Some Useful Formulae for Particle Size Distributions and Optical Properties

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Contents

1 Describing Particle Size **1**
1.1 Particle Size Distribution . . . 1
1.2 Metrics of Distribution Centre & Spread . . . 2
1.3 Effective Radius and Effective Variance . . . 3
1.4 Moments . . . 3
1.5 Area, Volume and Mass Distributions . . . 4
1.6 Non-spherical Particles . . . 5

2 Normal and Logarithmic Normal Distributions **6**
2.1 Definitions . . . 6
2.2 Properties of the Log-normal Distribution . . . 10
2.2.1 Median and Mode . . . 10
2.2.2 Limits . . . 11
2.2.3 Moments . . . 12
2.2.4 Derived Metrics . . . 12
2.2.5 Derivatives . . . 13
2.3 Log-normal Distributions of Area and Volume . . . 13

3 The Modified Gamma and Gamma Distributions **19**
3.1 Modified Gamma Distribution . . . 19
3.2 Gamma Distribution . . . 21

4 Other Distributions **24**
4.1 Inverse Modified Gamma Distribution . . . 24
4.2 Regularized Power Law . . . 25
4.2.1 Moments . . . 25

5 Modelling the Evolution of an Aerosol Size Distribution **27**

6 Optical Properties **28**
6.1 Volume Absorption, Scattering and Extinction Coefficients . . . 28
6.2 Back Scatter . . . 29
6.3 Phase Function . . . 29
6.4 Single Scatter Albedo . . . 29
6.5 Asymmetry Parameter . . . 29
6.6 Integration . . . 29
6.7 Formulae for Practical Use . . . 30
6.7.1 Extinction Limits for a Log-Normal Distribution . . . 32
6.8 Cloud Liquid Water Path . . . 34
6.9 Aerosol Mass . . . 35

List of Symbols **38**

References **39**

Index **40**

Note that this is a working draft. Comments that will be excluded from the final text are indicated by ▬ XXX ▬ .

1 Describing Particle Size

Atmospheric particles come in a variety of compositions and sizes. There are three main classes: aerosols, water droplets and ice crystals. But that is just a start, a complete description of an ensemble of particles would encompass the composition and geometry of each particle. Such an approach is impracticable. For example atmospheric aerosols have concentrations as high as $\sim 10,000$ particles per cm^3. An alternate approach is to use a statistical description of the particle size distribution. This is assisted by the fact that small liquid drops adopt a spherical shape so that for a chemically homogeneous particle the problem becomes one of representing the number distribution of particle radii. The particle size distribution can be represented in tabular form but it is usual to adopt an analytic functional. The success of this approach hinges upon the selection of an appropriate size distribution function that approximates the actual distribution. There is no *a priori* reason for assuming this can be done.

1.1 Particle Size Distribution

A measured distribution of particle sizes can be described by a histogram of the number of particles per unit volume within defined size bins. By making the bin size tend to zero a continuous function is formed called the radius number density distribution $N(r)$ which represents the number of particles with radii between r and $r + dr$ per unit volume. The difficulty with this representation is that it is unmeasurable! This is because there are an infinite number of radius values so that the probability of the radius being any one specific value is zero. The problem is avoided by using the differential radius number density distribution $n(r)$ defined by

$$n(r) \;=\; \frac{dN(r)}{dr}. \tag{1}$$

The same equation can be written in integral form as

$$dN(r) \;=\; \int_r^{r+dr} n(r)\, dr. \tag{2}$$

The total number of particles per unit volume, N_0, is given by

$$N_0 \;=\; \int_0^{\infty} n(r)\, dr. \tag{3}$$

The total surface area of the distribution or surface area density is defined

$$A_0 \;=\; 4\pi \int_0^{\infty} r^2 n(r)\, dr. \tag{4}$$

and similarly the volume density is found from

$$V_0 = \frac{4}{3}\pi \int_0^\infty r^3 n(r)\, dr. \tag{5}$$

It follows that the mass density is

$$M_0 = \rho V_0 \tag{6}$$

where ρ is the density of the substance forming the particles.

1.2 Metrics of Distribution Centre & Spread

Generally a size distribution is characterised by the total number of particles per unit volume, its centre and by its spread. The centre of a distribution can be represented by

the mean μ_0 is the arithmetic average defined by

$$\mu_0 = \frac{\int_0^\infty r n(r)\, dr}{N_0}. \tag{7}$$

the geometric mean μ_g is defined by

$$\mu_g = \exp\left(\frac{\int_0^\infty \ln r\ n(r)\, dr}{N_0}\right) \tag{8}$$

the mode is the location of the peak (maximum) value of a size distribution.

the median is the "middle" value of a data set, i.e. 50 % of particles are smaller than the median (and so 50 % of particles are larger).

For symmetric distributions the mean, mode and median have the same value. Most aerosol distributions are asymmetric having more larger particles than smaller so that

$$\text{mode} < \text{median} < \text{mean} \tag{9}$$

The spread of the size distribution is captured through

the variance σ_0^2 defined

$$\sigma_0^2 = \frac{\int_0^\infty (r-\mu_0)^2 n(r)\, dr}{N_0}. \tag{10}$$

The standard deviation, σ_0, of a distribution is the square root of the variance.

the geometric standard deviation σ_g defined

$$\sigma_g = \exp\left[\sqrt{\frac{\int_0^\infty (\ln r - \ln \mu_g)^2\, n(r)\, dr}{N_0}}\right]. \tag{11}$$

1.3 Effective Radius and Effective Variance

The mean along with other size distribution metrics can be helpful in describing a particle distribution, but by far the most useful for optical measurements is the effective radius, r_e. The advantage of using the effective radius comes from the fact that energy removed from a light beam by a particle is proportional to the particle's area (provided the radius of the particle is similar to, or larger than, the wavelength of the incident light). Weighting each radius by $\pi r^2 n(r)$ gives

$$r_\mathrm{e} = \frac{\int_0^\infty r\pi r^2\, n(r) dr}{\int_0^\infty \pi r^2\, n(r) dr}. \tag{12}$$

The effective radius is sometimes called the area-weighted mean radius. In an equivalent manner the effective variance for a distribution can be defined (Hansen 1971)

$$v_\mathrm{e} = \frac{\int_0^\infty (r - r_\mathrm{e})^2 \pi r^2 n(r)\, dr}{r_\mathrm{e}^2 \int_0^\infty \pi r^2 n(r)\, dr} \tag{13}$$

The factor r_e^2 is included in the denominator to make v_e dimensionless and a relative measure.

1.4 Moments

The i^th moment m_i of a distribution $n(r)$ is defined

$$m_i = \int_0^\infty (r - c)^i n(r)\, dr \tag{14}$$

where c is some constant. Choosing $c = 0$ gives the raw moment. Choosing c to be the distribution mean generates central moments. Moments depend upon the shape of the $n(r)$ and can be used to calculate some common metrics listed in Table 1. Note that the geometric mean (or variance) is not necessarily expressible in moments but Vogel (2020) provides expressions for most of the common distributions.

Table 1: Size distribution metrics expressed in terms of the raw moments.

Metric	**Formula**
number density	$N_0 = m_0$
surface area density	$a_{\mathrm{V}} = 4\pi m_2$
volume density	$v_{\mathrm{V}} = \frac{4}{3}\pi m_3$
mean radius	$\mu_0 = \frac{m_1}{m_0}$
variance	$\sigma_0^2 = \frac{m_2}{m_0} - \frac{m_1^2}{m_0^2}$
effective radius	$r_{\mathrm{e}} = \frac{m_3}{m_2}$
effective variance	$v_{\mathrm{e}} = \frac{m_2 m_4}{m_3^2} - 1$

1.5 Area, Volume and Mass Distributions

Analogous to the description of the distribution of particle number with radius it is also possible to describe particle area, volume or mass with equivalent expressions to Equations 2 and 3. This gives rise to multiple ways of expressing a particle distribution and it is often necessary to swap from one representation to another.

The area density distribution $A(r)$ represents the area of particles with radii between r and $r + dr$ per unit volume, i.e.

$$A(r) \;=\; \int_r^{r+dr} a(r)\, dr. \tag{15}$$

The differential area density distribution, $a(r)$ is

$$a(r) \;=\; \frac{dA}{dr}. \tag{16}$$

For spherical particles

$$a(r) \;=\; \frac{dA}{dN}\frac{dN}{dr} = 4\pi r^2 n(r). \tag{17}$$

The total particle area per unit volume, A_0, is then given by

$$A_0 \;=\; \int_0^{\infty} a(r)\, dr. \tag{18}$$

The volume density distribution $V(r)$ represents the volume of particles with radii between r and $r + dr$ per unit volume, i.e.

$$V(r) \;=\; \int_r^{r+dr} v(r)\, dr. \tag{19}$$

The differential volume density distribution, $v(r)$ represents the volume contained in particles whose radii lie between r and $r + dr$ per unit volume, i.e.

$$v(r) = \frac{dV}{dr}. \tag{20}$$

For spherical particles

$$v(r) = \frac{dV}{dN}\frac{dN}{dr} = \frac{4}{3}\pi r^3 n(r). \tag{21}$$

The total particle volume per unit volume, V_0, is given by

$$V_0 = \int_0^\infty v(r)\, dr. \tag{22}$$

The mass density distribution $M(r)$ represents the mass of particles with radii between r and $r + dr$ per unit volume, i.e.

$$M(r) = \int_r^{r+dr} m(r)\, dr. \tag{23}$$

The differential mass density distribution, $m(r)$ represents the mass contained in particles with radii between r and $r + dr$ per unit volume, i.e.

$$m(r) = \frac{dM}{dr}. \tag{24}$$

For spherical particles

$$m(r) = \frac{dM}{dN}\frac{dN}{dr} = \frac{4}{3}\pi r^3 \rho n(r), \tag{25}$$

where ρ is the density of the aerosol material. The total particle mass per unit volume, M_0, is

$$M_0 = \int_0^\infty m(r)\, dr. \tag{26}$$

1.6 Non-spherical Particles

Liquid particles are nearly always spherical, the principal deviant being raindrops where aerodynamic forces create a distortion. Solid particles are nearly always non-spherical having been formed by mechanical processes, evaporation, combustion or agglomeration. In these cases the particle size is described by a volume equivalent radius and some form of non-sphericity factor. For example the sphericity Ψ is defined as

$$\Psi = \frac{4\pi A}{p^2} \tag{27}$$

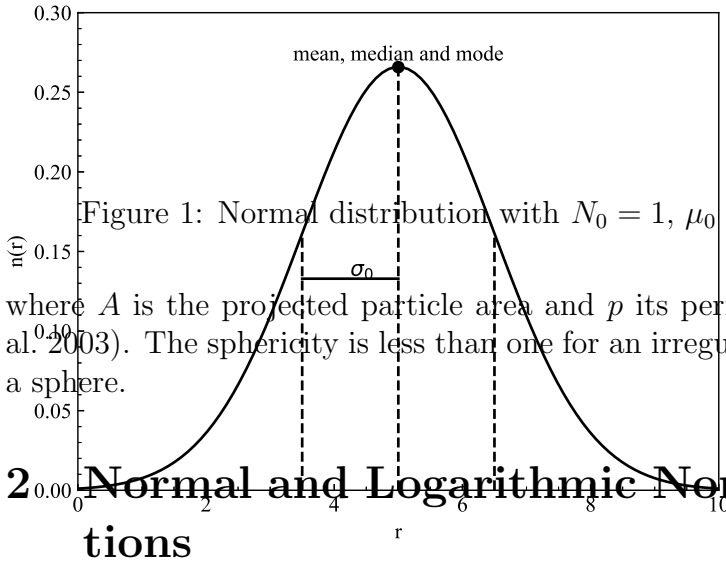


Figure 1: Normal distribution with $N_0 = 1$, $\mu_0 = 5$ and $\sigma_0 = 1.5$.

where A is the projected particle area and p its perimeter length (Riley et al. 2003). The sphericity is less than one for an irregular particle and one for a sphere.

2 Normal and Logarithmic Normal Distributions

2.1 Definitions

One particle distribution to consider adopting is the normal distribution

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_0} \exp\left[-\frac{(r-\mu_0)^2}{2\sigma_0^2}\right], \tag{28}$$

where μ_0 is the mean and σ_0 is the standard deviation of the distribution. An example is shown in Figure 1. The distribution is symmetric about the mean so the mean, median and mode of a normal distribution all have the same value.

Aside 2.1

Show that μ_0 and σ_0^2 are the mean and variance of the normal distribution. The following derivations all use the same substitution, i.e.

$$x = \frac{r-\mu_0}{\sqrt{2}\sigma_0}$$

Firstly

$$\begin{aligned}\int_{-\infty}^{\infty} n_r(r)\,dr &= \int_{-\infty}^{\infty} \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_0}\exp\left[-\frac{(r-\mu_0)^2}{2\sigma_0^2}\right]\,dr \\ &= \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_0}\int_{-\infty}^{\infty}\exp\left(-x^2\right)\sqrt{2}\sigma_0\,dx \\ &= \frac{N_0}{\sqrt{\pi}}\underbrace{\int_{-\infty}^{\infty}\exp\left(-x^2\right)\,dx}_{=\sqrt{\pi}} = N_0\end{aligned}$$

The mean is given by

$$\begin{aligned}\frac{\int_{-\infty}^{\infty} r n_r(r)\,dr}{\int_{-\infty}^{\infty} n_r(r)\,dr} &= \frac{1}{N_0}\int_{-\infty}^{\infty} r\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_0}\exp\left[-\frac{(r-\mu_0)^2}{2\sigma_0^2}\right]\,dr \\ &= \frac{1}{\sqrt{2\pi}}\frac{1}{\sigma_0}\int_{-\infty}^{\infty}\exp\left(-x^2\right)(\sqrt{2}\sigma_0 x+\mu_0)\sqrt{2}\sigma_0\,dx \\ &= \frac{\mu_0}{\sqrt{\pi}}\underbrace{\int_{-\infty}^{\infty}\exp\left(-x^2\right)\,dx}_{\sqrt{\pi}} + \frac{\sqrt{2}\sigma_0}{\sqrt{\pi}}\underbrace{\int_{-\infty}^{\infty} x\exp\left(-x^2\right)\,dx}_{=0} = \mu_0\end{aligned}$$

The variance is given by

$$\begin{aligned}\frac{\int_{-\infty}^{\infty}(r-\mu_0)^2 n_r(r)\,dr}{N_0} &= \frac{1}{N_0}\int_{-\infty}^{\infty}(r-\mu_0)^2\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_0}\exp\left[-\frac{(r-\mu_0)^2}{2\sigma_0^2}\right]\,dr \\ &= \frac{1}{\sqrt{2\pi}\sigma_0}\int_{-\infty}^{\infty} 2\sigma_0^2 x^2\exp\left(-x^2\right)\sqrt{2}\sigma_0\,dx \\ &= \frac{2}{\sqrt{\pi}}\sigma_0^2\underbrace{\int_{-\infty}^{\infty} x^2\exp\left(-x^2\right)\,dx}_{=\sqrt{\pi}/2} = \sigma_0^2\end{aligned}$$

The range of particle sizes generally covers several orders of magnitude. As a result fitting an ensemble of measured particle size with the normal distribution is often poor, typically indicated by a very large standard deviation.

A further drawback of the normal distribution is that it allows negative radii. Particle distributions are much better represented by a normal distribution of the logarithm of the particle radii. Letting $l = \ln(r)$ we have

$$n_l(l) = \frac{dN(l)}{dl} = \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\exp\left[-\frac{(l-\mu_l)^2}{2\sigma_l^2}\right] \tag{29}$$

where $N(l)$ represents the number of particles whose log radii lie between l and $l + dl$ per unit volume while μ_l and σ_l^2 are the mean, and variance of l respectively. As l is normally distributed the mean, median and mode in log space all have the same value, μ_l.

To express the log-normal distribution given in Equation 29 in terms of the linear radius interval dr note that

$$\frac{dl}{dr} = \frac{1}{r} \tag{30}$$

then

$$n(r) = \frac{dN(r)}{dr} = \frac{dN(l)}{dl}\frac{dl}{dr} = n_l(l)\frac{dl}{dr} = \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[-\frac{(\ln r - \mu_l)^2}{2\sigma_l^2}\right] \tag{31}$$

Figure 2 shows normal and log-normal distributions with the same mean and variance. When compared with a normal distribution the log-normal distribution has a stepper increase in number density approaching the mode from zero and a longer tail for radii greater than the mode. Thus the mode of the log-normal distribution is smaller than the mode of the normal distribution.

What complicates the representation of a log-normal distribution is that it is rarely expressed in terms μ_l and σ_l. Instead parameters related to radius are used. These two changes are considered in turn.

1. As the logarithm is a monotonic function, the median of the log radius distribution (μ_l) is the log of the median of the radius distribution r_m so

   $$\mu_l = \ln r_\mathrm{m} \tag{32}$$

   Hence the centre of the distribution (median, mean or mode) in log space is the natural logarithm of the median radius, r_m, in linear space and Equation 31 can be expressed

   $$n(r) = \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[-\frac{(\ln r - \ln r_m)^2}{2\sigma_l^2}\right] \tag{33}$$

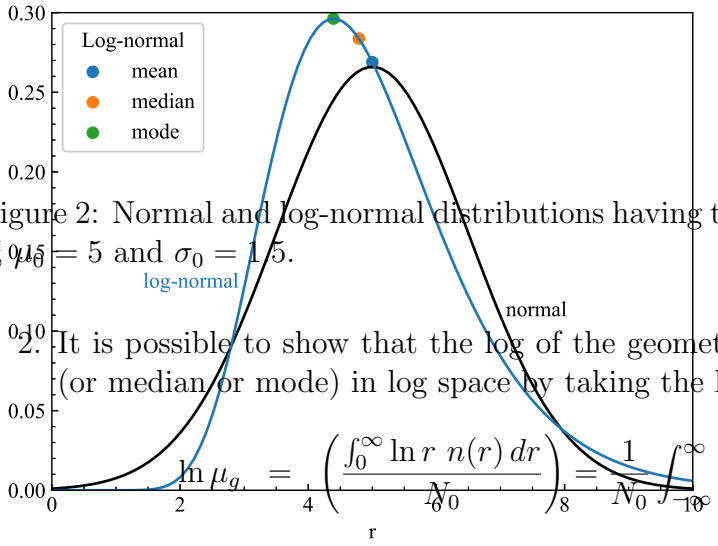


Figure 2: Normal and log-normal distributions having the same metrics $N_0 = 1.0$, $\mu_0 = 5$ and $\sigma_0 = 1.5$.

2. It is possible to show that the log of the geometric mean is the mean (or median or mode) in log space by taking the log of Equation 8 e.g.

$$\ln \mu_g = \left(\frac{\int_0^\infty \ln r \; n(r)\, dr}{N_0} \right) = \frac{1}{N_0} \int_{-\infty}^{\infty} ln_l(l)\, dr = \mu_l \tag{34}$$

Combining with Equation 32 gives

$$\mu_g = r_{\mathrm{m}} \tag{35}$$

that is, the geometric mean is the same as the median for a log-normal distribution. The geometric standard deviation, S, for a log-normal distribution is calculated by placing this expression into Equation 11 and combining with Equation 33

$$\begin{aligned} \ln^2 S &= \frac{\int_0^\infty (\ln r - \ln r_m)^2 \, \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_m)^2}{2\sigma^2} \right] dr}{N_0} \\ &= \frac{1}{\sqrt{2\pi}} \frac{1}{\sigma_l} \int_0^\infty \frac{(\ln r - \ln r_m)^2}{r} \exp\left[-\frac{(\ln r - \ln r_m)^2}{2\sigma_l^2} \right] dr \end{aligned} \tag{36}$$

Making the substitution $x = \frac{(\ln r - \ln r_m)}{\sqrt{2}\sigma_l}$ leads to

$$\begin{aligned} \ln^2 S &= \frac{1}{\sqrt{2\pi}} \frac{1}{\sigma_l} \int_{-\infty}^{\infty} 2\sigma_l^2 x^2 \exp\left(-x^2\right) \sqrt{2}\sigma_l \, dx \\ &= \frac{2}{\sqrt{\pi}} \sigma_l^2 \underbrace{\int_{-\infty}^{\infty} x^2 \exp\left(-x^2\right) dx}_{\sqrt{\pi}/2} = \sigma_l^2 \\ \Rightarrow \ln S &= \sigma_l \end{aligned} \tag{37}$$

From this definition S must be greater or equal to one otherwise the log-normal standard deviation is negative. When S is one the distribution is monodisperse. Typical aerosol distributions have S values in the range 1.5 - 2.0.

The log-normal distribution appears in the atmospheric literature using any of combination of r_{m} or μ and σ ($\equiv \sigma_l$) or S with perhaps the commonest being

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln S} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] \tag{38}$$

Be particularly careful about σ and S whose definitions are sometimes reversed!

2.2 Properties of the Log-normal Distribution

2.2.1 Median and Mode

The mode of the log-normal distribution, r_{M}, is related to the median. Let

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln S} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] = \frac{A}{r} \exp\left[B\right] \tag{39}$$

$$\text{where } A = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln S}, \; B = -\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2 S} \text{ and } \frac{dB}{dr} = -\frac{(\ln r - \ln r_{\mathrm{m}})}{r \ln^2 S}.$$

then

$$\frac{dn(r)}{dr} = -\frac{A}{r^2} \exp\left[B\right] + \frac{A}{r} \exp\left[B\right] \frac{dB}{dr} \tag{40}$$

$$= -\frac{A}{r^2} \exp\left[B\right] - \frac{A}{r^2} \exp\left[B\right] \frac{(\ln r - \ln r_{\mathrm{m}})}{\ln^2 S} \tag{41}$$

Setting the left hand side to zero so $r = r_{\mathrm{M}}$

$$0 = -\frac{A}{r_{\mathrm{M}}^2}\exp[B] - \frac{A}{r_{\mathrm{M}}^2}\exp[B]\frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})}{\ln^2 S} \quad (42)$$

$$-1 = \frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})}{\ln^2 S} \quad (43)$$

$$\ln r_{\mathrm{M}} = \ln r_{\mathrm{m}} - \ln^2 S \quad (44)$$

The peak value of the distribution (at the mode) is then

$$n(r) = \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\exp\left(\frac{\ln^2 S}{2} - \ln r_{\mathrm{m}}\right) \quad (45)$$

2.2.2 Limits

A common problem is identifying minimum and maximum values of $r_{\pm}$ over which to apply the log-normal, usually in some numerical work. If some fraction η of the peak value is chosen then distribution limits can be calculated from

$$\frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{r_{\pm}}\exp\left[-\frac{(\ln r_{\pm} - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] = \eta\frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{r_{\mathrm{M}}}\exp\left[-\frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] \quad (46)$$

Using Equation 44 to eliminate r_{m} gives

$$\ln^2 r_{\pm} - 2\ln r_{\mathrm{M}}\ln r_{\pm} + \ln^2 r_{\mathrm{M}} + 2\ln\eta\ln^2 S = 0 \quad (47)$$

Solving

$$\ln r_{\pm} = \frac{2\ln r_{\mathrm{M}} \pm \sqrt{4\ln^2 r_{\mathrm{M}} - 4(\ln^2 r_{\mathrm{M}} + 2\ln\eta\ln^2 S+)}}{2} \quad (48)$$

$$= \ln r_{\mathrm{M}} \pm \sqrt{-2\ln\eta\ln^2 S} = \ln r_{\mathrm{m}} - \ln^2 S \pm \sqrt{-2\ln\eta\ln^2 S} \quad (49)$$

The limits are

$$r_{\pm} = r_{\mathrm{M}}\exp\left(\pm\sqrt{-2\ln\eta\ln^2 S}\right) = \exp\left(\ln r_{\mathrm{m}} - \ln^2 S \pm \sqrt{-2\ln\eta\ln^2 S}\right) \quad (50)$$

The value in square root is positive as η is always less than one e.g. 10^{-3}.

2.2.3 Moments

The i-th raw moment of a log-normal distribution is given by

$$m_i = N_0 r_{\mathrm{m}}^i \exp\left(\frac{i^2 \ln^2 S}{2}\right) \qquad \left[\equiv N_0 \exp\left(i\mu + \frac{i^2\sigma_l^2}{2}\right)\right]. \tag{51}$$

The first five raw moments are

$$m_0 = N_0 \tag{52}$$

$$m_1 = N_0 r_{\mathrm{m}} \exp\left(\frac{1}{2}\ln^2 S\right) \qquad \left[\equiv N_0 \exp\left(\mu + \frac{1}{2}\sigma_l^2\right)\right] \tag{53}$$

$$m_2 = N_0 r_{\mathrm{m}}^2 \exp\left(2\ln^2 S\right) \qquad \left[\equiv N_0 \exp\left(2\mu + 2\sigma_l^2\right)\right] \tag{54}$$

$$m_3 = N_0 r_{\mathrm{m}}^3 \exp\left(\frac{9}{2}\ln^2 S\right) \qquad \left[\equiv N_0 \exp\left(3\mu + \frac{9}{2}\sigma_l^2\right)\right] \tag{55}$$

$$m_4 = N_0 r_{\mathrm{m}}^4 \exp\left(8\ln^2 S\right) \qquad \left[\equiv N_0 \exp\left(4\mu + \frac{16}{2}\sigma_l^2\right)\right] \tag{56}$$

2.2.4 Derived Metrics

The number density, surface area density, volume density, mean radius, variance, effective radius and effective variance of a log-normal distribution are given by

$$N_0 = m_0 \tag{57}$$

$$A_0 = 4\pi m_2 = 4\pi N_0 r_{\mathrm{m}}^2 \exp\left(2\ln^2 S\right) \qquad \left[\equiv 4\pi N_0 \exp\left(2\mu + 2\sigma^2\right)\right] \tag{58}$$

$$V_0 = \frac{4}{3}\pi m_3 = \frac{4}{3}\pi N_0 r_{\mathrm{m}}^3 \exp\left(\frac{9}{2}\ln^2 S\right) \qquad \left[\equiv \frac{4}{3}\pi N_0 \exp\left(3\mu + \frac{9}{2}\sigma^2\right)\right] \tag{59}$$

$$\mu_0 = \frac{m_1}{m_0} = r_{\mathrm{m}} \exp\left(\frac{1}{2}\ln^2 S\right) \tag{60}$$

$$\sigma_0^2 = \frac{m_2}{m_0} - \frac{m_1^2}{m_0^2} = r_{\mathrm{m}}^2 \exp\left(\ln^2 S\right)\left[\exp\left(\ln^2 S\right) - 1\right] \tag{61}$$

$$r_{\mathrm{e}} = \frac{m_3}{m_2} = r_m \exp\left(\frac{5}{2}\ln^2 S\right) \tag{62}$$

$$v_{\mathrm{e}} = \frac{m_2 m_4}{m_3^2} - 1 = \exp\left(\ln^2 S\right) - 1 \tag{63}$$

The log-normal parameters to form a size distribution with mean μ_0 and variance σ_0^2 are

$$r_m = \frac{\mu_0}{\sqrt{\exp\left(\ln^2 S\right)}} \qquad S = \exp\sqrt{\ln\left(1 + \frac{\sigma_0^2}{\mu_0^2}\right)} \tag{64}$$

2.2.5 Derivatives

The first derivatives of Equation 38 are

$$\frac{dn}{dN_0} = \frac{n}{N_0} \tag{65}$$

$$\frac{dn}{dr} = -\frac{n}{r}\left[1 + \frac{(\ln r - \ln r_\mathrm{m})}{\ln^2 S}\right] \tag{66}$$

$$\frac{dn}{dS} = \frac{n}{S \ln S}\left[\frac{(\ln r - \ln r_\mathrm{m})^2}{\ln^2 S} - 1\right] \tag{67}$$

The second derivatives are:

$$\frac{d^2n}{dN_0^2} = 0 \tag{68}$$

$$\frac{d^2n}{dr^2} = n\left[\frac{2}{r^2} + \frac{3\ln r - 3\ln r_\mathrm{m} - 1}{r^2 \ln^2 S} + \frac{(\ln r - \ln r_\mathrm{m})^2}{r^2 \ln^4 S}\right] \tag{69}$$

$$\frac{d^2n}{dS^2} = n\left[\frac{(\ln r - \ln r_\mathrm{m})^4}{S^2 \ln^6 S} - 5\frac{(\ln r - \ln r_\mathrm{m})^2}{S^2 \ln^4 S} - \frac{(\ln r - \ln r_\mathrm{m})^2}{S^2 \ln^3 S} + \frac{2}{S^2 \ln^2 S} + \frac{1}{S^2 \ln S}\right] \tag{70}$$

2.3 Log-normal Distributions of Area and Volume

Measurements made of particle area or particle volume are often fitted with log-normal distributions in area or volume. The log-normal area density distribution is

$$a(r) = \frac{A_0}{\sqrt{2\pi}} \frac{1}{\sigma_a} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu_a)^2}{2\sigma_a^2}\right] \tag{71}$$

where A_0 is the total aerosol surface area per unit volume, μ_a is the radius of the median area and σ_a the geometric standard deviation.

Aside 2.2

Show that log-normal distribution of area can be expressed in terms of a log-normal distribution of number.
Given the expression for the area of a number distribution

$$a(r) = 4\pi r^2 n(r) = 4\pi r^2 \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu_l)^2}{2\sigma_l^2}\right] \quad (72)$$

make the substitution $r^2 = \exp(2 \ln r)$ and complete the square to get

$$\begin{aligned} a(r) &= 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[-\frac{\ln^2 r - 2 \ln r \mu_l + \mu_l^2 - 4\sigma_l^2 \ln r}{2\sigma_l^2}\right] \\ &= 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[-\frac{\ln^2 r - 2 \ln r(\mu_l + 2\sigma_l^2) + \mu_l^2}{2\sigma_l^2}\right] \\ &= 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \times \\ &\quad \exp\left[-\frac{\ln^2 r - 2 \ln r(\mu_l + 2\sigma_l^2) + (\mu_l + 2\sigma_l^2)^2 - (\mu_l + 2\sigma_l^2)^2 + \mu_l^2}{2\sigma_l^2}\right] \\ &= 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[-\frac{\left(\ln r - (\mu_l + 2\sigma_l^2)\right)^2 - 4\mu_l\sigma_l^2 - 4\sigma_l^4}{2\sigma_l^2}\right] \end{aligned}$$

Giving

$$a(r) = 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[2\mu_l + 2\sigma_l^2\right] \exp\left[-\frac{\left(\ln r - (\mu_l + 2\sigma_l^2)\right)^2}{2\sigma_l^2}\right] \quad (73)$$

Equating Equations 71 and 73 gives

$$\begin{aligned} &\frac{A_0}{\sqrt{2\pi}} \frac{1}{\sigma_a} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu_a)^2}{2\sigma_a^2}\right] \\ &\qquad = 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_l} \frac{1}{r} \exp\left[2\mu_l + 2\sigma_l^2\right] \exp\left[-\frac{\left(\ln r - (\mu_l + 2\sigma_l^2)\right)^2}{2\sigma_l^2}\right] \end{aligned} \quad (74)$$

which is true if

$$\frac{A_0}{\sigma_a} = \frac{4\pi N_0}{\sigma_l} \exp\left[2\mu_l + 2\sigma_l^2\right] \quad (75)$$

and

$$\frac{(\ln r - \mu_a)^2}{2\sigma_a^2} = \frac{\left(\ln r - (\mu_l + 2\sigma_l^2)\right)^2}{2\sigma_l^2} \quad (76)$$

From Equation 58

$$A_0 = 4\pi N_0 \exp\left(2\mu_l + 2\sigma_l^2\right) \tag{77}$$

which gives $\sigma_a = \sigma_l$ when inserted into Equation 75. This shows that if the number density distribution is log-normal then the surface area density distribution is log-normal with the same geometric standard deviation. Applying this result to Equation 76 gives

$$\mu_a = \mu_l + 2\sigma_l^2. \tag{78}$$

This states that the area median radius is greater than the median radius.

Equivalent expressions can be calculated for a volume density distribution, $v(r)$ defined in Section 1.5. The log-normal volume density distribution is

$$v(r) = \frac{V_0}{\sqrt{2\pi}} \frac{1}{\sigma_v} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu_v)^2}{2\sigma_v^2}\right] \tag{79}$$

where V_0 is the total aerosol volume per unit volume, μ_v is the radius of the median volume and σ_v the geometric standard deviation.

Aside 2.3

Show that log-normal distribution of volume can be expressed in terms of a log-normal distribution of number.
Given the expression for the volume of a number distribution

$$v(r) = \frac{4}{3}\pi r^3 n(r) = \frac{4}{3}\pi r^3 \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[-\frac{(\ln r - \mu_l)^2}{2\sigma_l^2}\right] \quad (80)$$

make the substitution $r^3 = \exp(3\ln r)$ and complete the square to get

$$\begin{aligned} v(r) &= \frac{4}{3}\pi\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[-\frac{\ln^2 r - 2\ln r\mu_l + \mu_l^2 - 6\sigma_l^2\ln r}{2\sigma_l^2}\right] \\ &= \frac{4}{3}\pi\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[-\frac{\ln^2 r - 2\ln r(\mu_l + 3\sigma_l^2) + \mu_l^2}{2\sigma_l^2}\right] \\ &= \frac{4}{3}\pi\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\times \\ &\quad \exp\left[-\frac{\ln^2 r - 2\ln r(\mu_l + 3\sigma_l^2) + (\mu_l + 3\sigma_l^2)^2 - (\mu_l + 3\sigma_l^2)^2 + \mu_l^2}{2\sigma_l^2}\right] \\ &= \frac{4}{3}\pi\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[-\frac{\left(\ln r - (\mu_l + 3\sigma_l^2)\right)^2 - 6\mu_l\sigma_l^2 - 9\sigma_l^4}{2\sigma_l^2}\right] \end{aligned}$$

Giving

$$v(r) = \frac{4}{3}\pi\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[3\mu_l + 4.5\sigma_l^2\right]\exp\left[-\frac{\left(\ln r - (\mu_l + 3\sigma_l^2)\right)^2}{2\sigma_l^2}\right] \quad (81)$$

Equating with Equations 79 and 81 gives

$$\begin{aligned} &\frac{V_0}{\sqrt{2\pi}}\frac{1}{\sigma_v}\frac{1}{r}\exp\left[-\frac{(\ln r - \mu_v)^2}{2\sigma_v^2}\right] \\ &\quad = \frac{4}{3}\pi\frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_l}\frac{1}{r}\exp\left[3\mu_l + 4.5\sigma_l^2\right]\exp\left[-\frac{\left(\ln r - (\mu_l + 3\sigma_l^2)\right)^2}{2\sigma_l^2}\right] \end{aligned} \quad (82)$$

which is true if

$$\frac{V_0}{\sigma_v} = \frac{4}{3}\pi\frac{N_0}{\sigma_l}\exp\left[3\mu_l + 4.5\sigma_l^2\right] \quad (83)$$

and

$$\frac{(\ln r - \mu_v)^2}{2\sigma_v^2} = \frac{\left(\ln r - (\mu_l + 3\sigma_l^2)\right)^2}{2\sigma_l^2} \quad (84)$$

Table 2: Relationships area and volume density log-normal parameters and the number size distribution parameters, N_0, μ_l and σ_l.

Distribution	**Parameters**	**Relation to Number Density Parameters**
Area density	A_0	$= 4\pi N_0 \exp\left(2\mu_l + 2\sigma_l^2\right)$
	μ_a	$= \mu_l + 2\sigma_l^2$
	σ_a	$= \sigma_l$
Volume density	V_0	$= \frac{4}{3}\pi N_0 \exp\left(3\mu_l + 4.5\sigma_l^2\right)$
	μ_v	$= \mu_l + 3\sigma_l^2$
	σ_v	$= \sigma_l$

Table 3: Values derived for a log-normal number density size distribution with $S = 1.5$.

Median Radius	**Mean Radius**	**Effective Radius**	**Area Median Radius**	**Volume Median Radius**
1	1.3	3.3	2.6	4.2
2	2.5	6.6	5.2	8.5
5	6.4	17	13.	21
10	13	33	26	42

From Equation 59

$$V_0 = \frac{4}{3}\pi N_0 \exp\left(3\mu_l + 4.5\sigma_l^2\right) \tag{85}$$

which gives $\sigma_v = \sigma_l$ when inserted into Equation 83. This shows that is the number density distribution is log-normal then the volume density distribution is log-normal with the same geometric standard deviation. Applying this result to Equation 84 gives

$$\mu_v = \mu_l + 3\sigma_l^2. \tag{86}$$

They relationships area and volume density log-normal parameters and the number size distribution parameters are summarised in Table 2. They can be used to calculate the distribution centre metric for a log-normal distribution in number density that has been represented by either a log-normal area density distribution or a log-normal volume density distribution. Table

3 shows some derived values which underline how careful one has to be with terminology. For example two scientist could be at odds claiming 'the centre' of the size distribution was 1 or 8.4 μm. The issue could be resolved when it is realised scientist A is using the median radius to represent 'the centre' while scientist B was using volume median diameter.

3 The Modified Gamma and Gamma Distributions

3.1 Modified Gamma Distribution

The modified (or generalized) gamma distribution was introduced to represent particle size distributions in the Earth's atmosphere by Deirmendjian (1963) as

$$n(r) \quad = \quad ar^{\alpha} \exp\left(-br^{\gamma}\right). \tag{87}$$

The four constants a, α, b, γ are positive and real and α is an integer. It is a mathematically convenient model for size distributions of particle types ranging from aerosols and cloud droplets or ice particles to liquid and frozen precipitation (Petty and Huang 2011).

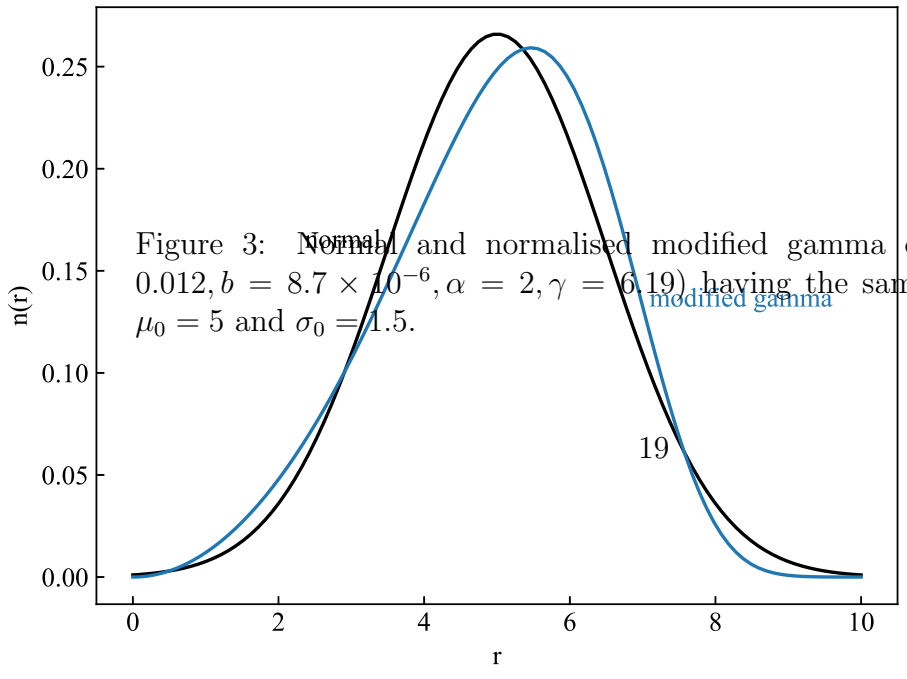


Figure 3: Normal and normalised modified gamma distributions ($a = 0.012, b = 8.7 \times 10^{-6}, \alpha = 2, \gamma = 6.19$) having the same metrics $N_0 = 1$, $\mu_0 = 5$ and $\sigma_0 = 1.5$.

The mode of the distribution occurs where $r = \left(\frac{\alpha}{b\gamma}\right)^{1/\gamma}$. The raw moments of a modified gamma distribution are (see 3·478/1 of Gradshteyn and Ryzhik 1994)

$$m_i = \frac{a}{\gamma} b^{-\frac{\alpha+i+1}{\gamma}} \Gamma\left(\frac{\alpha+i+1}{\gamma}\right). \tag{88}$$

The first five raw moments of the modified gamma distribution are

$$\begin{aligned}
m_0 &= \frac{a}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right) \\
m_1 &= \frac{a}{\gamma} b^{-\frac{\alpha+2}{\gamma}} \Gamma\left(\frac{\alpha+2}{\gamma}\right) \\
m_2 &= \frac{a}{\gamma} b^{-\frac{\alpha+3}{\gamma}} \Gamma\left(\frac{\alpha+3}{\gamma}\right) \\
m_3 &= \frac{a}{\gamma} b^{-\frac{\alpha+4}{\gamma}} \Gamma\left(\frac{\alpha+4}{\gamma}\right) \\
m_4 &= \frac{a}{\gamma} b^{-\frac{\alpha+5}{\gamma}} \Gamma\left(\frac{\alpha+5}{\gamma}\right)
\end{aligned}$$

The moments of the modified gamma distribution can be used to find the derived metrics:

$$N_0 = m_0 = \frac{a}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right) \tag{89}$$

$$A_0 = 4\pi m_2 = 4\pi \frac{a}{\gamma} b^{-\frac{\alpha+3}{\gamma}} \Gamma\left(\frac{\alpha+3}{\gamma}\right) \tag{90}$$

$$V_0 = \frac{4}{3}\pi m_3 = \frac{4}{3}\pi \frac{a}{\gamma} b^{-\frac{\alpha+4}{\gamma}} \Gamma\left(\frac{\alpha+4}{\gamma}\right) \tag{91}$$

$$\mu_0 = \frac{m_1}{m_0} = \frac{b^{-\frac{1}{\gamma}} \Gamma\left(\frac{\alpha+2}{\gamma}\right)}{\Gamma\left(\frac{\alpha+1}{\gamma}\right)} \tag{92}$$

$$\sigma_0^2 = \frac{m_2}{m_0} - \frac{m_1^2}{m_0^2} \tag{93}$$

$$= \frac{\frac{a}{\gamma} b^{-\frac{\alpha+3}{\gamma}} \Gamma\left(\frac{\alpha+3}{\gamma}\right)}{\frac{a}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right)} - \frac{\frac{a}{\gamma} b^{-\frac{\alpha+2}{\gamma}} \Gamma\left(\frac{\alpha+2}{\gamma}\right) \frac{a}{\gamma} b^{-\frac{\alpha+2}{\gamma}} \Gamma\left(\frac{\alpha+2}{\gamma}\right)}{\frac{a}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right) \frac{a}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right)} \tag{94}$$

$$= \frac{b^{-\frac{2}{\gamma}} \Gamma\left(\frac{\alpha+3}{\gamma}\right)}{\Gamma\left(\frac{\alpha+1}{\gamma}\right)} - \frac{b^{-\frac{2}{\gamma}} \Gamma^2\left(\frac{\alpha+2}{\gamma}\right)}{\Gamma^2\left(\frac{\alpha+1}{\gamma}\right)} \tag{95}$$

$$r_{\mathrm{e}} = \frac{m_3}{m_2} = \frac{b^{-1}\gamma\Gamma\left(\frac{\alpha+4}{\gamma}\right)}{\Gamma\left(\frac{\alpha+3}{\gamma}\right)} \tag{96}$$

$$v_{\mathrm{e}} = \frac{\Gamma\left(\frac{\alpha+3}{\gamma}\right)\Gamma\left(\frac{\alpha+5}{\gamma}\right)}{\Gamma\left(\frac{\alpha+4}{\gamma}\right)\Gamma\left(\frac{\alpha+4}{\gamma}\right)} - 1 \tag{97}$$

3.2 Gamma Distribution

When $\gamma = 1$ the modified gamma distribution becomes the gamma distribution.

$$n(r) = a_{\mathrm{u}} r^{\alpha} \exp\left(-br\right). \tag{98}$$

which has a mode value of $r = \frac{\alpha}{b}$. A version of this function is shown in Figure 4.

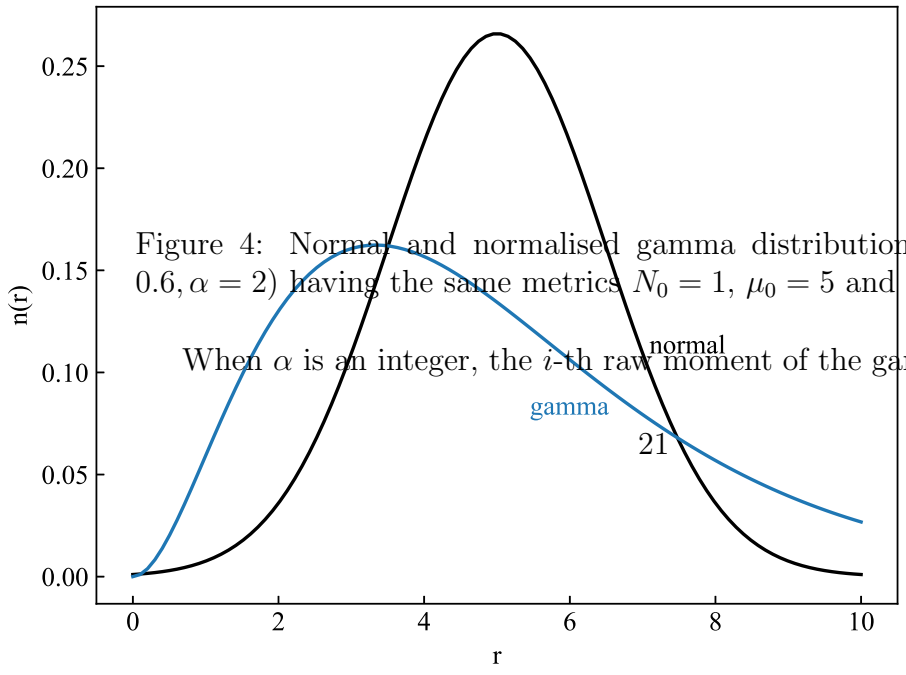


Figure 4: Normal and normalised gamma distributions ($a = 0.108, b = 0.6, \alpha = 2$) having the same metrics $N_0 = 1$, $\mu_0 = 5$ and $\sigma_0 = 1.5$.

When α is an integer, the i-th raw moment of the gamma distribution is

given by

$$m_i = a_{\mathrm{u}} b^{-\alpha-1-i} \Gamma(\alpha+i+1) \quad (99)$$
$$= a_{\mathrm{u}} b^{-\alpha-1-i} (\alpha+i)! \quad (100)$$

The first five raw moments are

$$\begin{aligned} m_0 &= a_{\mathrm{u}} b^{-\alpha-1} \alpha! \\ m_1 &= a_{\mathrm{u}} b^{-\alpha-2} (\alpha+1)! \\ m_2 &= a_{\mathrm{u}} b^{-\alpha-3} (\alpha+2)! \\ m_3 &= a_{\mathrm{u}} b^{-\alpha-4} (\alpha+3)! \\ m_4 &= a_{\mathrm{u}} b^{-\alpha-5} (\alpha+4)! \end{aligned}$$

The moments of the gamma distribution can be used to find the derived metrics:

$$N_0 = a_{\mathrm{u}} b^{-\alpha-1} \alpha! \quad (101)$$

$$A_0 = 4\pi m_2 = 4\pi a_{\mathrm{u}} b^{-\alpha-3} (\alpha+2)! \quad (102)$$
$$V_0 = \frac{4}{3}\pi m_3 = \frac{4}{3}\pi a_{\mathrm{u}} b^{-\alpha-4} (\alpha+3)! \quad (103)$$
$$\mu_0 = \frac{m_1}{m_0} = \frac{\alpha+1}{b} \quad (104)$$
$$r_{\mathrm{e}} = \frac{m_3}{m_2} = \frac{a_{\mathrm{u}} b^{-\alpha-4} (\alpha+3)!}{a_{\mathrm{u}} b^{-\alpha-3} (\alpha+2)!} \quad (105)$$
$$= \frac{\alpha+3}{b} \quad (106)$$
$$v_{\mathrm{e}} = \frac{m_2 m_4}{m_3^2} - 1 = \frac{a_{\mathrm{u}} b^{-\alpha-3} (\alpha+2)! a_{\mathrm{u}} b^{-\alpha-5} (\alpha+4)!}{a_{\mathrm{u}} b^{-\alpha-4} (\alpha+3)! a_{\mathrm{u}} b^{-\alpha-4} (\alpha+3)!} - 1 \quad (107)$$
$$= \frac{(\alpha+4)}{(\alpha+3)} - 1 \quad (108)$$
$$= \frac{1}{\alpha+3} \quad (109)$$

As N_0 does not equal unity, the parameter a_{u} is not the total particle number density. This can be rectified by including a normalising factor i.e.

$$n(r) = \frac{a}{b^{-\alpha-1}\alpha!} r^{\alpha} \exp(-br). \quad (110)$$

In this case a equates to the total particle number density. Note that the values of μ_0, r_{e} and v_{e} are independent of a_{u} and are the same for both representations of the gamma distribution.

Equations 106 and 109 can be used for an alternative formulation of the gamma distribution in r_e and v_e (Hansen and Travis 1974)

$$\alpha = \frac{1}{v_e} - 3 = \frac{1-3v_e}{v_e} \quad \text{and} \quad b = \frac{1}{r_e v_e} \tag{111}$$

$$\Rightarrow n(r) = a_u r^{\frac{1-3v_e}{v_e}} \exp\left(-\frac{r}{r_e v_e}\right) \tag{112}$$

or for the normalised version

$$n(r) = \frac{a}{(r_e v_e)^{\frac{1-2v_e}{v_e}} (\frac{1-3v_e}{v_e})!} r^{\frac{1-3v_e}{v_e}} \exp\left(-\frac{r}{r_e v_e}\right) \tag{113}$$

Figure 5 shows a family of distributions based on this expression where $r_e = 5$.

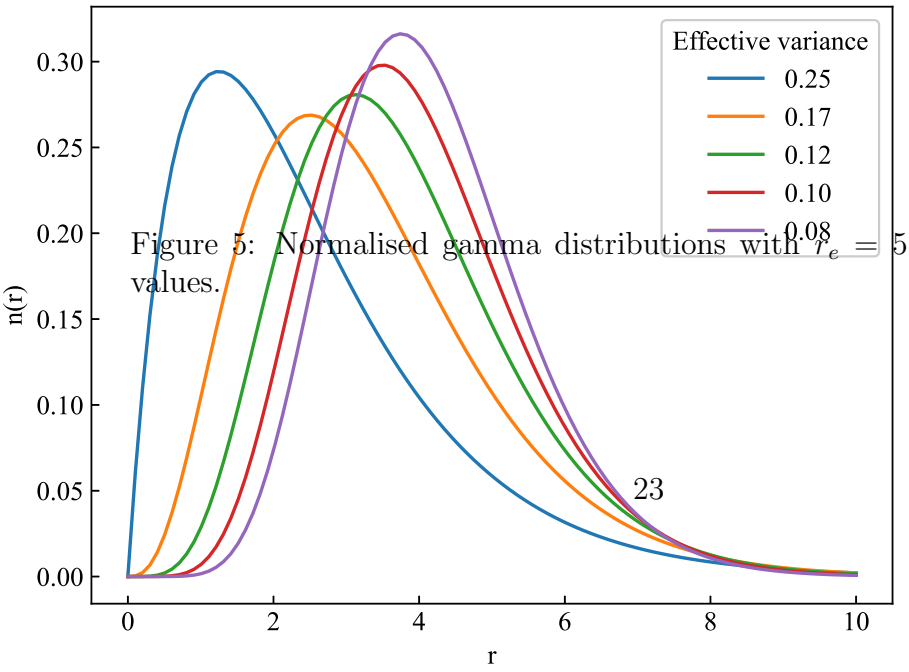


Figure 5: Normalised gamma distributions with $r_e = 5$ and a range of v_e values.

4 Other Distributions

4.1 Inverse Modified Gamma Distribution

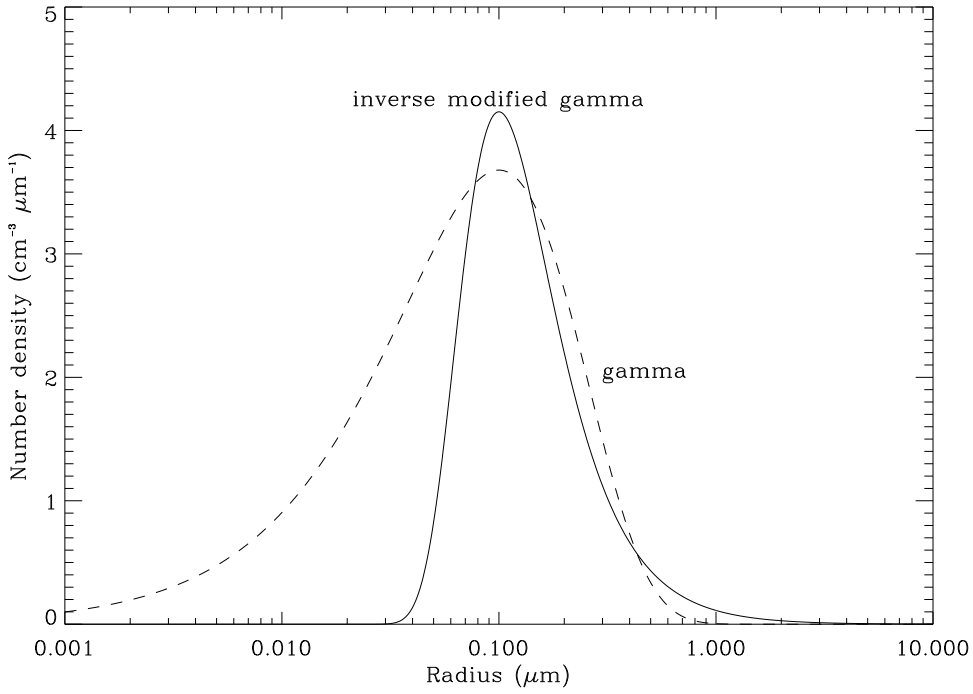


Figure 6: Normalised inverse modified gamma distribution ($\alpha = 2, b = 0.01, \gamma = 2$) and gamma distribution($b - 10$).

The inverse modified gamma distribution is defined by Deepak (1982) as

$$n(r) = ar^{-\alpha} \exp\left(-br^{-\gamma}\right). \tag{114}$$

The mode of the distribution occurs where $r = \left(\frac{\alpha}{b\gamma}\right)^{-1/\gamma}$ and it falls off slowly on the large radius side and exponentially on the small radius side. The raw moments are defined by

$$m_i = \frac{a}{\gamma} b^{-\frac{\alpha-1-i}{\gamma}} \Gamma\left(\frac{\alpha-1-i}{\gamma}\right). \tag{115}$$

The normalized inverse modified gamma distribution can be defined

$$n(r) = a\frac{r^{-\alpha}\exp\left(-br^{-\gamma}\right)}{\frac{1}{\gamma} b^{-\frac{\alpha-1}{\gamma}} \Gamma\left(\frac{\alpha-1}{\gamma}\right)}. \tag{116}$$

The raw moments of this distribution are given by

$$m_i = ab^{\frac{i}{\gamma}} \frac{\Gamma\left(\frac{\alpha-1-i}{\gamma}\right)}{\Gamma\left(\frac{\alpha-1}{\gamma}\right)}. \tag{117}$$

4.2 Regularized Power Law

The regularized power law is defined by Deepak (1982) as

$$n(r) = ab^{\alpha-2} \frac{r^{\alpha-1}}{\left[1+\left(\frac{r}{b}\right)^{\alpha}\right]^{\gamma}}, \tag{118}$$

where the positive constants a, b, α, γ mainly effect the number density, the mode radius, the positive gradient and the negative gradient respectively. The mode radius is given by

$$r = b\left(\frac{\alpha-1}{1+\alpha(\gamma-1)}\right)^{1/\alpha}, \tag{119}$$

and the raw moments by

$$m_i = a\frac{b^i}{\alpha} \frac{\Gamma(1+i/\alpha)\Gamma(\gamma-1-i/\alpha)}{\Gamma(\gamma)}. \tag{120}$$

Hence the distribution normalised so that the total number of particles is a is

$$n(r) = a\alpha\gamma b^{\alpha-2} \frac{r^{\alpha-1}}{\left[1+\left(\frac{r}{b}\right)^{\alpha}\right]^{\gamma}}. \tag{121}$$

4.2.1 Moments

The first three raw moments are

$$\begin{aligned} m_1 &= a\frac{b}{\alpha} \frac{\Gamma(1+1/\alpha)\Gamma(\gamma-1-1/\alpha)}{\Gamma(\gamma)} \\ m_2 &= a\frac{b^2}{\alpha} \frac{\Gamma(1+2/\alpha)\Gamma(\gamma-1-2/\alpha)}{\Gamma(\gamma)} \\ m_3 &= a\frac{b^3}{\alpha} \frac{\Gamma(1+3/\alpha)\Gamma(\gamma-1-3/\alpha)}{\Gamma(\gamma)} \end{aligned}$$

■ above formulae need to be checked and possibly simplified ■

The mean radius, the surface area density and the volume density of a regularized power law distribution are given by

For a regularized power law distribution the effective radius is

$$r_{\mathrm{e}} = \frac{m_3}{m_2} = \frac{a\frac{b^3}{\alpha}\frac{\Gamma(1+3/\alpha)\Gamma(\gamma-1-3/\alpha)}{\Gamma(\gamma)}}{a\frac{b^2}{\alpha}\frac{\Gamma(1+2/\alpha)\Gamma(\gamma-1-2/\alpha)}{\Gamma(\gamma)}} = b\frac{\Gamma(1+3/\alpha)\Gamma(\gamma-1-3/\alpha)}{\Gamma(1+2/\alpha)\Gamma(\gamma-1-2/\alpha)}, \quad (122)$$

5 Modelling the Evolution of an Aerosol Size Distribution

For retrieval purposes it is necessary to describe the evolution of an aerosol size distribution. Consider the case where an aerosol size distribution is described by three modes which are parametrized by a mode radius, $r_{m,i}$ and a spread, σ_i. We wish to alter the mixing ratios, μ_i, of each of the modes to achieve a given effective radius $r_{\rm e}$. How do we do this?

Firstly calculate the effective radius of each of the modes according to

$$r_{\rm e,i} = r_{m,i} \exp\left(\frac{5}{2}\ln^2 S_i\right). \tag{123}$$

If $r_{\rm e} \leq r_{\rm e,1}$ then $\mu = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}$ and $r_m = \begin{pmatrix} r_{\rm e,i}/\exp\left(\frac{5}{2}\ln^2 S_1\right) \\ r_{m,2} \\ r_{m,3} \end{pmatrix}$.

Similarly if $r_{\rm e} \geq r_{\rm e,3}$ then $\mu = \begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$ and $r_m = \begin{pmatrix} r_{m,1} \\ r_{m,2} \\ r_{\rm e,i}/\exp\left(\frac{5}{2}\ln^2 S_3\right) \end{pmatrix}$.

If $r_{\rm e,1} < r_{\rm e} < r_{\rm e,3}$ then μ_1 and is estimated by linearly interpolating between [0,1] as a function of $r_{\rm e}$ i.e.

$$\mu_1 = \frac{r_{\rm e} - r_{\rm e,1}}{r_{\rm e,3} - r_{\rm e,1}} \tag{124}$$

We now have two equations

$$\begin{aligned} \mu_1 + \mu_2 + \mu_3 &= 1 \\ \frac{\mu_1 r_{m,1}^3 \exp\left(\frac{9}{2}\ln^2 S_1\right) + \mu_2 r_{m,2}^3 \exp\left(\frac{9}{2}\ln^2 S_2\right) + \mu_3 r_{m,3}^3 \exp\left(\frac{9}{2}\ln^2 S_3\right)}{\mu_1 r_{m,1}^2 \exp\left(2\ln^2 S_1\right) + \mu_2 r_{m,2}^2 \exp\left(2\ln^2 S_2\right) + \mu_3 r_{m,3}^2 \exp\left(2\ln^2 S_3\right)} &= r_{\rm e} \end{aligned}$$

and two unknowns i.e. μ_2 and μ_3. The second equation is simplified by substitution i.e.

$$\frac{A\mu_1 + B\mu_2 + C\mu_3}{D\mu_1 + E\mu_2 + F\mu_3} = r_{\rm e} \tag{125}$$

and the two equations solved to give

$$\mu_2 = \frac{r_e E - B - \mu_1(A - B + r_e(E - D))}{C - B + r_e(E - F)} \tag{126}$$

$$\mu_3 = \frac{r_e F - C - \mu_1(A - C + r_e(F - D))}{B - C + r_e(F - E)} \tag{127}$$

6 Optical Properties

6.1 Volume Absorption, Scattering and Extinction Coefficients

The volume absorption coefficient, $\beta^{\text{abs}}(\lambda, r)$, the volume scattering coefficient, $\beta^{\text{sca}}(\lambda, r)$, and the volume extinction coefficient, $\beta^{\text{ext}}(\lambda, r)$, represent the energy removed from a beam per unit distance by absorption, scattering, and by both absorption and scattering. For a monodisperse aerosol they are calculated from

$$\text{monodisperse only} \begin{cases} \beta^{\text{abs}}(\lambda, r) = \sigma^{\text{abs}}(\lambda, r)N(r) = \pi r^2 Q^{\text{abs}}(\lambda, r)N(r), \\ \beta^{\text{sca}}(\lambda, r) = \sigma^{\text{sca}}(\lambda, r)N(r) = \pi r^2 Q^{\text{sca}}(\lambda, r)N(r), \\ \beta^{\text{ext}}(\lambda, r) = \sigma^{\text{ext}}(\lambda, r)N(r) = \pi r^2 Q^{\text{ext}}(\lambda, r)N(r), \end{cases} \quad (128)$$

where $N(r)$ is the number of particles per unit volume at some radius, r. The absorption cross section, $\sigma^{\text{ext}}(\lambda, r)$, the scattering cross section, $\sigma^{\text{ext}}(\lambda, r)$, and the extinction cross section, $\sigma^{\text{ext}}(\lambda, r)$, are determined from the extinction efficiency factor, $Q^{\text{ext}}(\lambda, r)$, extinction efficiency factor, $Q^{\text{ext}}(\lambda, r)$, extinction efficiency factor, $Q^{\text{ext}}(\lambda, r)$, respectively.

For a collection of particles, the volume coefficients are given by

$$\beta^{\text{abs}}(\lambda) = \int_0^\infty \sigma^{\text{abs}}(\lambda, r)n(r)\,dr = \int_0^\infty \pi r^2 Q^{\text{abs}}(\lambda, r)n(r)\,dr, \quad (129)$$

$$\beta^{\text{sca}}(\lambda) = \int_0^\infty \sigma^{\text{sca}}(\lambda, r)n(r)\,dr = \int_0^\infty \pi r^2 Q^{\text{sca}}(\lambda, r)n(r)\,dr, \quad (130)$$

$$\beta^{\text{ext}}(\lambda) = \int_0^\infty \sigma^{\text{ext}}(\lambda, r)n(r)\,dr = \int_0^\infty \pi r^2 Q^{\text{ext}}(\lambda, r)n(r)\,dr, \quad (131)$$

where $n(r)$ represents the number of particles with radii between r and $r+dr$ per unit volume. It is also useful to define the quantities per particle i.e.

$$\bar{\sigma}^{\text{abs}}(\lambda) = \int_0^\infty \sigma^{\text{abs}} n(r)\,dr \Big/ \int_0^\infty n(r)\,dr = \frac{\beta^{\text{abs}}(\lambda)}{N_0}, \quad (132)$$

$$\bar{\sigma}^{\text{back}}(\lambda) = \int_0^\infty \sigma^{\text{sca}} n(r)\,dr \Big/ \int_0^\infty n(r)\,dr = \frac{\beta^{\text{sca}}(\lambda)}{N_0}, \quad (133)$$

$$\bar{\sigma}^{\text{ext}}(\lambda) = \int_0^\infty \sigma^{\text{ext}} n(r)\,dr \Big/ \int_0^\infty n(r)\,dr = \frac{\beta^{\text{ext}}(\lambda)}{N_0}, \quad (134)$$

where $\bar{\sigma}^{\text{abs}}(\lambda)$, $\bar{\sigma}^{\text{back}}(\lambda)$ and $\bar{\sigma}^{\text{ext}}(\lambda)$ are the mean absorption cross section, the mean scattering cross section and the mean extinction cross section respectively.

6.2 Back Scatter

■ to be done ■

6.3 Phase Function

The phase function represents the redistribution of the scattered energy.

For a collection of particles, the phase function is given by

$$P(\lambda,\theta) \;=\; \frac{1}{\beta^{\rm sca}}\int_0^\infty \pi r^2 Q^{\rm sca}(\lambda,r)P(\lambda,r,\theta)n(r)\,dr. \tag{135}$$

6.4 Single Scatter Albedo

The single scatter albedo is the ratio of the energy scattered from a particle to that intercepted by the particle. Hence

$$\omega(\lambda) \;=\; \frac{\beta^{\rm sca}(\lambda)}{\beta^{\rm ext}(\lambda)}. \tag{136}$$

6.5 Asymmetry Parameter

The asymmetry parameter is the average cosine of the scattering angle, weighted by the intensity of the scattered light as a function of angle. It has the value 1 for perfect forward scattering, 0 for isotropic scattering and -1 for perfect backscatter.

$$g \;=\; \frac{1}{\beta^{\rm sca}}\int_0^\infty \pi r^2 Q^{\rm sca}(\lambda,r)g(\lambda,r)n(r)\,dr \tag{137}$$

6.6 Integration

The integration of an optical properties over size is usually reduced from the interval $r = [0,\infty]$ to $r = [r_{\rm l}, r_{\rm u}]$ as $n(r) \to 0$ as $r \to 0$ and $r \to \infty$. Numerically an integral over particle size becomes

$$\int_{r_{\rm l}}^{r_{\rm u}} f(r)n(r)\,dr \;=\; \sum_{i=1}^{n} w_i f(r_i) \tag{138}$$

where w_i are the weights at discrete values of radius, r_i.

For a log normal size distribution the integrals are

$$\beta^{\rm ext}(\lambda) \;=\; \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\int_{r_{\rm l}}^{r_{\rm u}} rQ^{\rm ext}(\lambda,r)\exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_{\rm m}}{\sigma}\right)^2\right]dr \tag{139}$$

$$
\begin{aligned}
&= \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\sum_{i=1}^{n} w_i r_i Q^{\mathrm{ext}}(\lambda, r_i)\exp\left[-\frac{1}{2}\left(\frac{\ln r_i - \ln r_\mathrm{m}}{\sigma}\right)^2\right] \\
&= \sum_{i=1}^{n} w_i' Q^{\mathrm{ext}}(\lambda, r_i) \\
\beta^{\mathrm{abs}}(\lambda) &= \sum_{i=1}^{n} w_i' Q^{\mathrm{abs}}(\lambda, r_i) \\
\beta^{\mathrm{sca}}(\lambda) &= \sum_{i=1}^{n} w_i' Q^{\mathrm{sca}}(\lambda, r_i) \\
P(\lambda,\theta) &= \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\frac{1}{\beta^{\mathrm{sca}}}\int_{r_\mathrm{l}}^{r_\mathrm{u}} r Q^{\mathrm{sca}}(\lambda, r) P(\lambda, r, \theta)\exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_\mathrm{m}}{\sigma}\right)^2\right] dr \\
&= \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\frac{1}{\beta^{\mathrm{sca}}}\sum_{i=1}^{n} w_i r_i Q^{\mathrm{sca}}(\lambda, r_i) P(\lambda, r_i, \theta)\exp\left[-\frac{1}{2}\left(\frac{\ln r_i - \ln r_\mathrm{m}}{\sigma}\right)^2\right] \\
&= \frac{1}{\beta^{\mathrm{sca}}}\sum_{i=1}^{n} w_i' Q^{\mathrm{sca}}(\lambda, r_i) P(\lambda, r_i, \theta) \\
g &= \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\frac{1}{\beta^{\mathrm{sca}}}\int_{0}^{\infty} r Q^{\mathrm{sca}}(\lambda, r) g(\lambda, r)\exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_\mathrm{m}}{\sigma}\right)^2\right] dr \\
&= \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\frac{1}{\beta^{\mathrm{sca}}}\sum_{i=1}^{n} w_i r_i Q^{\mathrm{sca}}(\lambda, r_i) g(\lambda, r_i)\exp\left[-\frac{1}{2}\left(\frac{\ln r_i - \ln r_\mathrm{m}}{\sigma}\right)^2\right] \\
&= \frac{1}{\beta^{\mathrm{sca}}}\sum_{i=1}^{n} w_i' Q^{\mathrm{sca}}(\lambda, r_i) g(\lambda, r_i)
\end{aligned}
$$

where

$$
w_i' = \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}} r_i \exp\left[-\frac{1}{2}\left(\frac{\ln r_i - \ln r_\mathrm{m}}{\sigma}\right)^2\right] w_i \tag{140}
$$

6.7 Formulae for Practical Use

As part of the retrieval process it is helpful to have analytic expression for the partial derivatives of β^{ext} (Equation 139) with respect to the size distribution parameters (N_0, r_m, σ).

$$
\frac{\partial \beta^{\mathrm{ext}}}{\partial N_0} = \frac{1}{\sigma}\sqrt{\frac{\pi}{2}}\int_0^\infty r Q^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_\mathrm{m})^2}{2\sigma^2}\right] dr, \tag{141}
$$

$$
\frac{\partial \beta^{\mathrm{ext}}}{\partial r_\mathrm{m}} = \frac{N_0}{r_\mathrm{m}\sigma^3}\sqrt{\frac{\pi}{2}}\int_0^\infty (\ln r - \ln r_\mathrm{m}) r Q^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_\mathrm{m})^2}{2\sigma^2}\right] dr, \tag{142}
$$

$$\begin{aligned}\frac{\partial\beta^{\rm ext}}{\partial\sigma} &= -\frac{N_0}{\sigma^2}\sqrt{\frac{\pi}{2}}\int_0^\infty rQ^{\rm ext}(\lambda,r)\exp\left[-\frac{(\ln r-\ln r_{\rm m})^2}{2\sigma^2}\right]\,dr \\ &\quad +\frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\int_0^\infty \frac{(\ln r-\ln r_{\rm m})^2}{\sigma^3}rQ^{\rm ext}(\lambda,r)\exp\left[-\frac{(\ln r-\ln r_{\rm m})^2}{2\sigma^2}\right]\,dr, \\ &= \frac{N_0}{\sigma^2}\sqrt{\frac{\pi}{2}}\int_0^\infty \left[\frac{(\ln r-\ln r_{\rm m})^2}{\sigma^2}-1\right]rQ^{\rm ext}(\lambda,r)\exp\left[-\frac{(\ln r-\ln r_{\rm m})^2}{2\sigma^2}\right]\,dr.\end{aligned} \tag{143}$$

To linearise the retrieval it is better to retrieve T $(=\ln N_0)$ rather than N_0. In addition to limit the values of $r_{\rm m}$ and σ to positive quantities it is better to retrieve $l_{\rm m}$ $(=\ln r_{\rm m})$ and G $(=\ln\sigma)$. In terms of these new variables volume extinction coefficient for a log normal size distribution is

$$\beta^{\rm ext}(\lambda)=\frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l}Q^{\rm ext}(l,\lambda)\exp\left[-\frac{(l-l_{\rm m})^2}{2\exp(2G)}\right]\,dl. \tag{144}$$

The partial derivatives of the transformed parameters (Equation 144) are

$$\frac{\partial\beta^{\rm ext}}{\partial T} = \frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l}Q^{\rm ext}(l,\lambda)\exp\left[-\frac{(l-l_{\rm m})^2}{2\exp(2G)}\right]\,dl, \tag{145}$$

$$\frac{\partial\beta^{\rm ext}}{\partial l_{\rm m}} = \frac{\exp T}{\exp(3G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty (l-l_{\rm m})e^{2l}Q^{\rm ext}(l,\lambda)\exp\left[-\frac{(l-l_{\rm m})^2}{2\exp(2G)}\right]\,dl, \tag{146}$$

$$\begin{aligned}\frac{\partial\beta^{\rm ext}}{\partial G} &= -\frac{\exp T}{\exp(G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l}Q^{\rm ext}(l,\lambda)\exp\left[-\frac{(l-l_{\rm m})^2}{2\exp(2G)}\right]\,dl \\ &\quad +\frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty \frac{(l-l_{\rm m})^2}{\exp(2G)}e^{2l}Q^{\rm ext}(l,\lambda)\exp\left[-\frac{(l-l_{\rm m})^2}{2\exp(2G)}\right]\,dl, \\ &= \frac{\exp T}{\exp(G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty \left[\frac{(l-l_{\rm m})^2}{\exp(2G)}-1\right]e^{2l}Q^{\rm ext}(l,\lambda)\exp\left[-\frac{(l-l_{\rm m})^2}{2\exp(2G)}\right]\,dl.\end{aligned} \tag{147}$$

6.7.1 Extinction Limits for a Log-Normal Distribution

■ to be done ■

In section 2.2.2 the limits for calculating the integral of a log-normal distribution where found as a fraction of the mode value. A similar problem occurs when determining β^{ext}.

If we consider the extinction efficiency to be a function of $1/x$ for size parameters less than 6 and 2 greater than 6 then there are three cases to consider.

i Both r_- and r_+ are such that the x values of interest are less than 6.

ii Both r_- and r_+ are such that the x values of interest straddle 6 but mode is < 6.

iii Both r_- and r_+ are such that the x values of interest straddle 6 but mode is > 6.

iv Both r_- and r_+ are such that the x values of interest are greater than 6.

Case i The function of interest becomes

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln S} \frac{1}{r^2} \exp\left[-\frac{(\ln r - \ln r_{\text{m}})^2}{2\ln^2 S}\right] \tag{148}$$

noting that r_{m} is now just a parameter and not the median of this distribution. Let

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln S} \frac{1}{r^2} \exp\left[-\frac{(\ln r - \ln r_{\text{m}})^2}{2\ln^2 S}\right] = \frac{A}{r^2} \exp[B] \tag{149}$$

$$\text{where } A = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln S},\ B = -\frac{(\ln r - \ln r_{\text{m}})^2}{2\ln^2 S} \text{ and } \frac{dB}{dr} = -\frac{(\ln r - \ln r_{\text{m}})}{r\ln^2 S}.$$

then

$$\frac{dn(r)}{dr} = -2\frac{A}{r^3}\exp[B] + \frac{A}{r^2}\exp[B]\frac{dB}{dr} \tag{150}$$

$$= -2\frac{A}{r^3}\exp[B] - \frac{A}{r^3}\exp[B]\frac{(\ln r - \ln r_{\text{m}})}{\ln^2 S} \tag{151}$$

Setting the left hand side to zero so $r = r_{\text{M}}$

$$0 = -2 - \frac{(\ln r_{\text{M}} - \ln r_{\text{m}})}{\ln^2 S} \tag{152}$$

$$\ln r_{\text{M}} = \ln r_{\text{m}} - 2\ln^2 S \tag{153}$$

The peak value of the distribution (at the mode) is then

$$\begin{aligned}
n(r_{\mathrm{M}}) &= \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{(\exp(\ln r_{\mathrm{m}} - 2\ln^2 S))^2}\exp\left[-\frac{(\ln r_{\mathrm{m}} - 2\ln^2 S - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] \\
&= \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{(\exp(2(\ln r_{\mathrm{m}} - 2\ln^2 S)))}\exp\left[-\frac{(-2\ln^2 S)^2}{2\ln^2 S}\right] \qquad (154) \\
&= \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{(\exp(2(\ln r_{\mathrm{m}} - 2\ln^2 S)))}\exp\left[-2\ln^2 S\right] \qquad (155) \\
&= \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\exp\left[-2\ln^2 S - 2(\ln r_{\mathrm{m}} - 2\ln^2 S)\right] \qquad (156) \\
&= \frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\exp\left[-2(\ln r_{\mathrm{m}} - \ln^2 S)\right] \qquad (157)
\end{aligned}$$

If some fraction η of the peak value is chosen then distribution limits can be calculated from

$$\begin{aligned}
\frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{r_\pm^2}\exp\left[-\frac{(\ln r_\pm - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] &= \eta\frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\exp\left[-2(\ln r_{\mathrm{m}} - \ln^2 S)\right] \\
\exp\left[-\frac{(\ln r_\pm - \ln r_{\mathrm{m}})^2}{2\ln^2 S}\right] &= \eta r_\pm^2\exp\left[-2(\ln r_{\mathrm{m}} - \ln^2 S)\right] \\
-\frac{(\ln r_\pm - \ln r_{\mathrm{m}})^2}{2\ln^2 S} &= \ln\eta + 2\ln r_\pm - 2\ln r_{\mathrm{m}} + 2\ln^2 S \\
\ln^2 r_\pm - 2\ln r_{\mathrm{m}}\ln r_\pm + \ln^2 r_{\mathrm{m}} &= -2\ln^2 S\ln\eta - 4\ln^2 S\ln r_\pm + 4\ln^2 S\ln r_{\mathrm{m}} - 4\ln^4 S \\
\ln^2 r_\pm + (4\ln^2 S - 2\ln r_{\mathrm{m}})\ln r_\pm + \ln^2 r_{\mathrm{m}} &= -2\ln^2 S\ln\eta + 4\ln^2 S\ln r_{\mathrm{m}} - 4\ln^4 S
\end{aligned}$$

$$\ln^2 r_\pm + (4\ln^2 S - 2\ln r_{\mathrm{m}})\ln r_\pm + \ln^2 r_{\mathrm{m}} + 2\ln^2 S\ln\eta - 4\ln^2 S\ln r_{\mathrm{m}} + 4\ln^4 S = 0$$

which has solution

$$\begin{aligned}
\ln r_\pm &= -\frac{(4\ln^2 S - 2\ln r_{\mathrm{m}})}{2} \\
&\quad \pm\frac{\sqrt{(4\ln^2 S - 2\ln r_{\mathrm{m}})^2 - 4\left(\ln^2 r_{\mathrm{m}} + 2\ln^2 S\ln\eta - 4\ln^2 S\ln r_{\mathrm{m}} + 4\ln^4 S\right)}}{2} \\
&= -2\ln^2 S + \ln r_{\mathrm{m}} \pm\sqrt{-2\ln^2 S\ln\eta} \qquad (158)
\end{aligned}$$

The value in square root is positive as η is always less than one e.g. 10^{-3}.

Case ii r_- as in case (i). r_+ found through

$$\begin{aligned}
\frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\frac{1}{r_+}\exp\left[-\frac{(\ln r_+ - \ln r_\mathrm{m})^2}{2\ln^2 S}\right] &= \eta\frac{N_0}{\sqrt{2\pi}}\frac{1}{\ln S}\exp\left[-2(\ln r_\mathrm{m} - \ln^2 S)\right] \\
\frac{1}{r_+}\exp\left[-\frac{(\ln r_+ - \ln r_\mathrm{m})^2}{2\ln^2 S}\right] &= \eta\exp\left[-2(\ln r_\mathrm{m} - \ln^2 S)\right] \\
-\frac{(\ln r_+ - \ln r_\mathrm{m})^2}{2\ln^2 S} &= \ln\eta + \ln r_+ - 2(\ln r_\mathrm{m} - \ln^2 S) \\
-\ln^2 r_+ + 2\ln r_\mathrm{m}\ln r_+ - \ln^2 r_\mathrm{m} &= 2\ln^2 S\ln\eta + 2\ln^2 S\ln r_+ - 4\ln^2 S(\ln r_\mathrm{m} - \ln^2 S)
\end{aligned}$$

$$\ln^2 r_+ + 2(\ln^2 S - \ln r_\mathrm{m})\ln r_+ + \ln^2 r_\mathrm{m} + \ln^2 S(4\ln^2 S + 2\ln\eta - 4\ln r_\mathrm{m}) = 0$$

$$\ln r_+ = -(\ln^2 S - \ln r_\mathrm{m}) \pm \sqrt{(\ln^2 S - \ln r_\mathrm{m})^2 - \ln^2 S(4\ln^2 S + 2\ln\eta - 4\ln r_\mathrm{m}) - \ln^2 r_\mathrm{m}}.$$

6.8 Cloud Liquid Water Path

The mass l of liquid per unit area in a cloud with a homogeneous size distribution is given by

$$l = \rho\int_0^\infty \frac{4}{3}\pi r^3 n(r)\,dr \times z \tag{159}$$

where ρ is the density of the cloud material (water or ice) and z is the vertical distance through the cloud. The liquid water path is usually expressed as $\mathrm{g\,m^{-2}}$. Note that

$$\tau = \beta^\mathrm{ext} \times z \tag{160}$$

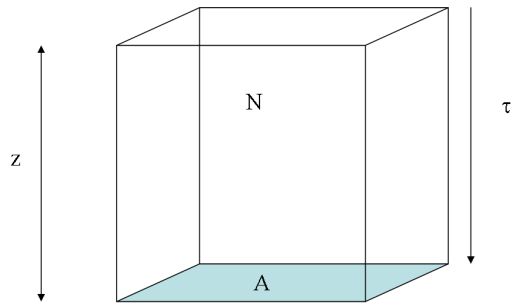
so

$$l = \rho\tau\frac{\int_0^\infty \frac{4}{3}\pi r^3 n(r)\,dr}{\beta^\mathrm{ext}} \tag{161}$$

$$= \frac{4}{3}\pi\rho\tau\frac{\int_0^\infty r^3 n(r)\,dr}{\int_0^\infty \pi r^2 Q^\mathrm{ext}(\lambda, r)n(r)\,dr}. \tag{162}$$

For drops large with respect to wavelength we assume $Q^\mathrm{ext}(\lambda, r) = 2$. Hence

$$l = \frac{4}{6}\rho\tau\frac{\int_0^\infty r^3 n(r)\,dr}{\int_0^\infty r^2 n(r)\,dr} = \frac{2}{3}\rho\tau r_e \tag{163}$$



So for a water cloud ($\rho = 1$ g cm^{-3}) of $\tau = 10$, $r_e = 15\,\mu$m we get

$$l = \frac{2}{3} \times 1 \times 10 \times 15 \text{ g cm}^{-3}\mu\text{m} = 100 \text{ g m}^{-2} \quad (164)$$

While for an ice cloud (-45 °C, $\rho = 0.920$ g cm^{-3}) of $\tau = 1$, $r_e = 25\,\mu$m we get

$$l = \frac{2}{3} \times 0.92 \times 1 \times 25 \text{ g cm}^{-3}\mu\text{m} = 15 \text{ g m}^{-2} \quad (165)$$

.

6.9 Aerosol Mass

Consider the measurement of optical depth and effective radius made by a imaging instrument. How can this be related to the mass of aerosol present in the atmosphere? Consider a volume observed by the instrument whose area is A. If ρ is the density of the aerosol and Z is the height of this volume then the total mass of aerosol, M, in the box is given by

$$M = \rho \times v \times N \times A \times Z \quad (166)$$

where N is the number of particles per unit volume and v is the average volume of each particle. If we divide both sides by A we obtain the mass per unit area m, i.e.

$$m = \rho \times v \times N \times Z \quad (167)$$

This formula can re-expressed in terms of more familiar optical measurements of the volume. First note that the optical depth is related to the β^{ext} by

$$\tau = \beta^{\text{ext}} \times Z \quad (168)$$

so that

$$m = \frac{\rho \times v \times N \times \tau}{\beta^{\text{ext}}} \quad (169)$$

For a given size distribution $n(r)$ the average volume of each particle is

$$v = \frac{\int_0^\infty \frac{4}{3}\pi r^3 n(r)\,dr}{N} \tag{170}$$

so that the mass per unit area is given by

$$m = \frac{\rho\tau}{\beta^{\text{ext}}}\int_0^\infty \frac{4}{3}\pi r^3 n(r)\,dr \tag{171}$$

The important thing to note here is that N disappears explicitly from the equation.

For optical measurement it is more common to know the effective radius rather than the full size distribution. In terms of r_{e} the mass per unit area is given by

$$m = \frac{4\pi\rho\tau}{3\beta^{\text{ext}}}\int_0^\infty r^3 n(r)\,dr \times \frac{\int_0^\infty r^2 n(r)\,dr}{\int_0^\infty r^2 n(r)\,dr} = \frac{4\rho\tau}{3\tilde{Q}^{\text{ext}}} r_{\text{e}} \tag{172}$$

which uses an area weighted extinction efficiency

$$\tilde{Q}^{\text{ext}} = \frac{\beta^{\text{ext}}}{\pi\int_0^\infty r^2 n(r)\,dr} = \frac{\int_0^\infty \sigma^{\text{ext}} n(r)\,dr}{\int_0^\infty \pi r^2 n(r)\,dr} = \frac{\int_0^\infty \pi r^2 Q^{\text{ext}} n(r)\,dr}{\int_0^\infty \pi r^2 n(r)\,dr} \tag{173}$$

If the particles with the size distribution are mostly much larger than the wavelength then $Q^{\text{ext}} \to 2$ and $\tilde{Q}^{\text{ext}} \approx 2$. With this assumption Equation 163 becomes identical to Equation 172 whose derivation made the same approximation.

If the aerosol size distribution is log-normal with number density N_0, mode radius r_m and spread σ then the integral in Equation 171 can be completed analytically i.e.

$$m = \frac{\rho\tau}{\beta^{\text{ext}}}\frac{4}{3}\pi N_0 r_{\text{m}}^3 \exp\left(\frac{9}{2}\sigma^2\right) \tag{174}$$

Typically ρ is in $\text{g}\,\text{cm}^{-3}$, N_0 is in cm^{-3}, r_{m} is in μm, and β^{ext} is in km^{-1} so that the units of m are

$$\frac{\frac{\text{g}}{\text{cm}^3}\frac{1}{\text{cm}^3}\mu\text{m}^3}{\frac{1}{\text{km}}} = \frac{\text{g}}{10^{-6}\,\text{m}^3}\frac{1}{10^{-6}\,\text{m}^3}10^{-18}\,\text{m}^3 10^3\,\text{m} = 10^{-3}\,\text{g m}^{-2} \tag{175}$$

Table 4 list the bulk density of some aerosol components.

If the effective radius, r_{e}, is known rather than r_{m} then we can use the relationship between r_{e} and r_m

$$r_{\text{e}} = r_m \exp\left(\frac{5}{2}\sigma^2\right), \tag{176}$$

Substance	**Density (g cm^{-3})**	**Reference**
Ice	0.92	
Volcanic ash	2.42±0.79	Bayhurst et al. (1994)
Water	1	

Table 4: Density of some materials that form aerosols.

to get

$$m = \frac{4\rho\tau\pi N_0}{3\beta^{\text{ext}}} r_{\text{e}}^3 \exp\left(-\frac{15}{2}\sigma^2\right) \exp\left(\frac{9}{2}\sigma^2\right) = \frac{4\rho\tau\pi N_0}{3\beta^{\text{ext}}} r_{\text{e}}^3 \exp\left(-3\sigma^2\right). \quad (177)$$

Equating this expression to Equation 163 gives

$$\tilde{Q}^{\text{ext}} = \frac{\beta^{\text{ext}}}{\pi N_0 r_{\text{e}}^2 \exp\left(-3\sigma^2\right)}. \quad (178)$$

which is true for a log-normal distribution.

For a multi-mode log-normal distribution where the i^{th} model is parameterised by N_i, r_i, σ_i and density ρ_i we have

$$m = \frac{\tau \times \rho \times N \times v}{\beta^{\text{ext}}} = \frac{\tau \sum_{i=1}^{n} \rho_i N_i v_i}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\text{ext}}} \quad (179)$$

where $\bar{\sigma}_i^{\text{ext}}$ is the extinction cross section per particle for the i^{th} mode. Remember the volume per particle for the i^{th}mode is

$$v_i = \frac{4}{3}\pi r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right) \quad (180)$$

Hence

$$m = \tau \frac{\sum_{i=1}^{n} \rho_i N_i \frac{4}{3}\pi r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right)}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\text{ext}}} \quad (181)$$

$$= \frac{4}{3}\pi\tau \frac{\sum_{i=1}^{n} \rho_i N_i r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right)}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\text{ext}}} \quad (182)$$

List of Symbols

r	radius
l	natural log of radius
r_{e}	effective radius
r_{m}	median radius
r_{M}	mode radius
μ_0	arithmetic mean
μ_g	geometric mean
σ_0	standard deviation
σ_g	geometric standard deviation
v_{e}	effective variance
m_{i}	i^{th} moment of a distribution
N_0	number of particles per unit volume
$N(r)$	the number of particles with radii between r and $r + dr$ per unit volume
$n(r)$	differential radius number density distribution
A_0	particle area per unit volume, surface area density
$A(r)$	the surface area of particles with radius between r and $r + dr$ per unit volume
$a(r)$	differential area density distribution
V_0	volume density
$A(r)$	the volume of particles with radius between r and $r + dr$ per unit volume
$v(r)$	differential volume density distribution
M_0	mass density
$M(r)$	the mass of particles with radius between r and $r + dr$ per unit volume
$m(r)$	differential mass density distribution

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Index

asymmetry parameter, 29

central moment, 3
cloud liquid water path, 34

effective radius, 3
effective variance, 3

gamma distribution, 21
geometric mean, 2
geometric standard deviation, 2
geometric variance, 2

inverse modified gamma distribution, 24

logarithmic normal distribution, 6

mean, 2
median, 2
mode, 2
modified gamma distribution, 19
moment, 3

normal distribution, 6

phase function, 29

raw moment, 3
regularized power law, 25

single scatter albedo, 29
size distribution, 1
standard deviation, 2

variance, 2
volume absorption coefficient, 28
volume extinction coefficient, 28
volume scattering coefficient, 28