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Formation of inhomogeneities in two-phase low-Mach-number compressible turbulent fluid flows

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Abstract

A new phenomenon of turbulent thermal diffusion is discussed. This effect is related to the dynamics of small inertial particles in low-Mach-number compressible turbulent fluid flows. Turbulent thermal diffusion is caused by the correlation between temperature and velocity fluctuations of the surrounding fluid and leads to relatively strong nondiffusive mean flux of inertial particles in regions with mean temperature gradients. It is shown that turbulent thermal diffusion under certain conditions can cause a large-scale instability of spatial distribution of particles. Particles are concentrated in the vicinity of the minimum (or maximum) of the mean temperature of the surrounding fluid depending on the ratio of material particle density to that of the surrounding fluid. At large Reynolds and Peclet numbers the turbulent thermal diffusion is much stronger than the molecular thermal diffusion. Turbulent thermal diffusion can be important in various naturally occurring and industrial multiphase flows. In particular, this effect may cause formation of inhomogeneities in spatial distribution of fuel droplets in internal combustion engines. It is conceivable to suggest that the effect of turbulent thermal diffusion can play an important role in a process of soot formation in flames and in atmospheric dynamics of pollutants, e.g. smog and aerosol clouds formation. © 1998 Elsevier Science Ltd. All rights reserved.

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1. Introduction

It is generally believed that turbulence promotes mixing (see for example McComb, 1990; Stock, 1996). However, experiments show formation of long-living inhomogeneities in concentration distribution of small inertial particles or gaseous admixtures in turbulent fluid flows

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(see, for example, Eaton and Fessler, 1994; Patterson et al., 1994). The origin of these inhomogeneities is not always clear but their influence on the mixing can be hardly overestimated.

Problem of formation of aerosol clouds is of fundamental significance in many areas of environmental sciences, physics of the atmosphere and meteorology. It is well-known that turbulence results in decay of inhomogeneities of aerosols concentration due to turbulent diffusion, whereas the opposite process, e.g. the preferential concentration of aerosols in atmospheric turbulent fluid flow still remains unexplained.

The main goal of this study is to bring attention to a new recently discovered phenomenon, i.e. the turbulent thermal diffusion (Elperin et al., 1996a). This phenomenon is related to the dynamics of small inertial particles in turbulent fluid flow in the presence of mean temperature gradient. The essence of this effect is that under certain conditions the initial spatial distribution of small inertial particles evolves into a highly inhomogeneous large-scale pattern where domains with increased particles concentration border on domains depleted of particles. Remarkably, similar phenomenon can occur in a turbulent fluid flow of gaseous mixtures (Elperin et al., 1997a).

In the present paper we have derived an equation for the turbulent flux of particles advected by a compressible turbulent fluid flow with low Mach numbers. It is shown that turbulent thermal diffusion may contribute to the formation of large-scale inhomogeneous structures in a particles distribution. The large-scale dynamics are studied by considering the stability of the equilibrium solution of the derived evolution equation for the mean number density of the particles in the limit of large Péclet numbers. The resulting equation is reduced to an eigenvalue problem for a Schrödinger equation with a variable mass, and a modified Rayleigh–Ritz variational method is used to estimate the lowest eigenvalue (corresponding to the growth rate of the instability). This estimate is in good agreement with obtained numerical solution of the Schrödinger equation.

Turbulent thermal diffusion can be important in various naturally occurring and industrial multiphase flows. It is generally believed that in conventional spark-ignition engines the fuel–air mixture is essentially homogeneous when it enters the cylinder. However, the turbulence can result in formation of an inhomogeneous concentration distribution which will have profound effects on the combustion process (Heywood, 1987; 1988; Reitz and Rutland, 1995). In fuel injection engines, e.g. diesel engines, the fuel is injected into the cylinder in the form of small liquid droplets. Turbulence induced inhomogeneities in the spatial distribution of the evaporating fuel droplets have strong effects upon combustion, and soot, and emissions formation in internal combustion engines and liquid (or solid) fuel combustors (Butler et al., 1981; Heywood, 1987; Glassman, 1988).

The effect of turbulent thermal diffusion can be a reason for smog and aerosol clouds formation and must be taken into account in the analysis of atmospheric dispersion of pollutants.

Note that inertia of particles also causes intermittency in spatial distribution of small inertial particles advected by a turbulent incompressible fluid flow (Elperin et al., 1996b). The inertia of particles results in divergent velocity field of particles and causes self-excitation (i.e. exponential growth) of small-scale fluctuations of concentration of small particles in a turbulent fluid flow. Under certain conditions the growth rates of the higher moments of

particles concentration is higher than those of the lower moments (Elperin et al., 1996b), i.e. particles spatial distribution is intermittent. The self-excitation of fluctuations of particles concentration is important in turbulent fluid flows of different nature with inertial particles or droplets (e.g. in atmospheric turbulence, combustion and in a laboratory turbulence). In particular, this effect may cause formation of small-scale inhomogeneities in spatial distribution of fuel droplets in internal combustion engines. Also this effect results in formation of small-scale inhomogeneities in droplet clouds (“inch clouds”) which were discovered recently (Baker, 1992).

2. The governing equations

Consider in detail the physics of the phenomenon of turbulent thermal diffusion. Evolution of the number density $n_p(t, \mathbf{r})$ of small particles in a turbulent flow is determined by equation:

$$\frac{\partial n_p}{\partial t} + \nabla \cdot (n_p \mathbf{U}) = -\nabla \cdot \mathbf{J}_M, \quad (1)$$

where \mathbf{U} is a random velocity field of the particles which they acquire in a turbulent fluid velocity field, the flux of particles \mathbf{J}_M is given by

$$\mathbf{J}_M = -D \left[\nabla n_p + k_t \frac{\nabla T_f}{T_f} + k_p \frac{\nabla P_f}{P_f} \right].$$

The first term in the formula for the flux of particles describes molecular diffusion, while the second term accounts for the flux of particles caused by the temperature gradient ∇T_f (molecular thermal diffusion for gases or thermophoresis for particles, see e.g. Reist, 1993), and the third term determines the flux of particles caused by the pressure gradient ∇P_f (molecular barodiffusion). Here D is the coefficient of molecular diffusion, $k_t \propto n_p$ is the thermal diffusion ratio, and Dk_t is the coefficient of thermal diffusion, $k_p \propto n_p$ is the barodiffusion ratio, and Dk_p is the coefficient of barodiffusion, T_f and P_f are the temperature and pressure of the surrounding fluid, respectively.

We consider here the case of large Reynolds and Peclet numbers and do not take into account the effect of particles upon the carrying fluid flow. The velocity of particle \mathbf{U} depends on the velocity of the surrounding fluid, and it can be determined from the equation of motion for a particle. This equation represents a balance of particle inertia with the fluid drag force produced by the motion of the particle relative to the surrounding fluid. Solution of the equation of motion for small particles with $\rho_p \gg \rho$ yields:

$$\mathbf{U} = \mathbf{v}(t, \mathbf{Y}(t)) - \tau_p \left[\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right] + O(\tau_p^2), \quad (2)$$

(Maxey, 1987; Maxey et al., 1996), where \mathbf{v} is the velocity of the surrounding fluid, $\mathbf{Y}(t)$ is the position of the particle, τ_p is the characteristic time of coupling between the particle and surrounding fluid (Stokes time), ρ_p is the material density of particles, and ρ is the density of the fluid. For instance, for spherical particles of radius a_* the Stokes time is $\tau_p = m_p / (6\pi a_* \rho \nu)$, where ν is the kinematic viscosity of the surrounding fluid, and m_p is the particle mass. The

second term in Eq. (2) describes the difference between the local fluid velocity and particle velocity arising due to the small, but finite, inertia of the particle.

In this study we consider low Mach numbers compressible turbulent flow $\nabla \cdot \mathbf{v} \neq 0$. The velocity field of particles is also compressible, i.e. $\nabla \cdot \mathbf{U} \neq 0$. Eq. (2) for the velocity of particles and the Navier–Stokes equation for the fluid for large Reynolds numbers yield

$$\nabla \cdot \mathbf{U} = \nabla \cdot \mathbf{v} - \tau_p \nabla \cdot \left(\frac{d\mathbf{v}}{dt} \right) + O(\tau_p^2) = \nabla \cdot \mathbf{v} + \tau_p \nabla \cdot \left(\frac{\nabla P_f}{\rho} \right) + O(\tau_p^2). \quad (3)$$

Here we used that $\nabla \cdot \mathbf{F} = 0$, where \mathbf{F} is the stirring force in the Navier–Stokes equation (see Appendix A).

We study the large-scale dynamics of small inertial particles and average Eq. (1) over an ensemble of random velocity fluctuations. For this purpose we use the stochastic calculus which has been previously employed in magnetohydrodynamics (Zeldovich et al., 1988; Kleorin and Rogachevskii, 1994; Rogachevskii and Kleorin, 1997) and in the problems of passive scalar transport in incompressible (Zeldovich et al., 1988; Avellaneda and Majda, 1994) and compressible (Elperin et al., 1995; 1996a, b, c; 1997a, b, c) turbulent flows. Within the stochastic calculus the solution of Eq. (1) is reduced to the analysis of the evolution of the concentration field $n_p(t, \mathbf{r})$ along the Wiener path, ξ :

$$\xi(t, t_0) = \mathbf{x} - \int_0^{t-t_0} \mathbf{U}[t_s, \xi(t, t_s)] ds + (2D)^{1/2} \mathbf{w}(t - t_0), \quad (4)$$

where $t_s = t - s$, and $\mathbf{w}(t)$ is a Wiener process. Eq. (4) describes a set of random trajectories which pass through the point \mathbf{x} at time t . A solution of Eq. (1) with the initial condition $n_p(t = t_0, \mathbf{x}) = n_0(\mathbf{x})$ is given by Feynman–Kac formula

$$n_p(t, \mathbf{x}) = M\{G(t, t_0)n_0[\xi(t, t_0)]\} \quad (5)$$

(see Appendix B and Schuss, 1980), where

$$G(t, t_0) = \exp \left[- \int_{t_0}^t b_*(\sigma, \xi(t, \sigma)) d\sigma \right], \quad (6)$$

$b_* \equiv \nabla \cdot \mathbf{U}$, and $M\{\cdot\}$ denotes the mathematical expectation over the Wiener paths. Hereafter we neglect small molecular thermal diffusion (see below). It can be taken into account in the mean velocity of particles.

Using the procedure described in detail in Appendix C we arrive at the equation for the mean field $N = \langle n_p \rangle$:

$$\frac{\partial N}{\partial t} + \nabla \cdot [N \mathbf{V}_{\text{eff}} - \hat{D} \nabla_m N] = 0, \quad (7)$$

where $\hat{D} \equiv D_{\text{pm}} = D \delta_{\text{pm}} + \langle \tau u_p u_m \rangle$, $\mathbf{V}_{\text{eff}} = \mathbf{V}_p - \langle \tau b \mathbf{u} \rangle$, and $b = \nabla \cdot \mathbf{u}$, and τ is the momentum relaxation time of random velocity field \mathbf{u} , which depends on the scale of turbulent motion. We use here for simplicity the δ -correlated in time random process to describe a turbulent velocity field. However, the results remain valid also for the velocity field with a finite correlation time, if the mean number density of the particles varies slowly in comparison with the correlation

time of the turbulent flow (see e.g. Dittrich et al., 1984). Eq. (7) was derived for $Pe \gg 1$. It is shown that for $Pe \ll 1$ (see Appendix D) and the arbitrary velocity field the equation for the mean field coincides with Eq. (7).

3. Turbulent flux of particles

Now we derive an equation for N^2 . Multiplication of Eq. (7) by N and simple manipulations yield

$$\frac{\partial N^2}{\partial t} + (\mathbf{V} \cdot \mathbf{S}) = -N^2(\mathbf{V} \cdot \mathbf{V}_{\text{eff}}) - I_D,$$

where $\mathbf{S}_m = N^2(\mathbf{V}_{\text{eff}})_m - D_{\text{mp}}\mathbf{V}_p N^2$, $I_D = 2D_{\text{mp}}(\mathbf{V}_m N)(\mathbf{V}_p N)$. The latter equation implies that if $\mathbf{V} \cdot \mathbf{V}_{\text{eff}} < 0$, a perturbation of the equilibrium distribution of inertial particles can grow in time, i.e. $(\partial/\partial t)\int N^2 d^3r > 0$. However, the total number of particles is conserved. Therefore the growth of N^2 when $\mathbf{V} \cdot \mathbf{V}_{\text{eff}} < 0$ is accompanied by formation of an inhomogeneous spatial distribution of the inertial particles whereby regions with an increased concentration of particles coexist with regions depleted from particles.

Now we calculate the velocity \mathbf{V}_{eff} . Using the equations of state $P_f = \kappa T_f \rho / m_\mu$ and Eq. (3) we obtain

$$\langle \tau \mathbf{u} b \rangle \approx \langle \tau (\mathbf{V} \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle + (\tau_p v_T^2 / T_*) \langle \tau \tilde{\mathbf{u}} \Delta \Theta \rangle,$$

where $v_T^2 = \kappa T_* / m_\mu$, m_μ is the mass of molecules of surrounding fluid and $T_f(t, \mathbf{r})$ is the temperature field with a characteristic value T_* , Θ are fluctuations of temperature. We neglect here the second moments $\sim \langle \tilde{\mathbf{u}} \rho \rangle$, since the mean turbulent flux of mass of the surrounding fluid vanishes in a finite domain surrounded by solid boundaries. Here ρ and $\tilde{\mathbf{u}}$ are fluctuations of the density and velocity of the fluid. On the other hand, the mean turbulent heat flux is nonzero in the presence of an external mean temperature gradient $\mathbf{V}T \neq 0$ (see below). To obtain an equation for $\langle \tau \mathbf{u} b \rangle$, we take into account that the fluctuating component of the particle velocity \mathbf{u} can be expressed in terms of the turbulent velocity of fluid $\tilde{\mathbf{u}}$: $\mathbf{u} = \tilde{\mathbf{u}} - \tau_p (d\tilde{\mathbf{u}}/dt - \langle d\tilde{\mathbf{u}}/dt \rangle)$ [see Eq. (2)]. Therefore, the velocity \mathbf{V}_{eff} is given by

$$\mathbf{V}_{\text{eff}} = \mathbf{V}_p - \langle \tau (\mathbf{V} \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle - \frac{\tau_p v_T^2}{T_*} \langle \tau \tilde{\mathbf{u}} \Delta \Theta \rangle,$$

where we neglect terms $\sim O(\tau_p^2)$. The latter formula shows that \mathbf{V}_{eff} depends on the mean turbulent heat flux $\langle \tilde{\mathbf{u}} \Theta \rangle$ that is determined by the well known equation

$$\langle \tilde{\mathbf{u}}(\mathbf{x}) \Theta(\mathbf{x}) \rangle = -\chi_T \mathbf{V}T$$

(see e.g. McComb, 1990), where the total temperature is $T_f = T + \Theta$, $T = \langle T_f \rangle$ is the mean temperature field, $\chi_T \sim u_0 l_0 / 3$ is the coefficient of turbulent thermal diffusivity. Note that herein we do not consider a situation with very high gradients when gradient transport assumption is violated. The above formula for the mean turbulent heat flux is written in the \mathbf{r} -space. The corresponding second moment in \mathbf{k} -space is given by

$$\langle \tilde{u}_m(\mathbf{k})\Theta(-\mathbf{k}) \rangle = -\tau(k)\langle \tilde{u}_m(\mathbf{k})\tilde{u}_n(-\mathbf{k}) \rangle \left(\frac{\partial T}{\partial R_n} \right),$$

where \mathbf{R} is a large-scale variable, and a spectrum of the turbulent velocity field and correlation time $\tau(k)$ can be chosen as Kolmogorov's spectrum:

$$\langle \tilde{u}_m\tilde{u}_n \rangle = \frac{2}{3k_0} \left(\frac{\langle \tilde{u}^2 \rangle}{8\pi k^2} \right) \left(\frac{k}{k_0} \right)^{-5/3} \left(\delta_{mn} - \frac{k_mk_n}{k^2} \right), \quad \tau(k) = 2\tau_0 \left(\frac{k}{k_0} \right)^{-2/3},$$

where $k_0 < k < k_0 Re_*^{3/4}$ (see e.g. McComb, 1990), $Re_* = ReF_0^{1/2}$, $Re = l_0 u_0 / \max(\nu, \chi)$ is the Reynolds number, $l_0 = k_0^{-1}$ is the maximum scale of turbulent motions, u_0 is the characteristic velocity in this scale, χ is the coefficient of molecular thermal conductivity, and $F_0(\mathbf{r}) = \langle \tilde{\mathbf{u}}^2(\mathbf{r}) \rangle / u_0^2$. Multiplying the equation for $\langle \tilde{u}_m(\mathbf{k})\Theta(-\mathbf{k}) \rangle$ by $-k^2\tau(k)$ and integrating in \mathbf{k} -space we obtain $\langle \tau \tilde{u}_m \Delta \Theta \rangle = \alpha \ln(Re_*) \nabla T$, where $\alpha = 2/3$. Finally we arrive at the following equation for the effective velocity

$$\mathbf{V}_{\text{eff}} = \mathbf{V}_p - \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle - \frac{\alpha \tau_p \nu_T^2}{T_*} \ln(Re_*) \nabla T. \quad (8)$$

Eq. (7) with this effective velocity \mathbf{V}_{eff} can be rewritten in the form

$$\frac{\partial N}{\partial t} + \nabla \cdot (N \mathbf{V}_p) = -\nabla \cdot (\mathbf{J}_T + \mathbf{J}_M), \quad (9)$$

where

$$\mathbf{J}_T = -D_T \left[\frac{k_T}{T} \nabla T - \frac{k_P}{P} \nabla P + F_0 \nabla N \right], \quad (10)$$

$$k_T = N[F_0 + T(\eta_0 + \sigma f)], \quad (11)$$

$$\eta_0 = \frac{3\alpha}{Pe} \left(\frac{m_p}{m_\mu} \right) \left(\frac{1}{T_*} \right) \ln Re, \quad (12)$$

where $\sigma = \eta_0 / (2 \ln Re)$, $f = \ln F_0$, $D_T = u_0 l_0 / 3$ is the coefficient of turbulent diffusion, k_T can be interpreted as a turbulent thermal diffusion ratio, and $D_T k_T$ is the coefficient of turbulent thermal diffusion, $k_P = F_0 N$ can be interpreted as turbulent barodiffusion ratio, and $D_T k_P$ is the coefficient of turbulent barodiffusion. We use here an identity

$$\frac{\tau_p \nu_T^2}{l_0 u_0} = \frac{1}{Pe} \left(\frac{m_p}{m_\mu} \right),$$

and $Pe = u_0 l_0 / D_*$ is the Peclet number and the molecular diffusion coefficient $D_* = \kappa T_* / (6\pi a_* \rho \nu)$. Note that for $Re \gg 1$ and $Pe \gg 1$ both turbulent diffusion coefficients are much larger than the corresponding molecular coefficients (i.e. $D_T \gg D$, and $D_T k_T \gg D k_t$, and $D_T k_P \gg D k_p$).

Using Eq. (2) the particles mean velocity can be written in the form

$$(\mathbf{V}_p)_i = \mathbf{V}_i - \tau_p \frac{\partial \mathbf{V}_i}{\partial t} - \tau_p \frac{\partial}{\partial x_j} \langle u_i u_j \rangle + \tau_p \langle u_j b \rangle. \quad (13)$$

Taking into account Eq. (13) the turbulent flux of particles \mathbf{J}_T^* in isotropic turbulence is given by

$$\mathbf{J}_T^* = \mathbf{J}_T - \frac{\tau_p}{3} \nabla \langle \mathbf{u}^2 \rangle, \quad (14)$$

where \mathbf{J}_T is determined by Eq. (10) and

$$k_T = N \left[F_0 \left(1 + \frac{\tau_p}{\tau_0} \right) + T(\eta_0 + \sigma f) \right], \quad k_P = NF_0 \left(1 + \frac{\tau_p}{\tau_0} \right).$$

The second term in Eq. (14) describes the effect of turbophoresis (see Caporaloni et al., 1975; Reeks, 1983).

Now we will show that turbulent thermal diffusion results in large-scale pattern formation whereby initial spatial distribution of particles in a turbulent incompressible flow of fluid evolves under certain conditions into large-scale inhomogeneous distribution due to excitation of an instability. One of the most important conditions for the instability is inhomogeneous spatial distribution of mean temperature of the surrounding fluid. In particular, the instability can be excited in the vicinity of the minimum in the mean temperature distribution. This results in particles concentrated in the vicinity of the minimum (or maximum) of the mean temperature of the surrounding fluid depending on the ratio of material particle density ρ_p to that of the surrounding fluid ρ .

The mechanism of the instability for $\rho_p \gg \rho$ is as follows. The inertia causes particles inside the turbulent eddy to drift out to the boundary regions between eddies (the regions with decreased velocity of the turbulent fluid flow and maximum of pressure of the surrounding fluid). Thus, inertial particles are accumulated in regions with maximum pressure of the turbulent fluid. Indeed, the inertia effect results in $\nabla \cdot \mathbf{U} \propto \tau_p \Delta P \neq 0$. On the other hand, for large Peclet numbers $\nabla \cdot \mathbf{U} \propto -dn_p/dt$. This means that in regions with maximum pressure of turbulent fluid (i.e. where $\Delta P < 0$) there is accumulation of inertial particles (i.e. $dn_p/dt > 0$). Similarly, there is an outflow of inertial particles from regions with minimum pressure of fluid. In a homogeneous and isotropic turbulence without large-scale external gradients of temperature a drift from regions with increased (decreased) concentration of inertial particles by a turbulent flow of fluid is equiprobable in all directions. Therefore, fluctuations of pressure (temperature) of the surrounding fluid is not correlated with turbulent velocity field and there exists only turbulent diffusion flux of inertial particles.

The situation is drastically changed when there is a large-scale inhomogeneity of the temperature of the turbulent flow. In this case the mean heat flux $\langle \tilde{\mathbf{u}} \Theta \rangle \neq 0$. Therefore fluctuations of both temperature and velocity of fluid are correlated. Fluctuations of temperature cause fluctuations of pressure of fluid. The pressure fluctuations result in fluctuations of the concentration of inertial particles. Indeed, increase (decrease) of the pressure of surrounding fluid is accompanied by accumulation (outflow) of the particles. Therefore,

direction of mean flux of particles coincides with that of heat flux, i.e. $\langle \tilde{\mathbf{u}} n_p \rangle \propto \langle \tilde{\mathbf{u}} \Theta \rangle \propto -\nabla T$. The mean flux of the inertial particles is directed to the minimum of the mean temperature and the inertial particles are accumulated in this region.

The additional turbulent nondiffusive fluxes of particles can also be estimated as follows. We average Eq. (1) over the ensemble of the turbulent velocity field and subtract the obtained averaged equation from Eq. (1). This yields an equation for the turbulent component q of particles number density

$$\frac{\partial q}{\partial t} - D\Delta q = -\nabla \cdot (N\mathbf{u} + \mathbf{Q}), \quad (15)$$

where $n_p = N + q$, $\mathbf{Q} = \mathbf{u}q - \langle \mathbf{u}q \rangle$. Eq. (15) is written in a frame moving with the mean velocity \mathbf{V}_p . The magnitude of $\partial q/\partial t - D\Delta q + \nabla \cdot \mathbf{Q}$ can be estimated as q/τ , where τ is the turnover time of the turbulent eddies. Thus the turbulent field q is of the order of $q \sim -\tau N(\nabla \cdot \mathbf{u}) - \tau(\mathbf{u} \cdot \nabla)N$. Now we calculate the turbulent flux of particles $\mathbf{J}_T = \langle \mathbf{u}q \rangle$:

$$\mathbf{J}_T \sim -N\langle \tau \mathbf{u}(\nabla \cdot \mathbf{u}) \rangle - \langle \tau \mathbf{u} u_j \rangle \nabla_j N. \quad (16)$$

Here $\nabla \cdot \mathbf{u} = \nabla \cdot \tilde{\mathbf{u}} - \tau_p \nabla \cdot (d\tilde{\mathbf{u}}/dt)$. Using Eq. (8) we can reduce the turbulent flux of particles (16) to Eq. (10).

Compressibility of the background fluid is important when the size of particles are smaller than one micron (or for the gaseous admixture). In this case the effect of particles inertia is very small and the main contribution to the effect of the turbulent thermal diffusion is due to the compressibility of the background fluid. On the other hand, when the size of particles larger than 5–10 microns the effect of particles inertia is very important and the contribution to the effect of the turbulent thermal diffusion caused by particles inertia is much larger than that due to compressibility of the background fluid [i.e. $T(\eta_0 + \sigma f) \ll F_0$, see Eq. (11)]. Certainly, the compressibility ($\nabla \cdot \mathbf{v} \neq 0$) of the background fluid cannot be ignored completely, since otherwise we cannot satisfy the continuity equation and the equation of state simultaneously in the presence of a nonzero mean temperature gradient.

4. Formation of large-scale inhomogeneities

Let us study the large-scale instability. Evolution of the mean field N is determined by Eq. (9). Substitution

$$N(t, \mathbf{r}) = N_* \Psi_0(Z) \exp(\gamma_0 t) \exp\left[-\frac{1}{2} \int \chi_0(Z) dZ + i\mathbf{k} \cdot \mathbf{r}_\perp\right] + N_0(\mathbf{r})$$

reduces Eq. (9) to the eigenvalue problem for the Schrödinger equation

$$\frac{1}{m_0} \Psi_0''(Z) + [W_0 - U_0(Z)] \Psi_0(Z) = 0, \quad (17)$$

where $W_0 = -\gamma_0$, $A' = dA/dZ$, and the potential U_0 is given by

$$U_0 = \frac{1}{m_0} \left(\frac{\chi_0^2}{4} + \frac{\chi_0'}{2} + \kappa_0 \right),$$

and

$$\chi_0 = f' + \frac{T'}{T} - \frac{P'}{P} + \frac{1}{F_0}(\eta_0 + \sigma f)T',$$

$$\kappa_0 = k^2 - \left(\frac{T'}{T} \right)' + \left(\frac{P'}{P} \right)' + \frac{f'P'}{P} - \frac{1}{F_0}(\eta_0 + \sigma f)T'' - \frac{f'T'}{T} \left(1 + \frac{\sigma T}{F_0} \right).$$

Here $m_0 = \exp[-f(Z)]$ the axis Z is directed along mean temperature gradient, the wave vector \mathbf{k} is normal to the axis Z . In derivation of Eq. (17) we take into account that for an isotropic turbulence $\langle u_m(\mathbf{x})u_n(\mathbf{x}) \rangle = u_0^2 \exp(f)\delta_{mn}/3$. Equilibrium distribution of the mean number density $N_0(\mathbf{r})$ is determined by equation $\hat{D}\nabla_m N_0 = \mathbf{V}_{\text{eff}}N_0$. Eq. (17) is written in the dimensionless form, the coordinate is measured in units Λ_T , time t is measured in units Λ_T^2/D_T , the wave number k is measured in units Λ_T^{-1} , the temperature T is measured in units of temperature difference δT in the scale Λ_T , and concentration N is measured in units N_* .

Now we use quantum mechanical analogy for the analysis of the large-scale pattern formation in the concentration field N of the inertial particles. The instability can be excited ($\gamma_0 > 0$) if there is a region of well potential where $U_0 < 0$. The positive value of W_0 corresponds to the turbulent diffusion, whereas a negative value of W_0 results in the excitation of the instability. Consider the case $P'/P \ll T'/T$. The potential U_0 can be rewritten as

$$U_0 = \frac{1}{4m_0} \left[\left(f' - \frac{T'}{T} \right)^2 + \left(\frac{T'}{T} - \frac{\sigma T f'}{F_0} \right)^2 + \left(\frac{T'}{T} + \frac{1}{F_0}(\eta_0 + \sigma f)T' \right)^2 + 4k^2 + 2f'' - 2\frac{T''}{T} - \frac{2}{F_0}(\eta_0 + \sigma f)T'' - \left(\frac{\sigma T f'}{F_0} \right)^2 \right]. \tag{18}$$

The potential U_0 can be negative if

$$2f'' - 2\frac{T''}{T} - \frac{2}{F_0}(\eta_0 + \sigma f)T'' - \left(\frac{\sigma T f'}{F_0} \right)^2 < 0. \tag{19}$$

In order to estimate the first energy level W_0 we use a modified variational method (e.g. a modified Rayleigh–Ritz method). The modification of the regular variational method is required, since Eq. (17) can be regarded as the Schrödinger equation with a variable mass $m_0(Z)$. Now we rewrite Eq. (17) in the form

$$\hat{H}\Psi_0 = W_0\Psi_0, \quad \hat{H} = U_0 - \frac{1}{m_0} \frac{d^2}{dZ^2}. \tag{20}$$

The modified variational method employs an inequality

$$W_0 \leq I, \quad I = \int m_0\Psi^*\hat{H}\Psi dZ, \tag{21}$$

where ψ is an arbitrary function that satisfies a normalization condition

$$\int m_0 \Psi^* \Psi dZ = 1. \quad (22)$$

The inequality (21) can be proved if one uses the expansion

$$\Psi = \sum_{p=0}^{\infty} a_p \Psi_0^{(p)},$$

where

$$\sum_{p=0}^{\infty} |a_p|^2 = 1$$

and $\int m_0 (\Psi_0^{(p)})^* \Psi_0^{(k)} dZ = \delta_{pk}$. The eigenfunctions $\Psi_0^{(p)}$ satisfy the equation $\hat{H} \Psi_0^{(p)} = W_p \Psi_0^{(p)}$.

We chose the trial function Ψ in the form

$$\Psi = A_0 \exp[-\alpha(Z - Z_0)^2/2], \quad A_0 = \left(\frac{\alpha + b_0}{\pi}\right)^{1/4} \exp\left(\frac{\alpha b_0 Z_0^2}{2(\alpha + b_0)}\right), \quad (23)$$

where the unknown parameters α and Z_0 can be found from the condition of minimum of the function $I(\alpha, Z_0)$ [see Eq. (21)]. Here we use the following spatial distributions of $f(Z)$ and $T(Z)$:

$$f(Z) = -b_0 Z^2 \exp(-\beta_0 Z^2), \quad (24)$$

$$T(Z) = (T_* + Z^2 + aZ) \exp(-\epsilon_0 Z^2), \quad (25)$$

where $\beta_0 \ll 1$ and $\epsilon_0 \ll 1$. These distributions satisfy the necessary condition (19) for excitation of the instability. We consider a case $T_*^{-1} \ll b_0$.

Note that the mean temperature gradient and the inhomogeneity of the turbulence can be specified independently due to the following reason. The hydrodynamic turbulence is generally determined by equations (A4)–(A6) with two independent external sources \mathbf{F}_1 and Q_1 (see Appendix A). Only in the special case of a turbulent convection is there one independent source Q_1 . In this case $\mathbf{F}_1 = \rho_1 \mathbf{g}$, where \mathbf{g} is the acceleration of gravity. Only in this special case the mean temperature gradient and the inhomogeneity of the turbulence cannot be specified independently. On the other hand, in a general case the external sources \mathbf{F}_1 and Q_1 are independent. The latter allows us to specify the mean temperature gradient and the inhomogeneity of the turbulence independently.

Substituting (23) and (25) into Eq. (21) yields

$$I = -\eta_0 + \frac{1}{2\alpha^{3/2}} [\alpha^2(\alpha - b_0)^{1/2} + b_0(\alpha + b_0)^{1/2} [b_0 + 2\alpha(b_0 Z_0^2 - 1)]] \exp\left(-\frac{\alpha b_0 Z_0^2}{\alpha - b_0}\right) + \eta_0^2 \frac{(\alpha - b_0)^{1/2}}{4(\alpha - 2b_0)^{5/2}} [2(\alpha - 2b_0) + (2\alpha Z_0 + a(\alpha - 2b_0))^2] \exp\left(\frac{\alpha^2 b_0 Z_0^2}{(\alpha - b_0)(\alpha - 2b_0)}\right). \quad (26)$$

Here we consider the case of $k \ll 1$. This implies long-wave perturbations in the horizontal plane. Thus, the modified Rayleigh–Ritz method allows us to estimate the growth rate of the instability. For example, when $b_0 \ll \eta_0$ (i.e. the inhomogeneity of turbulence is not strong), the growth rate of the instability is given by:

$$\gamma = \frac{3}{2} b_0. \tag{27}$$

Thus, it is shown here that the equilibrium distribution of the number density of particles is unstable. The instability results in the formation of an inhomogeneous distribution of the number of density particles. The exponential growth during the linear stage of the instability can be damped by the nonlinear effects (e.g. hydrodynamic interaction between particles and a turbulent fluid flow, a change of temperature distribution in the vicinity of the temperature inversion layer).

Characteristic size of the inhomogeneity in Z -direction when $\eta_0 \geq a_0$ is of the order of

$$l_z \sim A_T \left\{ 1 + \left[\frac{3\alpha}{Pe} \left(\frac{m_p}{m_\mu} \right) \left(\frac{\delta T}{T_*} \right) \ln Re_* \right]^{-1/2} \right\}.$$

Remarkably $l_z \rightarrow \infty$ when $Pe \rightarrow \infty$, i.e. this effect exists for large, but finite, Peclet numbers.

The obtained results are valid in the case when the density of surrounding fluid is much less than the material density of particles ($\rho \ll \rho_p$). However, the results of this study can be easily generalized to include the case $\rho \geq \rho_p$ using an equation of motion of particles in fluid flow (see e.g. McComb, 1990). This equation of motion takes into account contributions due to the pressure gradient in the fluid surrounding the particle (caused by acceleration of the fluid) and the virtual (“added”) mass of the particles relative to the ambient fluid. A solution of this equation coincides with Eq. (2) except for the transformation $\tau_p \rightarrow \beta \tau_p$, where

$$\beta = \left(1 + \frac{\rho}{\rho_p} \right) \left(1 - \frac{3\rho}{2\rho_p + \rho} \right).$$

For $\rho \geq \rho_p$ turbulent thermal diffusion ratio k_T in Eq. (12) must be multiplied by β . Therefore the additional mass flux of particles is directed towards the mean temperature gradient [see Eq. (10)] and particles are accumulated in the vicinity of the maximum of mean temperature of surrounding fluid since $\beta < 0$. In the opposite case when $\rho \ll \rho_p$, $\beta \simeq 1$ and particles are accumulated in the vicinity of the mean temperature minimum.

Eq. (17) was solved numerically with turbulent kinetic energy and mean temperature profiles given by Eqs. (24) and (25). The extremum of turbulent kinetic energy is located at $Z = 0$, temperature minimum is located at $Z = -a/2$ [see Eqs. (24) and (25)], and $Z = -H$ is a location of an impenetrable boundary for the particles. The boundary condition at $Z = -H$ is determined by equation $(\mathbf{J}_T)_Z = 0$, which yields the condition for Ψ_0

$$\frac{d\Psi_0}{dZ} = \left(f' - \frac{1}{2} \chi_0 \right) \Psi_0 \quad \text{at} \quad Z = -H. \tag{28}$$

Eq. (28) provides zero flux of the particles through a horizontal boundary plane $Z = -H$. The second boundary condition is $\Psi_0(Z = \infty) = 0$. As an example, Figs. 1 and 2 show the

dependence of the growth rate of the instability versus b_0 for $k \ll 1$ for different η_0 . These values of the parameters satisfy the necessary condition (19) for the excitation of the instability. These numerical results are in agreement with the analytical estimates obtained by means of the modified Rayleigh–Ritz method for $b_0 > 0$. Remarkably the numerical results show that the instability is excited (e.g. $\gamma > 0$) even in a homogeneous turbulent flow ($b_0 = 0$).

5. Discussion

The effect of turbulent thermal diffusion may be of relevance in combustion. In particular, this effect may cause formation of inhomogeneities in spatial distribution of fuel droplets in internal combustion engines (see e.g. Heywood, 1988; Patterson et al., 1994; Reitz and Rutland, 1995). Indeed, characteristic parameters of turbulence in a cylinder of internal combustion engine are: maximum scale of turbulent flow $l_0 \sim 0.5\text{--}1$ cm; velocity in the scale l_0 : $u_0 \sim 100$ cm/s; Reynolds number $Re \sim (0.7\text{--}7) \times 10^3$; and characteristic values of mean temperature distribution: scale $A_T \sim 13\text{--}18$ cm; and dimensionless mean spatial temperature variation $\delta T/T_* \sim 0.3\text{--}0.5$ (see e.g. Heywood, 1988; Patterson et al., 1994; Reitz and Rutland, 1995). Then the characteristic time of formation of inhomogeneities in spatial distribution of droplets of radius $a_* = 30 \mu\text{m}$ is $\sim (3\text{--}6) \times 10^{-2}$ s. Notably, this time is comparable with the duration of an engine cycle. These turbulence induced inhomogeneities in the spatial distribution of the evaporating fuel droplets have strong effects upon combustion, soot and emissions formation (see e.g. Butler et al., 1981; Glassman, 1988).

It is conceivable to suggest that the effect of turbulent thermal diffusion can play an important role in a process of soot formation in flames and in atmospheric dynamics of combustion pollutants, e.g. smog formation. Observations of the vertical distributions of pollutants in the atmosphere show that maximum concentrations can occur within temperature inversion layers (see e.g. Seinfeld, 1986; Jaenicke, 1987). Using the characteristic parameters of the atmospheric turbulent boundary layer: maximum scale of turbulent flow $l_0 \sim 10^3\text{--}10^4$ cm; velocity in the scale l_0 : $u_0 \sim 30\text{--}100$ cm/s; Reynolds numbers $Re \sim 10^6$ and of the temperature inversion: scale $A_T \sim 3 \times 10^4$ cm and dimensionless mean spatial temperature variation $\delta T/T_* \sim (1\text{--}3) \times 10^{-2}$ (see e.g. Seinfeld, 1986; Jaenicke, 1987), we obtain that the characteristic

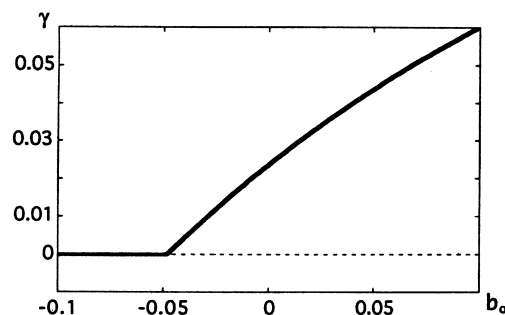


Fig. 1. Dependence of the growth rate of the instability versus b_0 for $a = -2$, $H = 8.1$, $Re = 10^5$, $T_* = 300$, and $\eta_0 = 15$.

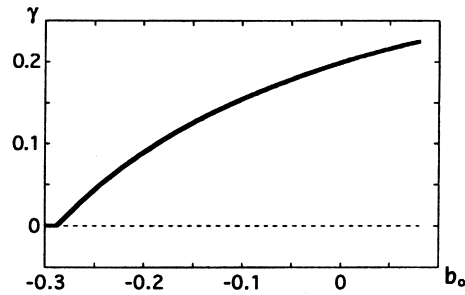


Fig. 2. Dependence of the growth rate of the instability versus b_0 for $a = -2$, $H = 8.1$, $Re = 10^5$, $T_* = 300$, and $\eta_0 = 30$.

time of excitation of the instability of concentration distribution of aerosols with material density $\rho_p \sim 2 \text{ g/cm}^3$ and radius $a_* = 10 \text{ }\mu\text{m}$ varies in the range from 0.3 to 3 hours. This value is in compliance with the characteristic time of growth of inhomogeneous structures in atmosphere. It is essential that this time strongly depends on the aerosol size, i.e. $\sim a_*^{-2}$.

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Appendix A

A.1. Governing equations for small Mach numbers

The fluid velocity \mathbf{v} is determined by the nondimensional Navier–Stokes equation

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} = -M^{-2} \frac{\nabla P_f}{\rho_f} + Re^{-1} [\nabla \mathbf{v} + \zeta \nabla(\text{div } \mathbf{v})] + \frac{\mathbf{F}}{\rho_f}, \tag{A1}$$

where velocity, coordinate, temperature and density are measured in the units v_* , l_* , T_* , ρ_* , respectively, $M = v_*/c_s$ is the Mach number, $c_s = (T_*/m_\mu)^{1/2}$ is the sound speed, and $\zeta = 1/3 + \zeta_b/v$, ζ_b is a bulk viscosity, $Re = v_* l_*/\nu$ is the Reynolds number, ν is the kinematic viscosity. The stirring force \mathbf{F} is measured in the units $F_* = \rho_* v_*^2/l_*$ and the pressure is measured in the units $P_* = \rho_* T_*/m_\mu$. A solution of equation (A1) can be sought in the form of power series of Mach number

$$\phi = \sum_{k=0}^{k=\infty} M^{2k} \phi_{k+1}, \tag{A2}$$

where functions $\phi = (\rho_f; T_f; P_f; \mathbf{v})$. Substitution of the expansion (A2) into equation (A1), continuity equation and equation for the temperature field, and comparing the terms of the same order in M^{2k} yields a set of equations

$$\nabla P_1 = 0, \quad (\text{A3})$$

$$\frac{\partial \mathbf{v}_1}{\partial t} + (\mathbf{v}_1 \cdot \nabla) \mathbf{v}_1 = -\frac{1}{\rho_1} \nabla P_2 + Re^{-1} [\nabla \mathbf{v}_1 + \zeta \nabla (\text{div } \mathbf{v}_1)] + \frac{1}{\rho_1} \mathbf{F}_1, \quad (\text{A4})$$

$$\frac{\partial \rho_1}{\partial t} + \nabla \cdot (\rho_1 \mathbf{v}_1) = 0, \quad (\text{A5})$$

$$\frac{\partial T_1}{\partial t} + (\mathbf{v}_1 \cdot \nabla) T_1 = -(\gamma - 1) T_1 (\nabla \cdot \mathbf{v}_1) + Pe^{-1} \nabla T_1 + Q_1, \quad (\text{A6})$$

where Q is an external heat source, $P_1 = \rho_1 T_1$ and $P_2 = \rho_2 T_1 + \rho_1 T_2$, and γ is the specific heats ratio. Equations (A5)–(A6) coincide with those for the total fields ρ_f and T_f . Note that equation (A3) appears in the order of M^{-2} , whereas equations (A4)–(A6) appear in the order of M^0 .

Appendix B

B.1. Solution of Eq. (1)

Show that Eq. (5) is a solution of Eq. (1). We expand the function $n_p(t, \xi_{\Delta t})$ in equation (C1) in the Taylor series in the vicinity of the point \mathbf{x} :

$$n_p(t, \xi_{\Delta t}) \simeq n_p(t, \mathbf{x}) + \frac{\partial n_p}{\partial x_m} (\xi_{\Delta t} - \mathbf{x})_m + \frac{1}{2} \frac{\partial^2 n_p}{\partial x_m \partial x_s} (\xi_{\Delta t} - \mathbf{x})_m (\xi_{\Delta t} - \mathbf{x})_s + \dots \quad (\text{B1})$$

Using the equation for the Wiener trajectory we obtain

$$[\xi(t_2, t_1) - \mathbf{x}]_m = - \int_0^{t_2-t_1} U_m(t_s, \xi_s) ds + (2D)^{1/2} \mathbf{w}_m(t_2 - t_1), \quad (\text{B2})$$

where $\xi(t_2, t_2 - s) \equiv \xi_s$. Expanding the velocity $U_m(t_s, \xi_s)$ in the Taylor series in the vicinity of the point \mathbf{x} , and using equation (B2) we get

$$U_m(t_s, \xi_s) = U_m(t_s, \mathbf{x}) + \frac{\partial U_m}{\partial x_1} \left[- \int_0^s U_1(t_\sigma, \xi_\sigma) d\sigma + (2D)^{1/2} \mathbf{w}_1(s) \right]. \quad (\text{B3})$$

Substitution of equation (B3) into (B2) yields

$$\begin{aligned}
 (\xi_{\Delta t} - \mathbf{x})_m = & - \int_0^{\Delta t} U_m(t_s, \xi_s) ds + \int_0^{\Delta t} \frac{\partial U_m}{\partial x_1} \Big|_{(t_s, \mathbf{x})} ds \int_0^s U_1(t_\sigma, \xi_\sigma) d\sigma \\
 & - \sqrt{2D} \int_0^{\Delta t} \frac{\partial U_m}{\partial x_1} \Big|_{(t_s, \mathbf{x})} w_1(s) ds + \sqrt{2D} w_m.
 \end{aligned}
 \tag{B4}$$

Integrals in formula (B4) can be evaluated by means of the “mean value” theorem. The result is given by

$$(\xi_{\Delta t} - \mathbf{x})_m \simeq -U_m(t_*, \mathbf{x})\Delta t + \sqrt{2D}w_m + O[(\Delta t)^2],
 \tag{B5}$$

where t_* is within the interval $(t, t + \Delta t)$. Substitution of equation (B5) into (B1) yields an expression for the field $n_p(t, \xi_{\Delta t})$

$$n_p(t, \xi_{\Delta t}) \simeq n_p(t, \mathbf{x}) + \frac{\partial n_p}{\partial x_m} [-U_m(t_*, \mathbf{x})\Delta t + \sqrt{2D}w_m] + Dw_m w_s \frac{\partial^2 n_p}{\partial x_m \partial x_s} + O[(\Delta t)^2].
 \tag{B6}$$

Expanding function $b_*(\sigma, \xi_\sigma)$ in the Taylor series in the vicinity of the point \mathbf{x} , using equation (B5), evaluating the integral

$$\int_t^{t+\Delta t} b_*(\sigma, \xi_\sigma) d\sigma$$

by means of the “mean value” theorem, we calculate the Green function $G(t + \Delta t, t)$ accurate up to $\sim \Delta t$. The result is given by

$$G(t + \Delta t, t) \simeq 1 - b_*(t_3, \mathbf{x})\Delta t,
 \tag{B7}$$

where t_3 is within the interval $(t, t + \Delta t)$. Combination of equations (B6), (B7) and (C1), and averaging over Wiener trajectories yields expression for the field $n_p(t + \Delta t, \mathbf{x})$. Now we calculate the value $[n_p(t + \Delta t, \mathbf{x}) - n_p(t, \mathbf{x})]/\Delta t$ for $\Delta t \rightarrow 0$. This procedure yields Eq. (1). Therefore, Eq. (5) is a solution of Eq. (1).

Appendix C

C.1. Derivation of the mean-field equation for large Peclet number

Now let us derive an equation for the mean field $N = \langle n_p \rangle$ using Eq. (1). The procedure of derivation is outlined in the following:

1. If the total field n_p is specified at instant t , then we can determine the total field $n_p(t + \Delta t)$ at near instant $t + \Delta t$ by means of substitutions $t \rightarrow t + \Delta t$ and $t_0 \rightarrow t$ in Eq. (5). The result is given by

$$n_p(t + \Delta t, \mathbf{x}) = M\{G(t + \Delta t, t)n_p[t, \boldsymbol{\xi}(t + \Delta t, t)]\}, \quad (C1)$$

were

$$G(t + \Delta t, t) = \exp\left[-\int_t^{t+\Delta t} b_*(\sigma, \boldsymbol{\xi}_\sigma) d\sigma\right],$$

$$\boldsymbol{\xi}(t + \Delta t, t) \equiv \boldsymbol{\xi}_{\Delta t} = \mathbf{x} - \int_0^{\Delta t} \mathbf{U}(t_\sigma, \boldsymbol{\xi}_\sigma) d\sigma + (2D)^{1/2}\mathbf{w}(\Delta t),$$

and $t_\sigma = t + \Delta t - \sigma$, and $\boldsymbol{\xi}(t_2, t_1) \equiv \boldsymbol{\xi}_{t_2-t_1}$, i.e. $\boldsymbol{\xi}_\sigma = \boldsymbol{\xi}(t + \Delta t, t_\sigma)$.

2. Expansion of the functions $n_p(t, \boldsymbol{\xi}_{\Delta t})$ and the velocity $U_m(t_\sigma, \boldsymbol{\xi}_\sigma)$ in the Taylor series in the vicinity of the point \mathbf{x} allows us to express the field $n_p(t, \boldsymbol{\xi}_{\Delta t})$ in terms of the field $n_p(t, \mathbf{x})$. Indeed, expand function $n_p(t, \boldsymbol{\xi}_{\Delta t})$ of equation (C1) in the Taylor series in the vicinity of the point \mathbf{x} :

$$n_p(t, \boldsymbol{\xi}_{\Delta t}) \simeq n_p(t, \mathbf{x}) + \frac{\partial n_p}{\partial x_m}(\boldsymbol{\xi}_{\Delta t} - \mathbf{x})_m + \frac{1}{2} \frac{\partial^2 n_p}{\partial x_m \partial x_s}(\boldsymbol{\xi}_{\Delta t} - \mathbf{x})_m(\boldsymbol{\xi}_{\Delta t} - \mathbf{x})_s + \dots \quad (C2)$$

Using the equation for the Wiener trajectory we obtain

$$[\boldsymbol{\xi}(t_2, t_1) - \mathbf{x}]_m = -\int_0^{t_2-t_1} U_m(t_s, \boldsymbol{\xi}_s) ds + (2D)^{1/2}\mathbf{w}_m(t_2 - t_1), \quad (C3)$$

where $\boldsymbol{\xi}(t_2, t_2-s) \equiv \boldsymbol{\xi}_s$. Expanding the velocity $U_m(t_s, \boldsymbol{\xi}_s)$ in the Taylor series in the vicinity of the point \mathbf{x} , and using equation (C3) yields

$$U_m(t_s, \boldsymbol{\xi}_s) \simeq U_m(t_s, \mathbf{x}) - U_1 \frac{\partial U_m}{\partial x_1} s + (2D)^{1/2} \frac{\partial U_m}{\partial x_1} \mathbf{w}_l(s) + \dots \quad (C4)$$

Substituting equation (C4) into (C3) and calculating the integrals in equation (C3) accurate up to terms $\sim (t_2 - t_1)^2$ yields

$$[\boldsymbol{\xi}(t_2, t_1) - \mathbf{x}]_m \simeq -(t_2 - t_1)U_m + \frac{1}{2}(t_2 - t_1)^2 U_1 \frac{\partial U_m}{\partial x_1}$$

$$- \sqrt{2D} \frac{\partial U_m}{\partial x_1} \int_0^{t_2-t_1} w_1 ds + \sqrt{2D} w_m(t_2 - t_1) + \dots \quad (C5)$$

The combination of equations (C5) and (C2) yields the field $n_p(t, \boldsymbol{\xi}_{\Delta t})$

$$n_p(t, \boldsymbol{\xi}_{\Delta t}) = n_p(t, \mathbf{x}) + \frac{\partial n_p}{\partial x_m} \left[-U_m \Delta t + \frac{1}{2} U_1 \frac{\partial U_m}{\partial x_1} (\Delta t)^2 + \sqrt{2D} w_m - \sqrt{2D} \frac{\partial U_m}{\partial x_1} \int_0^{\Delta t} w_1 ds \right]$$

$$+ \frac{1}{2} \frac{\partial^2 n_p}{\partial x_m \partial x_s} [U_m U_s (\Delta t)^2 + 2D w_m w_s - \sqrt{2D} \Delta t (U_m w_s + U_s w_m)]. \quad (C6)$$

Here we keep the terms up to $\geq O[(\Delta t)^2]$.

3. Now we expand the function $b_*[\sigma, \xi(t + \Delta t, \sigma)]$ in the Taylor series in the vicinity of the point \mathbf{x} , and calculate the integral

$$\int_t^{t+\Delta t} b_*[\sigma, \xi(t + \Delta t, \sigma)] d\sigma.$$

The result is given by

$$\int_t^{t+\Delta t} b_*(\sigma, \xi_\sigma) d\sigma \simeq b_*(t, \mathbf{x})\Delta t - \frac{1}{2} U_q \frac{\partial b_*}{\partial x_q} (\Delta t)^2 + \sqrt{2D} \frac{\partial b_*}{\partial x_q} \int_t^{t+\Delta t} w_q d\sigma + \dots \quad (C7)$$

Here we also keep terms $\geq O[(\Delta t)^2]$. Using equation (C7) we calculate the function $G(t + \Delta t, t)$ accurate up to $\sim (\Delta t)^2$

$$G(t + \Delta t, t) \simeq 1 - b_*(t, \mathbf{x})\Delta t + \frac{1}{2} U_q \frac{\partial b_*}{\partial x_q} (\Delta t)^2 + \frac{1}{2} b_*^2 (\Delta t)^2 - \sqrt{2D} \frac{\partial b_*}{\partial x_q} \int_t^{t+\Delta t} w_q d\sigma. \quad (C8)$$

4. The substitution of equation (C8) and (C6) into equation (C1) allows us to determine the number density $n_p(t + \Delta t, \mathbf{x})$. Note that the velocity \mathbf{U} is determined by the turbulent velocity \mathbf{v} of surrounding fluid [see Eq. (2)]. In order to determine the mean field N we average the obtained equation for the number density $n_p(t + \Delta t, \mathbf{x})$ over the turbulent velocity \mathbf{U} (i.e. $N = \langle n \rangle$). Note that $\mathbf{U} = \mathbf{V}_p + \mathbf{u}$, where $\mathbf{V}_p = \langle \mathbf{U} \rangle$ is the mean velocity and \mathbf{u} is the random component of the velocity of particles. It is important to note that the Wiener random process $\mathbf{w}(t)$ and the turbulent velocity $\mathbf{u}(t, \mathbf{x})$ are independent random processes and, therefore, we can change the order of averaging: $\langle M\{f\} \rangle \rightarrow M\{\langle f \rangle\}$ (see Zeldovich et al., 1988). On the contrary, the random processes $\mathbf{w}(t)$ and $\mathbf{u}(t, \xi_{\Delta t})$ are correlated. We also assume that the velocities \mathbf{u} in both intervals $(0, t)$ and $(t, t + \Delta t)$ are independent, because we consider the random flow with short time of the renewal. It is assumed also that the velocity \mathbf{u} in small intervals $(0, \Delta t)$; $(\Delta t, 2\Delta t)$; $(2\Delta t, 3\Delta t)$; ..., is constant (time-independent) and changes every small time interval Δt . Note that the averaging over the Wiener paths corresponds to the averaging over the molecular processes with very small characteristic scales. On the other hand, $\langle f \rangle$ determines the averaging over the turbulent velocity field with scales that are larger than molecular ones.

5. Now we calculate

$$\frac{N(t + \Delta t, \mathbf{x}) - N(t, \mathbf{x})}{\Delta t},$$

and pass to the limit $\Delta t \rightarrow 0$. Here $N = \langle n_p \rangle$. The result is given by

$$\frac{\partial N}{\partial t} + [(\mathbf{V} - \langle \tau(\mathbf{u} \cdot \nabla) \mathbf{u} \rangle - 2\langle \tau b \mathbf{u} \rangle) \cdot \nabla] N = B_{\text{eff}} N + D_{\text{pm}} \frac{\partial^2 N}{\partial x_p \partial x_m}, \quad (C9)$$

where $B_{\text{eff}} = -(\nabla \cdot \mathbf{V}) + \langle \tau(\mathbf{u} \cdot \nabla) b \rangle + \langle \tau b^2 \rangle$. In such a procedure the turbulent velocity field \mathbf{u} with very short time of the renewal tends to δ -correlated in the time random

process:

$$\langle u_m(t, \mathbf{x})u_n(t', \mathbf{y}) \rangle = 2\delta(t - t')\langle \tau u_m(\mathbf{x})u_n(\mathbf{y}) \rangle$$

(see Zeldovich et al., 1988). Here we take into account that the time of the renewal Δt may depend on scale. Using the identity

$$\left\langle \tau u_p \frac{\partial}{\partial x_p} u_m \right\rangle = \frac{\partial}{\partial x_p} \langle \tau u_p u_m \rangle - \langle \tau \mathbf{u}(\nabla \cdot \mathbf{u}) \rangle$$

we obtain Eq. (7) for the mean field N . Here we neglect a weak dependence of τ on coordinate.

Appendix D

D.1. Derivation of the mean-field equation for small Peclet numbers

The solution of the equation of the convective diffusion (1) in form (5) by averaging over the Wiener trajectories is valid for arbitrary Peclet numbers. However, using a δ -correlated in time process for the turbulent velocity field is justified only for $Pe \gg 1$. In this Appendix we will show that for small Peclet numbers the equation for the mean particles number density has the same form as in the case $Pe \gg 1$.

The equation of the convective diffusion (1) can be rewritten in the form

$$\frac{\partial n}{\partial t} + \nabla \cdot (n\mathbf{U}) = D\Delta n. \quad (\text{D1})$$

Fields \mathbf{U} , b_* and n can be presented in the form $\mathbf{U} = \mathbf{V}_p + \mathbf{u}$, $b_* = B + b$, and $n = N + q$, where $\mathbf{V} = \langle \mathbf{U} \rangle$, $B = \langle b_* \rangle = \nabla \cdot \mathbf{V}_p$, $N = \langle n \rangle$, and q is a random component of the number density of particles, the angular brackets mean statistical averaging.

Averaging of equation (D1) over the ensemble of the turbulent fluctuations we obtain an equation for the mean field N

$$\frac{\partial N}{\partial t} + \nabla \cdot (\mathbf{V}_p N) - D\Delta N = -\nabla \cdot \langle q\mathbf{u} \rangle. \quad (\text{D2})$$

Subtracting equation (D2) from (D1) yields an equation for the turbulent field q

$$\frac{\partial q}{\partial t} - D\Delta q = -(\mathbf{u} \cdot \nabla)N - bN, \quad (\text{D3})$$

where the term Bq in the left part of equation (D3) is excluded by means of substitution $q \rightarrow q \exp(-Bt)$. Equation (D3) is written in a frame moving with the mean particles velocity \mathbf{V}_p . Here we neglect a small quadratic in the fluctuating field terms $\nabla \cdot \langle q\mathbf{u} \rangle - \nabla \cdot (q\mathbf{u})$.

These terms yield effects that are of the order of $\sim Pe^2$, whereas linear in the fluctuating field terms are of the order of $\sim Pe$. A solution of equation (D3) with the initial condition $q(t = 0, \mathbf{x}) = q_0(\mathbf{x})$ is given by

$$q(t, \mathbf{x}) = \int q_0(\mathbf{z})G_*(t, \mathbf{x} - \mathbf{z})d^3z - \int u_m(t', \mathbf{z})\frac{\partial N}{\partial z_m}G_*(t - t', \mathbf{x} - \mathbf{z})d^3z dt' - \int b(t', \mathbf{z})N(t, \mathbf{y})G_*(t - t', \mathbf{x} - \mathbf{z})d^3z dt', \tag{D4}$$

where $G_*(\tau, \mathbf{y})$ is the Green function of the diffusion equation:

$$G_*(\tau, \mathbf{y}) = (2\pi D\tau)^{-3/2} \exp\left(-\frac{y^2}{2\pi D\tau}\right).$$

Now let us calculate the second moment $\langle qu_n \rangle$ by means of equation (D4). The result is given by

$$\langle q(t, \mathbf{y})u_n(t, \mathbf{x}) \rangle = \int \langle q_0(\mathbf{z})u_n(t, \mathbf{x}) \rangle G_*(t, \mathbf{y} - \mathbf{z})d^3z - \int \langle u_n(t, \mathbf{x})u_m(t', \mathbf{z}) \rangle \frac{\partial N}{\partial z_m}(t', \mathbf{z})G_*(t - t', \mathbf{y} - \mathbf{z})d^3z dt' - \int \langle u_n(t, \mathbf{x})b(t', \mathbf{z}) \rangle N(t', \mathbf{z})G_*(t - t', \mathbf{y} - \mathbf{z})d^3z dt'. \tag{D5}$$

Note that $\langle q_0u_n \rangle = 0$, because q_0 and \mathbf{u} are not correlated. Now we introduce the fast $\mathbf{r} = \mathbf{x} - \mathbf{z}$ and slow $\mathbf{R} = (\mathbf{x} + \mathbf{z})/2$ variables. The derivative

$$\frac{\partial N}{\partial z_m} \simeq \frac{\partial N}{\partial R_m} + N\left(\frac{r}{R}\right).$$

It follows from equation (D5) that

$$\langle q(t, \mathbf{y})u_n(t, \mathbf{x}) \rangle = -\frac{\partial N}{\partial R_m} \int \langle u_m u_n \rangle G_*(\tau, \mathbf{r})d^3r d\tau - N \int \langle u_n b \rangle G_*(\tau, \mathbf{r})d^3r d\tau. \tag{D6}$$

Substitution of equation (D6) into equation (D2) yields an equation for the mean particles number density

$$\frac{\partial N}{\partial t} + \mathbf{V} \cdot (\mathbf{V}_{\text{eff}}N) = \frac{\partial}{\partial R_m} \left(D_{mn} \frac{\partial N}{\partial R_n} \right), \tag{D7}$$

where

$$D_{mn} = D\delta_{mn} + \int \langle u_n u_m \rangle G_*(\tau, \mathbf{r})d^3r d\tau, \tag{D8}$$

$$\mathbf{V}_{\text{eff}} = \mathbf{V} - \int \langle b\mathbf{u} \rangle G_*(\tau, \mathbf{r})d^3r d\tau. \tag{D9}$$

Comparison of equations (D7)–(D9) obtained for $Pe \ll 1$ with Eq. (7) derived for $Pe \gg 1$ shows that these equations coincide in form. Therefore, the described above phenomena for $Pe \gg 1$ can also occur for $Pe \ll 1$. However, the instability which can occur at $Pe \gg 1$ is suppressed for $Pe \ll 1$ by strong diffusion.

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