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DERIVATION OF EQUATIONS TO DETERMINE FREQUENCY
DISTRIBUTIONS OF PARTICLE SIZE

G. A. SEHMEL

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DERIVATION OF EQUATIONS TO DETERMINE FREQUENCY
DISTRIBUTIONS OF PARTICLE SIZE

By

G. A. Sehmel

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TABLE OF CONTENTS

	<u>Page</u>
<u>ABSTRACT</u>	1
<u>INTRODUCTION</u>	1
<u>SUMMARY</u>	2
<u>GRAPHICAL REPRESENTATION OF SIZE DISTRIBUTION DATA</u>	3
<u>DEVELOPMENT OF METHOD</u>	11
<u>FITTING EXPERIMENTAL DATA</u>	13
<u>FITTING STANDARD FORMS</u>	13
<u>CONCLUSIONS</u>	19
<u>NOMENCLATURE</u>	21
<u>REFERENCES</u>	21

LIST OF FIGURES

<u>Figure No.</u>		<u>Page</u>
1	Graphical Representation of Cumulative Size Distributions	6
2	Calculated Distribution Function Between 3 and 17 Micron	8
3	Comparison of the Initial Estimate and the Calculated Distribution Function	10
4	Typical Experimental and Calculated Size Distributions	14
5	Typical Frequency Distribution Functions for Experimental Data	15
6	Frequency Distribution Functions for Assumed Standard Distributions	17
7	Calculated Size Distributions for Assumed Standard Distributions	18

LIST OF TABLES

<u>Table No.</u>		
I.	Calculation of Cumulative Size Distribution	5
II.	Calculation of Initial Approximation of the Distribution Function	9
III.	Precision in Fitting Standard Distributions	20

DERIVATION OF EQUATIONS TO DETERMINE FREQUENCY
DISTRIBUTIONS OF PARTICLE SIZE

ABSTRACT

A method was developed for obtaining a mathematical expression which represents particle size frequency distributions when the measured distributions are originally specified in terms of size increments. The method is particularly applicable when a measured size distribution does not correspond to the normal, log-normal, and Weibull distributions. Nevertheless, the method is also applicable to these distributions over the limits of the experimental data. The method is an iterative least squares technique to fit a partial distribution function over the size range of the experimental data. Particle size data are represented which were initially grouped into seven size increments, but the method should have applicability to any number of size increments.

INTRODUCTION

Several distribution functions have been used to represent particle size distributions. These include the log-normal^[1], mixed exponential^[1,2], Mugele-Evans upper-limit function^[1,2], normal^[1,2], Nukiyama-Tanasawa^[1,2], Rosin-Ramler^[1,2], and Weibull.^[3] These expressions are all empirical except for the log-normal which may be justified for some types of particles on the bases of processes associated with their physical formation. Although the log-normal distribution is often used to fit data because experience shows frequent agreement, one may be as equally justified to choose another distribution function as long as the data are fitted accurately.

An accurate, even though empirical, representation of particle size data is often required in small particle studies in which the particle behavior is a marked function of particle size. For example, inertial impaction^[4] of particles on a surface is proportional to the square of the particle size, the deposition of particles^[5] from a turbulent air stream upon the surfaces of a conduit may be proportional to greater than the fourth power of the particle size, and the errors^[4] associated with anisokinetically sampling aerosols from an air stream are a function of particle size.

Particle behavior in these instances can most readily be studied by using uniform aerosols from controlled condensation processes, evaporation of uniform drops, dispersed polystyrene latex spheres, pollens, or classified powders. If the physical properties of the aerosols from these processes do not meet experimental criteria, an aerosol of non-uniform size must be used. In this case, the correct number of each size particle may have to be determined from an aerosol containing many particle sizes.

A problem in particle research is to identify the number and hence behavior of each size particle. In some experiments, even when a uniform aerosol could be used, eventual contamination of the equipment with particles of several sizes may ultimately require the use of particle sizing and size distribution analysis.

Much work has been done on the development of automatic devices and instrumental aids to increase the accuracy and the rapidity with which particle size analyses can be made. In general, the output from each method is the same as that obtained through the use of a calibrated graticule in the eyepiece of a microscope. Particles are sized and each is recorded as being within one of a series of size increments. The data must then be processed by some method of interpolation to determine the number of particles of any required size. These methods of interpolation are often based upon representing the data by normal, log-normal, or Weibull distributions. In a recent study^[4] these three methods of representing the data were found inadequate and a systematic method of representing the data was required.

The purpose of the present paper is to present the method developed for fitting grouped particle size data in which no function relationship to describe the distribution can be postulated a priori. The method should have general applicability to any particle size analyses. The applicability of the method to both experimental data and assumed classical distributions will be shown.

SUMMARY

A method was developed to yield an expression for the particle size distributions between the limits of the experimentally determined size range. The method is based on a first estimation of the size frequency distribution, integration to determine the calculated number of particles in any size range, comparison with the experimentally determined number of particles, and iteration

until the size distribution as determined by least squares techniques converges. Computer processing facilitates the calculations.

The observation was made during the development of the method that the logarithm of the quotient of the percentages of particles in any size increment divided by the width of the size increment in microns, was nearly linear with the particle size at the mid-point of the increment, or at most could be represented by a cubic expression on semi-log paper. The cubics approximated the true size distributions.

The method is to determine by least squares techniques the relationship between the logarithm of these quotients of percentages per unit size and the average particle size in each size increment. Next, the analytical least squares equation is integrated to obtain the calculated percent of particles in each size increment. The calculated, C_i , and experimental percentage, E_i , in each size increment are compared and the comparison used as a basis for a first iteration. That is for each average size, $\bar{x}_i$, in an increment, Δx_i , the next estimate of the frequency distribution, f_i , at $\bar{x}$, is

$$f_{i+1} = f_i + \frac{E_i - C_i}{\Delta x_i} \quad (1)$$

The calculation is repeated and by the fifth iteration the constants converge in the least squares equation.

GRAPHICAL REPRESENTATION OF SIZE DISTRIBUTION DATA

The true distribution of particle sizes would be determined by taking a representative sample of particles and identifying each particle as having a diameter lying within an infinitesimal diameter increment. In practice, the infinitesimal diameter increments must be increased to a finite class increment. The experimentally determined sizes must be determined to an accuracy commensurate with the ultimate use of the data. The problems and errors associated with particle sizing will not be considered here. However, the importance of these problems and errors are not to be minimized. They may be so large as to preclude a refined analysis of the data. Nevertheless, the assumption is made that the technique to be developed for calculating particle distribution functions will not be limited by gross inaccuracies in the input data.

The case will be considered in which particles were sized into seven size increments. For this case, the accuracy and precision of the data are comparable

for each size increment due to a greater relative portion of the sample actually sized for increments with fewer particles.

An example showing the analysis of data obtained in seven increments will be discussed in terms of an example calculation. This example will show the inadequacies of existing analysis techniques and will be used to show the development of the present technique. The example is discussed in terms of the data in Table I in which are shown the seven increments in column one. In column two are shown the limits of each size increment. The first increment is for all particles below 3 micron diameter. Particles smaller than 3 micron could not be sized since the self-illumination of fluorescent light used in the study was insufficient to adequately define the smaller particles. The upper size limits of each of the adjacent seven increments are related by the square root of two. This corresponds to the relationship between the circles of a Porton graticule used in the eyepiece of a microscope. Particles greater in size than the 25 microns of the seventh increment were observed in the study, but the number of these particles were few and did not have a significance to the study. Thus, a particle size distribution is to be analyzed of which the particles of interest are between 3 and 25 microns.

In column 3 of Table I are shown the number of particles sized and counted for each increment. These number data have been converted to the cumulative percent of particles less than the upper limit of each size increment in column 4. These cumulative percentages are usually plotted on normal or log-normal paper, as a function of particle size in order to characterize a size distribution. If the cumulative data exhibit a straight line relationship, the distribution can easily be characterized in terms of a mean, μ , and a standard deviation, σ . However, if a straight line relationship is not indicated, a force-fit of a μ and σ to the data may cause significant errors in the interpretation and ultimate use of the data. Such was the situation for the data in Table I.

The cumulative percentages are shown plotted on log-probability coordinates in Figure 1. Two facts can be seen immediately. First, the data do not show a linear relationship. Secondly, the data point for the largest particle size is located at 17.8 microns. This means that some method of extrapolation to 25 microns is needed to represent the data obtained in the seventh size increment. The extrapolation will be considered later. For the moment, the determination of the percentages of particles within one-half micron of each micron integer size will be considered. These percentages, f_x , can be determined from

TABLE ICALCULATION OF CUMULATIVE SIZE DISTRIBUTION

<u>Increment Number</u>	<u>Diameter* Of Size Increment (microns)</u>		<u>Number of Particles In Size Increment Y_i</u>	<u>Cumulative % Less Than Upper Limit Of Size Increment F</u>
	<u>x_L</u>	<u>to x_H</u>		
1	0	3.14	3200	72.2
2	3.14	4.45	533	84.2
3	4.45	6.28	201	88.7
4	6.28	8.90	94	90.9
5	8.90	12.56	90	92.9
6	12.56	17.8	193	97.9
7	17.8	25.2	124	100.0
TOTAL			4435	

* Diameters of succeeding increments
increase by the factor of $\sqrt{2}$.

Figure 1

Graphical Representation of Cumulative Size Distributions

Figure 1 by evaluating the slopes of the cumulative distribution curve:

$$f_x = \frac{F_{x+\epsilon} - F_{x-\epsilon}}{2\epsilon} \quad (2)$$

where $\epsilon = 0.5$ microns. As ϵ approaches zero, the distribution of the slopes becomes the distribution function, f , which has the property that the percentage of particles that have diameters between x and $x + dx$ can be written

$$dF = f(x)dx \quad (3)$$

with the normalizing condition,

$$\int_0^{\infty} f(x)dx = 100. \quad (4)$$

The distribution function, f , calculated from the curve in Figure 1 is shown in Figure 2. The distribution function extends from 3.6 to 17.3 microns and suggests that f is an exponential cubic function of particle size.

The distribution function can be approximated assuming f is a linear function of x . This linearity is characterized by the ratio of the incremental percentage of particles to the length of the size increment and graphing this ratio as a function of the average particle size in each increment. The purpose of this approximation is to obtain an easily determined first approximation of f . These calculations are shown for the example problem in Table II. The size increments are shown in column 2 and are seen to increase with increment number from 1.3 to 7.4 microns. The first increment is meaningless for approximating slopes for sizes below 3 microns since the smallest size particle present is unknown. The linear slopes and increments midpoints shown in columns 5 and 6 are plotted in Figure 3 as the data symbol X. Included in this figure is the distribution function, f , from Figure 2. The data symbols are seen to be located along the curve, f . By analogy, this means that the symbol for f at the midpoint, 21.5 microns, of the seventh increment should approximate the distribution function extending from 17.8 to 25.2 microns. It will be determined that the distribution function can be represented by the broken line.

To summarize, an example size distribution has been cited which does not follow the standard expressions used to characterize particles. The distribution function has been determined to be approximately an exponential cubic

Figure 2
Calculated Distribution Function Between
3 and 17 Micron

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TABLE II
CALCULATION OF INITIAL APPROXIMATION OF THE
DISTRIBUTION FUNCTION

<u>Increment Number</u>	<u>Size Increment ($x_H - x_L$) (micron)</u>	<u>Incremental % Of Particles Of Sizes Within Δx E_i</u>	<u>E_i ($x_H - x_L$)</u>	<u>Average Diameter For Increment $\bar{x}$</u>
1	(3.14)	(72.2)	--	--
2	1.31	12.0	9.16	3.80
3	1.83	4.5	2.46	5.37
4	2.62	2.2	0.839	7.59
5	3.66	2.0	0.546	10.73
6	5.24	4.3	0.821	15.18
7	7.4	2.8	0.389	21.5

Figure 3

Comparison of the Initial Estimate and the
Calculated Distribution Function

function of particle size and the true f can be approximated by assuming f is a linear function of x for each size increment. An analytical expression for f will be calculated in order that the percentage of each integral micron particle size can be obtained.

The method of Irani^[6] for abnormalities in the log-normal distribution may fit these data, but Irani's methods would require more complex calculations than the simple method to be outlined. This simplicity and general applicability are the prime worth of the method. The applicability will be shown.

DEVELOPMENT OF METHOD

The present method of calculating an analytical expression for the distribution function was based on the observation that f could be initially approximated by assuming that f is a linear function for each increment of particle size. The second assumption is that the true distribution function is a smooth continuous function without rapid increases or decreases for any particle size. This means that the distribution function can be represented by a simple function. The assumption is made that the simple functional form for the distribution function is a cubic in the examples cited:

$$f(x) = \exp(a + bx + cx^2 + dx^3), \quad (5)$$

in which the random variable, x , is particle size and the parameters are a , b , c , and d . For the case under consideration there are seven observations and four parameters or there are three degrees of freedom not used for fitting. The use of seven parameters would yield a perfect fit for the case of the seven size increments while the use of a greater number of size increments would increase the degrees of freedom.

The terminology used in the method is as follows: The number of particles in each size increment is y_i , the total number of particles counted is N , Δx_i are the increment intervals, and $\bar{x}_i$ are the midpoints of the intervals. In the method, the $\bar{x}_i$ are held constant and an iterative technique is used to determine the quantity,

$$Y_i = \ln[f_i] \quad (6)$$

where f_i is the frequency distribution at $\bar{x}_i$. Least squares techniques are used to minimize the quantity Q ,

$$Q = \sum_{i=2}^n \left[Y_i - (a + b\bar{x}_i + c\bar{x}_i^2 + d\bar{x}_i^3) \right]^2, \quad (7)$$

with respect to the parameters in the distribution function. This technique determines the constants a , b , c , and d .

The values of f_i are initially determined by assuming the linearity

$$f_{i,1} = \frac{100 y_i}{N \Delta x_i}. \quad (8)$$

Having now obtained the parameters from an initial least squares fit, the distribution function

$$f_i = \exp(a + bx_i + cx_i^2 + dx_i^3) \quad (9)$$

can be integrated between the limits of each class interval. Thus, the calculated fraction, C_i , in any interval is calculated from the first estimation of the distribution function by the equation

$$C_i = \int_{x_L}^{x_H} f(x) dx \quad (10)$$

in which the limits of integration are the size limits of each size increment. Values of C_i can be calculated for class increments two through seven, but are meaningless for increment one.

For the second trial to determine the true distribution function, the values of the initial approximations of the distribution function are adjusted to minimize the difference between the calculated, C_i , and the experimental, E_i , percentage of particles in each size increment. The adjustments are

$$f_{i,2} = f_{i,1} + \frac{E_i - C_i}{\Delta x_i} \quad (11)$$

where the subscript 1 refers to the first estimate and 2 refers to the second estimate of f_i . For the second trial, equation (6) for Y_i is

$$Y_i = \ln[f_{i,2}] \quad (12)$$

and the procedure repeated of calculating a least squares curve for the distribution function, integration of the distribution function, and comparison of calculated and experimental percentages in each increment to determine new input data for the least squares fit. The parameters in the distribution function were found to converge by the fifth iteration. This procedure has been applied to experimental data and to assumed standard distributions.

FITTING EXPERIMENTAL DATA

The method outlined above was used to process the data in Table I. The solid and dashed distribution curve shown in Figure 3 was determined in this manner. This curve permits calculation of percentages of particles having diameters between 3 and 25 microns. However, the cumulative plot is somewhat desirable. In this case the function can be integrated beyond the size limits used to determine the analytical expression. This was done in the present situation for particle sizes between 25 and 50 micron. This permits the graphing of a point at 25 microns, but does not suggest that this point can be used to predict the percentage of particles above 25 microns. This extrapolation is strictly a method to represent all the cumulative data from 3 to 25 microns on a single graph as is shown by the solid and dashed curve of Figure 1.

The method was applied to obtain experimental distribution functions for typical curves shown as A, B, and C in Figure 4. Curve B is the example curve discussed. These three curves show a wide range of size distributions, and all are fitted by the technique. The distribution functions for these curves are shown in Figure 5. The first estimates are good approximations to the distribution functions and the distribution functions for a variety of forms are fitted by the technique. The technique has now been shown to represent three non-standard distributions, but one may want to know how good is the technique for predicting a standard distribution. The fitting of several standard distributions will be considered.

FITTING STANDARD FORMS

Size distributions which are normally distributed, log-normally distributed, and Weibull distributed have been fitted by the technique. These expressions each have only two parameters as compared to the four parameters of the cubic expression used for the distribution function. That is, the two parameters in the normal and log-normal distributions are the mean, μ , and the var-

Typical Experimental and Calculated Size Distributions

Figure 5

Typical Frequency Distribution Functions
For Experimental Data

iance, σ . For the Weibull distribution the cumulative distribution, F , is given by

$$F = 1 - \exp(-ax^b) \quad (13)$$

in which a and b are the two parameters. The distribution functions for the Weibull distribution is

$$f = (ax^b \ln x) \exp(-ax^b) \quad (14)$$

and for the normal distribution is

$$f = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{1}{2}\left(\frac{x-\mu}{\sigma}\right)^2\right]. \quad (15)$$

For the log-normal distribution the variable x in the normal distribution is replaced by the logarithm of the variable x .

The following procedure was used to test the ability of the present method to fit three assumed standard distributions. Straight lines were drawn on graph paper representing normal, log-normal and Weibull distributions and the percentage indicated within each size class was determined. These fractions were used as the raw data for calculating the size distribution from the cubic expression of equation (5). The initial approximations, the ratio of percentage divided by increment width, for the distribution functions are shown as circles in Figure 6. Also shown in the lower left hand corner is a graph with a line for which the distribution function was originally assumed to be straight in order to calculate the raw data. The curves obtained after the fifth iteration of the method are shown for each of the four areas. The curve shapes ranged from convex and concave to having a point of inflection. These curves were integrated over particle sizes from 3 to 50 microns. No particles greater than 50 microns were assumed to be present. The results of these integrations after summations are the cumulative distributions shown in Figure 7. The calculated percentage from 25 to 50 microns is included in both the calculated curve and as an eighth increment of data in order to plot the data point at 25 microns.

The calculated curves are seen to fit the experimental points. However, the calculated curves are not the same straight lines from which the raw data were obtained. This non-linearity can be seen on the graphs with log-normal and Weibull coordinates. The original straight line distributions are represented

Figure 6
Frequency Distribution Functions For Assumed
Standard Distributions

Figure 7
Calculated Size Distributions for
Assumed Standard Distributions

by the broken line extension of the straight line portions of the solid curves. This deviation of the solid curve from the broken straight line is caused by the extrapolation of the distribution function beyond the limits for which the constants were determined. The calculated results are valid even though straight lines are not obtained. The precision of fit of the distribution function for these four standard distributions can be shown by the deviation of the calculated from the experimental percentage of particles in each increment. The largest deviation shown in Table III is only six per cent -- an acceptable value in any case of real data due to the experimental problems and errors associated with sizing particles.

CONCLUSIONS

A method has been presented for calculating particle size distributions from grouped data. The method yields an objective analysis of the data by minimizing investigator bias. The method can be used when the data can not be represented by a recognized functional form, or when numerous size distribution analyses are required. The method utilizing a cubic expression for the distribution function has been shown to satisfactorily fit experimental, normal, log-normal, and Weibull described data. If data were obtained in more than seven groups, the number of constants in the distribution function could be increased to improve the fit. The principal limitations are that the true distribution is smooth, without discontinuity, without large fluctuations for small size increments, and is representative only within the range of sizes for which it was established.

The method has been found to predict within a few per cent over 80 size distributions can be used to calculate any of the commonly calculated characteristics of a size distribution.

TABLE III

PRECISION IN FITTING STANDARD DISTRIBUTIONS

Deviation of Calculated from Experimental Data
For Assumed* Straight Line Distributions,

$$\frac{100(C_i - E_i)}{E_i}$$

<u>Increment</u>	<u>f</u>	<u>Normal</u>	<u>Log-Probability</u>	<u>Weibull</u>
2	0.97	0.26	4.06	0.34
3	-0.64	0.61	-3.97	-1.26
4	-0.44	1.87	-1.58	6.35
5	1.40	-2.08	5.54	-5.65
6	-0.83	0.75	-2.79	2.18
7	-0.43	-0.57	-0.50	-1.06

* Data determined graphically.

NOMENCLATURE

- C_i = calculated percentage of particles within an interval Δx_i
 E_i = experimental percentage within an interval Δx_i
 f = frequency distribution function
 F = cumulative distribution function
 i = subscript for particles within an interval
 N = total number of particles in an experimental distribution
 Q = sum of squares, see Equation (7)
 x, x_i = random variable which is particle diameter size
 $\bar{x}_i$ = midpoint size of an increment
 y_i = number per cent of particles experimentally determined of size within the increment, Δx_i
 Y_i = See equation (6)
 Δx_i = increment width
 ϵ = differential increase
 μ = mean size in normal distribution
 σ = variance

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