Letter

Variance of the velocity in suspensions of particles does not diverge

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In fluid dynamics, a classic calculation pertaining to suspensions of uniform spheres predicted that the variance of the velocity should diverge with system size. Experiments, on the other hand, find that while the variance grows with system size in small systems, it asymptotes to a fixed value for sufficiently large systems. Here, I show that this discrepancy between theory and experiment is resolved by accounting for the inertia of the particles. Using the unsteady Stokes‘ Green’s functions, the leading order contribution to the variance is determined to be finite for systems of infinite extent.

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In a suspension of particles, the movement of one particle can influence the motion of nearby particles through the fluid flow that its motion generates. For particles undergoing random movements, such as Brownian motion, these fluid-mediated interactions can impact the magnitude of the average fluctuations in the system. Experiments have shown that the variance of particle velocity in a suspension increases with particle volume fraction [1]. This velocity variance directly influences particle diffusion and will, therefore, impact the behavior of suspensions directly and through its impact on the Peclet number and other relevant dynamic parameters [2]. For example, under shear, suspended particles can migrate perpendicular to the flow in a Peclet number–dependent way [3], which is important in many situations, including blood flow dynamics [4]. These motions will be affected by enhanced diffusion resulting from increased velocity variance. Building off the work in [5–7], Caflisch and Luke (CL) sought to compute the velocity variance of a suspension of sedimenting particles [2]. Surprisingly, they found that the variance should diverge with system size. While experiments find that early on, the velocity variance scales like the system size, these large-scale fluctuations die out such that, on long timescales, the variance is independent of the system size for large systems [1]. Consequently, the theoretical prediction has become known as the Caflisch-Luke paradox.

In this paper, I show that the CL paradox arises due to the neglect of inertia, and that a proper accounting of it leads to a finite velocity variance at an infinite system size. First, I will consider the inertial dynamics of a single sphere of radius a and density ρ_s randomly fluctuating in a fluid of viscosity η and density ρ. If the random fluctuations are small, so that the particle moves significantly less than the particle radius before being redirected, then the particle‘s acceleration will dominate the nonlinear convective term in the material derivative. Consequently, the fluid motion should be well-described by the unsteady Stokes’ equations

$$\rho \frac{\partial \mathbf{v}}{\partial t} = \eta \nabla^2 \mathbf{v} - \nabla p \,,$$
$$\nabla \cdot \mathbf{v} = 0 \,, \tag{1}$$

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where $\mathbf{v}$ is the fluid velocity and p is the pressure. If the fluid is otherwise quiescent, then the disturbance flow from the particle can be described by a Green's function that consists of the unsteady Stokeslet and its Laplacian [8]. If the particle is centered at $\mathbf{x}_m$ (where the m is used as an index for the particle), then the velocity at location $\mathbf{x}$ in the fluid is given by $\mathbf{v}(\mathbf{x},t) = \hat{\mathbf{B}}^0 \cdot \hat{\mathbf{S}}^0(\mathbf{x},\mathbf{x}_m)e^{-i\omega t}$, with

$$\hat{S}^0_{ij} = \frac{\delta_{ij}}{r}\left[\frac{2e^{-\lambda(r-a)}}{D}\left(1+\frac{1}{\lambda r}+\frac{1}{\lambda^2 r^2}\right)-\frac{2}{\lambda^2 r^2}\right] \\ -\frac{\hat{x}_i\hat{x}_j}{r^3}\left[\frac{2e^{-\lambda(r-a)}}{D}\left(1+\frac{3}{\lambda r}+\frac{3}{\lambda^2 r^2}\right)-\frac{6}{\lambda^2 r^2}\right], \tag{2}$$

where $\hat{\mathbf{x}} = \mathbf{x} - \mathbf{x}_m$, $r = |\hat{\mathbf{x}}|$, and $D = 1 + \lambda a + \frac{1}{3}\lambda^2 a^2$. This Green's function corresponds to motions with frequency ω, and $\lambda = \sqrt{-i\rho\omega/\eta}$ has a positive real part. General motions can be described using Fourier or Laplace transforms. The value of $\hat{\mathbf{B}}^0$ is related to the force that the fluid exerts on the particle $\mathbf{F}_p$ [8]:

$$\hat{\mathbf{B}}^0 = -\frac{D}{\left(1+\lambda a+\frac{1}{9}\lambda^2 a^2\right)}\mathbf{F}_p = -b(\lambda a)\mathbf{F}_p\,, \tag{3}$$

where I have introduced the function $b(\lambda a)$.

If the only additional force that acts on the particle is a time-dependent random force $\boldsymbol{\xi}(t)$, then the dynamic equation for the particle velocity $\mathbf{v}_p$ is [8]

$$\rho_s V_p \frac{d\mathbf{v}_p}{dt} = \boldsymbol{\xi}(t) - \zeta\mathbf{v}_p - \frac{1}{2}\rho V_p\frac{d\mathbf{v}_p}{dt} - 6\sqrt{\pi\rho\eta}\int_{-\infty}^{t}\left(\frac{d\mathbf{v}_p}{dt}\right)_{t=s} ds\,, \tag{4}$$

where $\zeta = 6\pi\eta a$ is the Stokes drag coefficient, and the last two terms are the acceleration reaction force and the Boussinesq-Basset memory integral, respectively. The presence of the acceleration reaction force leads to an effective mass for the particle given by $m_e = (\rho_s + \frac{1}{2}\rho)V_p$, where the volume of the particle is $V_p = 4\pi a^3/3$. I will use the notion that the random forces have an expectation value given by $\langle\xi_i(t)\xi_j(t')\rangle = c^2\delta_{ij}\delta(t-t')$ and use $\mathbf{v}_p(\mathbf{x},t) = \tilde{\mathbf{v}}_p(\mathbf{x})e^{-i\omega t}$ in Eq. (4) to determine the Fourier transform of the velocity, from which the particle velocity variance is then found to be

$$\langle v_{p,i}(t)v_{p,j}(t)\rangle = \frac{c^2\delta_{ij}}{\zeta m_e}q(\alpha)\,, \tag{5}$$

where

$$q(\alpha) = \frac{4}{9\pi}\int_0^\infty \frac{y\,dy}{1+\sqrt{2\alpha}y+\alpha y^2+\frac{2\sqrt{2\alpha}}{9}y^3+\frac{4}{81}y^4} \tag{6}$$

is an integral over frequency with $y = \sqrt{\omega}$ that depends on the density of the fluid divided by the effective density of the sphere $\alpha = \rho/(\rho_s + \frac{1}{2}\rho)$. The functional form of q is shown in Fig. 1(a). I have written the variance so that it depends inversely on the effective particle mass, which is the proper mass to use for particles in fluid, as pointed out by Hinch [9]. In this form, the variance resembles what is expected from equipartition with the particle energy depending on the effective mass. We should then have that $\langle v_{p,i}v_{p,j}\rangle = kT\delta_{ij}/m_e$, where kT is thermal energy. Consequently, the inverse of q must set the variance of the force as $c^2 = \zeta kT/q$ [Fig. 1(b)]. In the limit where the fluid density is negligible, $\rho_s \gg \rho$, we get the expected value $c^2 = 2\zeta kT$. This limit gives the same result as is obtained if the memory term in Eq. (4) is ignored. As the relative density of the fluid becomes more appreciable, the variance of the force increases. For the neutrally buoyant case, $\alpha = 2/3$ and $c^2 = 6.464\zeta kT$.

I will now address the case of a suspension of these particles. While CL considered the variance in a suspension of sedimenting particles, the density difference between the fluid and the particles was

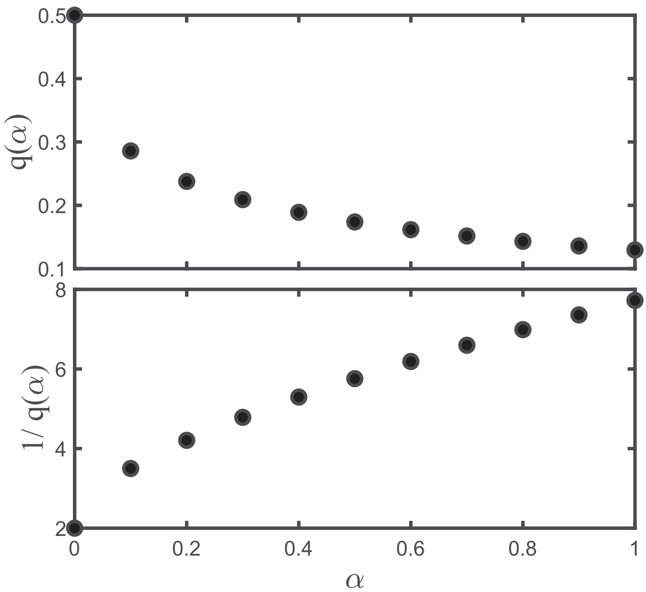

FIG. 1. The dependence of the variance of the force on α. (a) The frequency dependent integral q as a function of the relative fluid density α. (b) The inverse of q, which defines the variance of the random force.

inconsequential to their result, which leads to a similar divergence for the case where the particles are neutrally buoyant but still experiencing random fluctuations. Without loss of generalization, I will therefore consider particles of the same density as the fluid, $\rho_s = \rho$, and ask what the velocity variance is in an otherwise stationary suspension. To determine the variance, the fluid flow created by the random motions of the particles must be determined, and its effect on the rest of the suspension must be accounted for. Following the work in [10], I frame the calculation in the context of a multipole expansion of the force distribution exerted on the fluid due to the presence of the spheres. Since the equations in Eq. (1) are linear, I can use superposition to define the velocity at any point in the fluid as a sum over the disturbance velocities produced by the spheres. For a sphere centered at $\mathbf{x}_m$, the disturbance velocity at $\mathbf{x}$ can be described by a multipole expansion as

$$\mathbf{u}_m(\mathbf{x}) = \frac{1}{8\pi\eta}\sum_k \hat{\mathbf{B}}_m^k \odot \hat{\mathbf{S}}^k(\mathbf{x},\mathbf{x}_m)e^{-i\omega t}\,. \tag{7}$$

In this expression, $\hat{\mathbf{B}}_m^k$ is related to the kth moment of the force distribution on the surface of the sphere located at $\mathbf{x}_m$, and the $\hat{\mathbf{S}}^k$ are the corresponding Green's functions, with $\hat{\mathbf{S}}^0$ given by Eq. (2) and the $k \geqslant 1$ functions defined analogously to the steady Stokes' $\hat{\mathbf{S}}^k$ given in [10]. Here I am using the notation that $\odot$ represents a full contraction of $\hat{\mathbf{B}}^k$ with $\hat{\mathbf{S}}^k$, so that

$$\hat{\mathbf{B}}^k \odot \hat{\mathbf{S}}^k = \hat{B}^k_{jkl\ldots}\hat{S}^k_{ijkl\ldots}\,, \tag{8}$$

where Einstein notation has been employed. These Green's functions are the unsteady Stokes analog to the Green's functions described in [10], with the crucial one that we will need here being $\hat{\mathbf{S}}^0$ given in Eq. (2).

As before, the value of $\hat{\mathbf{B}}_m^0$ is set by the zeroth moment of the force of the fluid on the particle, as defined by Eq. (3). The strength of the antisymmetric part of the $k = 1$ multipole is set by the torque on the mth sphere. The other k-multipole strengths are set by the kth derivatives of the disturbance flows from the other particles, evaluated at the center of the mth particle [10]. For the case under consideration, where the fluid is stationary in the absence of the spheres, the velocity field the mth particle experiences is given by the disturbance flows from all the other particles (it is the full flow minus the disturbance flow from the mth particle):

$$\tilde{\mathbf{v}}_{-m}(\mathbf{x}) = \sum_{n\neq m} \tilde{\mathbf{u}}_n = \frac{1}{8\pi\eta} \sum_{n\neq m} \sum_k \hat{\mathbf{B}}_n^k \odot \hat{\mathbf{S}}^k(\mathbf{x}, \mathbf{x}_n)\,, \tag{9}$$

and the velocity at any point in the fluid is

$$\tilde{\mathbf{v}}(\mathbf{x}) = \frac{1}{8\pi\eta} \sum_n \sum_k \hat{\mathbf{B}}_n^k \odot \hat{\mathbf{S}}^k(\mathbf{x}, \mathbf{x}_n)\,. \tag{10}$$

While experiments typically measure the variance of the suspended particle velocities, there are, in fact, two separate variances that can be computed: the variance of the fluid velocity and the variance of the particle velocities. The dynamics of the full suspension depend on both of these. In the original CL calculation, the authors decomposed the particle velocities into a sedimentation velocity, a velocity due to the Stokeslets, and a correction factor required to satisfy the boundary conditions on the surface of the spheres. The term that led to the divergence in the CL variance was the contribution of the $k = 0$ multipole to the variance of the fluid velocity.

A general definition of the fluid velocity variance is the most straightforward. From Eq. (10), the general form for the variance in frequency space is

$$\langle \mathbf{v}(\mathbf{x},\omega)\mathbf{v}(\mathbf{x},\omega)\rangle = \frac{1}{(8\pi\eta)^2} \sum_{k,k'} \sum_{m,n} \left\langle \left[\hat{\mathbf{B}}_m^k \odot \hat{\mathbf{S}}^k(\mathbf{x}, \mathbf{x}_m)\right]\left[\hat{\mathbf{B}}_n^k \odot \hat{\mathbf{S}}^k(\mathbf{x}, \mathbf{x}_n)\right]^*\right\rangle_f\,, \tag{11}$$

where the * denotes the complex conjugate, since $\hat{\mathbf{B}}^k$ and $\hat{\mathbf{S}}^k$ are complex. The brackets inside the sum represent that we should perform an ensemble average of these terms over all possible, nonoverlapping positions of the spheres, and the subscript f denotes that this average should be taken only over the fluid phase. At low frequencies, the Green's functions die off like $r^{-(k+1)}$ (just like the steady Stokes' Green's functions), while at higher frequencies they die more rapidly [8]. Consequently, the fluid velocity variance is dominated by the effect from the $k = 0$ term. Using only the contribution from this multipole, and considering that the force that the fluid exerts on the mth particle is $\mathbf{F}_m(t) = \rho V_p(d\mathbf{v}_m/dt) - \boldsymbol{\xi}$, the frequency-dependent variance determined from Eqs. (3) and (11) is

$$\langle \tilde{v}_i \tilde{v}_j\rangle_f \approx \frac{9a^2 bb^* h(\omega)}{16\zeta^2} \sum_m \left\langle \hat{S}_{ik}^0(\mathbf{x}, \mathbf{x}_m)\hat{S}_{jk}^{0*}(\mathbf{x}, \mathbf{x}_m)\right\rangle_f\,, \tag{12}$$

where $h(\omega) = c^2/2\pi + (\rho V_p\omega)^2\langle \tilde{v}_{p,i}\tilde{v}_{p,j}\rangle$, and I have assumed that the velocities of the particles are uncorrelated with each other. Equation (12) shows that the fluid velocity variance depends explicitly on the particle velocity variance. Therefore, to uniquely determine the fluid velocity variance, we must calculate the particle velocity variance.

In the formalism from [10] that we are using, the particle velocity is set by equating the zeroth-order term in the Taylor series expansion of the background flow about a particle's center with the velocity produced by the zeroth-order multipole [10]. If we continue to neglect higher-order multipoles, then $\hat{\mathbf{B}}^0 = \zeta D(\mathbf{v}_p - \mathbf{v}_{-m})$, where $\mathbf{v}_p$ is the velocity of the particle with its center at $\mathbf{x}_m$. (Note that the Faxen force enters this equation through the $k = 2$ multipole, which is why it is not included here [10].) Consequently, the velocity of the mth particle satisfies

$$\tilde{\mathbf{v}}_m = \frac{\tilde{\boldsymbol{\xi}}_m}{\zeta D} + \sum_{n\neq m}\left(\frac{3a}{4\zeta}\tilde{\boldsymbol{\xi}}_n - \frac{\lambda^2 a^3}{6}\tilde{\mathbf{v}}_n\right)\cdot \hat{\mathbf{S}}^0(\mathbf{x}_m, \mathbf{x}_n). \tag{13}$$

The variance of the particle velocity is then given by

$$\left(1 + \frac{(\lambda\lambda^* a^3)^2}{36}\sum_{n\neq m}\left\langle \hat{S}_{ik}^0 \hat{S}_{jk}^{0*}\right\rangle\right)\langle \tilde{v}_{p,i}\tilde{v}_{p,j}\rangle = \frac{c^2\delta_{ij}}{2\pi\zeta^2 DD^*} + \frac{9a^2c^2}{32\pi\zeta^2}\sum_{n\neq m}\left\langle \hat{S}_{ik}^0 \hat{S}_{jk}^{0*}\right\rangle, \tag{14}$$

where I have used $\langle\tilde{\xi}_i\tilde{\xi}_j\rangle = \delta_{ij}/2\pi$ and assumed that perpendicular components of a particle's velocity are uncorrelated. The first term on the right-hand side is the single particle variance, while the second term represents the variance induced by the fluid-mediated interactions between particles. To complete the calculation, we need to determine the ensemble average of the sum on the Green's functions. For a uniform suspension of particles at volume fraction ϕ, the sum can be replaced by an integral

$$\sum_{n\neq m}\left\langle \hat{S}^0_{ik}\hat{S}^{0*}_{jk}\right\rangle = \frac{3\phi}{4\pi a^3}\int_0^\infty g(\mathbf{x}')\hat{S}^{0*}_{ik}(\mathbf{x}')\hat{S}^0_{jk}(\mathbf{x}')d^3x' \,, \tag{15}$$

with $g(\mathbf{x}')$ as the pair distribution function for the particles [11]. For a uniform distribution of particles at low density, $g(\mathbf{x}') = 0$ when $|\mathbf{x}'| < 2a$ and equals one everywhere else. The integral is then

$$\begin{aligned}\int_{|\mathbf{x}'|>2a}\hat{S}^0_{ik}(\mathbf{x}')\hat{S}^{0*}_{jk}(\mathbf{x}')d^3x' = \frac{\pi a\delta_{ij}}{3k^4DD^*}\bigg[&e^{-2k}(1+4k+8k^2+16k^3)\\ &-2\left(1+3k+4k^2+\frac{2}{3}k^3\right)e^{-k}\cos k\\ &-2k\left(1-\frac{1}{3}k-\frac{2}{3}k^2\right)e^{-k}\sin k + DD^*\bigg],\end{aligned} \tag{16}$$

where $k = \sqrt{\rho\omega a^2/2\eta}$.

To compute the variance, we need to integrate Eq. (14) over frequency. I approximate the sum in the parentheses on the left-hand side using the large frequency limit of Eq. (16), so that $\frac{(\lambda\lambda^* a^3)^2}{36}\sum_{n\neq m}\langle\hat{S}^0_{ik}\hat{S}^{0*}_{jk}\rangle \approx \phi/36$. The integral of Eq. (16) relating to frequency was computed numerically and found to be approximately $(23.7\eta/\rho a)\delta_{ij}$. The particle velocity variance to first order in ϕ using the value of c^2 corresponding to neutrally buoyant particles is then

$$\langle v_{p,i}v_{p,j}\rangle \approx \frac{kT\delta_{ij}}{m_e}(1+1.07\phi)\,. \tag{17}$$

This is the principal result of this paper. The particle velocity variance is shown to be finite and to depend on the volume fraction of the particles.

We can now finish the calculation of the fluid-phase velocity variance. From Eq. (14), the leading order contribution to the frequency-dependent particle velocity variance is $1/2\pi\zeta^2DD^*$. In addition, the fluid-phase ensemble average of the sum in Eq. (12) is

$$\frac{3\phi}{4\pi a^3(1-\phi)}\int_{|\mathbf{x}'|\geqslant a}\hat{S}^0_{ik}(\mathbf{x}')\hat{S}^{0*}_{jk}(\mathbf{x}')d^3x' = \frac{8\phi\delta_{ij}}{a^2DD^*(1-\phi)}\left(\frac{1}{9}+\frac{1}{(\lambda+\lambda^*)a}\right). \tag{18}$$

In this equation, the integral is over all distances larger than a, because particle centers must be at least a radius away from any point in the fluid. The factor of $1/(1-\phi)$ comes from fluid-phase averaging [12]. Putting all of this together and numerically integrating Eq. (12) over the frequency, the fluid-phase velocity variance is

$$\langle v_iv_j\rangle_f \approx \frac{0.528c^2\phi\delta_{ij}}{\zeta\rho V_p(1-\phi)} \approx \frac{5.11\phi kT\delta_{ij}}{m_e(1-\phi)}\,. \tag{19}$$

From this we see that the leading order contribution to the fluid velocity variance is finite and comparable to $\phi/(1-\phi)$ times the expected variance of the single particle velocity.

In retrospect, it is now clear where the CL calculation went wrong. The steady Stokes' equations are time-independent. Therefore, when CL used the Stokeslet derived from the steady Stokes' equations, all frequencies were treated equally. Indeed, the calculation done above will diverge exactly like the CL calculation if the steady Stokes' Green's functions are used. However, high-frequency solutions to the unsteady Stokes‘ equations are damped out on shorter lengthscales than the corresponding low-frequency solutions. Therefore, high-frequency motions of the spheres contribute significantly less power to the velocity variations than is predicted by the CL calculation and lead to a finite rather than infinite result. Because the variance scales inversely with fluid density, ignoring inertial effects treats the fluid density as negligible, which leads to a divergent variance in Eq. (19).

Here I have shown that standard fluid mechanics theory predicts a finite variance for the velocities of particles in suspension and that the previously found divergence came from neglecting inertia, which treats all frequencies equally, and therefore overestimates the contribution of high frequencies to the variance. Other groups have found that stratification in a suspension can also lead to finite fluctuations [13]. The theory presented here, though, does not require any special particle distributions to produce finite results. Experiments observe that, for small system sizes, ones where the system size $L < 20a\phi^{-1/3}$, the variance does grow with system size, but for systems larger than this, the variance is independent of size [1]. At these larger system sizes, the variance increases with the particle volume fraction. Some experiments suggest that these fluctuations may increase as $\phi^{2/3}$ [1], whereas the theory here predicts linear dependence. This discrepancy between the theory presented here and the previous experiments may stem from the manner in which many of the experiments were carried out. Of all the experimental data, Segré *et al.* (1997) [14] provides the strongest evidence for the two-thirds power law (see Fig. 4 in [1]). However, these experiments used Particle Image Velocimetry (PIV) to measure the sedimenting particle velocities. In PIV, two subsequent images are compared to generate the velocity field. About specific pixels in the first image, an interrogation window is defined. This window is then rastered over the subsequent image, and the maximum in the cross-correlation between the initial window image and the rastered window defines the average displacement of the particles in that window. Consequently, PIV measures the local spatial average of the particle velocity. The variance of that velocity is then the variance of a spatially averaged velocity field, not the variance of the individual particles. In [14], the authors mention that each velocity vector was the average over 2 to 4 particles. Keeping the same number of particles in the interrogation window as the volume fraction was being changed requires that the length of each side of the interrogation windows be scaled roughly as $1/\phi^{1/3}$. Since the fluid velocity produced by a moving particle dies off roughly as $1/r$, the variance of the spatially averaged velocity would likely scale like $\phi^{2/3}$, as these authors found. Consistent with this view, the vertical velocity variance of suspensions of rising bubbles, which was measured using a dual impedance probe providing a much more local measure of the bubble velocity, was found to increase approximately linearly with volume fraction [15], in rough agreement with the results given in Eq. (17). The normalized bubble velocity variance was found to increase approximately as 0.44ϕ in these experiments. The same authors also measured the vertical variance of the fluid velocity and found that it increased approximately like 2ϕ. This result agrees well with what we find in Eqs. (17) and (19), which predict that the dependence of the fluid velocity variance on volume fraction should be about five times larger than that for the variance of the particle velocity, although this result was for neutrally buoyant particles, not bubbles at higher Reynolds number. New experiments are needed to determine how well the theory presented here predicts the variances of the fluid and particle velocities in suspensions. In order to avoid potential artifacts from averaging, single-particle tracking methods should be used. Since I used the zeroth order approximation to the pair distribution function, the scaling found here is expected to deviate from experiments at larger volume fractions. If the pair-distribution function's dependence on volume fraction is known, then Eq. (15) can be used to predict the variance over a larger range of particle densities.

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