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PULSE HEIGHT RESPONSE OF AN OPTICAL PARTICLE COUNTER TO MONODISPERSE AEROSOLS

By R. G. Wilmoth, S. S. Grice and V. Cuda

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The pulse height response of a right-angle-scattering optical particle counter has been investigated using monodisperse aerosols of polystyrene latex spheres, di-ctyl phthalate and methylene blue. The results confirm previous measurements for the variation of mean pulse height as a function of particle diameter and show good agreement with the relative response predicted by Mie-scattering theory. Measured cumulative pulse height distributions were found to fit reasonably well to a log-normal distribution with a minimum geometric standard deviation of about 1.4 for particle diameters greater than about 2 μm. The geometric standard deviation was found to increase significantly with decreasing particle diameter.
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SUMMARY

The pulse height response of a right-angle-scattering, optical particle counter has been investigated using monodisperse aerosols of polystyrene latex spheres, di-octyl phthalate and methylene blue. The results confirm previous measurements for the variation of mean pulse height as a function of particle diameter and show good agreement with the relative response predicted by Mie-scattering theory. Measured cumulative pulse height distributions were found to fit reasonably well to a log-normal distribution with a minimum geometric standard deviation of about 1.4 for particle diameters greater than about 2 μm. The geometric standard deviation was found to increase significantly with decreasing particle diameter.

INTRODUCTION

Optical particle counters are frequently used in air pollution studies to measure the size of atmospheric aerosols. These counters convert the light scattered from single aerosol particles into electrical pulses whose frequency is proportional to the concentration and whose height is proportional to the size of the particles. To accurately determine the size distribution when sampling unknown aerosols, it is necessary to know the instrument response as a function of particle size, particle refractive index, and particle shape. It is also important to know the resolution of the counter as a function of particle size.

The response of the most common types of counters (forward-scatter and right-angle-scatter) has been investigated (refs. 1 and 2) as a function of size and index of refraction. Although the size resolution of these counters was also investigated, insufficient information is presented to permit determination of the accuracy of specific instruments for size distribution measurements. The purpose of this paper is to present detailed measurements of the pulse height distributions produced by one type of optical particle counter, viz, right-angle scattering type, to illustrate the nonuniformity of pulses produced when sampling a monodisperse aerosol. Results are presented as a function of particle diameter and index of refraction, and the effects of instrument resolution on the ability to distinguish between sizes and types of particles are discussed. Also discussed are some of the difficulties encountered in attempting to use state-of-the-art techniques for the generation and sampling of monodisperse aerosols.
Two methods of aerosol generation were used for the experiment. Aerosols of uniform polystyrene latex (PSL) spheres were generated by nebulizing a
suspension containing 0.1 percent by weight of these spheres with a commercial compressed air nebulizer. The resulting drops were then mixed with dry, filtered air in a drying chamber and the aerosol stream was introduced into the particle counter inlet as shown in figure 1. Particle concentrations generated by this method were typically about 8 particles/cm³ with the number of multiplets (more than one PSL sphere per drop) estimated to be less than 1 percent.

Aerosols of di-octyl phthalate (DOP) and methylene blue were generated by a vibrating-orifice monodisperse aerosol generator² shown schematically in figure 2. A dilute, liquid solution was fed through a 10 μm diameter orifice vibrating at an ultrasonic frequency causing a breakup of the liquid jet into uniform drops. The drops were then mixed with a sufficient quantity of dry, filtered air (about 2 m³/hr) to allow the solvent to evaporate. The electrical charge incurred on the drops during dispersion was neutralized by a Kr-85 radioactive source located in the drying column. The diameter, \( D_d \), of the liquid drops is given by (ref. 1).

\[
D_d = \left( \frac{6Q}{\pi f} \right)^{1/3}
\]

where \( Q \) is the volumetric liquid flow rate, and \( f \) is the frequency at which the orifice is vibrated. The final particle diameter, \( D_p \), is then given by

\[
D_p = C^{1/3} D_d
\]

where \( C \) is the volumetric concentration of nonvolatile material in the liquid solution. The theoretical particle concentration, \( n_o \), is given by

\[
n_o = \frac{f}{Q_a}
\]

where \( Q_a \) is the total volume rate of airflow through the generator. At the conditions of the experiments (\( f = 150 \text{ kHz} \), \( Q_a = 2 \text{ m}^3/\text{hr} \)), equation (3) gives about 270 particles/cm³. However, the actual concentrations measured by the particle counter were a factor of 5 to 6 lower than the theoretical concentration due to losses in the drying column and sampling system.

For one set of experiments with aerosols from the vibrating-orifice generator, sampling was accomplished through an isokinetic (IK) sampler as shown in figure 3. This sampler was designed to permit isokinetic flow at the low flow rate of the optical particle counter (about 0.2 m³/hr) from the high flow

¹ Thermo-Systems, Inc. Model 3050

² Measured as the number of 2.5 μm diameter PSL spheres per drop.
rate of the aerosol generator (about 2 m³/hr). However, some difficulty was encountered in sampling the larger particles (> about 3 µm diameter) resulting in excessive loss of particles due either to break-up, or to impaction in the sampling lines. Therefore, an additional set of experiments were performed by placing the 1.25 inch diameter, flexible hose directly over the inlet to the particle counter. These experiments will be referred to as non-isokinetic (NIK) samples.

Optical Particle Counter

The optical particle counter ² used for the present work detects particles by sensing the light scattered at near right angles to the incident light beam. A schematic of the optical system is shown in figure 1. White light is projected through the aerosol stream illuminating a cylindrical volume of about 2.6 mm³. Light pulses scattered into a 47.5° cone, whose axis is 90° from the incident beam axis, are collected and converted to electrical pulses by a photomultiplier tube. The uniformity in the height (amplitude) of these pulses depends on the baseline electronic and optical noise and on the uniformity of light intensity in the illuminated volume (ref. 2).

The counter is designed to detect particles with diameters in the range of 0.5 to 10 µm at a sampling rate of 3 l/min. The maximum particle concentrations that can be measured without significant (<10 percent) coincidence losses, i.e., more than one particle in the viewing volume at one time, is about 100 particles/cm³. Further details of the counter are given in references 1 and 3.

Pulse Height Detection System

Pulses from the optical particle counter were fed into a pulse discriminator-detector which gave a positive 13-volt pulse every time the height of an input pulse exceeded the discriminator voltage level (see fig. 5). A unique feature of the discriminator-detector was that it did not require each input pulse to return to a voltage less than the discriminator level before the next pulse could be detected. Thus, overlap between input pulses did not prevent these pulses from being counted separately. For typical pulse shapes produced by the particle counter (typically Gaussian with 100 µsec half-widths), the discriminator-detector could accurately measure count rates up to 5000 counts/sec. The discriminator-detector output pulses were used to trigger an electronic counter, and the counts were recorded on a digital printer. The normal counting period was 1 sec and the counts for several periods (26) were recorded and averaged for each data point.

By varying the discriminator voltage level, \( V_d \), and recording the counts at each level, a cumulative pulse height distribution was obtained. The range of the discriminator-detector was 0.01 volt (corresponding to zero setting)

² Royco Instruments, Inc., Model 220
to 10 volts with an accuracy of ±0.01 volt. Although the baseline electronic noise out of the particle counter was about 0.05 volt, it was found that the frequency of this noise was too high to trigger the discriminator-detector even when the discriminator level was set to its minimum value. Thus, accurate particle count rates could be measured for pulse heights down to 0.01 volt.

The output of the optical particle counter was also fed directly into an uncalibrated discriminator-amplifier built into the electronic counter. This discriminator level was adjusted to give the same total count rate as that with the discriminator-detector when set at \( V_d = 0 \). Thus, a rapid measurement of the total particle count could be made before and after each counting period at a prescribed value of \( V_d \). This allowed variations in particle concentration during a run to be taken into account when normalizing the cumulative pulse height distributions to the total pulse count.

**DATA ANALYSIS**

**Cumulative Pulse Height Distributions**

For each run, the pulse count was recorded at a number of values of the discriminator level. As mentioned previously, the pulse count at each \( V_d \) was taken as the average of several count readings taken at the same \( V_d \). Letting \( N(V>V_d) \) represent the total count of pulses whose height is greater than \( V_d \), the probability, \( P(V>V_d) \), of measuring a pulse with height greater than \( V_d \) is given by

\[
P(V>V_d) = \frac{N(V>V_d)}{N_T}
\]

where \( N_T \) is the total pulse count. It was assumed for these measurements that \( N_T \) was equal to the total count measured at \( V_d = 0 \). Although the actual minimum level was about 0.01 volt, it was found that the number of pulses with height ≤0.01 volt was always much less than the total pulse count.

**Curve Fitting to Log-Normal Distribution**

A particle size distribution frequently used in analysis of air pollution measurements is the log-normal distribution given by (ref. 4)

\[
y' = \frac{1}{\ln \sigma_g \sqrt{2\pi}} \exp \left[ -\frac{(\ln D_p - \ln M)^2}{2(\ln \sigma_g)^2} \right]
\]
where $y'$ is the number of particles with logarithmic diameters in the interval $d \ln D_p$ per unit $d \ln D_p$, $D_p$ is particle diameter, $M_g$ is the geometric mean diameter, and $\sigma_g$ is the geometric standard deviation. For convenience in comparing the resolution of the optical particle counter to typical particle size distributions for polydisperse aerosols, the measured pulse height distributions were assumed to obey a similar relation, viz,

$$
\frac{dN(V)}{d\ln V} = \frac{1}{\ln \sigma_g \sqrt{2\pi}} \exp \left[ -\frac{(\ln V - \ln M_g)^2}{2\ln^2 \sigma_g} \right] 
$$

(6)

where $dN(V)$ is the number of pulses in the interval $d\ln V$, and $V$ is pulse height. $M_g$ and $\sigma_g$ retain a similar meaning except they are now in terms of mean pulse height and pulse height standard deviation, respectively. To convert equation (6) to the form of a cumulative distribution, it is noted that

$$
P(V; V_d) = 1 - \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{X} e^{-u^2/2} du 
$$

(7)

where $X = (\ln V_d - \ln M_g)/\ln \sigma_g$ and $u = (\ln V - \ln M_g)/\ln \sigma_g$. Equation 7 is simply the form of the normal probability integral (ref. 5). Using measured values of $P(V > V_d)$ for each cumulative pulse height distribution, $X$ - values were found from table D of reference 5. This gave a set of values for $X$ versus $V_d$ which could be fit by the method of least squares to the equation

$$
\ln V_d = \ln \sigma_g + \ln M_g 
$$

(8)

from which values for $M_g$ and $\sigma_g$ were found. From equations 7 and 8, it can be shown that $M_g$ is the value of $V_d$ at which $P = 1/2$ and $\sigma_g = (\text{value of } V_d \text{ at which } P = 0.843)/M_g$.

Error Analysis

For the latex spheres, particle diameters given herein are taken to be the manufacturer's quoted values obtained from batch samples and are believed to be accurate to better than 1 percent. For aerosol particles generated by the vibrating-orifice technique, diameters were calculated from equations 1 and 2, and thus depend on the accuracy of $Q_k$, $f$ and $C$. Estimating the
uncertainties to be ±10 percent for $Q$, ±0.1 percent for $f$ and ±1 percent for $C$ gives an estimated uncertainty of about ±3 percent for the diameter of these particles.

In applying equations 1 and 2, it was also assumed that the liquid solvent did not contain any nonvolatile impurities. The presence of such impurities would cause the particle diameters to be larger than those calculated. To estimate this effect, measurements were made of the mean particle diameter resulting when using only pure solvent without the solute in the aerosol generator. From the results, the concentration of nonvolatile impurities was estimated to be about 0.001 percent. For the solution concentrations used to obtain particles less than 1 μm diameter, this resulted in underestimating those diameters by about 10 percent. However, for particles greater than 1 μm, these impurities had a negligible effect on the calculated diameter.

Sample standard deviations in average count rate were typically about ±13 percent of $N_T$. The uncertainties in determining the geometric mean pulse height and geometric standard deviation using the log-normal distribution fit were generally less than ±20 percent for $M_g$ and ±0.1 for $\sigma_g$.

EXPERIMENTS

A summary of the various aerosol types and sizes tested is given in table I. Also listed is the estimated degree of monodispersity expressed in terms of the geometric standard deviation for a log-normal size distribution. The index of refraction is shown for the PSL and DOP aerosols. Although the index of refraction for the methylene blue aerosol is not known, it is certain to have a significant imaginary component due to visible absorption. For the DOP aerosol, tests were performed using two different initial drop diameters, and with and without isokinetic sampling.

RESULTS AND DISCUSSION

Measured Cumulative Pulse Height Distributions

All cumulative distributions will be shown both as $P(V<V_d)$ versus $V_d$ and as $P(V>V_d)$ versus $V_d$. It is straightforward to convert between the two since $P(V<V_d) = 1 - P(V>V_d)$. Also shown for each distribution, is the least-squares-fit line for a log-normal distribution given by equation 8.

PSL Spheres. The pulse height distributions for PSL spheres are shown in figure 6. The agreement with the log-normal distribution form is generally good. A principal exception is the data for $D_p = 2.02$ μm which show a greater percentage than expected of pulse heights smaller than the mean, i.e.,
a flatter slope at small values of $V_d$. This is believed to be due primarily to two factors; (1) loss of the particles through impaction in the sampling lines, and (2) generation of smaller particles by breakup of liquid drops which did not completely evaporate in the drying chamber. The first factor results in a lower ratio of the total count rate to the standard deviation in count rate due to random fluctuations, i.e., signal-to-noise ratio. The second factor simply gives greater numbers of small particles probably with a very broad size distribution which is superimposed onto that due to the PSL spheres. Although the PSL spheres would not be expected to breakup, the original solution probably had a significant amount of nonvolatile impurities which could produce small solid particles.

**DOP Aerosol.** The results for the DOP aerosol sampled isokinetically are shown in figure 7 for two different drop sizes. The distributions are similar to those for PSL spheres, fitting the straight lines reasonably well for $D_p < 2 \mu m$, but again showing a significantly smaller slope for pulse heights less than the mean. Although different sampling systems were used, both PSL and DOP were sampled through relatively small diameter tubing with a significant bend in it just before it attached to the particle counter. DOP samples were also taken by placing the large sampling hose directly over the particle counter inlet. These results are shown in figure 8. The distributions for non-isokinetic sampling do not exhibit the large decrease in slope seen with the isokinetic samples for $D_p > 2 \mu m$. Although there do appear to be some characteristic variations in slope, these distributions fit the log-normal form reasonably well over the entire range of pulse heights.

**Methylene Blue Aerosol.** Pulse height distributions for methylene blue aerosol sampled isokinetically are shown in figure 9. Good agreement with the log-normal distribution is seen for all but $D_p = 5.23 \mu m$. Thus, it appears that impaction losses and particle breakup are less significant than for DOP aerosol. Since DOP is a plasticizer and plastic tubing was used for the sampling line, DOP was probably removed more efficiently than the methylene blue particles.

**Mean Pulse Heights**

The geometric mean pulse heights for PSL spheres are shown in figure 10. For comparison, the theoretical response is also shown, based on Mie-scattering theory taken from reference 1 for an index of refraction, $m$, of 1.6. Since Mie theory can only predict the relative response, the theoretical curves were equated to the measured response at $D_p = 0.79 \mu m$ given by the instrument manufacturer's calibration with PSL spheres (a value of 0.23 volt). All theoretical response curves presented in this paper were put on an absolute scale in the same manner. Thus, all comparisons with theory are relative to $D_p = 0.79 \mu m$ and $m = 1.6$. Figure 10 shows good agreement between theory and experiment for the relative mean pulse height as a function of particle diameter. For $D_p < 1 \mu m$, the measured pulse heights are slightly lower than
predicted, probably due to the contribution of small particles formed by impurities in the aerosol solution.

The results for DOP aerosol sampled isokinetically are shown in figure 11 for two drop diameters and compared to the theoretical response for \( m = 1.5 \). The agreement with theory is very good for the smaller drop diameter except at \( D_p < 1 \mu m \). The larger drop diameter generally gave larger pulse heights due to incomplete evaporation of the solvent, although the reverse is true for \( D_p < 1 \mu m \). However, it should be noted that the uncertainty in \( M_g \) is greater at smaller particle diameters due to the greater dispersion in the pulse height distributions. Also, the presence of nonvolatile impurities in the solution caused the actual diameter to be about 10 percent greater than those calculated for particles less than 1 \( \mu m \). Thus, no firm conclusion can be made about the results for \( D_p < 1 \mu m \). Direct or non-isokinetic sampling of DOP aerosol gave the results shown in figure 12. Some indication of the uncertainty in \( M_g \) can be seen from the repeated measurements with the smaller drops. However, the mean pulse heights with the small drops still give good agreement with theory while the larger drops give consistently higher pulse heights. Comparing figures 11 and 12, there appears to be no significant difference in the mean pulse heights obtained by isokinetic and non-isokinetic sampling. Thus, it seems that the difference for the two drop sizes might be due, in part at least, to the greater significance of impurities for the larger drops, i.e., for a given concentration of impurities in the solvent, the greatest increase in particle diameter over that calculated by equation 2 will occur for larger drops.

Mean pulse heights for methylene blue aerosol are shown in figure 13. Since the index of refraction was not known, the theoretical response for \( m = 1.5 \) was chosen arbitrarily for comparison. The measured pulse heights are typically a factor of 10 less than those predicted for non-absorbing spheres, i.e., no imaginary component. Since methylene blue is known to absorb light in the visible region, a theoretical curve for \( m = 1.5 - 0.11 \) is also shown to illustrate the qualitative effect of absorption. Although this value for \( m \) gives much better agreement between theory and experiment than that for the nonabsorbing case, it should be noted that this particular combination of real and imaginary parts is not necessarily unique, i.e., about the same order of magnitude difference in response could be predicted by simply using a lower value for the real part of \( m \) and no imaginary part (see refs. 1 and 6). This illustrates the importance of knowing the index of refraction of the aerosol under study before attempting to deduce size information from measured pulse heights.

**Pulse Height Dispersion**

The variation in the dispersion of the pulse height distributions is illustrated by the variation of the log-normal geometric standard deviation with geometric mean pulse height shown in figure 14. Correlation with mean pulse height allows a more direct comparison for all of the aerosols tested than correlation with particle diameter. Although there is considerable
scatter in the results, the decrease of $\sigma_g$ with increasing $M_g$ is consistent in all cases, with $\sigma_g$ reaching a limit of about 1.4 at about 1 volt (corresponding to $D_p = 2 \mu m$). Thus, the correlation with mean pulse height seems to be justifiable even for widely different aerosols. These results are qualitatively similar to those found in reference 2 for a forward-scatter particle counter. However, the magnitude of the deviations shown in figure 14 are much larger since the right-angle counter is more sensitive to the position of the particle in the viewing volume. Improvement in the pulse height dispersion for right-angle counters has been reported in reference 2 using a sheath of filtered air around the aerosol stream to "focus" the particles.

An estimate of the average standard deviation is given by the faired line through all the data points in figure 14. The effect of this deviation on the resolution of the instrument for particle size determination is illustrated in figure 15. Here the faired line for $\sigma_g$ has been used together with the theoretical response curve for $m = 1.5$ to calculate bands containing 68 and 98 percent of the pulses for a given particle diameter. The increase in $\sigma_g$ as particle diameter decreases accounts for the significant widening of the bands for $D_p < 1 \mu m$. Although these results are based on monodisperse aerosols, they can be used to predict, at least qualitatively, some aspects of the results expected for polydisperse aerosol. First, for a given pulse height, there will be contributions from a range of particle diameters resulting in a measured size distribution which is broader than the true distribution. In fact, the minimum broadening for this instrument varies from about 0.2 $\mu m$ at the low end of the size range (corresponding to a geometric standard deviation of about 2.5) to about 4 $\mu m$ at the upper end of the size range (corresponding to geometric standard deviation of 1.4). Second, since the relative broadening (in terms of $\Delta \ln D_p$) is greater for smaller particles, there will be a greater relative contribution of pulses from small particles resulting in a size distribution that is skewed to smaller diameters with a mean diameter less than the true value. The exact amount of this effect could be estimated by a convolution of the instrument pulse height response distribution over the actual size distribution. Finally, the dispersion in pulse heights places a lower limit on the minimum number of pulses which must be counted to obtain a prescribed accuracy for the mean value. For example, based on t-statistics, the minimum number of pulses required to give a 98 percent confidence interval of ±10 percent of $M_g$ varies from about 500 at $D_p = 0.5 \mu m$ to about 50 at $D_p = 10 \mu m$.

CONCLUDING REMARKS

The response of a right-angle-scattering optical particle counter to PSL and DOP aerosols confirms previous measurements for the variation of mean pulse height as a function of particle diameter and shows good agreement
with the relative response predicted by Mie-scattering theory. However, the need to know the aerosol index of refraction before attempting to deduce accurate size information, particularly for absorbing particles, is emphasized by the results with methylene blue aerosol.

The resolution of the particle counter is found to be severely limited by the pulse height dispersion resulting from nonuniform illumination in the viewing volume and nonuniform optical response. For a monodisperse aerosol \( \sigma < 1.1 \), the minimum measured dispersion corresponds to a geometric standard deviation of about 1.4 for particle diameters greater than about 2 μm and increases with decreasing particle diameter. For a polydisperse aerosol, the effect of this dispersion would be to broaden the distribution and shift it towards smaller diameters, thus, indicating a mean diameter smaller than the true value.

In the course of the experiments, it was also found that the use of state-of-the-art techniques for generation of monodisperse aerosols requires extreme care to minimize the effects of impurities and to ensure proper sampling of the aerosol.
REFERENCES


### TABLE I

**AEROSOL TYPES AND SIZES TESTED**

<table>
<thead>
<tr>
<th>TYPE</th>
<th>DIAMETER, µm</th>
<th>GEOMETRIC STANDARD DEVIATION</th>
<th>INDEX OF REFRACTION</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polystyrene</td>
<td>0.500</td>
<td>1.005</td>
<td>1.59</td>
</tr>
<tr>
<td>Latex (PSL)</td>
<td>0.760</td>
<td>1.006</td>
<td></td>
</tr>
<tr>
<td>Spheres</td>
<td>1.101</td>
<td>1.005</td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.202</td>
<td>1.007</td>
<td></td>
</tr>
<tr>
<td>di-Octyl Phthalate</td>
<td>0.69</td>
<td>&lt; 1.1 (Ref. 3)</td>
<td>1.49</td>
</tr>
<tr>
<td>(DOP)</td>
<td>1.18</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2.01</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.44</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>5.88</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Methylene Blue</td>
<td>0.60</td>
<td>&lt; 1.1 (Ref. 3)</td>
<td>Not known for these experiments</td>
</tr>
<tr>
<td></td>
<td>1.05</td>
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<td></td>
</tr>
<tr>
<td></td>
<td>1.79</td>
<td></td>
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</tr>
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<td>3.06</td>
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</tr>
<tr>
<td></td>
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</tr>
</tbody>
</table>
Figure 1.- Nebulizer aerosol generation system.
Figure 2. - Schematic of vibrating orifice aerosol generator.
Figure 3.- Isokinetic sampling system.
Figure 4.- Schematic of optical particle counter.
Figure 5.- Pulse height discriminator-detector system.
Figure 6. - Cumulative pulse height distributions for PSL spheres.
Figure 7. - Cumulative pulse height distributions for DOP aerosol. (Isokinetic sampling.)
Figure 7. - Concluded.
Figure 8. - Cumulative pulse height distributions for DOP aerosol. (Non-isokinetic sampling.)
Figure 8. - Concluded.
Figure 9. - Cumulative pulse height distributions for methylene blue aerosol. (Isokinetic sampling, \( D_d = 21.7 \mu m \).)
Figure 10. - Variation of geometric mean pulse height with particle diameter. (PSL spheres.)
Figure 11. - Variation of geometric mean pulse height with particle diameter. (DOP aerosol, Isokinetic sampling.)
Figure 12. - Variation of geometric mean pulse height with particle diameter. (DOP aerosol, Non-isokinetic sampling.)
Figure 13. - Variation of geometric mean pulse height with particle diameter. (Methylene blue aerosol, Isokinetic sampling, $D_d = 21.7 \mu m$.)
Figure 14. - Variation of geometric standard deviation with geometric mean pulse height for various aerosols.
Figure 15. - Variation in pulse height dispersion with particle diameter. (m = 1.5.)