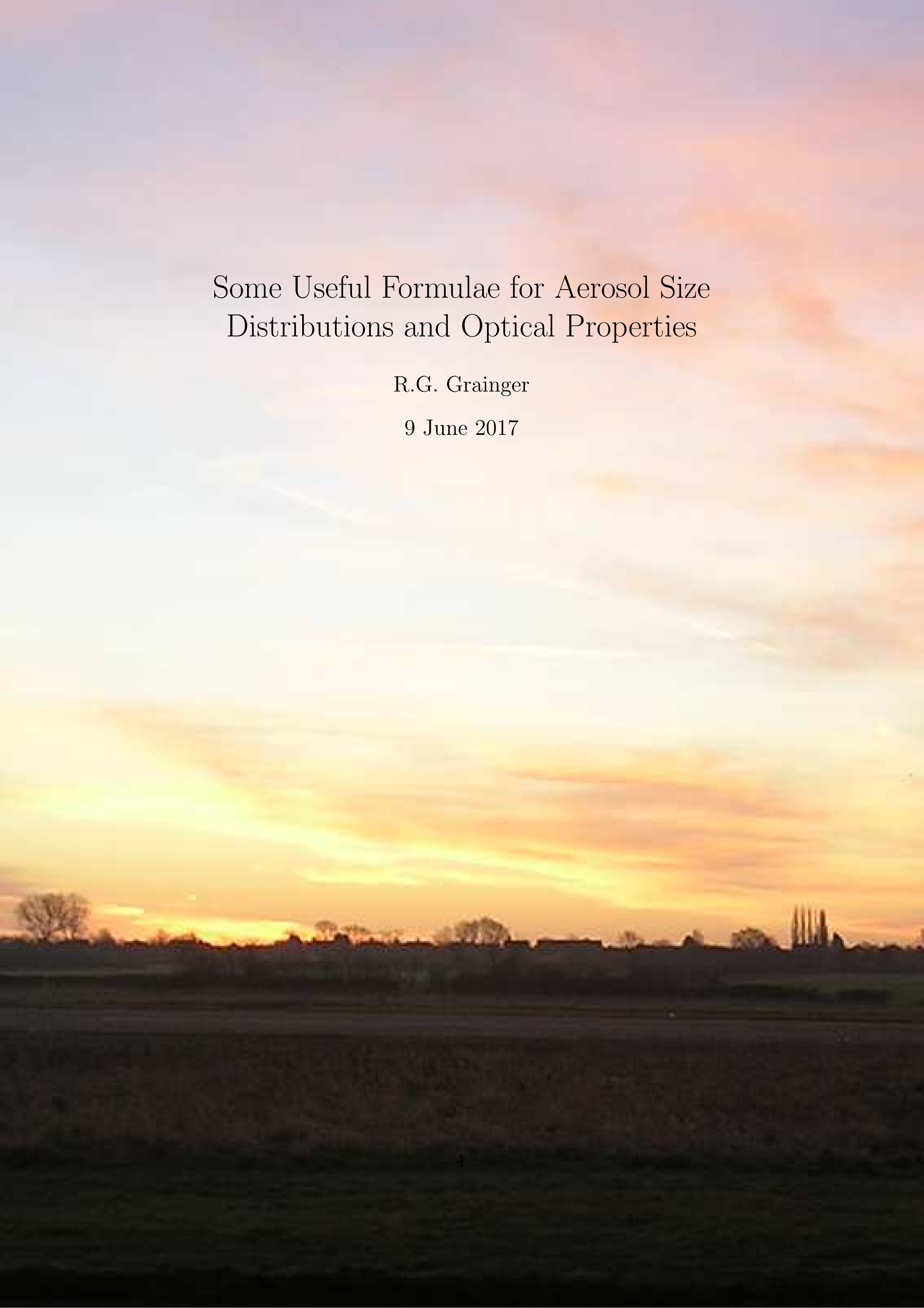
Some Useful Formulae for Aerosol Size Distributions and Optical Properties

R.G. Grainger

9 June 2017



Acknowledgements

I would like to acknowledge helpful comments that improved this manuscript from Dwaipayan Deb.

Contents

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

1 Describing Aerosol Size

A complete description of an ensemble of particles would describe the composition and geometry of each particle. Such an approach for atmospheric aerosols whose concentrations can be $\sim 10,000$ particles per cm^3 is impracticable in most cases. The simplest alternate approach is to use a statistical description of the aerosol. This is assisted by the fact that small liquid drops adopt a spherical shape so that for a chemically homogeneous aerosol the problem becomes one of representing the number distribution of particle radii. The 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 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 Size Distribution

The distribution of particle sizes can be represented by a differential radius number density distribution, $n(r)$ which represents the number of particles with radii between r and $r + dr$ per unit volume, i.e.

$$N(r) = \int_r^{r+dr} n(r)\, dr. \tag{1}$$

Hence

$$n(r) = \frac{dN(r)}{dr}. \tag{2}$$

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

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

1.2 Moments, Mean, Mode, Median and Standard Deviation

The moments of a size distribution are simple metrics to express the shape of the distribution. The i^{th} moment m_i of $n(r)$ is defined[1]

$$m_i = \frac{1}{N_0} \int_0^{\infty} (r - c)^i n(r)\, dr \tag{4}$$

[1]Many texts omit the normalising term before the integral as for a probability distribution function $N_0 = 1$.

where c is some constant. The raw moment is the moment where $c = 0$. The moments about the mean are called central moments.

Generally a size distribution is characterised by its centre and by its spread. The centre of a distribution can be represented by the

mean, μ_0 defined by

$$\mu_0 = \frac{\int_0^\infty r n(r)\, dr}{\int_0^\infty n(r)\, dr} = \frac{1}{N_0} \int_0^\infty r n(r)\, dr. \tag{5}$$

The mean is the first raw moment of a size distribution.

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

median 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).

while the spread is captured through the variance, σ_0^2, defined

$$\sigma_0^2 = \frac{1}{N_0} \int_0^\infty (r - \mu_0)^2 n(r)\, dr. \tag{6}$$

The variance is the second central moment of the size distribution. Finally, the standard deviation, σ_0, of a distribution is the square root of the variance.

1.3 Geometric Mean and Standard Deviation

The geometric mean μ_g of a set of n numbers $\{r_1, r_2, \ldots, r_n\}$ is

$$\mu_g = \sqrt[n]{r_1 \times r_2 \times \ldots \times r_n} \tag{7}$$

The geometric mean can also be written

$$\mu_g = \exp\left[\ln\left(\sqrt[n]{r_1 \times r_2 \times \ldots \times r_n}\right)\right] = \exp\left(\frac{\sum_{i=1}^n \ln r_i}{n}\right) \tag{8}$$

The geometric standard deviation σ_g or S is defined

$$\sigma_g = S = \exp\left[\sqrt{\frac{\sum_{i=1}^n (\ln r_i - \ln \mu_g)^2}{n}}\right] \tag{9}$$

that is, the geometric standard deviation is the exponential of σ, the standard deviation of $\ln r$. So $\ln(\sigma_g) = \ln(S) = \sigma$.

1.4 Effective radius

The usefulness of effective radius, r_{e}, 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 larger than the wavelength of the incident light). Weighting each radius by $\pi r^2 n(r)$ gives

$$r_{\mathrm{e}} \;=\; = \frac{\int_0^\infty r\pi r^2\, n(r) dr}{\int_0^\infty \pi r^2\, n(r) dr} = \frac{\int_0^\infty r^3\, n(r) dr}{\int_0^\infty r^2\, n(r) dr}. \tag{10}$$

This makes clear why the effective radius is sometimes called the area-weighted mean radius. More often the effective radius is defined as the ratio of the third moment of the drop size distribution to the second moment, i.e.

$$r_{\mathrm{e}} \;=\; \frac{m_3}{m_2} = \frac{\int_0^\infty r^3\, n(r) dr}{\int_0^\infty r^2\, n(r) dr}. \tag{11}$$

Although this is identical mathematically it misses why the effective radius is such useful metric and physically descriptive in radiative transfer.

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 1-3.

The distribution of particle area can be represented by a differential area density distribution, $a(r)$ which represents the area of particles whose radii lie between r and $r + dr$ per unit volume, i.e.

$$A(r) \;=\; \int_r^{r+dr} a(r)\, dr. \tag{12}$$

Hence

$$a(r) \;=\; \frac{dA}{dr}. \tag{13}$$

For spherical particles

$$a(r) \;=\; \frac{dA}{dN}\frac{dN}{dr} = 4\pi r^2 n(r). \tag{14}$$

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

$$A_0 = \int_0^\infty a(r)\,dr. \tag{15}$$

The distribution of particle volume can be represented by a differential volume density distribution, $a(r)$ which represents the volume contained in particles whose radii lie between r and $r + dr$ per unit volume, i.e.

$$V(r) = \int_r^{r+dr} v(r)\,dr. \tag{16}$$

Hence

$$v(r) = \frac{dV}{dr}. \tag{17}$$

For spherical particles

$$v(r) = \frac{dV}{dN}\frac{dN}{dr} = \frac{4}{3}\pi r^3 n(r). \tag{18}$$

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

$$V_0 = \int_0^\infty v(r)\,dr. \tag{19}$$

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

$$M(r) = \int_r^{r+dr} m(r)\,dr. \tag{20}$$

Hence

$$m(r) = \frac{dM}{dr}. \tag{21}$$

For spherical particles

$$m(r) = \frac{dM}{dN}\frac{dN}{dr} = \frac{4}{3}\pi r^3 \rho n(r), \tag{22}$$

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

$$M_0 = \int_0^\infty m(r)\,dr. \tag{23}$$

2 Normal and Logarithmic Normal Distributions

2.1 Definitions

One particle distribution to consider adopting is the normal distribution

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma_0} \exp\left[-\frac{(r-\mu_0)^2}{2\sigma_0^2}\right], \tag{24}$$

where μ_0 is the mean and σ_0 is the standard deviation of the distribution. The size of particles in an aerosol generally covers several orders of magnitude. As a result the normal distribution fit of measured particle sizes often has a very large standard deviation. Another drawback of the normal distribution is that it allows negative radii. Aerosol distributions are much better represented by a normal distribution of the logarithm of the particle radii. Letting $l = \ln(r)$ we have

$$n_l(l) = \frac{dN(l)}{dl} = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \exp\left[-\frac{(l-\mu)^2}{2\sigma^2}\right] \tag{25}$$

where the mean, μ, and the standard deviation, σ, of $l = \ln(r)$ are defined

$$\mu = \frac{\int_{-\infty}^{\infty} l n_l(l)\, dl}{\int_{-\infty}^{\infty} n_l(l)\, dl} = \frac{1}{N_0} \int_{-\infty}^{\infty} l n_l(l)\, dl \tag{26}$$

$$\sigma^2 = \frac{1}{N_0} \int_{-\infty}^{\infty} (l-\mu)^2 n_l(l)\, dl \tag{27}$$

It is common for the lognormal distribution to be expressed in terms of the radius. Noting that

$$\frac{dl}{dr} = \frac{1}{r} \tag{28}$$

then in terms of radius rather than log radius we have

$$n(r) = \frac{dN(r)}{dr} = \frac{dN(l)}{dl}\frac{dl}{dr} = n_l(l)\frac{dl}{dr} = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[-\frac{(\ln r-\mu)^2}{2\sigma^2}\right] \tag{29}$$

It is also common to express the spread of the distribution using the geometric standard deviation, S. From this definition S must be greater or

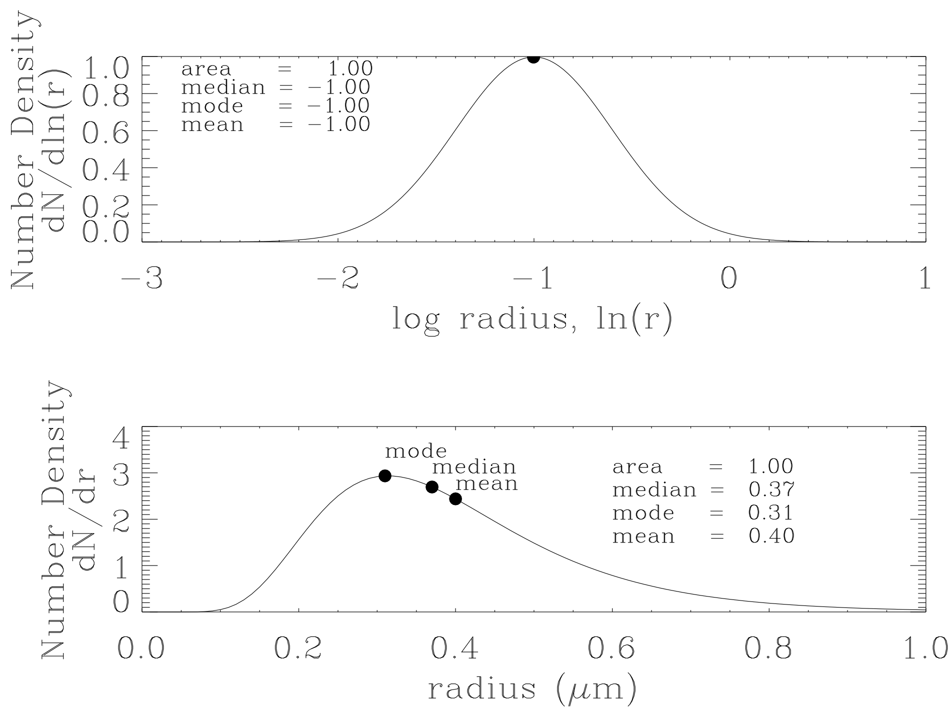


Figure 1: Log-normal distribution with parameters $N_0 = 1$, $\mu = -1$ and $\sigma = 0.4$ plotted in log space (top panel) and linear space (bottom panel). Indicated are the distribution mode, median and mean in log and linear space respectively.

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 lognormal distribution appears in the atmospheric literature using any of combination of r_{m} or μ and σ or S with perhaps the commonest being

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln(S)} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2(S)}\right] \tag{30}$$

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

2.2 Properties of the Lognormal Distribution

2.2.1 Normal versus Lognormal

Figure 1 shows the particle number density per unit log(radius) where the distribution is Gaussian. Also shown in the figure is the particle number density distribution plotted per unit radius. In performing the transform the area (ie. total number of particles) is conserved and the median, mean or mode in log space is the natural logarithm of the median radius, r_{m}, in linear space.

It is possible to show the geometric mean, r_{g}, of the radius is the same as the median in linear space, i.e.

$$r_{\mathrm{g}} = (r_1 \times r_2 \times \ldots \times r_n)^{\frac{1}{n}} \tag{31}$$

$$\Rightarrow \ln(r_{\mathrm{g}}) = \ln(r_1 \times r_2 \times \ldots \times r_n)^{\frac{1}{n}} = \frac{\ln(r_1) + \ln(r_2) + \ldots + \ln(r_n)}{n} \tag{32}$$

which has the continuous form

$$\ln(r_{\mathrm{g}}) = \frac{1}{N_0} \int_{-\infty}^{\infty} l n_l(l)\, dl = \mu = \ln(r_{\mathrm{m}}) \tag{33}$$

$$\Leftrightarrow r_{\mathrm{g}} = r_{\mathrm{m}} \tag{34}$$

2.2.2 Median and Mode

It can be shown that the mode of the log-normal distribution, r_{M}, is related to the median. Let

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln(S)} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2(S)}\right] = \frac{A}{r} \exp[B] \tag{35}$$

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

then

$$\frac{dn(r)}{dr} = -\frac{A}{r^2} \exp[B] + \frac{A}{r} \exp[B] \frac{dB}{dr} \tag{36}$$

$$= -\frac{A}{r^2} \exp[B] - \frac{A}{r^2} \exp[B] \frac{(\ln r - \ln r_{\mathrm{m}})}{\ln^2(S)} \tag{37}$$

Setting the left hand side to zero so r becomes r_{M}

$$0 = -\frac{A}{r_{\mathrm{M}}^2} \exp[B] - \frac{A}{r_{\mathrm{M}}^2} \exp[B] \frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})}{\ln^2(S)} \tag{38}$$

$$-1 = \frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})}{\ln^2(S)} \tag{39}$$

$$\ln r_{\mathrm{M}} = \ln r_{\mathrm{m}} - \ln^2(S) = \ln(r_m) - \sigma^2 \tag{40}$$

2.2.3 Derivatives

The first and second derivatives of Equation 25 are

$$\frac{dn_l}{dN_0} = \frac{n_l}{N_0} \tag{41}$$

$$\frac{dn_l}{dl} = -\frac{(l-\mu)}{\sigma^2}n_l \tag{42}$$

$$\frac{dn_l}{d\sigma} = n_l\left[\frac{(l-\mu)^2}{\sigma^3} - \frac{1}{\sigma}\right] \tag{43}$$

$$\frac{d^2n_l}{dN_0^2} = 0 \tag{44}$$

$$\frac{d^2n_l}{dl^2} = n_l\left[\frac{(l-\mu)^2}{\sigma^4} - \frac{l}{\sigma^2}\right] \tag{45}$$

$$\frac{d^2n_l}{d\sigma^2} = n_l\left\{\left[\frac{(l-\mu)^2}{\sigma^3} - \frac{1}{\sigma}\right]^2 - 3\frac{(l-\mu)^2}{\sigma^4} + \frac{1}{\sigma^2}\right\} \tag{46}$$

If using Equation 30 the derivatives are :

$$\frac{dn}{dN_0} = \frac{n}{N_0} \tag{47}$$

$$\frac{dn}{dr} = -\frac{n}{r}\left[1 + \frac{(\ln r - \ln r_{\mathrm{m}})}{\ln^2(S)}\right] \tag{48}$$

$$\frac{dn}{dS} = \frac{n}{S\ln(S)}\left[\frac{(\ln r - \ln r_{\mathrm{m}})^2}{\ln^2(S)} - 1\right] \tag{49}$$

The second derivatives are:

$$\frac{d^2n}{dN_0^2} = 0 \tag{50}$$

$$\frac{d^2n}{dr^2} = n\left[\frac{2}{r^2} + \frac{3\ln r - 3\ln r_{\mathrm{m}} - 1}{r^2\ln^2(S)} + \frac{(\ln r - \ln r_{\mathrm{m}})^2}{r^2\ln^4(S)}\right] \tag{51}$$

$$\frac{d^2n}{dS^2} = n\left[\frac{(\ln r - \ln r_{\mathrm{m}})^4}{S^2\ln^6(S)} - 5\frac{(\ln r - \ln r_{\mathrm{m}})^2}{S^2\ln^4(S)} - \frac{(\ln r - \ln r_{\mathrm{m}})^2}{S^2\ln^3(S)} + \frac{2}{S^2\ln^2(S)} + \frac{1}{S^2\ln(S)}\right] \tag{52}$$

2.2.4 Moments

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

$$m_i = N_0\exp\left(i\mu + \frac{i^2\sigma^2}{2}\right). \tag{53}$$

The first three moments are

$$
\begin{aligned}
m_1 &= \int_0^\infty r n(r)\,dr = N_0 \exp\left(\mu + \frac{1}{2}\sigma^2\right) = N_0 r_\mathrm{m} \exp\left(\frac{1}{2}\sigma^2\right) \equiv N_0 r_\mathrm{m} \exp\left(\frac{1}{2}\ln^2 S\right) \\
m_2 &= \int_0^\infty r^2 n(r)\,dr = N_0 \exp\left(2\mu + 2\sigma^2\right) = N_0 r_\mathrm{m}^2 \exp\left(2\sigma^2\right) \equiv N_0 r_\mathrm{m}^2 \exp\left(2\ln^2 S\right) \\
m_3 &= \int_0^\infty r^3 n(r)\,dr = N_0 \exp\left(3\mu + \frac{9}{2}\sigma^2\right) = N_0 r_\mathrm{m}^3 \exp\left(\frac{9}{2}\sigma^2\right) \equiv N_0 r_\mathrm{m}^3 \exp\left(\frac{9}{2}\ln^2 S\right)
\end{aligned}
$$

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

$$
\begin{aligned}
\text{mean} &= \frac{1}{N_0}\int_0^\infty r n(r)\,dr = \frac{1}{N_0} m_1 = \frac{1}{N_0} N_0 \exp\left(\mu + \frac{1}{2}\sigma^2\right), && (54) \\
&= r_\mathrm{m} \exp\left(\frac{1}{2}\sigma^2\right) \equiv r_\mathrm{m} \exp\left(\frac{1}{2}\ln^2 S\right), && (55)
\end{aligned}
$$

$$
\begin{aligned}
\text{area} &= \int_0^\infty 4\pi r^2 n(r)\,dr = 4\pi m_2 = 4\pi N_0 \exp\left(2\mu + 2\sigma^2\right), && (56) \\
&= 4\pi N_0 r_\mathrm{m}^2 \exp\left(2\sigma^2\right) \equiv 4\pi N_0 r_\mathrm{m}^2 \exp\left(2\ln^2 S\right), && (57)
\end{aligned}
$$

and

$$
\begin{aligned}
\text{volume} &= \int_0^\infty \frac{4}{3}\pi r^3 n(r) dr = \frac{4}{3}\pi m_3 = \frac{4}{3}\pi N_0 \exp\left(3\mu + \frac{9}{2}\sigma^2\right), && (58) \\
&= \frac{4}{3}\pi N_0 r_\mathrm{m}^3 \exp\left(\frac{9}{2}\sigma^2\right) \equiv \frac{4}{3}\pi N_0 r_\mathrm{m}^3 \exp\left(\frac{9}{2}\ln^2 S\right). && (59)
\end{aligned}
$$

For a lognormal distribution the effective radius is

$$
\begin{aligned}
r_\mathrm{e} &= \frac{m_3}{m_2} = \frac{\exp\left(3\mu + \frac{9}{2}\sigma^2\right)}{\exp\left(2\mu + \frac{4}{2}\sigma^2\right)} = \exp\left(\mu + \frac{5}{2}\sigma^2\right), && (60) \\
&= r_m \exp\left(\frac{5}{2}\sigma^2\right) \equiv r_m \exp\left(\frac{5}{2}\ln^2 S\right). && (61)
\end{aligned}
$$

2.3 Log-Normal Distributions of Area and Volume

The log-normal area density distribution is

$$
a(r) = \frac{A_0}{\sqrt{2\pi}}\frac{1}{\sigma_a}\frac{1}{r}\exp\left[-\frac{(\ln(r) - \mu_a)^2}{2\sigma_a^2}\right] \qquad (62)
$$

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. These constants can all be related to the description of a log-normal number density distribution starting from

$$a(r) \quad = \quad 4\pi r^2 n(r) = 4\pi r^2 \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu)^2}{2\sigma^2}\right] \tag{63}$$

Making the substitution $r^2 = \exp(2\ln r)$ and completing the square gives

$$a(r) \quad = \quad 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[2\mu + 2\sigma^2\right] \exp\left[-\frac{\left(\ln r - (\mu + 2\sigma^2)\right)^2}{2\sigma^2}\right] \tag{64}$$

Equating with Equations 62 and 63 gives

$$\begin{aligned} & \frac{A_0}{\sqrt{2\pi}} \frac{1}{\sigma_a} \frac{1}{r} \exp\left[-\frac{(\ln(r) - \mu_a)^2}{2\sigma_a^2}\right] \\ = \quad & 4\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[2\mu + 2\sigma^2\right] \exp\left[-\frac{\left(\ln r - (\mu + 2\sigma^2)\right)^2}{2\sigma^2}\right] \end{aligned} \tag{65}$$

which is true if

$$\frac{A_0}{\sigma_a} = \frac{4\pi N_0}{\sigma} \exp\left[2\mu + 2\sigma^2\right] \tag{66}$$

and

$$\frac{(\ln(r) - \mu_a)^2}{2\sigma_a^2} \quad = \quad \frac{\left(\ln r - (\mu + 2\sigma^2)\right)^2}{2\sigma^2} \tag{67}$$

From Equation 56

$$A_0 \quad = \quad 4\pi N_0 \exp\left(2\mu + 2\sigma^2\right) \tag{68}$$

which gives $\sigma_a = \sigma$ when inserted into Equation 66. This shows that is 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 67 gives

$$\mu_a = \mu + 2\sigma^2. \tag{69}$$

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

$$v(r) \quad = \quad \frac{V_0}{\sqrt{2\pi}} \frac{1}{\sigma_v} \frac{1}{r} \exp\left[-\frac{(\ln(r) - \mu_v)^2}{2\sigma_v^2}\right] \tag{70}$$

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. These constants can all be related to the description of a log-normal number density distribution starting from

$$v(r) \quad = \quad \frac{4}{3}\pi r^3 n(r) = \frac{4}{3}\pi r^3 \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu)^2}{2\sigma^2}\right] \tag{71}$$

Making the substitution $r^3 = \exp(3 \ln r)$ and completing the square gives

$$v(r) \quad = \quad \frac{4}{3}\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[-\frac{(\ln r - \mu)^2 - 6\sigma^2 \ln r}{2\sigma^2}\right] \tag{72}$$

Complete the square

$$v(r) \quad = \quad \frac{4}{3}\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[3\mu + 4.5\sigma^2\right] \exp\left[-\frac{\left(\ln r - (\mu + 3\sigma^2)\right)^2}{2\sigma^2}\right] \tag{73}$$

Equating with Equations 70 and 73 gives

$$\begin{aligned} & \frac{V_0}{\sqrt{2\pi}} \frac{1}{\sigma_v} \frac{1}{r} \exp\left[-\frac{(\ln(r) - \mu_v)^2}{2\sigma_v^2}\right] \\ = \quad & \frac{4}{3}\pi \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[3\mu + 4.5\sigma^2\right] \exp\left[-\frac{\left(\ln r - (\mu + 3\sigma^2)\right)^2}{2\sigma^2}\right] \end{aligned} \tag{74}$$

which is true if

$$\frac{V_0}{\sigma_v} = \frac{4}{3}\pi \frac{N_0}{\sigma} \exp\left[3\mu + 4.5\sigma^2\right] \tag{75}$$

and

$$\frac{(\ln(r) - \mu_v)^2}{2\sigma_v^2} \quad = \quad \frac{\left(\ln r - (\mu + 3\sigma^2)\right)^2}{2\sigma^2} \tag{76}$$

From Equation 58

$$V_0 \quad = \quad \frac{4}{3}\pi N_0 \exp\left(3\mu + 4.5\sigma^2\right) \tag{77}$$

which gives $\sigma_v = \sigma$ when inserted into Equation 75. 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 76 gives

$$\mu_v = \mu + 3\sigma^2. \tag{78}$$

They relationships area and volume density log-normal parameters and the number size distribution parameters are summarised in Table 1.

Lastly Table 2 shows derived values for a log-normal distributions over a range of r_{m} and with $S = 1.5$.

Table 1: Relationships area and volume density log-normal parameters and the number size distribution parameters, N_0, μ and σ.

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

Table 2: Values derived for a log-normal number density size distribution with $S = 1.5$.

Median Radius μm	**Mean Radius** μm	**Effective Radius** μm	**Area Median Radius** μm	**Volume Median Radius** μm
0.1	0.1	0.3	0.3	0.4
0.2	0.3	0.7	0.5	0.8
0.5	0.6	1.7	1.3	2.1
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
20	25	67	52	85
50	64	166	131	211
100	127	332	261	423

2.4 A Different Basis State

While the shape of the lognormal distribution is controlled its median radius, r_{m}, and its geometric standard deviation, S. An alternative is the effective radius, r_{e}, and volume per particle, $v = V/N_0$. If these parameters are adopted r_{m} and S can be recovered as follows:

$$S = \exp\left[\sqrt{-\frac{\ln\left(\frac{3v}{4\pi r_{\mathrm{e}}^3}\right)}{3}}\right] \tag{79}$$

$$r_{\mathrm{m}} = \left(\frac{3v}{4\pi}\right)^{(5/6)} \frac{1}{r_{\mathrm{e}}^{(3/2)}} \tag{80}$$

3 Other Distributions

3.1 Gamma Distribution

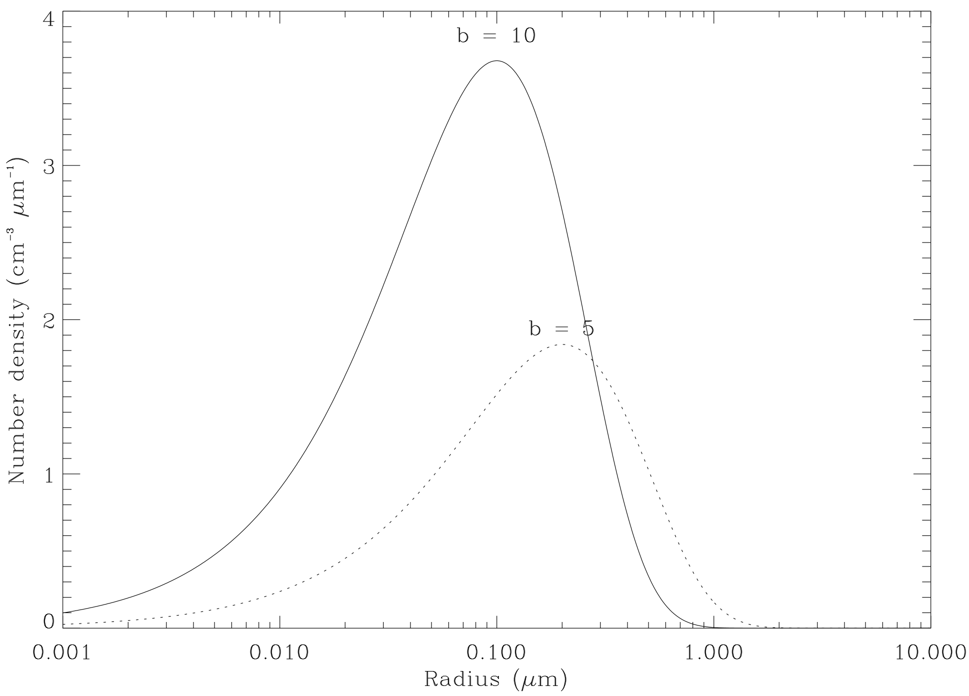


Figure 2: Normalised gamma distribution.

The gamma distribution is given by Twomey (1977) as

$$n(r) = ar\exp(-br), \tag{81}$$

where a and b are positive constants. The mode of the distribution occurs where $r = b^{-1}$ and it falls off slowly on the small radius side and exponentially on the large radius side. The i-th moment of the gamma distribution is given by

$$\begin{aligned} m_i &= ab^{-2-i}\Gamma(2+i), &(82)\\ &= ab^{-2-i}(1+i)!. &(83)\end{aligned}$$

If the constant a is used to denote the total number density then the normalised distribution (see Figure 2) can be expressed

$$n(r) = ab^2 r\exp(-br), \tag{84}$$

which has moments defined by

$$m_i = ab^{-i}(1+i)!. \tag{85}$$

3.2 Modified Gamma Distribution

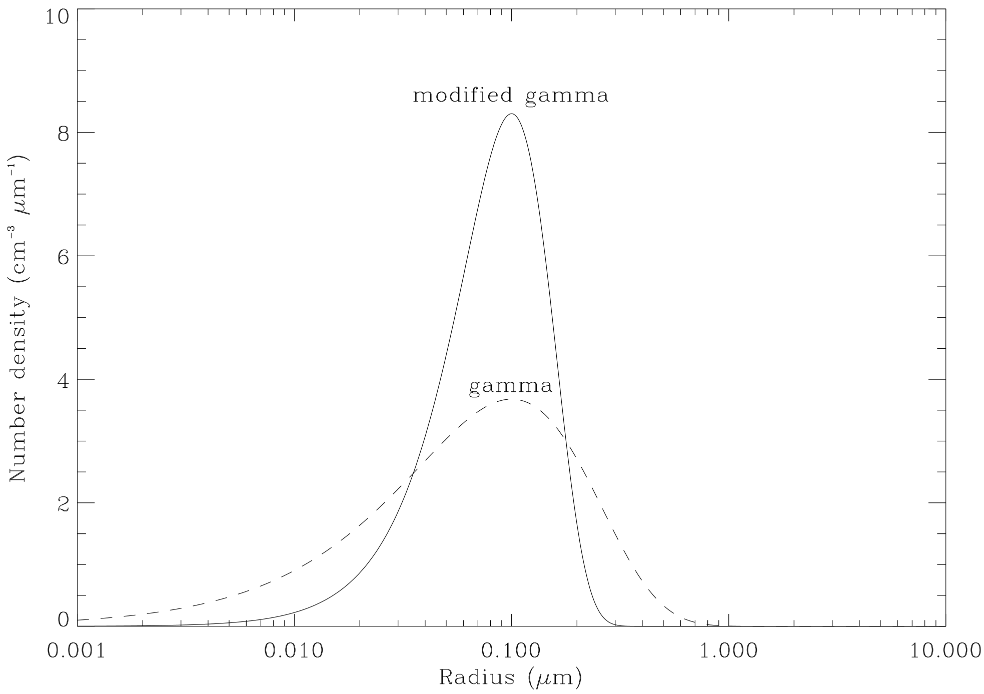


Figure 3: Normalised modified gamma distribution ($\alpha = 2, b = 100, \gamma = 2$) and gamma distribution ($b = 10$).

The modified gamma distribution is given by Deirmendjian (1969) as

$$n(r) = ar^{\alpha}\exp\left(-br^{\gamma}\right). \tag{86}$$

The four constants a, α, b, γ are positive and real and α is an integer. The mode of the distribution occurs where $r = \left(\frac{\alpha}{b\gamma}\right)^{1/\gamma}$ and the moments of this distribution are[2]

$$m_i = \frac{a}{\gamma}b^{-\frac{\alpha+1+i}{\gamma}}\Gamma\left(\frac{\alpha+1+i}{\gamma}\right). \tag{87}$$

[2]See $3 \cdot 478/1$ of Gradshteyn and Ryzhik (1994).

Hence the integral of the modified gamma distribution is

$$\int_0^\infty n(r)\,dr = \frac{a}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right). \tag{88}$$

In order that the first parameter, a, is the total aerosol concentration it is convenient to define the normalized modified gamma distribution (see Figure 3) as

$$n(r) = a \frac{r^\alpha \exp\left(-br^\gamma\right)}{\frac{1}{\gamma} b^{-\frac{\alpha+1}{\gamma}} \Gamma\left(\frac{\alpha+1}{\gamma}\right)}, \tag{89}$$

where a, α, b, γ are positive and real but α is no longer constrained to be an integer. The moments of this distribution are given by

$$m_i = ab^{-\frac{i}{\gamma}} \frac{\Gamma\left(\frac{\alpha+i+1}{\gamma}\right)}{\Gamma\left(\frac{\alpha+1}{\gamma}\right)}. \tag{90}$$

3.3 Inverse Modified Gamma Distribution

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

$$n(r) = ar^{-\alpha} \exp\left(-br^{-\gamma}\right). \tag{91}$$

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 moments are defined by

$$m_i = \frac{a}{\gamma} b^{-\frac{\alpha-1-i}{\gamma}} \Gamma\left(\frac{\alpha-1-i}{\gamma}\right). \tag{92}$$

The normalized inverse modified gamma distribution can be defined

$$n(r) = a \frac{r^{-\alpha} \exp\left(-br^{-\gamma}\right)}{\frac{1}{\gamma} b^{-\frac{\alpha-1}{\gamma}} \Gamma\left(\frac{\alpha-1}{\gamma}\right)}. \tag{93}$$

The moments of this distribution are given by

$$m_i = ab^{\frac{i}{\gamma}} \frac{\Gamma\left(\frac{\alpha-1-i}{\gamma}\right)}{\Gamma\left(\frac{\alpha-1}{\gamma}\right)}. \tag{94}$$

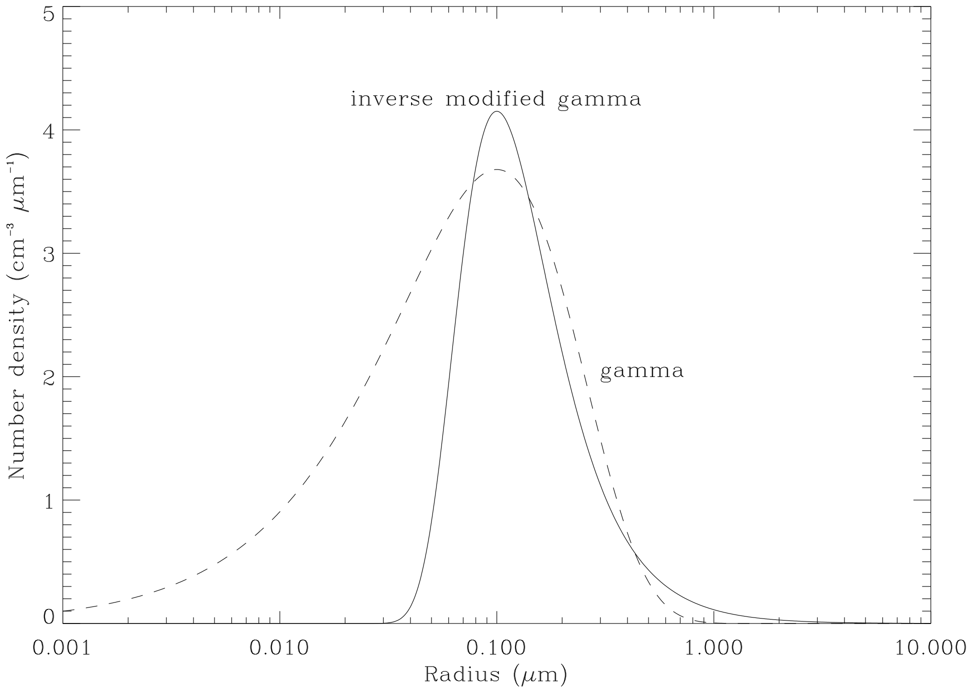


Figure 4: Normalised inverse modified gamma distribution ($\alpha = 2, b = 0.01, \gamma = 2$) and gamma distribution($b - 10$).

3.4 Regularized Power Law

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

$$n(r) \quad = \quad ab^{\alpha-2} \frac{r^{\alpha-1}}{\left[1 + \left(\frac{r}{b}\right)^{\alpha}\right]^{\gamma}}, \tag{95}$$

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

$$r \quad = \quad b\left(\frac{\alpha - 1}{1 + \alpha(\gamma - 1)}\right)^{1/\alpha}, \tag{96}$$

and the moments by

$$m_i \quad = \quad a\frac{b^i}{\alpha}\frac{\Gamma(1 + i/\alpha)\Gamma(\gamma - 1 - i/\alpha)}{\Gamma(\gamma)}. \tag{97}$$

Hence the normalised distribution is

$$n(r) = a\alpha\gamma b^{\alpha-2} \frac{r^{\alpha-1}}{\left[1+\left(\frac{r}{b}\right)^{\alpha}\right]^{\gamma}}. \tag{98}$$

4 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_e. How do we do this?

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

$$r_\mathrm{e,i} = r_{m,i} \exp\left(\frac{5}{2}\ln^2 S_i\right). \tag{99}$$

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

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

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

$$\mu_1 = \frac{r_\mathrm{e} - r_\mathrm{e,1}}{r_\mathrm{e,3} - r_\mathrm{e,1}} \tag{100}$$

We now have two equations

$$\mu_1 + \mu_2 + \mu_3 = 1 \tag{101}$$

$$\frac{\mu_1 r_{m,1}^3 \exp\left(\frac{9}{2}\ln^2 S_1\right) + \mu_2 r_{m,2}^3 \exp\left(\frac{9}{2}\ln^2 S_2\right) + \mu_3 r_{m,3}^3 \exp\left(\frac{9}{2}\ln^2 S_3\right)}{\mu_1 r_{m,1}^2 \exp\left(2\ln^2 S_1\right) + \mu_2 r_{m,2}^2 \exp\left(2\ln^2 S_2\right) + \mu_3 r_{m,3}^2 \exp\left(2\ln^2 S_3\right)} = r_e \tag{102}$$

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

$$\frac{A\mu_1 + B\mu_2 + C\mu_3}{D\mu_1 + E\mu_2 + F\mu_3} = r_\mathrm{e} \tag{103}$$

and the two equations solved to give

$$\mu_2 = \frac{r_e E - B - \mu_1(A - B + r_e(E - D))}{C - B + r_e(E - F)} \tag{104}$$

$$\mu_3 = \frac{r_e F - C - \mu_1(A - C + r_e(F - D))}{B - C + r_e(F - E)} \tag{105}$$

5 Optical Properties

5.1 Volume Absorption, Scattering and Extinction Coefficients

The volume absorption coefficient, $\beta^{\rm abs}(\lambda, r)$, the volume scattering coefficient, $\beta^{\rm sca}(\lambda, r)$, and the volume extinction coefficient, $\beta^{\rm ext}(\lambda, r)$, 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

$$\text{monodisperse only} \quad \left\{ \begin{array}{l} \beta^{\rm abs}(\lambda, r) = \sigma^{\rm abs}(\lambda, r)N(r) = \pi r^2 Q^{\rm abs}(\lambda, r)N(r), \\ \beta^{\rm sca}(\lambda, r) = \sigma^{\rm sca}(\lambda, r)N(r) = \pi r^2 Q^{\rm sca}(\lambda, r)N(r), \\ \beta^{\rm ext}(\lambda, r) = \sigma^{\rm ext}(\lambda, r)N(r) = \pi r^2 Q^{\rm ext}(\lambda, r)N(r), \end{array} \right. \tag{106}$$

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

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

$$\beta^{\rm abs}(\lambda) = \int_0^\infty \sigma^{\rm abs}(\lambda, r)n(r)\,dr = \int_0^\infty \pi r^2 Q^{\rm abs}(\lambda, r)n(r)\,dr, \tag{107}$$

$$\beta^{\rm sca}(\lambda) = \int_0^\infty \sigma^{\rm sca}(\lambda, r)n(r)\,dr = \int_0^\infty \pi r^2 Q^{\rm sca}(\lambda, r)n(r)\,dr, \tag{108}$$

$$\beta^{\rm ext}(\lambda) = \int_0^\infty \sigma^{\rm ext}(\lambda, r)n(r)\,dr = \int_0^\infty \pi r^2 Q^{\rm ext}(\lambda, r)n(r)\,dr, \tag{109}$$

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.

$$\bar{\sigma}^{\rm abs}(\lambda) = \int_0^\infty \sigma^{\rm abs} n(r)\,dr \Big/ \int_0^\infty n(r)\,dr = \frac{\beta^{\rm abs}(\lambda)}{N_0}, \tag{110}$$

$$\bar{\sigma}^{\rm sca}(\lambda) = \int_0^\infty \sigma^{\rm sca} n(r)\,dr \Big/ \int_0^\infty n(r)\,dr = \frac{\beta^{\rm sca}(\lambda)}{N_0}, \tag{111}$$

$$\bar{\sigma}^{\rm ext}(\lambda) = \int_0^\infty \sigma^{\rm ext} n(r)\,dr \Big/ \int_0^\infty n(r)\,dr = \frac{\beta^{\rm ext}(\lambda)}{N_0}, \tag{112}$$

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

5.2 Back Scatter

■ to be done ■

5.3 Phase Function

The phase function represents the redistribution of the scattered energy.

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

$$P(\lambda,\theta) \quad = \quad \frac{1}{\beta^{\mathrm{sca}}}\int_0^\infty \pi r^2 Q^{\mathrm{sca}}(\lambda,r)P(\lambda,r,\theta)n(r)\,dr. \tag{113}$$

5.4 Single Scatter Albedo

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

$$\omega(\lambda) \quad = \quad \frac{\beta^{\mathrm{sca}}(\lambda)}{\beta^{\mathrm{ext}}(\lambda)}. \tag{114}$$

5.5 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.

$$g \quad = \quad \frac{1}{\beta^{\mathrm{sca}}}\int_0^\infty \pi r^2 Q^{\mathrm{sca}}(\lambda,r)g(\lambda,r)n(r)\,dr \tag{115}$$

5.6 Integration

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

$$\int_{r_{\mathrm{l}}}^{r_{\mathrm{u}}} f(r)n(r)\,dr \quad = \quad \sum_{i=1}^{n} w_i f(r_i) \tag{116}$$

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

For a log normal size distribution the integrals are

$$\beta^{\mathrm{ext}}(\lambda) \quad = \quad \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\int_{r_{\mathrm{l}}}^{r_{\mathrm{u}}} rQ^{\mathrm{ext}}(\lambda,r)\exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_{\mathrm{m}}}{\sigma}\right)^2\right]dr \tag{117}$$

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

where

$$
w_i' = \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}} r_i \exp\left[-\frac{1}{2}\left(\frac{\ln r_i - \ln r_{\mathrm{m}}}{\sigma}\right)^2\right] w_i \tag{118}
$$

5.7 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 117) with respect to the size distribution parameters (N_0, r_{m}, σ).

$$
\frac{\partial \beta^{\mathrm{ext}}}{\partial N_0} = \frac{1}{\sigma}\sqrt{\frac{\pi}{2}}\int_0^{\infty} r Q^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr, \tag{119}
$$

$$
\frac{\partial \beta^{\mathrm{ext}}}{\partial r_{\mathrm{m}}} = \frac{N_0}{r_{\mathrm{m}}\sigma^3}\sqrt{\frac{\pi}{2}}\int_0^{\infty} (\ln r - \ln r_{\mathrm{m}}) r Q^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr, \tag{120}
$$

$$\begin{aligned}
\frac{\partial \beta^{\mathrm{ext}}}{\partial \sigma} &= -\frac{N_0}{\sigma^2}\sqrt{\frac{\pi}{2}}\int_0^\infty rQ^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr \\
&\quad + \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\int_0^\infty \frac{(\ln r - \ln r_{\mathrm{m}})^2}{\sigma^3} rQ^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr, \\
&= \frac{N_0}{\sigma^2}\sqrt{\frac{\pi}{2}}\int_0^\infty \left[\frac{(\ln r - \ln r_{\mathrm{m}})^2}{\sigma^2} - 1\right] rQ^{\mathrm{ext}}(\lambda, r)\exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr.
\end{aligned} \tag{121}$$

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_{m} and σ to positive quantities it is better to retrieve l_{m} $(= \ln r_{\mathrm{m}})$ and G $(= \ln \sigma)$. In terms of these new variables volume extinction coefficient for a log normal size distribution is

$$\beta^{\mathrm{ext}}(\lambda) = \frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l} Q^{\mathrm{ext}}(l, \lambda)\exp\left[-\frac{(l - l_{\mathrm{m}})^2}{2\exp(2G)}\right] dl. \tag{122}$$

The partial derivatives of the transformed parameters (Equation 122) are

$$\frac{\partial \beta^{\mathrm{ext}}}{\partial T} = \frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l} Q^{\mathrm{ext}}(l, \lambda)\exp\left[-\frac{(l - l_{\mathrm{m}})^2}{2\exp(2G)}\right] dl, \tag{123}$$

$$\frac{\partial \beta^{\mathrm{ext}}}{\partial l_{\mathrm{m}}} = \frac{\exp T}{\exp(3G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty (l - l_{\mathrm{m}}) e^{2l} Q^{\mathrm{ext}}(l, \lambda)\exp\left[-\frac{(l - l_{\mathrm{m}})^2}{2\exp(2G)}\right] dl, \tag{124}$$

$$\begin{aligned}
\frac{\partial \beta^{\mathrm{ext}}}{\partial G} &= -\frac{\exp T}{\exp(G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l} Q^{\mathrm{ext}}(l, \lambda)\exp\left[-\frac{(l - l_{\mathrm{m}})^2}{2\exp(2G)}\right] dl \\
&\quad + \frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty \frac{(l - l_{\mathrm{m}})^2}{\exp(2G)} e^{2l} Q^{\mathrm{ext}}(l, \lambda)\exp\left[-\frac{(l - l_{\mathrm{m}})^2}{2\exp(2G)}\right] dl, \\
&= \frac{\exp T}{\exp(G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty \left[\frac{(l - l_{\mathrm{m}})^2}{\exp(2G)} - 1\right] e^{2l} Q^{\mathrm{ext}}(l, \lambda)\exp\left[-\frac{(l - l_{\mathrm{m}})^2}{2\exp(2G)}\right] dl.
\end{aligned} \tag{125}$$

5.8 Cloud Liquid Water Path

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

$$l = \rho \int_0^\infty \frac{4}{3}\pi r^3 n(r)\, dr \times z \tag{126}$$

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

$$\tau = \beta^{\mathrm{ext}} \times z \tag{127}$$

so

$$l = \rho\tau \frac{\int_0^\infty \frac{4}{3}\pi r^3 n(r)\, dr}{\beta^{\mathrm{ext}}} \tag{128}$$

$$= \frac{4}{3}\pi\rho\tau \frac{\int_0^\infty r^3 n(r)\, dr}{\int_0^\infty \pi r^2 Q^{\mathrm{ext}}(\lambda, r) n(r)\, dr}. \tag{129}$$

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

$$l = \frac{4}{6}\rho\tau \frac{\int_0^\infty r^3 n(r)\, dr}{\int_0^\infty r^2 n(r)\, dr} = \frac{2}{3}\rho\tau r_e \tag{130}$$

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

$$l = \frac{2}{3} \times 1 \times 10 \times 15 \text{ g cm}^{-3}\mu\text{m} = 100 \text{ g m}^{-2} \tag{131}$$

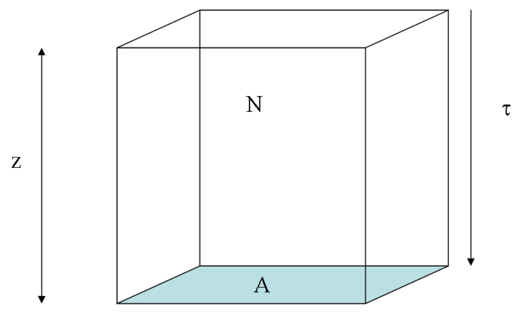
While for an ice cloud (-45 °C, $\rho = 0.920$ g cm^{-3}) of $\tau = 1$, $r_e = 25\,\mu$m we get

$$l = \frac{2}{3} \times 0.92 \times 1 \times 25 \text{ g cm}^{-3}\mu\text{m} = 15 \text{ g m}^{-2} \tag{132}$$

.

5.9 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

$$M = \rho \times v \times N \times A \times Z \tag{133}$$

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.

$$m = \rho \times v \times N \times Z \tag{134}$$

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

$$\tau = \beta^{\text{ext}} \times Z \tag{135}$$

so that

$$m = \frac{\rho \times v \times N \times \tau}{\beta^{\text{ext}}} = \frac{\rho \times v \times \tau}{\bar{\sigma}^{\text{ext}}} \tag{136}$$

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

$$v = \frac{\int_0^\infty \frac{4}{3}\pi r^3 n(r)\, dr}{N} \tag{137}$$

so that the mass per unit area is given by

$$m = \frac{\rho\tau}{\beta^{\text{ext}}} \int_0^\infty \frac{4}{3}\pi r^3 n(r)\, dr \tag{138}$$

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

In terms of r_{e} the mass per unit area is given by

$$m = \frac{4\pi\rho\tau}{3\beta^{\text{ext}}} \int_0^\infty r^3 n(r)\, dr \times \frac{\int_0^\infty r^2 n(r)\, dr}{\int_0^\infty r^2 n(r)\, dr} = \frac{4\rho\tau}{3\tilde{Q}^{\text{ext}}} r_{\text{e}} \tag{139}$$

Substance	**Density (g cm^{-3})**	**Reference**
Ice	0.92	
Volcanic ash	2.42±0.79	Bayhurst, Wohletz and Mason (1994)
Water	1	

Table 3: Density of some materials that form aerosols.

which uses an area weighted extinction efficiency

$$\tilde{Q}^{\text{ext}} = \frac{\beta^{\text{ext}}}{\pi \int_0^\infty r^2 n(r)\, dr} = \frac{\int_0^\infty \sigma^{\text{ext}} n(r)\, dr}{\int_0^\infty \pi r^2 n(r)\, dr} = \frac{\int_0^\infty \pi r^2 Q^{\text{ext}} n(r)\, dr}{\int_0^\infty \pi r^2 n(r)\, dr} \tag{140}$$

If the size distribution is much larger than the wavelength then $Q^{\text{ext}} \to 2$ and $\tilde{Q}^{\text{ext}} \approx 2$ and Equation 130 becomes identical to Equation 139.

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 138 can be completed analytically i.e.

$$m = \frac{\rho\tau}{\beta^{\text{ext}}} \frac{4}{3} \pi N_0 r_{\text{m}}^3 \exp\left(\frac{9}{2}\sigma^2\right) \tag{141}$$

Typically ρ is in g 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

$$\frac{\frac{\text{g}}{\text{cm}^3} \frac{1}{\text{cm}^3} \mu\text{m}^3}{\frac{1}{\text{km}}} = \frac{\text{g}}{10^{-6}\,\text{m}^3} \frac{1}{10^{-6}\,\text{m}^3} 10^{-18}\,\text{m}^3 10^3\,\text{m} = 10^{-3}\,\text{g m}^{-2} \tag{142}$$

Table 3 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

$$r_{\text{e}} = r_m \exp\left(\frac{5}{2}\sigma^2\right), \tag{143}$$

to get

$$m = \frac{4\rho\tau\pi N_0}{3\beta^{\text{ext}}} r_{\text{e}}^3 \exp\left(-\frac{15}{2}\sigma^2\right) \exp\left(\frac{9}{2}\sigma^2\right) = \frac{4\rho\tau\pi N_0}{3\beta^{\text{ext}}} r_{\text{e}}^3 \exp\left(-3\sigma^2\right). \tag{144}$$

Equating this expression to Equation 130 gives

$$\tilde{Q}^{\text{ext}} = \frac{\beta^{\text{ext}}}{\pi N_0 r_{\text{e}}^2 \exp\left(-3\sigma^2\right)}. \tag{145}$$

which is true for a log-normal distribution.

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

$$m = \frac{\tau \times \rho \times N \times v}{\beta^{\mathrm{ext}}} = \frac{\tau \sum_{i=1}^{n} \rho_i N_i v_i}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\mathrm{ext}}} \tag{146}$$

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

$$v_i = \frac{4}{3}\pi r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right) \tag{147}$$

Hence

$$m = \tau \frac{\sum_{i=1}^{n} \rho_i N_i \frac{4}{3}\pi r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right)}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\mathrm{ext}}} \tag{148}$$

$$= \frac{4}{3}\pi\tau \frac{\sum_{i=1}^{n} \rho_i N_i r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right)}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\mathrm{ext}}} \tag{149}$$

References

Bayhurst, G. K., Wohletz, K. H. and Mason, A. S. (1994). A method for characterizing volcanic ash from the December 15, 1989 eruption of Redoubt volcano, Alaska, *in* T. J. Casadevall (ed.), *Volcanic Ash and Aviation Safety, Proc. 1st Int. Symp. on Volcanic Ash and Aviation Safety*, USGS Bulletin 2047, Washington, pp. 13–17.

Deepak, A. (ed.) (1982). *Atmospheric Aerosols*, Spectrum Press, Hampton, Virginia.

Deirmendjian, D. (1969). *Electromagnetic scattering on spherical polydispersions*, Elsevier, New York.

Gradshteyn, I. S. and Ryzhik, I. M. (1994). *Table of integrals, series, and products*, Academic Press, London.

Twomey, S. (1977). *Atmospheric aerosols*, Elsevier, Amsterdam, The Netherlands.

Index