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Mechanisms of formation of aerosol and gaseous inhomogeneities in the turbulent atmosphere

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Abstract

New mechanisms of formation of large-scale (more than 100–200 m) and small-scale (about 1 cm) inhomogeneities in spatial distributions of aerosols, droplets and gaseous admixtures are discussed. The large-scale inhomogeneities are formed in the vicinity of the temperature inversion layers due to excitation of the large-scale instability. This effect is caused by additional nondiffusive turbulent flux of particles in the vicinity of the temperature inversion. The characteristic time of excitation of the instability of concentration distribution of particles varies in the range from 0.3 to 3 h depending on the particle size and parameters of both, the atmospheric turbulent boundary layer and the temperature inversion layer. The derived equation for the effective velocity of particles provides parameterization of these turbulence effects, and it can be directly incorporated into existing atmospheric models. Small-scale inhomogeneities are related with a self-excitation of fluctuations of inertial particles concentration in turbulent atmosphere. This effect is responsible for the intermittency in spatial distribution of particles and gases.

Keywords: Turbulent atmosphere; Preferential concentration of aerosols and gases

1. Introduction

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 both cumulus and stratiform clouds are strongly inhomogeneous (see, e.g., Paluch and Baumgardner, 1989; Baumgardner and Baker, 1992; Baumgardner and Colpitt, 1995;

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Korolev and Mazin, 1993; Haman and Malinowski, 1996; and references therein). Inhomogeneous distributions of ice particles and aerosols are observed in clouds as well (see, e.g., Hobbs and Rangno, 1985). The importance of the effects of atmospheric turbulence on microphysical processes in clouds was discussed in a number of studies (see, e.g., Pruppacher and Klett 1978; Jonas 1996; Pinsky and Khain 1996, 1997; Telford 1996; Shaw et al., 1998; and references therein).

One of the mechanisms that determine 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, for example, the preferential concentration of particles in atmospheric turbulent fluid flow still remains poorly understood.

The main goal of this paper is to bring attention of the atmospheric science community to three recently discovered phenomena caused by atmospheric turbulence that are of great importance in cloud physics and environmental science. In particular, these phenomena cause the formation of inhomogeneities in spatial distribution of particles, droplets and gaseous admixtures in turbulent atmosphere. These phenomena include:

(1) Turbulent thermal diffusion of small inertial particles (Elperin et al., 1996a). This phenomenon results in formation of aerosol inhomogeneities in spatial distribution of particles concentration in scales larger than the maximum scale of turbulent atmospheric fluid motions.

(2) Turbulent barodiffusion and turbulent thermal diffusion of gaseous admixtures (Elperin et al., 1995, 1997b). This phenomenon causes formation of inhomogeneities in spatial distribution of gaseous admixtures concentration in scales larger than the maximum scale of turbulent atmospheric fluid motions.

(3) Self-excitation of fluctuations of particles and gaseous admixtures concentration in scales smaller than the maximum scale of turbulent atmospheric fluid motions (Elperin et al., 1996b, 1998d). This phenomenon results in formation of small-scale inhomogeneities and intermittency in spatial distribution of particles and gaseous admixtures concentration in turbulent atmosphere.

In this study, additional developments in the theory of these phenomena and their applications in the atmospheric physics are discussed.

2. Formation of aerosol inhomogeneities

An effect of turbulent thermal diffusion of small inertial particles (Elperin et al., 1996a) that results in formation of large-scale inhomogeneities of aerosols in turbulent atmosphere is discussed in this section. Mechanism of the effect of turbulent thermal diffusion is associated with both, particle inertia and correlation between velocity and temperature fluctuations of atmospheric fluid in the presence of a nonzero mean fluid temperature gradient. Turbulent thermal diffusion causes additional nondiffusive turbulent flux of aerosols in the vicinity of the temperature inversion layer. Turbulent thermal diffusion under certain conditions can cause a large-scale instability of spatial distribution of aerosols. Aerosols are concentrated in the vicinity of the minimum of the mean atmospheric temperature.

Let us discuss this effect. Evolution of the number density $n_p(t,\mathbf{r})$ of small inertial particles in turbulent atmospheric fluid flow is determined by the equation:

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

where $\mathbf{v}_p$ is a random velocity field of the particles that they acquire in turbulent atmospheric fluid velocity field, D is the coefficient of molecular diffusion. We consider the case of large Reynolds ($Re = u_0 l_0 / \nu$) and Peclet ($Pe = u_0 l_0 / D$) numbers. Here, l_0 is the maximum scale of atmospheric turbulent motions, u_0 is the characteristic velocity in this scale, ν is the kinematic viscosity. Note that although Eq. (1) does not formally include coagulation, it is applicable to a volume-averaged density of particles. This can be seen by multiplying the stochastic coagulation equation by the volume of the particles and integrating over the size distribution of the aerosols. The integral term weighted with the particle mass vanishes, and we get the continuity Eq. (1) for the volume-averaged density of particles. Thus, the equation can be seen as describing an average effect that does not take explicitly into account the size distribution of the particles.

The velocity of particles $\mathbf{v}_p$ 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 (Maxey and Corrsin 1986; Maxey, 1987):

$$\mathbf{v}_p = \mathbf{v} + \mathbf{W} - \tau_p \left[\frac{\partial \mathbf{v}}{\partial t} + \left[(\mathbf{v} + \mathbf{W}) \cdot \vec{\nabla} \right] \mathbf{v} \right] + O\left(\tau_p^2\right), \tag{2}$$

where $\mathbf{v}$ 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), ρ_p is the material density of particles, 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 m_p is the particle mass. The term $\propto \tau_p$ in Eq. (2) is due to the small, but finite inertia of the particle.

We consider a low-Mach-number compressible turbulent fluid flow $\vec{\nabla} \cdot \mathbf{v} \neq 0$, that is, $\vec{\nabla} \cdot \mathbf{v} = -(\mathbf{v} \cdot \vec{\nabla}\rho)/\rho$. In the atmosphere without temperature inversion, the characteristic density stratification length $\Lambda_\rho = |\vec{\nabla}\rho/\rho|^{-1} \simeq 8$ kilometers, and therefore $\vec{\nabla} \cdot \mathbf{v}$ is small. However, in the atmosphere with temperature inversion, the characteristic density stratification length in the vicinity of the temperature inversion layer $\Lambda_\rho \sim \Lambda_T \simeq 250$–$300$ m, where $\Lambda_T = |\vec{\nabla} T/T|^{-1}$. This implies that in the vicinity of the temperature inversion layer, the value of velocity divergence $\vec{\nabla} \cdot \mathbf{v}$ is not so small.

The velocity field of particles is also compressible, that is, $\vec{\nabla} \cdot \mathbf{v}_p \neq 0$. Eq. (2) for the velocity of particles and Navier–Stokes for atmospheric fluid yield

$$\vec{\nabla} \cdot \mathbf{v}_p = \vec{\nabla} \cdot \mathbf{v} - \tau_p \vec{\nabla} \cdot \left(\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} \right) + O\left(\tau_p^2\right) = \vec{\nabla} \cdot \mathbf{v} + \tau_p \vec{\nabla} \cdot \left(\frac{\vec{\nabla} P_f}{\rho} \right) + O\left(\tau_p^2\right). \tag{3}$$

where P_f is atmospheric fluid pressure.

The mechanism of formation of particles inhomogeneities for $\rho_p \gg \rho$ is as follows. The inertia of particles results in that particles inside the turbulent eddy are carried out to the boundary regions between the eddies by inertial forces (i.e., regions with low vorticity or high strain rate, see, e.g., Maxey et al., 1996). On the other hand, the inertia effect causes $\vec{\nabla}\cdot \mathbf{v}_p \propto \tau_p \Delta P_f \neq 0$ [see Eq. (3)]. In addition, for large Peclet numbers $\vec{\nabla}\cdot \mathbf{v}_p \propto -\mathrm{d}n_p/\mathrm{d}t$ [see Eq. (1)]. Therefore, $\mathrm{d}n_p/\mathrm{d}t \propto -\tau_p \Delta P_f$. This means that in regions where $\Delta P_f < 0$, there is accumulation of inertial particles (i.e., $\mathrm{d}n_p/\mathrm{d}t > 0$). Similarly, there is an outflow of inertial particles from the regions with $\Delta P_f > 0$. In a 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. Location of these regions is not correlated with the turbulent velocity field. Therefore, they do not contribute into the large-scale flow of inertial particles.

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 θ and velocity $\tilde{\mathbf{u}}$ of fluid, are correlated. Note that the fluctuations of particles velocity $\mathbf{u} = \tilde{\mathbf{u}} + O(\tau_p)$ [see Eq. (2)]. Fluctuations of temperature cause fluctuations of pressure of fluid and vise versa (see the equation of state $P_f = \rho \kappa_B T_f / m_\mu$, where κ_B is the Boltzmann constant, and m_μ is the mass of molecules of atmospheric fluid, P_f and T_f are the pressure and temperature of fluid, respectively). 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, that is, $\langle \tilde{\mathbf{u}} n_p \rangle \propto \langle \tilde{\mathbf{u}} \theta \rangle \propto -\vec{\nabla} T$, where $T = \langle T_f \rangle$ is the mean temperature of atmospheric fluid with the characteristic value T^*, and $T_f = T + \theta$. 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). This effect is more pronounced when atmospheric turbulent fluid flow is inhomogeneous in the direction of the mean temperature gradient. Thus, in turbulent atmosphere, the effect of turbulent thermal diffusion causes accumulation of aerosol particles in the vicinity of temperature inversion. On the other hand, turbulent diffusion results in relaxation of the particle inhomogeneities. Thus, two competitive mechanisms of particles transport, that is, the mixing by turbulent diffusion and accumulation of particles due to turbulent thermal diffusion exist simultaneously together with the effect of gravitational settling of particles.

Now we study these effects quantitatively. We consider large-scale dynamics of small inertial particles and average Eq. (1) over an ensemble of random atmospheric fluid velocity fluctuations. It yields the equations for the mean number density of particles $N = \langle n_p \rangle$

$$\frac{\partial N}{\partial t} + \vec{\nabla}\cdot \left[N(\mathbf{V} + \mathbf{W} + \mathbf{V}_{\mathrm{eff}}) - D_T \vec{\nabla} N \right] = 0, \tag{4}$$

where the effective velocity is given by

$$\mathbf{V}_{\mathrm{eff}} = -\langle \tau \mathbf{u} (\vec{\nabla}\cdot \mathbf{u}) \rangle = -(2/3) W A(Re, a^*) \Lambda_P \ln(Re) (\vec{\nabla} T)/T, \tag{5}$$

where we neglected small molecular flux of particles and compressibility of a surrounding fluid. Here $A(Re,a^*)=1$ for $a^* < a_{cr}$, and $A(Re,a^*)=1-3\ \ln(a^*/a_{cr})/\ln(Re)$ for $a^* \geq a_{cr}$, and $a_{cr}=r_d(\rho/\rho_p)^{1/2}$, and $Re=l_0 u_0/\nu$ is the Reynolds number, and $r_d = l_0 Re^{-3/4}$ is the viscous scale of turbulent fluid flow, and $\mathbf{V}$ is the mean velocity of atmospheric fluid, $D_T = u_0 l_0/3$ is the coefficient of turbulent diffusion. Thus, for example, $a_{cr} \sim 20\ \mu$m for $Re=10^7$, $l_0=100$ m and $\rho_p = 1$ g/cm^3. The effective velocity $\mathbf{V}_{eff}$ of particles determines a turbulent contribution to particle velocity due to both, effect of inertia and mean temperature gradient. This additional particle velocity can cause formation of inhomogeneities in the spatial distribution of inertial particles. The deviation of the function $A(Re,a^*)$ from the unity is caused by the deviation from Eq. (2).

The turbulent flux of particles is given by

$$\mathbf{J}_T = N\,\mathbf{V}_{eff} - D_T \vec{\nabla} N, \tag{6}$$

and we used Eqs. (4) and (5). The additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{eff}$ 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 Θ of the particles number density

$$\frac{\partial \Theta}{\partial t} - D\Delta\Theta = -\vec{\nabla}\cdot(N\mathbf{u} + \mathbf{Q}), \tag{7}$$

where $n_p = N + \Theta$, $\mathbf{Q} = \mathbf{u}\Theta - \langle \mathbf{u}\Theta \rangle$. Eq. (7) 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 turnover time of turbulent eddies. Thus, the turbulent field Θ is of the order of

$$\Theta \sim -\tau N(\vec{\nabla}\cdot\mathbf{u}) - \tau(\mathbf{u}\cdot\vec{\nabla})N.$$

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

$$\mathbf{J}_T \sim -N\langle \tau\mathbf{u}(\vec{\nabla}\cdot\mathbf{u})\rangle - \langle \tau\mathbf{u}u_j\rangle \nabla_j N. \tag{8}$$

The first term in Eq. (8) describes the additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{eff}$. 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,c).

Remarkably, Eq. (5) 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. (5) that the ratio $|\mathbf{V}_{eff}/W|$ is of the order of

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

where δT is the temperature difference in the scale ΛT, and T^* is the characteristic temperature. The ratio $|\mathbf{V}_{eff}/W|$ for typical atmospheric parameters (see below) and for

Table 1
The ratio $|\mathbf{V}_{\mathrm{eff}}/W|$ for typical atmospheric parameters and for different temperature gradients and different particle sizes

	1 K/100 m	1 K/200 m	1 K/300 m	1 K/1000 m
$a_* = 30$ μm	2.7	1.35	0.9	0.27
$a_* = 50$ μm	2.43	1.22	0.81	0.243
$a_* = 100$ μm	2.06	1.03	0.687	0.206
$a_* = 200$ μm	1.7	0.85	0.567	0.17
$a_* = 300$ μm	1.5	0.75	0.5	0.15
$a_* = 500$ μm	1.2	0.6	0.4	0.12

different temperature gradients and different particle sizes is presented in Table 1, where we used Eq. (5). It is seen from Table 1 that the additional particle velocity $\mathbf{V}_{\mathrm{eff}}$ is of the order of W or larger than the terminal fall velocity W. In the atmosphere without temperature inversion, the effective particle velocity is directed opposite to the terminal fall velocity, and thus the effective particle velocity decreases the effective sedimentation velocity in 10–30%. In the atmosphere with a temperature inversion, the effective particle velocity $\mathbf{V}_{\mathrm{eff}}$ 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 $\mathbf{V}_{\mathrm{eff}}$ 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. The characteristic time of formation of inhomogeneities of particles is

$$\tau_{\mathrm{f}} \sim \Lambda_T / |V_{\mathrm{eff}} - W|.$$

The latter equation implies that $\tau_{\mathrm{f}} \propto a_*^{-2}$. This dependence is caused by the particle size dependence of the terminal fall velocity. The formation of inhomogeneities is possible when $|\mathbf{V}_{\mathrm{eff}}| > |\mathbf{W}|$. 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

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

Therefore, it is important to take into account the additional turbulent nondiffusive flux of particles due to the effective velocity $\mathbf{V}_{\mathrm{eff}}$ in atmospheric phenomena (e.g., atmospheric aerosols, cloud formation and smog formation). Observations of the vertical distributions of aerosols in the atmosphere show that maximum concentrations can occur within temperature inversion layers (see, e.g., Lu and Turco, 1994; and references therein). Using the characteristic parameters of the atmospheric turbulent boundary layer

(Seinfeld, 1986; Jaenicke, 1987): 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 $Re \sim 10^6$–10^7, we obtain that for particles with material density $\rho_p \sim 1$–2 g/cm^3 and radius $a_* = 30$ μm, the characteristic time of formation of inhomogeneities of the order of 11 min for the temperature gradient 1 K/100 m and 106 min for the temperature gradient 1 K/200 m. For particles of the size $a^* = 100$ μm, the characteristic time of formation of inhomogeneities of the order of 1 min for the temperature gradient 1 K/100 m and 121 min for the temperature gradient 1 K/200 m (used in Table 1). These estimates are in compliance with the characteristic time of formation of inhomogeneous structures in atmosphere. We expect that the spatial density $m_p n_p$ of particles inside the inhomogeneous structures is of the order of the density ρ of surrounding fluid.

It is conceivable to suggest that a trapping of ice particles in ‘‘cold gas traps’’ observed in the recent experiments (see Banerecker and Neidhart, 1998) can be explained by the abovementioned effect due to the additional particle velocity $\mathbf{V}_{eff}$.

3. Generation of fluctuations of number density of inertial particles and gaseous admixtures

In this section, we discuss a new effect that causes a self-excitation of fluctuations of inertial particles or gaseous admixtures concentration in turbulent atmospheric fluid flow (Elperin et al., 1996b). This effect is responsible for the intermittency in particle spatial distribution. In particular, we have shown that the growth rates of the higher moments of particles concentration is higher than those of the lower moments.

Physics of self-excitation of fluctuations of particles concentration is as follows. The inertia of particles results in that particles inside the turbulent eddy are carried out to the boundary regions between the eddies by inertial forces (i.e., regions with low vorticity or high strain rate, see, e.g., Maxey et al., 1996). On the other hand, the inertia effect causes $\vec{\nabla} \cdot \mathbf{v}_p \propto \tau_p \Delta P_f \neq 0$ [see Eq. (3)]. In addition, for large Peclet numbers $\vec{\nabla} \cdot \mathbf{v}_p \propto -\mathrm{d} n_p / \mathrm{d} t$ [see Eq. (1)]. Therefore, $\mathrm{d} n_p / \mathrm{d} t \propto -\tau_p \Delta P_f$. This means that in regions where $\Delta P_f < 0$, there is accumulation of inertial particles (i.e., $\mathrm{d} n_p / \mathrm{d} t > 0$). 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 turbulent atmospheric fluid flow. Turbulent diffusion results in relaxation of fluctuations of particles concentration in large scales. However, in small scales where scale-dependent turbulent diffusion is small, the relaxation of fluctuations of particles concentration is very weak. Therefore, the fluctuations of particles concentration are localized in the small scales.

Divergent velocity field of particles is the main reason of a new effect, that is, self-excitation (exponential growth) of fluctuations of concentration of small particles in a turbulent fluid flow. Indeed, multiplication of Eq. (1) by n_p and simple manipulations yield

$$\frac{\partial \langle n_p^2 \rangle}{\partial t} \sim -\left\langle n_p^2 \left(\vec{\nabla} \cdot \mathbf{v}_p \right) \right\rangle - 2 D \left\langle \left(\vec{\nabla} n_p \right)^2 \right\rangle . \tag{9}$$

Eq. (1) implies that variation of particles concentration during the time interval $\tau_0 = l_0/u_0$, around the value $n_p^{(0)}$ is of the order of $\delta n_p \sim -n_p^{(0)}\tau_0(\vec{\nabla}\cdot \mathbf{v}_p)$, where u_0 is the characteristic velocity in the energy containing scale l_0. Substitution $n_p = n_p^{(0)} + \delta n_p$ into Eq. (9) yields $\partial\langle n_p^2\rangle/\partial t \sim 2\tau_0\langle n_p^2(\vec{\nabla}\cdot \mathbf{v}_p)^2\rangle$. Therefore, the growth rate of fluctuations of particles concentration $\gamma_0 \sim 2\tau_0\langle(\vec{\nabla}\cdot \mathbf{v}_p)^2\rangle$. This estimate in a good agreement with the analytical results obtained below [see Eq. (10) for $r = 0$].

The use of the technique (see, e.g., Elperin et al., 1995, 1996a,b, 1997a,b,c, 1998a,b,c,d) allows to derive the equation for the correlation function $\Phi(t,\mathbf{r}) = \langle n_p(t,\mathbf{x})n_p(t,\mathbf{x}+\mathbf{r})\rangle - N(t,\mathbf{x})N(t,\mathbf{x}+\mathbf{r})$:

$$\frac{\partial\Phi}{\partial t} = 2\left[D\delta_{mn} + D_{mn}(0) - D_{mn}(\mathbf{r})\right]\frac{\partial^2\Phi}{\partial r_m \partial r_n} + 2\langle\tau b(\mathbf{x})b(\mathbf{x}+\mathbf{r})\rangle\Phi$$
$$-4\langle\tau u_m(t,\mathbf{x})b(t,\mathbf{x}+\mathbf{r})\rangle\frac{\partial\Phi}{\partial r_m} + I, \tag{10}$$

where $I = 2\langle\tau b(\mathbf{x})b(\mathbf{x}+\mathbf{r})\rangle N^2$, and $D_{mn} = \langle\tau u_p u_m\rangle$, and $\mathbf{v}_p = \mathbf{V} + \mathbf{u}$, $\mathbf{V} = \langle\mathbf{v}_p\rangle$ is the mean velocity, $\mathbf{u}$ is the random component of the particles velocity, $b = \vec{\nabla}\cdot\mathbf{u}$, and τ is the momentum relaxation time of turbulent atmospheric fluid velocity field that depends on the scale of turbulent motions.

An asymptotic analysis of solutions of Eq. (10) yields the growth rate of fluctuations of particles concentration

$$\gamma_0 = \frac{2\left[c^2 + (q-a)^2\right]^2}{3q^4(3-p)^2 r_a^{2q}}\ln^2\left(\frac{Re}{Re^{(\mathrm{cr})}}\right), \tag{11}$$

(Elperin et al., 1996b), where $r_a = (\tau_p/\tau_0)^{1/(p-1)}$, $Re > Re^{(\mathrm{cr})}$ and the critical Reynolds number $Re^{(\mathrm{cr})}$

$$Re^{(\mathrm{cr})} \simeq r_a^{p-3}\exp\left[\frac{3-p}{c}\left(\pi k + \arctan\frac{q-a}{c} + \arctan(3/2 - \mu - a + c^*\zeta_i)\right)\right], \tag{12}$$

where $k = 1,2,3,\ldots$ Note that when $\tau_p \geq \tau_0$ in Eqs. (11) and (12), r_a is set equal 1. Here, we used a model of homogeneous and isotropic turbulent velocity field of inertial particles with $\nabla\cdot\mathbf{v}_p \neq 0$. The second moment of the velocity field $\langle\tau u_m(\mathbf{x})u_n(\mathbf{x}+\mathbf{r})\rangle$ is given by:

$$\langle\tau u_m(\mathbf{x})u_n(\mathbf{x}+\mathbf{r})\rangle$$
$$= D_T\left[\left[F(r) + F_c(r)\right]\delta_{mn} + \frac{rF'}{2}\left(\delta_{mn} - \frac{r_m r_n}{r^2}\right) + (rF_c')\frac{r_m r_n}{r^2}\right] \tag{13}$$

(for details, see Elperin et al., 1995, 1996b), where $F' = \mathrm{d}F/\mathrm{d}r$, $F(0) = 1 - F_c(0)$. The function $F_c(r)$ describes the compressible (potential) component whereas $F(r)$ corresponds to the vortical part of the turbulent velocity of particles. Incompressible $F(r)$ and compressible $F_c(r)$ components of the turbulent velocity field are given by

$$F(r) = (1-\varepsilon)(1-r^{q-1}), \quad F_c(r) = \varepsilon(1-r^{q-1}),$$

where $r_d < r \ll 1$, and r is measured in the units of l_0, $q = 2p-1$, p is the exponent in spectrum of turbulent kinetic energy of atmospheric fluid, $r_d = Re^{-1/(3-p)}$, $Re = l_0 u_0/\nu \gg 1$ is the Reynolds number, ν is the kinematic viscosity of atmospheric fluid (see Elperin et al., 1996c). The parameter r_d coincides with the viscous scale of Kolmogorov turbulence (for $p = 5/3$), that is, $r_d \approx Re^{-3/4}$. In Eqs. (11) and (12), parameters a, and μ and c are given by $a = [q - \sigma(2q^2 + 3q - 6)]/2(1+q\sigma)$, and $\mu = 15\sigma/(1+3\sigma)$, and $c = \sqrt{M(q,\sigma)}/2(q+3)(1+q\sigma)$, where the parameter compressibility is $\sigma = \varepsilon/(1-\varepsilon)$, and $M(q,\sigma) = b_1 B^2 + b_2 B + b_3$, and $B(q,\sigma) = 2\sigma(q+3) - 1$, and $b_1(q) = 4(q-1)(9-2q^2) - q^2$, and $b_2(q) = 2(q+2)[2(q-1)(4q^2+9q+9) - 3q]$, and $b_3(q) = 3(q+2)^2(2q-3)(2q+1)$. The range of values of σ is given by

$$\sigma_1 < \sigma < \sigma_2, \tag{14}$$

where $\sigma_i = (B_i + 1)/2(q+3)$, and B_i are the roots of the equation $M = 0$. For Kolmogorov energy spectrum of turbulent velocity field $p = 5/3$, we obtain $\sigma_1 \simeq 1/20$ and $\sigma_2 \simeq 7$. Here, we used that Eq. (2) is valid only in scales $r_a \ll r < l_0$ [where $\tau_p \ll \tau(k)$]. On the other hand, in scales $r_d < r \le r_a$ [where $\tau(k) \le \tau_p < \tau_0$], Eq. (2) is not valid and we have to solve the equation of motion for particles (see Elperin et al., 1996b, 1998d).

The analyzed effect of the self-excitation of fluctuations of droplets concentration is important in atmospheric turbulence. For instance, using the parameters of the atmospheric turbulent boundary layer: $u_0 \sim 30$–100 cm/s; $l_0 \sim 10^3$–10^4 cm, and a droplet size $a^* = 200$ μm, we find that the excited fluctuations of droplets concentration are localized in scales $l_f \sim r_a \sim 0.5$–3 cm. When the exponent of the spectrum of turbulent kinetic energy of atmospheric fluid $p = 5/3$ (Kolmogorov turbulence) and degree of compressibility of particles velocity field $\sigma = 0.1$, we obtain that the critical Reynolds number $Re^{(cr)} \sim 6 \times 10^5$ for droplets of radius $a^* = 200$ μm. In atmospheric turbulence, the Reynolds number $Re \sim 10^6$–10^7. Therefore, the fluctuations of droplets concentration can be excited in atmospheric turbulent fluid flow. The magnitude of fluctuations of particles number density can be estimated as follows. The nonlinear effects become important when $m_p n_p \sim \rho$. In this case, the interphase friction between particles and fluid results in strong effect of particles to the motion of fluid. This causes a saturation of the small-scale instability. Therefore, the magnitude of fluctuations $\sqrt{\langle n_p^2 \rangle} \sim \rho/m_p \sim 2.4 \times 10^8 a^{*-3}$, where the particle size a^* microns, and we used that $\rho/\rho_p = 10^{-3}$. For droplets of radius $a^* = 200$ μm, we obtain the magnitude of fluctuations $\sqrt{\langle n_p^2 \rangle} \sim 30\ cm^{-3}$. This corresponds to the liquid water content that is of the order of 1.

This phenomenon was also observed in laboratory (see Fessler et al., 1994). The developed theory is in a good agreement with these experiments. This effect was also observed recently in atmospheric turbulent flows (see Baker and Brenguier, 1999). The above described effect can cause formation of small-scale inhomogeneities in droplet clouds and may increase in droplets coagulation rates. In particular, recent numerical simulations demonstrated a considerable increase in particles coagulation rates in turbulent flows (see Sundaram and Collins, 1997; Reade and Collins, 1998; Zhou et al., 1998).

The small-scale instability described above causes formation of small-scale clusters. The instability is excited in the atmospheric turbulence for particles with size that is more than 100 μm. However, small-scale inhomogeneities can be formed without this small-scale instability. They can be created due to the source I in Eq. (10). In this case, the amplitude of fluctuations in small-scale inhomogeneities is much less than that caused by the small-scale instability (it is larger than the mean number density of particles in 2–3 times). This mechanism is essential for particles with the size $a^* \sim 10$–30 μm. The mechanism is also associated with a particle inertia.

Note also that for larger particles and droplets, the self-excitation of concentration fluctuations of inertial particles causes formation of anisotropic (elongated in the gravity acceleration direction) inhomogeneities. Probably, these inhomogeneities are related to the recently studied tracks collection of the falling droplets (see Maxey 1987; Wang and Maxey 1993; Khain and Pinsky 1995; Maxey et al., 1996; Pinsky and Khain 1996, 1997).

Now we discuss an effect of self-excitation of fluctuations of concentration of gaseous admixture and small non-inertial particles in a turbulent low-Mach-number compressible fluid flow. The reason for self-excitation of fluctuations of non-inertial particles concentration is that accumulation and outflow of the particles of the passive scalar in a small volume due to fluid compressibility are separated in time and are not balanced in compressible atmospheric fluid flow. Molecular diffusion breaks a symmetry between accumulation and outflow, that is, it breaks a reversibility in time and does not allow leveling of the total mass fluxes over the consecutive intervals of time. Certainly, the compressibility of the turbulent atmospheric fluid flow results only in a redistribution of the particles in the volume. In the whole volume, the total quantity of particles is conserved.

The growth rate of fluctuations of non-inertial particles concentration and the critical Reynolds number $Re^{(cr)}$ are determined by Eqs. (11) and (12), respectively whereby r_a is set equal 1. The characteristic scales of localization of the fluctuations of particles concentration are of the order

$$l_\mathrm{f} \sim l_0 Re^{-1/(3-p)} \exp(\pi n/c), \tag{15}$$

where $n \le k$. The analysis of fluctuations of gaseous admixtures concentration is similar to that for particles concentration. The growth rate γ_0, the critical Reynolds number $Re^{(cr)}$ and the characteristic scales l_f of fluctuations of gaseous admixtures concentration are determined by Eqs. (11) and (12) with $r_a = 1$ in which one need to change $1/(3-p) \to 2/(p+1)$.

The analyzed effect of the self-excitation of fluctuations of concentration of gaseous admixtures may cause formation of small-scale inhomogeneities in smoke clouds, gaseous pollutants and vapour distribution in turbulent atmosphere. The latter phenomenon may cause increase in droplet nucleation rates (see, e.g., Easter and Peters, 1994; Kulmala et al., 1997).

4. Discussion and conclusions

In this paper, we discussed new mechanisms of formation of large-scale (more than 100–200 m) and small-scale (about 1 cm) inhomogeneities in spatial distributions of aerosols, droplets and gaseous admixtures.

The large-scale inhomogeneities are formed in the vicinity of the temperature inversion layers due to excitation of the large-scale instability. This effect is caused by additional nondiffusive turbulent flux of particles in the vicinity of the temperature minimum (e.g., temperature inversion layer). The characteristic time of formation of large-scale inhomogeneities in concentration distribution of particles varies in the range from 0.3 to 3 h depending on parameters of both, the atmospheric turbulent boundary layer and the temperature inversion layer. This time τ_f depends on the particle size a^*, that is, for inertial particles the time $\tau_f \sim a^{*-2}$. We expect that the spatial density $m_p n_p$ of particles inside the inhomogeneous structures is of the order of the density ρ of surrounding fluid. However, for the droplets with the size more than 400 μm, the instability is not effective due to the sedimentation effects. In the absence of turbulence, the instability is not excited and large-scale inhomogeneities are not formed. Remarkably, the derived Eq. (5) for the effective velocity of particles provides parameterization of these turbulence effects, and it can be directly incorporated to existing atmospheric models.

Small-scale inhomogeneities are related with a self-excitation of fluctuations of inertial particles concentration in turbulent atmosphere. This effect is responsible for the intermittency in spatial distribution of inertial particles. The effect is caused by inertia of aerosols that results in divergent velocity field of particles. The inertia effect causes $\vec{\nabla} \cdot \mathbf{v}_p \propto \tau_p \Delta P_f \neq 0$ [see Eq. (3)]. On the other hand, for large Peclet numbers $\vec{\nabla} \cdot \mathbf{v}_p \propto -\mathrm{d}n_p/\mathrm{d}t$ [see Eq. (1)]. Therefore, $\mathrm{d}n_p/\mathrm{d}t \propto -\tau_p \Delta P_f$. This means that in regions where $\Delta P_f < 0$, there is accumulation of inertial particles (i.e., $\mathrm{d}n_p/\mathrm{d}t > 0$). 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 particles concentration in large scales. However, in small scales where turbulent diffusion is very small, the relaxation of fluctuations of particles concentration is very weak. Therefore, the fluctuations of particles concentration are localized in the small scales. For instance, using the parameters of the atmospheric turbulent boundary layer: $u_0 \sim 30$–100 cm/s; $l_0 \sim 10^3$–10^4 cm, and a droplet size $a^* = 200$ μm, we find that the excited fluctuations of droplets concentration are localized in scales $l_f \sim r_a \sim 0.5$–3 cm. When the exponent of the spectrum of turbulent kinetic energy of atmospheric fluid $p = 5/3$ (Kolmogorov turbulence) and degree of compressibility of particle's velocity field $\sigma = 0.1$, we obtain that the critical Reynolds number (i.e., the threshold for generation of fluctuations of particles concentration) $Re^{(cr)} \sim 6 \times 10^5$ for droplets of radius $a^* = 200$ μm. In atmospheric turbulence, the Reynolds number $Re \sim 10^6$–10^7. Therefore, the fluctuations of droplets concentration can be excited in atmospheric turbulent fluid flow. This effect can cause formation of small-scale inhomogeneities in droplet clouds. This phenomenon was also observed in laboratory (see Fessler et al., 1994). The developed theory is in a good agreement with these experiments. Small-scale inhomogeneities of gaseous admixtures concentration can be formed due to compressibility of a low-Mach-number turbulent atmospheric flow.

The effect of preferential concentration of small inertial particles and gaseous admixtures may exist in all types of clouds and can act as a universal turbulence induced mechanism that substantially increases the rate of the collection processes, especially in the early stages of cloud development.

In conclusion, it is of interest to note that since equation of convective heat transfer in inelastic approximation coincides in form with the equation for particles number density, small-scale inhomogeneities of temperature distribution can be excited in turbulent atmospheric flows (see, e.g., Elperin et al., 1997d).

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