DETERMINATION OF PARTICLE-SIZE DISTRIBUTION BY MEASURING THE TRANSMISSION OF AN AEROSOL IN SEDIMENTATION

G. Roy

"Détermination de la Distribution Granulométrique des Particules par la Mesure de la Transmission d'un Aerosol Sedimentant" DREV Report 4278/F2, File 3618B-005, November, 82, 39 pp.
ABSTRACT

The results of a feasibility study for determining aerosol size distribution using the extinction-sedimentation inversion technique are presented. It was found that the height of the dissemination chamber may affect the value of the results.
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LIST OF SYMBOLS

A  numerically determined constant
C  numerically determined constant
Cs sedimentation rate correction factor
d  particle diameter (variable)
dg geometric mean diameter (by number of particles)
dm mean particle diameter (by weight)
F( ) particle distribution (log probability)
g 9.18 m/sec
h height of laser source and filter papers
H height of dissemination chamber
H( ) step function (descending)
I(t) light intensity measured as a function of time
Io light intensity in the absence of aerosol
k Boltzman constant
K(xn, n) diffusion efficiency
l mean free path of air molecules
L length of optical path
m mass (weight) of deposits on filter paper
M total mass (weight) of particles
Mfl weight of disseminated aerosol present at beginning of sedimentation
n index of refraction
n(r) distribution function
N number of particles per unit volume
r radius of a particle (variable)
 rp particle with radius p
r_F particle with radius equal to \( \left( \frac{9h_n}{2 \rho g \tau_F} \right)^{1/2} \)
Mr particle with radius equal to \( \left( \frac{9h_n}{2 \rho g \tau_F} \right)^{1/2} \)
S filter paper area
S_o area of dissemination chamber floor
t time (variable)
t_1 time during which filter paper 1 uncovered
t_D time dissemination starts
T room temperature
T_c characteristic diffusion agglomeration time
v_s rate of a particle under the influence of gravity
V_c volume of dissemination chamber
X_n \( \frac{2 \pi r}{\sqrt{1 + \frac{h}{(p - \rho)}}} \)
\( \frac{9}{2} \left( \frac{h}{(p - \rho)} \right)^{1/2} g \) wavelength
\( \eta \) viscosity of air
\( \rho \) density of air
\( \rho_p \) density of particles
\( \sigma \) collision cross section

\( \tau \) optical density

\( \tau_0 \) optical density at \( t = 0 \)
1.0 INTRODUCTION

Particle size is a factor which determines the ability of an aerosol to diffuse light. An aerosol's ability to diffuse light will vary according to its particle-size distribution.

Several methods exist for measuring the particle-size distribution of an aerosol. First of all, there are optical methods using a wide variety of instruments from the microscope to the Doppler effect laser velocimeter. There are also non-optical methods such as the use of fine grids, cascade impactors, aerosol sedimentation as a function of time, and the Model TA II, Coulter counter made by Coulter Electronics Inc.

This report presents the results of a feasibility study for setting up an optical method based on measuring the extinction coefficient to determine the particle-size distribution of aerosols undergoing sedimentation in the large DREV silo-shaped chamber.

This measurement technique, known as ESIT (Sedimentation-extinction Inversion Technique) is applied to an aerosol, powdered glass No. 3419-2 made by Ferro Industrial Products, Ltd., to determine its particle-size distribution. In order to check whether the validity conditions of this technique were met, a study was made on
the mass of aerosol deposited by sedimentation on filter paper as a function of time.

The theoretical aspects of the ESIT technique and the evaluation of sediments on filter paper are described in Chapters 2 and 3. Finally, the test results obtained are presented in Chapter 4. Appendix A gives information on fitting a mathematical function to the measured optical density values. Appendix B deals with computerization of the study.

This work was performed at the DREV between March and August 1981 as part of NCP 21B05 "Aerosols."

2.0 OPTICAL TRANSMISSION OF AN AEROSOL UNDERGOING SEDIMENTATION

The theoretical aspect of the technique of particle-size distribution determination is described in this chapter. The rate of particle sedimentation under the influence of gravity is discussed first, followed by transmission of a monodisperse aerosol undergoing sedimentation, and finally, an analytical expression of particle-size distribution for polydisperse aerosols is developed.

2.1 Sedimentation

A spherical particle in an undisturbed viscous medium (absence of temperature gradient, wind, etc.) falls under
the influence of the gravitational field at a constant rate (ref. 1) equal to

\[ v_s = \frac{2\rho g r^2}{\mu} C_s \left( 1 - \frac{\rho}{\rho_p} \right) \]  

(1)

where \( r \) is the radius of the particle in cm.

\( \rho \) is the fluid density g/cm\(^3\).

\( \rho_p \) is the particle density g/cm\(^3\).

\( g \) is equal to 981 cm/s\(^2\).

\( \mu \) is the viscosity of the fluid g/cm.s.

with

\[ C_s = 1 + \frac{\ell}{r} \left( A_1 + A_2 e^{-A_3 r/\ell} \right) \]  

(2)

where \( \ell \) is the mean free path of the molecules in the fluid,

\( A_1 = 1.257 \),

\( A_2 = 0.400 \),

\( A_3 = 0.55 \).

Table 1 gives the value of coefficient \( C \) and the sedimentation rate of particles with different diameters.
TABLE I
TRANSPORTATION PROPERTIES OF AEROSOLS (Ref. 1)
SPHERICAL PARTICLES IN AIR AT 20°C, 1 atm

<table>
<thead>
<tr>
<th>d_p (µm)</th>
<th>C</th>
<th>v_s (cm/s) (ρ_p = 1 g/cm³)</th>
</tr>
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<tbody>
<tr>
<td>0.1</td>
<td>2.85</td>
<td>8.26 x 10⁻⁵</td>
</tr>
<tr>
<td>0.2</td>
<td>1.865</td>
<td>2.62 x 10⁻⁴</td>
</tr>
<tr>
<td>0.5</td>
<td>1.326</td>
<td>1.00 x 10⁻³</td>
</tr>
<tr>
<td>1.0</td>
<td>1.164</td>
<td>3.52 x 10⁻³</td>
</tr>
<tr>
<td>2.0</td>
<td>1.082</td>
<td>1.31 x 10⁻²</td>
</tr>
<tr>
<td>5.0</td>
<td>1.032</td>
<td>7.80 x 10⁻²</td>
</tr>
<tr>
<td>10.0</td>
<td>1.016</td>
<td>3.07 x 10⁻¹</td>
</tr>
<tr>
<td>20.0</td>
<td>1.008</td>
<td>1.22</td>
</tr>
<tr>
<td>50.0</td>
<td>1.003</td>
<td>7.58</td>
</tr>
<tr>
<td>100.0</td>
<td>1.00016</td>
<td>30.3</td>
</tr>
</tbody>
</table>

Thus, it appears that for particles larger than 2 microns, the coefficient C_s is more or less independent of particle diameter and tends toward 1. Hence, for particles larger than two microns, it is possible to obtain a simple analytical expression for the diameter of these particles as
a function of their sedimentation rate, namely:

\[ r = \left( \frac{9 \mu v_s}{2g(\rho_p - \rho)} \right)^{\frac{1}{2}} \]  \hspace{1cm} (3)

2.2 Optical Transmission Through A Monodisperse Aerosol in Particles in Process of Sedimentation

A monodisperse aerosol is an aerosol whose particles are all of the same diameter and hence fall at the same rate. Figure 1 represents optical transmission through a monodisperse aerosol of uniform concentration. At a time \( t_0 \), the aerosol is disseminated instantly. After a time \( t_1 - t_0 \), there is no longer any aerosol above a height \( H - h \) (\( h \) and \( h \) are defined in Figure 3). The sedimentation is given by:

\[ v_s = \frac{h}{t_1 - t_0} \]  \hspace{1cm} (4)

Substituting the value of \( v_s \) in Equation 2, we obtain the radius of the particles of which the aerosol is composed, namely:

\[ r = \left( \frac{9h \gamma}{2g (t_1 - t_0)(\rho_p - \rho)} \right)^{\frac{1}{2}} \]  \hspace{1cm} (5)
Figure 1. Transmission as a function of time of a monodisperse aerosol undergoing sedimentation.
(a) Transmission (%), arbitrary value. (b) Arbitrary unit time.
Now that we know the ratio $I_{\text{min}}/I_0$, where $I_{\text{min}}$ and $I_0$ are the light intensities in the presence and in the absence of aerosol, we can use Beer's law to find out the number of particles (per unit volume) which have been disseminated.

$$I = I_0 e^{-T}$$

where

$$T = \ln \frac{I}{I_0}$$

(6)

where (ref. 1)

$$T = \int_0^\infty K(x_n, n) r^2 n(r)dr dl$$

(7)

where $K(x_n, n)$ is the effectiveness of diffusion,

$n$ is the index of refraction,

$x_n$ is equal to $2\pi r/\lambda$,

$\lambda$ is the wavelength of the incident light,

$n(r)$ is the distribution function,

$r$ is the radius of the particle,

$L$ is the length of the optical path.

The function $K(x_n, n)$ cannot be evaluated with Mie's theory. It is of the oscillating type and tends toward 2 for high values of $x_n$. 

For a monodisperse aerosol, the distribution function can be written with the aid of a Dirac function centered on the radius \( r_p \) of the particles:

\[
p_n(r) = N \delta(r - r_p)
\]

where \( N \) is the number of particles per unit volume.

After integration, and isolating \( N \), it appears that

\[
N = \frac{T}{L \pi r^2 K(x_n, n)} = \frac{T}{2 \pi r^2 L}
\]

for high values of \( x_n \).

Hence, it is possible to determine the size and number of particles of a monodisperse aerosol by studying its sedimentation rate and the optical transmission through the aerosol.

2.3 Optical Transmission through a Polydisperse Aerosol in Process of Sedimentation

An analysis of the foregoing type can be done for a polydisperse aerosol by using summations. However, Deepak
and Vaughan (Ref. 2) developed a more analytical method which is relatively simple to use and gives the distribution function directly.

2.3.1 General Form of Distribution Function

Let us consider an aerosol disseminated instantaneously and distributed uniformly at time \( t = 0 \). After a time \( t \), all particles with a diameter greater than \( r(t) \) would be below level \( h \).

\[
r(t) = \frac{9}{2} \left( \frac{uh}{(\rho_p - \rho) gt} \right)^{\frac{1}{2}} = \left( \frac{a}{t} \right)^{\frac{1}{2}}
\]  

(10)

The law of optical transmission (Beer) becomes a function of time:

\[
\frac{I(t)}{I_0} = e^{-T(t)}
\]  

(11)

where

\[
T(t) = \pi L \int_{r_1}^{r_2} K(x_n, n) r^2 n(r) \, dr
\]  

(12)
at a time $t + \Delta t$, we have:

$$T(t + \Delta t) = \pi L \int_{r_1}^{r_2} K(x, n) \, r^2 \, n(r) \, dr$$  \hspace{1cm} (13)

Using the mean value theorem to evaluate these two integrals and the definition of the derivative, we obtain:

$$\frac{dT(t)}{dt} = \lim_{\Delta t \to 0} \frac{T(t + \Delta t) - T(t)}{\Delta t}$$  \hspace{1cm} (14)

$$= \pi L K(x(t), n) \, n(r(t)) \, r^2(t) \frac{dr(t)}{dt}$$  \hspace{1cm} (15)

From the latter equation, the distribution function can be isolated:

$$n(r(t)) = \frac{dT(t)}{dt} / \pi L \frac{dr(t)}{dt} K(x(t), n) \, r^2(t)$$  \hspace{1cm} (16)
2.3.2 Equation of Distribution Function

If we are to define the equation of the distribution function \([16]\) completely, we must find out the derivatives with respect to time of the optical density and critical radius of the particles.

The choice of a function to represent optical density enables the distribution function to be defined analytically. We chose the following function to fit the experimental points:

\[
T(t) = \frac{T_0}{A(e^{Ct} - 1) + 1} \tag{17}
\]

\(T_0\) is the value of the optical density of \(t = 0\), and the constants \(A\) and \(C\) are determined with the least squares method after transformation of equation 17 (See Appendix A).

The expressions for the derivative are obtained with the aid of equations 17 and 10.

\[
\frac{dT(t)}{dt} = \frac{-ACT_0 e^{Ct}}{(A(e^{Ct} - 1) + 1)^2} \tag{18}
\]

\[
\frac{dr(t)}{dt} = -\frac{1}{2} a^\frac{1}{2} t^{-3/2}
\]

The expression for the distribution function becomes:

\[
n(r(t)) = \frac{2ACT_0 e^{Ct} t^{5/2}}{(A(e^{Ct} - 1) + 1)^2 K(x(t), \ n)x^{3/2}} \tag{19}
\]
It is preferable to express variable $t$ as a function of $r$ to prevent confusion:

$$n(r) = \frac{ACTe^{\frac{\alpha}{r^2}}}{\left(A(e^\frac{\alpha}{r^2} - 1) + 1\right)^2 \pi L r^5} \tag{20}$$

For the latter equation, the diffusion effectiveness $K$ has been established at 2. Equation 20 is valid for particles with radii greater than 1 micron.

3.0 THEORETICAL MASS DEPOSITED AS A FUNCTION OF TIME

The theory described in Chapter 2 which enables the particle-size distribution function to be calculated assumes that the particles undergo sedimentation under ideal conditions, i.e.:

- no agglomeration by diffusion or caused by differences in sedimentation rate of particles with different sizes,
- still air,
- spherical particles,
- negligible loss by diffusion on walls.

One way of detecting the presence of processes affecting aerosol sedimentation rate, and hence delimiting the range of validity of the technique studied, consists of
measuring the actual mass deposited as a function of time and comparing it to the calculated mass.

The mass deposited by sedimentation is calculated for a discrete distribution and then for continuous distribution.

3.1 Calculation of Mass Deposited by Sedimentation: Discrete Distribution

Let $n$ be the number of particles with radius $r$ per unit volume, let $v_s$ be the sedimentation rate, $t$ the time variable, $h$ the height of the aerosol column above the filter at time $t=0$, and $S$ the surface area of the filter paper.

If the filter paper is empty at time $t=0$, the number of particles deposited on the paper after a time $t$ is given by:

$$N_p = nSv_s t \bar{H}(t - h/v_s)$$

(21)

The function $\bar{H}(t - h/v_s)$ is a descending step function; it has the value 1 for times shorter than $h/v_s$ and value 0 for times greater than $h/v_s$. This function represents the fact that, after time $h/v_s$, all the particles are deposited.

If the filter paper is empty at a time $t_1$, the expression for the number of particles deposited after time $t$ is
If we now consider particles with different diameters, expression 22 becomes

\[ N_p = (t - t_1) \sum_{i=1}^{P} n_i v_i \bar{H}(t - \frac{h}{v_i} - t_1) \]  

(23)

where \( n_i \) is the number of particles per unit volume, and \( v_i \) is the sedimentation for particles of type \( i \).

The mass deposited rather than the number of particles on the filter paper is measured. The mass deposited is equal to the mass flux through surface \( A \) multiplied by elapsed time, namely,

\[ m = \frac{8\rho_p^2}{27} (t - t_1) \frac{Sg}{\mu} \sum_{i=1}^{P} n_i r^5 \bar{H}(t - \frac{9h\rho}{2\rho_p g r_i^2} - t_1) \]  

(24)

where the velocity \( v_1 \) has been replaced by the expression \[ \frac{2\rho_p g r^2}{\mu} \] and \( r \) is the particle radius.

3.2 Calculation of Mass Deposited by Sedimentation: Continuous Distribution

Let \( n(r) \) be a particle-size distribution which is continuous with respect to particle radius such that:
where \( N \) is the total number of particles. The number of particles deposited on the filter paper at a time \( t \) when the latter has been uncovered at a time \( t_1 \) after the beginning of sedimentation is given by:

\[
N_p = (t - t_1) \int_0^\infty n(r) \nu_s(r) \bar{H} \left( t - \frac{h}{\nu_s(r)} - t_1 \right) dr
\]  

Hence, the mass deposited on the filter paper is:

\[
m = \frac{8\pi \rho^2}{27} (t - t_1) \frac{g \rho_s}{\mu} \int_0^n(r) r^5 \bar{H} \left( t - \frac{9h\mu}{2\rho_p g r^2} - t_1 \right) dr
\]  

Time \( t_1 \) corresponds to a maximum radius \( r_M \) equal to \( \left( \frac{g h \mu}{2 \rho_p g t_1} \right)^{\frac{1}{2}} \), which implies that at time \( t_1 \) the particles whose radius is greater than \( r_M \) are already deposited on the plate covering the filter paper. At a time \( t_F \), where the filter paper is once again "hidden", all particles with radius greater than \( r_F = \left( \frac{g h \mu}{2 \rho_p g t_F} \right)^{\frac{1}{2}} \), but less than \( r_M \), are deposited. Their contribution to the mass of sediment on the filter is:

\[
m_{(r_M > r > r_F)} = \frac{8\pi \rho^2 g \rho_s}{27 \mu} \int_0^{r_F} \left( \frac{h}{\nu_s(r)} - t_1 \right) n(r) r^5 dr
\]
where

\[ v_g(r) = \frac{2 \rho p g r^2}{\mu} \]

Particles smaller than \( r_F \) are deposited continuously over the interval \([t_1, t_F]\). Their contribution to the total mass deposited on the filter paper is:

\[ m(r < r_F) = \frac{8g}{27} \rho p^2 (r_F - t_1) \frac{g}{\mu} \int_0^{r_F} n(r)r^5 dr \]  

The total mass deposited on the filter is hence written:

\[ m = m(r < r_F) + m(r > r_F) \]  

3.3 **Choice of Distribution Function**

The mass deposited on the filter paper can be evaluated if the particle-size distribution function is known. It should be noted that the converse is also true. The distribution function can be determined by measuring the mass deposited as a function of time (ref. 3).

Two distribution functions are presented in this section. The first was established with a model TA II Coulter counter (by Coulter Electronics, Inc.) before dissemination of the powder. The second was measured with the ESIT method.
3.3.1 Distribution Function Measured by Coulter Counter

The Coulter counter measures the changing conductivity of an electrolyte placed between two electrodes when a particle passes between them. The signal recorded is proportional to the volume of the particle. After data discrimination and processing, the computer furnishes the percentages of the mass of all the particles analyzed which have a diameter smaller than $d$.

An initial distribution measurement (before dissemination) of the particles in terms of the percentage of the mass of particles whose diameter is less than $d$ was obtained for powdered glass No. 3419-2. The measured density of these glass balls is 4.2 g/cm$^3$. The particle-size distribution in terms of mass can be represented by a distribution function of the log probability type (Figure 2).

\[
F(m) = \frac{M}{\sqrt{2\pi} \log \sigma_m} e^{-\frac{(\log d - \log d_m)^2}{2 \log^2 \sigma_m}}
\]

where $F(m)$ is the mass of the particles with diameter $d$,

$M$ is the total mass of particles,

$\sigma_m$ is the geometric standard deviation by mass,

$d_m$ is the mean geometric diameter by mass.
50% of the particles have a smaller mass than that of a particle with diameter $d$.

![Graph showing particle distribution](image)

**Figure 2** - Particle distribution according to a log probability scale. (a) Diameter (D), microns. (b) Percentage of mass below D.

The curve in Figure 2 as determined by the least squares method best represents the values measured with the Coulter counter.

Thus, using the mathematical expression representing the best curve, or proceeding graphically, we find a mean diameter per mass of 10.5 microns. The geometric standard
deviation per mass is calculated as follows (Ref. 4):

\[
\sigma_m = \frac{d_{84.13\%} - d_{50\%}}{d_{50\%} - d_{15.87\%}} \tag{32}
\]

where the diameters indicated correspond to 15.87\%, 50\% and 84.13\% of the mass. The mass is 2.42 for #3419-2 powdered glass.

### 3.3.2.1 Log Probability Type Distribution in Terms of Number of Particles

In the preceding section, a log probability distribution in terms of mass was adjusted to the measurements of the Coulter counter. This distribution in terms of mass must be converted into distribution in terms of number of particles.

The number of particles with diameter \(d\) per unit volume is given by:

\[
F(d) = \frac{N}{\sqrt{2\pi} \cdot \log \sigma_g} e^{-\frac{(\log d - \log d_g)^2}{2 \cdot \log^2 \sigma_g}} \tag{33}
\]

where \(N\) is the total number of particles per unit volume,

\(\sigma_g\) is the geometric standard deviation by number of particles,

\(d_g\) is the mean geometric diameter.
50% of the particles have a diameter smaller than \( d_g \).

The geometric standard deviation by mass is the same as the standard deviation by number (Ref. 4). The mean geometric diameter by number is linked to the mean geometric by mass by the relation:

\[
\log d_g = \log d_m - 6.9078 \log \sigma_g
\]  \hspace{1cm} (34)

Using the values for \( d_m \) and \( \sigma_g \) of the previous section, we obtain a value of 1.82 microns for the mean geometric diameter by number.

3.3.1.3 Relation between function \( F(d) \) and Function \( n(r) \)

The function \( F(d) \) cannot be used directly to evaluate deposits on filter papers. In fact, as well as the change in variable from \( d \) to \( r \), we must remember that a log probability distribution is the result of another change in variable (Ref. 5):

\[
n(r)dr = \frac{N}{\sqrt{2\pi} \log \sigma_g} e^{- \frac{(\log r - \log r_g)^2}{2 \log \sigma_g}} d\log r
\]  \hspace{1cm} (35)
hence

\[ n(r) = \frac{F(r)}{r} \cdot \log e \quad (36) \]

The number of particles per unit volume \( N \) is unknown, and will be discussed in the next section.

3.3.1.4 Particle Density in Dissemination Chamber

Evaluation of the mass deposited on the filter papers calls for knowledge of the function \( n(r) \). The relative proportions of the number of particles was established with the Coulter counter but their density is unknown.

A mass of powder \( M_i \) is disseminated in the chamber at time \( t_D \) for a period of 30 s. During and after dissemination, during another period of 30 s, the inside of the chamber is turbulent because the mixing fans are on. Particle losses occur during mixing and, as a result, the mass of powder disseminated is no longer \( M_i \) but rather \( M_p \). The mass \( M_p \) is determined by measuring the mass \( M_{fl} \) of aerosol deposited by sedimentation on a filter paper uncovered at the beginning of sedimentation.

\[ M_p = \frac{h}{H-h} \cdot \frac{S_c}{S} \cdot M_{fl} \quad (35) \]
where $S_c$ and $S$ are the four areas of the chamber and the filter paper and the heights $h$ and $H$ are defined in Figure 3.

The particle density is given by

$$N = \frac{M/V}{\rho \ c} = \frac{4 \pi \rho C}{3} \int_0^3 G(r) r^3 dr$$

(36)

where $V_c$ is the chamber volume and

$$G(r) = \frac{(\log r - \log r_g)^2}{2 \log r_g}$$

(37)

3.3.2 Calculation of Mass Deposited Using the Distribution Function Measured by the ESIT Method

Calculation of the mass deposited on filter papers using the distribution function measured by the ESIT method is relatively simple. One need only substitute the distribution function $n(r)$ established in equation 20 in equations 28 and 29.

It should be noted that, to calculate the mass deposited, it is not necessary to determine the particle density as this is included implicitly in the expression of $n(r)$. 
4.0 MEASUREMENTS AND DISCUSSION

A particle-size determination method was presented in Chapter 2 and in Chapter 3 evaluation of mass deposits by sedimentation was formulated mathematically. In this chapter, the experimental and theoretical results are presented and discussed.

4.1 Setup and Experimental Method

Figure 3 illustrates the setup used. Despite the fact that the ceiling of the silo is semi-hemispherical in form, it was considered that the aerosol was distributed uniformly in a cylinder 11.5 m high. An He-Ne laser (632.8 nm, 15 mW dc) by Spectra Physics, and a Model RP313-1 laser precision pyroelectric detector whose aperture was reduced to 0.5 cm diameter were placed 10 m from the ceiling of the chamber. The detector was coupled to a Model RK 3441, power ratiometer by Laser Precision; this instrument was connected to a graphic recorder.

Ten Gelman filter papers with diameters 4.7 cm were placed in cylindrical boxes 12 cm deep. Each of the ten boxes was covered with an aluminum plate. Nine of the filter papers were uncovered one by one at different times \( t_N \) with the aid of strings attached to the aluminum plates. The tenth filter paper served as a reference and was used to determine the measurement error of the mass deposited.
The aerosol was disseminated with nitrogen at a pressure of 1.03 MPa. The aerosol was in a plastic bottle whose neck was perforated with small holes. This was placed 10 m from the floor.

The sequence of events during an experiment was the following:

- switch on mixing fans at $t_M'$
- disseminate aerosol for 30 s at $t_D$,
- switch off fans at time $t_s$, namely 30 s after the end of dissemination
- uncover filter papers at time $t_N'$
- evacuate silo at time $t_F$.

Since the height/diameter ratio of the boxes is high, the deposits on the filter papers are undisturbed when the silo is evacuated at time $t_F$.

Two sedimentation experiments were performed with powdered glass 3419-2 under the conditions described in Section 1.1. Table II presents the operating conditions for the two experiments.
Figure 3. Experimental setup.
(a) Expansion valve, 150 psi. (b) Nitrogen cylinder. (c) Plastic bottle perforated with holes. (d) Mixing fan. (e) Detector. (f) Cover. (g) Box. (h) Filter paper. (i) Thumbtack. (j) He-Ne source. (k) Graphic recorder.
Table II

<table>
<thead>
<tr>
<th>Mass dispersed on Filter I</th>
<th>Mass collected (g)</th>
<th>Operating Time (s)</th>
<th>Duration of experiment (s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>E1</td>
<td>1310</td>
<td>0.0217</td>
<td>30</td>
</tr>
<tr>
<td>E1</td>
<td>995</td>
<td>0.0112</td>
<td>90</td>
</tr>
</tbody>
</table>

4.2 Optical Transmission and Distribution Established by ESIT Method

Figure 4 represents the optical transmission through the powdered glass 3419-2 undergoing sedimentation as a function of time in the quantities given in Table II. Figure 5 represents the optical density in the natural base, namely, $-\ln \frac{I}{I_0}$. The curves drawn in continuous lines are the best representing the experimental points according to mathematical expression 17. The details of calculation of the coefficients of the best curve are given in Appendix A.
Figure 4. Transmission as a function of time of an He-Ne laser beam (0.6328 microns) through powdered glass #3419-2 undergoing sedimentation.

(a) Transmission (%). (b) Time (sec).

E1: 1310 g disseminated, mixing period after dissemination: 30 sec.

E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figure 5. Variation as a function of time of optical density produced by dissemination of powdered glass #3419-2.
(a) Optical density (-ln I/I₀) (b) Time (sec).
E1: 1310 g disseminated, mixing period after dissemination: 30 sec.
E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figure 6. Distribution functions obtained by ESIT method and with Coulter counter.

(a) Distribution functions (cm$^{-4}$)  (b) Particle diam. (cm)
(c) Coulter counter  (d) ESIT

El: 1310 g disseminated, mixing period after dissemination: 30 sec.

E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figure 7. Distribution functions obtained by ESIT method with Coulter counter.
(a) Distribution functions (cm$^{-4}$)  (b) Particle diam. (cm)
(c) Coulter counter (d) ESIT
E1: 1310 disseminated, mixing period after dissemination: 30 sec.
E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figures 6 and 7 represent, on different scales, the distribution functions measured by the ESIT Method (Equation 20) using the mathematical expressions of the best curves of optical density. The initial distribution functions of the powders before dissemination determined by a Coulter counter and calculated in Section 3.3.1.3 were plotted in continuous lines. It will be noted that for values with high diameters, the distribution curves are parallel and relatively close to each other. The curves begin to have a different shape at 10 microns. It should be noted that, for diameters smaller than 4.6 microns, the distribution functions obtained by the ESIT Method come from extrapolation of the optical density curves.

4.3 Comparison of Experimental and Theoretical Sediments

Figures 8, 9, and 10 represent the experimental sediments measured and those established with the distribution functions measured by the Coulter counter and ESIT Method. The curves of the experimental sediments intersect each other; however, this intersection is insignificant because it is within the margin of error. The curves of the sediments calculated with the distribution functions established by the ESIT Method also intersect; this is because the distribution functions intersect.

Figures 11 and 12 regroup the results presented in Figures 8, 9, and 10 by experiment. The sediments obtained
Figure 8. Sediment on filters as a function of start of exposure, with maximum exposure time 5430 sec.
(a) Time (sec) (b) weight (g)
E1: 1310 g disseminated, mixing period after dissemination: 30 sec.
E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figure 9. Sediment (calculated with Coulter counter) on filters as a function of start of exposure, with maximum exposure time 5430 sec.

(a) Time (sec), (b) weight (g)

E1: 1310 g disseminated, mixing period after dissemination: 30 sec.
E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figure 10. Sediment (calculated by ESIT method) on filters at beginning of exposure time, with maximum exposure time of 5430 sec.
(a) Time (sec) (b) weight (g)
E1: 1310 g disseminated, mixing period after dissemination: 30 sec.
E2: 995 g disseminated, mixing period after dissemination: 90 sec.
Figure 11. Comparison of experimental and theoretical sediments.
(a) Experiment El, 1310 g disseminated, mixing time after dissemination 30 sec. (b) weight (g) (c) experimental sediments (d) theoretical sediments, Coulter counter (e) theoretical sediments, ESIT method (f) time (sec)
in experiment E1, and those obtained with the aid of the distribution function measured with the Coulter counter are in relatively good agreement for the filter papers uncovered at the beginning of sedimentation. The agreement is less good for the results of Experiment E2. The mixing fans operated for an additional 60 s during experiment E2, which favored losses on walls and particle agglomeration. Since the particles were larger, they fell more rapidly and hence the quantity of sediment was larger.

The relative discrepancy between the experimental and theoretical sediments (Coulter counter) increases with time. This is due to agglomeration by diffusion and agglomeration produced by the difference in falling rate of particles with different sizes. Hence, the ratio between the experimental and theoretical sediments is greater than 2 after 1,000 s.

Agglomeration by diffusion of a monodisperse aerosol can be represented mathematically by the following expression:

\[ \frac{dN}{dt} = -\sigma v N^2 \]  

(38)

where \( N \) is the particle density, \( \sigma \) is the collision cross-section and \( v \) is the brownian motion of the particles. The characteristic agglomeration time by diffusion is given
Figure 12. Comparison of experimental and theoretical sediments. (a) Experiment E2, 995 g disseminated, mixing period after dissemination 90 sec. (b) weight (g) (c) experimental sediments (d) theoretical sediments, Coulter counter (e) theoretical sediments, ESIT method (f) time (sec)
by:

\[
T_c = (\sigma v N)^{-1} \\
= N \pi d^2 \frac{2kT}{\rho_p \sigma d^{3/6}}
\]  

(39)

Thus, for \( N = 6.4 \times 10^3 \) cm\(^{-3}\), \( d = 1.82 \) microns, and at room temperature, \( T_c \) is approximately \( 1 \times 10^4 \) s, which is a very long period of time. This leads to the conclusion that, due to the relatively broad particle distribution and the height of the silo, agglomeration by diffusion is far less than that caused by the difference in falling rate of particles with different sizes. With reference to this fact, the calculations by Friedlander, (ref. 1) show that the diffusion agglomeration process is dominant for particles with radii smaller than 1 micron and that agglomeration caused by the difference in falling rate predominates for particles with diameters larger than 1 micron. Note that these calculations are based on the interaction of particles with radii 1 micron with particles with radii varying between 0.1 and 10 microns.

4.4 Discussion

It was shown in the previous section that the measured sediments and the calculated sediments (Coulter counter) diverge due to particle agglomeration.
Since agglomeration is a cumulative process its effect is less significant at the beginning of the experiment. This explains why the results before 1000 s agree relatively well with the theoretical value of the deposits.

The ESIT Method is not valid if there are interactions between particles. Hence, the results obtained with this technique are no longer valid after 1000 s. After this time, all the particles larger than 10 microns are deposited. As a result, the distribution functions obtained by the ESIT Method are not valid for particles whose diameters are smaller than 10 microns.

5.0 CONCLUSION

The limits of the ESIT technique for determining particle size were established by comparing theoretical sediments and experimental sediments on filter papers as a function of time. The experiments show that the considerable height of the silo appreciably increases the particle sedimentation time so that agglomeration becomes important. The distribution function obtained is not valid for particles smaller than 10 microns. It would be possible to determine the sizes of particles smaller than 10 microns by moving the laser and the detector to the upper end of the dissemination chamber. The height of the aerosol column above the measurement line would be smaller and the agglomeration caused by the difference in particle falling
rate would be less important. Moreover, since the experiment would last a shorter time, agglomeration by diffusion would be less.

6.0 ACKNOWLEDGEMENTS

The author would like to give special thanks to Mr. A J. Evans, CDE, Porton Down, England, working at the DREV under a TTCP scholarship from 1980 to 1981, who gave useful technical advice during these experiments. The author also wishes to thank Mr. P. Pelletier from the Explosives Section, Propulsion Division, who set up a program enabling a line to be fitted on a log probability scale.
APPENDIX A

Best curve with shape \( Y = A_f + \frac{A_0 - A_f}{A(e^{Ct} - 1) + 1} \)

We will first attempt to linearize this equation and then determine the coefficients giving the minimum standard deviation.

A1.1 Linearization of Equation

Let
\[
Y = A_f + \frac{A_0 - A_f}{A(e^{Ct} - 1) + 1}
\]

[A-1]

where
- \( t \) is the time variable
- \( A_0 \) is the value of \( y \) when \( t = 0 \)
- \( A_f \) is the value of \( y \) when \( t = \infty \)
- \( A \) and \( C \) are coefficients to be determined.

Modifying [A-1], we have:
\[
Y_+ = \frac{A_0 - A_f}{Y - A_f} = A(e^{Ct} - 1) + 1
\]

[A-2]
Deriving the expression on the right

\[ Y'_4 = ACe^{Ct} \quad \text{[A-3]} \]

and taking the natural logarithm of the latter equation, we obtain:

\[ Z = \ln Y'_4 = Ct + D \quad \text{or} \quad D = \ln AC \quad \text{[A-4]} \]

which is a linear equation.

**Al.2 Calculation of Coefficients for a Minimum Standard Deviation**

For a linear equation of type \( Z = Ct + D \), the coefficients for a minimum standard deviation are calculated as follows (ref. 6):

\[
C = \frac{s_0 \mu_1 - s_1 \mu_0}{s_0 \sigma_2 - s_1^2}, \quad D = \frac{s_2 \mu_0 - s_1 \mu_1}{s_0 \sigma_2 - s_1^2} \quad \text{[A-5]} \]

where

- \( s_0 \) is the number of experimental points,
- \( s_1 \) is \( \sum t'_1 \),
- \( s_2 \) is \( \sum t'_1^2 \),
- \( s_0 \) is \( \sum z'_1 \),
- \( s_1 \) is \( \sum t'_1 z'_1 \).
However, in the case which concerns us, some explanations need to be given.

The value of coefficient $A_0$ is given directly by the coordinates of the first experimental point $(0, y_{1s})$, i.e. the optical density at time $t_0$. When the time tends toward infinity, all the particles are deposited and hence coefficient $A_f$ equals zero.

Equations A-3 and A-4 suggest that first its slope and then its logarithm be taken to linearize the equation. In concrete terms, we have equation

$$z_i = \ln A_0 \frac{1/y_{i+1} - 1/y_i}{t_{i+1} - t_i}$$

where $y_i$ and $y_{i+1}$ are experimental points taken at $t_i$ and $t_{i+1}$. It should be noted that since the slope has to be calculated the number of $z_i$ is equal to $n - 1$ where $n$ is the number of experimental points.
APPENDIX B

Computerization of ESIT Method

Enter experimental points on computer with point reader

Correct experimental points against all rotation errors or translation errors produced by poor positioning of the sheet which contains the experimental points, on the point reader

Calibrate time axis and normalize transmission.

Calibrate optical density and calculate coefficients of best curve

Specify the following parameters:
- length of optical path
- height above detector
- density of material of which particles are composed
- viscosity of air

Calculate particle diameter
Calculate distribution function
The results of a feasibility study for determining aerosol size distribution using the extinction-sedimentation inversion technique are presented. It was found that the height of the dissemination chamber may affect the value of the results.