Some exact and approximate solutions of the nonlinear Boltzmann equation with applications to aerosol coagulation

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Abstract. The nonlinear kinetic equation describing aerosol coagulation and deposition is studied analytically by a new method based upon successive coalescences. The solution is represented as an infinite series, each term of which denotes a generation of collisions and a consequent change in the aerosol number and volume distribution function. The method is illustrated by using a constant coagulation kernel and a deposition term which depends upon a power of the particle volume (zero or unity). These limitations on the physical processes inhibit use of the results for direct comparison with experiment but do provide evidence that the method may be extended to physically more realistic processes.

   The success of the method, as shown by its rapid convergence, rests upon a novel use of the central limit theorem of statistics applied to causal systems. The techniques are of general interest in the solution of the nonlinear Boltzmann equation in the kinetic theory of gases and in the case studied here show that the volume distribution of successive generations of coalescences tends rapidly to a gamma distribution.

   The success of the central limit theorem method leads us to study the solutions that can be obtained by assuming that the volume distribution function follows the gamma distribution throughout its history. Closed form solutions are obtained with a constant coagulation kernel and a deposition rate that is proportional to any power of particle volume. Numerical results are obtained for several cases which indicate that the values $m = 0$ and 1 for the deposition rate are not unreasonable for the purposes of general comment.

1. Introduction

The resurgence of interest in the behaviour of aerosols in the atmosphere and in closed containers, such as nuclear reactor buildings, has led to further mathematical studies of the underlying coagulation equation of Müller (Friedlander 1977, Mulholland and Baum 1980). The state of the art on mathematical developments up to 1972 has been comprehensively surveyed by Drake (1972). In his article, Drake considers both exact and approximate techniques for solving the nonlinear kinetic balance equation for the volume distribution function $n(v, t)$ of aerosol particles at time t. The purpose of the present article therefore is to extend some of the results reported by Drake and to introduce a new technique which appears to offer the promise of gaining further insight into the effect of the coagulation and deposition mechanisms on the particle distribution function.

The kinetic equation of Müller is made more general by the addition of a deposition term. This equation is then cast into a more suitable form under the assumption of a constant coagulation kernel and a diffusion coefficient which is proportional to v^m. For $m = 0$ and 1 it is found that exact solutions can be obtained for the kinetic equation. The value $m = 0$ corresponds physically to a leakage term caused for example by a crack. Although the value $m = 1$ does not correspond to any physical process it enables the accuracy of approximate methods to be assessed which may have wider application. Such an approximation is obtained by considering the coagulation process as a series of successive coalescences. The central limit theorem of statistics is used to sum the resulting series and this is found to lead to very accurate results for $n(v, t)$ when compared with the exact results mentioned earlier.

The results of the central limit theorem calculations suggest that the gamma distribution will be very useful for representing the volume distribution function during its lifetime. We therefore examine some exactly soluble models involving this function which are applicable for arbitrary values of m and from these results can comment on the value of the $m = 0$ and 1 cases in studying real situations.

Although the work described in this paper is related to aerosols, the techniques can profitably be used to understand the more general nonlinear Boltzmann equation for gases. Models of this equation are being studied which lead to analytically soluble results for molecular distribution functions. Futcher *et al* (1980), Hendriks *et al* (1980) and Futcher and Hoare (1980) have discussed several methods for solving the nonlinear Boltzmann equation which bear a strong resemblance to those developed independently in aerosol work. The results of the present work should therefore be considered in this wider context.

2. The coagulation equation

If we consider a medium in which the spatial variation of aerosol density can be neglected, then a balance equation for the particle volume density distribution function $n(v, t)$ can be written as

$$\frac{\partial n(v, t)}{\partial t} = \frac{1}{2} \int_0^v \mathrm{d}u \, K(u, v - u) n(u, t) n(v - u, t) - n(v, t) \int_0^\infty \mathrm{d}u \, K(u, v) n(u, t) \qquad (1)$$

where the initial condition $n(v, 0)$ is prescribed.

$K(u, v)$ is the coagulation kernel for the mechanism of interest, e.g. Brownian, sedimentation, turbulent agglomeration, etc (Hidy and Brock 1970).

Equation (1) has been solved exactly and approximately by several authors when various modelled forms are used for $K(u, v)$. It is often of interest, however, to include in equation (1) the effects of deposition, by diffusion or sedimentation, to the boundaries of the system; this is particularly important in closed vessels. In this case an additional term of the form

$$-R(v) n(v, t) \qquad (2)$$

can be added to the right-hand side of equation (1) where the nature of $R(v)$ depends upon the mechanism of deposition.

This more general type of coagulation equation has not been studied in much detail from the analytical point of view, but clearly such an analysis is desirable. The two most

common forms of $R(v)$ arise from sedimentation and Brownian diffusion. Thus if we consider a closed vessel of surface area S, volume V and height H, we can write:
Stokes settling

$$R(v) = V_s(v)/H$$

where $V_s(v)$ is the Stokes velocity, the form of which is

$$V_s(v) = (3/4\pi)^{2/3}(2\rho g/q\eta)v^{2/3}$$

with ρ the particle density, g the acceleration due to gravity and η the coefficient of viscosity of the host medium. Thus for this case $R(v) \propto v^{2/3}$ and $m = \frac{2}{3}$.
Brownian diffusion

$$R(v) = \frac{SD(v)}{V\delta}$$

where δ is the diffusion boundary layer thickness and $D(v)$ is the diffusion coefficient for Brownian motion; this takes the form

$$D(v) = (4\pi/3)^{1/3}(kT/6\pi\eta v^{1/3}).$$

$R(v)$ in this case is therefore proportional to $v^{-1/3}$ and $m = -\frac{1}{3}$.

The behaviour of $n(v, t)$ in equation (1) with these additional terms is clearly of some practical interest.

There is also the question of leakage from a closed container. If it is assumed that particles leak out through a crack or hole at a rate which is independent of their volume, then an appropriate leakage rate can be given as

$$R(v) = LS = R_0$$

where L is the leakage rate per unit area per unit time. In this case $m = 0$.

To make further progress, we have found it convenient to approximate $K(u, v)$ by a constant K and to set $R(v) = R_m v^m$. Thus by changing m both diffusion ($m = -\frac{1}{3}$) and sedimentation ($m = \frac{2}{3}$) can be simulated. Other values of m have been suggested for diffusion but the general principle remains. The constant coagulation kernel restricts the coagulation mechanism to a Brownian-like form but this is not unrealistic in practice. The equation for study is therefore of the form

$$\frac{\partial n(v, t)}{\partial t} = -R_m v^m n(v, t) - Kn(v, t)\int_0^\infty \mathrm{d}u\, n(u, t) + \frac{K}{2}\int_0^v \mathrm{d}u\, n(u, t)n(v-u, t). \tag{3}$$

In what follows we shall show how some exact and approximate solutions of equation (3) may be obtained.

3. Exact solutions

We note that two quantities of well-defined physical interest are

$$N(t) = \int_0^\infty \mathrm{d}u\, n(u, t) \tag{4}$$

i.e. the total particle number density, and

$$\phi(t) = \int_0^\infty du \, un(u, t) \tag{5}$$

i.e. the fraction of material in suspension per unit volume. Calculation of these two averages over $n(v, t)$ is a major task of this paper.

Because of the definition of $N(t)$, we can rewrite equation (3) as follows

$$\partial n(v, t)/\partial t = -[R_m v^m + KN(t)]n(v, t) + \frac{K}{2}\int_0^v du \, n(u, t)n(v - u, t). \tag{6}$$

It would seem reasonable, therefore, to seek solutions in the form

$$n(v, t) = N_0\Psi(v, t) \exp\left\{-R_m v^m t - K\int_0^t dt' \, N(t')\right\} \tag{7}$$

where $N_0 = N(0)$. Inserting equation (7) into (6) and rearranging leads to

$$\frac{\partial}{\partial t}\Psi(v, t) = \frac{N_0 K}{2}\exp\left(-K\int_0^t dt' \, N(t')\right)\int_0^v dv'\Psi(v', t)\Psi(v - v', t)$$
$$\times \exp(R_m t[v^m - v'^m - (v - v')^m]). \tag{8}$$

Clearly, the ansatz (7) will only be suitable for $m = 0$ and 1 and we therefore note that what follows in this section is not directly related to the diffusion and sedimentation processes discussed above. However when $m = 0$ we can simulate the leakage process and find

$$\frac{\partial}{\partial t}\Psi(v, t) = \frac{N_0 K}{2}\exp\left\{-R_0 t - K\int_0^t dt' \, N(t')\right\}\int_0^v dv' \, \Psi(v', t)\Psi(v - v', t) \tag{9}$$

and when $m = 1$

$$\frac{\partial}{\partial t}\Psi(v, t) = \frac{N_0 K}{2}\exp\left(-K\int_0^t dt' \, N(t')\right)\int_0^v dv' \, \Psi(v', t)\Psi(v - v', t). \tag{10}$$

Now if we define a generalised time in the case of $m = 0$ as

$$\theta(t) = 1 - \frac{KN_0}{2}\int_0^t dt' \, e^{-R_0 t'} \exp\left\{-K\int_0^{t'} dt'' \, N(t'')\right\} \tag{11}$$

and in the case of $m = 1$ as

$$\theta(t) = 1 - \frac{KN_0}{2}\int_0^t dt' \exp\left\{-K\int_0^{t'} dt'' \, N(t'')\right\} \tag{12}$$

then, in both cases, we find that $\Psi(v, t)$ or $\Psi(v, \theta)$ as we now call it obeys the equation

$$-\frac{\partial}{\partial\theta}\Psi(v, \theta) = \int_0^v dv' \, \Psi(v', \theta)\Psi(v - v', \theta) \tag{13}$$

subject to $\Psi(v, 1) = \Psi_0(v)$.

Equation (13) is readily solved by Laplace transforms and leads to

$$\Psi(v, \theta) = \frac{1}{2\pi i} \int_L \frac{ds\, e^{sv}}{\theta - 1 + (1/\bar{\Psi}_0(s))} \tag{14}$$

where

$$\bar{\Psi}_0(s) = \int_0^\infty dv\, e^{-sv} \Psi_0(v). \tag{15}$$

We conclude that the nature of $\Psi(v, \theta)$ and hence $n(v, t)$ depends crucially on the initial distribution. This is in contrast to linear problems where the initial distribution generally affects only the amplitude of the solution and not its functional form (Mulholland and Baum 1980). We shall consider below some special solutions of equation (14) and also an approximate but generally more flexible technique.

4. Rectangular initial distribution

To illustrate the dependence of the solution on the initial conditions let us consider a uniform distribution of particle sizes up to a cut-off value $v_{max} = \xi$. Thus

$$n(v, 0) = N_0/\xi; \qquad 0 \leq v \leq \xi$$
$$= 0; \qquad v > \xi. \tag{16}$$

The corresponding Laplace inversion is from equation (14) given by

$$\Psi(v, \theta) = \frac{1}{2\pi i} \int_L \frac{ds\, e^{sv}(1 - e^{-s\xi})}{s\xi - 1 + \theta + (1 - \theta)\, e^{-s\xi}}. \tag{17}$$

The poles of the integrand are given by the roots of

$$s\xi - 1 + \theta + (1 - \theta)\, e^{-s\xi} = 0. \tag{18}$$

If we put $Z = (\theta - 1)\, e^{-s\xi}$ then equation (18) can be written

$$Z\, e^Z = a \tag{19}$$

where

$$a = -(1 - \theta)\, e^{-(1-\theta)}. \tag{20}$$

But $-1 \leq a \leq 0$ and from other work (Sengupta and Srikantiah 1974) we know that this equation has one real root Z_0 and an infinite number of complex conjugate roots $Z_n = \alpha_n \pm i\beta_n$. The integral in (17) can thus be written

$$\Psi(v, \theta) = \frac{1}{\xi}(1 - \theta)^{(v-\xi)/\xi} \left\{ \left(1 - \frac{\theta}{1 + Z_0}\right)(-Z_0)^{-v/\xi} \right.$$
$$\left. + \sum_{n=1}^\infty \left[\left(1 - \frac{\theta}{1 + Z_n}\right)(-Z_n)^{-v/\xi} + \left(1 - \frac{\theta}{1 + Z_n^*}\right)(-Z_n^*)^{-v/\xi} \right] \right\} \tag{21}$$

where Z_n^* is the complex conjugate of Z_n. The numerical evaluation of $N(t)$ in this case is tedious but straightforward.

5. The gamma distribution

A very flexible and useful form of initial condition may be introduced via the gamma distribution (Scott 1968). Thus we assume that

$$n(v, 0) = \frac{N_0}{\hat{v}\Gamma(\nu+1)}\left(\frac{v}{\hat{v}}\right)^\nu e^{-v/\hat{v}}. \tag{22}$$

It has been found that $\nu = \frac{8}{3}$ gives a fair representation of some aerosol distributions (Levin 1954). In equation (22) the parameters v and ν may be defined in terms of the moments

$$\bar{v}_m = \frac{1}{N}\int_0^\infty dv\, v^m n(v) \tag{23}$$

as follows

$$\bar{v}_1 = \hat{v}(\nu+1) \tag{24}$$

and

$$\bar{v}_m = \bar{v}_1^m\, \Gamma(m+\nu+1)/(\nu+1)^m \Gamma(\nu+1). \tag{25}$$

When the gamma distribution is used in equation (14) we find

$$\Psi(v, \theta) = \frac{1}{\hat{v}}\, e^{-v/\hat{v}} \frac{1}{2\pi i}\int_L \frac{dZ\, e^{vZ/\hat{v}}}{Z^{\nu+1}-1+\theta}. \tag{26}$$

If ν is an integer the residue theorem leads to simple results. Thus, for example, for $\nu = 0$

$$\Psi(v, \theta) = (1/\hat{v})\, e^{-v/\hat{v}}. \tag{27}$$

For $\nu = 1$

$$\Psi(v, \theta) = (1/\hat{v})\, e^{-v/\hat{v}} \sinh\{(v/\hat{v})(1-\theta)^{1/2}\}/(1-\theta)^{1/2}. \tag{28}$$

For $\nu = 2$ the solution contains complex conjugate roots, which can be rearranged to give

$$\Psi(v, \theta) = \frac{e^{-v/\hat{v}}}{\hat{v}(1-\theta)^{2/3}}\left\{\frac{1}{3}\exp\left(\frac{v}{\hat{v}}(1-\theta)^{1/3}\right) - \frac{2}{25}\exp\left(-\frac{v(1-\theta)^{1/3}}{\sqrt{3}\hat{v}}\right)\right.$$
$$\left.\times\left[\cos\left(\frac{2v(1-\theta)^{1/3}}{\sqrt{3}\hat{v}}\right) + 4\sin\left(\frac{2v(1-\theta)^{1/3}}{\sqrt{3}\hat{v}}\right)\right]\right\}. \tag{29}$$

For ν fractional, in addition to a number of poles, a branch point arises in the Z plane at $Z = 0$ and it is necessary to introduce cuts. For example, when $\nu = \frac{1}{2}$ we get poles at $(1-\theta)^{2/3}$, $(1-\theta)^{2/3}(-1\pm i\sqrt{3})/2$. This leads to

$$\Psi(v, \theta) = \frac{2e^{-v/\hat{v}}}{3\hat{v}(1-\theta)^{1/3}}\{\exp[(v/\hat{v})(1-\theta)^{2/3}]$$
$$-\exp[-(v/2\hat{v})(1-\theta)^{2/3}][\cos((\sqrt{3}v/2\hat{v})(1-\theta)^{2/3})$$
$$+\sqrt{3}\sin((\sqrt{3}v/2\hat{v})(1-\theta)^{2/3})]\}$$
$$-\frac{1}{\pi\hat{v}}\,e^{-v/\hat{v}}\int_0^\infty \frac{dt\, t^{3/2}\, e^{-vt/\hat{v}}}{t^3+(1-\theta)^2}. \tag{30}$$

To obtain $N(t)$ and $\phi(t)$ it is necessary to substitute $n(v, t)$ back in the definitions given by equations (4) and (5) and evaluate numerically the resulting nonlinear equation. However the case $\nu = 0$ can be done without this complication and it is readily seen that the problem reduces to the solution of the following differential equations:

$m = 0$

$$\mathrm{d}N/\mathrm{d}t = -R_0N - \tfrac{1}{2}KN^2 \tag{31}$$

and

$$\mathrm{d}\phi/\mathrm{d}t = -R_0\phi \tag{32}$$

$m = 1$

$$\mathrm{d}N/\mathrm{d}t = -R_1\phi - \tfrac{1}{2}KN^2 \tag{33}$$

and

$$\mathrm{d}\phi/\mathrm{d}t = -2R_1\phi^2/N. \tag{34}$$

These equations represent the exact solutions for $m = 0$ and 1, respectively with an initial condition

$$n(v, 0) = (N_0/v_0)\,\mathrm{e}^{-v/v_0}. \tag{35}$$

The solutions of these equations are given in the Appendix (but see § 7).

6. The method of successive coalescences

The main difficulty in evaluating the Laplace transform of equation (14) lies in the nature of the initial condition. However, if we rewrite the equation in the form

$$\bar{\Psi}(s, \theta) = \frac{\bar{\Psi}_0(s)}{1 - (1 - \theta)\bar{\Psi}_0(s)} \tag{36}$$

and expand by the binomial theorem, the following series arises:

$$\Psi(v, \theta) = \sum_{n=0}^{\infty} (1 - \theta)^n \frac{1}{2\pi i} \int_L \mathrm{d}s\, \mathrm{e}^{sv}\bar{\Psi}_0(s)^{n+1}. \tag{37}$$

The physical meaning of this series is readily appreciated since the nth term corresponds to the distribution of those particles that have had n coalescences. We therefore call this method *the method of successive coalescences*. The solution is written

$$\Psi(v, \theta) = \sum_{n=0}^{\infty} (1 - \theta)^n F_{n+1}(v). \tag{38}$$

To make the method useful we either have to assume that the series converges rapidly, have some interest in the first few F_n or devise a method for summing the series. We shall discuss below the latter possibility.

6.1. The central limit theorem

It is well known (Papoulis 1962) that if we have a Fourier transform

$$\bar{F}_n(k) = \prod_{j=1}^{n} g_j(k) \tag{39}$$

then its inverse will be

$$F_n(\xi) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk \ e^{i\xi k} \prod_{j=1}^{n} g_j(k). \tag{40}$$

It follows from the convolution theorem that

$$F_2(\xi) = \int_{-\infty}^{\infty} d\xi_1 \ G_1(\xi_1)G_2(\xi - \xi_1) \tag{41}$$

and in general

$$F_n(\xi) = \int_{-\infty}^{\infty} d\xi_1 \ G_1(\xi_1) \dots \int_{-\infty}^{\infty} d\xi_{n-1} \ G_{n-1}(\xi_{n-1})G_n\left(\xi - \sum_{j=1}^{n-1} \xi_j\right) \tag{42}$$

or

$$F_n(\xi) = \int_{-\infty}^{\infty} d\xi' \ G_n(\xi')F_{n-1}(\xi')$$

where $G_0 = 1$, $G_1(\xi) = F_1(\xi)$ and

$$G_n(\xi) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk \ e^{i\xi k}g_n(k). \tag{43}$$

Now the central limit theorem states that $F_n(\xi)$ may be approximated by a normal distribution, provided

(1)
$$m_{j3} = \int_{-\infty}^{\infty} d\xi \ \xi^3 G_j(\xi) < C \tag{44}$$

(2)
$$\delta_j'^2 = \int_{-\infty}^{\infty} d\xi \ (\xi - m_{j1})^2 G_j(\xi) \tag{45}$$

satisfies

$$\delta_n^2 = \sum_{j=1}^{n} \delta_j'^2 \to \infty \qquad \text{as } n \to \infty. \tag{46}$$

When these conditions are satisfied

$$F_n(\xi) \sim (1/\sqrt{2\pi}\delta_n) \exp\{-(\xi - m_n)^2/2\delta_n^2\} \tag{47}$$

where $n \gg 1$ and

$$m_n = \sum_{j=1}^{n} m_{j1} \tag{48}$$

$$m_{j1} = \int_{-\infty}^{\infty} d\xi \ \xi G_j(\xi). \tag{49}$$

6.1.1. Causal functions. If the functions $G_n(\xi)$ are causal, i.e. they vanish when their argument becomes negative, the CLT needs modification. The result is that

$$F_n(\xi) \sim \frac{\xi^\beta \ e^{-\xi/\gamma}}{\gamma^{\beta+1}\Gamma(\beta + 1)} \qquad \xi \geq 0 \tag{50}$$

where

$$g_j(s) = \int_0^\infty d\xi\, e^{-s\xi} G_j(\xi) \tag{51}$$

and

$$\beta + 1 = m_n^2/\delta_n^2 \tag{52}$$

$$\gamma = \delta_n^2/m_n. \tag{53}$$

Equation (50) is the so-called 'gamma distribution' and is just the form we require for the successive coalescence problem if we recognise that all $g_j(s)$ are identical. Thus

$$m_{j1} = m_1 = \int_0^\infty d\xi\, \xi G(\xi) \tag{54}$$

hence

$$m_n = nm_1 \tag{55}$$

$$\delta_j'^2 = \int_0^\infty d\xi(\xi - m_1)^2 G(\xi) = \delta'^2 \tag{56}$$

hence

$$\delta_n^2 = n\delta'^2. \tag{57}$$

From these parameters we readily find that

$$\beta + 1 = nm_1^2/\delta'^2 \tag{58}$$

$$\gamma = \delta'^2/m_1. \tag{59}$$

It should be noted that, in practice, the convolutions rapidly tend to the CLT form of $F_n(\xi)$ even for initial distributions that are markedly non-normal. We see therefore that

$$F_{n+1}(v) \sim \frac{v^{\bar\beta_n}\, e^{-v/\bar\gamma_n}}{\bar\gamma_n^{\bar\beta_n+1}\,\Gamma(\bar\beta_n + 1)} \tag{60}$$

where

$$\bar\beta_n + 1 = (n+1)m_1^2/\delta'^2 \tag{61}$$

$$\bar\gamma_n = \delta'^2/m_1. \tag{62}$$

If the initial distribution is $\Psi(v, 0)$ then

$$m_1 = \int_0^\infty dv\, v\, \Psi(v, 0) \tag{63}$$

and

$$\delta'^2 = \int_0^\infty dv(v - m_1)^2 \Psi(v, 0). \tag{64}$$

Thus the solution can be written as

$$\Psi(v, \theta) = \sum_{n=0}^\infty \frac{(1-\theta)^n v^{\bar\beta_n} \exp(-v/\bar\gamma_n)}{\bar\gamma_n^{\bar\beta_n+1}\,\Gamma(\bar\beta_n + 1)}. \tag{65}$$

6.2. Initial conditions

If three different initial conditions are considered we find the following.

(i) Gamma distribution

$$\bar{\beta}_n + 1 = (n+1)(\nu+1) \tag{66}$$

$$\bar{\gamma}_n = \hat{v}. \tag{67}$$

(ii) Delta distribution

$$\Psi(v, 0) = \sum_i w_i \delta(v - \hat{v}_i) \tag{68}$$

$$\bar{\beta}_n + 1 = (n+1)\left(\sum_i w_i \hat{v}_i\right)^2 \bigg/ \left[\sum_i w_i \hat{v}_i^2 - \left(\sum_i w_i \hat{v}_i\right)^2\right] \tag{69}$$

$$\bar{\gamma}_n = \left[\sum_i w_i \hat{v}_i^2 - \left(\sum_i w_i \hat{v}_i\right)^2\right] \bigg/ \sum_i w_i \hat{v}_i. \tag{70}$$

(NB this can be solved exactly from equation (43) and the solution comes out in terms of sums of delta functions.)

(iii) Uniform distribution

$$\bar{\beta}_n + 1 = 3(n+1)\left(\frac{v_1 + v_2}{v_1 - v_2}\right)^2 \tag{71}$$

$$\bar{\gamma}_n = \frac{(v_1 - v_2)^2}{6(v_1 + v_2)}. \tag{72}$$

If we consider the gamma distribution, we note that for $\nu = 0$

$$\Psi(v, \theta) = (1/\hat{v}) \exp(-\theta v/\hat{v}) \tag{73}$$

which is exact. Similarly, $\nu = 1$ gives the exact result. Indeed, a little thought shows that if the initial distribution is a gamma one then this is preserved in collisions, just as a gaussian is preserved over the range $(-\infty, \infty)$. For the gamma distribution then we have an exact solution which is equivalent to that given by the contour integral of equation (26). The speed of convergence to the gamma form for other initial distributions can be readily checked by evaluating F_1, F_2, etc exactly, via equation (37).

As an example of the rapidity of convergence to the gamma distribution, we have taken the uniform distribution of (iii) above and performed the convolutions. The result is easily seen to be

$$F_n(v) = \frac{1}{(v_2 - v_1)^n} \sum_{\nu=0}^n \binom{n}{\nu} (-)^\nu \frac{1}{(n-1)!} [v - nv_1 - \nu(v_2 - v_1)]^{n-1} H(v - nv_1 - \nu(v_2 - v_1)) \tag{74}$$

where $H(v)$ is the Heaviside step function.

Figure 1 shows the $F_n(v)$ in the exact case and for the approximate gamma case of equation (65). The percentage error is given also. It is readily observed that after $n = 3$ the distributions reduce to a gamma form except in the 'wings' where the amplitude is small and hence makes little contribution to the total number density. The error over the dominant part of the distribution is less than 10%. These results are true for other initial conditions and give us confidence in the usefulness of the method of successive
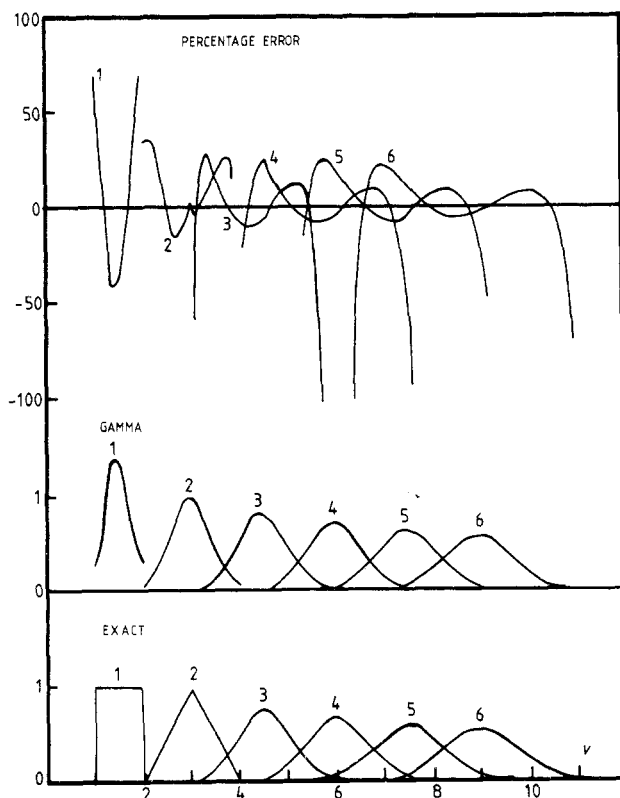
Figure 1. Example of the central limit theorem. EXACT denotes the shape of successive coalescences $F_n(v)$ following a rectangular initial distribution. GAMMA is the corresponding approximation found from the CLT. PERCENTAGE ERROR gives the fractional error, defined by

$$\frac{F_{\text{EXACT}} - F_{\text{GAMMA}}}{F_{\text{EXACT}}} \times 100.$$

coalescences and in the gamma distribution. Indeed, it would seem that an extension to arbitrary kernel would be successful. This can be illustrated by writing the basic equation as

$$\frac{\partial n_l(v,t)}{\partial t} + \left[R_m v^m + \int_0^\infty du\, K(u,v) n_{l-1}(u,t) \right] n_l(v,t)$$

$$= \tfrac{1}{2} \int_0^v du\, K(u, v-u) n_{l-1}(u,t) n_{l-1}(v-u,t) \tag{75}$$

where values $n_0, n_1, \ldots$ etc, describe sequential coalescences. We have not explored this equation further but it will prove useful if the terms converge rapidly.

As far as the central limit theorem is concerned, it is most appropriate to evaluate the first few terms in the expansion in successive coalescences exactly and then sum the remainder by the CLT. Such a technique has proved to be of great value in neutron scattering problems (Williams 1966, Syros 1966).

It should not be inferred that, because each term in the series of successive coalescences tends to a gamma distribution, the total volume density distribution is also

a gamma distribution. Indeed the exact solutions for 1, and 2 show that this is not the case. Nevertheless, we explore in the next section some consequences of making this assumption.

7. An approximation using the gamma distribution

A shortcoming of the analysis in the previous sections lies in the use of $m = 1$ for the deposition term. This value does not correspond to any physical process and therefore it is important to establish that results deduced from it are not so special or pathological as to be of little value in the general case. To this end we seek an approximate solution of equation (3) for arbitrary m and investigate its properties for any pathological effects. The natural occurrence of the gamma distribution leads us to investigate the possibility that the particle volume distribution function can be approximated over its complete lifetime by a function of the form

$$n(v, t) = \frac{N(t)}{\hat{v}(t)} \frac{1}{\Gamma(\nu + 1)} \left(\frac{v}{\hat{v}(t)}\right)^{\nu} \exp(-v/\hat{v}(t)) \tag{76}$$

where ν is fixed, but $N(t)$ and $\hat{v}(t)$ are determined by equation (3). Equation (76) has the added property of similarity, that is we may write, with $(\nu + 1)\hat{v} = \phi/N$ (Schumann 1940),

$$n(v, t) = \frac{N^2(t)}{\phi(t)} \psi\left(\frac{N(t)v}{\phi(t)}\right) \tag{77}$$

where

$$\psi(x) = \frac{(\nu + 1)^{\nu+1}}{\Gamma(\nu + 1)} x^{\nu} \exp[-(\nu + 1)x]. \tag{78}$$

It is known that many distribution functions possess the similarity property in limiting situations and so this adds further to our expectation that the gamma distribution will be acceptable for general guidance.

To find equations for N and $\hat{v}$ we insert equation (76) into equation (3) and take zeroth- and first-volume moments. This leads to

$$\frac{dN}{dt} + R_m N\hat{v} \frac{\Gamma(\nu + m + 1)}{\Gamma(\nu + 1)} = -\frac{K}{2} N^2 \tag{79}$$

and

$$\frac{d}{dt}(N\hat{v}) + R_m N\hat{v}^{m+1} \frac{\Gamma(m + \nu + 2)}{\Gamma(\nu + 2)} = 0. \tag{80}$$

It is shown in the Appendix how an exact solution of these coupled nonlinear differential equations can be obtained. However, we note that for $m = 0$ and 1 the equations, and hence the solutions, are identical with the exact ones as obtained from equations (31) and (34). This gives confidence in their overall applicability.

The results in the Appendix show that we may write the solutions of equations (79) and (80) as follows

$$\tilde{N}(t) = \frac{\tilde{\phi}^q(t)}{[1 + \Lambda(1 - \tilde{\phi}^{q(1+m)-m})]^{1/(1+m)}} \tag{81}$$

and

$$\tau = \Lambda \int_{\tilde{\phi}}^{1} \frac{\mathrm{d}y\, y^{qm-m-1}}{[1+\Lambda(1-y^{q(1+m)-m})]^{m/(1+m)}} \tag{82}$$

where $\tilde{N} = N/N_0$, $\tilde{\phi} = \phi/\phi_0$, $\tilde{v} = \hat{v}/v_0$, $q = (\nu+1)/(m+\nu+1)$ and

$$\Lambda = \frac{(1+m)(\nu+1)}{(\nu+1-m^2)}\bar{\Lambda} \tag{83}$$

$$\bar{\Lambda} = \frac{KN_0^{1+m}\Gamma(\nu+1)(1+\nu)^m}{2R_m\phi_0^m\Gamma(\nu+m+1)} \tag{84}$$

$$\tau = \frac{(1+m)(\nu+m+1)}{(\nu+1-m^2)}\bar{\tau} \tag{85}$$

$$\bar{\tau} = KN_0t/2. \tag{86}$$

Physically, $\bar{\Lambda}$ denotes the ratio of the coagulation rate to the deposition rate at zero time. For very large ν we can write

$$\Lambda = \Lambda_\infty = (1+m)KN_0^{1+m}$$

$$\tau = \tau_\infty = (1+m)\bar{\tau}.$$

From the above equations it is possible to study the way in which N and ϕ vary with time for a range of initial distributions and arbitrary values of m. For $\nu = 0$ the values of $m = 0$ and 1 give exact solutions, and the other values of m do not lead to exact solutions but it is very likely that they do not deviate appreciably from the truth. These points can be illustrated numerically. Thus we show in figures 2, 3 and 4 values of $\tilde{N}$, $\tilde{\phi}$ and $\tilde{v}$, respectively for $\nu = 0$ and $\bar{\Lambda} = 10^2$ and a range of values of m.

Figure 2 shows the reduced number density $\tilde{N}$ as a function of reduced time $\bar{\tau}$ for $m = 1, \frac{2}{3}, 0, -\frac{1}{3}$ and -0.595. The values of 0 and 1 are chosen because they correspond
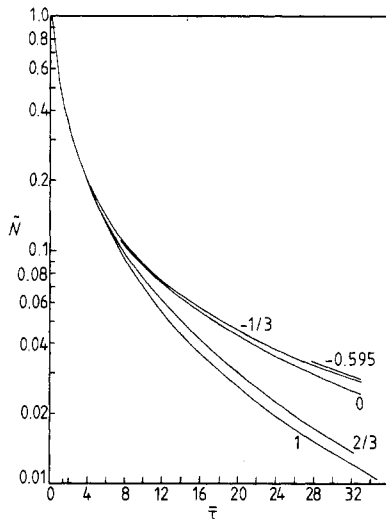
Figure 2. The reduced number density $\tilde{N}$ as a function of the reduced time $\bar{\tau}$. $\bar{\Lambda} = 10^2$, $\nu = 0$ and the value of m is indicated on the curve.
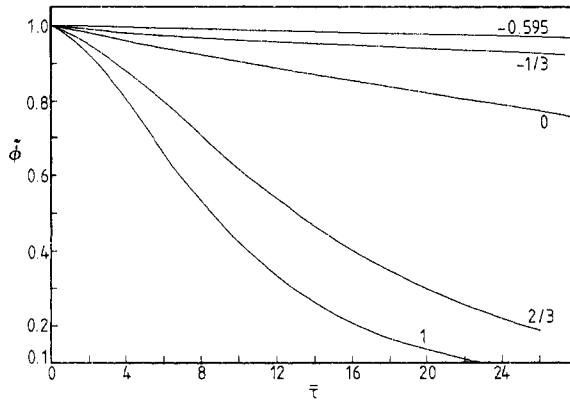
Figure 3. The reduced volume fraction $\tilde{\phi}$ as a function of the reduced time $\bar{\tau}$. $\bar{\Lambda} = 10^2$, $\nu = 0$ and the value of m is indicated on the curve.
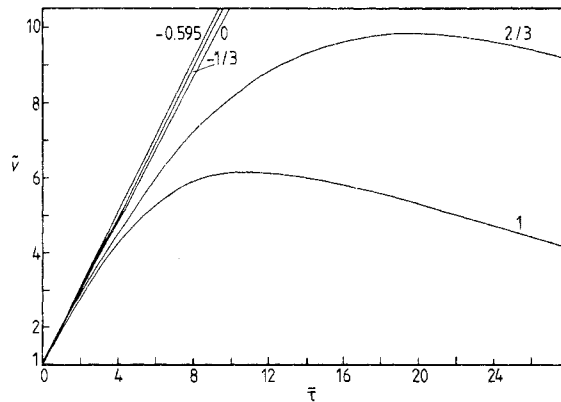

Figure 4. The reduced mean particle volume $\tilde{v}$ as a function of the reduced time $\bar{\tau}$. $\bar{\Lambda} = 10^2$, $\nu = 0$ and the value of m is indicated on the curve.

to the special models considered for the exact solution. $\frac{2}{3}$ and $-\frac{1}{3}$ are chosen because they are the values generally associated with settling and diffusion, respectively. The value of -0.595 has been suggested as being superior to $-\frac{1}{3}$ in practical cases. We observe that, as m decreases, the rate of reduction of suspended particles also decreases. The results for $m = 1$ and $\frac{2}{3}$ are not significantly different nor are the results for $m = 0$ and $-\frac{1}{3}$. The corrected value of -0.595 seems to make little difference up to $\bar{\tau} = 30$ although its effect will grow for longer times. There appears to be no special significance attaching to $m = 0$ or 1 and so results obtained from their use can be considered as typical of general m.

Figure 3 shows the reduced volume fraction as a function of reduced time $\bar{\tau}$. This quantity is much more sensitive to values of m although $m = 1$ and $\frac{2}{3}$ are still similar in general behaviour as are $m = 0$ and $-\frac{1}{3}$. If the value $m = -0.595$ is correct then it suggests that deposition by diffusion is a much less efficient process than the normal Brownian effect would predict.

Figure 4 is interesting because it seems to be the most sensitive indicator of aerosol behaviour to changes in values of m. The figure shows the mean particle volume $\tilde{v}(t)$

normalised to its value at $t = 0$ as a function of reduced time. For $m = 1$ and $\frac{2}{3}$ the mean volume increases with time up to a maximum and then decreases. This effect can be explained physically as being due to the coagulation process initially causing an increase in particle size, but later when the settling process becomes dominant, the larger particles are removed more rapidly than the smaller ones thereby reducing the average size of particle remaining in suspension. For $m < 0$, the maximum is absent due to the fact that for diffusion the diffusion coefficient becomes smaller as the average particle size increases. Detailed studies indicate that, for $m = 0$, the value of $\hat{v}(\infty)$ tends monotonically to a constant value. The existence of a maximum in $\tilde{v}$ can be proved from equations (79) and (80) by noting that at the maximum

$$d\hat{v}/dt = 0 \tag{87}$$

or

$$N \, d\phi/dt = \phi \, dN/dt. \tag{88}$$

Hence from equations (79) and (80)

$$R_m\left(\frac{\Gamma(m+\nu+2)}{\Gamma(\nu+2)} - \frac{\Gamma(m+\nu+1)}{\Gamma(\nu+1)}\right)\phi^m = \frac{K}{2}N^{1+m}. \tag{89}$$

Thus for a maximum to exist at finite $\bar{\tau}$

$$\frac{\Gamma(m+\nu+2)}{\Gamma(\nu+2)} > \frac{\Gamma(m+\nu+1)}{\Gamma(\nu+1)} \tag{90}$$

or $m > 0$.

In general, therefore, the value $m = 1$ simulates the behaviour of $m = \frac{2}{3}$. $m = 0$ does not completely cover the properties of $m = -\frac{1}{3}$ but over the initial period of the aerosol life it gives a reasonable description of the effect of diffusion which is not dissimilar to leakage.

We have also calculated values of $\tilde{N}$, $\tilde{\phi}$ and $\tilde{v}$ for other values of ν. The basic results are similar to the case of $\nu = 0$ but there are, of course, variations in detail. We note that as $\nu \to \infty$ the values of $\tilde{N}$ and $\tilde{\phi}$ tend to finite limits independent of ν. Examination of equation (78) shows that this case corresponds to $\psi(x) = \delta(x - 1)$. Thus as ν increases, the distribution narrows to a delta function. The gamma distribution can therefore describe a wide variety of similarity functions.

An extension of this technique would be to leave ν as an unknown and to determine it by taking a further moment of equation (3). The resulting coupled nonlinear differential equations would have to be solved numerically, but could prove to give an accurate description of $n(v, t)$.

8. Conclusion and summary

The purpose of the present paper has been threefold. Firstly, to examine the mathematical problems caused by the addition of a deposition term to the coagulation equation, secondly to test the efficiency of the central limit theorem for summing the effects of successive coalescences and thirdly to check the value of using a form of the deposition law proportional to v^m where $m = 0$ or 1.

We may conclude that significant difficulties are introduced when a deposition term is added. In particular, the usual canonical transformation for casting the constant

coagulation problem into a form suitable for Laplace transforms is only successful for $m = 0$ or 1. In such cases, however, exact solutions can be obtained and we have illustrated this by calculating the volume distribution as a function of time for a variety of initial conditions. To calculate the number density and volume fraction, however, usually requires the subsequent solution of a set of nonlinear equations.

The difficulty of inverting the Laplace transform for a general source distribution has been overcome by the application of the central limit theorem of statistics. This enables the volume distribution of successive coagulations to be approximated by a gamma distribution. Numerical calculations show that after a few, possibly only three, generations of coagulation the distribution function closely resembles the gamma form. On this basis we propose a numerical algorithm that should converge well for more realistic deposition rates and coagulation kernels.

Finally, to assess the physical significance of using a deposition rate proportional to $v^m (m = 0, 1)$, we have assumed that the gamma distribution holds throughout the life of the aerosol but with a time dependent amplitude and mean. Equations for the amplitude and mean are obtained by taking moments of the coagulation equation and relating them to the particle number density and the suspended volume fraction. We find that the behaviour for $m = 1$ and $m = \frac{2}{3}$ is quite similar in general form as is the behaviour for $m = 0$ and $m = -\frac{1}{3}$. Thus these two values are physically reasonable and exact results obtained by their use are likely to give guidance about the more realistic cases. Indeed, $m = 0$ does realistically simulate leakage from a container.

Acknowledgments

Appendix 1. Solution of nonlinear differential equations

Equations (79) and (80) are a pair of coupled nonlinear differential equations which by suitable transformation can be solved exactly as we shall show.

Let us write the equations as

$$dN/dt = -A_{vm}N^{1-m}\phi^m - \tfrac{1}{2}KN^2 \tag{A1.1}$$

and

$$d\phi/dt = -B_{vm}\phi^{m+1}N^{-m} \tag{A1.2}$$

where

$$A_{vm} = R_m\Gamma(v + m + 1)/\Gamma(v + 2) \tag{A1.3}$$

$$B_{vm} = R_m\Gamma(v + m + 2)/(v + 1)\Gamma(v + 2). \tag{A1.4}$$

Dividing (A1.1) by (A1.2) we get

$$\frac{dN}{d\phi} = p\frac{N^{2+m}}{\phi^{m+1}} + q\frac{N}{\phi} \tag{A1.5}$$

where $p = K/2B_{vm}$ and $q = A_{vm}/B_{vm}$.

Now substituting $N = y^\alpha$ and $\phi = x^\beta$ we find that by setting $\alpha = -1/(1+m)$ and $\beta = -1/m$, equation (A1.5) reduces to

$$\frac{dy}{dx} - q\frac{(1+m)}{m}\frac{y}{x} = p\frac{(1+m)}{m}. \tag{A1.6}$$

This equation may be integrated exactly and the result when combined with (A1.2) leads to equations (81) and (82) of the text.

Appendix 2. Further considerations on the $m = 0$ case

If aerosol coagulation is taking place in a finite volume of gas, free to expand, as for example in a nuclear air blast, then the coagulation equation will have added to it a term of the form

$$-(\partial \ln V(t)/\partial t) n(v, t)$$

where $V(t)$ is the volume of the cloud at time t. Thus if

$$R_0 = \partial \ln V(t)/\partial t$$

the theory developed for $m = 0$ can immediately be extended by replacing $R_0 t$ by $\ln (V(t)/V(0))$. The time dependence of $V(t)$ will be determined by the nature of the blast and the atmospheric conditions.

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