

Intermittent Distribution of Inertial Particles in Turbulent Flows

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We consider inertial particles suspended in an incompressible turbulent flow. Because of particles' inertia their flow is compressible, which leads to fluctuations of concentration significant for heavy particles. We show that the statistics of these fluctuations is independent of details of the velocity statistics, which allows us to predict that the particles cluster on the viscous scale of turbulence and describe the probability distribution of concentration fluctuations. We discuss the possible role of the clustering in the physics of atmospheric aerosols, in particular, in cloud formation.

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When observing air bubbles in water or dust in air, one often notices that inertial particles are not distributed homogeneously in a flow. This is used for flow visualization. If the flow is turbulent, the concentration of the suspended particles fluctuates. Here we develop a statistical theory of this phenomenon based on a Lagrangian description of turbulence (see [1–3] and references therein). We describe the initial growth of concentration fluctuations from a uniform state and its saturation due to finite-size effects, imposed either by the Brownian motion or by a finite distance between the particles.

To illustrate the fact that the flow of inertial particles is compressible, consider a spherical particle so small that the flow around it is viscous. The particle's velocity \mathbf{v} is related to the fluid velocity \mathbf{u} by the equation $d\mathbf{v}/dt - \beta d\mathbf{u}/dt = (\mathbf{u} - \mathbf{v})/\tau_s$. We defined $\beta = 3\rho/(\rho + 2\rho_0)$ and the Stokes time $\tau_s = a^2/(3\nu\beta)$, where a is the radius of the particle, ρ and ρ_0 are densities of the ambient fluid and the particle, respectively [4,5]. Both \mathbf{v} and \mathbf{u} are evaluated along the particle's trajectory $\mathbf{q}(t, \mathbf{r})$ that satisfies $\partial_t \mathbf{q} = \mathbf{v}$ and $\mathbf{q}(0, \mathbf{r}) = \mathbf{r}$. Since $a \ll r_v$, where r_v is the viscous scale of the flow, then one can solve the system for \mathbf{v} and \mathbf{q} perturbatively in a small parameter $(\beta - 1)\tau_s/\tau_v = (\beta - 1)a^2/\beta r_v^2$:

$$\mathbf{v} = \mathbf{u} + (\beta - 1)\tau_s[\partial_t \mathbf{u} + (\mathbf{u} \cdot \nabla) \mathbf{u}]. \quad (1)$$

The velocity field $\mathbf{v}(t, \mathbf{r})$ of spatially distributed particles is compressible even if the fluid flow is incompressible [5]: $(\nabla \cdot \mathbf{v}) = (\beta - 1)\tau_s \nabla[(\mathbf{u} \cdot \nabla) \mathbf{u}]$. The effect of the (small) compressible component of the \mathbf{v} flow can be enhanced by large parameters (Reynolds and Schmidt numbers).

Particle concentration satisfies the diffusion-advection equation with the diffusivity κ due to Brownian motion:

$$\partial_t n + \nabla(\mathbf{v}n) - \kappa \nabla^2 n = 0. \quad (2)$$

Every particle produces a perturbation of the flow that decays as an inverse distance from the particle. Since

particles move coherently within the viscous scale r_v , the condition $a \int_a^{r_v} n(r)r^{-1}d^3r \approx nar_v^2 \ll 1$ has to be satisfied to neglect their interaction. This condition is more restrictive than $nar_v^3 \ll 1$. If $nar_v^2 \ll 1$, the concentration field can be considered passive; i.e., \mathbf{v} is independent of n in Eq. (2). We consider velocity \mathbf{v} as an arbitrary random field with a statistics stationary in time and homogeneous in space. We presume only that velocity has finite temporal correlations and is spatially smooth below the viscous scale r_v . Velocity gradients produce inhomogeneities in the concentration while diffusion tends to smooth it out. Comparing the second and the third terms in Eq. (2) one concludes that velocity gradient λ dominates the motion at the scales larger than the diffusion scale, $\sqrt{\kappa/\lambda}$. Diffusion makes the field n smooth at scales smaller than $\sqrt{\kappa/\lambda}$. The account of diffusion is equivalent to the consideration of concentration coarse-grained over the diffusion scale [6]. Note that the diffusivity of macroscopic particles is usually much smaller than the viscosity of the ambient fluid (the Schmidt number ν/κ is large) so the diffusion scale is much smaller than the viscous scale: $\sqrt{\kappa/\lambda} \ll r_v \approx \sqrt{\nu/\lambda}$. Since $\sqrt{\kappa/\lambda}$ can be even smaller than the distance between particles, we define the scale of coarse-graining r_d as the largest between $\sqrt{\kappa/\lambda}$ and $n^{-1/3}$. We neglect the fluctuations of diffusion scale because they do not change the dependence of the concentration on large parameters, which are either time in the transient regime or Reynolds and Schmidt numbers in the steady state.

Evolution of an arbitrary initial condition $n(0, \mathbf{r})$ according to Eq. (2) ultimately results in a unique steady state of the concentration fluctuations [6]. Without any loss of generality, we consider a homogeneous in space initial concentration choosing the units so that $n_0 = 1$. Let us describe the initial stage of the fluctuation growth. The term $n_0(\nabla \cdot \mathbf{v})$ in Eq. (2) can be viewed as a source producing fluctuations on the scale r_v . At $t \leq \lambda^{-1} \ln(r_v/r_d)$ those fluctuations have not yet compressed down to r_d so that diffusion (and the coarse-graining) is irrelevant. We call this stage the ideal case. In the Lagrangian frame, Eq. (2) then becomes the ordinary differential equation

$dn/dt = -n(\nabla \cdot \mathbf{v})$. Here $(\nabla \cdot \mathbf{v})$ is a function of time, which fluctuates in a random flow. If the Lagrangian correlation time of the fluid velocity, \mathbf{u} , is finite, which is true for most flows of interest, then $(\nabla \cdot \mathbf{v}) \propto \nabla[(\mathbf{u} \cdot \nabla)\mathbf{u}]$ has also a finite correlation time, τ . At $t \gg \tau$, the concentration logarithm, $X(t) \equiv \ln[n(t)/n(0)] = -\int_0^t (\nabla \cdot \mathbf{v}) dt'$, is a sum of a large number of random variables. The theory of large deviations [7] assures that the probability density function (PDF) has the form $\mathcal{P}(X) \propto \exp[-ts(X/t)]$, where s is a non-negative convex function. To calculate the moments of the concentration in the Eulerian frame one has to take every Lagrangian element with its own weight proportional to its volume, i.e., to the inverse concentration:

$$\langle n^\alpha(t, \mathbf{r}) \rangle \propto \int dX \exp[(\alpha - 1)X - ts(X/t)]. \quad (3)$$

At large times, this integral can be found using the saddle-point approximation. The saddle-point X_α is given by $s'(X_\alpha/t) = \alpha - 1$, which implies $X_\alpha \propto t$. Hence the moments generally behave exponentially in time: $\langle n^\alpha(t) \rangle \propto \exp[\gamma(\alpha)t]$.

Let us show that the conclusion on exponential behavior of moments is enough to establish the most interesting properties of this stage of evolution. The number of particles is conserved; i.e., $\langle n \rangle$ is time independent. Hence $\gamma(1) = 0$. It is also obvious that $\gamma(0) = 0$. Because of the Hölder inequality, the function $\gamma(\alpha)$ is convex. Therefore, it follows that $\gamma(\alpha)$ is negative for $0 < \alpha < 1$ and positive otherwise. Low-order moments decay, whereas high-order and negative moments grow. The decay rate is $\langle \log|n| \rangle/t = d\gamma(\alpha)/d\alpha|_{\alpha=0} < 0$; i.e., n decays almost everywhere. Since the mean concentration is conserved, n has to grow in some (smaller and smaller) regions, which implies growth of high moments. The growth of passive scalar fluctuations in the case of a short-correlated in time compressible flow has been described in [8].

Let us now give a more formal analysis which includes the account of coarse-graining and describes the saturation of the growth. Since the concentration fluctuations are produced on the scale r_v and are enhanced by compressing down to r_d we need few basic facts about the Lagrangian statistics below the viscous scale of turbulence [3,6,9–13]. As long as the distance between two trajectories, $\mathbf{R} = \mathbf{q}_1 - \mathbf{q}_2$, is much smaller than r_v , it satisfies the equation $\partial_t \mathbf{R} = \mathbf{v}(t, \mathbf{q}_1) - \mathbf{v}(t, \mathbf{q}_2) \approx \sigma \mathbf{R}$ with the rate-of-strain matrix $\sigma_{\alpha\beta}(t) = \nabla_\beta v_\alpha$. The solution, $\mathbf{R}(t) = W(t)\mathbf{R}_0$, is expressed via the matrix W , statistics of which can be described universally at times much larger than the correlation time of σ . It is convenient to represent the Lagrangian transformation as stretching or contraction along fixed orthogonal directions followed by a rotation: $W = M\Lambda N$, where M and N are orthogonal, while Λ is diagonal. For our purpose, we need only the PDF of the eigenvalues $\exp(\rho_i)$ of the matrix Λ which is given by the large deviation theory [13]

$$\begin{aligned} \mathcal{P}(t, \rho_i) &= Z^{-1}(t)\theta(\rho_1 - \rho_2), \dots, \theta(\rho_{d-1} - \rho_d) \\ &\times \exp[-tS(\rho_1/t - \lambda_1, \dots, \rho_d/t - \lambda_d)]. \end{aligned} \quad (4)$$

Thus, at $t \gg \tau$, the statistics of stretching/contraction is characterized by a single function S of d variables. This entropy function is convex and non-negative. We assume S to be nonzero at least in some interval, which means that the flow is random. At large times, $\mathcal{P}(t, \rho_i)$ has a sharp maximum at $\rho_i = \lambda_i t$. The constants λ_i are called the Lyapunov exponents.

When one considers the advected fields, the Lagrangian trajectories fixed by their final rather than initial positions appear in the solution. Indeed, consider the Green's function $G(t, \mathbf{r} | t' = 0, \mathbf{r}')$ of (2). At $\kappa = 0$ it is supported on the Lagrangian trajectory $\mathbf{q}(0 | t, \mathbf{r})$ fixed by its final position \mathbf{r} . Finite diffusivity leads to a finite value of the Green's function in a region around the nondiffusive trajectory. As long as it is smaller than r_v one can expand the velocity in the equation on G in Taylor's series near $\mathbf{q}(t' | t, \mathbf{r})$. Since the zeroth order term corresponds to the mere sweeping of the density one must keep the first order term $\mathbf{v}(t', \mathbf{q}(t' | t, \mathbf{r}) + \mathbf{x}) - \mathbf{v}(t', \mathbf{q}(t' | t, \mathbf{r})) = \tilde{\sigma} \mathbf{x}$. The velocity gradient is now taken at the trajectory fixed by the final point, and its statistical properties are generally different from the ones of σ . The resulting equation is easily resolved in Fourier space

$$G(t, \mathbf{r} | 0, \mathbf{r}') = \int dk \frac{\exp[i\mathbf{k} \cdot [\mathbf{r}' - \mathbf{q}(0 | t, \mathbf{r})] - \mathbf{k}' I \mathbf{k}]}{(2\pi)^d \det \tilde{W}(t, \mathbf{r})}. \quad (5)$$

Here the matrix \tilde{W} determines the evolution of patches coming to the final point \mathbf{r} . It satisfies $\partial_t \tilde{W}(t | T, \mathbf{r}) = \tilde{\sigma} \tilde{W}(t | T, \mathbf{r})$ with $\tilde{W}(0 | T, \mathbf{r}) = 1$. One can show that the matrix $I = \kappa \int_0^t dt' \tilde{W}^{-1}(t' | t, \mathbf{r}) \tilde{W}^{-1,t}(t' | t, \mathbf{r})$ is the inertia tensor of a patch of particles, evaluated at $t = 0$, provided the patch is a sphere with the center at the point \mathbf{r} at time t . The particles perform Lagrangian motion in the common velocity field and independent Brownian motions (cf. [13]). The expression for G is purely dynamical since no averaging has been done in Eq. (5). We observe that the size of the region that makes the main contribution to the concentration grows as the largest eigenvalue of the matrix I ; i.e., the square of the linear size grows as $\kappa \int_0^t dt' \exp[-2\tilde{\rho}_d(t')]$. Since the Taylor expansion of velocity is valid as long as the range of Green's function around $\mathbf{q}(0 | t, \mathbf{r})$ is smaller than r_v , the applicability condition of Eq. (5) is

$$\kappa \int_0^t dt' \exp[-2\tilde{\rho}_d(t')] \ll r_v^2. \quad (6)$$

To express the advected field in the Lagrangian terms one ought to relate \tilde{W} to W . Introducing the spatial argument into W as $W_{ij}(t | t', \mathbf{r}) = \partial q_i(t | t', \mathbf{r}) / \partial r_j$ one

has $\tilde{W}(T | T, \mathbf{r}) = W(T | 0, \mathbf{q}(0 | T, \mathbf{r}))$. Indeed, what started at $\mathbf{q}(0 | T, \mathbf{r})$ comes finally to \mathbf{r} . Let us stress the difference in Eulerian and Lagrangian averages in the compressible case. All points equally contribute to an Eulerian average which is just the space integral (assuming ergodicity). In a Lagrangian average each trajectory comes with its own weight determined by the local rate of volume change. The volume average of a function $f(\tilde{W})$ is $\int d\mathbf{r} f(\tilde{W}(t_0, \mathbf{r})) = \int d\mathbf{x} f(W(t_0 | 0, \mathbf{x})) \det W(t_0 | 0, \mathbf{x})$. Since $\det W = \exp(\sum \rho_i)$, then the PDF of the eigenvalues $\exp \tilde{\rho}_i$ of \tilde{W} is given by (4) with $\rho_i \rightarrow \tilde{\rho}_i$ and multiplied by $\exp(\sum \tilde{\rho}_i)$. It is normalized due to $(\det W) = 1$ and has a maximum at $\tilde{\rho}_i = \tilde{\lambda}_i t$ with $\tilde{\lambda}_i$ generally different from λ_i [6].

One can now return to (5) and find the time of applicability of the ideal-case description for different moments. Formula (5) gives $n(t, \mathbf{r}) = \int G(t, \mathbf{r} | 0, \mathbf{r}') d\mathbf{r}' = 1 / \det \tilde{W}(t, \mathbf{r}) = \exp[-\sum \tilde{\rho}_i]$, the same expression as for nondiffusing particles. The condition (6) thus defines the applicability condition for the ideal-case regime. Let us now specify which $\tilde{\rho}_i$ correspond to the realizations determining a given moment $\langle n^\alpha \rangle$. The configurations that satisfy (6) contribute

$$\langle n^\alpha \rangle_{\text{id}} = \int d\tilde{\rho}_i \exp \left[-(\alpha - 1) \sum \tilde{\rho}_i - tS \right]. \quad (7)$$

This expression gives (3) after changing the integration variables $\sum \tilde{\rho}_i = -X$. The saddle point that determines (7) corresponds to $\tilde{\rho}_d = -c_\alpha t$, where c_α is an α -dependent constant. When $\kappa \int_0^t dt' \exp[2c_\alpha t'] \ll r_v^2$ the saddle-point satisfies the constraint (6). Whether the ideal description eventually breaks down depends on the sign of c_α . Note that the moments with $\alpha > 1$ grow in an ideal case which requires $\sum \tilde{\rho}_i < 0$ according to (7). Since $d\tilde{\rho}_d \leq \sum \tilde{\rho}_i < 0$, then $c_\alpha > 0$ at least for $\alpha > 1$. The physical meaning is transparent: the growth of these moments requires compression which eventually brings diffusion into play. The ideal approximation breaks down after $t_\alpha^* = c_\alpha^{-1} \ln(r_v/r_d)$. This expression is exact in the limit of large Schmidt numbers. Note that t_* depends on the order of the moment. For an entropy quadratic in $\tilde{\rho}$, c_α is a linear function of α . Since $\langle n^\alpha \rangle > \langle n^\alpha \rangle_{\text{id}}$ then the steady-state moments can be estimated from below by $(r_v/r_d)^{\gamma(\alpha)/c_\alpha}$. Note that $\gamma_\alpha \sim \alpha c_\alpha$ at large α which tells that the concentration PDF is less intermittent in a steady state than during the initial stage of growth.

The sign of c_α depends on α and the details of the entropy. Nevertheless, one can make universal statements about $\langle n^\alpha \rangle_{\text{id}}$ at large times, when (6) is effectively reduced to $\tilde{\rho}_d > 0$. The integral (7) is exponential $\langle n^\alpha \rangle_{\text{id}} \sim \exp[\tilde{\gamma}(\alpha)t]$ with convex $\tilde{\gamma}(\alpha)$. Considering the argument of the exponent in (7) one finds that $\tilde{\gamma}(\alpha)$ is negative for $\alpha > 0$ and positive for large enough negative α , so it has one zero at α_b . The boundary $\alpha_b < 0$ for $\tilde{\lambda}_d < 0$ and becomes equal to zero at high compressibility when $\tilde{\lambda}_d > 0$

and the normalization of the PDF of $\tilde{\rho}_i$ is determined by $\tilde{\rho}_d > 0$. Therefore large enough negative moments continue to grow exponentially and become infinite in the steady state, which corresponds to the formation of the power-law asymptotic behavior for the PDF of the concentration near $n = 0$: $\mathcal{P}(n) \propto n^{-\alpha_b - 1}$. Negative moments are determined by the regions void of particles where diffusion is irrelevant.

We now consider the moments determined by configurations that violate (6), in particular those with $\alpha > 0$. At large enough times, the particles that originated from different viscous intervals at $t = 0$ come into contact at $t = t_0$. Such contributions are not fully correlated and partially cancel each other. For the Reynolds number of the order unity, the velocity is decorrelated at $r \geq r_v$. Advection becomes equivalent to Brownian motion, for which all the contributions are canceled out, and the growth stops at $t \approx \lambda^{-1} \ln(r_v/r_d)$. The saturated moments are proportional to powers of the large parameter r_v/r_d . If the turbulence is developed, that is, $\text{Re} \gg 1$, then fluid velocity has power-law correlations at $r > r_v$: $\delta u \propto r^x$ with $x \leq 1$ ($x = 1/3$ in the Kolmogorov phenomenology of the energy cascade). The compressible component of the particles' flow is proportional to $(\mathbf{u} \cdot \nabla) \mathbf{u}$ and scales as follows at $r > r_v$: $\delta v \propto r^{2x-1}$. As we shall show below, the behavior of concentration is determined by the relative compressibility $\delta v/\delta u \propto r^{x-1}$ which is maximal at the viscous scale. The result is that the velocity modes from the inertial interval do not increase the level of concentration fluctuations beyond what has been produced on the viscous scale.

Unfortunately, we still lack the formalism to describe Lagrangian motion in the inertial interval with the same degree of universality as in the viscous interval. However, to understand the most essential properties of the concentration fluctuations one can use the standard model of a short-correlated Gaussian velocity with the variance $\langle v_\alpha(t, \mathbf{r},) v_\beta(0, 0) \rangle = \delta(t)[V_0 \delta_{\alpha\beta} - \mathcal{K}_{\alpha\beta}(\mathbf{r})]$ and $\mathcal{K}_{\alpha\beta} = [ru' + (d+1)u - c]r^2 \delta_{\alpha\beta} - (ru' + 2u - c)r_\alpha r_\beta$ [12,14]. The functions $u(r)$ and $c(r)$ are smooth at $r \ll r_v$: $u(r) - u(0) \propto r^2$ and $c(r) - c(0) \propto r^2$. In the inertial interval, $r_v \ll r \ll L$, $u(r) - u(0) \propto r^{-\gamma}$ and $c(r) - c(0) \propto r^{-3\gamma}$, where $0 \leq \gamma \leq 2$. Kolmogorov value is $\gamma = 2/3$. Conditions $u \propto r^{-2}$ and $c/u \rightarrow 0$ provide decorrelation at $r \gg L$. For nonzero γ , the ratio $\epsilon = c/u$ (measure of relative compressibility) increases as r decreases in the inertial interval and saturates in the viscous interval at ϵ_0 . The pair correlation function of the concentration $f = \langle n(t, 0)n(t, \mathbf{r}) \rangle$ satisfies a closed equation

$$\partial_t f = \hat{L}f = \nabla_\alpha \nabla_\beta (\mathcal{K}_{\alpha\beta} f) + 2\kappa \nabla^2 f. \quad (8)$$

One can prove that the operator \hat{L} has a nonpositive, continuous, and nondegenerate spectrum [6]. At large times, the correlation function thus relaxes to the zero-energy eigenmode f_{st} of \hat{L} :

$$f_{st} = \exp \left[\int_r^\infty \frac{xc(x) dx}{x^2 u(x) + 2(d-1)\kappa} \right]. \quad (9)$$

Assuming $c \propto r^{-2-\delta}$, $\delta > 0$ at $r \gg L$ we obtain there $f_{st} - 1 \propto (L/r)^\delta$. The behavior of f_{st} in the inertial interval, $r_v \ll r \ll L$, crucially depends on whether u and c have the same scaling exponents. As we have seen, that requires the velocity field to be spatially smooth ($\gamma = 0$) which takes place only in 2D vorticity cascade (up to logarithmic corrections). That type of turbulence is believed to be realized at the scales larger than few hundreds kilometers in the atmosphere so that our theory may be relevant for the description of clustering of atmospheric balloons observed in [15]. In this case, $f_{st}(r)$ behaves as a negative power of r . The fluctuation variance can be estimated as $f_{st}(r_d)$, which is then proportional to a positive power of the Reynolds and Schmidt numbers:

$$\langle n^2 - 1 \rangle \simeq (L/r_d)^{\epsilon_0}, \quad (10)$$

If, however, the velocity is not smooth in the inertial interval, u and c have different scaling exponents, and Eq. (9) shows that the main growth of f_{st} occurs below the viscous scale. Hence $\langle n^2 \rangle$ is independent of the Reynolds number. For example, for the Kolmogorov scaling of energy cascade, the solution (9) has the form $\ln f_{st} \propto a^4 r^{-4/3} r_v^{-8/3} \beta^{-2}$ at $r > r_v$. Therefore, the fluctuations of the concentration are mainly produced in the interval of scales $r \lesssim r_v$, where the fluid velocity is smooth:

$$\langle n^2 - 1 \rangle \simeq (r_v/r_d)^{\epsilon_0}. \quad (11)$$

For both (10) and (11), one can estimate the degree of compressibility as $\epsilon_0 \simeq \beta^{-2}(a/r_v)^4$. Since by definition $r_d \geq a$, significant fluctuations are possible only for heavy particles with $\beta \approx 2\rho/3\rho_p < (a/r_v)^2 \ln^{1/2}(r_v/r_d)$.

We thus conclude that the fluctuations of concentration are produced by the scales $r < r_v$ where the velocity is spatially smooth. For the α th moment of the concentration one has in the steady state

$$\langle n^\alpha \rangle \simeq (r_v/r_d)^{\beta_\alpha \epsilon_0}. \quad (12)$$

Apart from the expression for the particle velocity (1), all the rest of our theory is valid for $\epsilon \simeq 1$ as well. The moments are due to configurations which compress a region of size r_v into that with the smallest size reaching r_d at the time of observation. This picture agrees with the above statement that $\beta_\alpha \propto \alpha$ at large α .

The fluctuations of concentration may influence different physical and chemical phenomena that involve inertial particles. Consider a simple (yet important) example of a gas with the concentration $x(\mathbf{r}, t)$ that can condensate on aerosol particles: $dx/dt = -n(x - x_{eq})$. Being interested in the evolution of the spatial average $\bar{x}(t)$ one is

tempted to replace $n(\mathbf{r}, t)$ by \bar{n} , which would give an exponential decay of $x - x_{eq} \propto \exp(-\bar{n}t)$. The true decay in a turbulent flow has to be generally slower. The power tail of $\mathcal{P}(n)$ near zero gives $\bar{x} - x_{eq} \propto \int \exp(-nt) P(n) dn \propto t^{\alpha_b}$, as long as one may neglect the diffusion of the gas from the regions with low concentration of particles.

The regions of high concentration also play an important role. For example, in the atmospheric boundary layer, the viscous scale of 3D turbulence is $r_v \simeq 10^{-1}-10^{-2}$ cm so that we expect the concentration of atmospheric aerosols to have significant fluctuations on the scale of a millimeter or so. One important consequence of the phenomenon described is that solid aerosols (like lead particles from the exhaust pipes) may enter respiratory tracks in dense millimeter bunches rather than being uniformly distributed, this may have repercussions for human health. Another potential application of our theory is the description of the statistics of cloud formation both because solid particles can serve as cloud condensation nuclei and because the concentration of water droplets also may have large fluctuations in a turbulent atmosphere. A particular long-standing problem is to describe how millimeter-size droplets (necessary to trigger rain) appear from a suspension of micron-size droplets. We believe that clustering of droplets on a viscous scale of turbulence described here is of paramount importance for the proper theory of precipitation; the quantitative description requires further work.

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