

Changes in Aerosol Particle Size and Number by Coagulation: A Review

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COLLISION MECHANISMS

The delivery and distribution of very small particles throughout rooms and enclosures has become an important component of military and security operations in the urban environment. However, the effectiveness of an RNP deployment will be significantly affected by entity-entity collisions and the evolution of the particle size distribution. It is essential therefore to understand the mechanisms and rates of coagulation processes occurring in a decaying turbulent field.

Following Friedlander (2000), the collision rate between particles of types i and j can be written as

$$N_{ij} = \beta(v_i, v_j) n_i n_j \quad (1)$$

where β is the collision frequency function between particles of the corresponding volumes (i and j). β has dimensions of cm^3/s . Entity-entity collision can be driven by thermal motion of the fluid molecules (Brownian motion), by fluid motion (both laminar and turbulent), and by differential sedimentation (requiring a difference in size or density).

The collision frequency function for Brownian coagulation was developed by Smoluchowski (1917). If the participating particle size is significantly larger than the mean free path of the gas molecules ($\approx 0.06 \mu\text{m}$ in air at 0°C), and if the Stokes-Einstein diffusion coefficient is employed, then

$$\beta(v_i, v_j) = \frac{2kT}{3\mu} \left(\frac{1}{v_i^{1/3}} + \frac{1}{v_j^{1/3}} \right) (v_i^{1/3} + v_j^{1/3}). \quad (2)$$

For a monodisperse system, $v_i = v_j$, and then $\beta = \frac{8kT}{3\mu}$. This is valid for the continuum

regime where the Knudsen number (Kn) is less than 0.1. In this case, the initial rate of disappearance of particles is given by:

$$\frac{dn}{dt} = -\frac{4kT}{3\mu} n^2 \quad (3)$$

A collision efficiency factor (λ) can be incorporated into eq. (3) to account for the possibility that not all collisions result in aggregate formation; see, e.g., Swift and Friedlander (1964). Computed collision efficiencies in hydrosols were compared by Kusters et al. (1997); for solid spherical entities, λ decreases sharply with increasing particle size.

An attractive feature of (3) is that it is easily solved to yield:

$$\frac{n}{n_0} = \frac{1}{\frac{4kT}{3\mu} n_0 t + 1} \quad (4)$$

Thus, for example, we can estimate the time required for the number concentration of particles to be reduced to $n_0/2$ in air at 20°C:

Initial concentration per cm ³ , n_0	$t_{1/2}$, s
1x10 ⁸	33.6
1x10 ⁷	336
1x10 ⁶	3357

The actual rate of particle disappearance will be affected by several factors, including the breakdown of continuum theory (as particles approach each other), deviations from sphericity, and electrical charge. Shahub and Williams (1988) reported that van der Waals, viscous, and electrostatic forces interact in a complex way and significantly alter the coagulation rate (from that predicted by classical theory). For electrostatic forces, weakly bipolar atmospheric aerosols yield a net effect that is nearly a wash. However, Friedlander (2000) indicates that a strongly charged (bipolar) aerosol will yield a greatly enhanced coagulation rate. The collision rate correction factor, W , is given by

$$W = \frac{1}{y} (e^y - 1) \quad \text{where} \quad y = \frac{z_i z_j e^2}{\epsilon_0 k T (R_i + R_j)}.$$

z is the number of charges on the colliding particles, e is the fundamental electrostatic unit of charge, and ϵ_0 is the dielectric constant of the medium (air: 1.0006). To illustrate, consider a pair of 2 μm particles in air, each carrying 20 charges, but of opposite sign. For this case, $y = -5.69$ and $W = 0.175$; the collision rate enhancement is $1/W$ which is a factor of 5.7. If, on the other hand, ions of like charge are preferentially adsorbed upon the particle surface, coagulation can be very effectively inhibited. Xiong et al. (1992) describe how alkali metals such as Na, K, and Rb can be used to significantly narrow the particle size distribution for TiO_2 formed by the oxidation of TiCl_4 . They note that the particulate scavenging of ions is a much more rapid process than coagulation.

Fluid motion can also drive interparticle collisions; the collision frequency function for particles “i” and “j” in a laminar shear field with a velocity gradient, dU/dz , was derived by Smoluchowski (1917):

$$\beta(v_i, v_j) = \frac{4}{3} (R_i + R_j)^3 \frac{dU}{dz} \quad (5)$$

And again, the rate of disappearance of monodisperse particles can be written as a simple ordinary differential equation (assuming that the dispersed-phase volume fraction, ϕ , is constant):

$$\frac{dn}{dt} = -\frac{4\phi}{\pi} \frac{dU}{dz} n \quad (6)$$

This equation has been tested many times for hydrosols, usually in some type of Couette device with (nearly) uniform velocity gradient. For concentric cylinder apparatuses, dU/dz can be assigned a single value that can be varied by changing the speed of the (outer) cylinder. Eq. (6) is also easily integrated, yielding:

$$n = n_0 \exp\left(-\frac{4\phi}{\pi} \frac{dU}{dz} t\right) \quad (7)$$

This result is not likely to be of significant interest for impulsively deployed aerosols, however, since no part of the flow field could be described as a simple laminar current.

Saffman and Turner (1956) developed the collision frequency function for small particles in isotropic turbulence:

$$\beta(v_i, v_j) = 1.3 \left(\frac{\varepsilon}{\nu}\right)^{1/2} (R_i + R_j)^3. \quad (8)$$

ε is the dissipation rate per unit mass and ν is the kinematic viscosity of the fluid. Note the similarity of this equation to (5). A few words regarding the dissipation rate are in order. For isotropic turbulence, the dissipation rate is:

$$\varepsilon = 2\nu \overline{s_{ij}s_{ij}} \quad (9)$$

where s_{ij} is the fluctuating strain rate. The strain rate is difficult to measure because it requires velocities with spatial separation. However, it is a critical parameter of turbulent flows; for a given fluid it determines the eddy size(s) in the dissipation range of wavenumbers. By definition, the wavenumber that corresponds to the beginning of the dissipation range (in the three-dimensional spectrum of turbulent energy) is:

$\kappa_d = \frac{1}{\eta}$, where the Kolmogorov microscale is given by $\eta = \left(\frac{v^3}{\varepsilon} \right)^{1/4}$. Therefore, in air

with $\varepsilon = 100 \text{ cm}^2/\text{s}^3$, $\eta \approx 0.077 \text{ cm}$ and $\kappa_d \approx 13 \text{ cm}^{-1}$. Under laboratory conditions, the dissipation rate is generally in the range of 10 to perhaps $10^4 \text{ cm}^2/\text{s}^3$; ε can also be estimated with Taylor's inviscid approximation: $\varepsilon \approx Au^3/l$. For pipe flows, Delichatsios and Probst (1975) used the relation: $\varepsilon \approx 4V_*^3/d_{\text{pipe}}$, where V_* is the shear, or friction, velocity. This relationship for dissipation rate came from experimental work carried out by Laufer (1954). In atmospheric turbulence, the dissipation rate is

inversely proportional to height in neutral air: $\varepsilon = \frac{V_*^3}{K_a z}$. For unstable air, ε decreases

with height near the surface, becoming constant near the top of the surface layer which is the lowest part of the planetary boundary layer. Panofsky and Dutton (1984) note that in daytime with strong winds, surface layer simplifications are valid to a height of about 100 m.

In cases where dispersed particles differ in size and mass, interparticle collision can also occur by turbulent inertia and differential sedimentation. The collision frequency functions for these two cases are:

$$\beta(v_i, v_j) = 5.7(R_i^3 + R_j^3) \left| \tau_i - \tau_j \right| \frac{\varepsilon^{3/4}}{v^{1/4}} \quad (10)$$

where τ is a characteristic time (mass of particle/ $6\pi\mu R$) and

$$\beta(v_i, v_j) = \pi\alpha(R_i + R_j)^2 (V_i - V_j). \quad (11)$$

Unless (or until) there is considerable disparity in the sizes of the particles, these collision mechanisms will be minor contributors to the processes of interest. In the deployment of RNP's, we might expect (11) to become increasingly important with time, but of little significance initially. Finally, Williams (1988) noted that the actual collision frequency under the influence of gravity would be less than indicated by (11) because the distortion of the velocity field affects the trajectories of approaching particles.

SELF-PRESERVING SIZE DISTRIBUTIONS

Swift and Friedlander (1964) and Friedlander and Wang (1966) developed a technique for solving certain types of coagulation problems based upon a similarity transformation. They observed that after long times, the solutions to such problems may become independent of the initial particle size distribution. Thus,

$$n(v, t) = \frac{N^2}{\phi} \psi\left(\frac{v}{\bar{v}}\right) \quad \text{where } \bar{v} \text{ is the average particle volume.}$$

ψ is a dimensionless function that is invariant with time. The particle size distribution must also satisfy the following:

$$N = \int_0^{\infty} n(v, t) dv, \text{ i.e., the total number of particles must be obtained}$$

by integrating the distribution over all possible volumes. In addition, the dispersed-phase volume fraction can be determined:

$$\phi = \int_0^{\infty} n(v, t) v dv.$$

Finally, it is usually taken that the distribution function is zero for both $v=0$ and $v \rightarrow \infty$. Friedlander (2000) shows results for the Brownian coagulation case and provides a comparison with experimental data obtained with a tobacco smoke aerosol. The agreement is reasonable. The principal problem with this technique is that while a transformation may be found for the collision kernel of interest, an appropriate solution may not necessarily exist.

An important question in this context is the length of time required for the size distribution to become self-preserving. Vemury et al. (1994) report that for Brownian coagulation in the continuum regime, τ_c was found to be on the order of 12 to 13; since

$$T_c = \frac{\tau_c}{K_c n_0}, \text{ and } K_c = \frac{2kT}{3\mu}, \text{ one can estimate the time required given a specific medium}$$

and an initial number concentration of particles. For air at 20°C with $n_0 = 1 \times 10^7$ particles per cm^3 , $T_c \approx 8000$ s.

DYNAMIC BEHAVIOR OF THE PARTICLE SIZE DISTRIBUTION

Processes of the type that are of interest here lend themselves to analysis by population balance. In the chemical process industries, population balances were first used for the analysis of crystal nucleation and growth by Hulburt and Katz (1964), among others. In RNP deployment we can expect aggregation, sedimentation, and perhaps Coulombic deposition. Furthermore, small particles will be carried about by eddies of all sizes (from integral to dissipative scales). In a decaying, inhomogeneous turbulent flow of the type created by impulsive insertion of RNP's, the general problem is quite intractable. Some alternative approaches will be discussed later. Friedlander (2000) notes that if the Reynolds decomposition and time-averaging are employed with the general population balance, the result is:

$$\frac{\partial \bar{n}}{\partial t} + \nabla \bar{n} + \frac{\partial}{\partial v} (\bar{n} q) + \frac{\partial}{\partial v} (\bar{n}' q') = -\nabla \bar{n}' V' + D \nabla^2 \bar{n} +$$

$$\frac{1}{2} \int_0^v \beta(v^*, v - v^*) n(v^*) n(v - v^*) dv^* - \int_0^{\infty} \beta(v, v^*) n(v) n(v^*) dv^* +$$

$$\frac{1}{2} \int_0^v \beta(v^*, v - v^*) \overline{n'(v^*) n'(v - v^*)} dv^* - \int_0^\infty \beta(v, v^*) \overline{n'(v) n'(v^*)} dv^* - V_s \frac{\partial \bar{n}}{\partial z} \quad (12)$$

The familiar problem of closure rears its head here. The turbulent fluxes are often represented as though they were mean-field, gradient transport processes; e.g.,

$$\overline{n' V_i'} \approx -D_T \frac{\partial \bar{n}}{\partial x_i} \quad (13)$$

where D_T is an eddy diffusivity. However, we should remember that such analogies have little physical basis; coupling between the turbulence and the mean field variables is very weak.

A dynamic equation that includes aggregation and sedimentation for a system that is spatially homogeneous (well-mixed) can be written as:

$$\frac{dn(v)}{dt} = \frac{1}{2} \int_0^v \beta(v, v - \bar{v}) n(v) n(v - \bar{v}) d\bar{v} - n(v) \int_0^\infty \beta(v, \bar{v}) n(\bar{v}) d\bar{v} - \frac{V_s(v)}{h} n(v) \quad (14)$$

$n(v)$ is the particle size distribution (number concentration as a function of volume), β is the collision frequency function, V_s is the settling velocity, and h is the vertical “depth” of the system. Note that (14) does not include diffusion or convective transport. If the settling particles follow Stokes law, and if buoyancy is neglected, then

$$\frac{4}{3} \pi R^3 \rho_p g = 6 \pi \mu R V_s \quad (15)$$

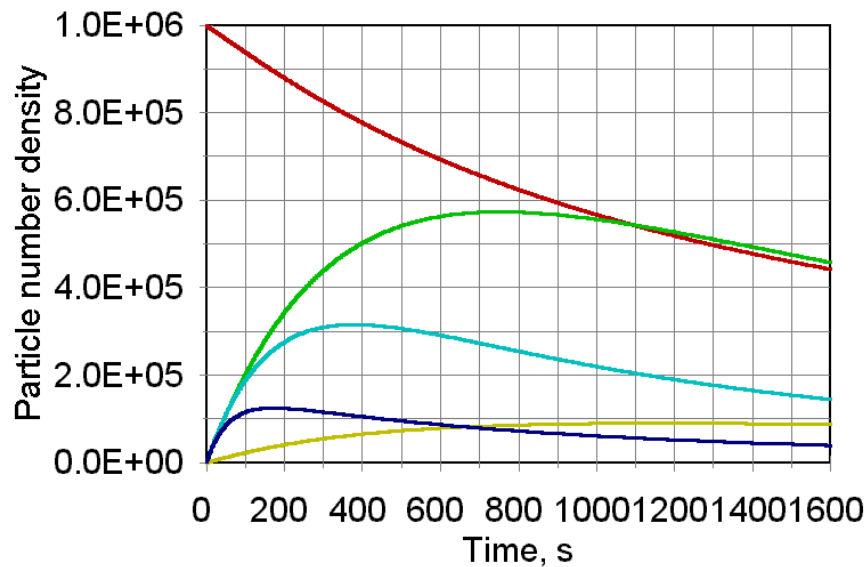
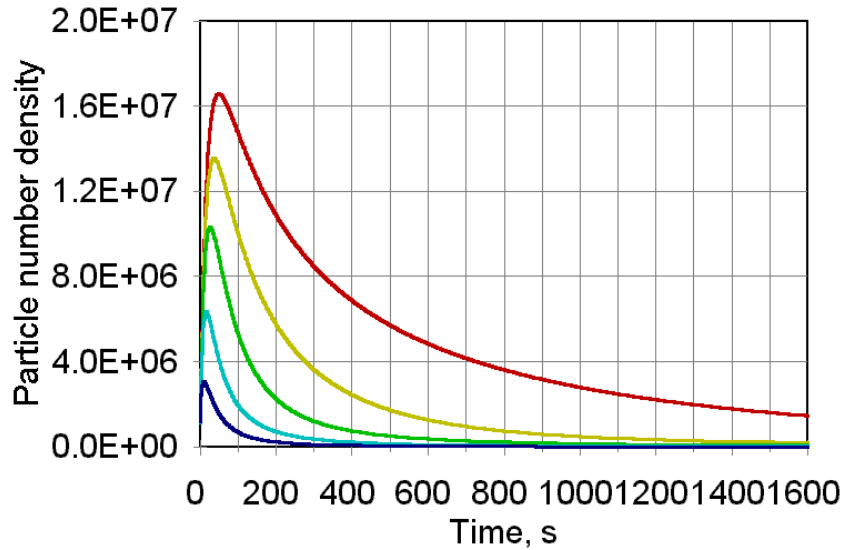
However, the right-hand side of (15) should be modified for smaller particles to account for non-continuum effects. Seinfeld (1986) provided a table of values for the Cunningham correction factor; for a particle with a diameter of 0.1 μm , the Stokes drag should be divided by 2.85. Thus, V_s would be increased by 285%.

Farley and Morel (1986) recast eq. (14) in discrete form for application to a limited number of logarithmically spaced particle classes:

$$\frac{dn_k}{dt} = \frac{1}{2} \sum_{i+j=k} \alpha(i, j) \beta(i, j) n_i n_j - n_k \sum_{i=1}^m \alpha(i, k) \beta(i, k) n_i - \frac{V_s(k)}{h} n_k \quad (16)$$

$\alpha = 1$ if $i \neq j$, and 2 if $i = j$. With a discrete model of this type, a collision does not necessarily produce a particle in the next larger class; consequently, particle volume may not be conserved with eq. (16), even if disappearance by sedimentation is removed. One method of compensation is to use weighting fractions so that only a portion of i - j collisions yields production in higher classes. Additional collision frequencies can be added to (16) to account for turbulence-induced coagulation, or other phenomena. However, Williams (1988) notes that there is no *a priori* reason to assume that the resultant coagulation kernel should merely be the sum of the individual mechanisms. The most attractive aspect of the modeling approach described above is that influences of the initial particle size distribution, settling velocities, and collision efficiencies could be very

rapidly compared (qualitatively). A simulation program was developed to illustrate this; the algorithm considers just Brownian motion and it uses eight particle classes with mean diameters corresponding to: 0.375, 0.75, 1.5, 3, 6, 12, 24, and 48 μm . This is a logarithmic spacing as recommended by Gelbard and Seinfeld (1978). The figures provided below give some indication of the wide variations possible in the evolution of the particle size distribution.



Figures 1A and B. Comparison of simulation results showing the changes in population of the five largest particle classes. In the upper figure (1A) the particles are initially placed in the 0.75 μm class and the loss of larger particles by settling is enhanced. In the lower figure (1B), the initial particles are fewer in number and spread among the first

four classes, but loss by sedimentation is suppressed. The reduced particle number density and the inhibited settling used in 1B result in sluggish dynamics.

A comparison of these preliminary results with those computed by Lindauer and Castleman (1971) indicates that the simulation performs surprisingly well. However, a number of modifications would clearly be appropriate, including allocating the classes or bins according to: $V_{n+1}=2V_n$. For spherical particles or entities this corresponds to $d_{n+1}=1.26d_n$. Therefore, covering particle diameters ranging from 0.4 to 10 μm would require 15 classes and extension to 40 μm , 21. This alteration should make it easier to achieve conservation of volume, where appropriate.

OTHER ASPECTS OF PSD MODELING

Gelbard et al. (1980) observed that numerical solutions for dynamic aerosol balances require approximation of the continuous size distribution by some finite set of classes or sections. They addressed the question as to whether a “sectional representation” can in fact produce an accurate solution for a dynamic aerosol problem. They were able to show that for the limiting case in which the section size (or class interval) decreases, that the finite representation reduced to the classic coagulation equation. By comparison with experimental (power plant plume) data, they demonstrated that the discrete approximation yielded satisfactory results.

Direct numerical simulation has become (at least somewhat) feasible due to recent increases in computing power. Reade and Collins (2000), for example, devised a simulation for a “periodic” volume (a particle whose trajectory causes it to leave through a bounding surface immediately re-enters the domain on the opposite side) using 262,144 initial particles. They considered isotropic turbulence with a Reynolds number (based upon the Taylor microscale) of 54. Their results show that a finite Stokes number* (St, the ratio of the stop distance and a characteristic dimension of the system) results in a much broader particle size distribution (psd) than does either limiting case (St=0 or St $\rightarrow\infty$). Furthermore, they found that the standard deviation of the psd decreased with increasing St. Reade and Collins used their results to test collision kernels written in power law form (collision diameter raised to a power, p). They found that dynamic psd behavior could not be adequately represented with a constant value of p; the conclusion is that the dynamic behavior of real particles may not correspond closely to idealized collision mechanisms.

Sandu (2002) employed a discretization of the coagulation equation in which the integral terms were approximated by Newton-Cotes sums. A polynomial of order n was used to interpolate the function at the nodes (collocation). This resulted in a system of coupled ordinary differential equations which was solved with a semi-implicit Gauss-Seidel iteration. The technique was said to offer improved accuracy over earlier approaches.

* *The Stokes number is important in inertial deposition. For impaction upon a cylindrical fiber, $St = \frac{\rho_p d_p^2 V}{18\mu d}$.*

Fernandez-Diaz et al. (2000) improved the semi-implicit technique developed by Jacobson et al. (1994) that produced unwanted numerical diffusion (unphysical broadening of the particle size distribution). Fernandez-Diaz et al. attacked this problem by devising different partition coefficients for the bins; they noted that the coagulation of i- and j-type particles might not necessarily result in a new entity of volume $v_i + v_j$. In fact, the new entity could have a volume corresponding to $(v_i + v_j)$ where the volumes were both from the bottom (minimum) of the original bins, or from the top (maximum) of each. Therefore, they assumed that each bin could be characterized by the geometric mean of its limits; i.e., $v_k = \sqrt{v_{k-} v_{k+}}$. This results in each bin having a width of 1 in the new size space. In addition, particles were uniformly distributed throughout the bin and the volumes of the bins varied as: $v_x = v_1 [1 + b(x - 1)]^a$, where a and b were appropriately chosen. It appeared that this technique better approximated populations in the larger entity sizes than that achieved with geometrical spacing of bins.

A SIMPLIFIED STARTING POINT

Finally, an elementary starting point for comparison of gross effects will be described. We consider a highly simplified model that provides partial connection between the particle number density and the fluid mechanics (dissipation rate). Differential sedimentation could be added, and the model could be compartmentalized (with exchange between the subunits) to handle inhomogeneous turbulence. If we presume that the collision kernels are additive (which is suspect, as noted previously) and neglect particle size variation, then:

$$\frac{dn}{dt} = - \left(\frac{4}{3} \frac{kT}{\mu} + 5.2 \left(\frac{\varepsilon}{\nu} \right)^{1/2} R^3 \right) n^2 \quad \text{with} \quad (17)$$

$$\frac{d}{dt} \left(\frac{3}{2} u^2 \right) = -\varepsilon \approx -A \frac{u^3}{l}. \quad (18)$$

In eq. (18) the dissipation rate is represented with Taylor's inviscid approximation; u is a characteristic velocity and l is the integral length scale. Such a model would be valid only initially, and only for the initial period of decay (of turbulence in a box); for advanced times, the dissipation rate estimate would need to be replaced with an equation of the type:

$$\varepsilon \approx C \nu u^2 / l^2. \quad (19)$$

Tennekes and Lumley (1972) recommend making the transition to the final period of decay at

$$\text{Re} = \frac{ul}{\nu} = 10. \quad (20)$$

This modeling approach might be useful for qualitative purposes such as assessment of the initial effects of dissipation rate, particle number density, and particle size. It would also be possible to include a loss term in (17) to account for deposition onto surfaces, should that be necessary.

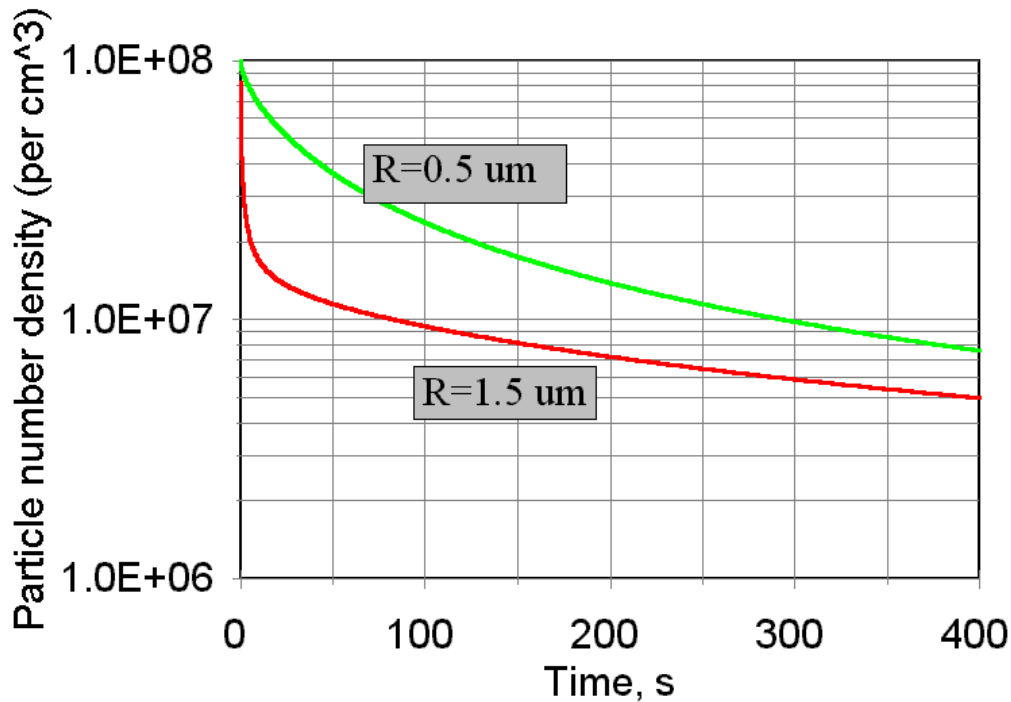


Figure 2. Illustration of the effects of particle size upon the (simultaneous) solution of equations (17) and (18). Clearly, turbulence is very effective in the initial rate of reduction of larger particles (with $R=1.5 \mu\text{m}$); the times required for an order of magnitude reduction can be compared: $t_{10\%}(1.5)/t_{10\%}(0.5) \approx 85/290 = 0.29$.

An important question in this context is whether or not eq. (18) can adequately represent the decay of turbulent energy in enclosures. Several experiments were carried out using constant temperature anemometry (CTA) with decaying turbulence in box (with a volume of 136 liters). Turbulent flow was created with a centrifugal blower with a nominal discharge velocity of about 8 to 9 m/s. At a predetermined time, the box was isolated from the blower, and the air velocity was recorded at a point 16 cm from the wall and 16 cm above the floor (bottom). Of course, every experiment of this type is unique; it would be necessary to use an ensemble if one were seriously interested in characterizing the mean velocity. Results from a single run are shown below in Figure 3. The box was isolated from the blower at $t=2$ s, and the decay was monitored until the mean velocity had declined by more than two orders of magnitude.

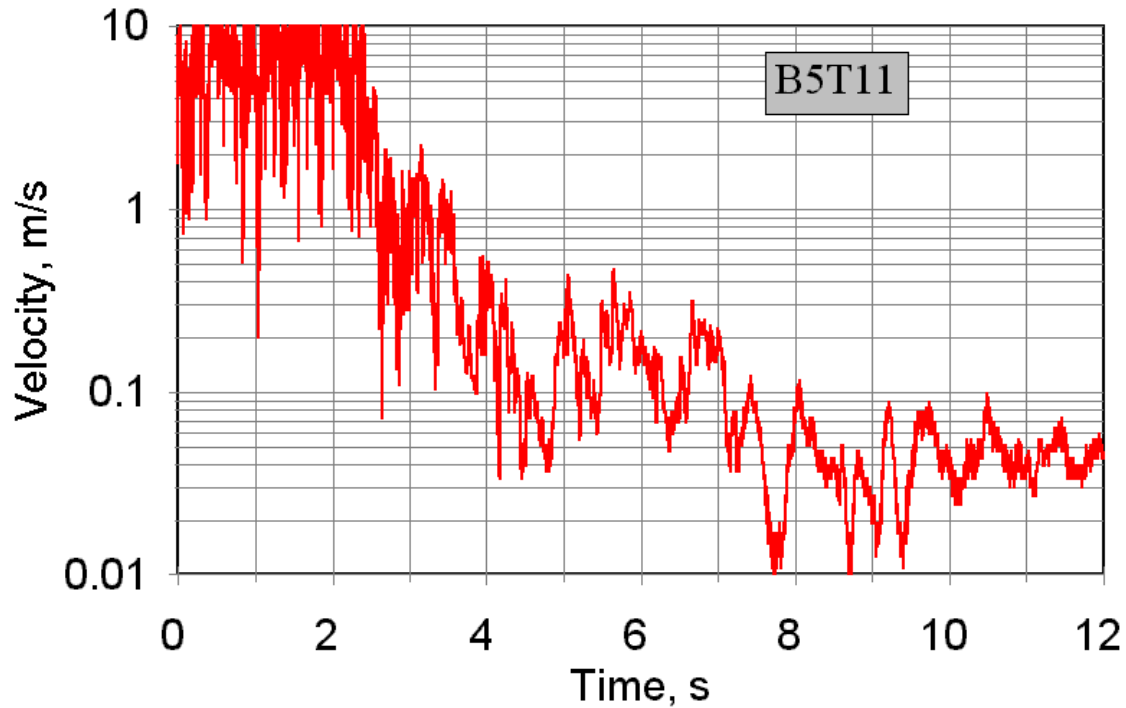


Figure 3. CTA results for decaying turbulence in a box. The centrifugal blower was isolated from the test chamber at $t=2$ s. After 10 s ($t=12$), the mean velocity has fallen to about 4 or 5 cm/s.

In eq. (18), the constant, A , has been set to 1.5 as indicated by experiment. The integral length scale, l , is generally taken to correspond to the size of the largest eddies present in the flow. In an enclosure such as a rectangular box, the smallest of the principal dimensions, length, width, and height (L , W , h), would be a very rough approximation. For the test apparatus, the minimum dimension (size) is about 36 cm. Eq. (18) was solved for integral lengths of 12, 20, and 36 cm and the results are shown in Figure 4 below. Note that the curve for 20 cm corresponds reasonably with the experimental data in Figure 3 above (at $t=4$ s, $u \approx 0.2$ m/s, at $t=6$ s, $u \approx 0.1$ m/s, and at $t=10$ s, $u \approx 0.05$ m/s).

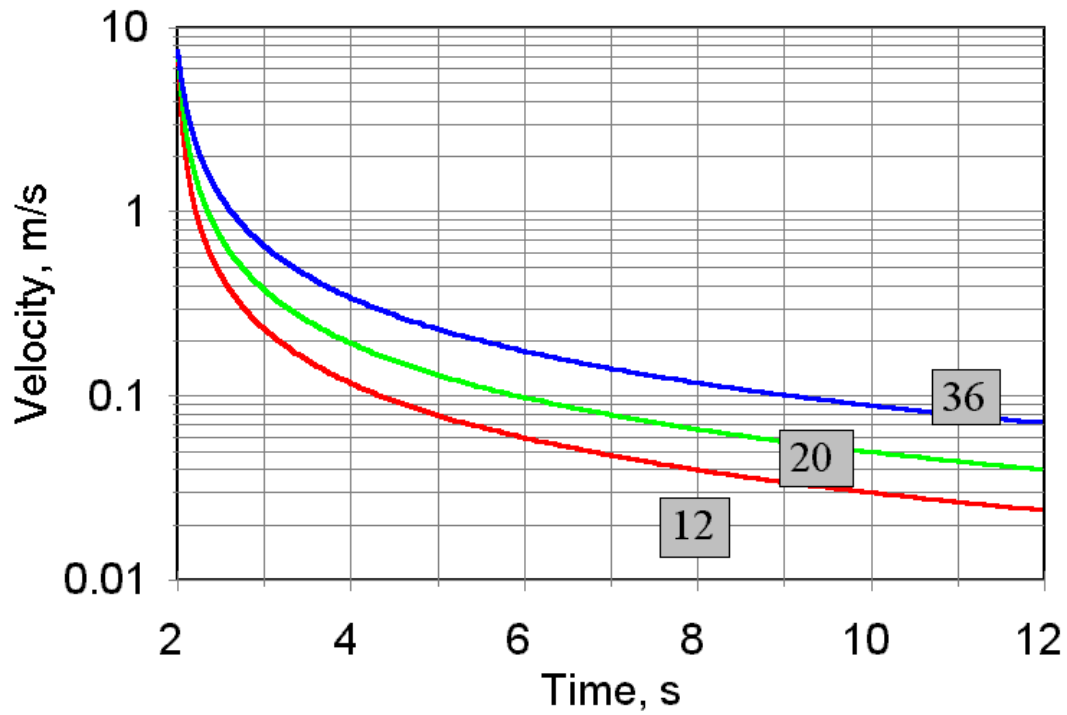


Figure 4. Results from a simplified model for decaying turbulence in a box using Taylor's inviscid approximation for the dissipation rate. The three curves are for integral length scales (l 's) of 12, 20, and 36 cm.

These data suggest that eq. (18) is an appropriate approximation for turbulent energy decay, at least for systems of small scale. We should also observe that the Reynolds number, as given by eq. (20), would still have a value of about 500 at $t=12$ s; the final period of decay would begin when the velocity, u , was about 0.08 cm/s. Based upon the results shown in Figure 4, $u \approx 0.08$ cm/s would not be attained until $t \approx 500$ s. At that point, Taylor's approximation for ϵ would have to be replaced by eq. (19).

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