Identification of unimodal size distributions of aerosol particles

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Practically all known unimodal aerosol particle size distributions are generalized by two compact dependences. Equations (approximate and analytical whenever possible) relating the parameters of these distributions to the average particle size and geometrical characteristics of the probability density functions are obtained. The modified technique for determining the particle size spectra and results obtained by processing histograms are presented for the case study of the sedimentation analysis.

The disperse aerosol systems are widely spread in both natural and technological environments. In connection with the rapid development of powder technologies, mechanics of multiphase flows, as well as of the laser methods for diagnostics of the parameters of aerosol particles, call for an adequate description of the polydisperse systems. It is an urgent problem that the description would allow the identification of the particle size distributions to be done based on experimental histograms. The possibility of approximating the distributions by a suitable probability density functions is also important in different fields of physics. There exist different ways of solving these problems because of a wide variety of the particle size distributions known from literature (which are traditionally different for different subfields of aerosol mechanics and optics). In addition, the complete set of relations between these characteristics and the distribution parameters is absent.

The fraction of particles of different size in a polydisperse ensemble is fully determined by its differential number density function $f(x)$, where x is the parameter characterizing the size of an individual particle (radius, diameter, volume, mass, cross section, some equivalent size for nonspherical particles, etc.). In this case, $\mathrm{d}f = f(x)\,\mathrm{d}x$ is the fraction of particles, whose size ranges within the $(x, x + \mathrm{d}x)$ interval.[1] The function $f(x)$ has the meaning of the probability density distribution, that is, it is normalized to unity

$$\int_0^\infty f(x)\mathrm{d}x = 1. \qquad (1)$$

The analysis of voluminous literature data on the particle size distributions of different polydisperse systems has shown that almost all natural and artificial aerosols with the unimodal distribution function can be described using either the generalized gamma-distribution (GGD) or the lognormal distribution (LND).[1–6] The equation for the GGD proposed by Shifrin[2] in 1951 can be written in the form

$$f(x) = ax^{\alpha}\exp\left(-bx^{\beta}\right), \qquad (2)$$

where $a > 0$ is the normalization factor; α, β, and b are the parameters of distribution ($\alpha > -1$, $b > 0$, $\mathrm{sgn}(\alpha) = \mathrm{sgn}(\beta)$).

The normalizing factor GGD as determined from the condition (1) has the form

$$a = \beta b^{\frac{\alpha+1}{\beta}}\,\Gamma^{-1}\left(\frac{\alpha+1}{\beta}\right),$$

where Γ is the gamma-function.

If $\beta = 1$ GGD transforms into the widely used gamma-distribution (GD)

$$f(x) = ax^{\alpha}\exp(-bx), \qquad (3)$$

whose normalizing factor is $a = b^{\alpha+1}\,\Gamma^{-1}(\alpha + 1)$. Note that at integer α, $\Gamma(\alpha + 1) = \alpha!$.

By varying the parameters α, β, and b, one can obtain, from the generalized gamma-distribution (2), most of the distributions used in the literature. Table 1 summarizes the most widely used distributions.

In the majority of cases, these equations have no theoretical sense. They are more or less good empirical approximations of the actual distributions. Nevertheless, their practical significance is obvious, since they allow one to describe the particle size distribution of a polydisperse ensemble with a limited set (no more than three) of parameters. Multiparameter equations have gained no practical utility because of laborious fit in case of approximating the histograms.

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Table 1. Empirical distribution laws of polydisperse particles

α	b	β	$f(x)$	Distribution	Applications
2	b	3	$ax^2 \exp(-bx^3)$	Smolukhovsky–Schuman	Atmospheric physics (clouds, precipitation), colloid systems
0	b	1	$a \exp(-bx)$	Martin(Marshall–Palmer)	Atmospheric physics (clouds, precipitation), grinding products
2	b	2	$ax^2 \exp(-bx^2)$	Maxwell–Boltzmann	Soot dispersion in flames
1	b	2	$ax \exp(-bx^2)$	Romashov	Industrial dusts
–4	0	–	ax^{-4}	Junge	Atmospheric physics (haze, surface aerosol)
2	b	β	$ax^2 \exp(-bx^\beta)$	Nukiyama–Tanasawa	Fluid spraying
β–4	b	1	$ax^{\beta-4} \exp(-bx)$	Rosin–Rammler	Fluid spraying, grinding products
α	b	2	$ax^\alpha \exp(-bx^2)$	Weining	Grinding products

The lognormal distribution belongs to the class of Captain distributions[3] derived based on the Gauss distribution. In this case, the particles are distributed over the size according to the lognormal law, and the equation for the distribution function has the form[1]:

$$f(x) = \frac{1}{\sqrt{2\pi}x\,\mathrm{Ln}(\sigma_p)} \exp\left(-\frac{\left(\mathrm{Ln}(x) - \mathrm{Ln}(x_p)\right)^2}{2\mathrm{Ln}^2(\sigma_p)}\right), \quad (4)$$

where $\mathrm{Ln}(x_p)$ is the mathematical expectation of the log particle size (x_p is the mean geometric size); $\mathrm{Ln}^2(\sigma_p)$ is the root-mean-square deviation of the log size (in the non-Russian literature, σ_p is usually called the standard geometric deviation). In contrast to GGD obtained empirically, LND has certain physical grounds. In 1941 Kolmogorov has shown theoretically that LND is the limiting case of a rather general scheme of a random grinding process.[4] It is natural to assume that LND can also be a result of the random coagulation process of drops in a two-phase flow in the limiting case of the large number of interactions between particles. The lognormal law is widespread in nature. It governs the distributions of particles suspended in air and water, ground rock particles, the distributions formed due to chemical sedimentation and screen analysis, the size distribution of grains of the placer gold, etc.[5] It is interesting to note that the LND function provides, for example, quite a satisfactory description of the weight growth in schoolchildren after vacation.[3]

The graphical analysis performed in Ref. 6 has shown a good mutual approximation of LND and GD (Fig. 1). The larger the parameter α of the gamma-distribution, the better the approximation. The higher values of the parameter α correspond to smaller variance of the log particle size.

It is convenient to write the equation for LND (4) in a compact form:

$$f(x) = \frac{a}{x} \exp\left[-b\,\mathrm{Ln}^2(\beta x)\right], \quad (5)$$

where the parameters a, β, and b are related to x_p, σ_p as

$$a = \frac{1}{\sqrt{2\pi}\,\mathrm{Ln}(\sigma_p)}; \quad b = \frac{1}{2\mathrm{Ln}^2(\sigma_p)}; \quad \beta = \frac{1}{x_p}.$$

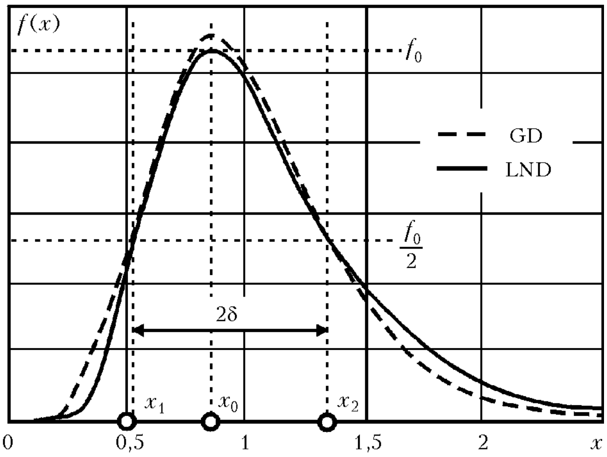

Fig. 1. Geometric characteristics of the distribution $f(x)$.

The parameters a, β, and b of the distributions (2), (3), and (5) cannot be illustrated graphically. In the practice of the disperse system analysis, it is convenient to evaluate the shape of the distribution using the geometric characteristics of the probability density function, such as the modal size x_0 (the point of maximum of the probability density function), $f_0 = f(x_0) = f_{\max}(x)$, the distribution half-width δ and asymmetry ε (see Fig. 1):

$$\delta = \frac{x_2 - x_1}{2}; \quad \varepsilon = \frac{x_0 - x_1}{x_2 - x_1} = \frac{x_0 - x_1}{2\delta}, \quad (6)$$

where

$$f(x_1) = f(x_2) = f_0/2.$$

For the gamma-distribution, the relationships between its parameters and the geometric characteristics of $f(x)$ (except for asymmetry) can be found in Ref. 2. In this paper, these equations are obtained for GGD, GD, and LND (Table 2). Along with the distribution function, averaged characteristics are widely used for description of the disperse media. Most of these characteristics are determined as follows:

$$x_{mn} = \left[\int_0^\infty x^m f(x)\,\mathrm{d}x \bigg/ \int_0^\infty x^n f(x)\,\mathrm{d}x\right]^{\frac{1}{m-n}}, \quad (7)$$

where m and n are integer numbers denoting the order of the moment of the distribution function.

Table 2. Geometric characteristics of the probability density and some mean particle sizes as expressed through the parameters of the distributions

Parameters	GGD	GD	LND
x_0	$\left(\frac{\alpha}{b\beta}\right)^{1/\beta}$	$\frac{\alpha}{b}$	$\frac{1}{\beta}\exp\left(-\frac{1}{2b}\right)$
f_0	$a\left(\frac{\alpha}{b\beta}\right)^{\alpha/\beta}\exp\left(-\frac{\alpha}{\beta}\right)$	$a\left(\frac{\alpha}{b}\right)^{\alpha}\exp(-\alpha)$	$a\beta\exp\left(\frac{1}{4b}\right)$
2δ	$\frac{2.34}{\sqrt{\alpha\beta}}x_0$	$\frac{2.34}{\sqrt{\alpha}}x_0$	$2\mathrm{sh}\left(\sqrt{\frac{\mathrm{Ln}\,2}{b}}\right)x_0$
ε	$0.304\alpha^{(3/b-1)/16}\beta^{0.358}$	$0.304\alpha^{0.125}$	$\left[1+\exp\left(\sqrt{\frac{\mathrm{Ln}\,2}{b}}\right)\right]^{-1}$
x_{10}	$\Gamma\left(\frac{2+\alpha}{\beta}\right)\left[b^{1/\beta}\Gamma\left(\frac{1+\alpha}{\beta}\right)\right]^{-1}$	$\frac{1+\alpha}{b}$	$\frac{1}{\beta}\exp\left(\frac{1}{4b}\right)$
x_{32}	$\Gamma\left(\frac{4+\alpha}{\beta}\right)\left[b^{1/\beta}\Gamma\left(\frac{3+\alpha}{\beta}\right)\right]^{-1}$	$\frac{3+\alpha}{b}$	$\frac{1}{\beta}\exp\left(\frac{5}{4b}\right)$
x_{43}	$\Gamma\left(\frac{5+\alpha}{\beta}\right)\left[b^{1/\beta}\Gamma\left(\frac{4+\alpha}{\beta}\right)\right]^{-1}$	$\frac{4+\alpha}{b}$	$\frac{1}{\beta}\exp\left(\frac{7}{4b}\right)$

The most widely used characteristics are:

– x_{10} – arithmetical mean size;

– x_{20} – root-mean-square size;

– x_{32} – mean volume-surface size;

– x_{43} – mass-average size (mathematical expectation x for the differential function of the mass particle size distribution $g(x)$).

By substituting Eqs. (2), (3), and (5) into Eq. (7) one can obtain the equation for calculating x_{mn} for GGD, GD, and LND, respectively:

$$\text{GGD:}\quad x_{mn} = b^{\frac{n-m}{\beta}}\,\Gamma\left(\frac{m+1+\alpha}{\beta}\right)\Gamma^{-1}\left(\frac{n+1+\alpha}{\beta}\right);$$

$$\text{GD:}\quad x_{mn} = b^{n-m}\,\frac{m+\alpha}{n+\alpha}\,\frac{\Gamma(m+\alpha)}{\Gamma(n+\alpha)};$$

$$\text{LND:}\quad x_{mn} = \left[\beta^{n-m}\exp\left(\frac{m^2-n^2}{4b}\right)\right]^{\frac{1}{m-n}}.$$

The particular dependences for x_{10}, x_{32}, x_{43} on the parameters of the distributions are given in Table 2.

Table 3 presents the equations for estimation of the GD and LND parameters from the geometric characteristics of the distribution function $f(x)$.

The practical application of this approach can be illustrated by reconstruction of the distribution function from the data obtained from the gravitational-sedimentation analysis of ferrosilicon particles. First, it should be noted that since the series of experimental values is usually limited, it is necessary to use some mean particle sizes of the obtained mass fractions for refinement of the derived dependences. In particular, for gravitational sedimentation the equation for the effective size of fractions can be derived from the following concepts.

Table 3. The parameters of the distributions expressed through the geometric characteristics of the probability density function

Parameter	GD	LND
α	$\left(\frac{1.17}{\delta}x_0\right)^2$	–
b	$x_0\left(\frac{1.17}{\delta}\right)^2$	$\mathrm{Ln}\,2\left[\mathrm{Ar\,sh}\left(\frac{\delta}{x_0}\right)\right]^{-2}$
β	–	$\frac{1}{x_0}\exp\left\{-\frac{\left[\mathrm{Ar\,sh}\left(\frac{\delta}{x_0}\right)\right]^2}{2\,\mathrm{Ln}\,2}\right\}$

Assume that N fractions of particles with the size $(D_j - D_{j+1})$ are deposited in the Stokes mode, that is, $v_i = \varphi D_i^2$, where v is the particle velocity, D is the particle diameter, and φ is the constant dependent on the medium characteristics and the density of the particulate matter. Then, from the law of conservation of mass, we have

$$c_m\bar{D}^2 = \sum_{i=1}^{N} c_{mi}D_i^2, \qquad (8)$$

where c_m, $\bar{D}$ are the mass concentration and the mean effective diameter of particles in the considered range of the particle size; c_{mi} is the mass concentration of particles of the ith fraction.

From Eq. (8) it follows that

$$\bar{D} = \sqrt{\frac{\sum_{i=1}^{N} c_{mi} D_i^2}{c_m}} = \sqrt{\sum_{i=1}^{N} z_i D_i^2}.$$

Assuming that the particle size distribution is uniform in the range (D_j, D_{j+1}), that is, $z_i = \text{const}$ and taking account that $\sum_{i=1}^{N} z_i = 1$, we obtain

$$\bar{D} = \sqrt{\frac{\sum_{i=1}^{N} D_i^2 \Delta D}{D_{j+1} - D_j}}. \tag{9}$$

Passing on from summation to integration in Eq. (9), we obtain

$$\bar{D} = \sqrt{\frac{\int_{D_j}^{D_{j+1}} D^2 dD}{D_{j+1} - D_j}} = \sqrt{\frac{\left(D_{j+1}^3 - D_j^3\right)}{3\left(D_{j+1} - D_j\right)}}. \tag{10}$$

The comparison of Eqs. (7) and (10) shows that, in the case of gravitational sedimentation, the root-mean-square size D_{20} should be used as the characteristic point on the histogram columns.

Figure 2 shows the initial data processed by Eq. (10) (closed circles) and the corresponding histogram of mass fraction of ferrosilicon particles, along with the reconstructed particle size distribution functions in the GD (curve *1*) and GGD (curve *2*) classes after scaling the normalized dependences to the values of the initial parameters. For obtaining these dependences, from the graphical approximation the following parameters were determined: $x_0 \approx 35$, $x_1 \approx 17.5$, $x_2 \approx 62$ μm. Equations (6) were used to calculate the distribution half-width δ and asymmetry ε, which, in their turn, have allowed the estimation of α, b, and β for GD and GGD to be done by the equations from Table 2. Then the normalizing factor, a, was determined from the parameters of the distributions by use of the above dependences.

Thus, the data shown in Fig. 2 have yielded the following parameters of the distribution laws:

for GD: $\alpha = 3.387$, $b = 0.097$;

for GGD: $\alpha = 1.739$, $\beta = 1.947$, $b = 0.001$.

Thus, the approach presented allows one to determine adequate particle size distribution functions in approximating the experimental data on the particle size distributions. It also enables one to obtain estimates of the parameters of distributions within the class of distribution functions chosen, as well as to evaluate the average parameters of the polydisperse system under study.

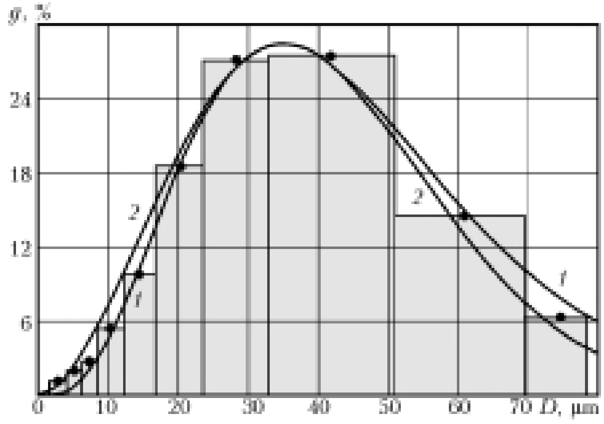

Fig. 2. Initial data and reconstructed particle size distribution functions.

Acknowledgments

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