Statistical mechanical descriptions of turbulent coagulation

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A fundamental tenet of statistical mechanics is that the rate of collision of two objects is related to the expectation value of their relative velocities. In pioneering work by Saffman and Turner [J. Fluid Mech. 1, 16 (1956)], two different formulations of this tenet are used to calculate the collision kernel $\Gamma$ between two arbitrary particle size groups in a turbulent flow. The first or spherical formulation is based on the radial component $w_r$ of the relative velocity $w$ between two particles: $\Gamma^\text{sph} = 2\pi R^2 \langle |w_r| \rangle$, where $w_r = w \cdot R/R$, $R$ is the separation vector, and $R = |R|$. The second or cylindrical formulation is based on the vector velocity itself: $\Gamma^\text{cyl} = 2\pi R^2 \langle |w| \rangle$, which is supported by molecular collision statistical mechanics. Saffman and Turner obtained different results from the two formulations and attributed the difference to the form of the probability function of $w$ used in their work. A more careful examination reveals that there is a fundamental difference between the two formulations. An underlying assumption in the second formulation is that the relative velocity at any instant is locally uniform over a spatial scale on the order of the collision radius $R$, which is certainly not the case in turbulent flow. Therefore, the second formulation is not expected to be rigorously correct. In fact, both our analysis and numerical simulations show that the second formulation leads to a collision kernel about 25% larger than the first formulation in isotropic turbulence. For a simple uniform shear flow, the second formulation is about 20% too large. The two formulations, however, are equivalent for treating the collision rates among random molecules and the gravitational collision rates. © 1998 American Institute of Physics. [S1070-6631(98)01410-X]

I. INTRODUCTION

The rate of coagulation in turbulent dispersions of small solid particles and droplets is important to many areas of meteorology and engineering. Examples include precipitation and cloud processing of aerosols, production of titanium-dioxide pigments, fine spray combustion, and formation of industrial emissions. The overall coagulation rate of finite-size particles in fluid turbulence is governed by three consecutive and interrelated processes: (1) geometric collision due to particle–turbulence interactions, (2) collision efficiency due to local particle–particle aerodynamic interactions, and (3) coagulation efficiency as determined by surface sticking characteristics. The starting point of turbulent coagulation is a formulation of the average collision kernel that measures the percentage of particle pairs that will adhere to form larger particles per unit time and volume. There is more than one standard way to express the collision kernel, and the question of whether they are equivalent has not been made clear, particularly when turbulent coagulation is considered. In this paper, we will use the geometric collision rate as a way to clarify and compare two popular formulations used in the pioneering work of Saffman and Turner, although these formulations are also used as a basis to study other aspects of the coagulation processes.

The paper by Saffman and Turner is probably the most cited paper in turbulent coagulation literature. In that paper, Saffman and Turner presented two formulations of the collision kernel $\Gamma$ between two arbitrary particle size groups (we shall limit our discussions to body-force-free, inertialless particles of size much smaller than the Kolmogorov length). In the first formulation, the average collision kernel is described as the average volume of fresh fluid entering a collision sphere per unit time,

$$\Gamma^\text{sph} = 2\pi R^2 \langle |w_r| \rangle. \tag{1}$$

The collision sphere is defined, relative to a reference particle, as a sphere of radius $R = r_1 + r_2$, centered on the reference particle [Fig. 1(a)]. Here $r_1$ and $r_2$ are the radii of the two particle size groups, $w_r$ is the radial component of the relative velocity $w$, namely, $w_r = w \cdot R/R$, $R$ is the separation vector, and $R = |R|$. Since this first formulation, Eq. (1), is based on a spherical geometry, we shall call it the spherical formulation. One important assumption of Eq. (1) is that the relative velocity $w$ is incompressible, thus influx and outflux over the sphere surface are equal. The collision kernel is then half the surface area multiplied by the average magnitude of the radial relative velocity.

In the second formulation, the collision kernel is described in terms of the relative velocity directly and is defined as the cylindrical volume passing through a reference particle per unit time, with cross-sectional area $\pi R^2$ and length $|w|$ [Fig. 1(b)]. The average collision kernel is then

$$\Gamma^\text{cyl} = \pi R^2 \langle |w| \rangle. \tag{2}$$
where $u_i$ is the flow velocity and $s_{ij}$ is the local rate of strain tensor. Although the velocity gradients can be assumed to be uniform over a spatial scale $R$, the relative velocities will depend on the orientation of $R$ over the surface of the collision sphere. Thus, the spherical formulation is general, whereas the cylindrical formulation reduces to the spherical formulation under the circumstances stated. For collision of random molecules in statistical mechanics, we will demonstrate that the two formulations lead to the same result.

It should be noted that both formulations have been widely cited in the literature, but the difference in the two formulations has never been made clear. The spherical formulation was used recently in the studies of Koziol and Leighton,\textsuperscript{4} Hu and Mei,\textsuperscript{5} Wang et al.,\textsuperscript{6} and Zhou et al.;\textsuperscript{7} while the cylindrical formulation or its equivalent form was used in the studies of Kruijts and Kusters\textsuperscript{8} and Sundaram and Collins.\textsuperscript{9} We have shown recently through direct numerical simulations that the spherical formulation gives the correct collision kernel, under the assumptions that particles can overlap and are retained in the system after collisions.\textsuperscript{5} Therefore, we argue that the cylindrical formulation should not be used for treating turbulent coagulation. An estimation of the level of error involved in the cylindrical formulation will be given later in this paper.

II. ANALYSIS

Now let us demonstrate the above points. In isotropic turbulence, the statistical averages of $|w_i|$ and $|w|$ are independent of the direction of $R$. Therefore, we may limit ourselves to the case of $R$ aligned with the $x$ direction. In this case, we have

$$\langle w^2 \rangle = \langle w_x^2 \rangle = R^2 \left( \frac{\partial u_x}{\partial x} \right)^2 = \frac{R^2}{15} \bar{\varepsilon} = \sigma^2,$$

and

$$\langle w^2 \rangle = \langle w_x^2 \rangle + \langle w_z^2 \rangle,$$

where we use the notation $w = (w_x, w_y, w_z)$, $\bar{\varepsilon}$ is the average rate of viscous dissipation per unit mass, and $\nu$ is the fluid kinematic viscosity. For isotropic turbulence, it can be shown that

$$\langle w_x^2 \rangle = \langle w_z^2 \rangle = 2 \sigma^2,$$

and

$$\langle w_x w_z \rangle = \langle w_y w_z \rangle = 0.$$

Note that Eq. (6) follows simply from the fact that

$$\left( \frac{\partial u_x}{\partial x} \right)^2 = \left( \frac{\partial u_z}{\partial x} \right)^2 = 2 \left( \frac{\partial u_y}{\partial x} \right)^2,$$

for isotropic turbulence field. It follows that

$$\langle w_x^2 \rangle = 5 \sigma^2.$$

We shall now make the assumption that the relative velocities follow a Gaussian probability distribution, although it is now well known that this is not the case.\textsuperscript{11} This simple assumption, however, is consistent with the original work of Saffman and Turner\textsuperscript{1} and allows us to evaluate the relative
velocity statistics in Eqs. (1) and (2) explicitly in terms of $\sigma$. Furthermore, for the first- and second-order velocity moments of concern here, the Gaussian assumption is a reasonable approximation. Equation (4) implies that the probability distribution of $w_r$ may be written as

$$p(w_r) = \frac{1}{\sqrt{2\pi \sigma}} \exp \left( -\frac{w_r^2}{2\sigma^2} \right). \tag{10}$$

then

$$\langle |w_r| \rangle = \int_{-\infty}^{\infty} |w_r| p(w_r) dw_r = \sqrt{\frac{2}{\pi}} \sigma. \tag{11}$$

Combining Eqs. (1), (4), and (11), we arrive at the well-known result of Saffman and Turner:

$$\Gamma_{sph, turb} = 2\pi R^2 \sqrt{\frac{2}{\pi} \frac{R}{\sqrt{15} \tau_k}} = 1.294 \frac{R^3}{\tau_k}, \tag{12}$$

where $\tau_k = \sqrt{\frac{1}{\epsilon}}$ is the Kolmogorov time scale. Similarly, considering Eqs. (4), (6), and (7), we can write the probability distribution of $w$ as

$$p(w) = \frac{1}{\sqrt{2\pi \sigma}} \exp \left( -\frac{w^2}{2\sigma^2} \right) \frac{1}{\Gamma_{sph, turb}} \exp \left( -\frac{w^2 + w_r^2}{4\sigma^2} \right). \tag{13}$$

Therefore,

$$\langle |w| \rangle = \int dw_x \int dw_y dw_z p(w) \sqrt{w_x^2 + w_y^2 + w_z^2} = \sigma \left( \sqrt{\frac{\pi}{2}} + \sqrt{\frac{2}{\pi}} \right). \tag{14}$$

A detailed derivation of the above result is given in the Appendix. Substituting Eq. (14) into Eq. (2), we have

$$\Gamma_{cyl, turb} = \pi R^2 \left( \sqrt{\frac{\pi}{2}} + \sqrt{\frac{2}{\pi}} \right) \frac{R}{\sqrt{15} \tau_k} = 1.664 \frac{R^3}{\tau_k}. \tag{15}$$

This result is almost the same as that given by Saffman and Turner based on the isotropic form of the probability distribution for $p(w)$:

$$p(w) = \frac{1}{(2\pi \sigma_1^2)^{3/2}} \exp \left( -\frac{w \cdot w}{2\sigma_1^2} \right), \tag{16}$$

with $\sigma_1^2 = w^2$. Their result is

$$\Gamma_{cyl, turb, ST} = \pi R^2 \sqrt{\frac{40}{3\pi} \frac{R}{\sqrt{15} \tau_k}} = \sqrt{\frac{8\pi R^3}{9 \tau_k}} = 1.671 \frac{R^3}{\tau_k}. \tag{17}$$

In summary, our result based on the cylindrical formulation and the correct Gaussian probability function is 28.6% larger than the result obtained from the spherical formulation, while $\Gamma_{cyl, turb, ST} = 1.291 \Gamma_{sph, turb}$. Saffman and Turner realized the inconsistency between the two formulations, but attributed this inconsistency to the simplified probability distribution given by Eq. (16). Here we have clarified that this is not the reason. Sundaram and Collins made an effort to derive an expression of the collision kernel for finite-inertia particles. They assumed that $p(w)$ was independent of the orientation of the separation vector, which led their formulation to be equivalent to the cylindrical formulation. The variances of the relative velocity components used in their work [Eq. (2.3) in their paper] are not correct. The above two problems happen to cancel each other in their formulation when applied to fluid elements.

To confirm the above results, we computed directly $\langle |w_r| \rangle$, $\langle |w| \rangle$, and other related statistics for turbulent flow fields generated by direct numerical simulations and used in our previous work (see Zhou et al. for numerical simulation details). The results are listed in Table I. The separation $R$ was set to the Kolmogorov length $\eta = (v^3/\epsilon)^{1/4}$. The first row shows the grid resolutions and Taylor microscale Reynolds numbers for DNS turbulent flow fields. Let us first check the Gaussian assumption by comparing the ratios $\langle |w_r| \rangle/\sqrt{\langle w_r^2 \rangle}$ and $\langle |w| \rangle/\sqrt{\langle w \cdot w \rangle}$. These ratios are equal to 0.7979 and 0.9173 for Gaussian turbulence, and the simulation results give similar values but deviate slightly more from the Gaussian results as the flow Reynolds number increases. Finally, the ratio $\langle |w| \rangle/2\langle |w_r| \rangle$ corresponds to $\Gamma_{cyl, turb}/\Gamma_{sph, turb}$ and changes from 1.271 to 1.228 as the flow Reynolds number increases, comparable to the predicted value of 1.286 based on the Gaussian assumption.

One can further show that the two formulations give different results for a simple shear flow with velocity field given as $u = (\gamma z, 0, 0)$. For this flow field, the averages over the collision sphere surface can be directly evaluated. Using spherical polar coordinates, we have $w_r = \gamma (r_1 r_3 / R) = (\gamma R / 2) \cos \theta \sin 2\theta$ and $|w| = r_3 (d\varphi / d\phi) = \gamma R |\cos \theta|$ on the surface of the collision sphere, where $\theta$ is the polar angle and $\psi$ is the azimuthal angle. Here $r_1$, $r_2$, and $r_3$ are the vector components of $R$. Therefore,

$$\langle |w_r| \rangle = \frac{1}{4\pi} \int_0^{2\pi} d\psi \int_0^\pi d\theta \cdot \sin \theta \cdot \frac{\gamma R}{2} |\cos \psi \cdot \sin 2\theta| = \frac{2}{3\pi} \gamma R, \tag{18}$$

and

$$\langle |w| \rangle = \frac{1}{4\pi} \int_0^{2\pi} d\psi \int_0^\pi d\theta \cdot \sin \theta \cdot \gamma R |\cos \theta| = \frac{\gamma R}{2}. \tag{19}$$

Turbulent Flow Field: Statistics of Two-point Relative Velocities

| $R$ values | $\langle |w_r| \rangle$ | $\langle |w| \rangle$ |
|-----------|----------------|----------------|
| 32$^+$ | 1.3426 | 1.0692 |
| 64$^+$ | 3.0175 | 1.9069 |
| 96$^+$ | 3.4115 | 2.6546 |
| 128$^+$ | 15.2749 | 9.5984 |

| $R$ values | $\langle |w_r| \rangle$ | $\langle |w| \rangle$ |
|-----------|----------------|----------------|
| 32$^+$ | 0.7729 | 0.7743 |
| 64$^+$ | 0.8729 | 0.8568 |
| 96$^+$ | 1.2705 | 1.2413 |
| 128$^+$ | 1.2406 | 1.2278 |

$^+$ Data taken from Zhou et al. [1]
It follows that
\[ \Gamma_{\text{sph}}^{\text{shear}} = \frac{4}{3} \pi R^3 = 1.333 \gamma R^3, \]  
(20)
\[ \Gamma_{\text{cyl}}^{\text{shear}} = \frac{\pi}{4} \gamma R^3 = 1.571 \gamma R^3, \]  
(21)
therefore, the cylindrical formulation leads to a result 18% larger than the spherical formulation. The spherical formulation gives the correct, well-known result of von Smoluchowski.\(^{12}\)

We shall now present two physical situations for which the two formulations actually give identical results. The first one concerns the collision kernel of random molecules such as in statistical mechanics.\(^{7}\) In this case there is no spatial correlation of particle velocities and the relative velocity statistics do not depend on the orientation of \( \mathbf{R} \), namely,
\[ \langle w_x^2 \rangle = \langle w_y^2 \rangle = \langle w_z^2 \rangle = \sigma^2. \]  
(22)
Then the probability distribution of \( w \) is
\[ p(w) = \frac{1}{(\sqrt{2} \pi \sigma)^3} \exp \left( -\frac{w^2}{2\sigma^2} \right), \]  
(23)
leading to
\[ \langle |w| \rangle = \frac{\sqrt{\pi}}{4} \sigma = 2\sqrt{2} \sigma. \]  
(24)
where \( w = |w| \). Therefore, the two formulations give the identical result:
\[ \Gamma_{\text{random}}^{\text{sph}} = \Gamma_{\text{random}}^{\text{cyl}} = \sqrt{8} \pi R^2 \sigma. \]  
(25)

The second case concerns the gravitational collision kernel in which the relative velocity is uniform and equal to \( w = (0,0,\Delta V) \), where \( \Delta V \) is the differential settling rate. Obviously the cylindrical formulation gives
\[ \Gamma_{\text{cyl}}^{\text{gravity}} = \pi R^2 \Delta V. \]  
(26)
While the spherical formulation will predict
\[ \Gamma_{\text{sph}}^{\text{gravity}} = 2 \pi R^2 \frac{1}{4 \pi} \int_0^{2\pi} \sin \theta d\theta \int_0^\pi \sin \theta |\Delta V \cos \theta| d\psi \]  
\[ = \pi R^2 \Delta V, \]  
(27)
which is identical to the prediction based on the cylindrical formulation.

### III. CONCLUSIONS

From the above four example problems, we can draw the following conclusions.

1. The spherical formulation in Eq. (1) always gives the correct prediction under the assumptions that particles can overlap in space and are retained in the system after collisions, as shown by Wang et al.\(^6\)

2. The cylindrical formulation in Eq. (2) is equivalent to the spherical one when the relative velocity is an uncorrelated isotropic random field or uniform over the spatial scale of \( \mathbf{R} \).

3. The cylindrical formulation can overpredict the collision kernel by 20% or more in a turbulent flow field.

4. The Gaussian assumption results in reasonable predictions of low-order relative velocity statistics.

As a final note, we would like to suggest an alternative expression for the collision kernel when weak particle inertia and gravitational effect are included. Saffman and Turner\(^1\) gave the following expression:

\[ \Gamma_{\text{ST}} = 2 \sqrt{2} \pi R^2 \frac{1}{6} \left( \frac{\rho_s}{\rho_p} \right)^2 \left( \frac{\Delta V}{D}\right)^2 \left( \frac{Du}{Dt}\right)^2 \]  
(28)
where \( \rho_p \) and \( \rho_f \) are particle and fluid densities, \( \tau_i = (\rho_s/\rho_f)(d_i/18\nu) \) are the Stokes response times of the particles, \( Du/Dt \) is the \( x \) component of the fluid acceleration, and \( g \) is the gravitational acceleration. The first term represents the effect of local shear in the turbulent flow (the shear term); the second term results from the differential response of the particles to local fluid acceleration (the acceleration term); and the last term is due to differential settling by gravity (the gravity term).

This expression, although widely cited in the literature, has a number of inconsistencies due to the use of the cylindrical formulation and the isotropic form of the probability density function for \( w \). A more consistent expression is

\[ \Gamma = 2 \sqrt{2} \pi R^2 \frac{1}{15} \left( \frac{\rho_s}{\rho_p} \right)^2 \left( \frac{\Delta V}{D}\right)^2 \left( \frac{Du}{Dt}\right)^2 \]  
(29)
where \( \lambda_D \) is the longitudinal Taylor-type microscale of fluid acceleration (see Hu and Mei\(^5\)). In addition to the terms of same physical origins as in Eq. (28), we have included a term accounting for the combined effect of spatial variation of fluid acceleration and particle inertia (the coupling term), according to Hu and Mei.\(^5\) This coupling term does not appear in the work of Saffman and Turner because they assumed that the local fluid accelerations are perfectly correlated in space.

Equation (29) represents several improvements over Eq. (28): (1) the shear term, for the reason discussed in this pa...
per, is corrected; (2) the gravity term in Saffman and Turner is not correct due to their incorrect probability function for \( \mathbf{w} \) if the gravity effect is considered alone; and (3) the coupling term derived by Hu and Mei is more important than the acceleration term when monodisperse particles are considered. We note that the only term identical to that in Saffman and Turner’s expression is the acceleration term when monodisperse particles are considered.

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### APPENDIX: THE DERIVATION OF \( \langle |\mathbf{w}| \rangle \) FOR A GAUSSIAN RELATIVE VELOCITY FIELD

In this appendix, we provide a detailed derivation for \( \langle |\mathbf{w}| \rangle \), the average magnitude of the relative velocity between two colliding particles. We would like to demonstrate that \( \langle |\mathbf{w}| \rangle \) can be derived analytically for a general Gaussian relative velocity field, with the correct nonisotropic probability function given by Eq. (13). The derivation requires several nontrivial steps of integration by parts. This was probably the reason for Saffman and Turner to use the isotropic probability function, Eq. (23), instead.

We start by substituting Eq. (13) into the first part of Eq. (14). A change of variables from \( (w_x, w_y) \) to cylindrical coordinates \((\rho, \theta)\) is made to reduce the triple integration to double integration:

\[
\langle |\mathbf{w}| \rangle = \int d\rho \int d\theta \int dw_x dw_y p(\mathbf{w}) \sqrt{w_x^2 + w_y^2 + w_z^2} \]

\[
= \frac{1}{(2\pi)^{2/2}} \int_{-\infty}^{\infty} \exp\left( -\frac{w_z^2}{2\sigma^2} \right) dw_z \times \int_{0}^{\infty} \exp\left( -\frac{\rho^2}{4\sigma^2} \right) 2\pi \rho d\rho \cdot \sqrt{w_x^2 + \rho^2}.
\]

At this point, we can simplify the expression by setting \( x = w_x/(2\sigma) \) and \( y = \sqrt{(w_z^2 + \rho^2)/(2\sigma)} \). The integration can be written in terms of \( x \) and \( y \) as

\[
\langle |\mathbf{w}| \rangle = \frac{4\sqrt{2}\pi}{\sqrt{\pi}} \int_{0}^{\infty} e^{-x^2} dx \int_{x}^{\infty} y d(-e^{-y^2}).
\]

Integration by parts then gives

\[
\langle |\mathbf{w}| \rangle = \frac{4\sqrt{2}\pi}{\sqrt{\pi}} \left( \int_{0}^{\infty} e^{-2x^2} dx + \int_{0}^{\infty} dx \int_{x}^{\infty} e^{-(x^2 + y^2)} dy \right).
\]

The first integral can be easily carried out and the symmetry property of the second integrand allows us to write

\[
\langle |\mathbf{w}| \rangle = \frac{4\sqrt{2}\pi}{\sqrt{\pi}} \left( \frac{1}{4} + \frac{1}{8} \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} e^{-(x^2 + y^2)} dy \right).
\]

A change of variables from \((x, y)\) to cylindrical coordinates allows the second integration to be performed analytically as

\[
\langle |\mathbf{w}| \rangle = \frac{4\sqrt{2}\pi}{\sqrt{\pi}} \left( \frac{1}{4} + \frac{1}{8} \int_{-\infty}^{\infty} e^{-\rho^2/2} 2\pi \rho d\rho \right)
\]

\[
= \sigma \left( \sqrt{\frac{\pi}{2}} + \sqrt{\frac{2}{\pi}} \right),
\]

which is the final result used in the paper.