

AD 626228

AFCRL-65-836

MIE SCATTERING COEFFICIENTS FOR SELECTED AEROSOL SIZE DISTRIBUTIONS

Final Report

1 April 1964 through 31 October 1965

Prepared by

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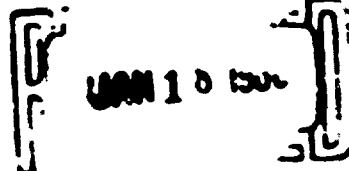
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ABSTRACT

Formulas are derived for volume scattering coefficients applicable to aerosols. Different types of size distribution for the aerosol, such as normal distribution, log-normal distribution, and exponential distribution, are assumed.

Numerical results obtained by the author and those published in the literature are given and discussed.

The volume scattering coefficients become smooth functions if the size distribution is very wide. Hence, simpler mathematical formulations can be used than those obtained from the exact Mie theory. However, only the number of aerosol particles can be obtained from an evaluation of experimental data, but details of the size distribution cannot be extracted from experimental data.

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1. INTRODUCTION

The scattering functions for single particles with real index of refraction n are known for many values of n , and a very large range of size parameters a ($a = 2\pi r/\lambda$). Total scattering coefficient, angular scattering coefficients, and degree of polarization are known. Because of the electronic computer technique, such computations pose no problem and scattering coefficients can be generated for any desired value of n and size range. For aerosol studies, where integrations over a particle size range and wavelength range are needed, the minor fluctuations, the so-called ripples, are unimportant. Smoothed values without ripples are often completely sufficient. Hence, this problem is well understood (van deHulst, 1957).

The scattering, absorption, and polarization coefficients for single particles with complex index of refraction $\tilde{n} = n - i\kappa$ are also known for many values of \tilde{n} and a . However, less extensive data have been generated than for real n . The results are well understood; absorption reduces the fluctuations, the ripples vanish first, and the major fluctuations become weaker the larger the absorption, i. e., κ . There exists a steep increase of the scattering coefficients in the Rayleigh range for $a < 1$ and a very simple functional behavior for $a > 1$. This applies to the total as well as to the angular scattering coefficients. Again the problem is understood; data exist to derive the general behavior and machine computations pose no difficulties (Deirmendjian, 1963).

The most important problem of today is the scattering from a small volume containing an aerosol of uniform refractive index \tilde{n} or n . The particles inside the volume are not of uniform size, but possess a size distribution. The size distribution can be described by reasonably simple functions. As such, one may assume an exponential law (as in Junge's distribution function), or a Gaussian distribution function, or any other which has been proven to exist (Deirmendjian, 1963; Dettmar et al, 1963). A literature survey was published by Fenndorf (1963).

In this report we discuss some theoretical derivations for representing the scattering coefficients for assumed size distributions and show some of the results of such computations.

2. TOTAL AND ANGULAR VOLUME-SCATTERING COEFFICIENTS

The total volume scattering coefficient β_T for an aerosol with uniform size distribution is

$$\beta_T = \pi r^2 K \{a, n\} N \quad (1)$$

where

r = radius of aerosol particle

a = size parameter; $a = (2\pi r)/\lambda = kr$

N = number of particles of radius r

n = refractive index of aerosol particle

K = total Mie scattering coefficient.

The angular volume scattering coefficient β_θ is correspondingly

$$\beta_\theta = \pi r^2 i_\theta \{a, n, \theta\} N \quad (2)$$

where i_θ is the angular Mie scattering coefficient for a single particle in the direction θ ; $\theta = 0$ degree for forward scattering.

The relation between i_θ and K is (see Penndorf, 1960)

$$\int_0^{4\pi} i_\theta d\omega = K \quad (3)$$

Hence

$$\int_0^{4\pi} \beta_\theta d\omega = \beta_T \quad (4)$$

In the following, the formulas are written for K but by replacing K with i_θ the results for β_T follow automatically for β_θ ; we used the symbol β for β_T in the following for simplification only.

The computations of the volume-scattering coefficient for an aerosol containing particles of non-uniform sizes; i. e., and aerosol with a specific size distribution follows from

$$\frac{d\beta}{da} = \pi r_i^2 K |a_i, n| \frac{dN_i}{da} \quad (5)$$

where r_i = radius at $r \pm (\frac{1}{2}) \Delta r$ or $a \pm (\frac{1}{2}) \Delta a$, and dN_i/da = number of particles between $a_i \pm \Delta a$.

This can be written as

$$\frac{d\beta}{da} = \pi a_i^2 \frac{1}{k_i^2} K \frac{dN}{da} = \frac{\pi a^2}{k_i^2} K \frac{dN}{da} \quad (6)$$

It is understood that all these considerations refer to a specific wavelength λ , and the integration over the selected wavelength range should be performed if a wide band is chosen. We will come back to it later. It is also understood that n remains unchanged for the full wavelength range selected.

We write the size distribution function $p |a|$ as

$$\frac{dN}{dr} = p |r| \quad (7)$$

or with $dr = \frac{1}{k} da$

$$\frac{dN}{da} = \frac{1}{k} p |a| \quad (8)$$

The total number of particles follows from equation (8) as

$$N = \int dN = \frac{1}{k} \int_0^{\infty} p |a| da \quad (9)$$

For $p |a|$ we shall later on introduce specific population functions.

Using equation (8) we can now write

$$\frac{d\beta}{da} = \frac{\pi a^2}{k^3} K p |a| \quad (10)$$

a relationship which holds for a fixed interval da and a fixed wavelength λ .

The integration over all a yields

$$\beta = \frac{\pi}{k^3} \int_0^{\infty} a^2 K |a| p |a| da . \quad (11)$$

In this form β is valid for a specific λ only, because k contains λ .

Next we define a new total Mie coefficient K_p for a size distribution, so that equation (1) can be used

$$\beta = \frac{\pi a_0}{k^2} K_p |a_0| N . \quad (12)$$

This equation (12) is identical to equation (11) and it follows

$$K_p |a_0| = \frac{1}{a_0^2 k N} \int_0^{\infty} a^2 K |a| p |a| da . \quad (13)$$

In this form a_0 denotes the a value at which the distribution function reaches a maximum; i. e., dN/da reaches a maximum.

Introducing N from equation (9) into equation (13) leads to

$$K_p |a_0| = \frac{1}{a_0^2} \frac{\int_0^{\infty} a^2 K |a| p |a| da}{\int_0^{\infty} p |a| da} . \quad (14)$$

In the case of angular scattering consideration we define an angular Mie scattering coefficient as

$$i_{\theta p} |a_0| = \frac{1}{a_0^2} \frac{\int_0^{\infty} a^2 i_{\theta} |a| p |a| da}{\int_0^{\infty} p |a| da} \quad (15)$$

3. AVERAGE SIZE

3.1 AVERAGE SIZE \bar{a} FOR $p(r)$

The average radius \bar{r} of particles having a size distribution $p(r)$ is the arithmetic mean value of all the radii, if N is the total number in the sample.

Thus

$$\bar{r} = \frac{1}{N} \sum_i (r_1 + r_2 + \dots + r_i) = \frac{1}{N} \sum_i r_i (\Delta N_i) \quad (16)$$

Replacing r by a/k and the sum by the integral leads to

$$\bar{a} = \frac{1}{kN} \int_0^{\infty} a p(a) da \quad (17)$$

Since

$$N = \frac{1}{k} \int_0^{\infty} p(a) da$$
$$\bar{a} = \frac{\int_0^{\infty} a p(a) da}{\int_0^{\infty} p(a) da} \quad (18)$$

3.2 AVERAGE SIZE \bar{a} FOR NORMAL DISTRIBUTION

The normal distribution is

$$p(a) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left(-\frac{u^2}{2\sigma^2}\right) \quad (19)$$

with

$$u = (a - a_0)/\sigma \quad \text{and} \quad a = \sigma u + a_0$$

Equation (18) can be solved

$$\int_0^{\infty} ap|a|da = \frac{a_0}{\sigma} + \sqrt{\frac{2}{\pi}}$$

and

$$\int_0^{\infty} p|a|da = \frac{1}{\sigma}$$

Hence

$$\bar{a} = a_0 + \sigma \sqrt{\frac{2}{\pi}} = a_0 + 0.798 \sigma \quad (20)$$

Thus, for a normal distribution, the average size is determined by the a_0 and the standard deviation σ . \bar{a} is always larger than a_0 .

4. K_p AND $i_{\theta p}$ FOR NORMAL DISTRIBUTION

We select a population function for which the aerosol particles have a normal distribution with respect to a . The total number of particles in the sample is N .

$$p(a) = \frac{N}{\sigma\sqrt{2\pi}} \exp(-u^2/2\sigma) = (N/\sigma) \phi(u) \quad (21)$$

where $\phi(u)$ denotes the normal distribution.

The maximum of the distribution is given by

$$p(a_0) = (N/\sigma\sqrt{2\pi}) = (0.399 N/\sigma) \quad (22)$$

Equation (14), using the result of the last section, namely

$$\int_0^{\infty} p(a) da = 1/\sigma$$

becomes

$$K_p(a) = \frac{\sigma}{a_0^2} \int_0^{\infty} a^2 K(a) p(a) da \quad (23)$$

or

$$K_p(a_0) = \frac{N}{a_0^2} \int_0^{\infty} a^2 K(a) \phi\left\{\frac{1}{2}\left(\frac{a-a_0}{\sigma}\right)^2\right\} da$$

The integral has to be solved numerically because K is not a simple function of a ; $\phi(u)$ is tabulated in standard text books on statistics.

In the case of angular scattering consideration, K_p is replaced in equation (23) by $i_{\theta p}$ and K by i_{θ} . The general formalism remains unaltered.

In many aerosols the so called logarithmic distribution (log-normal) has been found. In this case the quantity u in equation (21) is defined as

or $u = [\ln(a/a_0)]/\sigma$

$$a = a_0 \exp(\sigma u). \quad (24)$$

We can solve $\int_0^\infty p|a| da$ which is

$$\int_0^\infty p|a| = \frac{N}{\sigma\sqrt{2\pi}} \int_0^\infty \exp\left(-\frac{1}{2\sigma^2} [\ln(a/a_0)]^2\right) da \quad (25)$$

Such an equation can be brought to the form $\int_0^\infty e^m e^{-Am^2} dm$ for which a solution is given by Gröbner and Hofreiter (1950, p. 57)

$$\int_0^\infty p|a| = N a_0 \left[\exp\left(\frac{\sigma^2}{2}\right) \right] [1 - \phi(-\sigma/\sqrt{2})] \quad (26)$$

with $\phi(x) = \int_0^x e^{-t^2} dt$, the error integral, which is tabulated in standard textbooks.

The solution for angular scattering is now obtained from equations (15) and (26) as

$$i_{\theta_p} |a_0| = \frac{1}{a_0^3 \sigma \sqrt{2} [\exp(\sigma^2/2)] [1 - \phi(-\sigma/\sqrt{2})]} \int_0^\infty a^2 i_{\theta_p} \exp\left(-\frac{1}{2\sigma^2} \left[\ln \frac{a}{a_0}\right]^2\right) da \quad (27)$$

Because of the Mie function in i_{θ_p} , a numerical evaluation of the integral has to be performed.

5. EXPONENTIAL POPULATION FUNCTION

Junge (1963) has shown that a good agreement for the natural aerosol in the range $0.1 < r < 20\mu$ is given by

$$\frac{dN}{d(\log r)} = N_0 r^\gamma \quad (28)$$

A good agreement is normally found for $\gamma = -3$. In all cases γ is negative and $|\gamma| \geq |1|$.

Such a function leads to

$$\frac{dN}{dr} = N_0 (\log e) r^{\gamma-1}$$

Replacing r by a

$$p(a) = \frac{dN}{da} = N_0 (\log e) \left(\frac{1}{k}\right)^\gamma a^{\gamma-1} = CN_0 a^{\gamma-1} \quad (29)$$

Such an exponential size distribution is only valid within specified limits of r (and therefore a). Hence,

$$K_p(a_0) = \frac{1}{a_0^2} \frac{\int_{a_1}^{a_2} a^2 K(a) p(a) da}{\int_{a_1}^{a_2} p(a) da} \quad (30)$$

The denominator can be written as

$$\int_{a_1}^{a_2} p(a) da = CN_0 \int_{a_1}^{a_2} a^{\gamma-1} da = \frac{CN_0}{\gamma} (a_2^\gamma - a_1^\gamma)$$

Since the number of particles at the upper end (large r) a_2 is very small and determined by the limitations of the instrument, we can set the integral equal to

$$= \frac{CN_0}{\gamma} a_1^\gamma$$

Thus equation (30) becomes

$$\begin{aligned}
 K_p \{a_0\} &= - \frac{\gamma}{a_0^2 C N_0 a_1^\gamma} \int_{a_1}^{a_2} a^2 K \{a\} p \{a\} da \\
 &= - \frac{\gamma}{a_0^2 a_1^\gamma} \int_{a_1}^{a_2} a^2 K \{a\} a^{\gamma-1} da \\
 &= - \frac{\gamma}{a_0^2 a_1^\gamma} \int_{a_1}^{a_2} K \{a\} a^{\gamma+1} da .
 \end{aligned} \tag{31}$$

For selected values of γ , it becomes even simpler. For $\gamma = -1$, we obtain

$$K_p \{a\} = \left(\frac{a_1}{a_2} \right) \int_{a_1}^{a_2} K \{a\} da$$

Since in this population function $a_0 = a_1$

$$K_p \{a_1\} = \frac{1}{a_1} \int_{a_1}^{a_2} K \{a\} da \tag{32}$$

For $\gamma = -2$ we obtain

$$K_p \{a_1\} = 2 \int_{a_1}^{a_2} a^{-1} K \{a\} da \tag{33}$$

For $\gamma = -3$ we obtain

$$K_p \{a_1\} = 3 a_1 \int_{a_1}^{a_2} a^{-2} K \{a\} da \tag{34}$$

and for $\gamma = -4$

$$K_p \{a_1\} = 4 a_1^2 \int_{a_1}^{a_2} a^{-3} K \{a\} da . \quad (35)$$

Similar formulas can be written for $i_{\theta p}$; for example, for $\gamma = -3$ one obtains

$$i_{\theta p} \{a_1\} = 3 a_1 \int_{a_1}^{a_2} a^{-2} i_{\theta} \{a\} da . \quad (36)$$

It follows from the foregoing theoretical considerations that we can compute Mie scattering functions for aerosols, provided the general size distribution law can be assumed from other observations or general physical considerations. If, however, no knowledge on the general size distribution law exists, the experimental observations cannot be interpreted.

6. RESULTS OF COMPUTATIONS

Using the theoretical derivations, we show in several examples what values are obtained for K_p and i_{θ_p} .

For a normal distribution, equation (23), we choose $a_0 = 5$, $\sigma = 1.0$, and $da = 0.2$, and use the K values for $n = 1.33$ obtained by Penndorf (1956).

In this case we obtain $K_p(5.0) = 3.69$. The value for $K(5.0) = 3.59$. In table I we give some examples for $n = 1.33$ and $a_0 = 6.0(0.2) 7.0$.

TABLE I

TOTAL SCATTERING COEFFICIENTS K_p FOR
SOME SIZE DISTRIBUTIONS
WITH $\sigma = 1.0$

a_0	6.0	6.2	6.4	6.6	6.8	7.0
K_p	3.84	3.82	3.83	3.75	3.70	3.63
K	3.89	3.89	3.96	3.96	3.84	3.74

As expected, smooth variations in K_p result for size distributions.

Kerker (1964) conducted experiments and theoretical computations for refractive indices ($n = 1.43, 1.51, \text{ and } 2.07$), a size range $a_0 = 1.9(0.1) 15$, and width parameter $\sigma_0 = 0.000(0.05) 0.155$.

He used a logarithmic normal distribution and a_0 and σ_0 are the parameter for such a size distribution.

Experimental data are obtained and compared with the theoretical computations. He found that the experimental angular scattering data for the two polarized components in the range $\theta = 30$ to 130 degrees can be fitted to the theoretical data; he obtained the modal size and the width parameter of the distribution function. The uniqueness of the solution has not posed a problem. The experimental data always fitted one set of theoretical data with the least deviations.

His results show that for very narrow distribution ($\sigma_0 \leq 0.155$) theoretical computations can be applied to experimental data and the important parameters; namely, refractive index n and size-distribution parameters will be obtained.

For atmospheric aerosols the situation is not so good, because we have a mixture of refractive indices and very broad size distribution.

For an exponential distribution (Junge's aerosol distribution) we have computed data based on our theoretical results and find the following values for K_p (see table II) for the interval from $\alpha = 0.1$ to $\alpha = 50$ using $\gamma = -3$.

TABLE II
FOR EXPONENTIAL DISTRIBUTION
WITH $K_p = -3$

n	1.33	1.40	1.44	1.486	1.5	2.0
K_p	1.26	1.48	1.61	1.75	1.79	3.13

The value for $n = 2.0$ is not so accurate as those for the interval $n = 1.33$ to 1.5.

The effect of different values of γ , i. e., different steepness of the size distribution has been computed for the interval $\alpha = 0.1$ to $\alpha = 30$ and $n = 1.33$. We find for $\gamma = -2$, $K_p = 1.93$; $\gamma = -3$, $K_p = 1.17$, and for $\gamma = -4$, $K_p = 0.38$.

Such a result is expected because the steeper the function the more aerosol particles are contained in the very small α range.

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1. ORIGINATING ACTIVITY <i>(Corporate author)</i> Research and Development Division Avco Corporation, Wilmington, Massachusetts		2a. REPORT SECURITY CLASSIFICATION Unclassified
		2b. GROUP
3. REPORT TITLE Mie Scattering Coefficients for Selected Aerosol Size Distributions		
4. DESCRIPTIVE NOTES <i>(Type of report and inclusive dates)</i> Final Report Scientific Period covered: 1 Apr. 1964 - 31 Oct. 1965		
5. AUTHOR(S) <i>(Last name, first name, initial)</i> Penndorf, Rudolf Approved 15 Dec. 1965		
6. REPORT DATE 31 October 1965	7a. TOTAL NO. OF PAGES 23	7b. NO. OF REFS 11
8a. CONTRACT OR GRANT NO. AF19(628)-4072	9a. ORIGINATOR'S REPORT NUMBER(S) RAD-TR -65-39	
b. PROJECT AND TASK NO. 4479-19		
c. DOD ELEMENT 62405484	9b. OTHER REPORT NO(S) <i>(Any other numbers that may be assigned this report)</i>	
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11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Hq. AFCRL, OAR (CRO) United States Air Force L. G. Hanscom Field, Bedford, Mass.	
13. ABSTRACT <p>Formulas are derived for volume scattering coefficients applicable to aerosols. Different types of size distribution for the aerosol, such as normal distribution, log-normal distribution, and exponential distribution, are assumed.</p> <p>Nominal results obtained by the author and those published in the literature are given and discussed.</p> <p>The volume scattering coefficients become smooth functions. The size distribution is very wide. Hence, simpler mathematical formulation can be used than those obtained from the exact Mie theory. However, only the number of aerosol particles can be obtained from an evaluation of experimental data, but details of the size distribution cannot be extracted from experimental data</p>		

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