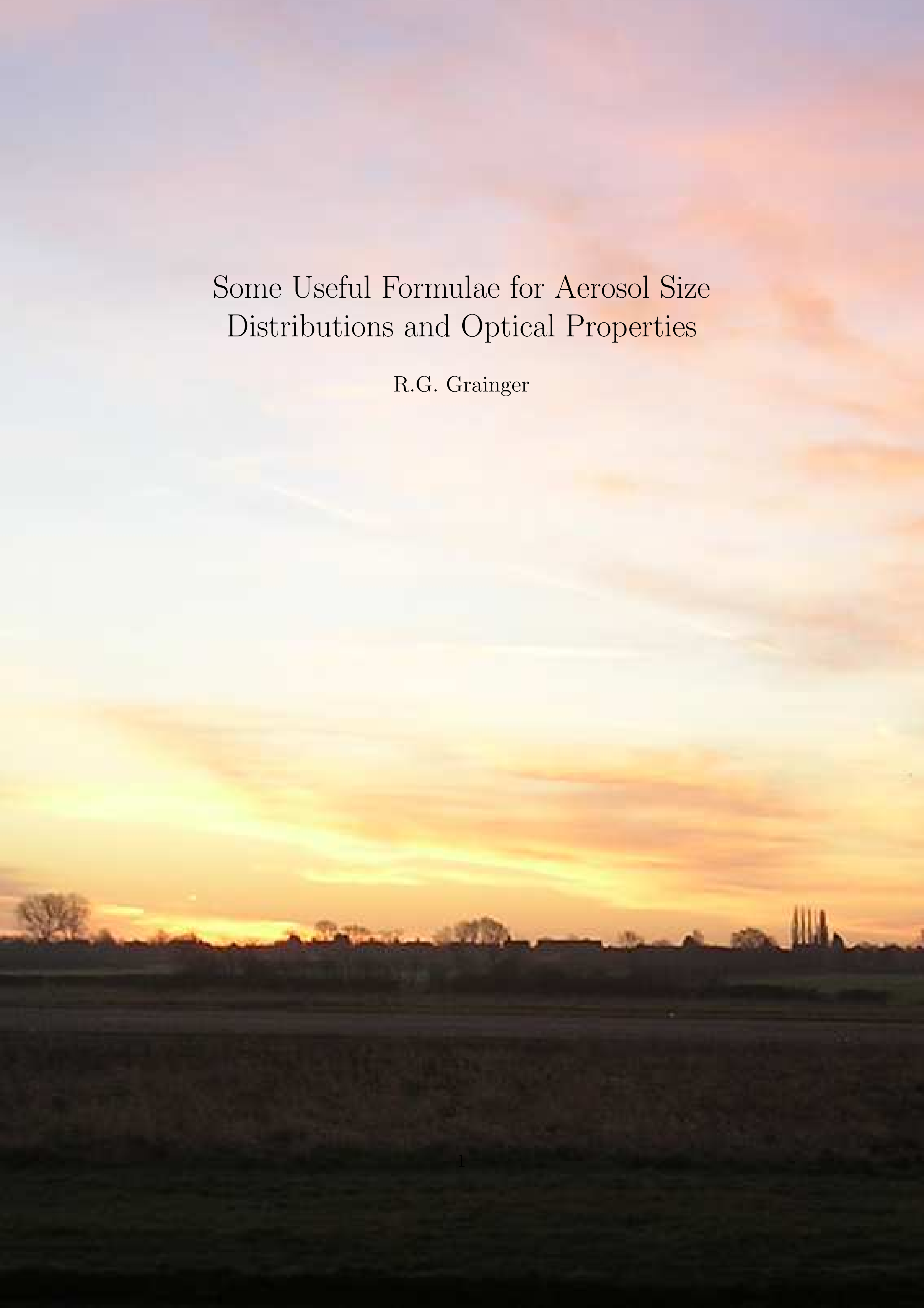

Some Useful Formulae for Aerosol Size Distributions and Optical Properties

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Contents

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

1 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 (less than 100 μm ■ check ■) 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 Distributions

The distribution of particle sizes can be represented by a differential radius 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 Normal Distribution

One particle distribution to consider adopting is the normal distribution

$$n(r) = \frac{N_0}{\sqrt{2\pi}}\frac{1}{\sigma_0}\exp\left[-\frac{(r-\mu_0)^2}{2\sigma_0^2}\right] \tag{4}$$

where the mean, μ_0, and the standard deviation, σ_0, are defined

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

$$= \frac{1}{N_0}\int_0^\infty r n(r)\,dr \tag{6}$$

$$\sigma_0^2 = \frac{1}{N_0} \int_0^{\infty} (r - \mu_0)^2 n(r)\, dr \tag{7}$$

1.3 Logarithmic Normal 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 Normal distribution is that it allows negative radii. In fact aerosol distributions are much better represented by a normal distribution of the logarithm of the particle radii. Letting $l = \ln(r)$ we have

$$n_l(l) = \frac{dN(l)}{dl} = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \exp\left[-\frac{(l-\mu)^2}{2\sigma^2}\right] \tag{8}$$

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

$$\mu = \frac{\int_{-\infty}^{\infty} l n_l(l)\, dl}{\int_{-\infty}^{\infty} n_l(l)\, dl} \tag{9}$$

$$= \frac{1}{N_0} \int_{-\infty}^{\infty} l n_l(l)\, dl \tag{10}$$

$$\sigma^2 = \frac{1}{N_0} \int_{-\infty}^{\infty} (l-\mu)^2 n_l(l)\, dl \tag{11}$$

Noting that

$$\frac{dl}{dr} = \frac{1}{r} \tag{12}$$

then in terms of radius rather than log radius we have

$$n(r) = \frac{dN(r)}{dr} \tag{13}$$

$$= \frac{dN(l)}{dl} \frac{dl}{dr} \tag{14}$$

$$= n_l(l) \frac{dl}{dr} \tag{15}$$

$$= \frac{N_0}{\sqrt{2\pi}} \frac{1}{\sigma} \frac{1}{r} \exp\left[-\frac{(\ln(r)-\mu)^2}{2\sigma^2}\right] \tag{16}$$

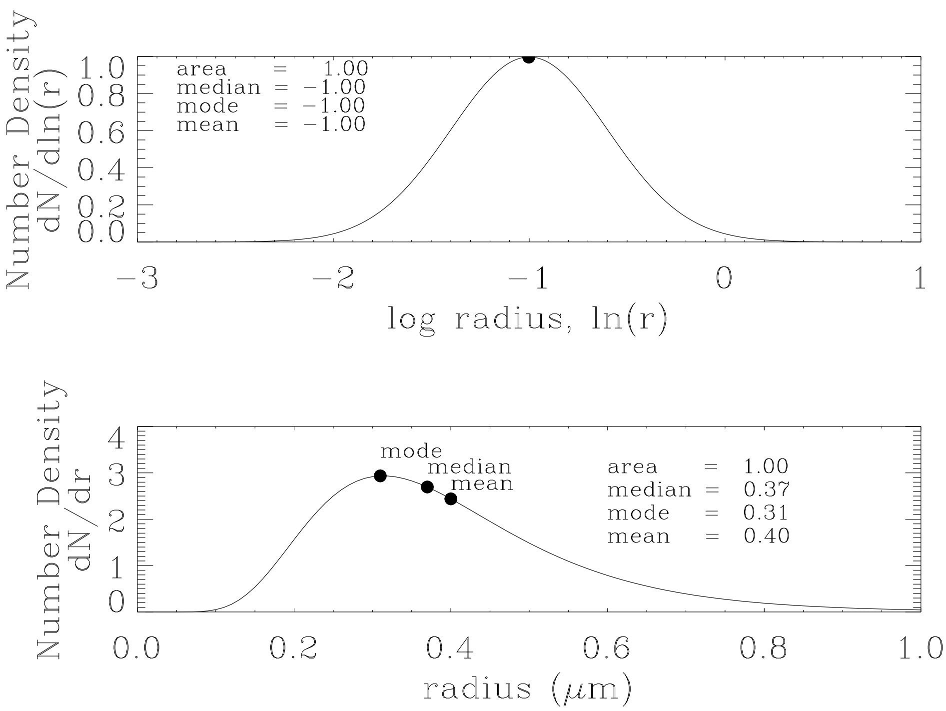


Figure 1: Log-normal distribution with parameters $N_0 = 1$, $\mu = -1$ and $\sigma = 0.4$ plotted in log space (top panel) and linear space (bottom panel). Indicated are the distribution mode, median and mean in log and linear space respectively.

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.

$$r_{\mathrm{g}} = (r_1 \times r_2 \times \ldots \times r_n)^{\frac{1}{n}} \tag{17}$$

$$\Rightarrow \ln(r_{\mathrm{g}}) = \ln (r_1 \times r_2 \times \ldots \times r_n)^{\frac{1}{n}} \tag{18}$$

$$= \frac{\ln(r_1) + \ln(r_2) + \ldots + \ln(r_n)}{n} \tag{19}$$

which has the continuous form

$$\ln(r_{\mathrm{g}}) = \frac{1}{N_0} \int_{-\infty}^{\infty} l n_l(l)\, dl \tag{20}$$

$$= \mu \tag{21}$$
$$= \ln(r_{\mathrm{m}}) \tag{22}$$
$$\Leftrightarrow r_{\mathrm{g}} = r_{\mathrm{m}} \tag{23}$$

It is also common to express the spread of the distribution using the geometric standard deviation, S. The geometric standard deviation is related to the standard deviation of $\ln r$ by $\sigma = \ln(S)$. From this definition S must be greater or equal to one otherwise the log-normal standard deviation is negative [redacted] see S&P for a little more on how many particles lie within one geometric standard deviation [redacted] . When S is one the distribution is monodisperse. Typical aerosol distributions have S values of about 1.5.

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

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln(S)} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2(S)}\right] \tag{24}$$

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

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

$$n(r) = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln(S)} \frac{1}{r} \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2(S)}\right] = \frac{A}{r}\exp[B] \tag{25}$$

$$\text{where} \quad A = \frac{N_0}{\sqrt{2\pi}} \frac{1}{\ln(S)} \tag{26}$$

$$B = -\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\ln^2(S)} \tag{27}$$

$$\text{and} \quad \frac{dB}{dr} = -\frac{(\ln r - \ln r_{\mathrm{m}})}{\ln^2(S) r} \tag{28}$$

then

$$\frac{dn(r)}{dr} = -\frac{A}{r^2}\exp[B] + \frac{A}{r}\exp[B]\frac{dB}{dr} \tag{29}$$

$$= -\frac{A}{r^2}\exp[B] - \frac{A}{r^2}\exp[B]\frac{(\ln r - \ln r_{\mathrm{m}})}{\ln^2(S)} \tag{30}$$

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

$$0 = -\frac{A}{r_{\mathrm{M}}^2}\exp[B] - \frac{A}{r_{\mathrm{M}}^2}\exp[B]\frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})}{\ln^2(S)} \tag{31}$$

$$-1 = \frac{(\ln r_{\mathrm{M}} - \ln r_{\mathrm{m}})}{\ln^2(S)} \tag{32}$$

$$\ln r_{\mathrm{M}} = \ln r_{\mathrm{m}} - \ln^2(S) = \ln(r_m) - \sigma^2 \tag{33}$$

1.4 Properties of the Lognormal Distribution

The i-th moment of a distribution is defined as

$$m_i = \int_0^\infty r^i n(r)\,dr, \tag{34}$$

and for a lognormal distribution is given by

$$m_i = N_0 \exp\left(i\mu + \frac{i^2\sigma^2}{2}\right). \tag{35}$$

Hence the mean radius, the surface area density and the volume density of a lognormal distribution are given by

$$\begin{aligned}
\text{mean} &= \frac{1}{N_0}\int_0^\infty rn(r)\,dr, && (36)\\
&= \frac{1}{N_0}m_1, && (37)\\
&= \frac{1}{N_0}N_0\exp\left(\mu + \frac{1}{2}\sigma^2\right), && (38)\\
&= r_{\mathrm{m}}\exp\left(\frac{1}{2}\sigma^2\right), && (39)\\
&= r_{\mathrm{m}}\exp\left(\frac{1}{2}\ln^2 S\right), && (40)
\end{aligned}$$

$$\begin{aligned}
\text{area} &= \int_0^\infty 4\pi r^2 n(r)\,dr, && (41)\\
&= 4\pi m_2, && (42)\\
&= 4\pi N_0 \exp\left(2\mu + 2\sigma^2\right), && (43)\\
&= 4\pi N_0 r_{\mathrm{m}}^2 \exp\left(2\sigma^2\right), && (44)\\
&= 4\pi N_0 r_{\mathrm{m}}^2 \exp\left(2\ln^2 S\right), && (45)
\end{aligned}$$

and

$$\begin{aligned}
\text{volume} &= \int_0^\infty \frac{4}{3}\pi r^3 n(r) dr, && (46)\\
&= \frac{4}{3}\pi m_3, && (47)\\
&= \frac{4}{3}\pi N_0 \exp\left(3\mu + \frac{9}{2}\sigma^2\right), && (48)\\
&= \frac{4}{3}\pi N_0 r_{\mathrm{m}}^3 \exp\left(\frac{9}{2}\sigma^2\right), && (49)\\
&= \frac{4}{3}\pi N_0 r_{\mathrm{m}}^3 \exp\left(\frac{9}{2}\ln^2 S\right). && (50)
\end{aligned}$$

The effective radius (or area-weighted mean radius) of an aerosol distribution, r_{e}, is defined as the ratio of the third moment of the drop size distribution to the second moment, i.e.

$$\begin{aligned} r_{\mathrm{e}} &= \frac{m_3}{m_2}, && (51) \\ &= \frac{\int\limits_0^\infty r^3\, n(r) dr}{\int\limits_0^\infty r^2\, n(r) dr}. && (52) \end{aligned}$$

For a lognormal distribution this becomes

$$\begin{aligned} r_{\mathrm{e}} &= \frac{\exp\left(3\mu + \frac{9}{2}\sigma^2\right)}{\exp\left(2\mu + \frac{4}{2}\sigma^2\right)}, && (53) \\ &= \exp\left(\mu + \frac{5}{2}\sigma^2\right), && (54) \\ &= r_m \exp\left(\frac{5}{2}\sigma^2\right), && (55) \\ &= r_m \exp\left(\frac{5}{2}\ln^2 S\right). && (56) \end{aligned}$$

1.5 Gamma Distribution

The gamma distribution is given by Twomey (1977) as

$$n(r) = ar \exp\left(-br\right), \quad (57)$$

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

$$\begin{aligned} m_i &= ab^{-2-i}\Gamma\left(2+i\right), && (58) \\ &= ab^{-2-i}\left(1+i\right)!. && (59) \end{aligned}$$

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

$$n(r) = ab^2 r \exp\left(-br\right), \quad (60)$$

which has moments defined by

$$m_i = ab^{-i}\left(1+i\right)!. \quad (61)$$

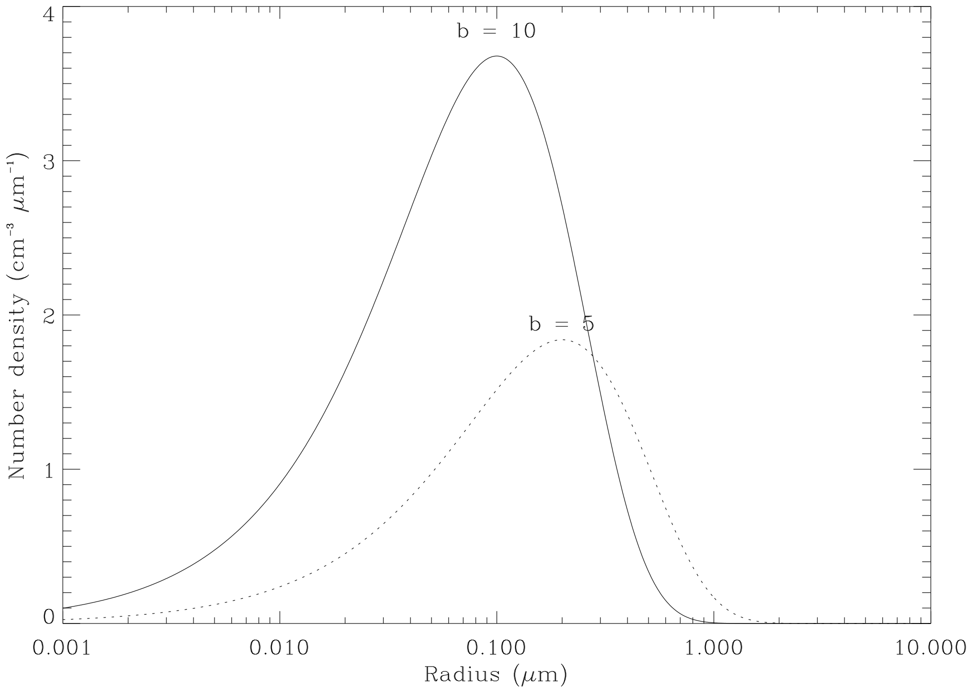


Figure 2: Normalised gamma distribution.

1.6 Modified Gamma Distribution

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

$$n(r) \quad = \quad ar^{\alpha}\exp\left(-br^{\gamma}\right). \tag{62}$$

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[1]

$$m_i \quad = \quad \frac{a}{\gamma}b^{-\frac{\alpha+1+i}{\gamma}}\Gamma\left(\frac{\alpha+1+i}{\gamma}\right). \tag{63}$$

Hence the integral of the modified gamma distribution is

$$\int_0^{\infty} n(r)\,dr \quad = \quad \frac{a}{\gamma}b^{-\frac{\alpha+1}{\gamma}}\Gamma\left(\frac{\alpha+1}{\gamma}\right). \tag{64}$$

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

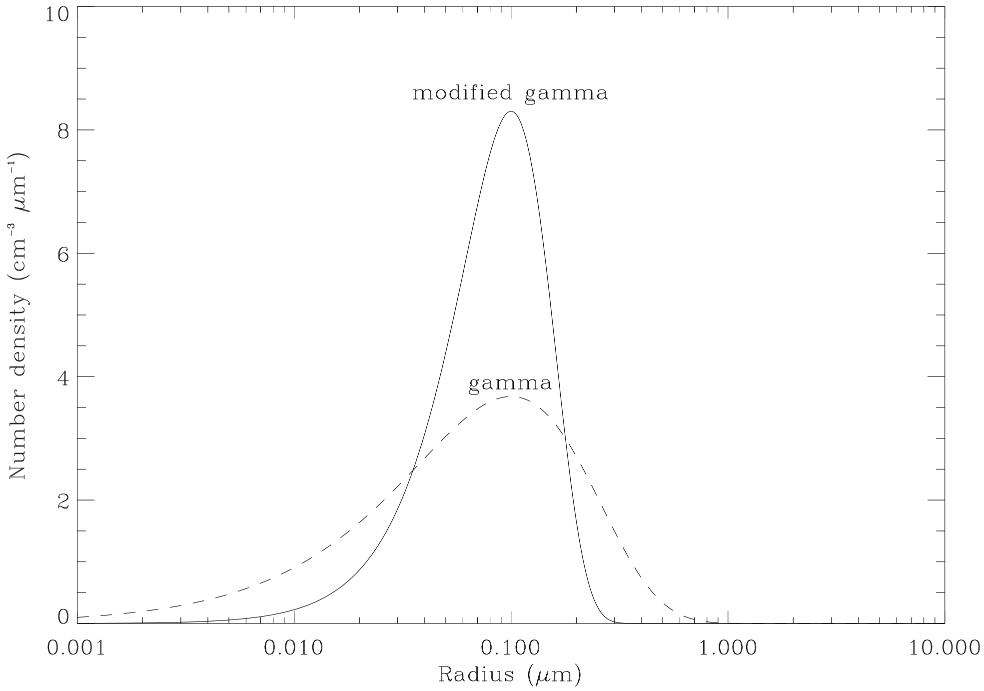


Figure 3: Normalised modified gamma distribution ($\alpha = 2, b = 100, \gamma = 2$) and gamma distribution ($b = 10$).

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

$$n(r) \quad = \quad a\frac{r^{\alpha}\exp\left(-br^{\gamma}\right)}{\frac{1}{\gamma}b^{-\frac{\alpha+1}{\gamma}}\Gamma\left(\frac{\alpha+1}{\gamma}\right)}, \tag{65}$$

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

$$m_i \quad = \quad ab^{-\frac{i}{\gamma}}\frac{\Gamma\left(\frac{\alpha+i+1}{\gamma}\right)}{\Gamma\left(\frac{\alpha+1}{\gamma}\right)}. \tag{66}$$

1.7 Inverse Modified Gamma Distribution

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

$$n(r) \quad = \quad ar^{-\alpha}\exp\left(-br^{-\gamma}\right). \tag{67}$$

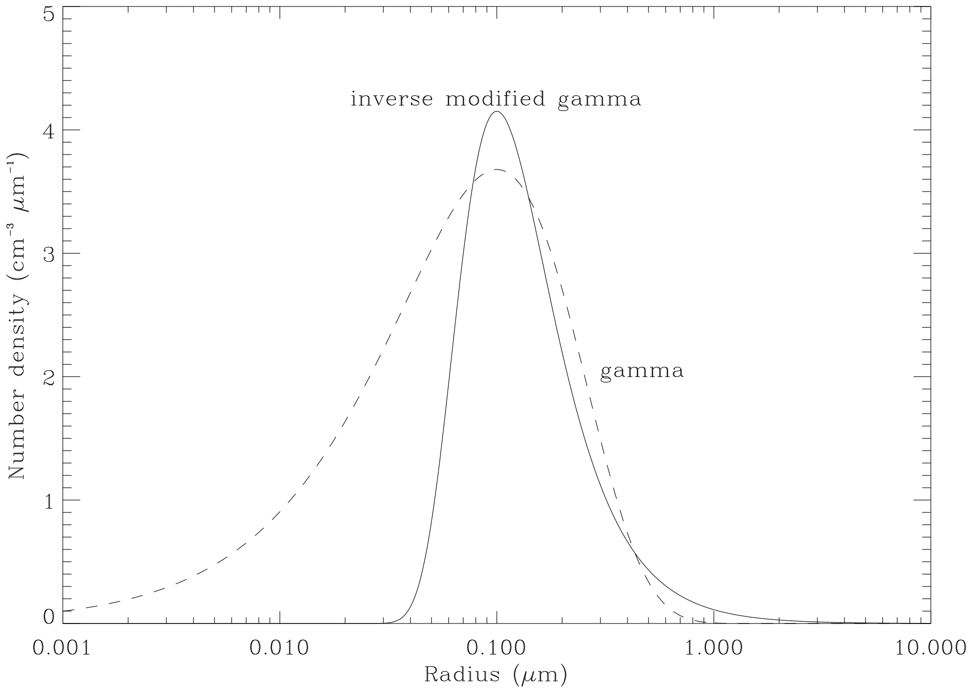


Figure 4: Normalised inverse modified gamma distribution ($\alpha = 2, b = 0.01, \gamma = 2$) and gamma distribution($b - 10$).

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

$$m_i = \frac{a}{\gamma} b^{-\frac{\alpha-1-i}{\gamma}} \Gamma\left(\frac{\alpha-1-i}{\gamma}\right). \tag{68}$$

The normalized inverse modified gamma distribution can be defined

$$n(r) = a \frac{r^{-\alpha} \exp\left(-br^{-\gamma}\right)}{\frac{1}{\gamma} b^{-\frac{\alpha-1}{\gamma}} \Gamma\left(\frac{\alpha-1}{\gamma}\right)}. \tag{69}$$

The moments of this distribution are given by

$$m_i = a b^{\frac{i}{\gamma}} \frac{\Gamma\left(\frac{\alpha-1-i}{\gamma}\right)}{\Gamma\left(\frac{\alpha-1}{\gamma}\right)}. \tag{70}$$

1.8 Regularized Power Law

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

$$n(r) = ab^{\alpha-2}\frac{r^{\alpha-1}}{\left[1+\left(\frac{r}{b}\right)^{\alpha}\right]^{\gamma}}, \tag{71}$$

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

$$r = b\left(\frac{\alpha-1}{1+\alpha(\gamma-1)}\right)^{1/\alpha}, \tag{72}$$

and the moments by

$$m_i = a\frac{b^i}{\alpha}\frac{\Gamma(1+i/\alpha)\Gamma(\gamma-1-i/\alpha)}{\Gamma(\gamma)}. \tag{73}$$

Hence the normalised distribution is

$$n(r) = a\alpha\gamma b^{\alpha-2}\frac{r^{\alpha-1}}{\left[1+\left(\frac{r}{b}\right)^{\alpha}\right]^{\gamma}}. \tag{74}$$

2 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

$$r_\mathrm{e,i} = r_{m,i} \exp\left(\frac{5}{2}\ln^2 S_i\right). \tag{75}$$

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.

$$\mu_1 = \frac{r_\mathrm{e} - r_\mathrm{e,1}}{r_\mathrm{e,3} - r_\mathrm{e,1}} \tag{76}$$

We now have two equations

$$\mu_1 + \mu_2 + \mu_3 = 1 \tag{77}$$

$$\frac{\mu_1 r_{m,1}^3 \exp\left(\frac{9}{2}\ln^2 S_1\right) + \mu_2 r_{m,2}^3 \exp\left(\frac{9}{2}\ln^2 S_2\right) + \mu_3 r_{m,3}^3 \exp\left(\frac{9}{2}\ln^2 S_3\right)}{\mu_1 r_{m,1}^2 \exp\left(2\ln^2 S_1\right) + \mu_2 r_{m,2}^2 \exp\left(2\ln^2 S_2\right) + \mu_3 r_{m,3}^2 \exp\left(2\ln^2 S_3\right)} = r_\mathrm{e} \tag{78}$$

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

$$\frac{A\mu_1 + B\mu_2 + C\mu_3}{D\mu_1 + E\mu_2 + F\mu_3} = r_\mathrm{e} \tag{79}$$

and the two equations solved to give

$$\mu_2 = \frac{r_e E - B - \mu_1(A - B + r_e(E - D))}{C - B + r_e(E - F)} \tag{80}$$

$$\mu_3 = \frac{r_e F - C - \mu_1(A - C + r_e(F - D))}{B - C + r_e(F - E)} \tag{81}$$

3 Optical Properties

3.1 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

$$\text{monodisperse only} \quad \begin{cases} \beta^{\text{abs}}(\lambda, r) = \sigma^{\text{abs}}(\lambda, r)N(r) = \pi r^2 Q^{\text{abs}}(\lambda, r)N(r), \\ \beta^{\text{sca}}(\lambda, r) = \sigma^{\text{sca}}(\lambda, r)N(r) = \pi r^2 Q^{\text{sca}}(\lambda, r)N(r), \\ \beta^{\text{ext}}(\lambda, r) = \sigma^{\text{ext}}(\lambda, r)N(r) = \pi r^2 Q^{\text{ext}}(\lambda, r)N(r), \end{cases} \tag{82}$$

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

$$\beta^{\text{abs}}(\lambda) = \int_0^\infty \pi r^2 Q^{\text{abs}}(\lambda, r) n(r)\, dr, \tag{83}$$

$$\beta^{\text{sca}}(\lambda) = \int_0^\infty \pi r^2 Q^{\text{sca}}(\lambda, r) n(r)\, dr, \tag{84}$$

$$\beta^{\text{ext}}(\lambda) = \int_0^\infty \pi r^2 Q^{\text{ext}}(\lambda, r) n(r)\, dr, \tag{85}$$

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 these quantities per particle i.e.

$$\bar{\sigma}^{\text{abs}}(\lambda) = \frac{\beta^{\text{abs}}(\lambda)}{N_0} = \int_0^\infty \sigma^{\text{abs}} n(r)\, dr, \tag{86}$$

$$\bar{\sigma}^{\text{sca}}(\lambda) = \frac{\beta^{\text{sca}}(\lambda)}{N_0} = \int_0^\infty \sigma^{\text{sca}} n(r)\, dr, \tag{87}$$

$$\bar{\sigma}^{\text{ext}}(\lambda) = \frac{\beta^{\text{ext}}(\lambda)}{N_0} = \int_0^\infty \sigma^{\text{ext}} n(r)\, dr, \tag{88}$$

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

3.2 Back Scatter

▬ to be done ▬

3.3 Phase Function

The phase function represents the redistribution of the scattered energy.

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

$$P(\lambda,\theta) \quad = \quad \frac{1}{\beta^{\mathrm{sca}}} \int_0^{\infty} \pi r^2 Q^{\mathrm{sca}}(\lambda,r) P(\lambda,r,\theta) n(r)\, dr. \tag{89}$$

3.4 Single Scatter Albedo

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

$$\omega(\lambda) \quad = \quad \frac{\beta^{\mathrm{sca}}(\lambda)}{\beta^{\mathrm{ext}}(\lambda)}. \tag{90}$$

3.5 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.

$$g \quad = \quad \frac{1}{\beta^{\mathrm{sca}}} \int_0^{\infty} \pi r^2 Q^{\mathrm{sca}}(\lambda,r) g(\lambda,r) n(r)\, dr \tag{91}$$

3.6 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

$$\int_{r_{\mathrm{l}}}^{r_{\mathrm{u}}} f(r) n(r)\, dr \quad = \quad \sum_{i=1}^{n} w_i f(r_i) \tag{92}$$

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

For a log normal size distribution the integrals are

$$\beta^{\mathrm{ext}}(\lambda) \quad = \quad \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}} \int_{r_{\mathrm{l}}}^{r_{\mathrm{u}}} r Q^{\mathrm{ext}}(\lambda,r) \exp\left[-\frac{1}{2}\left(\frac{\ln r - \ln r_{\mathrm{m}}}{\sigma}\right)^2\right] dr \tag{93}$$

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

where

$$
w'_i = \frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}} r_i \exp\left[-\frac{1}{2}\left(\frac{\ln r_i - \ln r_{\mathrm{m}}}{\sigma}\right)^2\right] w_i \tag{94}
$$

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

$$
\frac{\partial \beta^{\mathrm{ext}}}{\partial N_0} = \frac{1}{\sigma}\sqrt{\frac{\pi}{2}}\int_0^{\infty} r Q^{\mathrm{ext}}(\lambda, r) \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr, \tag{95}
$$

$$
\frac{\partial \beta^{\mathrm{ext}}}{\partial r_{\mathrm{m}}} = \frac{N_0}{r_{\mathrm{m}}\sigma^3}\sqrt{\frac{\pi}{2}}\int_0^{\infty} (\ln r - \ln r_{\mathrm{m}}) r Q^{\mathrm{ext}}(\lambda, r) \exp\left[-\frac{(\ln r - \ln r_{\mathrm{m}})^2}{2\sigma^2}\right] dr, \tag{96}
$$

$$\begin{aligned}
\frac{\partial\beta^{\mathrm{ext}}}{\partial\sigma} &= -\frac{N_0}{\sigma^2}\sqrt{\frac{\pi}{2}}\int_0^\infty rQ^{\mathrm{ext}}(\lambda,r)\exp\left[-\frac{(\ln r-\ln r_{\mathrm{m}})^2}{2\sigma^2}\right]dr \\
&\quad +\frac{N_0}{\sigma}\sqrt{\frac{\pi}{2}}\int_0^\infty \frac{(\ln r-\ln r_{\mathrm{m}})^2}{\sigma^3}rQ^{\mathrm{ext}}(\lambda,r)\exp\left[-\frac{(\ln r-\ln r_{\mathrm{m}})^2}{2\sigma^2}\right]dr, \\
&= \frac{N_0}{\sigma^2}\sqrt{\frac{\pi}{2}}\int_0^\infty \left[\frac{(\ln r-\ln r_{\mathrm{m}})^2}{\sigma^2}-1\right]rQ^{\mathrm{ext}}(\lambda,r)\exp\left[-\frac{(\ln r-\ln r_{\mathrm{m}})^2}{2\sigma^2}\right]dr.
\end{aligned} \tag{97}$$

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

$$\beta^{\mathrm{ext}}(\lambda) = \frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l}Q^{\mathrm{ext}}(l,\lambda)\exp\left[-\frac{(l-l_{\mathrm{m}})^2}{2\exp(2G)}\right]dl. \tag{98}$$

The partial derivatives of the transformed parameters (Equation 98) are

$$\frac{\partial\beta^{\mathrm{ext}}}{\partial T} = \frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l}Q^{\mathrm{ext}}(l,\lambda)\exp\left[-\frac{(l-l_{\mathrm{m}})^2}{2\exp(2G)}\right]dl, \tag{99}$$

$$\frac{\partial\beta^{\mathrm{ext}}}{\partial l_{\mathrm{m}}} = \frac{\exp T}{\exp(3G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty (l-l_{\mathrm{m}})e^{2l}Q^{\mathrm{ext}}(l,\lambda)\exp\left[-\frac{(l-l_{\mathrm{m}})^2}{2\exp(2G)}\right]dl, \tag{100}$$

$$\begin{aligned}
\frac{\partial\beta^{\mathrm{ext}}}{\partial G} &= -\frac{\exp T}{\exp(G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty e^{2l}Q^{\mathrm{ext}}(l,\lambda)\exp\left[-\frac{(l-l_{\mathrm{m}})^2}{2\exp(2G)}\right]dl \\
&\quad +\frac{\exp T}{\exp G}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty \frac{(l-l_{\mathrm{m}})^2}{\exp(2G)}e^{2l}Q^{\mathrm{ext}}(l,\lambda)\exp\left[-\frac{(l-l_{\mathrm{m}})^2}{2\exp(2G)}\right]dl, \\
&= \frac{\exp T}{\exp(G)}\sqrt{\frac{\pi}{2}}\int_{-\infty}^\infty \left[\frac{(l-l_{\mathrm{m}})^2}{\exp(2G)}-1\right]e^{2l}Q^{\mathrm{ext}}(l,\lambda)\exp\left[-\frac{(l-l_{\mathrm{m}})^2}{2\exp(2G)}\right]dl.
\end{aligned} \tag{101}$$

3.8 Cloud Liquid Water Path

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

$$l = \rho \int_0^\infty \frac{4}{3}\pi r^3 n(r)\, dr \times z \tag{102}$$

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

$$\tau = \beta^{\text{ext}} \times z \tag{103}$$

so

$$l = \rho\tau \frac{\int_0^\infty \frac{4}{3}\pi r^3 n(r)\, dr}{\beta^{\text{ext}}} \tag{104}$$

$$= \frac{4}{3}\pi\rho\tau \frac{\int_0^\infty r^3 n(r)\, dr}{\int_0^\infty \pi r^2 Q^{\text{ext}}(\lambda, r) n(r)\, dr}. \tag{105}$$

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

$$l = \frac{4}{6}\rho\tau \frac{\int_0^\infty r^3 n(r)\, dr}{\int_0^\infty r^2 n(r)\, dr} \tag{106}$$

$$= \frac{2}{3}\rho\tau r_e \tag{107}$$

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

$$l = \frac{2}{3} \times 1 \times 10 \times 15 \text{ g cm}^{-3}\mu\text{m} \tag{108}$$

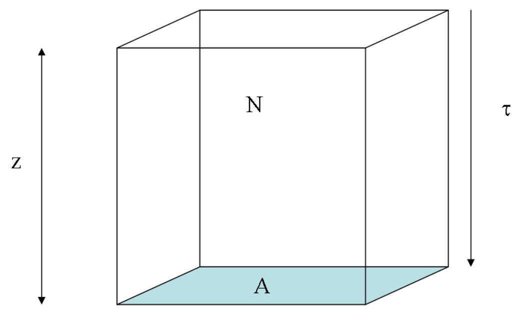
$$= 100\ \mathrm{g\,m^{-2}} \tag{109}$$

For an ice cloud (-45 °C) $\rho = 0.920$ g cm^{-3}.

3.9 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

$$M = \rho \times N \times v \times A \times Z \tag{110}$$



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.

$$m = \rho \times N \times v \times Z \tag{111}$$

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

$$\tau = \beta^{\text{ext}} \times Z \tag{112}$$

so that

$$m = \frac{\rho \times N \times v \times \tau}{\beta^{\text{ext}}} \tag{113}$$

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

$$v = \frac{\int_0^\infty \frac{4}{3}\pi r^3 n(r)\,dr}{N} \tag{114}$$

so that the mass per unit area is given by

$$m = \frac{\rho\tau}{\beta^{\text{ext}}}\int_0^\infty \frac{4}{3}\pi r^3 n(r)\,dr \tag{115}$$

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

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

$$m = \frac{\rho\tau}{\beta^{\text{ext}}}\frac{4}{3}\pi N_0 r_{\text{m}}^3 \exp\left(\frac{9}{2}\sigma^2\right) \tag{116}$$

Substance	**Density (g cm^{-3})**	**Reference**
Volcanic ash	2.42±0.79	Bayhurst, Wohletz & Mason (1994)
fill out!		

Table 1: Density of some materials that form aerosols.

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

$$\frac{\frac{\mathrm{g}}{\mathrm{cm^3}}\frac{1}{\mathrm{cm^3}}\mu\mathrm{m^3}}{\frac{1}{\mathrm{km}}} = \frac{\mathrm{g}}{10^{-6}\,\mathrm{m^3}}\frac{1}{10^{-6}\,\mathrm{m^3}}10^{-18}\,\mathrm{m^3}10^3\,\mathrm{m} = 10^{-3}\,\mathrm{g\ m^{-2}} \quad (117)$$

Table 1 list the 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 andr_m

$$r_\mathrm{e} = r_m \exp\left(\frac{5}{2}\sigma^2\right), \quad (118)$$

as well as the average extinction cross section $\bar{\sigma}^\mathrm{ext}$ defined

$$\bar{\sigma}^\mathrm{ext} = \frac{\beta^\mathrm{ext}(\lambda)}{N_0} \quad (119)$$

to get

$$m = \frac{\rho\tau}{\bar{\sigma}^\mathrm{ext}}\frac{4}{3}\pi r_\mathrm{e}^3 \exp\left(-\frac{15}{2}\sigma^2\right)\exp\left(\frac{9}{2}\sigma^2\right) = \frac{\rho\tau}{\bar{\sigma}^\mathrm{ext}}\frac{4}{3}\pi r_\mathrm{e}^3 \exp\left(-3\sigma^2\right) \quad (120)$$

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

$$m = \frac{\tau \times \rho \times N \times v}{\beta^\mathrm{ext}} \quad (121)$$

$$= \frac{\tau\sum_{i=1}^{n}\rho_i N_i v_i}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^\mathrm{ext}} \quad (122)$$

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^thmode is

$$v_i = \frac{4}{3}\pi r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right) \quad (123)$$

Hence

$$
\begin{aligned}
m &= \tau \frac{\sum_{i=1}^{n} \rho_i N_i \frac{4}{3}\pi r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right)}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\text{ext}}} && (124) \\
&= \frac{4}{3}\pi\tau \frac{\sum_{i=1}^{n} \rho_i N_i r_i^3 \exp\left(\frac{9}{2}\sigma_i^2\right)}{\sum_{i=1}^{n} N_i \bar{\sigma}_i^{\text{ext}}} && (125)
\end{aligned}
$$

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