "empties"), and observations of them, indicates that very little is known about their formation. Apparently the size and structure are very sensitive to humidity and concentration of the nebulized solution. If the effective refractive index of these particles were close to one, the differences in the PILLS IV and Climet data in Figures 9, 11, and 14, would be explained. The light scattering at large angles (used by the Climet) decreases relative to small angle scattering (used by the PILLS IV) as the refractive index decreases. Thus, such particles would appear to be smaller when sized with the Climet instrument. Another explanation not related directly to light scattering is turbulence encountered by particles in the inlet line to the Climet. If the "empties" are fragile, then breakage could result to form small fragments.

SECTION VII

CONCLUSIONS AND RECOMMENDATIONS

It appears that the major problem that cause the PILLS IV to disagree with other sizing devices is the dependance of the instrument counting volume upon σ_{14}/σ_{C14} at low values of σ_{14}/σ_{C14} as seen in Figure 22. It is interesting that as σ_{14}/σ_{C14} increases the sizing volume reaches a maximum volume and remains essentially constant. This suggests that the major discrepancies could be eliminated if σ_{C14}(D or R) were reduced so as to increase σ_{14}/σ_{C14}.

The calculations performed in this study for homoneneous spheres showed that if σ_{C14} for $R > 1.1$ were reduced by a factor of 4 then σ_{14}/σ_{C14} would always be close to 1 or greater. Then for $R > 1.1$ the counting volume would be constant; thus eliminating the sensitivity to particle composition. This improvement would also be accompanied by a slight increase in the counting volume to include more of the fringes of the 14° detector view. Particles in this portion of the volume would be sized incorrectly; however, their contribution to the total count would be small.

For $R < 1.1$, decreasing σ_{C14}(D) would also improve the instrument response; however, the present scheme of reducing the data must be modified. If σ_{C14} for $R < 1.1$ were adjusted so that $(\sigma_{14}/\sigma_{C14})^{1/5}$ is close to one, then from Figure 22 it can be seen that the counting volume V goes from 0 to V_{max} as D goes from D_C to $D_C + \Delta D_C$ and is essentially constant at V_{max} for larger diameters. In the range of D from D_C to $D_C + \Delta D_C$, the average counting volume for $(\sigma_{14}/\sigma_{C14})^{1/5} = 1$ is given by

$$\bar{V}/V_{max} = 1 + 5/\ln H + D_C/\Delta D_C$$
$$\approx \frac{1}{2} \text{ for small } \Delta D_C/D_C \ (<0.3).$$

Thus, for $(\sigma_{14}/\sigma_{C14})^{1/5}$ close to one, half the particles in V_{max} with $D_C \leq D < D_C + \Delta D_C$ would be counted in the channel associated with D_C. In addition all of the particles in V_{max} for $D_C + \Delta D_C \leq D \leq D_{1.1}$ would be counted. This means that in the highest channel ($k = K$), where $D_C + \Delta D_C = D_{1.1}$, only particles in the size range $D_C \leq D \leq D_C + \Delta D_C$ would be counted. The actual concentration in that size range is thus given by

$$(\Delta N/\Delta D)_K (\Delta D_C)_K = 2\ \Delta N_K/\left[n(V_{max})_K\right]$$

assuming that $\Delta N/\Delta D$ does not vary much over the size increment. ΔN_K is the number of counts in the highest channel and n is the number of laser pulses. The number of counts in the $(K-1)^{th}$ channel would be given by

$$\Delta N_{K-1} = n(V_{max})_{K-1}[(\Delta N/\Delta D)_K(\Delta D_c)_K + \tfrac{1}{2}(\Delta N/\Delta D)_{K-1}(\Delta D_c)_{K-1}]$$

The same logic would hold for lower channels, so we find that the actual size distribution could be determined; i.e.,

$$(\Delta N/\Delta D)_k = 2[\Delta N_k/n(V_{max})_k - \sum_{i=k+1}^{K}(\Delta N/\Delta D)_i(\Delta D_c)_i]/(\Delta D_c)_k. \quad (16)$$

The accuracy of the scheme for $R < 1.1$ depends upon keeping $[\sigma_{14}(D)/\sigma_{C14}(D)]^{1/5}$ close to one. The effect of deviation of its value from 1 would be to shift the particle diameter range counted in each channel by a factor $[1-(\sigma_{14}/\sigma_{C14})^{-1/5}]$. The errors introduced by this factor could be minimized by a simple on site adjustment using the PILLS IV itself. The instrument should be modified so that σ_{C14} could be varied by a known adjustable factor. The procedure for that adjustment would use the highest channel for which $R < 1.1$ (i.e., channel K where $(D_C)_K + (\Delta D_C)_K = D_{1.1} \approx 0.75$). This factor would be varied until $\sigma_{14}(D_{1.1})/\sigma_{C14}(D_{1.1}) = [D_{1.1}/(D_C)_K]^{-5}$. The value of $\sigma_{C14}(D_{1.1})$ that produces that condition could be identified when the count rate in channel K goes to zero while increasing $\sigma_{C14}(D_{1.1})$. This procedure would produce a measurement of $\sigma_{14}(D_{1.1})$ that would then be used to set σ_{C14} so that $\sigma_{14}(D_{1.1})/\sigma_{C14}(D_{1.1}) = 1$. According to the calculation of $\sigma_{14}(D)$ for homogeneous spheres illustrated in Figures 16-21 this could leave $\sigma_{14}(D)/\sigma_{C14}(D) \neq 1$ for lower D-values of particles for some refractive indices. However, the maximum expected error in particle size would be less than 30% and more typically less than 10%. Of course the errors involved in this procedure could be reduced with some representative measurements of $\sigma_{14}(D)$ for aerosol particles encountered in the different types of process streams. Such measurements could also provide the possibility of changing the form of $\sigma_{C14}(D)$ as well as its magnitude to something more appropriate for the aerosol under study.

Another recommendation to improve measurements with the PILLS IV is the development of a method to easily calibrate the detectors. This would eliminate the potential problem discussed in Section VI concerning small shifts in C_{14}, C_7, and C_0, producing large errors in counting particles with diameters close to $D_{1.1}$. One possible method would be the temporary insertion of a known diffuse light source into the instrument's view volume.

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TECHNICAL REPORT DATA
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1. REPORT NO. EPA-600/7-78-130	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Evaluation of the PILLS IV		5. REPORT DATE July 1978 6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) William E. Farthing and Wallace B. Smith		8. PERFORMING ORGANIZATION REPORT NO. SORI-EAS-78-611
9. PERFORMING ORGANIZATION NAME AND ADDRESS Southern Research Institute 2000 Ninth Avenue, South Birmingham, Alabama 35205		10. PROGRAM ELEMENT NO. EHE624 11. CONTRACT/GRANT NO. 68-02-2131, T.D. 11007
12. SPONSORING AGENCY NAME AND ADDRESS EPA, Office of Research and Development Industrial Environmental Research Laboratory Research Triangle Park, NC 27711		13. TYPE OF REPORT AND PERIOD COVERED Task Final; 5/77-3/78 14. SPONSORING AGENCY CODE EPA/600/13

15. SUPPLEMENTARY NOTES IERL-RTP project officer is William B. Kuykendal, Mail Drop 62, 919/541-2557.

16. ABSTRACT The report gives results of theoretical and experimental investigations of the operating characteristics of the PILLS IV (Particulate Instrumentation by Laser Light Scattering) in situ particle sizing instrument. Results of both investigations show large errors in sizing particles with the instrument. Attempts to correlate the experimental findings with qualitative theoretical explanations established a sensitivity to particle refractive index and detector response that seems to account for the observed characteristics. Further measurements would be required to test this explanation quantitatively. The prototype was designed to measure particle size using the ratio of intensities of light scattered from a particle at two small angles (14 and 7 degrees) with respect to an incident laser beam. The intensity ratio was chosen because of its relative independence of particle refractive index. However, the magnitude of the scattered intensity at 14 degrees is also used for several important decisions in the electronic processing logic which, for this particular optical system, render it especially sensitive to refractive index and detector variations for determinations of particle size distribution. Possible solutions to these problems were offered with only minor hardware changes.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Air Pollution	Air Pollution Control	13B
Measurement	Stationary Sources	14B
Size Determination	Particulate	
Dust	PILLS IV	11G
Lasers		20E
Light Scattering		20N,20F

18. DISTRIBUTION STATEMENT Unlimited	19. SECURITY CLASS *(This Report)* Unclassified	21. NO. OF PAGES 51
	20. SECURITY CLASS *(This page)* Unclassified	22. PRICE

EPA Form 2220-1 (9-73)