Turbulent barodiffusion, turbulent thermal diffusion, and large-scale instability in gases

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Two effects, turbulent barodiffusion and turbulent thermal diffusion in gases, are discussed. These phenomena are related to the dynamics of a gaseous admixture in compressible turbulent fluid flow with low Mach numbers. Turbulent barodiffusion causes an additional mass flux of the gaseous admixture directed to the maximum of the mean fluid pressure, while turbulent thermal diffusion results in an accumulation of the gaseous admixture in the vicinity of the minimum of the mean temperature of the surrounding fluid. At large Péclet and Reynolds numbers these additional turbulent fluxes are considerably higher than those caused by molecular barodiffusion and molecular thermal diffusion. It is shown that turbulent barodiffusion and turbulent thermal diffusion may contribute to the formation of large-scale inhomogeneous structures in a gaseous admixture advected by a low-Mach-number compressible turbulent velocity field. 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 gaseous admixture 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. [S1063-651X(97)00403-0]

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I. INTRODUCTION

The phenomena of molecular barodiffusion and molecular thermal diffusion in gases were discovered long ago (see, e.g., [1–3]). The equation for the number density n_p of a gaseous admixture in a surrounding fluid taking into account these effects is

$$\frac{\partial n_p}{\partial t} = -\nabla \cdot \mathbf{J}_M .$$

The molecular flux of the gaseous admixture

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

comprises three terms: the flux of the gaseous admixture due to molecular diffusion ($\sim \nabla n_p$), the flux caused by the temperature gradient ∇T_f of the surrounding fluid (molecular thermal diffusion), and the flux caused by the fluid pressure gradient ∇P_f (molecular barodiffusion; see, e.g., [3]). Here D is the coefficient of molecular diffusion, $k_t \propto n_p$ is the thermal diffusion ratio, and $k_p \propto n_p$ is the molecular barodiffusion ratio. Turbulent fluid flow results in a large increase of the effective diffusion coefficient at large Péclet and Reynolds numbers [4–7].

The main goal of this paper is to discuss two effects: turbulent barodiffusion and turbulent thermal diffusion in gases. These phenomena result in additional mass fluxes of the gaseous admixture advected by a compressible turbulent fluid flow with low Mach number. These effects are caused by the compressibility of the turbulent fluid flow. It is demonstrated that turbulent barodiffusion and turbulent thermal diffusion may contribute to the formation of large-scale inhomogeneous structures in a gaseous admixture. 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 gaseous admixture in the limit of large Péclet numbers.

Recently, it was found that the inertia of small particles advected by a turbulent fluid flow results in an additional flux of particles directed to 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 [8]. Turbulent thermal diffusion of small inertial particles is caused by nonzero divergence of the velocity field of inertial particles advected by fluid flow.

II. TURBULENT FLUX OF THE MASS OF A GASEOUS ADMIXTURE

We consider a mixture of two gaseous components with very different number densities: $n_p \ll n$. Instead of a gaseous component with a number density n_p, light particles can be considered. Hereafter the gas component with a number density $n = \rho_f / m_\mu$ is called a fluid, while the component with a number density n_p is called a gaseous admixture (m_μ is the mass of molecules of the surrounding fluid). The number density $n_p(t,\mathbf{r})$ of a gaseous admixture in a turbulent compressible flow is determined by

$$\frac{\partial n_p}{\partial t} + \nabla \cdot (n_p \mathbf{U}) = D \Delta n_p, \tag{1}$$

where $\mathbf{U}$ is a random velocity field of the gaseous admixture, which it acquires in a turbulent fluid velocity field $\mathbf{v}$. The density ρ_f of the surrounding fluid satisfies the continuity equation

$$\frac{\partial \rho_f}{\partial t} + \nabla \cdot (\rho_f \mathbf{v}) = 0. \tag{2}$$

In Eq. (2) we do not take into account the diffusion of the fluid in the gaseous admixture because $n_p \ll n$.

The velocity $\mathbf{v}$ of the surrounding fluid is assumed to be known. We neglect an influence of the gaseous admixture upon the velocity of the surrounding fluid because $\rho_p = m_p n_p \ll \rho_f$, where m_p is the mass of molecules (or particles) of the gaseous admixture. On the other hand, the velocity of the gaseous admixture $\mathbf{U}$ is determined by the velocity of the surrounding fluid, and it is found from the equation of motion $d\mathbf{U}/dt = [\mathbf{v} - \mathbf{U}]/\tau_p + \mathbf{g}$, where $\mathbf{g}$ is the acceleration of gravity, and τ_p is the characteristic time of momentum coupling between the gaseous admixture and the surrounding fluid. Here we neglect the small pressure gradient of the gaseous admixture and its viscous force. The solution of the equation of motion for a gaseous admixture when inertia effects are negligible yields the velocity $\mathbf{U} = \mathbf{v} + \mathbf{v}_s$. The second term in the velocity of the gaseous admixture describes a sedimentation of the admixture in a gravity field with a terminal fall velocity $\mathbf{v}_s = \tau_p \mathbf{g}$.

In order to derive the equation for the mean concentration of the gaseous admixture, Eq. (1) must be averaged over the ensemble of random velocity fluctuations. For this purpose we use the stochastic calculus that was applied in magnetohydrodynamics [9–11] and passive scalar transport in incompressible [9,10,12] and compressible [8,13] turbulent flows. In the stochastic calculus the solution of Eq. (1) with the initial condition $n_p(t=t_0, \mathbf{x}) = n_0(\mathbf{x})$ is given by

$$n_p(t,\mathbf{x}) = M\{G(t,t_0) n_0[\boldsymbol{\xi}(t,t_0)]\} \tag{3}$$

(see Appendix A), where

$$G(t,t_0) = \exp\left[- \int_{t_0}^{t} b_*(\sigma, \boldsymbol{\xi}(t,\sigma)) d\sigma \right], \tag{4}$$

$b_* \equiv \nabla \cdot \mathbf{U}$, and $M\{\ \}$ is the mathematical expectation over the Wiener paths $\boldsymbol{\xi}$,

$$\boldsymbol{\xi}(t,t_0) = \mathbf{x} - \int_0^{t-t_0} \mathbf{U}(t_s, \boldsymbol{\xi}_s) ds + (2D)^{1/2} \mathbf{w}(t-t_0), \tag{5}$$

where $t_s = t - s$, $\boldsymbol{\xi}_s \equiv \boldsymbol{\xi}(t, t-s)$, and $\mathbf{w}(t)$ is the Wiener random process. Equation (5) describes a set of random trajectories that pass through the point $\mathbf{x}$ at time t (see, e.g., [13]). The use of this technique (see Appendix B) yields the equation for the mean field $N = \langle n_p \rangle$,

$$\frac{\partial N}{\partial t} + \nabla \cdot (\mathbf{V}_{\rm eff} N - \hat{D} \nabla_m N) = 0, \tag{6}$$

where

$$\hat{D} = D_{pm} = D \delta_{pm} + \tau_0 \langle u_p u_m \rangle, \tag{7}$$

$$\mathbf{V}_{\rm eff} = \mathbf{V} - \tau_0 \langle \mathbf{u} (\nabla \cdot \mathbf{u}) \rangle, \tag{8}$$

$\mathbf{U} = \mathbf{V} + \mathbf{u}$, $\mathbf{V} = \langle \mathbf{U} \rangle$ is the mean velocity, $\mathbf{u}$ is the turbulent component of the velocity, and τ_0 is the characteristic time of turbulent motions in the energy containing scale l_0. Note that Eq. (6) is written in the form of a conservation law for the mass density of the gaseous admixture.

Now we calculate the effective velocity $\mathbf{V}_{\rm eff}$. The continuity equation (2) can be rewritten in the form

$$\left(1 + \frac{\tilde{\rho}}{\rho}\right)(\nabla \cdot \mathbf{u}) = -\frac{1}{\rho}(\mathbf{u} \cdot \nabla)\rho - \frac{1}{\rho}\frac{d\tilde{\rho}}{dt}, \tag{9}$$

where $\rho_f = \tilde{\rho} + \rho$, $\rho = \langle \rho_f \rangle$ is the mean density, and $\tilde{\rho}$ is the density fluctuations. We take into account that the turbulent velocity of the gaseous admixture coincides with that of the surrounding fluid because the random component of the terminal fall velocity $\mathbf{v}_s$ equals zero.

Consider the case of low Mach numbers $M \ll (l_0/L)^{1/2}$, where l_0 is the energy containing scale of turbulent fluid flow, L is the large scale (e.g., the inhomogeneity scale of the mean temperature or the mean density), $M = (\langle \mathbf{u}^2 \rangle)^{1/2}/c_s$ is the Mach number, and c_s is the sound speed. Since the fluctuations of pressure $\tilde{P} = c_s^2 \tilde{\rho} \sim \rho \langle \mathbf{u}^2 \rangle$, the ratio $\tilde{\rho}/\rho \sim M^2 \ll 1$. Therefore we can neglect terms $\sim \tilde{\rho}$ and Eq. (9) is reduced to

$$(\nabla \cdot \mathbf{u}) \simeq -\frac{1}{\rho}(\mathbf{u} \cdot \nabla)\rho. \tag{10}$$

The equation of state for the surrounding fluid for the mean fields $P = \rho T/m_\mu$ yields

$$\frac{\nabla T}{T} - \frac{\nabla P}{P} = -\frac{\nabla \rho}{\rho} \equiv \vec{\boldsymbol{\lambda}}. \tag{11}$$

Combining Eqs. (6)–(8), (10), and (11) allows us to rewrite Eq. (6) in the form

$$\frac{\partial N}{\partial t} + \nabla \cdot (\mathbf{J}_M + \mathbf{J}_T) = 0, \tag{12}$$

where the turbulent flux of the gaseous admixture is given by

$$\mathbf{J}_T = -D_T \left[\nabla N + N \frac{\nabla T}{T} - N \frac{\nabla P}{P} \right], \tag{13}$$

$D_T = u_0 l_0/3$ is the coefficient of turbulent diffusion, and u_0 is the characteristic velocity in the scale l_0. The molecular flux of the gaseous admixture

$$\mathbf{J}_M = -D \left[\nabla N + k_t \frac{\nabla T}{T} + k_p \frac{\nabla P}{P} \right] \tag{14}$$

comprises three terms: molecular diffusion ($\sim \nabla N$), molecular thermal diffusion ($\sim k_t \nabla T$, where k_t is the molecular thermal diffusion ratio), and molecular barodiffusion ($\sim k_p \nabla P$, where k_p is the molecular barodiffusion ratio; see, e.g., [3]). Comparing the molecular (14) and turbulent (13) fluxes of the gaseous admixture, we can interpret the new additional turbulent fluxes as fluxes caused by the effects of turbulent thermal diffusion ($\sim k_T \nabla T$, where $k_T = N$ is the turbulent thermal diffusion ratio) and turbulent barodiffusion ($\sim k_P \nabla P$, where $k_P = -N$ is the turbulent barodiffusion ratio).

Remarkably, the additional turbulent flux caused by the effect of turbulent thermal diffusion appears also for inertial particles advected by a turbulent flow [8]. The turbulent flux of small inertial particles of mass m_p is given by

$$\mathbf{J}_T^{(p)} = -D_T \left[\frac{k_T^{(p)}}{T} \nabla T + \nabla N \right], \tag{15}$$

$$k_T^{(p)} = N \frac{3}{\mathrm{Pe}} \left(\frac{m_p}{m_\mu} \right) \left(\frac{T}{T_*} \right) \ln \mathrm{Re}_* \tag{16}$$

(see [8]), where $k_T^{(p)}$ is the turbulent thermal diffusion ratio of small inertial particles, $D_T k_T^{(p)}$ is the coefficient of turbulent thermal diffusion, $\mathrm{Pe} = u_0 l_0 / D$ is the Péclet number, $\mathrm{Re}_* = \min\{\mathrm{Re}, \mathrm{Pe}_T\}$, $\mathrm{Re} = l_0 u_0 / \nu$ is the Reynolds number, $\mathrm{Pe}_T = l_0 u_0 / \chi$ is the thermal Péclet number, and χ is the coefficient of molecular thermal conductivity. The turbulent thermal diffusion of small inertial particles is caused by the correlation between temperature and velocity fluctuations of the surrounding fluid and leads to the relatively strong mean flux of small inertial particles in the direction of the regions with 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. Note that the velocity field of particles is divergent due to the finite inertia of particles advected by turbulent flow. For heavy particles (with sizes $\geqslant 1\,\mu$m) the turbulent thermal diffusion ratio $k_T^{(p)} \gg N$ at large Reynolds and Péclet numbers.

For light particles (or the gaseous admixture) the effects of inertia are negligible. However, compressibility of the surrounding fluid results in the new additional turbulent fluxes of the gaseous admixture caused by turbulent thermal diffusion and turbulent barodiffusion. Note that the turbulent flux of the gaseous admixture (13) can also be obtained by means of a simple dimensional analysis (see [14]).

III. MECHANISM OF THE FORMATION OF LARGE-SCALE INHOMOGENEITIES IN THE SPATIAL DISTRIBUTION OF THE CONCENTRATION OF A GASEOUS ADMIXTURE

Now we study the dynamics of the large-scale distribution of the concentration of the gaseous admixture in a small-scale turbulent fluid flow. Turbulent barodiffusion and turbulent thermal diffusion may result in the formation of inhomogeneous structures in a large-scale distribution of the gaseous admixture advected by a compressible turbulent fluid flow. The mechanism of this effect is as follows. In incompressible flow at any time, the mass of the fluid flowing into a small volume exactly equals the mass outflow from this volume. In the limit of infinite Péclet number the gaseous admixture is frozen into the flow of a surrounding fluid. Therefore, there is no accumulation of the gaseous admixture at any point of the volume.

The situation changes if $\nabla \cdot \mathbf{u} \neq 0$ in a turbulent fluid flow. In this case a mass of fluid flowing into a small volume does not equal the mass outflow from the volume at any instance. Therefore, at times smaller than a characteristic time of the turbulent velocity field there is an accumulation (or outflow) of the gaseous admixture. Note that the accumulation and outflow of the gaseous admixture in a small control volume are separated in time and molecular diffusion breaks a time-reversal symmetry between the accumulation and outflow. The latter can cause pattern formation in the concentration distribution of the gaseous admixture advected by a compressible turbulent fluid flow. Indeed, let us demonstrate this effect. For this purpose we derive the equation for n_p^2. Multiplication of Eq. (1) by n_p and simple manipulations yield

$$\frac{\partial n_p^2}{\partial t} + (\nabla \cdot \mathbf{A}) = -n_p^2 (\nabla \cdot \mathbf{U}) - 2D (\nabla n_p)^2, \tag{17}$$

where $\mathbf{A} = n_p^2 \mathbf{U} - D \nabla n_p^2$. Consider an evolution of mass of the gaseous admixture in a volume V_* in a Lagrangian frame. Integrating Eq. (17) over the volume V_*, we obtain

$$\frac{d}{dt} \int n_p^2 dV_* = -\int n_p^2 (\nabla \cdot \mathbf{v}) dV_* - 2D \int (\nabla n_p)^2 dV_*,$$

where we use that $\nabla \cdot \mathbf{U} = \nabla \cdot \mathbf{v}$. The latter equation shows that in an incompressible fluid flow $\int n_p^2 dV_*$ can only decrease with time due to molecular diffusion. On the other hand, in a compressible fluid flow the value $\int n_p^2 dV_*$ can grow when $\nabla \cdot \mathbf{v} < 0$. Thus the regions where $\nabla \cdot \mathbf{v} < 0$ contribute to the growth of $\int n_p^2 dV_*$. However, the total number of particles of the gaseous admixture in the whole system is conserved.

Now we elucidate the role of molecular diffusion and sedimentation. Integration of Eq. (1) over the volume V_* yields

$$\frac{d}{dt} \int n_p dV_* = D \oint (\nabla n_p) \cdot d\mathbf{S} - \oint n_p \mathbf{v}_s \cdot d\mathbf{S},$$

where we use the Gauss theorem $\int (\nabla \cdot \mathbf{B}) dV_* = \oint \mathbf{B} \cdot d\mathbf{S}$. Thus only molecular diffusion and sedimentation can change a mass of the gaseous admixture in the volume V_*. However, the direction of this change (the growth or decay of $\int n_p dV_*$ and $\int n_p^2 dV_*$) is determined by the sign of $\nabla \cdot \mathbf{v}$. When $D = 0$ and $\mathbf{v}_s = \mathbf{0}$ the particles of gaseous admixture are "frozen" into the surrounding fluid and $\int n_p dV_* = \mathrm{const}$. If $\nabla \cdot \mathbf{v} < 0$ and $D \neq 0$, a redistribution of the mass of the gaseous admixture occurs, so that regions with a high concentration of the gaseous admixture are adjacent to the regions with a low concentration. Therefore, the large-scale inhomogeneous structures in the spatial distribution of the gaseous admixture concentration are formed.

Note that there exists a certain similarity between the formation of the large-scale inhomogeneous structures in the spatial distribution of the gaseous admixture concentration (passive scalar) and generation of magnetic fields (passive vector) by a turbulent flow of conducting fluid. The magnetic flux $\int \mathbf{B} \cdot d\mathbf{S}$ through a surface corresponds to a mass $\int n_p dV_*$ of the gaseous admixture in the volume V_*. The magnetic energy $\int \mathbf{B}^2 dV_*$ corresponds to $\int n_p^2 dV_*$. The source of generation of magnetic energy is associated with $\partial v_k / \partial x_m$, i.e.,

$$\frac{\partial \mathbf{B}^2}{\partial t} \propto B_k B_m \frac{\partial v_k}{\partial x_m}$$

(see, e.g., [15]). On the other hand, compressibility ($\partial v_k / \partial x_k \neq 0$) of the fluid results in the formation of large-scale inhomogeneous structures in the spatial distribution of the gaseous admixture concentration (i.e., growth of n_p^2):

$$\frac{\partial n_p^2}{\partial t} \propto - n_p^2 \frac{\partial v_k}{\partial x_k}.$$

The effect of the magnetic diffusion is similar to that of molecular diffusion of a passive scalar. Indeed, the magnetic flux can be changed only if the magnetic diffusion is nonzero (finite electrical conductivity), which prevents the freezing of the magnetic field into the flow of a conducting fluid. Diffusion of the magnetic field breaks a reversibility in time of the magnetic structures evolution. Similarly, molecular diffusion breaks a reversibility in time so that accumulation and outflow of the gaseous admixture from a control volume cannot be compensated exactly. The latter results in the formation of the large-scale inhomogeneous structures in the spatial concentration distribution of the gaseous admixture.

All of the above considered conditions for the formation of the inhomogeneous structures in the concentration distribution of the gaseous admixture are only necessary, but not sufficient. The sufficient conditions can be determined only after the analysis of the large-scale stability of the equilibrium concentration distribution. This analysis is performed in the next section.

IV. ANALYSIS OF LARGE-SCALE INSTABILITY

In this section we analyze the formation of large-scale inhomogeneous structures in the spatial concentration distribution of the gaseous admixture concentration. Equation (12) for the mean number density of the gaseous admixture can be rewritten in the form

$$\frac{\partial N}{\partial t} + \nabla \cdot \{[\mathbf{v}_s - \boldsymbol{\lambda} D_T F_0(Z)] N\} = \nabla \cdot \{[D + D_T F_0(Z)] \nabla N\}, \tag{18}$$

where $\mathbf{V} = \mathbf{v}_s$, $F_0(Z) = \langle \mathbf{u}^2 \rangle / u_0^2$, $\langle u_p u_m \rangle = u_0^2 F_0(Z) \delta_{mn}/3$ (for details see [13]), and the vectors $\mathbf{g}$ and $\nabla \rho$ are directed against the Z axis. The equilibrium solution of Eq. (18) is given by

$$\nabla N_0 = N_0 \left(\frac{\mathbf{v}_s - \boldsymbol{\lambda} D_T F_0(Z)}{D + D_T F_0(Z)} \right).$$

Hereafter we consider the case Pe$\gg 1$, i.e., $D_T \gg D$. Now we study the stability of this equilibrium solution.

We seek the solution of Eq. (18) in the form

$$N(t,\mathbf{r}) = N_0(\mathbf{r}) + N(t,Z) \exp(i \mathbf{k} \cdot \mathbf{r}_\perp), \tag{19}$$

where the wave vector $\mathbf{k}$ is perpendicular to the Z axis. Substituting Eq. (19) into Eq. (18) yields

$$\frac{\partial N}{\partial t} = \frac{1}{m_0} \frac{\partial^2 N}{\partial Z^2} + \mu_0 \frac{\partial N}{\partial Z} - \frac{\kappa_0}{m_0} N, \tag{20}$$

where

$$\frac{1}{m_0} = F_0(Z), \quad \mu_0 = F_0' + \lambda F_0 + v_0, \quad \kappa_0 = k^2 - \lambda \frac{F_0'}{F_0} - \lambda'.$$

We consider the case $F_0(Z) \gg \mathrm{Pe}^{-1}$ for all Z. Equation (20) is written in dimensionless form, the coordinate Z is measured in units Λ_T, the time t is measured in units Λ_T^2/D_T, the wave number k and value λ are measured in units Λ_T^{-1}, $v_0 = v_s \Lambda_T / D_T$, Λ_T is the characteristic scale of the spatial temperature distribution, the temperature T is measured in units of temperature difference δT in the scale Λ_T, and the concentration N is measured in units N_*. The vector $\boldsymbol{\lambda} = \lambda \hat{\mathbf{e}}_z$, where $\hat{\mathbf{e}}_z$ is a unit vector directed along the axis Z.

Substitution of

$$N(t,Z) = \Psi_0(Z) \exp(\gamma t) \exp\left[-\frac{1}{2} \int \chi_0 dZ \right] \tag{21}$$

reduces Eq. (20) to the eigenvalue problem for the Schrödinger equation

$$\frac{1}{m_0} \Psi_0'' + [W_0 - U_0] \Psi_0 = 0, \tag{22}$$

where $W_0 = -\gamma$ and the potential U_0 is given by

$$U_0 = \frac{1}{m_0} \left(\frac{\chi_0^2}{4} + \frac{\chi_0'}{2} + \kappa_0 \right), \quad \chi_0 = \mu_0 m_0 = \frac{F_0'}{F_0} + \lambda + \frac{v_0}{F_0}. \tag{23}$$

Now we use a quantum-mechanical analogy for the analysis of inhomogeneity formation in a spatial distribution of the concentration of the gaseous admixture. The instability ($\gamma > 0$) can be excited if there is a region of a potential well where $U_0 < 0$. The positive value of W_0 corresponds to turbulent diffusion, whereas a negative value of W_0 results in the excitation of the instability. Now we introduce the function $f = \ln \langle \mathbf{u}^2 \rangle$, where $f' = F_0'/F_0$. The potential U_0 can be rewritten as

$$U_0 = \frac{1}{4 m_0} \{ (f' - \lambda)^2 + [\lambda + v_0 \exp(-f)]^2 + 4k^2 + 2f'' - 2\lambda' - \lambda^2 \}. \tag{24}$$

The potential U_0 can be negative if

$$2f'' - 2\lambda' - \lambda^2 < 0. \tag{25}$$

A. Estimation of the growth rate of the instability

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. (22) can be regarded as the Schrödinger equation with a variable mass $m_0(Z)$. Now we rewrite Eq. (22) in the form

$$\hat{H}\Psi_0 = W_0\Psi_0, \quad \hat{H} = U_0 - \frac{1}{m_0}\frac{d^2}{dZ^2}. \tag{26}$$

The modified variational method employs an inequality

$$W_0 \ll I, \quad I = \int m_0 \Psi^* \hat{H}\Psi\, dZ, \tag{27}$$

where Ψ is an arbitrary function that satisfies a normalization condition

$$\int m_0 \Psi^* \Psi\, dZ = 1. \tag{28}$$

The inequality (27) can be proved if one uses the expansion $\Psi = \Sigma_{p=0}^{\infty} a_p \Psi_0^{(p)}$, where $\Sigma_{p=0}^{\infty} |a_p^2| = 1$ 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

$$\Psi = A_0 \exp[-\alpha(Z - Z_0)^2/2],$$

$$A_0 = \left(\frac{\alpha + b_0}{\pi}\right)^{1/4} \exp\left(\frac{\alpha b_0 Z_0^2}{2(\alpha + b_0)}\right), \tag{29}$$

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. (27)]. Here we use the spatial distributions of $f(Z)$ and $\lambda(Z)$,

$$f(Z) = b_0 Z^2 \exp(-\beta_0 Z^2), \tag{30}$$

$$\lambda(Z) = (Z - \lambda_0)\exp(-\epsilon_0 Z^2), \tag{31}$$

where $\beta_0 \ll 1$ and $\epsilon_0 \ll 1$. These distributions satisfy the necessary condition (25) for excitation of the instability. Substituting Eqs. (29) and (31) into Eq. (27) yields

$$I = \frac{(\alpha + b_0)^{1/2}}{2\alpha^{3/2}}\left[2\alpha\left(Z_0 b - \frac{\lambda_0}{2}\right)^2 + (b - \alpha)^2 + 2\alpha k^2\right]$$
$$\times \exp\left(\frac{\alpha b_0 Z_0^2}{\alpha + b_0}\right) - \frac{v_0\lambda_0}{2} + \frac{v_0 \alpha Z_0}{2(\alpha + b_0)} + \frac{v_0^2}{4}$$
$$\times \left(\frac{\alpha + b_0}{\alpha + 2b_0}\right)^{1/2} \exp\left(-\frac{\alpha^2 b_0 Z_0^2}{(\alpha + b_0)(\alpha + 2b_0)}\right), \tag{32}$$

where $b = 1/2 - b_0 > 0$.

Thus the modified Rayleigh-Ritz method allows us to estimate the growth rate of the instability

$$\gamma = -W_0 \sim \frac{\lambda_0^2 \eta}{2}\left[1 - \frac{\eta}{2\sqrt{1 + c_0}} \exp\left(-\frac{c_0 \lambda_0^2 Y^2}{2(1 + c_0)}\right)\right]$$
$$- \frac{k^2}{\sqrt{1 - c_0}} \exp\left(\frac{c_0 \lambda_0^2 Y^2}{2(1 - c_0)}\right), \tag{33}$$

where $\eta = v_0/\lambda_0$, $c_0 = 2b_0$, $Z_0 = -\lambda_0 Y/(1 - c_0)$, and Y is determined from the equation

$$(Y + 1)^2 = 2\eta Y \sqrt{1 - c_0} \exp\left(-\frac{c_0 \lambda_0^2 Y^2}{2(1 - c_0)}\right).$$

Define a critical value of Y at which $\gamma = 0$, i.e.,

$$Y_{\rm cr}^2 = \frac{2(1 + c_0)}{c_0 \lambda_0^2} \ln\left(\frac{\eta}{2\sqrt{1 + c_0}}\right). \tag{34}$$

Then we can rewrite the growth rate of the instability in the form

$$\gamma \sim \frac{\lambda_0^2 \eta}{2}\left[1 - \left(\frac{4(1 + c_0)}{\eta^2}\right)^p\right], \tag{35}$$

where

$$p = \frac{1}{2}\left(\frac{Y^2}{Y_{\rm cr}^2} - 1\right).$$

Here we consider the case of $k \ll 1$. This implies long-wavelength perturbations in the horizontal plane. It is seen from Eqs. (34) and (35) that the instability is excited when

$$\frac{v_0}{\lambda_0} > 2\sqrt{1 + 2b_0} \tag{36}$$

and $Y > Y_{\rm cr}$. For example, when $b_0 \ll 1$ (i.e., the inhomogeneity of turbulence is very weak), the growth rate of the instability is given by

$$\gamma \sim b_0 \lambda_0^4 Y_0^2, \tag{37}$$

where $Y_0 = \eta - 1 + \sqrt{\eta(\eta - 2)}$. Thus it is shown here that the equilibrium distribution of concentration of the gaseous admixture is unstable. The instability results in the formation of an inhomogeneous distribution of concentration of the gaseous admixture. The exponential growth during the linear stage of the instability can be damped by the nonlinear effects (e.g., a hydrodynamic interaction between the gaseous admixture and a turbulent fluid flow and a change of temperature distribution in the vicinity of the temperature inversion layer).

B. Numerical study of the instability

Equation (22) was solved numerically with turbulent kinetic-energy and mean temperature profiles given by Eqs. (30) and (31). The extremum of turbulent kinetic energy is located at $Z = 0$, the temperature minimum is located at $Z = \lambda_0$ [see Eqs. (30) and (31)], and $Z = -H$ is the location of an impenetrable boundary for the gaseous admixture. The boundary condition at $Z = -H$ is determined by the equation $\mathbf{J}_T|_{Z=-H} = 0$, which yields the condition for Ψ_0

$$\frac{d\Psi_0}{dZ} = \left(f' - \frac{1}{2}\chi_0\right)\Psi_0 \quad \text{at} \quad Z = -H. \tag{38}$$

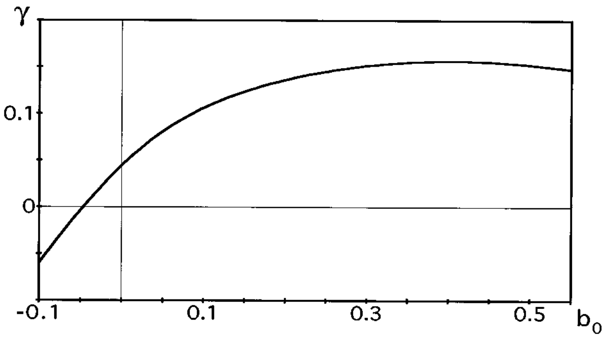

FIG. 1. Dependence of the growth rate of the instability versus b_0 for $\lambda_0=1$, $v_0=2.2$, and $H=7$.

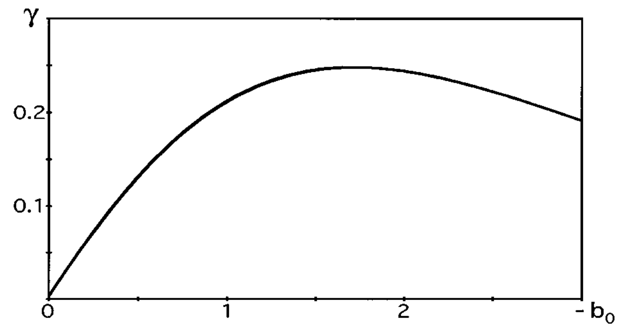

FIG. 3. Dependence of the growth rate of the instability versus $-b_0$ for $\lambda_0=-0.2$, $v_0=0$, and $H=0.9$.

Equation (38) provides zero flux of the gaseous admixture through a horizontal boundary plane $Z=-H$. The second boundary condition is $\Psi_0(Z=\infty)=0$. As an example, Fig. 1 shows the dependence of the growth rate of the instability versus b_0 for $\lambda_0=1$, $v_0=2.2$, and $k\ll 1$. These values of the parameters satisfy the necessary condition (36) for the excitation of the instability. The minimum of the function $f(Z)$ describing the spatial distribution of the turbulent kinetic energy is chosen to be located at the height $H=7$ from the horizontal boundary plane, where H is measured in units of Λ_T. These numerical results are in good agreement with the analytical estimates obtained by means of the modified Rayleigh-Ritz method (Sec. IV A). The instability is excited when $0<b_0<0.57$. For illustration, the spatial distributions of the potential $U(Z)$ are presented in Fig. 2(a) (for $b_0=0.1$) and in Fig. 2(b) (for $b_0=0.5$). In the vicinity of the upper bound for the parameter b_0, the spatial distributions of the potential $U(Z)$ sharply change. Note that the analytical estimate obtained by means of the modified Rayleigh-Ritz method yields the upper bound for $b_0=0.5$ in good agreement with the numerical results.

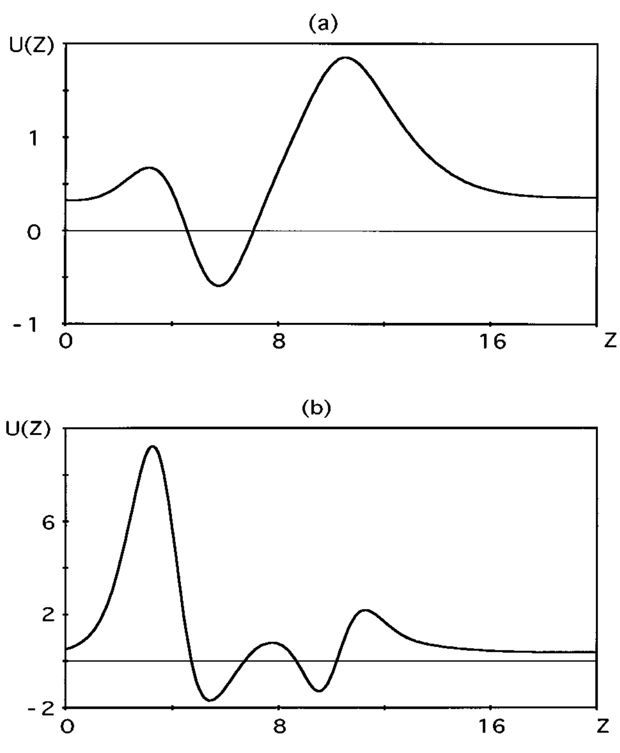

FIG. 2. Distributions of the potential $U(r)$ for $\lambda_0=1$, $v_0=2.2$, $H=7$, and different b_0: (a) $b_0=0.1$ and (b) $b_0=0.5$.

The effect of the boundary was neglected in the modified Rayleigh-Ritz method. Note that in the case $H=7$ the numerical simulation showed that the effect of the boundary is indeed very small. However, if the boundary is located not far from the extrema of the temperature and turbulent kinetic-energy distributions, the modified Rayleigh-Ritz method does not allow an estimate of the growth rate of the instability. However, this case can be interesting in view of its relevance to atmospheric turbulent transport and can be studied numerically. For example, the dependence of the growth rate of the instability versus $-b_0$ for $H=0.9$ is shown in Fig. 3. In this case the instability is excited even for very small terminal fall velocity (the calculations were performed for $v_0=0$). The gaseous admixture is accumulated in the region between the maximum of the turbulent kinetic energy (e.g., $b_0<0$) and minimum of the temperature, which is located under the maximum of turbulent kinetic energy (the calculations were performed for $\lambda_0=-0.2$). Note that the case $v_0=0$ describes the large-scale dynamics of the gaseous admixture when the terminal fall velocity is very small. On the other hand, $v_0\neq 0$ corresponds to the dynamics of light particles in a turbulent fluid flow.

V. CONCLUSION

We demonstrated the existence of two effects: turbulent barodiffusion and turbulent thermal diffusion. These effects occur during advection of the gaseous admixture by a compressible turbulent fluid flow with low Mach numbers. These phenomena result in the appearance of two additional turbulent nondiffusive fluxes in the equation for the total mass flux of the gaseous admixture. The magnitude of turbulent barodiffusion and turbulent thermal diffusion fluxes is much larger than the corresponding molecular fluxes at large Péclet and Reynolds numbers. The effects vanish in incompressible turbulent flow of a surrounding fluid. These phenomena can cause formation of the large-scale (mean-field) inhomogeneous structures in a spatial concentration distribution of the gaseous admixture. The effects of turbulent barodiffusion and turbulent thermal diffusion can be important in various natural and industrial turbulent compressible fluid flows, e.g., the formation of gaseous clouds in turbulent atmospheres in the vicinity of a temperature inversion layer.

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APPENDIX A: SOLUTION OF EQ. (1)

Here we show that Eq. (3) is a solution of Eq. (1). If the total field n_p is specified at time t, then we can determine the total field $n_p(t+\Delta t)$ at time $t+\Delta t$ by means of the substitutions $t\to t+\Delta t$ and $t_0\to t$ in Eq. (3). The result is given by

$$n_p(t+\Delta t,\mathbf{x})=M\{G(t+\Delta t,t)n_p[t,\boldsymbol{\xi}(t+\Delta t,t)]\}, \quad \text{(A1)}$$

where

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

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

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

We expand the function $n_p(t,\boldsymbol{\xi}_{\Delta t})$ in Eq. (A1) in a Taylor series in the vicinity of the point $\mathbf{x}$:

$$n_p(t,\boldsymbol{\xi}_{\Delta t})\simeq n_p(t,\mathbf{x})+\frac{\partial n_p}{\partial x_m}(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_m+\frac{1}{2}\frac{\partial^2 n_p}{\partial x_m\partial x_s}\times(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_m(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_s+\cdots. \quad \text{(A2)}$$

Using the equation for the Wiener trajectory we obtain

$$[\boldsymbol{\xi}(t_2,t_1)-\mathbf{x}]_m=-\int_0^{t_2-t_1}U_m(t_s,\boldsymbol{\xi}_s)ds+(2D)^{1/2}\mathbf{w}_m(t_2-t_1), \quad \text{(A3)}$$

where $\boldsymbol{\xi}(t_2,t_2-s)\equiv\boldsymbol{\xi}_s$. Expanding the velocity $U_m(t_s,\boldsymbol{\xi}_s)$ in a Taylor series in the vicinity of the point $\mathbf{x}$ and using Eq. (A3) we find that

$$U_m(t_s,\boldsymbol{\xi}_s)=U_m(t_s,\mathbf{x})+\frac{\partial U_m}{\partial x_l}\left[-\int_0^s U_l(t_\sigma,\mathbf{x})d\sigma+(2D)^{1/2}\mathbf{w}_l(s)\right]. \quad \text{(A4)}$$

Substitution of Eq. (A4) into Eq. (A3) yields

$$(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_m=-\int_0^{\Delta t}U_m(t_s,\boldsymbol{\xi}_s)ds+\int_0^{\Delta t}\frac{\partial U_m}{\partial x_l}\bigg|_{(t_s,\mathbf{x})}ds\int_0^s U_l(t_\sigma,\boldsymbol{\xi}_\sigma)d\sigma-\sqrt{2D}\int_0^{\Delta t}\frac{\partial U_m}{\partial x_l}\bigg|_{(t_s,\mathbf{x})}w_l(s)ds+\sqrt{2D}w_m. \quad \text{(A5)}$$

The integrals in formula (A5) can be evaluated by means of the ''mean value'' theorem. The result is given by

$$(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_m\simeq-U_m(t_*,\mathbf{x})\Delta t+\sqrt{2D}w_m+O((\Delta t)^2), \quad \text{(A6)}$$

where t_* is within the interval $(t,t+\Delta t)$. Substitution of Eq. (A6) into Eq. (A2) yields an expression for the field $n_p(t,\boldsymbol{\xi}_{\Delta t})$,

$$n_p(t,\boldsymbol{\xi}_{\Delta t})\simeq n_p(t,\mathbf{x})+\frac{\partial n_p}{\partial x_m}[-U_m(t_*,\mathbf{x})\Delta t+\sqrt{2D}w_m]+Dw_m w_s\frac{\partial^2 n_p}{\partial x_m\partial x_s}+O((\Delta t)^2). \quad \text{(A7)}$$

Expanding the function $b_*(\sigma,\boldsymbol{\xi}_\sigma)$ in a Taylor series in the vicinity of the point $\mathbf{x}$, using Eq. (A6), and evaluating the integral

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

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

$$G(t+\Delta t,t)\simeq 1-b_*(t_3,\mathbf{x})\Delta t, \quad \text{(A8)}$$

where t_3 is within the interval $(t,t+\Delta t)$. Combining Eqs. (A7), (A8) and (A1) and averaging over Wiener trajectories yields the expression for the field $n_p(t+\Delta t,\mathbf{x})$. Now we calculate the limit $[n_p(t+\Delta t,\mathbf{x})-n_p(t,\mathbf{x})]/\Delta t$ for $\Delta t\to 0$. This procedure yields Eq. (1). Therefore Eq. (3) is a solution of Eq. (1).

APPENDIX B: DERIVATION OF THE MEAN-FIELD EQUATION FOR LARGE PÉCLET NUMBER

We derive here the equation for the mean field. If the total field n_p is specified at time t, then we can determine the total field $n_p(t+\Delta t)$ at time $t+\Delta t$ by means of substitutions $t\to t+\Delta t$ and $t_0\to t$ in Eq. (3). The result is given by

$$n_p(t+\Delta t,\mathbf{x})=M\{G(t+\Delta t,t)n_p[t,\boldsymbol{\xi}(t+\Delta t,t)]\}, \quad \text{(B1)}$$

where

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

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

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

Expanding the function $n_p(t,\boldsymbol{\xi}_{\Delta t})$ [Eq. (B1)] in a Taylor series in the vicinity of the point $\mathbf{x}$ yields

$$n_p(t,\boldsymbol{\xi}_{\Delta t})\simeq n_p(t,\mathbf{x})+\frac{\partial n_p}{\partial x_m}(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_m+\frac{1}{2}\frac{\partial^2 n_p}{\partial x_m\partial x_s}\times(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_m(\boldsymbol{\xi}_{\Delta t}-\mathbf{x})_s+\cdots. \tag{B2}$$

Using the equation for the Wiener trajectory we obtain

$$[\boldsymbol{\xi}(t_2,t_1)-\mathbf{x}]_m=-\int_0^{t_2-t_1}U_m(t_s,\boldsymbol{\xi}_s)ds+(2D)^{1/2}\mathbf{w}_m(t_2-t_1), \tag{B3}$$

where $\boldsymbol{\xi}(t_2,t_2-s)\equiv\boldsymbol{\xi}_s$. Expanding the velocity $U_m(t_s,\boldsymbol{\xi}_s)$ in a Taylor series in the vicinity of the point $\mathbf{x}$ and using Eq. (B3) yield

$$U_m(t_s,\boldsymbol{\xi}_s)\simeq U_m(t_s,\mathbf{x})-U_l\frac{\partial U_m}{\partial x_l}s+(2D)^{1/2}\frac{\partial U_m}{\partial x_l}\mathbf{w}_l(s)+\cdots. \tag{B4}$$

Here we assume that the velocity $\mathbf{U}$ remains constant (time independent) at small time intervals $(0,\Delta t),(\Delta t,2\Delta t),\ldots$ and changes every small time Δt and that the velocity is statistically independent at different time intervals. Substituting Eq. (B4) into Eq. (B3) and calculating the integrals in Eq. (B3) accurate up to $\sim(t_2-t_1)^2$ terms yield

$$[\boldsymbol{\xi}(t_2,t_1)-\mathbf{x}]_m\simeq-(t_2-t_1)U_m+\frac{1}{2}(t_2-t_1)^2U_l\frac{\partial U_m}{\partial x_l}-\sqrt{2D}\frac{\partial U_m}{\partial x_l}\int_0^{t_2-t_1}w_l ds+\sqrt{2D}w_m(t_2-t_1)+\cdots. \tag{B5}$$

The combination of Eqs. (B5) and (B2) yields the formula for the field $n_p(t,\boldsymbol{\xi}_{\Delta t})$

$$n_p(t,\boldsymbol{\xi}_{\Delta t})=n_p(t,\mathbf{x})+\frac{\partial n_p}{\partial x_m}\left(-U_m\Delta t+\frac{1}{2}U_l\frac{\partial U_m}{\partial x_l}(\Delta t)^2+\sqrt{2D}w_m-\sqrt{2D}\frac{\partial U_m}{\partial x_l}\int_0^{\Delta t}w_l ds\right)+\frac{1}{2}\frac{\partial^2 n_p}{\partial x_m\partial x_l}[U_mU_l(\Delta t)^2+2Dw_mw_l-\sqrt{2D}\Delta t(U_mw_l+U_lw_m)], \tag{B6}$$

where we keep terms $\geqslant O((\Delta t)^2)$. Now we expand the function $b_*(\sigma,\boldsymbol{\xi}_\sigma)$ in a Taylor series in the vicinity of the point $\mathbf{x}$, use Eq. (B5), and calculate the integral

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

The result is given by

$$\int_t^{t+\Delta t}b_*(\sigma,\boldsymbol{\xi}_\sigma)d\sigma\simeq b_*(t,\mathbf{x})\Delta t-\frac{1}{2}U_q\frac{\partial b_*}{\partial x_q}(\Delta t)^2+\sqrt{2D}\frac{\partial b_*}{\partial x_q}\int_t^{t+\Delta t}w_q d\sigma+\cdots. \tag{B7}$$

Here we also keep terms $\geqslant O((\Delta t)^2)$. Using Eq. (B7) we calculate the function $G(t+\Delta t,t)$ accurate up to $\sim(\Delta t)^2$ terms

$$G(t+\Delta t,t)\simeq 1-b_*(t,\mathbf{x})\Delta t+\frac{1}{2}U_q\frac{\partial b_*}{\partial x_q}(\Delta t)^2+\frac{1}{2}b_*^2(\Delta t)^2-\sqrt{2D}\frac{\partial b_*}{\partial x_q}\int_t^{t+\Delta t}w_q d\sigma+\cdots. \tag{B8}$$

Substitution of Eqs. (B8) and (B6) into Eq. (B1) allows us to determine the number density $n_p(t+\Delta t,\mathbf{x})$,

$$n_p(t+\Delta t,\mathbf{x})=M\left\{n_p(t,\mathbf{x})+n_1(\Delta t)+n_2(\Delta t)^2+D\frac{\partial^2 n_p}{\partial x_m\partial x_l}w_mw_s\right\}, \tag{B9}$$

where

$$n_1=U_m\frac{\partial n_p}{\partial x_m}-b_*n_p,$$

$$n_2=\frac{\partial n_p}{\partial x_m}\left(\frac{1}{2}U_l\frac{\partial U_m}{\partial x_l}+b_*U_m\right)+\frac{1}{2}n_p\left(U_l\frac{\partial b_*}{\partial x_l}+b_*^2\right)+\frac{1}{2}\frac{\partial^2 n_p}{\partial x_m\partial x_l}U_mU_l.$$

Averaging Eq. (B9) over the ensemble of the turbulent velocities, we obtain the mean field $N(t+\Delta t,\mathbf{x}) = \langle n_p(t+\Delta t,\mathbf{x})\rangle$. Let us calculate the limit $[N(t+\Delta t,\mathbf{x}) - N(t,\mathbf{x})]/\Delta t$ for $\Delta t \to 0$. The result is given by

$$\frac{\partial N}{\partial t} + [(\mathbf{V} - \langle \tau(\mathbf{u}\cdot\nabla)\mathbf{u}\rangle - 2\langle \tau b\mathbf{u}\rangle)\cdot\nabla]N = B_{\mathrm{eff}}N + D_{pm}\frac{\partial^2 N}{\partial x_p \partial x_m}, \tag{B10}$$

where $B_{\mathrm{eff}} = -(\nabla\cdot\mathbf{V}) + \langle \tau(\mathbf{u}\cdot\nabla)b\rangle + \langle \tau b^2\rangle$. 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. (6) for the mean field N. Here we neglect a weak dependence of τ on the coordinates.

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