Resolving a paradox of anomalous scalings in the diffusion of granular materials

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Granular materials do not perform Brownian motion, yet diffusion can be observed in such systems when agitation causes inelastic collisions between particles. It has been suggested that axial diffusion of granular matter in a rotating drum might be “anomalous” in the sense that the mean squared displacement of particles follows a power law in time with exponent less than unity. Further numerical and experimental studies have been unable to definitively confirm or disprove this observation. We show two possible resolutions to this apparent paradox without the need to appeal to anomalous diffusion. First, we consider the evolution of arbitrary (non-point-source) initial data towards the self-similar intermediate asymptotics of diffusion by deriving an analytical expression for the instantaneous collapse exponent of the macroscopic concentration profiles. Second, we account for the concentration-dependent diffusivity in bidisperse mixtures, and we give an asymptotic argument for the self-similar behavior of such a diffusion process, for which an exact self-similar analytical solution does not exist. The theoretical arguments are verified through numerical simulations of the governing partial differential equations, showing that concentration-dependent diffusivity leads to two intermediate asymptotic regimes: one with an anomalous scaling that matches the experimental observations for naturally polydisperse granular materials, and another with a “normal” diffusive scaling (consistent with a “normal” random walk) at even longer times.

transport processes | granular flow

Granular materials are processed, mixed, separated, or otherwise manipulated for the purposes of a wide array of industries: agriculture, ceramics, chemicals, energy, manufacturing, minerals and ores, pharmaceuticals, and metallurgy, to name a few. It is estimated that the processing of particulate materials accounts for 10% of all the energy consumed on the planet (1). Despite the practical implications, the dynamics of flowing granular materials are poorly understood because they constitute a complex system away from equilibrium and so are capable of both chaotic dynamics and self organization (2). *Science* magazine lists the physics of granular matter as one of 125 unanswered questions in science at the beginning of the 21st century (3). When it comes to powder mixing processes in which segregation is not significant, diffusion is one of the fundamental mechanisms that achieves homogenization (4). Though granular materials consist of macroscopic particles that do not perform thermally driven Brownian motion, self and collective diffusion in the flow of groups of particles can arise from agitation processes that lead to nonequilibrium velocity fluctuations (5–9).

In particular, granular flow in a long rotating horizontal drum has proven to be a simple but important system to understand (10). Recently, it has been suggested that axial diffusion in this system is *anomalous* (11). Here, “anomalous diffusion” refers to a class of transport phenomena in which the mean squared displacement of particles follows a power law in time with exponent different from unity. A common mathematical approach to such processes is the framework of *fractional* partial differential equations that feature noninteger order derivatives (12). In the last decade, anomalous diffusion has been observed or suggested to be present in phenomena as diverse as tracer motion in chaotic

laminar flows (13), drainage of particulates from a silo (14), protein and chromosome transport through cellular membranes (15, 16), shaken granular media (17), motion of colloids in entangled actin networks (18), flow through disordered porous media (19) and across human transportation networks (20). Here, we focus on an apparent paradox of anomalous diffusion in agitated granular materials, and propose an explanation for the experimental observations without resorting to the framework of fractional partial differential equations.

The mean squared displacement of a tracer particle is one of the most reliable diagnostics for anomalous transport (21), though it has been argued that even more fundamental measures are required to distinguish anomalous diffusion from other physical processes (22). In the case of axial diffusion of grains in a tumbler (11), due to the opaque nature of the granular materials, measuring individual particle trajectories is a difficult task, which only recently was achieved using high-intensity synchrotron x-rays (23). Thus, in the rotating horizontal drum in (11), the cross-sectionally averaged concentration profile of dyed grains was instead measured by an optical projection technique, and the assessment of the diffusion process was made on the basis of the macroscopic theory by collapsing the axial concentration profiles $c(z, t)$ at different times t .

As early as 1954 it was proposed (24) that the concentration profiles of axially diffusing granular materials in a tumbler can be modeled by Fick’s laws, the first of which states, in one dimension, that the particle flux q is proportional to the gradient of concentration of particles (25), specifically $q = -D\partial c/\partial z$, where z is the axial coordinate, and D is the diffusivity, which can, in principle, depend on c and/or z . Conservation of mass, $\partial c/\partial t + \partial q/\partial z = 0$, then leads to the diffusion equation

$$\frac{\partial c}{\partial t} = \frac{\partial}{\partial z} \left(D(c, \dots) \frac{\partial c}{\partial z} \right), \quad [1]$$

which can also be derived as the continuum description of point particles undergoing a random walk with mean squared axial displacement $\langle (\Delta z)^2 \rangle \propto Dt$ (26, 27). For decades this macroscopic approach has been the accepted engineering model for granular diffusion (28–30). Moreover, Barenblatt’s *intermediate asymptotics* theory (31) indicates that solutions to Eq. 1 for arbitrary initial conditions under the requirement that $\int c(z, t) dz$ is constant will eventually converge to the self-similar regime in which $c(z, t) \simeq \mathfrak{C}(z/t^\alpha) t^{-\alpha}$ for some invariant profile $\mathfrak{C}(\cdot)$ and collapse exponent α ; in the linear case with D constant, $\alpha = 1/2$.

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However, the concentration profiles from the experiments in ref. (11) collapse under the rescaling $c(z, t) \rightarrow c(z/t^\alpha, t)t^\alpha$ for $\alpha \approx 0.3 < 1/2$, which does not appear to agree with Eq. 1 but is consistent with the fractional diffusion equation governing a *subdiffusive* process with $\langle(\Delta z)^2\rangle^{1/2} \propto t^{0.3}$ (12). Subsequent numerical simulations of the axial diffusion of granular materials through particle dynamics using the discrete element method (DEM) (32–34) found that the mean squared displacement of individual particles grows (asymptotically) linearly with time, consistent with normal (Fickian) diffusion; i.e., $\langle(\Delta z)^2\rangle \propto t$. X-ray measurements of individual particle trajectories in the most recent experiments (23) are in agreement with the simulations. Thus, there is a clear paradox between the microscopic and macroscopic theories of axial diffusion of granular materials.

Another set of experiments (35, 36) and DEM simulations (37) have provided some evidence for a concentration-dependent diffusivity in bidisperse mixtures, i.e., granular materials consisting of two kinds of particles differing either in size, density, shape, roughness, etc. Specifically, it was found that a band of larger particles diffuses anomalously in a background of smaller particles. A nonlinear normal diffusion model, specifically Eq. 1 with $D(c) \propto c$, was also proposed in ref. (11) because it possesses an exact self-similar solution of the form $c(z, t) = \mathcal{C}(z/t^\alpha)t^{-\alpha}$ with $\alpha = 1/3 < 1/2$ (38, 39). On this basis, it has been argued (36, 40) that concentration-dependent diffusivity of this form could explain the anomalous scalings in ref. (11), at least for bidisperse mixtures. However, the shape of the concentration profile predicted using this equation was found to be inconsistent with the experimental data in ref. (11) because of the behavior of the tails of the distribution. Thus, though there is considerable evidence that granular materials in tumblers should diffuse according to Fick's laws (or a nonlinear, concentration-dependent generalization of the latter), there is no clear theoretical understanding of why the experiments in ref. (11) do not support this type of macroscopic theory. The apparent paradox remains unresolved (unless one discards either the experimental or computational data), despite a growing number of numerical (34) and experimental (23) studies.

In this paper, we show that the paradox can be resolved in two ways: (i) by considering the evolution of arbitrary initial data towards the self-similar intermediate asymptotics of diffusion, and (ii) by accounting for concentration-dependent diffusivity of bidisperse mixtures in a different way than ref. (11). While (ii) provides a better explanation for the experiments in ref. (11), which were performed with either salt or sand as the granular material (each of which is naturally polydisperse), the result in (i) lays the theoretical foundations for preasymptotic anomalous collapse exponents for normal diffusion. This idea has broad implications for all diffusive transport phenomena being interrogated through their macroscopic (concentration) rather than microscopic (mean squared displacement) manifestations.

Model

Axial Diffusion of a Granular Material in a Tumbler. In any axial cross-section of a long rotating drum (length $2L$), the granular flow is composed of a thin surface shear layer (the *flowing layer*) in which material flows quickly down the slope (see white arrows in Fig. 1). Particles below it (the *fixed bed*) are in static equilibrium and perform solid body rotation in unison with the container (41–43). This flow rapidly reaches steady state (10, 42), at which point, to a good approximation, the volume fractions and macroscopic (averaged) densities of the material in the flowing layer and the bulk are constant and equal. For a monodisperse granular material, all particles are identical, and c is the concentration of the tagged particles (e.g., black in color), while $1 - c$ is the concentration of the nontagged particles (e.g., white in color). Following (24, 28), we consider a partially filled drum and appeal to Fick's laws to obtain an axial diffusion equation like

Fig. 1. Illustration of a partially filled cylindrical tumbler of length $2L$ rotating about its central axis (the z -axis) with angular velocity ω . The initial condition for the axial diffusion process is shown as the band of width $2l$ of dark ("tagged") granular material in the middle. Above the tumbler, an example concentration profile $c(z, t)$ of the tagged particles at three different times (lighter curves corresponding to later times) is depicted.

Eq. 1 for the cross-sectionally averaged concentration, where D is now an *axial* collective diffusivity* that can differ from the bare collective diffusivity because flow and hence velocity fluctuations only occur in the thin surface layer (29). To complete the problem statement, an initial distribution $c(z, 0) = c_i(z)$ is specified, and no-flux boundary conditions ($\partial c / \partial z = 0$ at $z = \pm L$) are applied at the ends of the domain $z \in [-L, +L]$ because the solid tumbler endwalls do not allow particles to leave.[†]

It is useful to introduce dimensionless variables by using the tumbler half-length L as the length scale and the diffusion time L^2/D_0 as the time scale:

$$Z = z/L, \quad T = t/(L^2/D_0), \quad C = c/C_0, \quad \mathcal{D} = D/D_0, \quad [2]$$

where C_0 is an arbitrary normalization factor so that $0 \leq C \leq 1$, D_0 is the typical experimentally determined magnitude of the diffusivity. Using the dimensionless variables from Eq. 2, Eq. 1 becomes

$$\frac{\partial C}{\partial T} = \frac{\partial}{\partial Z} \left(\mathcal{D} \frac{\partial C}{\partial Z} \right). \quad [3]$$

A different nondimensionalization is required for an infinite domain. In order to compare our results to experiments, we find it useful to think in terms of a finite cylinder, even if boundary effects are not important for parts of the discussion below.

Solution for Constant $\mathcal{D}$ and a Pulse Initial Condition. In axial diffusion experiments, it is common to take a symmetric pulse of width $2l$ as the initial condition, hence $C_i(Z) = H(Z + \ell) - H(Z - \ell)$, where $H(\cdot)$ is the Heaviside step function and $\ell = l/L$. The corresponding solution to Eq. 3 with $\mathcal{D} = 1$ subject to the finite-domain no-flux boundary conditions is well known [(44): section 2.2]:

$$C(Z, T) = \frac{1}{2} \sum_{n=-\infty}^{+\infty} \left[\operatorname{erf} \left(\frac{Z + \ell - 2n}{\sqrt{4T}} \right) - \operatorname{erf} \left(\frac{Z - \ell - 2n}{\sqrt{4T}} \right) \right]. \quad [4]$$

The special case of $n = 0$ (i.e., no summation) gives the solution for the boundary conditions $\lim_{|Z| \rightarrow \infty} C(Z, T) = 0$ (e.g., $L \rightarrow \infty$ but ℓ finite), which in turn has the early-time asymptotic expansion

*Note that collective diffusivity is distinct from the particle diffusivity, i.e., the proportionality constant between the mean squared displacement and time during self diffusion.

[†]On the other hand, if we are only concerned with early times, at which the boundary effects are negligible, then we could simply use the asymptotic boundary conditions $\lim_{|z| \rightarrow \infty} c = 0$.

$$C(Z, T) = H(Z + \ell) - H(Z - \ell) + \frac{1}{\sqrt{\pi}} \left\{ \exp \left\{ -\frac{(Z + \ell)^2}{4T} \right\} \left[-\frac{\sqrt{T}}{Z + \ell} + \mathcal{O}(T^{3/2}) \right] - \exp \left\{ -\frac{(Z - \ell)^2}{4T} \right\} \left[-\frac{\sqrt{T}}{Z - \ell} + \mathcal{O}(T^{3/2}) \right] \right\} \quad (T \rightarrow 0). \quad [5]$$

Clearly, there is *no* self-similar rescaling of the form $C(Z, T) \rightarrow C(Z/\sqrt{T}, T)\sqrt{T}$ that holds for all Z because we cannot express $(Z \pm \ell)^{-1}$ as $Z^{-1}(1 \pm \ell/Z)^{-1} \simeq Z^{-1}(1 \mp \ell/Z) \simeq Z^{-1}$ unless $|Z| \gg \ell$. However, for late times, Eq. 4 with $n = 0$ can be expanded as

$$C(Z, T) = \frac{\ell}{\sqrt{\pi T}} \left(1 - \frac{Z^2}{4T} \right) + \mathcal{O}(T^{-3/2}) \quad (T \rightarrow \infty). \quad [6]$$

Unlike the case in Eq. 5, this asymptotic solution profile is an invariant function of *only* the similarity variable $\xi = Z/\sqrt{T}$ under the normal diffusive rescaling $C(Z, T) \rightarrow C(Z/\sqrt{T}, T)\sqrt{T}$, if T is large enough. This feature is in agreement with the theory of intermediate asymptotics (31), which states that all solutions to Eq. 3 for arbitrary initial conditions evolve into a self-similar profile such as in Eq. 6. For the $n = 0$ case of Eq. 4, it has been suggested (33) that there is some delay until this convergence occurs. We now show how this delay, and also the approximate collapse exponent for times prior to reaching the intermediate asymptotic regime, can be calculated.

Results and Discussion

Transition to the Self-Similar Regime for Constant $\mathcal{D}$. Zel'dovich and Barenblatt (45) proposed that solutions to Eq. 3 starting from arbitrary initial data $C_i(Z)$ can be well approximated (for $\mathcal{D} = 1$ and $T > T^*$) by the point-source similarity solution

$$C(Z, T) = \frac{1}{\sqrt{4\pi(T + T^*)}} \exp \left\{ -\frac{(Z + Z^*)^2}{4(T + T^*)} \right\} \quad [7]$$

as long as the constants Z^* and T^* are chosen by requiring that the solution in Eq. 7 be optimal in an appropriate sense (45–48).[‡] It can be shown that Z^* and T^* depend only on the initial data and are given by the explicit formulæ

$$Z^* = -\frac{M_1}{M_0}, \quad T^* = \frac{1}{2} \left[\frac{M_2}{M_0} - \left(\frac{M_1}{M_0} \right)^2 \right], \quad [8]$$

where $M_k := \int_{-\infty}^{\infty} C_i(Z) Z^k dZ$ is the k th moment of the initial distribution.[§] For the step function initial condition of width 2ℓ , i.e., $C_i(Z) = H(Z + \ell) - H(Z - \ell)$, we have $M_0 = 2\ell$, $M_1 = 0$ and $M_2 = 2\ell^3/3$, which gives $Z^* = 0$ and $T^* = \ell^2/6$. Note that $T^* < 1$ (and typically $T^* \ll 1$) because we introduced a characteristic length scale in our nondimensionalization (recall Eq. 2). Physically, T^* corresponds approximately to the time needed for a given initial condition to evolve into the self-similar intermediate asymptotic profile, while Z^* is a spatial shift to account for possible asymmetry in the initial condition. Put another way, this approach determines T^* and Z^* such that the instan-

Fig. 2. Evolution of the scaling/collapse parameter α [Eq. 9] calculated from the optimal similarity solution as a function of time T for different initial pulse half-widths ℓ . Thin vertical lines denote the time-shift $T^* = \ell^2/6$ corresponding to the given value of ℓ . All curves approach $\alpha = 0$ at $T = 0$ slowly, so the graph is truncated at $T = 10^{-10}$. Note that the abscissa is logarithmic; the arrow indicates the direction of increasing ℓ .

taneous release of a point source at $T = -T^*$ and $Z = Z^*$ leads to a profile that approximates (for $T > T^*$) the solution for some non-point-source initial data specified at $T = 0$ (45, 46).

Thus, we cannot expect that the solution to Eq. 3 (with $\mathcal{D} = 1$) starting from non- δ -function initial data can be rescaled into an invariant profile for $T < T^*$. Nevertheless, suppose that $T = \mathcal{O}(T^*)$; i.e., the solution is “on the verge” of converging to the self-similar intermediate asymptotics but has not quite yet. Hence, if we were to try and succeed at collapsing the solution curves under the rescaling $C(Z, T) \rightarrow C(Z/T^\alpha, T)T^\alpha$ for some $\alpha > 0$, then according to Eq. 7 we must have balanced T^α with $\sqrt{T + T^*}$. In other words, α must be such that $T^\alpha \simeq \sqrt{T + T^*}$ or

$$\alpha(T) \simeq \frac{\ln(T + T^*)}{2 \ln T} = \begin{cases} \frac{1}{\ln T} \left[\ln T^* + \frac{T}{2T^*} + \mathcal{O}(T^2) \right], & T \rightarrow 0, \\ \frac{1}{2} + \frac{1}{\ln T} \left[\frac{T^*}{2T} + \mathcal{O}(T^{-2}) \right], & T \rightarrow \infty. \end{cases} \quad [9]$$

This equation provides an analytical expression for the *instantaneous collapse exponent* of the concentration profile. Fig. 2 shows $\alpha(T)$ for several choices of the half-width ℓ of the initial condition. For $T \gg T^*$, $\alpha \rightarrow 1/2$ as expected under the interpretation of intermediate asymptotics. However, for T on the order of T^* or smaller, there is an explicit time dependence in the presumed scaling exponent α .

Thus, let us consider now the behavior near $T = T^*$. From Eq. 9, $\alpha(T^*) = \ln(2T^*)/(2 \ln T^*)$, which possesses the limiting value of $1/2$ as $T^* \rightarrow 0$ (i.e., the limit in which the optimal self-similar solution is the exact solution). However, because $\alpha(T^*) \sim 1/2 + \ln 2/(2 \ln T^*)$ as $T^* \rightarrow 0$, it is clear that α quickly decreases from the limiting value of $1/2$ even for very small T^*

Fig. 3. Instantaneous collapse exponent $\alpha(T)$ at $T = T^*$ (solid) and the average collapse exponent (dashed) for times up to T^* , both shown as functions of T^* .

[‡]The general series solution to Eq. 3 ($\mathcal{D} = 1$) is $C(\eta, T) = e^{-\eta^2} \sum_{n=0}^{\infty} (T + T^*)^{-(n+1)/2} \mathcal{A}_n \mathcal{H}_n(\eta)$, where $\mathcal{H}_n(\cdot)$ is the n th Hermite polynomial, and $\eta = (Z + Z^*)/\sqrt{4(T + T^*)}$ is a similarity variable (47, 48). Eq. 7 is the first term in the series, and Z^* and T^* are chosen so that $\mathcal{A}_1 = \mathcal{A}_2 = 0$.

[§]If $C_i(Z) = \delta(Z)$, then $Z^* = T^* = 0$ and Eq. 7 is the exact self-similar solution for all T . If $C_i(Z)$ is an odd function, then $M_0 = 0$, and Eq. 8 must be modified (47, 48). This special case is not relevant to our discussion because the concentration is such that $0 \leq C \leq 1$ for all Z and T .

(see solid curve in Fig. 3). Thus, an attempt to collapse the concentration curves under a rescaling $C(Z, T) \rightarrow C(Z/T^\alpha, T)T^\alpha$ at times T such that $\alpha(T)$ has not reached the asymptotic regime can result in the observation of anomalous exponents, though the underlying process is ordinary diffusion. This result, which we highlight throughout the remainder of the paper, is one of the main points of our mathematical analysis.

From Fig. 2 we can also understand the effect of the localization of the initial condition. For larger ℓ (i.e., for less localized initial conditions) the convergence to the self-similar intermediate asymptotic scaling takes longer. Additionally, the observed collapse exponent at $T = T^*$ is increasingly different from $1/2$ for larger ℓ , meaning that one should wait *longer than* T^* before attempting to determine the approach to the expected diffusive scaling. The exponent α is always less than $1/2$ for $T = T^*$, so only *subdiffusive* scalings are observed for $T \lesssim T^*$. Furthermore, we find that the range $0.3 \lesssim \alpha \lesssim 0.4$ (as found in the experiments in ref. (11)) is robust over four orders of magnitude of ℓ . When collapsing experimental concentration profiles across a range of (early) times, one is not observing the instantaneous collapse exponent but rather some average value. Therefore, we also consider the running average of $\alpha(T)$ up to T^* , as a function of T^* . Fig. 3 shows the plot of this integral computed numerically. The range of the average of α is much narrower and better localized between 0.3 and 0.4, which is precisely within the margins of error of ref. (11).

The generic nature of the anomalous collapse exponents for $T < T^*$ can be confirmed easily by attempting to (numerically) collapse the solution from Eq. 4 (full summation) evaluated at some given times. Fig. 4 shows the result for (A) early and (B) late times. The collapse exponent used is selected to ensure a “good collapse” (see also Figs. S1–S3 in the *SI Text* and the discussion therein). Clearly, the best early time collapse exponent is ≈ 0.3 and, thus, would be interpreted as anomalous. Meanwhile, for late times up to $10T^*$, the best α is much closer to (but still smaller than) $1/2$, as expected from our theory.

Table 1 shows representative parameters from the recent experimental (11) and computational (32) investigations of axial diffusion of a granular pulse in a tumbler. We see that in both cases, the earliest times considered in the collapse are less than or on the order of T^* , meaning the profile has not fully converged to the self-similar intermediate asymptotics. This feature explains, for example, why the two earliest profiles in the DEM simulations in [(32); Fig. 4] show distinctly different collapse than the rest. Nevertheless, it is clear that the discrepancy in scalings is only partially accounted for by the scaling theory for a constant diffusivity that we have developed. Thus, we now examine the

Fig. 4. Solution to Eq. 3 with $\mathcal{D}(C) = 1$, $C(Z, 0) = H(Z + \ell) - H(Z - \ell)$ and $\ell = 0.05$ for times up to $10T^*$, where T^* is computed from Eq. 8. (A) Early times approximate collapse (10 curves corresponding to equispaced time points in $[0, 5T^*]$) and (B) late times approximate collapse (10 equispaced time points in $[5T^*, 10T^*]$). The collapse exponent changes with time but settles on a value near $\alpha \approx 1/2$ at late times. Dark profiles corresponds to early times, while lighter profiles corresponds to late times.

effects of bidispersity of the granular materials considered in (11, 32).

Bidisperse Case ($\mathcal{D} = \mathcal{D}(C)$). Accounting for polydispersity is important because even the “monodisperse” experiments in ref. (11) were performed using salt grains with particle diameters ranging from 0.300 mm to 0.420 mm (see Table 1). In addition, the ratio of large-to-small particle diameters used in the DEM simulations is typically 2, which further suggests the need to account for the influence of bidispersity. For a binary granular mixture consisting of particles of two different sizes, recent experiments (36) and simulations (34) have suggested that the diffusivity depends on the local concentration of, e.g., the smaller particles.[†] Fischer, et al. (36) attributed the latter effect to the larger momentum of the larger particles, which leads to enhanced lateral diffusivity for those particles experiencing more collisions with the larger particles. For example, it was suggested that $\mathcal{D}(C) = C^n$, where n is a positive integer, because Eq. 3 with $\mathcal{D}(C) = C^n$ possesses compact self-similar solutions with $\alpha = 1/(n + 2)$ (38, 39), which gives $\alpha = 1/3$ for $n = 1$. Though this value of α is close to the experimental collapse exponent, the experimental concentration distribution is observed to have long exponential tails (11), which is in conflict with the behavior of the compact solutions for $\mathcal{D}(C) = C^n$ that go to zero at finite z as a power function (38, 39).

Previously, Ristow and Nakagawa (35) proposed that $\mathcal{D}(C) = 1 \pm (C - 1/2)$ so that $\int_0^1 \mathcal{D}(C) dC = 1$ and there is no concentration dependence for a perfect mixture (i.e., $\mathcal{D}(1/2) = 1$). Neither experiments (35, 36) nor DEM simulations (34, 37) provide sufficiently well resolved data to determine the functional form of $\mathcal{D}(C)$ with any certainty or whether to pick the “+” or “−” sign in the proposed expression. Nevertheless, a concentration-dependent axial diffusivity with $\mathcal{D}(0) = 0$ (as in refs. (11, 36)) is not consistent with the theory of the monodisperse case because, even in the absence of small particles, the large particles self diffuse (and vice versa). In light of the discussion in ref. (36), the form $\mathcal{D}(C) = 1 \pm (C - 1/2)$ can be reinterpreted: if $C = C_l$ is the concentration of large particles then the “+” sign should be chosen because a locally higher than average concentration of large particles leads to enhanced lateral diffusivity beyond that of a perfect mixture ($C = 1/2$) for all nearby particles. Meanwhile, the opposite must be true for the concentration of small particles, namely $C_s = 1 - C_l$. Indeed, $\mathcal{D}(1 - C_s) = 1 + (1 - C_s - 1/2) = 1 - (C_s - 1/2)$, which shows the self consistency of the model and its interpretation.

Hence, a functional form $\mathcal{D}(C) = 1 + F(C)$, for some F such as in ref. (35), is not only reasonable but, as we now show, may be able to rationalize the experimental observations. We seek a self-similar solution of Eq. 3 with $\mathcal{D}(C) = 1 \pm (C - 1/2)$ under the transformation $Z = T^\alpha \zeta$, and $C(Z, T) = T^{-\alpha} \mathfrak{C}(\zeta)$, then Eq. 3 becomes

$$0 = \mathfrak{C} + \zeta \mathfrak{C}' + \underbrace{\frac{(2\mp 1)}{2\alpha} T^{1-2\alpha} \mathfrak{C}''}_{\textcircled{1}} + \underbrace{\frac{1}{2\alpha} T^{1-3\alpha} (\mathfrak{C}^2)''}_{\textcircled{2}}. \quad [10]$$

Clearly, there is no single self-similar solution $\mathfrak{C}$ to Eq. 10 that describes the intermediate asymptotics of this diffusion process because there is no unique choice of α that removes the explicit dependence on the time-variable T . Curiously, exact *traveling-wave solutions* [i.e., $C(Z, T) = \mathfrak{C}(Z - \lambda T)$ for some λ] to Eq. 3 with $\mathcal{D}(C) = 1 + \varepsilon C$ have been constructed in the context of heat

[†]In a bidisperse mixture, radial segregation occurs within a few revolutions of the drum, leading to the formation of a core of small particles, which then diffuses axially. Axial segregation typically takes place on a time scale much longer than those of radial segregation and axial diffusion, thus it is negligible for the purposes of our discussion (11, 35, 36).

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