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Turbulent Transport of Atmospheric Aerosols and Formation of Large-Scale Structures

T. Elperin[1], N. Kleeorin[1], I. Rogachevskii[1] and D. Sokoloff[2]

[1]The Pearlstone Center for Aeronautical Engineering Studies, Department of Mechanical Engineering, Ben-Gurion University of the Negev, Beer-Sheva 84105, P. O. Box 653, Israel
[2]Department of Physics, Moscow State University, 117234 Moscow, Russia

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Abstract. Turbulent transport of aerosols and droplets in a random velocity field with a finite correlation time is studied. We derived a mean-field equation and an equation for the second moment for a number density of aerosols. The finite correlation time of random velocity field results in the appearance of the high-order spatial derivatives in these equations. The finite correlation time and compressibility of the velocity field can cause a depletion of turbulent diffusion and a modification of an effective mean drift velocity. The coefficient of turbulent diffusion in the vertical direction can be depleted by 25 % due to the finite correlation time of a turbulent velocity field. The latter result is in compliance with the known anisotropy of the coefficient of turbulent diffusion in the atmosphere. The effective mean drift velocity is caused by a compressibility of particles velocity field and results in formation of large-scale inhomogeneities in spatial distribution of aerosols in the vicinity of the atmospheric temperature inversion. Results obtained by Saffman (1960) for the effect of molecular diffusivity in turbulent diffusion are generalized for the case of compressible and anisotropic random velocity field. A mechanism of formation of small-scale inhomogeneities in particles spatial distribution is also discussed. This mechanism is associated with an excitation of a small-scale instability of the second moment of number density of particles. The obtained results are important in the analysis of various atmospheric phenomena, e.g., atmospheric aerosols, droplets and smog formation.

1 Introduction

Mean-field theory for turbulent transport of particles and gases is of a great importance in view of numerous applications. In particular, this theory is applied for analysis of transport of aerosols, pollutants and cloud droplets in atmospheric turbulence of the Earth and other planets (see, e.g., Twomey, 1977; Csanady, 1980; Seinfeld, 1986; Elperin et al., 1997; Elperin et al., 2000a; and references therein), dust transfer in interstellar turbulence and turbulent transport of particles and gases in industrial flows (see, e.g., Piterbarg and Ostrovskii, 1997; Baldyga and Bourne, 1999; and references therein). Turbulent transport of aerosols in turbulent atmosphere was studied in a number of publications. However, a range of validity and applicability of mean-field equation for number density of aerosols still remain a subject of discussions. In particular, it is not elucidated why the mean-field equation for number density of aerosols does not contain high-order spatial derivatives and what is a role of the molecular diffusion.

Problem of formation and dynamics of aerosol and gaseous clouds is of fundamental significance in many areas of environmental sciences, physics of the atmosphere and meteorology. Analysis of experimental data shows that spatial distributions of droplets in cumulus and stratiform clouds is strongly inhomogeneous (see, e.g., Paluch and Baumgardner, 1989; Korolev and Mazin, 1993; Haman and Malinowski, 1996; and references therein). One of the mechanisms which determines formation and dynamics of clouds is the preferential concentration of atmospheric particles and droplets. However, in turbulent atmosphere a mechanism of concentration of atmospheric particles in nonconvective clouds is still a subject of active research. It is well-known that turbulence results in a decay of inhomogeneities of concentration due to turbulent diffusion, whereas the opposite process, e.g., the preferential concentration of particles in atmospheric turbulent fluid flow still remains poorly understood.

In the present study we derived a mean-field equation and an equation for the second moment for a number density of inertial particles (aerosols and droplets) advected by a turbulent velocity field with a finite correlation time. We have shown that the finite correlation time of random velocity field results in the appearance of the high-order spatial derivatives in the equations for a number density of inertial particles, and causes strong depletion of the coefficient of turbulent diffusion in the vertical direction and a modification of an effective mean drift velocity. The effective mean drift velocity

Correspondence to: T. Elperin

is caused by a compressibility of particles velocity field and results in formation of large-scale inhomogeneities in spatial distribution of aerosols in the vicinity of the atmospheric temperature inversion. A mechanism of formation of small-scale inhomogeneities in particles spatial distribution is also discussed.

2 Mean-field equations for a number density of small inertial particles

Number density $n(t, \mathbf{r})$ of small inertial particles advected by a turbulent compressible fluid flow is given by

$$\partial n / \partial t + \boldsymbol{\nabla} \cdot (n\mathbf{v}) = D\Delta n \ , \qquad (1)$$

where $\mathbf{v}$ is a random velocity of inertial particles or droplets which they acquire in turbulent atmospheric fluid velocity field, D is the coefficient of molecular (Brownian) diffusion.

The goal of the present study is to derive a mean-field equation for inertial particles advected by turbulent atmospheric velocity field with a finite correlation time. For the derivation of this equation we used an exact solution of the equation for $n(t, \mathbf{r})$ in the form of a functional integral for an arbitrary velocity field taking into account a small yet finite molecular diffusion (see Elperin et al., 2000b). This functional integral implies an averaging over random Brownian motions of a particle. The form of the exact solution used in the present paper allows us to separate the averaging over both, a random Brownian motions of a particle and turbulent atmospheric velocity field.

The derived mean-field equation for a number density of small inertial particles is given by

$$N(t, \mathbf{r}) = M_{\boldsymbol{\xi}}\{\langle G(t, s, \boldsymbol{\xi}) \exp(\boldsymbol{\xi}^* \cdot \boldsymbol{\nabla})\rangle\} N(s, \mathbf{r}) \ , \qquad (2)$$

(see, Elperin et al., 2000b), where $N(t, \mathbf{r}) = \langle n(t, \mathbf{r})\rangle$ is the mean number density of particles, the angular brackets $\langle\cdot\rangle$ denote the ensemble average over the random velocity field,

$$G(t, s, \boldsymbol{\xi}) = \exp[-\int_s^t b(\sigma, \boldsymbol{\xi}) \, d\sigma] \ ,$$

$b = \boldsymbol{\nabla} \cdot \mathbf{v}$, and $M_{\boldsymbol{\xi}}\{\cdot\}$ denotes the average over the Wiener paths

$$\boldsymbol{\xi} = \mathbf{x} - \int_0^{t-s} \mathbf{v}[t - \sigma, \boldsymbol{\xi}(t, \sigma)] \, d\sigma + (2D)^{1/2} \mathbf{w}(t - s) \ ,$$

$\boldsymbol{\xi}^* = \boldsymbol{\xi} - \mathbf{x}$ and $\mathbf{w}(t)$ is the Wiener process.

The derived mean-field equation for a number density of small inertial particles generally is a integro-differential equation. However, when the characteristic scale of variation of the mean number density of small inertial particles is much larger than the correlation length of a random velocity field the mean field equation has the form of a second-order differential equation in spatial derivatives.

Indeed, in order to simplify mean-field equation for a number density of small inertial particles we use two models of random velocity field: the model of the velocity field with a small correlation time and the model of a random velocity field with Gaussian statistics for the Wiener trajectories. In the model of the velocity field with a small correlation time we expand the functions $\boldsymbol{\xi}(t, s)$ and $G(\tau, \boldsymbol{\xi})$ in Taylor series of small correlation time τ. Thus an equation for the mean number density of small inertial particles is given by

$$\partial N / \partial t + (\mathbf{V}_{\text{eff}} \cdot \boldsymbol{\nabla}) N = D_{mn} \nabla_m \nabla_n N \ , \qquad (3)$$

where

$$\begin{aligned} D_{mn} &= D\delta_{mn} + \langle v_m v_n \rangle \tau - 2\langle b v_m v_n \rangle \tau^2 \\ &\quad + (D2\tau^2/3)(\Delta f_{mn} + \nabla_p \nabla_n f_{mp} \\ &\quad + \nabla_p \nabla_m f_{np})_{r=0} \ , \\ \mathbf{V}_{\text{eff}} &= \mathbf{W} - \langle \mathbf{v} b \rangle \tau + (4D/3)\langle (\nabla_p \mathbf{v})(\nabla_p b) \rangle \tau^2 \\ &\quad + 2\langle b(\mathbf{v} \cdot \boldsymbol{\nabla})\mathbf{v} \rangle \tau^2 \ , \end{aligned}$$

$\mathbf{W}$ is the terminal fall velocity, $f_{mn} = \langle v_m(t, \mathbf{x}) v_n(t, \mathbf{y})\rangle$, $\mathbf{r} = \mathbf{y} - \mathbf{x}$, $\boldsymbol{\nabla} = \partial / \partial \mathbf{r}$. The last term in the equation for D_{mn} describes interactions between turbulent diffusion and molecular diffusion for the mean concentration field and generalizes the result by Saffman (1960) to the case of compressible and anisotropic random velocity field.

In the model of a random velocity field with Gaussian statistics for the Wiener trajectories the equations for the mean number density of particles N has a form of Eq. (3) with

$$\begin{aligned} D_{mn} &= D_T\{\delta_{mn} - (3/2)[(V_{\text{eff}} - \mathbf{W})/u_0]^2 e_m e_n\} \ , \\ \mathbf{V}_{\text{eff}} &= \mathbf{W} - \tau \langle \mathbf{v} b \rangle \ . \end{aligned}$$

Here $D_T = u_0 l_0 / 3$ is the coefficient of turbulent diffusion, l_0 is the maximum scale of turbulent motions, u_0 is the characteristic velocity in the scale l_0, e_m is the unit vector in the direction opposite to the gravity $\mathbf{g}$. The last term in the equation for D_{mn} describes a depletion of the turbulent diffusion coefficient due to the finite correlation time of a random velocity field. The effective velocity $\mathbf{V}_{\text{eff}}$ of particles determines a turbulent contribution to particle velocity due to both, effect of inertia and mean temperature gradient. The effective velocity $\mathbf{V}_{\text{eff}}$ can be estimated as

$$\mathbf{V}_{\text{eff}} = -(2/3) W A(\text{Re}, a_*) \Lambda_P \ln(\text{Re}) (\boldsymbol{\nabla} T) / T \ , \qquad (4)$$

where $\Lambda_P = |\boldsymbol{\nabla} P_f / P_f|^{-1}$, P_f is the fluid pressure, $\text{Re} = l_0 u_0 / \nu$ is the Reynolds number, and ν is the kinematic viscosity, a_* is the size of a particle, $A(\text{Re}, a_*) = 1$ for $a_* < a_{\text{cr}}$, and $A(\text{Re}, a_*) = 1 - 3\ln(a_*/a_{\text{cr}})/\ln(\text{Re})$ for $a_* \geq a_{\text{cr}}$, and $a_{\text{cr}} = r_d (\rho/\rho_p)^{1/2}$, and $r_d = l_0 \text{Re}^{-3/4}$ is the viscous scale of turbulent fluid flow, ρ is the fluid density, ρ_p is the material density of particles, T is the mean fluid temperature. Thus, e.g., $a_{\text{cr}} \sim 20\mu$m for $\text{Re} = 10^7$, $l_0 = 100$m and $\rho_p = 1$ g / cm^3. The effective velocity $\mathbf{V}_{\text{eff}}$ can cause formation of inhomogeneities in the spatial distribution of inertial particles. The turbulent flux of particles is given by

$$\mathbf{J}_T = N \mathbf{V}_{\text{eff}} - D_T \boldsymbol{\nabla} N \ . \qquad (5)$$

The additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{\text{eff}}$ can be also estimated as follows.

We average Eq. (1) over the ensemble of the turbulent velocity fluctuations and subtract the obtained averaged equation from (1). This yields equation for the turbulent component $\Theta = n - N$ of the particles number density

$$\partial\Theta/\partial t - D\Delta\Theta = -\nabla\cdot(N\mathbf{v} + \mathbf{Q}) \,, \tag{6}$$

where $\mathbf{Q} = \mathbf{v}\Theta - \langle\mathbf{v}\Theta\rangle$. Equation (6) is written in a frame moving with the mean velocity $\mathbf{V}$. The magnitude of $\partial\Theta/\partial t - D\Delta\Theta + \nabla\cdot\mathbf{Q}$ can be estimated as Θ/τ, where τ is the correlation time of the velocity field. Thus the turbulent field Θ is of the order of

$$\Theta \sim -\tau N(\nabla\cdot\mathbf{v}) - \tau(\mathbf{v}\cdot\nabla)N \,.$$

Now we calculate the turbulent flux of particles $\mathbf{J}_T = \langle\mathbf{v}\Theta\rangle$:

$$\mathbf{J}_T \sim -N\langle\tau\mathbf{v}(\nabla\cdot\mathbf{v})\rangle - \langle\tau\mathbf{v}v_j\rangle\nabla_j N \,. \tag{7}$$

The first term in Eq. (7) describes the additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{\rm eff}$. Notably, similar turbulent cross-effects can occur in turbulent fluid flows with chemical reactions, or phase transitions, or fast rotation (see Elperin et al., 1998b; 1998c).

Remarkably, Eq. (4) for the effective velocity of particles provides local parameterization of these turbulence effects, and it can be directly incorporated to existing atmospheric numerical models. It is seen from Eq. (4) that the ratio $|\mathbf{V}_{\rm eff}/W|$ is of the order of

$$|\mathbf{V}_{\rm eff}/W| \sim (\Lambda_P/\Lambda_T)(\delta T/T_*)\ln \mathrm{Re}$$

(see Elperin et al., 1996a; 1998a; 2000a), where δT is the temperature difference in the scale Λ_T, and T_* is the characteristic temperature. Note that the additional particle velocity $\mathbf{V}_{\rm eff}$ is of the order of the terminal fall velocity W in the vicinity of the atmospheric temperature inversion (see Section 4). In the atmosphere without temperature inversion the effective particle velocity is directed opposite to the terminal fall velocity, and the effective particle velocity decreases the effective sedimentation velocity by 10 - 30 percents. In the atmosphere with a temperature inversion the effective particle velocity $\mathbf{V}_{\rm eff}$ is directed to the temperature minimum and it results in accumulation of particles in the vicinity of the temperature inversion.

The additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{\rm eff}$ results in formation of inhomogeneities of aerosols distribution whereby initial spatial distribution of particles in the turbulent atmosphere 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 the mean atmospheric temperature. In particular, the instability can be excited in the vicinity of the minimum in the mean temperature (see Elperin et al., 1996a; 1998a). The characteristic time of formation of inhomogeneities of particles is $\tau_f \sim \Lambda_T/|V_{\rm eff} - W|$. The formation of inhomogeneities is possible when $V_{\rm eff} > W$. The initially spatial distribution of the concentration of the inertial particles evolves into a pattern containing regions with increased (decreased) concentration of particles. Characteristic vertical size of the inhomogeneity is of the order of

$$l_f \sim \Lambda_T\left[\left(\frac{W\Lambda_P}{D_T}\right)\left(\frac{\delta T}{T_*}\right)\ln \mathrm{Re}\right]^{-\frac{1}{2}} .$$

Therefore, it is important to take into account the additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{\rm eff}$ in atmospheric phenomena (e.g., atmospheric aerosols, cloud formation and smog formation).

3 Formation of small-scale inhomogeneities in particles spatial distribution

In order to describe a formation of small-scale structures we derived equation for the second-order correlation function

$$\Phi(t,\mathbf{x},\mathbf{y}) = \langle\Theta(t,\mathbf{x})\Theta(t,\mathbf{y})\rangle$$

of number density of particles advected by a random velocity field with a small yet finite correlation time τ. The equation is given by

$$\partial\Phi/\partial t = [B(r) - 2U_m\nabla_m + D_{mn}\nabla_m\nabla_n]\Phi(t,\mathbf{r}) \,, \tag{8}$$

where

$$D_{mn} = 2D\delta_{mn} + 2\tau(\tilde f_{mn} + \mathrm{St}^2 Q_{mn}) \,, \tag{9}$$

$$Q_{mn} = \frac{1}{4}\left(\frac{\partial f_{pn}}{\partial r_s}\frac{\partial f_{ms}}{\partial r_p} - \tilde f_{ps}\frac{\partial^2 f_{mn}}{\partial r_p\partial r_s}\right) + 2A_mA_n + A_p\frac{\partial f_{mn}}{\partial r_p} - \tilde f_{mn}\frac{\partial A_p}{\partial r_p} - \frac{5}{3}\tilde f_{mp}\frac{\partial A_n}{\partial r_p} \,, \tag{10}$$

$$U_m = 2\tau\left[A_m - \frac{\mathrm{St}^2}{12}\left(\frac{\partial A_p}{\partial r_s}\frac{\partial f_{ms}}{\partial r_p} + 10A_s\frac{\partial A_m}{\partial r_s} + 12A_m\frac{\partial A_s}{\partial r_s}\right)\right] , \tag{11}$$

$$B(r) = -2\tau\left[\frac{\partial A_p}{\partial r_p} + \frac{\mathrm{St}^2}{2}\left(\frac{1}{6}\frac{\partial A_p}{\partial r_s}\frac{\partial A_s}{\partial r_p} - \left(\frac{\partial A_p}{\partial r_p}\right)^2\right)\right] , \tag{12}$$

and $A_m = \partial f_{mp}/\partial r_p$, $\tilde f_{mn} = f_{mn}(0) - f_{mn}(\mathbf{r})$, $\mathrm{St} = 2\tau/\tau_0$ is the Strouhal number, $\tau_0 = l_0/u_0$. For simplicity we assumed here that a random velocity field has a Gaussian statistics.

The correlation function f_{mn} for homogeneous, isotropic and compressible velocity field is given by

$$f_{mn} = (u_0^2/3)\{(F + F_c)\delta_{mn} + (rF'/2)P_{mn} + rF_c' r_{mn}\} \,, \tag{13}$$

where $P_{mn}(r) = \delta_{mn} - r_{mn}$, $r_{mn} = r_m r_n/r^2$ and $F' = dF/dr$. The function $F_c(r)$ describes the potential (compressible) component whereas $F(r)$ corresponds to the vortical part of a random velocity field. Note that the condition $\nabla\cdot\mathbf{v} \neq 0$ is associated either with a compressibility of a low-Mach-number compressible fluid flow or with particles inertia (see Maxey, 1987; Elperin et al., 1996a; 1998a). For inertial particles $\nabla \cdot \mathbf{v} = \tau_p \Delta P_f / \rho$ and the degree of compressibility of particles velocity is

$$\sigma = \langle (\nabla \cdot \mathbf{v})^2 \rangle / \langle (\nabla \times \mathbf{v})^2 \rangle = 12 \varepsilon \tau_p^2 / \nu = 12 \mathrm{Re} (\tau_p / \tau_0)^2 ,$$

where τ_p is the Stokes time and $\varepsilon = u_0^2/\tau_0$. The functions $F(r)$ and $F_c(r)$ are chosen as follows. In scales $0 \ll r < 1$ incompressible $F(r)$ and compressible $F_c(r)$ components of the random velocity field are given by $F(r) = (1 - r^2)/(1 + \sigma)$ and $F_c(r) = \sigma F(r)$, where r is measured in the units of the maximum scale of turbulent motions l_0. In scales $r \geq 1$ the functions $F = F_c = 0$.

Turbulent diffusion tensor D_{mn} is determined by the field of Lagrangian trajectories $\boldsymbol{\xi}$. Due to a finite correlation time of a random velocity field the field of Lagrangian trajectories $\boldsymbol{\xi}$ is compressible even if the velocity field is incompressible ($\sigma = 0$). Indeed, for $\sigma = 0$ we obtain $\langle (\nabla \cdot \boldsymbol{\xi})^2 \rangle = (20/3) \mathrm{St}^4$. We will show that the compressibility of the field of Lagrangian trajectories results in the excitation of a small-scale instability of the second moment of particles number density and formation of small-scale inhomogeneities of fluctuations of particles number density even for very small compressibility of a random velocity field with a finite correlation time.

Using Eq. (13) we calculate the functions $D_{mn}(r)$, $U_m(r)$ and $B(r)$. Thus, Eq. (8) can be rewritten in the form

$$\partial \Phi / \partial t = (1/m) \Phi'' + \lambda(r) \Phi' + B \Phi , \qquad (14)$$

where $\Phi' = \partial \Phi / \partial r, \quad \Phi'' = \partial^2 \Phi / \partial r^2, \quad r = |\mathbf{y} - \mathbf{x}|$,

$$1/m = 2(1 + X^2)/\mathrm{Pe} ,$$
$$\lambda = 2[2 + X^2(1 + 2C)]/(r\, \mathrm{Pe}) ,$$

$C = (c_2 + \mathrm{St}^2 c_5)/4\beta$, $X(r) = \sqrt{\mathrm{Pe}\beta}\, r$, and $\mathrm{Pe} = l_0 u_0 / D \gg 1$ is the Peclet number, $\beta = (c_3 + \mathrm{St}^2 c_6)/2$, and $B = c_1 + \mathrm{St}^2 c_4$, and

$$c_1 = \frac{20\sigma}{1+\sigma} , \quad c_2 = \frac{2(19\sigma + 3)}{3(1+\sigma)} ,$$
$$c_3 = \frac{2(3\sigma + 1)}{3(1+\sigma)} , \quad c_4 = 80 \left(\frac{\sigma}{1+\sigma} \right)^2 ,$$
$$c_5 = -\frac{1}{27(1+\sigma)^2} (12 - 1278\sigma - 3067\sigma^2) ,$$
$$c_6 = \frac{1}{27(1+\sigma)^2} (36 + 466\sigma + 2499\sigma^2) ,$$

the distance r is measured in units of l_0, time t is measured in units of τ_0. In order to obtain a solution of Eq. (14) we use a separation of variables, i.e., we seek a solution in the following form: $\Phi(t, r) = \hat{\Phi}(r) \exp(\gamma t)$, whereby γ is a free parameter to be specified by the boundary conditions $\hat{\Phi}(r = 0) = 1$ and $\hat{\Phi}(r \to \infty) = 0$. Since the function $\Phi(t, r)$ is the two-point correlation function, it has a global maximum at $r = 0$ and therefore it satisfies the conditions: $\hat{\Phi}'(r = 0) = 0$, and $\hat{\Phi}''(r = 0) < 0$, and $\hat{\Phi}(r = 0) > |\hat{\Phi}(r > 0)|$. Solution of Eq. (14) can be analyzed using an asymptotic analysis which is based on the separation of scales. In particular, the solution of Eq. (14) has different regions where the form of the functions $m(r)$ and $\lambda(r)$ are different. The functions $\hat{\Phi}(r)$ and $\hat{\Phi}'(r)$ in these different regions are matched at their boundaries in order to obtain continuous solution for the correlation function. Note that the most important part of the solution is localized in small scales (i.e., $r \ll 1$). Using the asymptotic analysis of the exact solution for $X \gg 1$ allowed us to obtain the necessary conditions of a small-scale instability of a number density of particles. The results obtained by this asymptotic analysis are presented below.

The solution of Eq. (14) has the following asymptotics: for $X \ll 1$ (i.e., in the scales $0 \leq r \ll 1/\sqrt{\mathrm{Pe}}$) the solution for the second moment $\hat{\Phi}$ is given by

$$\hat{\Phi}(X) = \{1 - (\kappa/6)[X^2 + O(X^4)]\} , \qquad (15)$$

where $\kappa = (B - \gamma)/2\beta$. For $X \gg 1$ (i.e., in the scales $1/\sqrt{\mathrm{Pe}} \ll r < 1$) the function $\hat{\Phi}$ is given by

$$\hat{\Phi}(X) = Re\{A X^{-C \pm \sqrt{C^2 - \kappa}}\} . \qquad (16)$$

When $C^2 - \kappa < 0$ the second-order correlation function for a number density of particles $\hat{\Phi}$ is given by

$$\hat{\Phi}(r) = A_3 r^{-C} \cos(\nu_I \ln r + \varphi) , \qquad (17)$$

where $\nu_I = \sqrt{\kappa - C^2}$, $\quad C > 0$ and φ is the argument of the complex constant A. For $r \geq 1$ the second-order correlation function is given by

$$\hat{\Phi}(r) = (A_4/r) \exp(-r\sqrt{3\gamma/2}) , \qquad (18)$$

for $\gamma > 0$. The total number of particles in a closed volume is conserved, i.e., particles can only be redistributed in the volume. It yields the condition $\int_0^\infty r^2 \hat{\Phi}(r)\, dr = \hat{\Phi}(k = 0) = 0$. The later yields $\varphi = -\pi/2$ for $\ln \mathrm{Pe} \ll 1$ and $\gamma \ll 1$. When $C^2 - \kappa > 0$ the solution $\hat{\Phi}(X) = A X^{-C \pm \sqrt{C^2 - \kappa}}$ cannot be matched with solutions (15) and (18). Thus the condition $C^2 - \kappa < 0$ is the necessary condition for the existence of the solution for the correlation function. The condition $C > 0$ provides the existence of the global maximum of the correlation function at $r = 0$. Matching functions Φ and Φ' at the boundaries of the above-mentioned regions yields coefficients A_k and γ. In particular, the growth rate of a small-scale instability of the the second-order correlation function for particles number density is given by

$$\gamma = c_1 + \mathrm{St}^2 c_4 - \frac{(c_2 + \mathrm{St}^2 c_5)^2}{4(c_3 + \mathrm{St}^2 c_6)} - \left(\frac{2\pi k}{\ln(\mathrm{Pe})} \right)^2 (c_3 + \mathrm{St}^2 c_6) , \qquad (19)$$

where $k = 1, 2, 3, \ldots$. Analysis shows that the small-scale instability can be excited in a very wide range of parameters St and σ. There is only a small range of values σ (e.g., $0.02 < \sigma < 0.2$) for which there is no instability of the second moment. The sufficient condition for the generation of fluctuations of particles number density is $\mathrm{Pe} > \mathrm{Pe}^{(\mathrm{cr})}$, where the critical Peclet number $\mathrm{Pe}^{(\mathrm{cr})}$ is given by $\mathrm{Pe}^{(\mathrm{cr})} = \mathrm{Pe}(\gamma = 0)$.

The causes for the excitation of a small-scale instability of fluctuations of particles number density are both, the compressibility of the velocity field and the compressibility of the field of Lagrangian trajectories. The compressibility of the field of Lagrangian trajectories when St $\neq 0$ implies that the number of particles flowing into a small control volume in a Lagrangian frame does not equal to the number of particles flowing out from this control volume during a correlation time. This can result in the depletion of turbulent diffusion.

The role of the compressibility of the velocity field is as follows. The condition $\nabla \cdot \mathbf{v} \neq 0$ is associated with particles inertia. For inertial particles one obtains $\nabla \cdot \mathbf{v} = \tau_p \Delta P_f / \rho$ (see Section 4 and Elperin et al. 1996a; 1998). The inertia of particles results in that particles inside the turbulent eddies are carried out to the boundary regions between the eddies by inertial forces (i.e., regions with low vorticity or high strain rate). For large Peclet numbers $\vec{\nabla} \cdot \mathbf{v} \propto -dn/dt$. Therefore, $dn/dt \propto -\tau_p \Delta P_f / \rho$. Thus there is accumulation of inertial particles (i.e., $dn/dt > 0$) in regions with $\Delta P_f < 0$. Similarly, there is an outflow of inertial particles from the regions with $\Delta P_f > 0$. This mechanism acts in a wide range of scales of a turbulent fluid flow. Turbulent diffusion results in relaxation of fluctuations of particle concentration in large scales. However, in small scales where turbulent diffusion is small, the relaxation of fluctuations of particle concentration is very weak. Therefore the fluctuations of particle concentration are localized in the small scales (see Elperin et al., 1996b; 1998a).

This phenomenon is considered for the case when density of fluid ρ is much less than the material density ρ_p of particles ($\rho \ll \rho_p$). When $\rho \geq \rho_p$ the results coincide with those obtained for the case $\rho \ll \rho_p$ except for the transformation $\tau_p \to \beta_* \tau_p$, where

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

For $\rho \geq \rho_p$ the value $dn/dt \propto -\beta_* \tau_p \Delta P/\rho$. Thus there is accumulation of inertial particles (i.e., $dn/dt > 0$) in regions with the minimum pressure of a turbulent fluid since $\beta_* < 0$. In the case $\rho \geq \rho_p$ we used the equation of motion of particles in fluid flow which 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.

When the second moment of particles number density grows in time the higher moments of passive scalar also grow. The growth rates of the higher moments of particles number density is larger than those of the lower moments, i.e., spatial distribution of particles number density is intermittent (see Elperin et al., 1996b; 1998a). This process can be damped by the nonlinear effects (e.g., two-way coupling between particles and turbulent fluid flow). Note that excitation of the second moment of a number density of particles requires two kinds of compressibilities: compressibility of the velocity field ($\sigma \neq 0$) and compressibility of the field of Lagrangian trajectories (St $\neq 0$), which is caused by a finite correlation time of a random velocity field. Compressibility of the field of Lagrangian trajectories determines the coefficient of turbulent diffusion (i.e. the coefficient D_{mn} near the second-order spatial derivative of the second moment of particles number density in Eq. (8) and causes depletion of turbulent diffusion in small scales even for $\sigma = 0$. On the other hand, compressibility of the velocity field determines a coefficient B(r) near the second moment of a passive scalar in Eq. (8). This term is responsible for the exponential growth of the second moment of a number density of particles.

4 Discussion

In the present study we derived a mean-field equation and an equation for the second moment for a number density of inertial particles (aerosols and droplets) advected by turbulent atmospheric velocity field with a small yet finite correlation time. The finite correlation time of the turbulent velocity field results in the appearance of the higher than the second-order spatial derivatives in these equations and causes a decrease of turbulent diffusion and a modification of an effective drift velocity. The effective mean drift velocity is caused by a compressibility of particles velocity field and results in formation of large-scale inhomogeneities in spatial distribution of aerosols in the vicinity of the atmospheric temperature inversion. A mechanism of formation of small-scale inhomogeneities in particles spatial distribution is also discussed here. This mechanism is associated with an excitation of a small-scale instability of the second moment of number density of particles.

The obtained results are important in some atmospheric phenomena (e. g., atmospheric aerosols, cloud formation and smog formation) and turbulent industrial flows. We considered turbulent velocity field with $\nabla \cdot \mathbf{v} \neq 0$ which is due to, e.g., particle inertia (see, e.g., Maxey, 1987; Elperin et al., 1996a; 1998a). The velocity of particles $\mathbf{v}$ depends on the velocity of the atmospheric 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 atmospheric fluid and gravity force. Solution of the equation of motion for small particles with $\rho_p \gg \rho$ yields

$$\mathbf{v} = \mathbf{u} + \mathbf{W} - \tau_p \{\partial \mathbf{u}/\partial t + [(\mathbf{u} + \mathbf{W}) \cdot \nabla]\mathbf{u}\} + O(\tau_p^2) ,$$

where $\mathbf{u}$ is the velocity of the atmospheric fluid, $\mathbf{W} = \tau_p \mathbf{g}$ is the terminal fall velocity, $\mathbf{g}$ is the acceleration due to gravity, τ_p is the characteristic time of coupling between the particle and atmospheric fluid (Stokes time). For instance, for spherical particles of radius a_* the Stokes time is $\tau_p = m_p/(6\pi a_* \rho \nu)$, where m_p is the particle mass. The velocity field of particles is compressible, i.e., $\nabla \cdot \mathbf{v} \neq 0$. Indeed, the equation for the velocity of particles and Navier-Stokes for atmospheric fluid yield $\nabla \cdot \mathbf{v} = \tau_p \Delta P_f / \rho + O(\tau_p^2)$, where P_f is atmospheric fluid pressure and we neglected small $\nabla \cdot \mathbf{u}$. Since $dn/dt \propto -\vec{\nabla} \cdot \mathbf{v} \sim -\tau_p \Delta P_f / \rho$, in regions where $\Delta P_f < 0$ there is accumulation of inertial particles (i.e., $dn/dt > 0$). Similarly, there is an outflow of inertial particles from the regions with $\Delta P_f > 0$. When there is a large-scale inhomogeneity of the temperature of the turbulent flow, the mean heat flux $\langle \mathbf{u}\theta \rangle \neq 0$. Therefore fluctuations of both, temperature θ and velocity $\mathbf{u}$ of fluid, are correlated. Fluctuations of temperature cause fluctuations of pressure of fluid and vise versa. The pressure fluctuations result in fluctuations of the number density of inertial particles. Indeed, increase (decrease) of the pressure of atmospheric fluid is accompanied by accumulation (outflow) of the particles. Therefore, direction of mean flux of particles coincides with that of heat flux, i.e. $\langle \mathbf{v}n \rangle \propto \langle \mathbf{u}\theta \rangle \propto -\nabla T$, where T is the mean temperature of atmospheric fluid. Therefore 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, e.g., in the vicinity of the temperature inversion layer. The latter results in formation of large-scale inhomogeneities in spatial distribution of aerosols in the vicinity of the atmospheric temperature inversion (for details see Elperin et al., 1996a; 1998).

Using the characteristic parameters of the atmospheric turbulent boundary layer: maximum scale of turbulent flow $l_0 \sim 10^3 - 10^4$ cm; velocity in the scale l_0 : $u_0 \sim 30 - 100$ cm/s; Reynolds number $\mathrm{Re} \sim 10^6 - 10^7$ we estimate the ratio $|\mathbf{V}_{\rm eff}/W|$ and the depletion of the turbulent diffusion coefficient. For particles with material density $\rho_p \sim 1 - 2$ g / cm^3 and radius $a_* = 30 \mu$m the ratio $|\mathbf{V}_{\rm eff}/W| \approx 0.9$ for the temperature gradient $1K/200$ m, where $W \sim 10^{-2} a_*^2$ cm / s and a_* is measured in microns. For these parameters the coefficient of turbulent diffusion in the vertical direction can be depleted by 25% due to the finite correlation time of a turbulent atmospheric velocity field. The latter result is in compliance with the known anisotropy of the coefficient of turbulent diffusion in the atmosphere. Thus, two competitive mechanisms of particles transport, i.e., the mixing by the decreased turbulent diffusion and accumulation of particles due to the effective velocity act simultaneously together with the effect of gravitational settling of particles. This can result in formation of large-scale inhomogeneities in spatial distribution of aerosols in the vicinity of the atmospheric temperature inversion. The characteristic time of excitation of the instability of concentration distribution of particles varies in the range from 0.3 to 3 hours depending on parameters of both, the atmospheric turbulent boundary layer and the temperature inversion layer. We expect that the spatial density $m_p n$ of particles inside the inhomogeneous structures is of the order of the density ρ of surrounding fluid.

The analyzed effect of self-excitation (exponential growth) of fluctuations of particles concentration is important in atmospheric turbulence. Using the parameters of the atmospheric turbulent boundary layer we find that the degree of compressibility of $\sigma \approx 1.2 \times 10^{-6} a_*^4$, where hereafter the size of a particle a_* is measured in microns, and the Stokes time $\tau_p \approx 10^{-5} a_*^2$ for particles with material density $\rho_p = 1$ g / cm^3. The instability is not excited when the degree of compressibility of particles velocity $0.02 < \sigma < 0.2$, i.e., for particles and droplets of the size $14.3 \mu m < a_* < 25.4 \mu m$. On the other hand, for droplets of the size $a_* > 25.4 \mu m$ the small-scale instability can be excited. This effect causes formation of small-scale inhomogeneities in droplet clouds which were recently observed in atmospheric turbulence in small cumulus clouds (see Baker and Brenguier, 1998). Notably, small-scale inhomogeneities in spatial distribution of inertial particles were observed also in laboratory turbulent flow (see Fessler et al., 1994; Hainaux et al., 2000).

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