United States
Environmental Protection
Agency

Industrial Environmental Research
Laboratory
Research Triangle Park NC 27711

EPA-600/7-78-189
September 1978

EPA

Analysis of Cascade Impactor Data for Calculating Particle Penetration

Interagency Energy/Environment R&D Program Report

RESEARCH REPORTING SERIES

Research reports of the Office of Research and Development, U.S. Environmental Protection Agency, have been grouped into nine series. These nine broad categories were established to facilitate further development and application of environmental technology. Elimination of traditional grouping was consciously planned to foster technology transfer and a maximum interface in related fields. The nine series are:

1. Environmental Health Effects Research
2. Environmental Protection Technology
3. Ecological Research
4. Environmental Monitoring
5. Socioeconomic Environmental Studies
6. Scientific and Technical Assessment Reports (STAR)
7. Interagency Energy-Environment Research and Development
8. "Special" Reports
9. Miscellaneous Reports

This report has been assigned to the INTERAGENCY ENERGY-ENVIRONMENT RESEARCH AND DEVELOPMENT series. Reports in this series result from the effort funded under the 17-agency Federal Energy/Environment Research and Development Program. These studies relate to EPA's mission to protect the public health and welfare from adverse effects of pollutants associated with energy systems. The goal of the Program is to assure the rapid development of domestic energy supplies in an environmentally-compatible manner by providing the necessary environmental data and control technology. Investigations include analyses of the transport of energy-related pollutants and their health and ecological effects; assessments of, and development of, control technologies for energy systems; and integrated assessments of a wide range of energy-related environmental issues.

EPA REVIEW NOTICE

This report has been reviewed by the participating Federal Agencies, and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the Government, nor does mention of trade names or commercial products constitute endorsement or recommendation for use.

This document is available to the public through the National Technical Information Service, Springfield, Virginia 22161.

EPA-600/7-78-189

September 1978

Analysis of Cascade Impactor Data for Calculating Particle Penetration

by

Phil A. Lawless

Research Triangle Institute
P. O. Box 12194
Research Triangle Park, North Carolina 27709

Contract No. 68-02-2612
Task No. 36
Program Element No. EHE624

EPA Project Officer: Leslie E. Sparks

Industrial Environmental Research Laboratory
Office of Energy, Minerals, and Industry
Research Triangle Park, NC 27711

Prepared for

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

ABSTRACT

The difficulties of analyzing cascade impactor data to obtain particle penetrations according to size are discussed. Several methods of analysis are considered and their merits weighed: interpolation, least-squares fitting, and spline fitting. The use of transforming functions prior to data fitting is also discussed. Recommendations for the use of the normal transformation and spline fitting method are made, and computer programs are provided to facilitate their use.

CONTENTS

FIGURES

ACKNOWLEDGMENT

The efforts of the Project Officer, Dr. L. E. Sparks, in the first stages of using the spline technique are gratefully acknowledged. He appreciated the power of the technique and stimulated the investigation.

1.0 Introduction to the Problem of Impactor Analysis

Multistage cascade inertial impactors are particle sizing devices which rely upon the inertial aerodynamic properties of particles of different diameters to separate them into distinct groups. By measuring the amount of particulate collected in each group, the size distribution of the sampled dust can be inferred. The widespread use of cascade impactors as particle sizing devices requires that a consistent reliable method for inferring the particle size distribution be used. This report addresses some of the problems and solutions encountered in the analysis of cascade impactor data.

1.1 Impactor Cut-points and the Cumulative Distribution

It is assumed for the purposes of this report that whatever type of cascade impactor is used, it has been adequately calibrated and carefully used to avoid the problems associated with such measurements. (See for example, References 1 and 2.) Each stage of an impactor has a characteristic diameter associated with it, depending on the operating conditions, called the cut diameter or cut-point which is the diameter particle collected with fifty percent efficiency. Ideally all particles of larger diameter would be collected with one hundred percent efficiency and none of the particles with smaller diameter would be collected on that stage. In practice, the collection efficiency curve is a moderately sharp function near the cut-point with substantial "wings" on either side of the cut-point. By knowing the precise nature of the collection efficiency curve, it should be possible to deconvolve the impactor stage weights to obtain a particle distribution. This approach would have to be specific to each impactor and could not reasonably be used unless each stage were calibrated for the flow conditions used. Generally, attempts to deconvolve real impactor data have not been successful.

Commonly, the impactor cut-points are assumed to represent infinitely sharp demarcations in the particle spectrum. This assumption will be carried on through this report.

Since each impactor stage separates the particle stream into fractions larger than the cut diameter and smaller than the cut diameter, the amount of dust on each stage represents the fraction of particles between adjacent cut diameters. If the total mass collected is determined as well as the mass on each stage, a cumulative fraction curve can be constructed which represents the cumulative particle size distribution. It is this cumulative distribution which the cascade impactor measures directly. The differential particle distribution, which is the derivative of the cumulative distribution, gives the abundance of particles for every diameter under consideration. Since control devices collect different diameter particles with varying degrees of efficiency, it is necessary to know both the inlet and outlet differential distributions in order to adequately evaluate the performance of control devices. A device may have an acceptable total collection efficiency, yet have so poor an efficiency for some particle diameters that remedial action is called for. Obtaining the differential distribution from the cumulative distribution is the heart of the impactor data reduction problem.

1.2 The Log-Normal Nature of the Distribution

If the particle size distribution from a single source, such as a boiler stack, is measured directly, it will be found to have a rather skewed distribution, with the particle frequency approaching zero at zero diameter and, for diameters above the mean diameter, approaching zero rather slowly. If the logarithm of particle diameter is used as the abscissa, rather than the particle diameter itself, the particle size distribution much more closely resembles the normal curve:

$$\phi\,(x) = \frac{1}{\sqrt{2\pi}}\; e^{-x^2/2}\;, \tag{1.1}$$

where $\phi\,(x)$ is the normally-distributed function of x. The resemblance is often so strong that the particles are said to have a log-normal size distribution.

Associated with the normal curve is the probability integral, defined as:

$$Q\,(x) = \int_{-\infty}^{x} \phi\,(t)\,dt\;, \tag{1.2}$$

where t is a variable of integration and ϕ is given by Equation 1.1. The probability integral corresponds to the cumulative size distribution measured by the cascade impactor. There are graph papers available with cumulative probability as one variable and logarithms as the other. The probability integral function, $Q(x)$, where x is a logarithmic variable, would plot as a straight line on such graph paper.

The cumulative fractions of cascade impactor particle measurements also generally exhibit a linear tread when plotted on such graph paper, and, in fact, a straight line plot is indicative of a true log-normal particle size distribution. A plot on log-normal (also called "log-probability") graph paper of a real impactor data set is shown in Figure 1.1.

Most real impactor data is only approximately log-normal and there are objections to considering them as such, particularly when the deviations become large. However, some data reduction schemes rely upon assuming a log-normal character for the cascade impactor data.

1.3 The Problem of Differentiating Sparse Data

Since it is the derivative of the cumulative distribution which provides the desired frequency distribution, a method of differentiating the data is needed. Unfortunately, there are no simple solutions to the problem. Direct approximation of the derivatives by finite differences has the disadvantages that the N data points produce only N-1 "derivatives" and that the derivatives so obtained are relatively crude approximations. Moreover, the derivative so calculated must be assigned to some particle diameter located between the two cut-points. (The well-known Mean Value Theorem states that the slope of the chord joining the end points of an interval is equal to the derivative of a function which passes through those end points at some point within the interval.) Commonly, this diameter is chosen to be the arithmetic mean of the logarithms of the cut-points, or equivalently, the geometric mean of the cut-points themselves. Thus, the points at which the "derivatives" are evaluated depend on the cut-points and may not be suitably located to compare the results from different impactors, for example, or for inlet and outlet samples from a control device. Nor is there any guarantee that the derivatives are correct for these particle diameters.

The general recommendation for differentiation of numerical data, and especially so when the data are sparse, is to fit a differentiable function to the data and obtain the derivatives analytically. Methods of doing this will be addressed in Section 3.

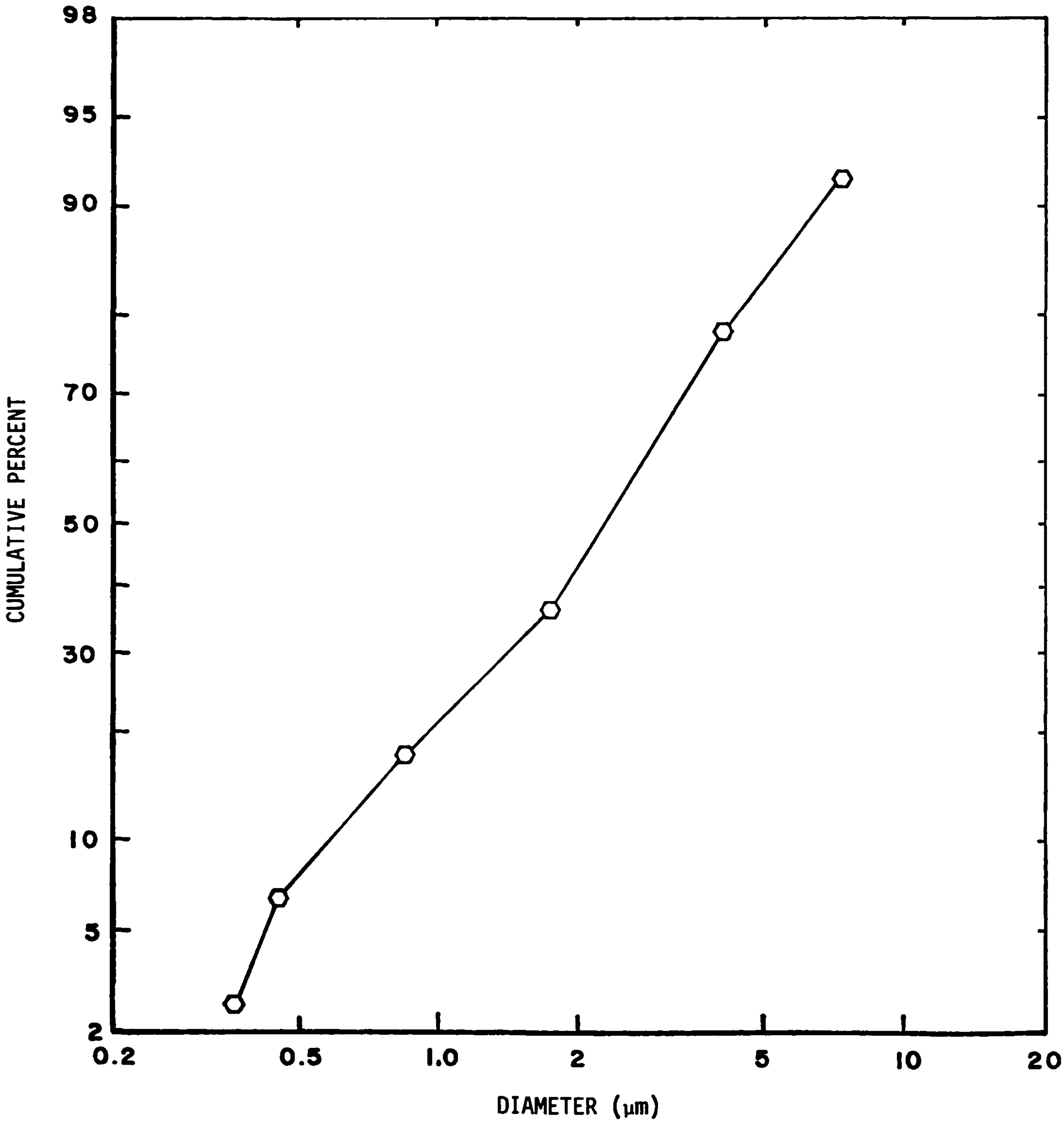

Figure 1.1. Real impactor data plotted on log-normal probability paper.

2.0 Recommendations

Cascade impactor cumulative distribution data can be analyzed in many ways to obtain a frequency distribution. Some of these ways have been investigated in this study and the following recommendations can now be made.

1. Prior to any curve fitting technique, the impactor data should be log-normal transformed: that is, the logarithm of the cut-point diameters whould be used instead of the diameters themselves and the normal transform of the cumulative probabilities should be used instead of the cumulative probabilities themselves. With this transformation, all of the techniques discussed in this study will fit a true log-normal distribution exactly, and the inherent errors in fitting near-log-normal distributions are substantially reduced. A calculator algorithm and a computer program for the normal transformation are provided in this report.
2. The natural cubic spline curve fitting method should be used to fit the transformed data. The properties of such splines are well-defined mathematically; these splines are the smoothest curves that can be passed through all the data points, and they are capable of fitting non-log-normal distributions with small error. A computer program for calculating a spline passing through given data points is provided in this report.
3. In cases where the impactor data are known (or suspected) to contain substantial random errors, a polynomial least-squares fit of the transformed data is recommended, with the polynomial degree selected judiciously. The variance of the data from the fitting polynomial should be calculated and used to establish confidence limits on the resulting frequency distribution. Least-squares fitting is <u>not</u> recommended in cases where the random error is small, because the spline fit will make better use of the data in such cases.

3.0 Approaches to Fitting Functions

3.1 Interpolating Polynomials

The simplest function which can pass through N data points and is differentiable is a polynomial of degree N-1. Such a polynomial exists and is unique (Reference 3). It is possible that the degree of the interpolating polynomial is actually less than N-1.

This polynomial can be written in a simple way: consider three functional values f_1, f_2, and f_3 at the respective points x_1, x_2, and x_3. The interpolating polynomial I (x) is:

$$I(x) = f_1 \frac{(x-x_2)(x-x_3)}{(x_1-x_2)(x_1-x_3)} + f_2 \frac{(x-x_1)(x-x_3)}{(x_2-x_1)(x_2-x_3)} + f_3 \frac{(x-x_1)(x-x_2)}{(x_3-x_1)(x_3-x_2)} \quad . \tag{3.1}$$

This reproduces the functional values exactly at their respective points. For more data points, a correspondingly higher degree polynomial can be generated.

The problem with using the interpolating polynomial for reducing impactor data is that it is generally a fifth degree or higher polynomial and, except at the cut-points, the value of the polynomial depends on the cumulative fractions at all of the cut-points. The first derivative will generally have 3 or more peaks, (the second derivative is a cubic or higher polynomial). Moreover, all the derivatives will be sensitive to slight changes in the impactor data, with the result that repeated measurements on the same particle size distribution may exhibit wide variations in the calculated distributions due to small perturbations in the stage weights.

It is possible to use piece wise interpolation of a few data points at a time in order to reduce the degree of the polynomial and the sensitivity of the derivative to small perturbations in the impactor data. Two such methods have been described recently (References 4 and 5). The problem with this approach is that generally the first (and higher) derivatives are discontinuous. Since this leads to unphysical particle size distributions, efforts must be made to smooth out the discontinuities.

In Reference 4, this smoothing is done by convolving the piecewise interpolating polynomials with a smoothing function, essentially performing a running average whose first derivative is a continuous function. The choice of the smoothing function is rather arbitrary, though the function used is reasonable on physical grounds. It appears that this particular smoothing function would allow discontinuities in the second derivative, equivalent to abrupt changes in slope of the particle size distribution. A higher order smoothing function could be chosen to produce a continuous second derivative as well.

In Reference 5, the smoothing procedure produces continuous first derivatives at the cost of allowing the interpolation to miss the data points. The interpolation passes close to, but not necessarily through the impactor cumulative fraction points. The method used also produces discontinuities in the second derivative of the interpolating function, in a way that cannot be easily corrected for. However, in this method, the logarithms of the cumulative fractions are the data points fitted, which reduces the magnitude of the discontinuities in the second derivatives.

One difficulty that applies to all interpolation schemes is that the interpolation is valid, or has meaning, only over the range of the data. Any effort to extrapolate the fitting fractions beyond the first and last data points is not only unjustified, but generally leads to poor results anyway. This is particularly troublesome when inlet and outlet impactor samples are obtained from a control device to assess its graded penetration, since the valid range will only include those data points common to both samples. For this reason, it is worthwhile to try to match the impactor cut-points at inlet and outlet.

3.2 The Method of Least-Squares Fitting

In contrast to interpolation methods, which effectively consider the impactor data as completely reliable, least-squares fitting methods assume that there are inherent errors in the data which partially conceal the true nature of the functional dependence of the variables. The method of least-squares assumes a functional dependence, either from theoretical considerations, educated guesses, or reasons of simplicity, and then finds the "best" function which fits the data. "Best" is defined in terms of the deviations of the data from the functional values, with the quantity

$$S^2 = \sum (y_i - f(x_i))^2 \qquad (3.2)$$

where y_i is the ordinate corresponding to abscissa x_i, being minimized for the "best" function f. It f is a linear function of the coefficients of powers of x, then a system of linear equations results which has a unique solution (Reference 3). This means that any functional dependence which can be expressed as a polynomial in x can be easily used with the method of least-squares.

Often the function used is in terms of variables transformed from the actual data. For instance, the logarithm of the diameter may be used as the independent variable, rather than the diameter itself. Such transformations may greatly simplify the required functional form and the method for solving the resulting equations. In particular, we will discuss the normal transformation in some detail later, by which log-normal functions are transformed to straight line functions.

If impactor data fall on a nearly straight line when plotted on log-probability graph paper, it may well be said that they should obey a linear relationship in those coordinates, and a linear least-squares fit might be used. If the data show an obvious, but gentle curvature in those coordinates, a quadratic least-squares fit, using powers of x up to the second, might be used. Higher powers could also be attempted. If there are N data points, then a least-squares fit with an N-1 degree polynomial will give a perfect fit and will in fact produce the interpolating polynomial for that data.

The choice of functional dependence is a matter for judgment, since there is no strong theoretical reason that the particle size distribution should indeed be log-normal. Since the method of least-squares is a method of accounting for random measurement errors, it is reasonable to use it only if the errors associated with the measurements are at least of the order of magnitude of the deviations of the data from the fitted function. The method cannot legitimately be used to remove "errors" that are in fact really present in the measured quantities.

If a functional dependence is assumed for the data, then there is some justification for extrapolating that dependence into the regions beyond the data at either end of the size distribution. This extrapolation must be done with due regard to the nature of the function; that is, a linear function extrapolates well, while higher order polynomial functions may quickly reach unphysical regions. Extrapolation is one of the useful properties of least-squares methods, particularly so with impactor data, where inlet and outlet samples may not have good overlap of cut-points.

The second major advantage of the least-squares method is that it provides an intrinsic measure of the total random error associated with the fit. If the deviation of the fitted function $f(x_i)$ from the measured datum y_i is defined as δ_i, then the variance of the fit is given by

$$\sigma^2 = (\textstyle\sum_i \delta_i^2)/(N-M-1), \tag{3.3}$$

where N is the number of data points and M is the degree of the fitting polynomial. It can be seen that if the number of data points equals M-1, the variance is undefined because all the deviations are zero. Thus, no measure of the error can be obtained for this case, which is equivalent to the interpolating polynomial.

The variance is a statistical measure of the random error in the fit and provides information for determining the overall confidence level that can be assigned to the data. A strategy for using the variance as a fitting parameter would be to fit successively higher degree polynomials to the data until the variance was minimized. A minimum usually exists because the denominator in Equation 3.3 approaches zero as the degree of the polynomial approaches N-1.

In summary, the least-squares method provides a statistical measure of the error in the data and the ability to extrapolate the fitted function, but at the cost of assuming a functional dependence for the data. As an aside, the method of Tschebyschev fitting is similar to the method of least-squares in that a functional dependence must be assumed. The procedure is different in that the maximum (absolute value) deviation of the data from the fitting function is minimized. The Tschebyschev method would be more applicable in situations where the randown error in the data is known to be small, but for which a certain functional form is desired (Reference 3).

3.3 Spline Methods of Analysis

A draftsman's spline is a thin flexible strip that can be made to conform to plotted data points by means of weighted pivots. The curve so constructed passes through all the data points and has continuous first and second derivatives. Mathematical splines are a class of polynomial functions with properties that are generalizations of those of the draftsman's spline.

The particular spline of interest in the analysis of impactor data is a third order natural spline. It consists of a series of cubic polynomials, joined at the data points with continuous first and second derivatives and with the second derivatives at the first and last data points equal to zero. Outside the data range, the cubic polynomial is reduced to a linear polynomial, so that the natural spline has some utility in situations calling for extrapolation.

Splines have well-defined mathematical properties, and odd-order splines have several special properties that are of interest in the impactor analysis problem. These are:

1. If F(x) is a continuous function, with continuous first and second derivatives, then the natural spline of third order, S(x), which attains the values $F(x_i)$ at points $a=x_1, < x_2 < \ldots x_i < x_N=b$ minimizes the integral

$$\int_a^b (F''(x)-S''(x))^2\, dx. \qquad \text{(Reference 6)}$$

2. Among the functions y(x) which have continuous first and second derivatives and interpolate to F(x) at the junction points, the third order natural spline minimizes

$$\int_a^b y''(x)^2\, dx. \qquad \text{(Reference 6)}$$

3. If Z(x), which has continuous first and second derivatives, does not interpolate to F(x) at all of the junction points, x_i, then the integral $\int_a^b (F''(x) - Z''(x))^2\, dx$ can be reduced by adding the third order spline W(x) such that Z+W interpolates all the function points $F(x_i)$. (Reference 6)

4. If the number of interpolating points in the inteval [a,b] is increased so that the maximum separation of any two adajcent points tends toward zero, then the spline and its first derivative, $S(x)$ and $S'(x)$, converge uniformly to $F(x)$ and $F'(x)$. (Reference 6)

The first and third properties describe the best approximation properties of the spline. The second property describes the "smoothness" of the spline curve itself, and the fourth property describes the behavior of the spline function and its first derivative at points away from the junction points.

These properties can be extended to higher order splines and higher order derivatives (Reference 7), but that is not of further interest at this time.

A fifth property that the third order natural spline has that is less mathematically defined is its insensitivity to variations at a single point. Changes at a single point in the value of the function being fitted changes the value of the spline function only at that point, changes the first spline derivatives mainly at the point and its two nearest neighbors, and changes the second derivatives of the spline mainly at the point and the two adjacent points on either side. Furthermore, the changes are largest at the central point, diminishing with distance from the point. Thus, an error in the original data does not propagate throughout the entire fitted region, as can happen with the interpolating polynomial.

The third order natural spline is calculated from the following set of equations.

If there are N data points with abscissas at $x_1 < \ldots x_k \ldots < x_N$ and values $F(x_k)$, and if $l_k = x_k - x_{k-1}$ $(k = 2,N)$, then the spline second derivatives $M_k = S''(x_k)$ are given by

$$\frac{l_k}{6} M_{k-1} + \frac{l_k + l_{k+1}}{3} M_k + \frac{l_{k+1}}{6} M_{k+1} \quad \frac{F(x_{k+1})-F(x_k)}{l_{k+1}} - \frac{F(x_k)-F(x_{k-1})}{l_k} \qquad (3.4)$$

with $M_1 = M_N = 0$.

This is a tridiagonal set of equations which can be solved easily by several methods. A program using successive overrelaxation is shown in the

Appendices. A program for programmable calculators is given in Reference 11. The value of the spline and its derivatives are determined by integration of these second derivatives from the junction points to the points of interest.

Because of its excellent approximating properties and relatively simple method of calculation, the spline fit is the best method for analyzing impactor data under any conditions where the random error is known to be small.

4.0 The Normal Transformation

All of the fitting methods described so far can be applied to any set of data, whether from impactors or not. There is one type of data for which these methods all exhibit intrinsic errors (and in which we are interested): log-normal distributions. We have stated that a log-normal distribution is one in which the logarithm of the independent variable is normally distributed. We now write it explicitly.

If y = log (d) and the cumulative function Q (y) is given by

$$Q(y) = \int_{-\infty}^{y} \frac{e^{-t^2/2}}{\sqrt{2\pi}}\, dt, \tag{4.1}$$

then Q is log-normally distributed in d. The variable t may be related to the mean value of log(d), m, and its standard deviation, σ, by the equations:

$$x = \frac{y-m}{\sigma}\ , \tag{4.2}$$

and

$$Q(x) = \int_{-\infty}^{x} \frac{e^{-t^2/2}}{\sigma\sqrt{2\pi}}\ . \tag{4.3}$$

Now, the point is that this function, Q, is linear near $x = 0$, but exhibits strong curvature (with infinitely many derivatives) for x away from zero. Thus, any fitting scheme based on polynomial approximations is bound to contain intrinsic errors in attempting to fit this function. Some analysis schemes, that of Reference 5 for example, reduce the errors by fitting log Q instead of Q itself; but the errors are still present. The spline can deliver an excellent fit to Q if the points of junction are close enough, but begins to deteriorate rapidly if they are too far apart.

Because the log-normal distribution is often very close to impactor-measured size disbributions, it would be worthwhile to require any fitting scheme to be able to reproduce the log-normal distribution as closely as possible under all conditions. Fortunately, all of the methods discussed before are capable of fitting the distribution exactly by use of the normal transformation.

The normal transformation is defined as the inverse function to the probability integral Q. In other words,

$$N(Q(y)) = y \ . \tag{4.4}$$

Whereas Q(y) ranges from 0 to 1, N(Q(y)) ranges from $-\infty$ to $+\infty$ in a one-to-one correspondence with y, i.e. it is a linear function. The normal transformation is equivalent, then, to plotting on probability paper, and any scheme that can fit a straight line can fit the transformed variable. The inverse function N is not analytic, but can be approximated by a polynomial expression:

$$-N(Q) = -y = t - \frac{c_0 + c_1 t + c_2 t^2}{1 + d_1 t + d_2 t^2 + d_3 t^3} \tag{4.5}$$

where $$t = \sqrt{\log\left(\frac{1}{Q^2}\right)} \qquad 0 \leq Q \leq .5$$

and
$$c_0 = 2.515517$$
$$c_1 = 0.802853$$
$$c_2 = 0.010328$$
$$d_1 = 1.432788$$
$$d_2 = 0.189269$$
$$d_3 = 0.001308$$

with an absolute error less than 4.5×10^{-4} (Reference 8). Since this is valid for Q over only half its range, the relation is needed that:

$$Q(-x) = 1 - Q(x) \ . \tag{4.6}$$

This function is easily evaluated on a pocket calculator, and is even available as a stored function on one model (Hewlett-Packard HP 32E). It is shown as a computer subroutine in the Appendices. A calculator program for the function is given in Reference 11.

With the use of this transformation, linear or higher order least-squares methods can be used to fit nearly log-normal data, and interpolating polynomials or splines can give good results with even widely spaced data points. A discussion of some of these results will be deferred to a later section.

Since it is the particle frequency distribution that is desired from the cumulative data, the derivative of the fit to the normal transfored data must itself be transformed back to the linear space of the impactor data. That is, if the transformed function F is given by

$$F(x) = N(f(n)) , \tag{4.7}$$

then the derivative of the function f is given by

$$f'(x) = e^{-F(x)^2/2} \cdot F'(x) . \tag{4.8}$$

Since all of the methods discussed give F(x) as well as F'(x), the inverse transformation is easy to evaluate. If the cumulative function f is also desired, it must be evaluated from the probability integral (Equation 4.1) with y = F(x). Polynomial approximations to the probability integral are available in Reference 8. Note that if F'(x) is constant, then f'(x) is a normally distributed function. In particular, for the natural spline fit, the extrapolation regions outside the range of the data for which the spline is of degree one represent normally distributed tails which join smoothly with the spline interpolation. This does not justify the extrapolation, but at least it guarantees a well-behaved extrapolation.

5.0 Results of Spline Fits of Test Data

The analysis of impactor data by splines was tested both with and without the normal transformation. Tests were performed on the ability of the analysis to reproduce the test frequency distributions for a single impactor and to reproduce the test penetrations for inlet and outlet impactors. The errors produced in fitting inlet and outlet impactor data were not always independent with the results that the propagated error in penetration could be smaller than the errors in the individual fits.

5.1 Method of Generating Test Data

Log-normal frequency distributions were specified by mass median diameters (MMD) and geometric standard deviations (σ_g). The logarithm of σ_g is the standard deviation that appears in the normal distribution (Equations 4.2 and 4.3). At a series of abscissas corresponding to the logarithms of particle diameters in the range of 0.2 to 20 μm, the frequency distribution appropriate to the MMD and σ_g was calculated by polynomial approximations (Reference 8). This formed the "true" data to which could be compared to the results of the impactor analysis. For testing the spline method without the normal transformation, an inlet distribution and an outlet distribution were used. The ratio of outlet to inlet at each diameter gave an effective penetration for that diameter.

These distributions were "sampled" by ideal impactors in the following way. Typical impactor cut points were chosen (independently) for the inlet and outlet samples. Then the cumulative fractions of the appropriate log-normal distribution were calculated for these cut points, one set for the inlet and one set for the outlet. Generally, six or seven cut-points were chosen.

Then the spline method was used to obtain a fit to each cumulative distribution, and the spline derivatives were evaluated at each diameter for which the "true" inlet or outlet was known. These derivatives were compared to the corresponding distribution values and the ratio of outlet to inlet at each diameter was compared to the "true" penetration.

When the normal transformation was used, a different set of data was required, since log-normal data would be transformed to a linear function, which the spline could fit exactly. For this case, two log-normal distributions were added together at the inlet. The two distributions could have their MMD's and σ_g's set separately and the ratio of their amplitudes was adjustable. The outlet distribution was allowed to remain a single log-normal distribution. For the normal-transformed data, the error in the penetration would then be in proportion to the error in the inlet distribution.

Two measures of error were used. The first was the root mean square relative deviation, defined as:

$$e = \left(\frac{1}{N}\sum_i\left(\frac{f_{true} - f_{calc}}{f_{true}}\right)^2\right)^{1/2} . \qquad (5.1)$$

The function f could present either a distribution function or a penetration calculation, for which true values were known. The number of points averaged was variable, usually ten to fifteen. The second measure of error was the relative maximum error, defined as:

$$\delta = \frac{\max\left(|f_{true} - f_{calc}|\right)}{\max\left(f_{true}\right)} . \qquad (5.2)$$

This measure took the maximum deviation and related it to the maximum value of the function (which was always positive).

The RMS error e gives a measure of the overall discrepancy between the true values and the fitted curve while the relative maximum error, δ, gives an error that is not exceeded in the range of the test. Both types of measure are useful, and in general, δ was of the order of twice e.

The geometric standard deviation of a log-normal distribution is defined as the ratio of particle sizes that are one standard deviation apart; for instance, the ratio of the size corresponding to a cumulative fraction of 0.841 to the size corresponding to a cumulative fraction of 0.500. Since over 68 percent of the total mass of the particles are included in the interval from one standard deviation below the MMD (MMD/σ_g) to one standard deviation above the mean (MMD x σ_g), the geometric standard deviation provides a quick measure of the sharpness of the distribution. A σ_g of 4, for example, means the distribution is spread over a substantial range from one-fourth the MMD to four times the MMD.

5.2 Results of Spline Fits to Untransformed Log-Normal Distributions

The spline was capable of fitting the untransformed log-normal distributions quite well, provided that the cut-points were suitably placed. The magnitude of errors in the penetration was about five percent if both the inlet and outlet impactor cut-points were well-distributed over the central part of the curve (MMD/σ_g to MMD x σ_g). As fewer impactor cut-points were placed in this region, so the error rose, reaching values of sixty percent or greater. If a distribution was broad, the impactor cut-points could be far apart without serious effect. If the distribution were sharp, the cut-points had to be closely spaced to keep the error low.

For these cases, a special advantage was found in matching the inlet and outlet impactor cut-points. Apparently, the errors in each fit compensated each other and the error (e) in penetration could be as low as one percent. Changing the location of a single cut-point by as little as twenty-five percent raised the RMS error to the level of five percent.

It is rarely possible to match inlet and outlet impactors cut-points when testing a control device, and it is also difficult to make sure the cut-points are well-distributed over each distribution and still maintain sufficient overlap to calculate the penetration over a wide range. It is also not reasonable to try to use the spline extrapolation in the untransformed case, because the extrapolations are constant values for the distribution, a very unphysical result. These problems lead us to look further, specifically to the transformed case.

5.3 Results of Spline Fits to Double Mode Transformed Distributions

The first tests of the normal-transformed spline fits used relatively broad distributions (MMD_1 = 20, σ_g = 4.5; MMD_2 = 3.0, σ_g = 2-3) in various ratios. The errors (e) in these cases were all less than one percent, often less than one-tenth percent. In those cases where the impactor cut-points were well-distributed over the distribution, the untransformed spline also performed creditably ($e < 0.05$). However, the transformed spline showed no sensitivity to choice of impactor cut-points in these tests. There were no significant differences between matched or unmatched cut-points, or between well-distributed and poorly distributed cut-points. No comparisons of the extrapolated fits were attempted for these tests. Even mixtures of positive and negative distributions were fitted well, as long as the total frequency distribution remained positive at all points.

Then a group of tests were performed combining a broad distribution (MMD = 10, σ_g = 3.5) with a very narrow distribution (MMD = 3, σ_g = 1.05), using normal cut-points. Very high errors resulted, with e's in the range of forty to two hundred percent, in proportion to the ratio of the sharper distribution to the broader distribution. In effect, the spline was completely missing the sharp peak in the distribution, because it fell almost totally between the impactor cut-points. The effect of this on the frequency distribution is shown in Figure 5.1. In this example, the total mass in the sharp peak is one-tenth the total mass in the main peak and produced a thirty-five percent error. If more mass is placed in the sharp peak, the error is greater, and if less mass is placed in the sharp peak, the error is less. The spacing of the cut-points does not have a significant effect in this case, as long as the sharp peak affects no more than one cut-point in the cumulative distribution.

This test provides a measure of the sensitivity of the spline technique to errors in the data points. If the distribution being fitted is truly log-normal, except that one data point is in error, then the spline will produce a distribution that is in error from the log-normal distribution. Based on the results of this series of tests, if x_i is the mass error at a single data point in the cumulative curve divided by the total mass of the distribution, then the RMS relative error, e_i, is given approximately by

$$e_i = 6.7\ x_i \tag{5.4}$$

This error holds mainly in the neighborhood of the erroneous point, and decreases to near zero beyond two cut-points away from the erroneous point. The effect of errors in all the data points could be approximated by summing the squares of the errors in the neighborhood of each point:

$$e_i^2 = \sum_{j = i-2}^{i+2} e_j^2 \tag{5.5}$$

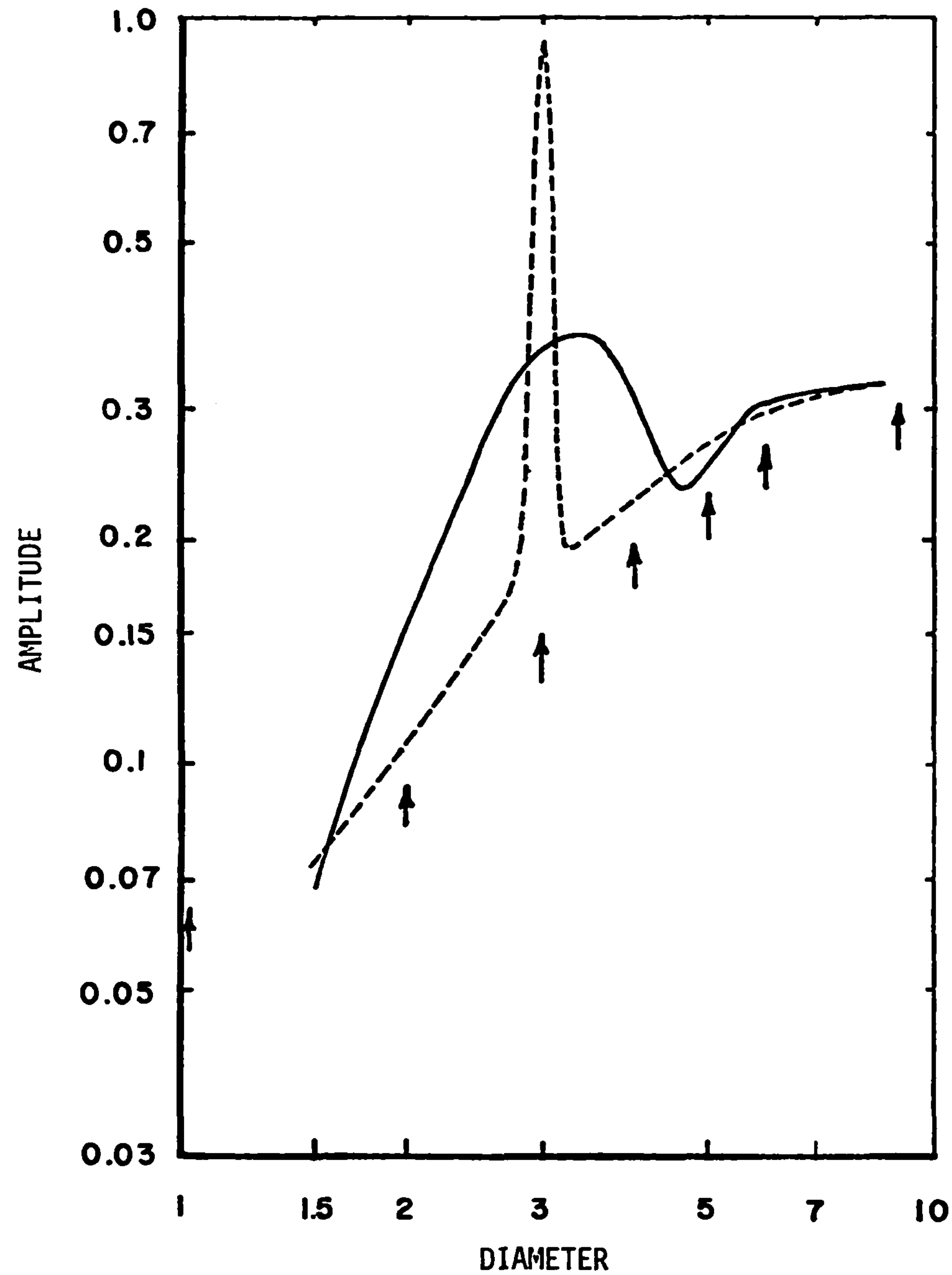

Figure 5.1. Spline fit of a single narrow peak on a broad peak. Dashed line is the true distribution (broad peak: MMD = 10, σg = 3; sharp peak: MMD = 3, σg = 1.05). Solid line is the spline curve. Arrows indicate location of impactor cut-points.

The usefulness of this error estimate is somewhat limited because of its empirical nature, but it does provide a method of assigning error bars to points in the frequency distribution if some estimate of the errors in the data can be made. For example, if the weighing error for each stage were 0.1 mg and the total mass collected were 20 mg, then the x_i's for each cut-point would be 0.005 and the overall error (Equation 5.5) at each cut-point would be 0.075 for interior cut-points (those located two or more points from the end points) and would decrease to approximately 0.035 (Equation 5.4) at the ends. These are very rough estimates based on the examination of limited data.

More precise error limits can be established from the mathematical properties of the spline, provided that a bound on the magnitude of the fourth derivative of the cumulative function can be estimated (Reference 10). There is no physical basis for doing this, and some arbitrary "smoothness" criterion would have to be used.

The final group of test data was aimed at establishing how narrow the secondary mode could be and still be resolved. It was quickly determined that if the distribution mean was no more than one standard deviation away from the second nearest cut-point, the RMS error was acceptably small. In other words, a secondary mode should be centered between cut-points and span them comfortably. Under such conditions, a narrow secondary peak in the distribution could have a peak amplitude twice that of the main broad peak with an RMS of the order of five percent. Smaller peak amplitudes in the secondary peak reduce the RMS error considerably, to less than one percent at an amplitude ratio of one to one.

Again, these observations are based on limited tests, but in general, a secondary peak that spans two or three cut-points will probably be accurately resolved from the data, assuming the data itself is free from error.

It should be noted that, in this section, we have referred to the RMS error in the particle size distribution, rather than in the penetration. The reason for this is that a constant log-normal distribution was assumed for the outlet distribution, which was fit exactly under the normal transformation. Therefore, no compensating errors could occur, and the error in the distribution is as good an indication of the fit as is the error in the penetration.

5.4 Spline Fits of Real Data

As a final demonstration of the curve fitting results, several sets of data for inlet and outlet impactor samples of an operating electrostatic precipitator (the EPA-IERL Pilot Scale Precipitator) are presented. Figures 5.2 through 5.6 are concerned with a single day's results while Figures 5.7 through 5.11 cover an average of six inlet and six outlet samples. (The impactor flow conditions were stable enough to allow direct averaging of the stage weights.)

In each group a comparison is shown between the results obtained with a least-squares fit of the log-normal-transformed data and the results with a natural cubic spline fit of the transformed data. In some cases, a linear least-squares fit gave a correlation coefficient just as good as the quadratic least-squares fit, and so the linear fit was used for simplicity. However, in most cases, the quadratic fit gives a substantially better correlation coefficient and a visually better fit of the data.

Figure 5.2 shows the inlet cumulative distribution, represented by the open circles, the fitted spline function passing through the points, and the quadratic least-squares line passing near the points. It can be seen that both fits represent the data well, except that the quadratic fit does poorly at the smallest particle sizes. If this were an isolated set of data, one might be inclined to use the quadratic fit and attribute the deviations to error in the data. However, the behavior of the data at the small sizes is consistent from run to tun, or in averages of many runs, indicating that random error cannot account for all the deviation.

The derivatives of the cumulative function fits are shown in Figure 5.3, with arrows indicating the cut-point diameters. The spline fit indicates a secondary peak in the distribution which the least-squares curve completely ignores. By the criteria of the preceding section, this secondary peak should be accurately represented because the data points are well-distributed over it. Whether the peak is actually in the particle distribution or represents an anomalous response of the impactor is a question that cannot be answered here.

Figures 5.4 and 5.5 show the cumulative and frequency distribution for the outlet impactor sample. Again the secondary peak is present and well-resolved in the spline fit.

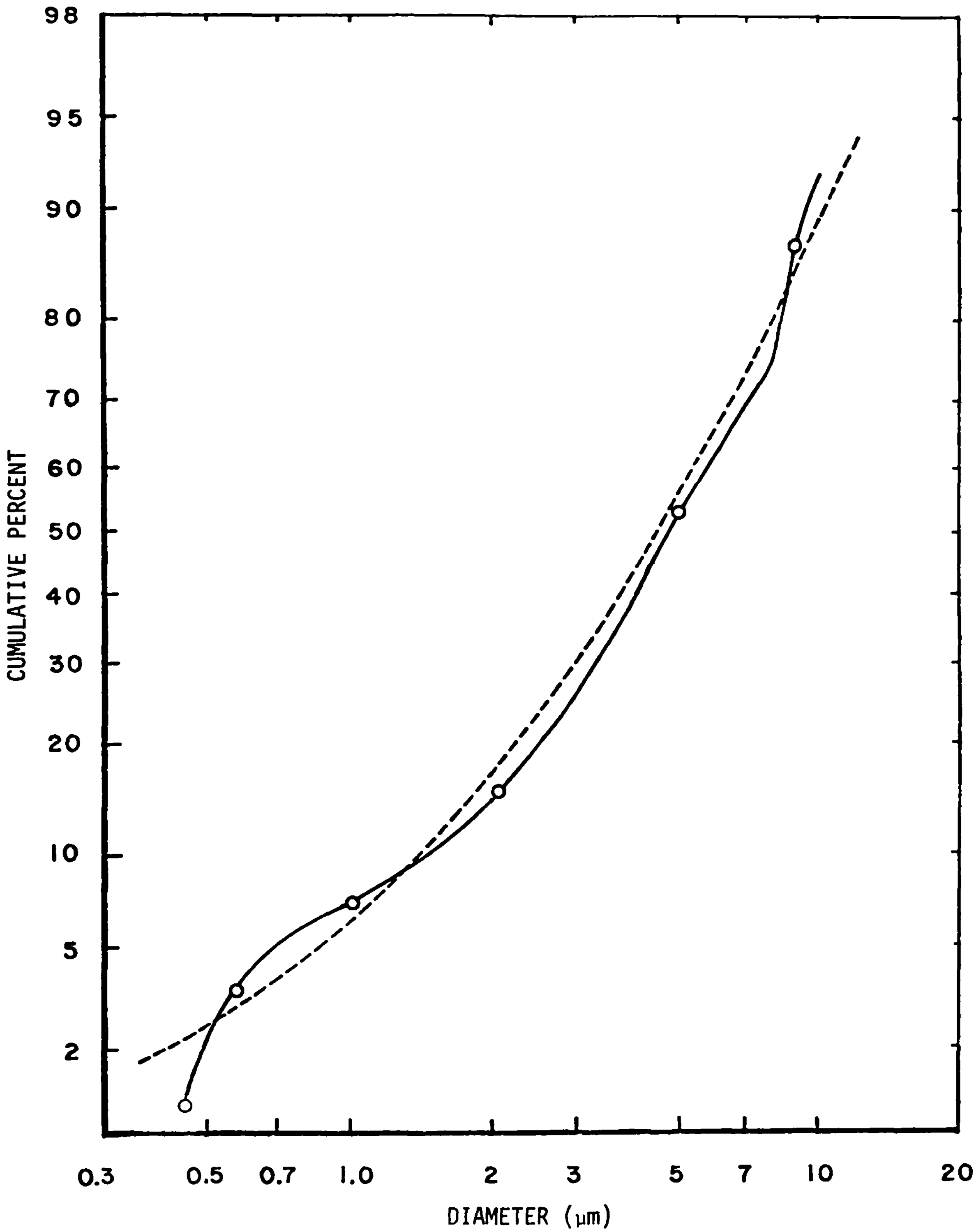

Figure 5.2. Inlet cumulative distribution for a single run. The solid curve is the spline fit passing through the data points (open circles) and the dashed line is the quadratic least-squares fitting curve.

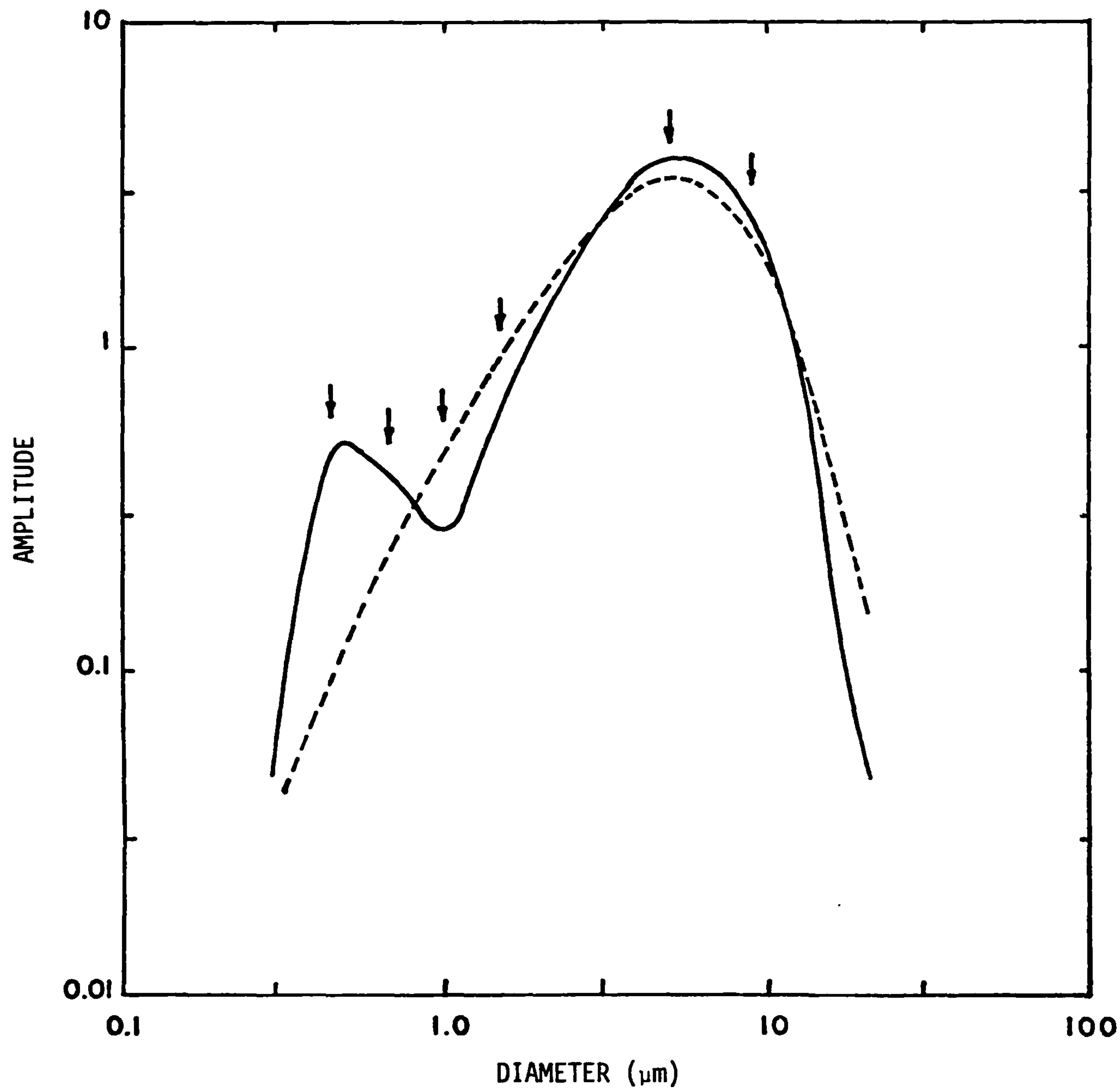

Figure 5.3. Inlet frequency distribution from Figure 5.2. The solid line is the derivative of the spline fit curve and the dashed line is the derivative of the least-squares curve. The arrows indicate the positions of the original data points.

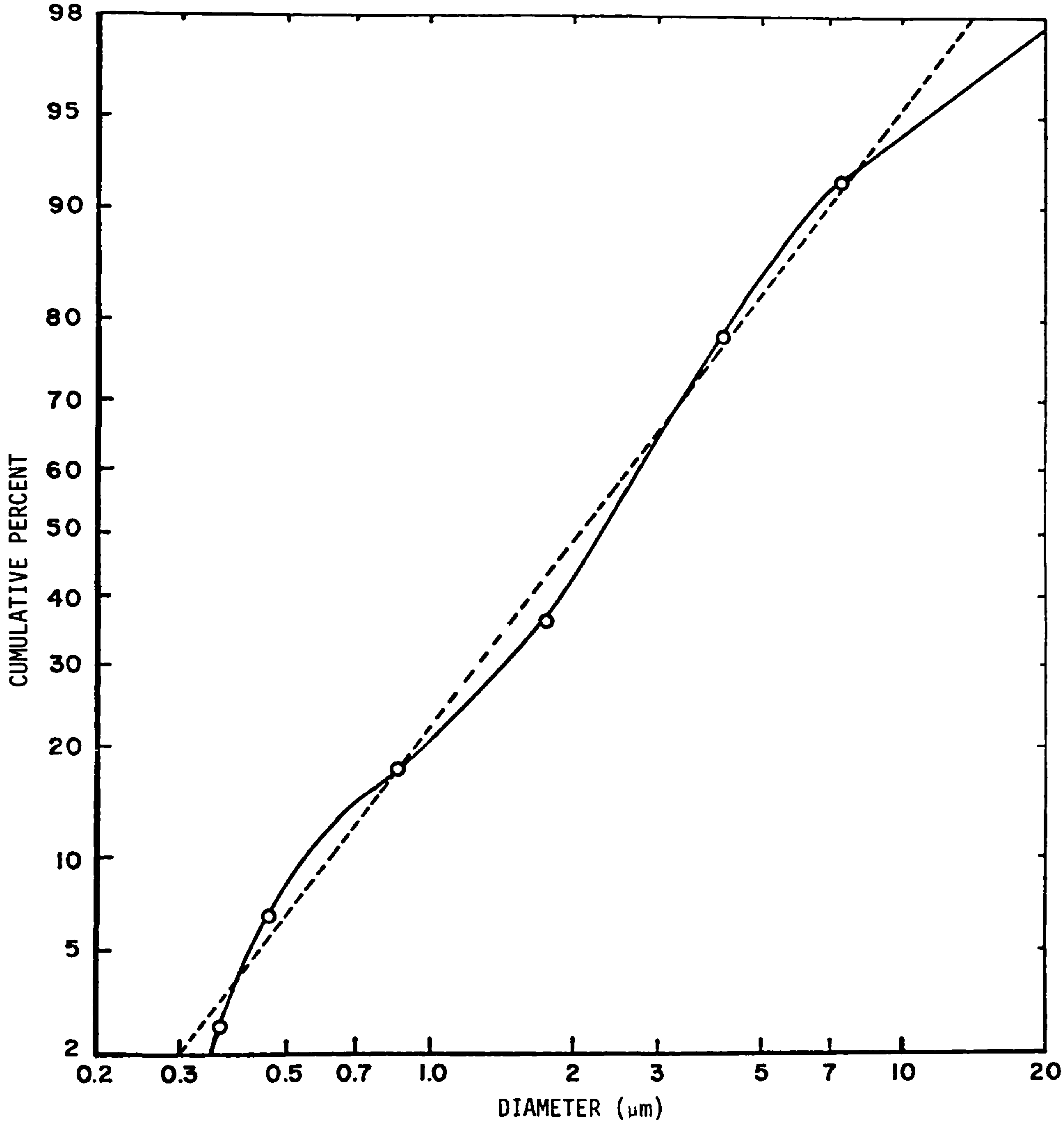

Figure 5.4. Outlet size distribution for a single run. The solid curve is the spline fit and the dashed line is a linear least-squares fit of the data.

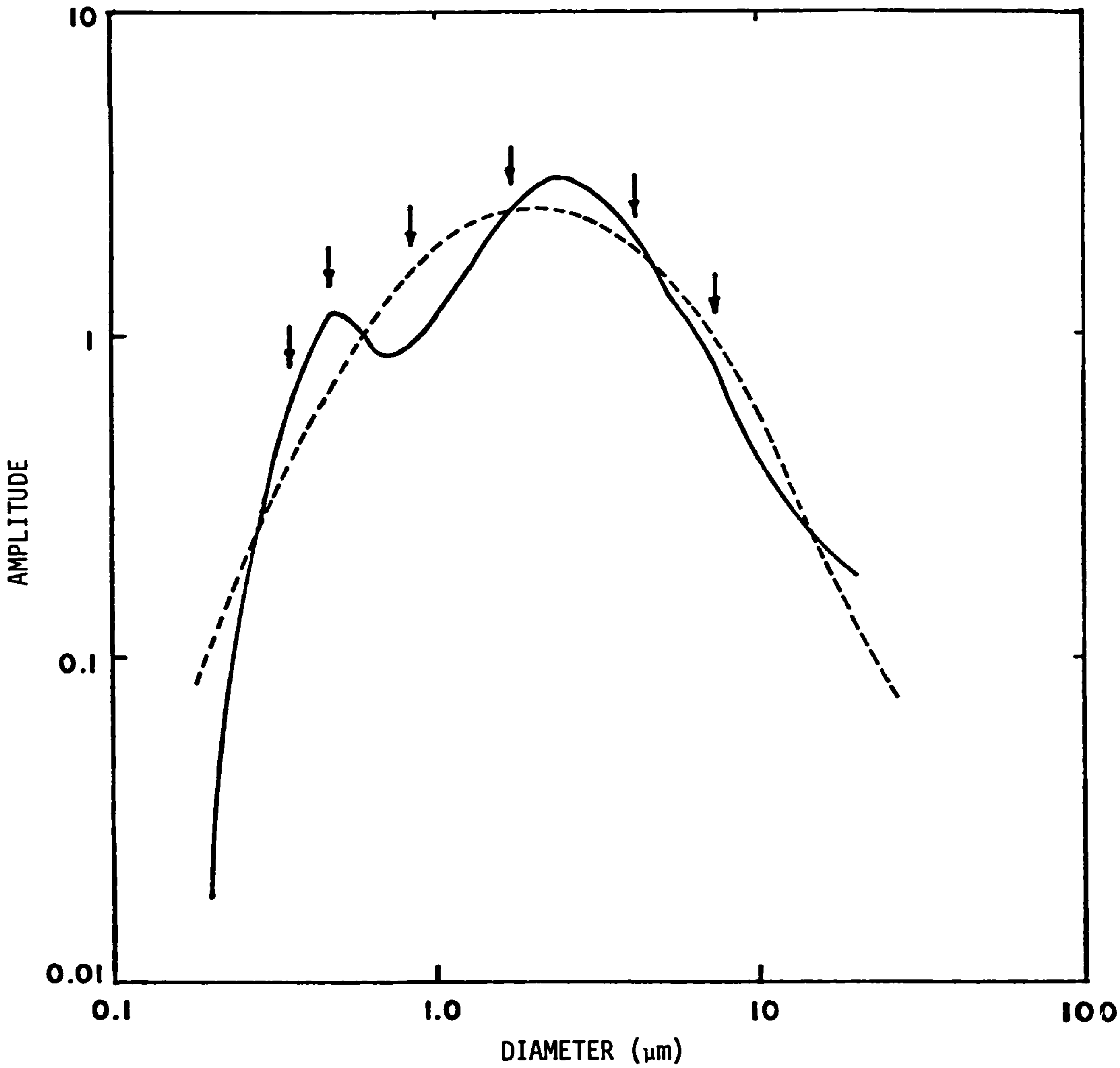

Figure 5.5. Outlet frequency distribution from Figure 5.4. The solid line is the spline curve derivative and the dashed line is the least-squares curve derivative. The arrows indicate the positions of the data points.

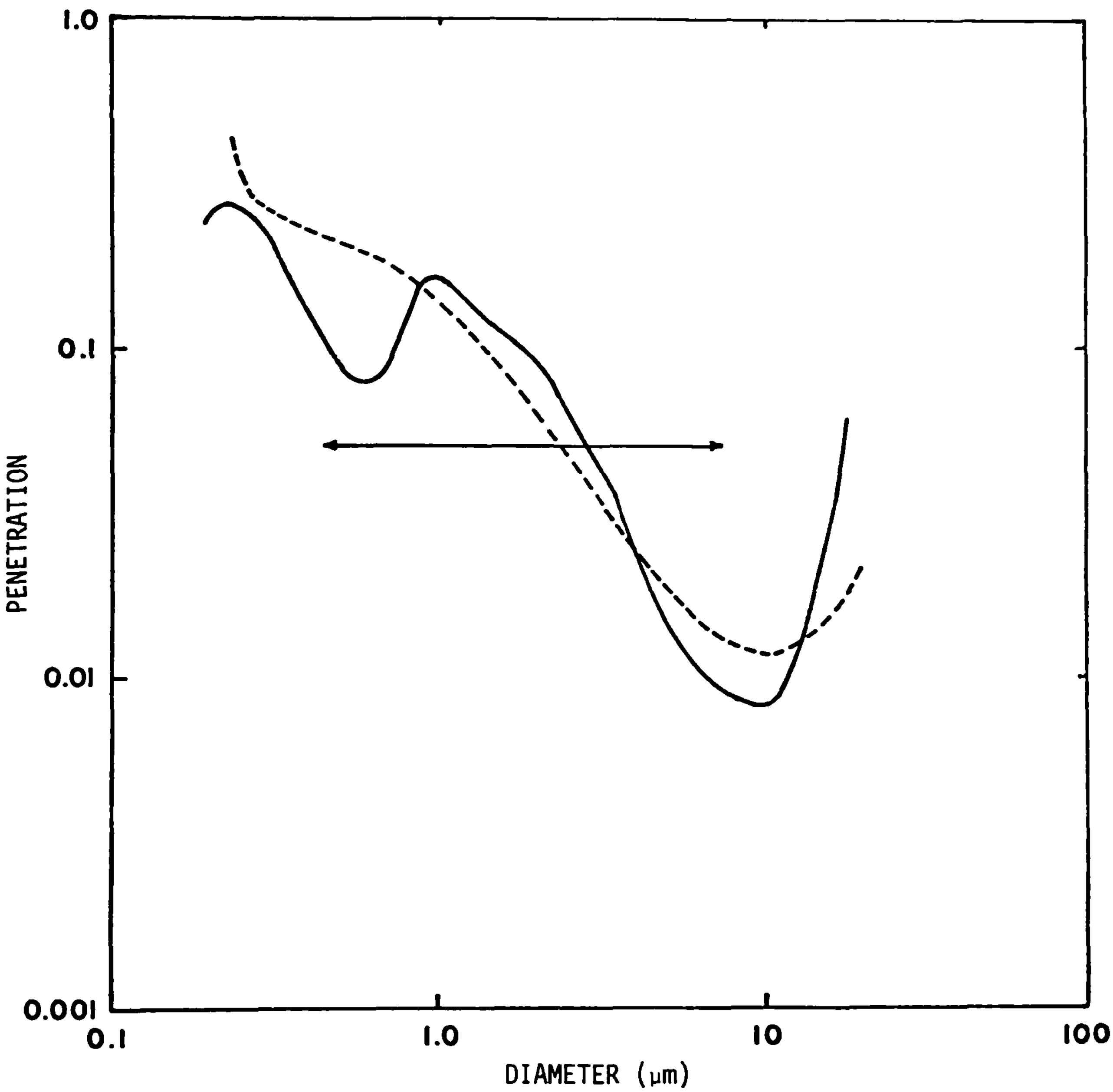

Figure 5.6. The penetration curve for a single run. The solid curve is the ratio of outlet to inlet spline derivatives multiplied by the ratio of dust concentrations at the outlet to the inlet. The dashed curve is the corresponding representation for the least-squares fits. The horizontal arrow shows the range of data common to both inlet and outlet samples.

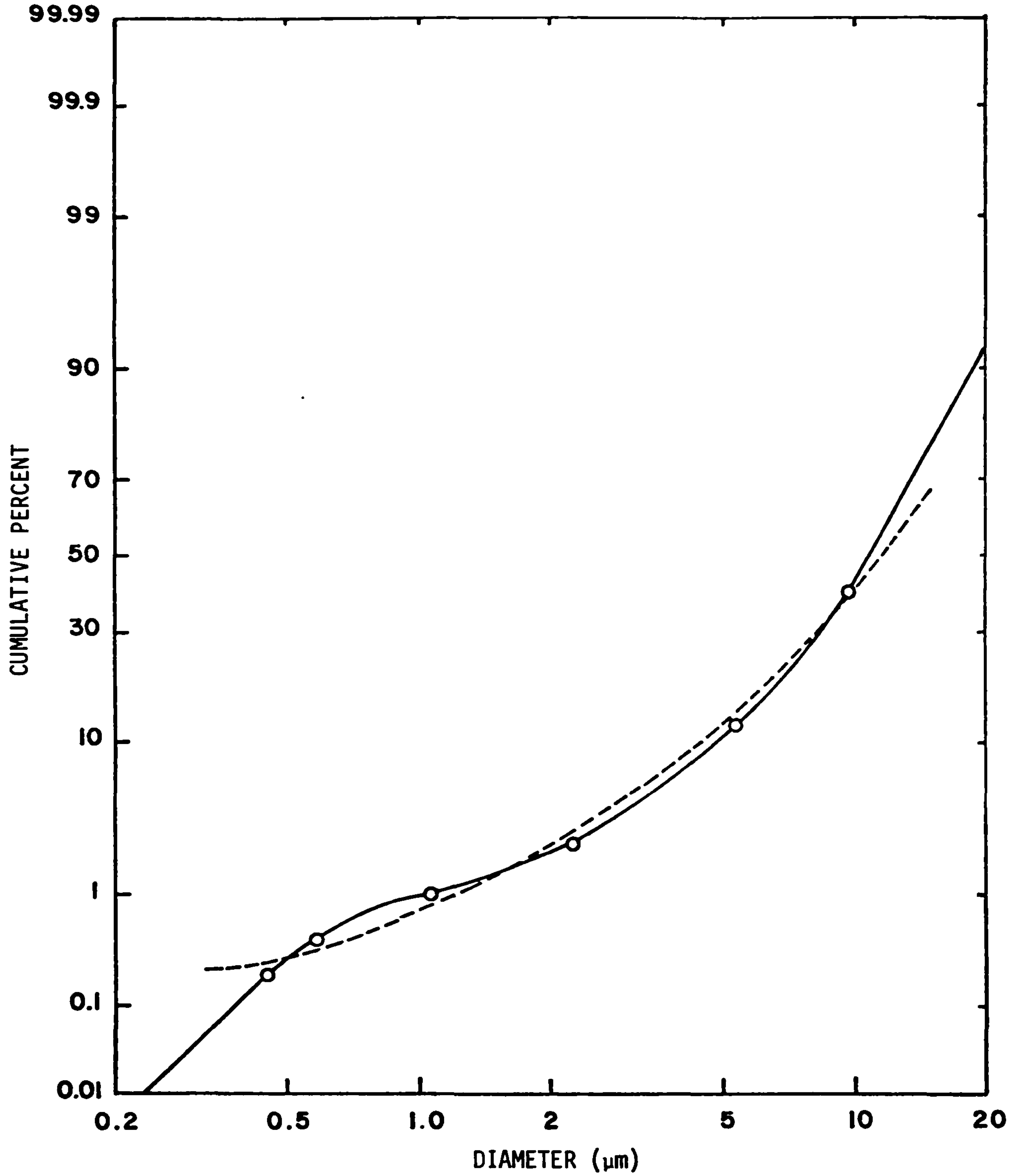

Figure 5.7. Inlet size distribution (average of six runs). Spline fit is the solid curve and quadratic least-squares fit is the dashed curve.

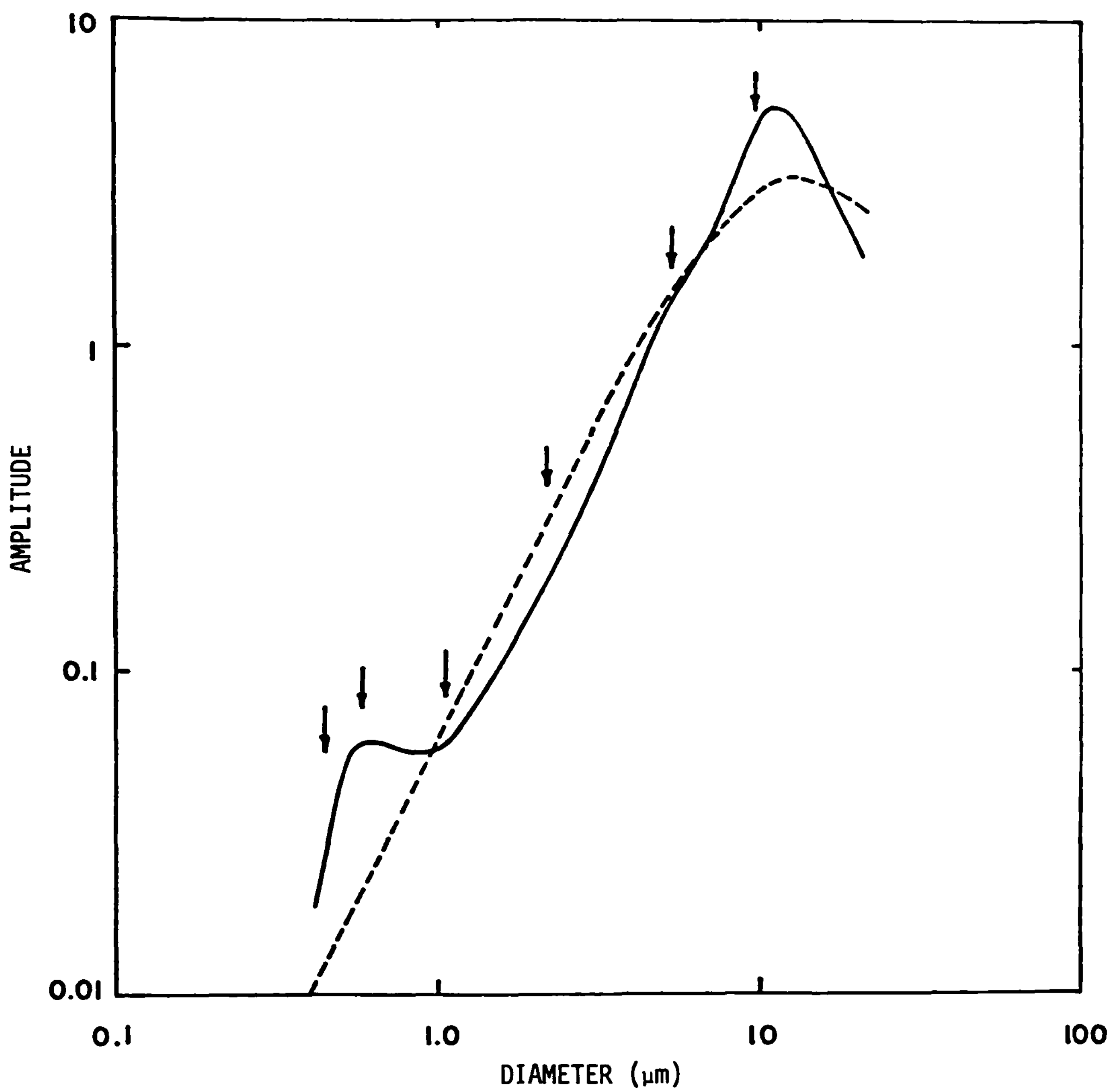

Figure 5.8. Inlet frequency distribution from Figure 5.7. Solid line is the spline derivative and the dashed line is the least-squares derivative. Arrows indicate the positions of the data points.

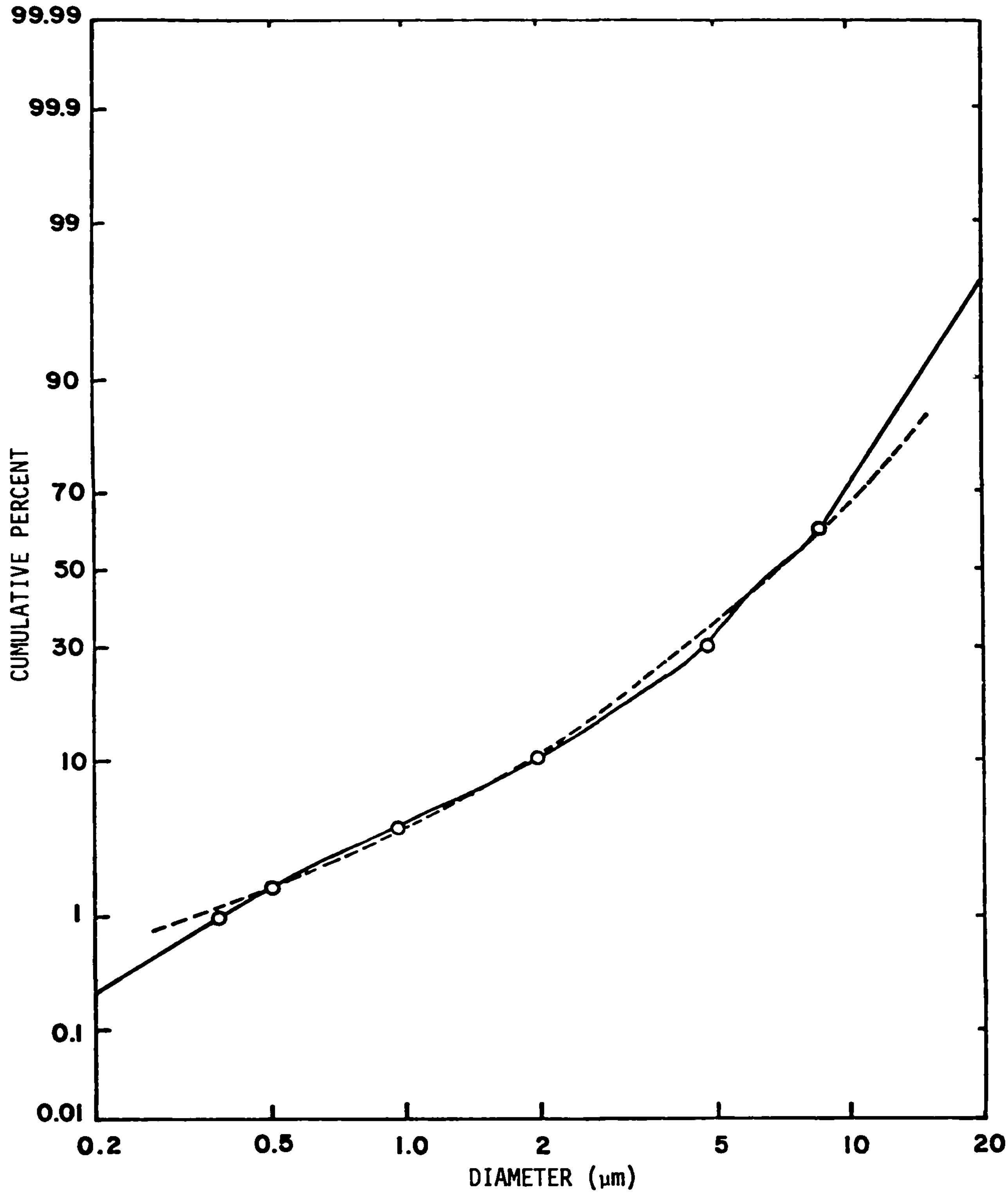

Figure 5.9. Outlet size distribution (average of six runs). Solid curve is the spline fit and the dashed curve is the least-squares fit.

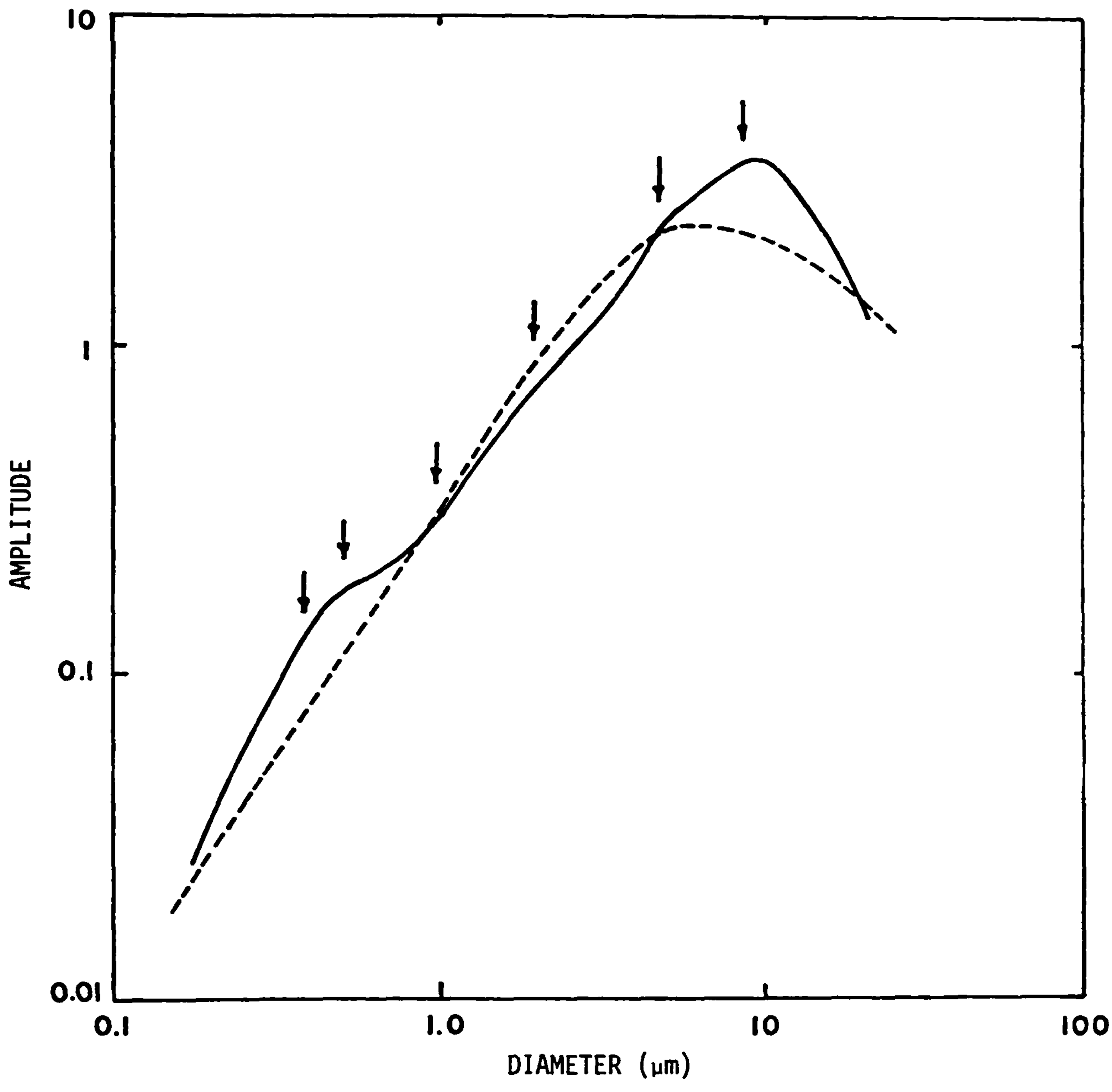

Figure 5.10. Outlet frequency distribution from Figure 5.9. Solid curve is from the spline fit and the dashed curve is from the least-squares fit. Arrows indicate the location of the original data points.

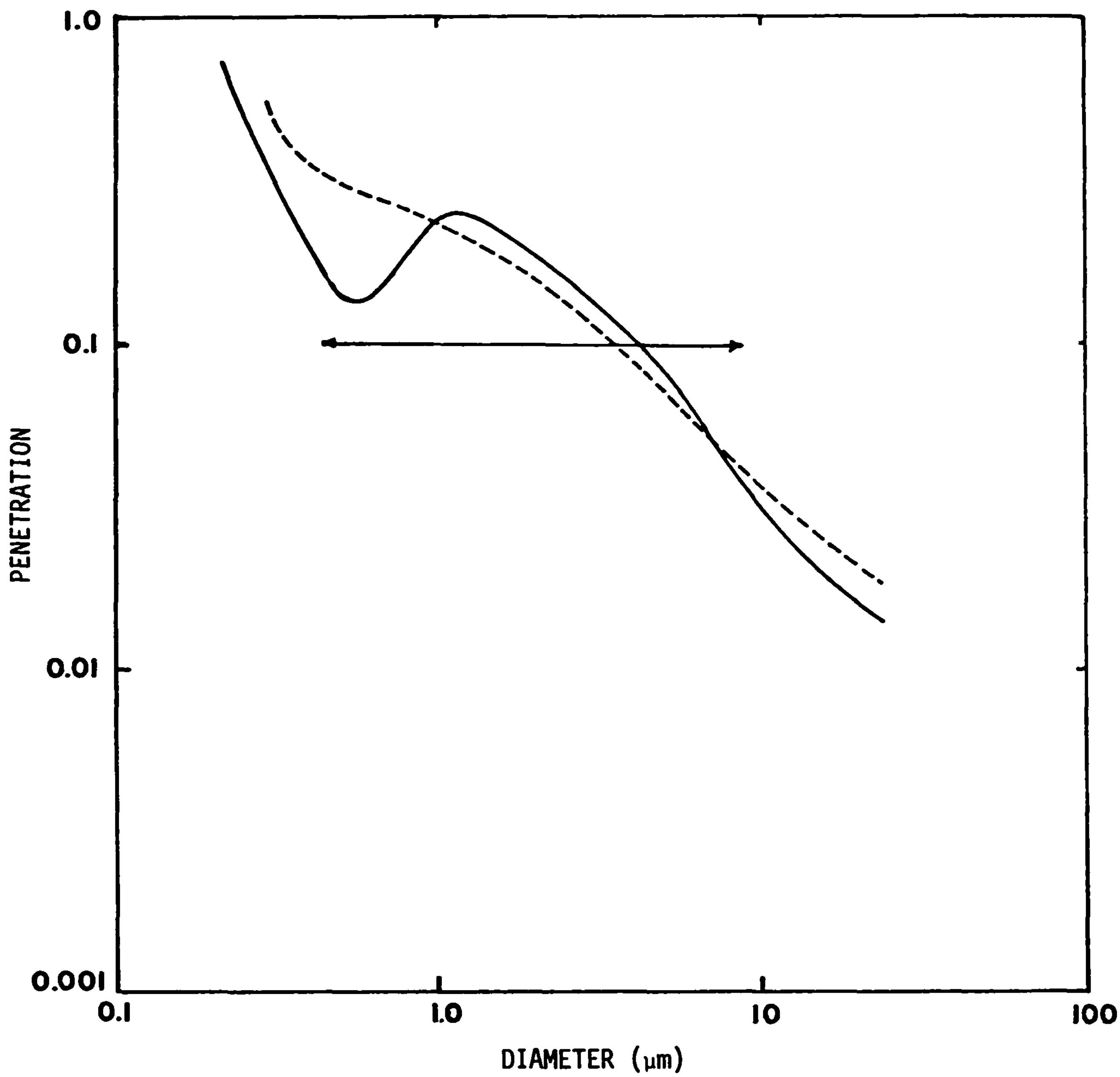

Figure 5.11. Penetration curves for the averaged data runs. Solid line is from the spline fits and the dashed line is from the least-squares fits. The horizontal arrow indicates the range of data common to both inlet and outlet samples.

The penetration curves in Figure 5.6 were computed by dividing the outlet frequency distribution at the points of interest by the inlet frequency distribution at the same points and multiplying by the ratio of the outlet dust concentration to the inlet dust concentration. The limits of the data range common to both samples are indicated by the arrows, but the extrapolated regions are well-behaved for reasonable distances outside the data range. The spline-derived penetration curve shows considerably more variation than the least-squares curve, due to the more detailed frequency distributions of the spline fits.

The second group of data, averaging six impactor runs in the inlet and outlet cumulative curves, is similar. The cumulative curves are somewhat smoother, and that is reflected in the frequency distribution curves for the spline fit. This smoothness may be due to the reduction of random error in the data by the averaging process. Note that the secondary peaks in the spline fits are less prominent, but still present. Again, the distribution of cut-points promises good representation of the secondary peaks.

The penetration curve, Figure 5.11, is also similar to Figure 5.6. Since similar conditions prevailed in these two data groups, temperature being the major difference, such a result is expected.

These data were not particularly difficult to fit, since a linear least-squares method also gave very reasonable (> 0.99) correlation coefficients in all cases. The additional resolution, obtained in the spline fits seems real, based on the consistency of the location of the secondary peaks in the frequency distribution, and the spline fits would be preferred in these cases. Spline fits of less consistent data would normally give very complex penetration curves, and a least-squares method might be preferable.

6.0 References

1. K. M. Cushing, G. E. Lacey, J. D. McCain, and W. B. Smith, "Particulate Sizing Techniques for Control Device Evaluation: Cascade Impactor Calibrations," U.S. Environmental Protection Agency, Office of Research and Development, Washington, D.C. 20460. EPA-600/2-76-280.

2. L. G. Felix, G. I. Clinard, G. E. Lacey, and J. D. McCain, "Inertial Cascade Impactor Substrate Media for Flue Gas Sampling," U.S. Environmental Protection Agency, Office of Research and Development, Washington, D.C. 20460. EPA-600/7-77-060.

3. E. L. Stiefel, "An Introduction to Numerical Mathematics," (Academic Press, New York, 1963).

4. G. R. Markowski and D. S. Ensor, "A Proceedure for Computing Particle Size Dependent Efficiency for Control Devices from Cascade Impactor Data," presented at the annual meeting of the Air Pollution Control Association, Toronto, Canada (1977).

5. J. D. McCain, G. Clinard, L. Felix and J. Johnson, "A Data Reduction System for Cascade Impactors," presented at the Symposium on Advances in Particle Sampling and Measurement, Asheville, N.C. (1978). Sponsored by Environmental Protection Agency, Industrial Environmental Research Laboratory, Research Triangle Park, N.C. 27713.

6. J. L. Walsh, J. H. Alberg, and E. N. Wilson, J. of Mathematics and Mechanics 11, 225 (1962).

7. C. DeBoor, J. Mathematics and Mechanics 12, 747 (1963).

8. M. Abramowitz and J. Stegen, "Handbook of Mathematical Functions" (Dover, New York, 1964).

9. T. N. E. Greville, "Mathematical Methods for Digital Computers, Vol. II," A. Ralston and H. S. Wilf, eds. (John Wiley and Sons, New York, 1967).

10. P. M. Prenter, "Splines and Variational Methods" (John Wiley and Sons, New York, 1975).

11. L. E. Sparks, "Cascade Impactor Data Reduction with SR-52 and TI-59 Programmable Calculators," EPA report in Press (1979).

7.0 Appendices

Two computer programs are included for use in implementing the log-normal transformation and the natural third order spline fit. Appendix 7.1 is a Fortran realization of the algorithm given in Section 4.0. The algorithm itself is taken from Reference 8. (As presented in Reference 8, the algorithm has a sign error; the version shown here operates correctly.) Appendix 7.2 is a Fortran program taken almost exactly from Reference 9; the structure is the same except for the addition of the extrapolation calculations and the omission of an integral calculation.

Appendix 7.1

Computer Routine for the Normal Transformation

```

C THIS FUNCTION CALCULATES THE INVERSE OF THE PROBABILITY INTEGRAL.  P MUST
C BE IN THE RANGE OF 0 TO 1.

      FUNCTION APROB(P)
      FLG = 0
      PS = P
      IF(P.LE.0.0) GO TO 1
      IF(P.GE.1.0) GO TO 2
      IF(P.GT.0.5) GO TO 3
    4 T = SQRT(ALOG(1.0/P**2))
      TS = T*T
      TC = TS *T
      APROB = -(TS*0.010328 + T*0.802853 + 2.515517)/(TC*0.001308 +
     1 TS*0.189269 + T*1.432788 + 1.0) + T
      IF(FLG.EQ.0) APROB =-APROB
      P = PS
      RETURN
    1 APROB = -1.0E37
      RETURN
    2 APROB = 1.0E37
      RETURN
    3 FLG = 1
      P = 1.0 - P
      GO TO 4
      END
```

Appendix 7.2

Spline Fit Subroutine

```
C THIS SUBROUTINE FITS A NATURAL CUBIC SPLINE THROUGH N DATA POINTS, PASSED BY
C MEANS OF THEIR COORDINATES IN VECTORS X AND Y.  THE ORDINATES IN X  MUST BE
C ARRANGED IN ASCENDING ALGEBRAIC ORDER.   THE M POINTS AT WHICH CALCULATIONS
C WILL BE PERFORMED ARE PASSED IN THE ARRAY T, IN ANY ORDER.

C THE VECTOR SS OUTPUTS THE VALUE OF THE SPLINE FIT AT EACH OF THE POINTS IN T,
C AND THE VECTOR SS1 OUTPUTS THE DERIVATIVE OF THE SPLINE FIT AT THE POINTS IN T
C .  THE SECOND DERIVATIVE IS ALSO AVAILABLE INTERNALLY, AND IT CAN BE OUTPUTTED
C . FOR POINTS OUTSIDE THE RANGE OF THE INPUT DATA, LINEAR EXTRAPOLATIONS FROM
C THE END POINTS OF THE FIT ARE PERFORMED, USING THE VALUE OF THE FITTED SLOPE
C AT EACH END POINT.


      SUBROUTINE  SPLINE(M,N,X,Y,T,SS,SS1)

      DIMENSION X(10),Y(10),T(12),SS(12),SS1(12),SS2(12),H(10),DELY(10),
     * H2(10),B(10),DELSQY(10),S2(10),C(10),S3(10)
      DATA  OMEGA/1.0717968/

C THE FINITE DIFFERENCE APPROXIMATIONS TO THE DERIVATIVES ARE MADE FOR EACH OF
C THE DATA POINTS, WITH THE SECOND DERIVATIVES AT THE END POINTS FIXED AT ZERO.

      N1 = N - 1
      DO 51 I = 1,N1
      H(I) = X(I+1) - X(I)
   51 DELY(I) = (Y(I+1) - Y(I))/H(I)
      DO 52 I = 2,N1
      H2(I) = H(I-1) + H(I)
      B(I) = 0.5*H(I-1)/H2(I)
      DELSQY(I) = (DELY(I) - DELY(I-1))/H2(I)
      S2(I) = 2.0*DELSQY(I)
   52 C(I) = 3.0*DELSQY(I)
      S2(1) = 0.0
      S2(N) = 0.0

C A RELAXATION TECHNIQUE IS USED  TO SOLVE FOR THE INTERIOR SECOND DERIVATIVES,
C REPEATED UNTIL THE RESIDUAL, ETA, IS SMALL ENOUGH.

    5 ETA = 0.0
      DO 10 I = 2,N1
      W = (C(I) - B(I)*S2(I-1) - (0.5 - B(I))*S2(I+1) - S2(I))*OMEGA
```

```
          IF(ABS(W).GT.ETA)  ETA = ABS(W)
   10     S2(I) = S2(I) + W
          IF(ETA.GE.1.0E-06) GO TO 5

    C THE FINITE APPROXIMATIONS TO THE THIRD DERIVATIVES ARE CALCULATED.

          DO 53 I = 1,N1
   53     S3(I) = (S2(I+1) - S2(I))/H(I)


    C THE VALUES OF THE SPLINE AND ITS DERIVATIVES ARE CALCULATED AT THE POINTS OF T

          DO 61 J = 1,M
          I = 1
          IF(T(J) - X(1)) 58,17,55
   55     IF(T(J) - X(N)) 57,59,62
   56     IF(T(J) - X(I)) 60,17,57
   57     I = I + 1
          GO TO 56

    C EXTRAPOLATION BELOW THE FIRST DATA POINT

   58     SS1ONE = DELY(1) - H(1)/(H(1) + H(2))*2.0*(DELY(2) - DELY(1))
          SS(J) = Y(1) + (T(J) - X(1))*SS1ONE
          SS1(J) = SS1ONE
          GO TO 61

    C EXTRAPOLATION ABOVE THE LAST DATA POINT

   62     SS1N = DELY(N1) + H(N1)*S2(N1)
          SS(J) = Y(N) + (T(J) - X(N))*SS1N
          SS1(J) = SS1N
          GO TO 61

    C CALCULATION IN THE RANGE OF THE DATA POINTS

   59     I = N
   60     I = I - 1
   17     HT1 = T(J) - X(I)
          HT2 = T(J) - X(I+1)
          PROD = HT1*HT2
          SS2(J) = S2(I) + HT1*S3(I)
          DELSQS= (S2(I) + S2(I+1) + SS2(J))/6.
          SS(J) = Y(I) + HT1*DELY(I) + PROD*DELSQS
          SS1(J) = DELY(I) + (HT1 + HT2)*DELSQS + PROD*S3(I)/6.
   61     CONTINUE

          RETURN
          END
```

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA-600/7-78-189	2.	3. RECIPIENT'S ACCESSION NO.
4. TITLE AND SUBTITLE Analysis of Cascade Impactor Data for Calculating Particle Penetration		5. REPORT DATE September 1978
		6. PERFORMING ORGANIZATION CODE
7. AUTHOR(S) Phil A. Lawless		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS Research Triangle Institute, P.O. Box 12194, Research Triangle Park, North Carolina 27709		10. PROGRAM ELEMENT NO. EHE624
		11. CONTRACT/GRANT NO. 68-02-2612, Task 36
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; 6-8/78
		14. SPONSORING AGENCY CODE EPA/600/13

15. SUPPLEMENTARY NOTES IERL-RTP project officer is Leslie E. Sparks, Mail Drop 61, 919/541-2925.

16. ABSTRACT The report discusses the difficulties of analyzing cascade impactor data to obtain particle penetrations according to size. It considers several methods of analysis (interpolation, least-squares fitting, and spline fitting) and weighs their merits. It also discusses the use of transforming functions prior to data fitting. It recommends the use of the normal transformation and spline fitting method, and provides computer programs to facilitate its use.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Impactors	Pollution Control	13B	13I
Dust	Interpolation	Particulate	11G	12A
Penetration	Least-squares	Cascade Impactors	14B	
Size Determination	Method	Normal Transformation		
Measurement	Computer Programs	Spline Fitting		09B
Analyzing	Data Processing			

18. DISTRIBUTION STATEMENT Unlimited	19. SECURITY CLASS *(This Report)* Unclassified	21. NO. OF PAGES 44
	20. SECURITY CLASS *(This page)* Unclassified	22. PRICE

EPA Form 2220-1 (9-73)